

**A STUDY OF VISCOELASTIC PROPERTIES
OF FLY ASH/NATURAL RUBBER COMPOSITES**

THANUNYA SAOWAPARK

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Thesis
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.....
Miss Thanunya Saowapark
Candidate

.....
Assoc. Prof. Chakrit Sirisinha,
Ph.D.
Major advisor

.....
Prof. Narongrit Sombatsompop,
Ph.D.
Co-advisor

.....
Mr. Pongdhorn Sae-Oui,
Ph.D.
Co-advisor

.....
Prof. Banchong Mahaisavariya,
M.D., Dip Thai Board of Orthopedics
Dean
Faculty of Graduate Studies
Mahidol University

.....
Assist. Prof. Panya Sunintaboon,
Ph.D.
Program Director
Doctor of Philosophy Program in
Polymer Science and Technology
Faculty of Science
Mahidol University

Thesis
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for the degree of Doctor of Philosophy (Polymer Science and Technology)
on
December 27, 2012

.....
Miss Thanunya Saowapark
Candidate

.....
Assoc. Prof. Jatuphorn Wootthikanokkhan,
Ph.D.
Chair

.....
Assoc. Prof. Chakrit Sirisinha,
Ph.D.
Member

.....
Mr. Pongdhorn Sae-Oui,
Ph.D.
Member

.....
Prof. Narongrit Sombatsompop,
Ph.D.
Member

.....
Prof. Banchong Mahaisavariya,
M.D., Dip Thai Board of Orthopedics
Dean
Faculty of Graduate Studies
Mahidol University

.....
Prof. Skorn Mongkolsuk,
Ph.D.
Dean
Faculty of Science
Mahidol University

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Thanunya Saowapark

A STUDY OF VISCOELASTIC PROPERTIES OF FLY ASH/NATURAL RUBBER COMPOSITES

THANUNYA SAOWAPARK 4938073 SCPO/D

Ph.D. (POLYMER SCIENCE AND TECHNOLOGY)

THESIS ADVISORSY COMMITTEE : CHAKRIT SIRISINHA, Ph.D.,
NARONGRIT SOMBATSOMPOP, Ph.D., PONGDHORN SAE-OUI, Ph.D.

ABSTRACT

Fly ash (FA) as a by-product of power station plants consists of silicon dioxide (SiO_2) similar to precipitated silica. Typically, FA particles are spherical with smooth surfaces having significant influences on mechanical and properties of polymeric composites. According to previous work, only the mechanical properties of FA filled NR have been studied and compared with precipitated silica. Thus, in this research, further investigation in properties of FA filled NR is extended by focusing mainly on the viscoelastic properties. The results obtained reveal that the incorporation of FA in NR increases storage modulus (G') and bulk viscosity under both oscillatory and steady shear flows. Moreover, the higher the FA loading, the larger the magnitude of viscous response in the FA filled NR compounds. The explanation is proposed in terms of a ball-bearing effect of FA particles associated with a molecular degradation of the rubber matrix as induced by the complex substances formed via fatty acids in the non-rubber component of NR together with heavy metals in FA particles. Such complex substances are believed to catalyze the degradation process of rubber molecules leading to storage instability of the compounds. Such a degradation process could effectively be suppressed by the addition of an amine-based antioxidant (6-PPD).

By the incorporation of silanes, namely, Si-69 (bis (3-triethoxysilylpropyl) tetrasulfide), Si-264 (3-thiocyanatopropyl triethoxy silane), and VTEO (vinyltriethoxysilane), the increase in G' of compounds is found mainly in the system with Si-69 silane. Also, the pre-surface treatment of milled FA particles is shown to be an appropriate technique for FA surface treatment. With various curing systems, the cure reversion phenomenon is observed in the sulfur system with increased FA loading. On the other hand, the peroxide curing system exhibits the marching phenomenon, particularly at high FA loading greater than 150 phr. Additionally, the NR vulcanizates filled with FA in all curing systems show progressive decrease in tensile strength with increased FA loading, due to poor filler-rubber interactions. By treating FA surfaces with silanes, a marginal improvement in mechanical properties of NR/FA composites is observed in all curing systems with Si-69 silane due to the free sulfur released from the Si-69 during the curing process. On the contrary, insignificant changes in mechanical properties with other silanes are attributed to the small amount of active silanol groups on FA surfaces.

KEY WORDS: FLY ASH/ NATURAL RUBBER/ VISCOELASTIC PROPERTIES/
FILLER/RHEOLOGY

430 pages

การศึกษาสมบัติสภาพหยุ่นหนืดของวัสดุเชิงประกอบของยางธรรมชาติที่มีเถ้าลอยเป็นสารตัวเติม

A STUDY OF VISCOELASTIC PROPERTIES OF FLY ASH/NATURAL RUBBER COMPOSITES

ธัญญา เสาวภาคย์ 4938073 SCPO/D

ปร.ค. (วิทยาศาสตร์และเทคโนโลยีพอลิเมอร์)

คณะกรรมการที่ปรึกษาวิทยานิพนธ์ : ชاکริต สิริสิงห, Ph.D., ณรงค์ฤทธิ์ สมบัติสมภพ, Ph.D., พงษ์ธร แซ่อู่, Ph.D.

บทคัดย่อ

เถ้าลอยนั้นเป็นวัสดุผลพลอยได้ที่ได้จากโรงงานผลิตกระแสไฟฟ้ามีองค์ประกอบคือซิลิกอนไดออกไซด์ เช่นเดียวกับซิลิกา โดยปกติแล้วอนุภาคเถ้าลอยมีลักษณะเป็นทรงกลมที่มีพื้นผิวเรียบซึ่งมีผลต่อสมบัติสภาพเชิงกลซึ่งในงานวิจัยก่อนหน้านี้ได้ทำการศึกษาสมบัติเชิงกลของยางธรรมชาติที่มีเถ้าลอยเป็นสารตัวเติมดังนั้นในงานวิจัยนี้จึงได้ศึกษาต่อเนื่องโดยทำการศึกษาสมบัติสภาพหยุ่นหนืดของยางธรรมชาติที่มีเถ้าลอยเป็นสารตัวเติม ผลการทดลองพบว่าการเติมเถ้าลอยลงในยางธรรมชาตินั้นทำให้โมดูลัสการสะสมและความหนืดภายใต้การไหลด้วยแรงเฉือนแบบคงตัว (Steady shear flow) และแบบออสซิลลาทอรี (Oscillatory flow) ของยางคอมพาวด์เพิ่มขึ้น นอกจากนี้พบว่าปริมาณของเถ้าลอยที่เพิ่มมากขึ้นส่งผลให้ปริมาณการตอบสนองต่อเชิงของไหล (Viscous response) ในยางธรรมชาติที่มีเถ้าลอยเป็นสารตัวเติมสูงมากขึ้น ซึ่งสามารถอธิบายได้จากผลของการกลิ้งแบบลูกปืน (Ball bearing effect) ของอนุภาคเถ้าลอยรวมกับการเกิดการเสื่อมสลายโมเลกุลยางธรรมชาติซึ่งถูกกระตุ้นโดยสารประกอบเชิงซ้อนที่เกิดจากกรดไขมันที่ประกอบอยู่ในส่วนที่ไม่ใช่ยาง (Non-rubber substances) ในยางธรรมชาติ กับโลหะหนักในอนุภาคเถ้าลอย โดยเชื่อว่าสารประกอบเชิงซ้อนดังกล่าวสามารถเร่งกระบวนการเสื่อมสลายของโมเลกุลยาง ซึ่งการเสื่อมสลายดังกล่าวสามารถถูกยับยั้งอย่างมีประสิทธิภาพได้โดยการเติมสารกันเสื่อมประเภทเอมีน (Amine-based antioxidant) จากการเติมสารควบคู่ไซเลน 3 ชนิด คือ Si-69 Si-264 และ VTEO พบว่ามีเพียงสารควบคู่ไซเลน Si-69 ที่ส่งผลให้โมดูลัสการสะสมของสารประกอบของยางธรรมชาติที่มีเถ้าลอยเพิ่มขึ้น นอกจากนี้พบว่าวิธีการทำการปรับปรุงพื้นผิวของเถ้าลอยที่มีการบดก่อนเป็นวิธีที่เหมาะสมในการปรับปรุงพื้นผิวของเถ้าลอย ในส่วนผลของระบบการคงรูป พบว่าการคงรูปด้วยกัมมะถันเกิดปรากฏการณ์ที่โมดูลัสของยางมีค่าลดลงเมื่อปริมาณของเถ้าลอยนั้นเพิ่มขึ้น ในทางตรงกันข้ามการคงรูปด้วยเปอร์ออกไซด์แสดงการเกิดปรากฏการณ์ที่โมดูลัสของยางมีค่าเพิ่มขึ้น โดยเฉพาะเมื่อมีปริมาณของเถ้าลอยมากกว่า 150 ส่วน เมื่อเทียบกับยาง 100 ส่วน โดยน้ำหนัก นอกจากนี้ยางธรรมชาติที่มีเถ้าลอยเป็นสารตัวเติมในทุกะบบการคงรูปแสดงการลดลงของแรงดึงอย่างต่อเนื่องเมื่อปริมาณของเถ้าลอยเพิ่มขึ้นเนื่องจากอันตรกิริยาระหว่างสารตัวเติมและยางธรรมชาตินี้น้อย เมื่อทำการปรับปรุงเถ้าลอยด้วยสารควบคู่ไซเลน พบว่าสมบัติเชิงกลเพิ่มขึ้นเล็กน้อยเมื่อใช้สารควบคู่ไซเลน Si-69 ภายใต้ระบบการคงรูปด้วยกัมมะถันและเปอร์ออกไซด์เนื่องจากกัมมะถันอิสระที่ปล่อยออกจากสารควบคู่ไซเลน Si-69 ระหว่างการคงรูป ในทางตรงกันข้าม สารควบคู่ไซเลนชนิดอื่นไม่ส่งผลต่อสมบัติเชิงกลของยางที่คงรูป เนื่องจากปริมาณที่น้อยของหมู่ไซลานอลบนผิวของอนุภาคเถ้าลอย

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LIST OF ABBREVIATIONS

CV	Conventional vulcanization
DCP	Dicumyl peroxide
EV	Efficient vulcanization
FA	Fly ash
FTIR	Fourier transform infrared spectroscopy
GPC	Gel permeation chromatography
IR or PI	Polyisoprene rubber or synthetic cis-1,4-polyisoprene
LOI	Loss of ignition
LVE	Linear viscoelastic region
MMFA	Mae Moh fly ash
MW	Molecular weight
NR	Natural rubber
PP	Parallel plate rheometer
PSi	Precipitated silica
RKFA	Rocktron fly ash
RPA	Rubber process analyzer
SEM	Scanning electron microscope
Semi-EV	Semi-efficient vulcanization
Si-264, TCPTS	3-thiocyanatopropyl triethoxy silane
Si-69 or TESPT	bis(3-triethoxysilylpropyl)tetrasulfide silane
VTEO	Vinyltriethoxysilane
XPS	X-ray photoelectron spectroscopy
XRD	X-ray diffractometer
XRF	X-ray fluorescence spectrometer

CHAPTER I

INTRODUCTION

Fillers are added to rubber for a variety of purposes, of which the most important are enhancements in reinforcement, processability and material costs [1]. Typically, fillers used in rubber industry are classified into reinforcing and non-reinforcing fillers. In some cases, fillers are divided into black and non-black types. There are basically two categories of reinforcing fillers - carbon black and mineral or 'white' fillers such as fumed and precipitated silica [2]. In rubber industry, due to its fine particle size (high specific surface area), the silica is widely used as non-black reinforcing filler to improve mechanical properties of vulcanizates, particularly hardness and resistances to tension, tear and abrasion. Although silica is still well accepted among rubber technologists, some other fillers from the natural resources as alternative reinforcing fillers in NR has been used to partly replace silica in rubber compounds [3]. Such fillers include clay, lignin, black rice husk ash (BRHA) and white rice husk ash (WRHA), and cellulose fiber.

Ash residues are wastes of coal-fired power plants; these include fly ash (FA) and bottom ash [4, 5]. The FA is a relatively inexpensive by-product, and its usages have the benefit of decreasing environmental problems. Furthermore, the FA has been used in industry as a consequence of such advantages as low cost, smooth spherical surface and good processability of the filled materials [6]. Generally, FA could function as either semi-reinforcing or non-reinforcing fillers in polymeric systems, depending on particle size, specific surface area and surface chemistry of FA particles. Moreover, in previous study [7-12], it has been reported that the FA particles consist of silicon dioxide similar to the silica, but with the lower content of silicon dioxide than silica. According to previous work, Sombatsompop [3] focused on the mechanical properties of FA filled NR and SBR by comparing the mechanical properties with precipitated silica. It was recommended that the FA particles could be used as filler in NR system at loading range of 0-30 phr of silica concentration, but not

in SBR system. Consequently, the addition of FA to rubber is able to enhance properties of rubber products in similar fashion to that of commercial silica, but with smaller magnitude. However, the presence of heavy metals in FA particles may play role on degradation process of rubber molecules to some extent. Such degradation process could effectively be suppressed by the addition of antioxidant.

It is well known that the FA is difficult to be dispersed in NR compounds in the same way as commercial silica, attributed to its poor interaction between the polar surface of FA and non-polar NR [13]. Thus, a surface treatment of FA particles is required to achieve good properties of vulcanizates. So far, the chemical treatment of FA surface has become the effective method to improve rubber-filler interaction, and to reduce filler-filler interaction (filler transient network). Among those chemical treatments, silane coupling agents are commonly used to improve the dispersion [14]. In previous study [15], it has been reported that the addition of 2.0 - 4.0 %wt Si-69 silane to NR/FA composites could offer optimum properties of the composites. Apart from the filler dispersion, vulcanization system is one of the important factors affecting the properties of rubber products. Generally, the vulcanization systems can be classified as sulfur and non-sulfur system, mainly peroxide vulcanization. The resultant FA/NR vulcanizates of each system give different mechanical and thermal aging properties.

Thus, the present work aims to further investigate the previous work [3, 15] by focusing on the viscoelastic behavior of NR/FA composites. The viscoelastic properties of NR/FA composites were studied by both steady and oscillatory shear rheometers. In addition, tensile properties and phase morphology were investigated and correlated to the viscoelastic behavior.

CHAPTER II

OBJECTIVES

The present research aims to study the viscoelastic properties of FA filled NR composites measured from oscillatory and steady shear rheometers. The scope of this work consists of three main parts, as follows:

2.1. Effect of fly ash loading

The objective of this section is to study the effect of FA loading on viscoelastic properties, aging properties and storage stability of FA filled NR compounds. Additionally, the influence of non-rubber component on such properties in NR compounds will be investigated.

2.2. Effect of silane coupling agent

The second part of this study is to investigate the effect of silane coupling agent (i.e., silane loading, treatment methods and types of silane) on viscoelastic properties of FA filled NR compounds. Three different silane used are Bis(3-triethoxysilylpropyl) tetrasulfide (Si-69 or TESPT), 3-thiocyanatopropyl triethoxy silane (Si-264 or TCPTS) and Vinyltriethoxysilane silane (VTEO).

2.3. Effect of curing systems

The last part aims to study the effects of curing systems (i.e., sulfur and peroxide) on the viscoelastic and mechanical properties of FA filled NR composites. For sulfur vulcanization, the properties caused by the use of conventional vulcanization (CV), efficient vulcanization (EV) and semi-efficient vulcanization (semi-EV) will be investigated.

CHAPTER IV

MATERIALS AND METHODS

4.1 Materials

All chemicals used in the present study are presented in Table 4.1.

Table 4.1 Compounding ingredients used in the present work

Compounding ingredients	Trade name/Grade	Manufacturer/Supplier
Natural rubber (NR)	STR 5L	Union Rubber Products Corp., Ltd., Thailand
Synthetic cis-1,4-polyisoprene (IR)	Nipol IR 2200	Nippon Zeon, Japan
Fly ash (FA)	As-received	Mae Moa power station of KNR group, Thailand
	MinTron	RockTron International Limited, UK
Precipitated silica (PSi)	Tokusil®233	Tokuyama Siam Silica Co. Ltd., Thailand
Spherical silica	SIDISTAR	Elkem Materials, Norway
Bis(3-triethoxysilylpropyl) tetrasulfide	Si-69 ,TESPT	JJ-Dequssa Co. Ltd., Thailand
3-thiocyanatopropyl triethoxy silane	Si-264, TCPTS	JJ-Dequssa Co. Ltd., Thailand
Vinyltriethoxysilane	VTEO	Evonilk Degussa GmbH Inorganic Material, Germany
Zinc oxide (ZnO)	Commercial grade	Chemmin Co. Ltd., Thailand

Table 4.1 Compounding ingredients used in the present work (cont.)

Compounding ingredients	Trade name/Grade	Manufacturer/ Supplier
Magnesium oxide (MgO)	Starmag #150	Konoshima Chemical Co. Ltd., Japan
Stearic acid	Commercial grade	Chemmin Co. Ltd., Thailand
N-(1, 3-dimethyl)-N'-phenyl-p-phenylenediamine (6-PPD)	Santogard 6-PPD	Flexsys Co. Ltd., Thailand
Sulfur	Commercial grade	Chemmin Co. Ltd., Thailand
N-tert-butyl-2-benzothiazolesulfenamide (TBBS)	Santocure TBBS	Flexsys Co. Ltd., Thailand
Toluene	Commercial grade	Labscan Co. Ltd., Thailand
Ethanol	Commercial grade	Labscan Co. Ltd., Thailand
Dicumyl peroxide (DCP) (98%)	Percumyl D	Chemmin Company, Ltd., Thailand
Copper (II) oxide (CuO)	Puriss	Fluka, Germany
Manganese (II) oxide (MnO)	99% purity	Aldrich, USA
Iron (III) oxide (Fe ₂ O ₃)	Puriss	Fluka Analytical, USA
Tetrahydrofuran (THF)	HPLC	Labscan Co. Ltd., Thailand

4.2 Instruments

The instruments used in the present study are summarized in Table 4.2.

Table 4.2 List of instruments used in the present study

Instrument	Model	Manufacture/Supplier
Laboratory 2-roll mill	LRM150	Labtech Engineering Company Ltd., Thailand
Rubber process analyzer	RPA 2000	Alpha Technology, USA
Capillary rheometer	Rheo-tester 2000	Goettfert, Germany
Parallel plate rheometer (PP)	MCR 500	Anton Paar, Germany
X-ray diffractometer (XRD)	D8 advance	BrukerAXS, Germany
Scanning electron microscope (SEM)	LEO 1455 VP	LEO Electron Microscopy Ltd., UK
	JSM-6500F	JEOL Ltd., Japan
Universal extensometer	5566	Instron Universal, USA
Electrical hydraulic hot press	G 30H	Wabash Genesis Series Hydraulic Press, USA
Particle size analyzer	Mastersizer 2000	Marvern Instruments, UK
Ultrapycnometer	Ultrapycnometer1000	Quantachrome, USA
Gel permeation chromatography (GPC)	CO-1560	JASCO, Japan
X-ray fluorescence spectrometer (XRF)	S4 Pioneer	BrukerAXS, Germany
Ball mill	-	Saturn Chemical Co. Ltd., Thailand
Fourier Transform Infrared spectroscopy (FTIR)	Spectrum One	Perkin Elmer, USA
X-ray photoelectron spectroscopy (XPS)	AXIS Ultra DLD	Kratos, UK

4.3 Characterization of Fly Ash (FA)

The FA was characterized using instruments as follows:

4.3.1 X-Ray Fluorescence Spectrometer (XRF)

In the present study, the FA is supplied by Mae Moa Power Station, Thailand. The XRF technique was used to determine the chemical composition of the FA. The FA was first prepared in a form of pressed powder pellets using a hydraulic press in aluminum cups, where the FA was put on top of boric acid. The Rh x-ray tube was used at accelerating voltages up to 55 kV and current values of up to 100 mA.

4.3.2 Particle Size Analyzer

Particle size of FA and its distribution were determined using a particle size analyzer in a dry dispersion mode. The FA was put in a chamber with sample preparation devices, dispersed in air at the given air pressure of 1 bar, and fed at the 40% of feeding rate into the measurement zone. The average particle size and its distribution of the FA were measured.

4.3.3 Density

FA density was determined by the gas displacement technique using ultrapycnometer. Helium gas (99.999 %) was used in micro cell at test temperature of 22.5 °C

4.3.4 X-Ray Diffractometer (XRD)

Typically, FA consists of silicon dioxide (SiO_2) divided into 2 phases, crystallized (or quartz) and amorphous phases. The XRD was used for investigating the phase of FA. Diffraction patterns were collected from the 2θ range of 15-30 degree, and the results of 2θ against counts per second were plotted.

4.3.5 Scanning Electron Microscope (SEM)

The SEM was used at an accelerating voltage of 10 kV of secondary electron imaging (SEI) to examine the morphology of FA particles. The FA powder was mounted on aluminum stub, and sputter-coated with thin layer of gold to avoid electrostatic charging during the examination.

4.3.6 Fourier Transform Infrared Spectroscopy (FTIR)

The FA was prepared in KBr disc. A mixture of FA and KBr was fused into a transparent disk using a hydraulic press. The absorbance was measured in the wavenumber of 4000 - 400 cm^{-1} for examining the functional groups on the FA surfaces.

4.3.7 X-ray Photoelectron Spectroscopy (XPS)

XPS was conducted to determine the chemical state of metal in FA particles. FA powder was pressed onto a small square of carbon tape. The instrument work function was calibrated to give a binding energy for the metal such as Cu, Fe and Mn. The XPS instrument presents data as counts versus binding energy.

4.4 Surface Treatment of Fly Ash

In this study, there were 3 silane treatment methods used for modifying the FA surfaces:

1. *In-situ surface treatment*: The FA was treated simultaneously during the mixing process of rubber compounds.
2. *Pre – surface treatment*: The FA was pre-treated by a silane solution before charging to the mixer. Silanes used in this study are bis(3-triethoxysilylpropyl) tetrasulfide (Si-69 or TESPT), 3-thiocyanatopropyl triethoxy silane (Si-264 or TCPTS) and vinyltriethoxysilane silane (VTEO). The silane solution was prepared with ethanol before wetting on the FA surfaces. For instance, for a silane content of 2.0 wt%, 2.0 g of the silane was mixed with 100 ml of ethanol, and then stirred for 30 min. The 100g of FA were then added into the solution with a further 15 min stirring to ensure a uniform distribution of the silane on the FA surfaces. The treated FA was then dried at

100°C for 12 h in an oven until a constant weight was reached. Treated FA was then charged to the mixer with the similar mixing procedure to that of the in-situ treatment technique.

3. *Ball milled with pre – surface treatment*: The FA was first ball-milled followed by the pre-treatment with silanes. The ball-milling time was varied from 0 to 12.5 hours. Particle size of milled FA was determined with the use of particle size analyzer with the procedure as mentioned earlier. Milled FA was treated and then charged to the mixer in a similar manner to the pre-treatment method as mentioned previously.

4.5 Preparation of Compounds

4.5.1 Preparation of FA Filled NR Compounds without Curatives

The formulations of FA filled NR compounds used for studying the effect of filler loading on properties of FA filled NR compounds are given in Table 4.3. It must be noted that the IR compounds were used for comparing the properties with NR in order to exclude a non-rubber effect. Additionally, the spherical silica (SIDISTAR) offering the ball-bearing effect was used to compare its properties with the FA as shown in Table 4.4.

Furthermore, the effect of antioxidant on properties of FA filled NR compounds was studied. The antioxidant used in this work was N-(1,3-dimethyl)-N'-phenyl-p-phenylenediamine (6-PPD). The formulation of the compounds is shown in Table 4.5.

In order to investigate the effect of heavy metals (Cu, Fe and Mn) on compound properties, both NR and IR were used for a comparison purpose. The formulation as shown in Table 4.6 was used to prepare the rubber compounds.

For the study of silane structure effect, three silane coupling agents namely, Si-69, Si-264 and VTEO were used with various loadings as presented in Table 4.7.

Table 4.3 Formulations of FA filled rubber compounds

Materials	Amount (phr)
NR*	100.0
FA	0, 50.0, 100.0, 150.0 and 200.0

* IR used for comparing the non-rubber effect

Table 4.4 Formulations of silica filled rubber compounds without curatives

Materials	Amount (phr)
NR	100.0
Silica (Precipitated silica and SIDISTAR)	equivalent to the total SiO ₂ content in FA

Table 4.5 Formulations of FA filled NR compounds used for the study of antioxidant effect

Materials	Amount (phr)
NR	100.0
FA	0, 50.0, 100.0, 150.0 and 200.0
6-PPD	4.0

Table 4.6 Formulations of FA filled rubber compounds used for the study of metals effect

Materials	Amount (phr)
IR	100.0
FA	0, 50.0, 100.0, 150.0 and 200.0
Metals (Cu, Fe and Mn)	2.0 and 8.0 ppm

Table 4.7 Formulations of FA filled rubber compounds used for the study of silane structure effect

Materials	Amount (phr)
NR	100.0
FA	100.0
Silane coupling agents (Si-69, Si-264 and VTEO)	0 – 8.0 *

* % w/w: only used for the amount of silane coupling agents loading

4.5.2 Preparation of FA Filled NR Compounds with Curatives

In the present study, the vulcanization systems, both sulfur and peroxide, of FA filled NR compounds with and without silane coupling agents were used. For the sulfur vulcanization systems, the NR compounds cured with conventional vulcanization (CV), efficient vulcanization (EV) and semi-efficient vulcanization (semi-EV) were prepared according to the formulations as given in Tables 4.8 and 4.9.

Table 4.8 Formulations of FA filled rubber compounds with sulfur curatives

Materials	CV (phr)	Semi EV (phr)	EV (phr)
NR	100.0	100.0	100.0
FA	0, 50.0, 100.0, 150.0 and 200.0	0, 50.0, 100.0, 150.0 and 200.0	0, 50.0, 100.0, 150.0 and 200.0
6-PPD	2.0	2.0	2.0
Stearic acid	2.0	2.0	2.0
ZnO	4.0	4.0	4.0
TBBS	0.75	1.80	2.25
Sulfur	2.25	1.20	0.75
Silane coupling agents (Si-69, Si-264 and VTEO)*	3.0*	3.0*	3.0*

* % w/w: only used for the compounds with silane coupling agents

Table 4.9 Formulations of FA filled rubber compounds with peroxide

Materials	Amount (phr)
NR	100.0
FA	0, 50.0, 100.0, 150.0 and 200.0
6-PPD	2.0
DCP	2.0
Silane coupling agents (Si-69, Si-264 and VTEO)	3.0*

* % w/w: only used for the compounds with silane coupling agent

4.6 Mixing Procedure

4.6.1 FA Filled NR Compounds without Curatives

The mixing procedure used for preparing compounds as given earlier in Tables 4.3 - 4.9 is presented in Table 4.10. In the initial step, a raw rubber was masticated for 1 minute on 2-roll mill at set temperature of 60 °C. Afterward, the filler, either FA or silica, was added to the masticated rubber. Finally, the compound was discharged at 13th minute to ensure good filler dispersion and distribution. The 6-PPD as antioxidant (if any) was premixed with FA prior to being charged onto 2-roll mill. For investigating the silane coupling agent and metal effects, the compounding chemicals were charged to the compounds as presented in Table 4.10.

4.6.2 FA Filled NR Compounds with Curatives

The mixing sequence for preparing the compounds is shown in Table 4.10. In the first step, raw rubber was masticated for 1 minute on 2-roll mill at set temperature of 60 °C. Afterward, the FA was charged to the masticated rubber. Finally, the curatives were added to the NR compounds and, the compound was discharged at 13th minute.

Table 4.10 Mixing procedure of rubber compounds with and without curatives

Step	Action	Time (min)
1	Load rubber	0
2	Add silane coupling agent (if any)	1
	Add metal (if any)	
3	Add FA (if any)	8
	Add curatives (if any)	
4	Discharge rubber compound	13

4.7 Determination of Viscoelastic Properties and Storage Stability

The properties of FA filled NR compounds were studied in the views of viscoelastic and mechanical properties. In addition, the storage stability of these compounds was studied with the storage duration of 12 weeks. Viscoelastic properties of compounds under shear flow were measured using the Rubber Process Analyzer (RPA2000), rate-controlled capillary rheometer and parallel-plate rheometer.

4.7.1 Rubber Process Analyzer (RPA2000)

Approximately 5 g of rubber compound was placed in the test chamber under conditions as mentioned in sections 4.7.1.1 to 4.7.1.4. The test temperature of 70 °C was used throughout the study. The storage modulus (G'), damping factor ($\tan \delta$) and complex viscosity (η^*) as functions of time, shear strain and frequency were recorded.

4.7.1.1 Time Sweep Test

Time sweep test was typically performed with the duration of 30 minutes at the angular frequency of 10 rad/s and 5 % strain to investigate a thermal stability of compounds to ensure that any change of both strain and frequency sweep tests is not caused by a thermal degradation.

4.7.1.2 Strain Sweep Test

For a strain sweep test, the linear viscoelastic (LVE) regions at 1 rad/s and 100 rad/s were determined to be used in a subsequent frequency sweep test. The strain was varied in the range of 0.5- 1200 % and 0.5 -30 % at 1 rad/s and 100 rad/s, respectively.

4.7.1.3 Frequency Sweep Test

In the frequency sweep test, the angular frequency of 1 to 100 rad/s was varied under a small strain within the LVE region.

4.7.1.4 Storage Stability and Aging Test

Storage stability of compounds was investigated using the RPA2000 with the test procedure as illustrated in Figure 4.1. At the beginning, the strain sweep test from 0.5-1200% strain at 70 °C was performed on specimens with various storage duration. Afterward, the compounds were conditioned in the RPA2000 test chamber at elevated temperature of 120 °C under a low strain of 0.28 % for 1 hour. Finally, the strain sweep test with similar test condition to the first stage was conducted. By this means, any alteration in viscoelastic properties attributed to thermal ageing could be monitored.

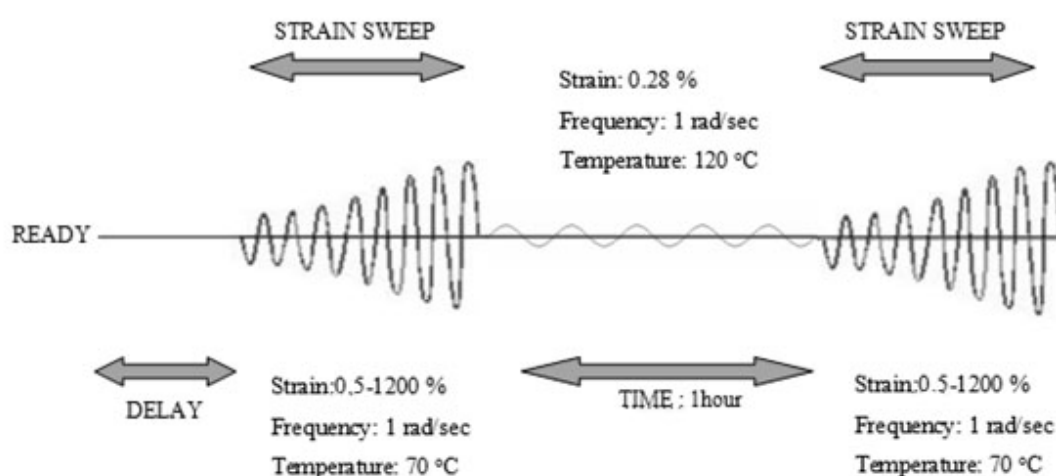


Figure 4.1 Schematic of aging study used in the present study

4.7.2 Capillary Rheometer

A rate-controlled capillary rheometer was utilized to measure rheological properties of compounds under steady shear flow at the test temperature of 70 °C. Shear rate was varied in the range of 10-1000 s⁻¹. A Bagley correction using 3 die lengths of 10, 20 and 30 mm with a given die diameter of 2 mm was carried out to eliminate the entrance effect. The small pieces of compounds were packed in the barrel and preheated for 5 minutes. Apparent shear stress and apparent viscosity were recorded as a function of apparent shear rate.

Apparent shear stress, apparent shear rate, and apparent viscosity were calculated from Equations 4.1, 4.2 and 4.3, respectively.

$$\tau_{app} = \frac{\Delta PR}{2L} \quad (4.1)$$

where τ_{app} : apparent shear stress (Pa)

ΔP : pressure drop (Pa)

R : radius of die (m)

L : length of die (m)

$$\dot{\gamma}_{app} = \frac{4Q}{\pi R^3} \quad (4.2)$$

where $\dot{\gamma}_{app}$: apparent shear rate (s⁻¹)

Q : volumetric flow rate (m³/s)

$$\eta_{app} = \tau_{app} / \dot{\gamma}_{app} \quad (4.3)$$

where η_{app} : apparent shear viscosity (Pa.s)

To eliminate the elastic and non-parabola flow pattern effects, the Bagley and Rabinowitsch corrections were performed. The corrected values of shear stress, shear rate and shear viscosity were calculated from Equations 4.4 to 4.6.

$$\tau_{corr} = \frac{\Delta PR}{2(L + eR)} \quad (4.4)$$

where τ_{corr} : corrected shear stress (Pa)

e : effective length as determined from Bagley plot

$$\dot{\gamma}_{corr} = \frac{4Q}{\pi R^3} \left[\frac{3n+1}{4n} \right] \quad (4.5)$$

where $\dot{\gamma}_{corr}$: corrected shear rate (s^{-1})

n : Power law index, as determined from the slope of a logarithmic plot of apparent shear stress vs. apparent shear rate

$$\eta_{corr} = \tau_{corr} / \dot{\gamma}_{corr} \quad (4.6)$$

where η_{corr} : corrected shear viscosity (Pa.s)

In practice, the Cox-Merz concept [112] as shown in Equation 4.7 was employed for linking rheological data collected in steady shear experiment to those in oscillatory shear experiment for uncured compounds. The complex viscosity (η^*) measured from the oscillatory shear flow was superimposed with the apparent shear viscosity (η_{app}) from the capillary results. Moreover, to eliminate the result of elastic behavior affecting rheological properties, the relationship between oscillatory and steady shear flow was drawn in terms of dynamic viscosity (η') and true shear viscosity (η_{true}).

$$\eta_{app}(\dot{\gamma}) = \eta^*(\omega) \Big|_{\omega=\dot{\gamma}} \quad (4.7)$$

where η_{app} : apparent or uncorrected viscosity at steady shear rate of $\dot{\gamma}$ (s^{-1}) in capillary rheometer
 η^* : dynamic complex viscosity at an oscillary frequency of ω (rad/s)

4.8 Determination of Molecular Weight

4.8.1 Gel Permeation Chromatography (GPC)

The molecular weights of masticated NR and IR were analyzed by GPC technique. A commercial standard polyisoprene (polymer standard service GmbH, Germany) was used as a standard for a calibration curve. The rubber samples were dissolved in tetrahydrofuran (THF) at concentration of 0.1 % w/v under stirring overnight. Afterward, the rubber solutions were filtered with filter paper and 0.45-micron nylon membrane (SARTORIOUS AG, Germany). The obtained rubber solutions were injected to GPC using THF as an eluent under the flow rate of 0.5 ml/min.

4.8.2 Parallel Plate Rheometer (PP) and RPA2000

Molecular weight (MW) of the extracted NR and IR was determined by the parallel plate rheometer. It is known that the polymer viscosity is affected strongly by MW, especially at low shear rate. Such shear viscosity at the shear rate approaching zero is known as the zero shear viscosity (η_0). Basically, the relationship between η_0 and MW is expressed as shown in Equation 4.8 [113-116].

$$\eta_0 = kM^a \quad (4.8)$$

where k is a consistency constant, a is the value of the exponent (approximately 1.0 for low MW polymers, and 3.4 as the MW increases beyond a critical value, M_c).

In this work, the rubbers filled with FA compounds were extracted by toluene and then centrifuged for a sedimentation of FA particles. Afterward, the NR (or IR) solution was separated by filtering with filter paper, and allowed for drying at room temperature. The η_0 of the extracted rubber films was measured using the parallel plate rheometer with the frequency sweep tests in the range of 0.01 - 100 rad/s at 70 °C with a 25-mm grooved rotor (to prevent slippage between polymer melt and rotor surface) under a small strain within the linear viscoelastic (LVE) region whereas the RPA2000 with the frequency sweep tests in the range of 0.5 - 160 rad/s at the similar temperature. The MW of rubber both RPA2000 and PP were calculated using Equation 4.8.

4.9 Determination of Bound Rubber Content

Bound rubber content was determined by extracting rubber compounds with toluene. Approximately 1 g of compounds was immersed in 50 ml toluene for 7 days at room temperature, and the toluene was renewed every 3 days. After filtering, the insoluble portion was dried for 1 day in air at room temperature and in an oven at 70°C until a constant weight was obtained. The percentage of bound rubber content was calculated as expressed in Equation 4.9.

$$R_b = \frac{w_{fg} - w \left[m_f / (m_f + m_p) \right]}{w \left[m_p / (m_f + m_p) \right]} \times 100 \quad (4.9)$$

where R_b = bound rubber content (%)

w_{fg} = weight of FA and gel (g)

m_f = weight of FA in the compound (g)

m_p = weight of rubber in the compound (g)

w = weight of specimen (g)

4.10 Determination of Cure Characteristics

Cure characteristics of rubber compounds were determined by the RPA2000 at the test temperature of 155 °C with frequency of 6.28 rad/s and % strain deformation of 15 %. Approximately 5 g of uncured rubber was placed on the disk of the rheometer cavity under oscillating arc of 1°. The torque required to oscillate the disk was recorded as a function of time. The scorch time (ts_2) and optimum cure time (tc_{90}) are calculated from the time in which the rheometer torque reaches 2 units from the minimum torque (M_L) and 90% of the maximum torque (M_{HF}) of a cure curve, respectively. The typical of cure curve is shown in Figure 4.2.

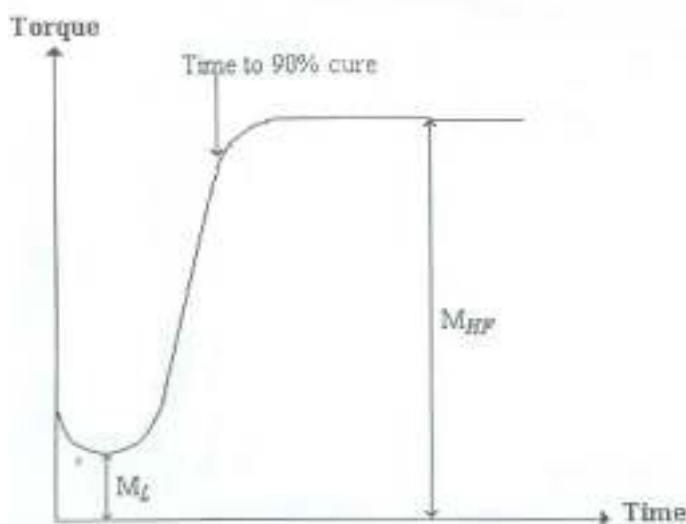


Figure 4.2 Typical of cure curve of rubber compound

4.11 Preparation of Rubber Vulcanizates

Rubber vulcanizate sheets with 1 mm in thickness were prepared by compression molding at 155°C under pressure of 15 MPa. Cure time used was tc_{90} as pre-determined from the cure curve.

4.12 Determination of Crosslink Density

The rubber vulcanizates were cut into square shape and weigh approximately 0.5-0.7 g before immersing it into toluene. The sample was kept in a dark place for 7 days. The swollen sample was removed from the toluene and the excessive toluene was blotted with a paper towel. Then, the swollen sample was placed in clean weighing bottle and weighed accurately. At least 3 samples for each vulcanizates were tested and the average values were reported. Crosslink density was calculated by Flory–Rehner equation as expressed in Equation 4.10 [117].

$$-\ln(1-V_r) - V_r \chi V_r^2 = 2V_s \eta_{\text{swell}} (V_r^{1/3} - 2V_r/f) \quad (4.10)$$

where V_r = the volume fraction of rubber in swollen gel

V_s = the molar volume fraction of toluene (106.2)

χ = the rubber–solvent interaction parameter (0.393)

η_{swell} = the crosslink density of rubber (mol cm^{-3})

f = the functionality of crosslinks (four for sulfur curing system)

4.13 Determination of Tensile Properties

Tensile properties of rubber vulcanizates were measured according to ASTM D412-98. The 1 mm thick vulcanized sheets were cut using the ASTM type C die into the dumbbell shape specimens having dimension as shown in Figure 4.3. The specimen was gripped properly to achieve uniform tension distribution over the cross section. The machine was started and the distance between the extensometer was measured continuously. Crosshead speed and load cell used were 500 mm/min and 1 kN, respectively. The modulus at 100% elongation (M100), tensile strength and elongation at break (EB) were determined. At least 5 specimens were tested, and the average values were reported.

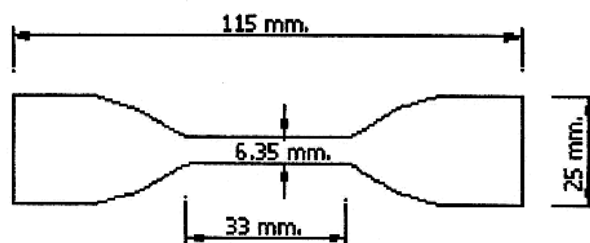


Figure 4.3 Dimensions of the tensile specimen

The tensile properties were calculated as shown in Equations 4.11 to 4.13:

$$M100 = \text{stress at 100\% elongation} \quad (4.11)$$

$$\text{Tensile strength} = \text{the stress at rupture} = F/A \quad (4.12)$$

$$\% \text{ EB} = 100 [L - L_0] / L_0 \quad (4.13)$$

where F = measured force (N)

A = cross-sectional area of unstrained specimen (m^2)

L = observed distance between grips of the extended specimen (m)

L_0 = original distance between the extensometer (m)

4.14 Morphological Observation

Samples were placed in liquid nitrogen for 2 minutes, and then fractured into two pieces to create fresh fractured surfaces. The fracture ends of rubber specimens were subsequently put in sample box with silica gel to prevent the moisture adsorption, then mounted on aluminum stub and sputter-coated with thin layer of gold to avoid electrostatic charging during the SEM examination.

The SEM observation was performed at an accelerating voltage of 10 kV to examine the morphology of FA/NR composites and FA dispersion in NR.

CHAPTER V

RESULTS AND DISCUSSION

The experiments in this research are divided into 3 main parts. The first part focuses on the effect of FA loading on the viscoelastic properties and storage stability of NR filled with FA. The second part aims to investigate effect of silane coupling agent on the viscoelastic properties of FA filled NR compounds. Finally, the effect of curing system (i.e., sulfur and peroxide) on the viscoelastic properties and mechanical properties of FA filled NR composites is studied.

5.1 Characterizations of FA

In the present study, the FA is supplied by the Mae Moa Power Station, Thailand. Before use of the FA in this work, it is necessary to characterize overall characteristics of FA including particle size and its distribution as well as chemical composition.

5.1.1 Chemical Compositions of FA

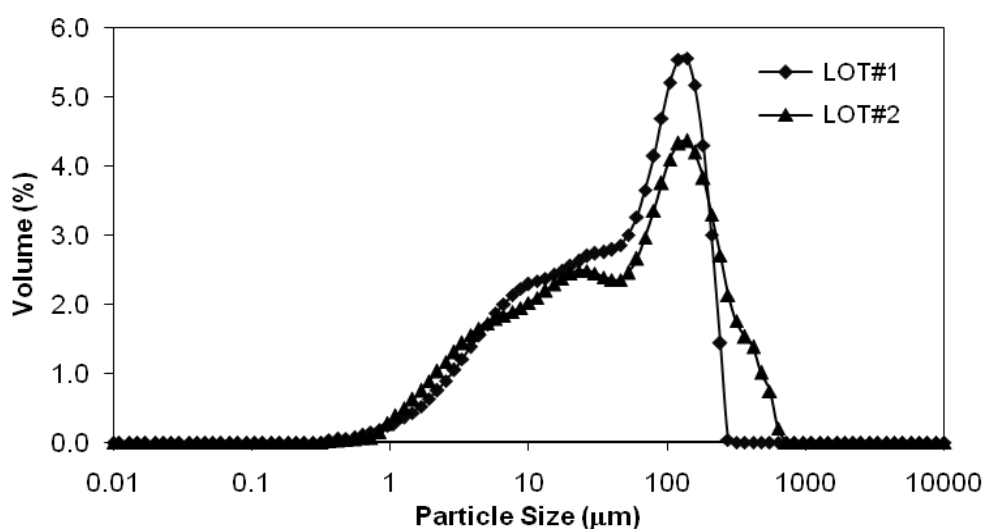
The chemical composition of FA as determined from the X-ray fluorescence spectrometer (XRF) is shown in Table 5.1. It is evident that the largest amount of composition in FA is silicate (SiO_2) or silica in the range of 30.0-38.0 % w/w.

5.1.2 FA Particle Size and Its Distribution

Particle size analyzer was used to measure the size and distribution of FA particles. The particle size distribution of FA used is rather broad with the sizes ranging from submicron to 200 microns as illustrated in Figure 5.1. The reported mean particle size of FA is of 50.311 (lot#1) and 47.858 (lot#2) microns.

Table 5.1 Chemical compositions of FA used in the present study

Compositions	Content (%)		Compositions	Content (%)	
	Lot#1	Lot#2		Lot#1	Lot#2
SiO ₂ (silica)	37.6	30.83	BaO	0.130	0.17
Al ₂ O ₃	22.4	14.28	SrO	0.123	0.04
CaO	15.1	23.31	MnO	0.101	0.17
Fe ₂ O ₃	14.5	15.68	ZnO	0.0301	0.03
K ₂ O	2.97	2.90	ZrO ₂	0.0297	0.02
SO ₃	2.71	6.32	Cr ₂ O ₃	0.0240	0.01
MgO	2.29	2.47	Rb ₂ O	0.0193	0.01
Na ₂ O	1.33	2.46	CuO	0.0187	0.01
TiO ₂	0.447	0.43	NiO	0.0121	0.01
P ₂ O ₅	0.171	0.34	Y ₂ O ₃	0.00431	0.01

**Figure 5.1** Particle size distribution of FA used in the present study

5.1.3 FA Density

The FA density used in the present study was determined from Ultrapycometer, and the results reveal the reported average density of $2.4244 \pm 0.0023 \text{ g/cm}^3$

5.1.4 Morphology of FA

Figure 5.2 depicts SEM micrographs of FA particles used in the present study. It is clear that the FA particles are spherical, encapsulating many sub-spheres. The geometry of FA particles plays strong roles on viscoelastic behavior of FA filled compounds. Full discussion on this point will be given in the subsequent sections.

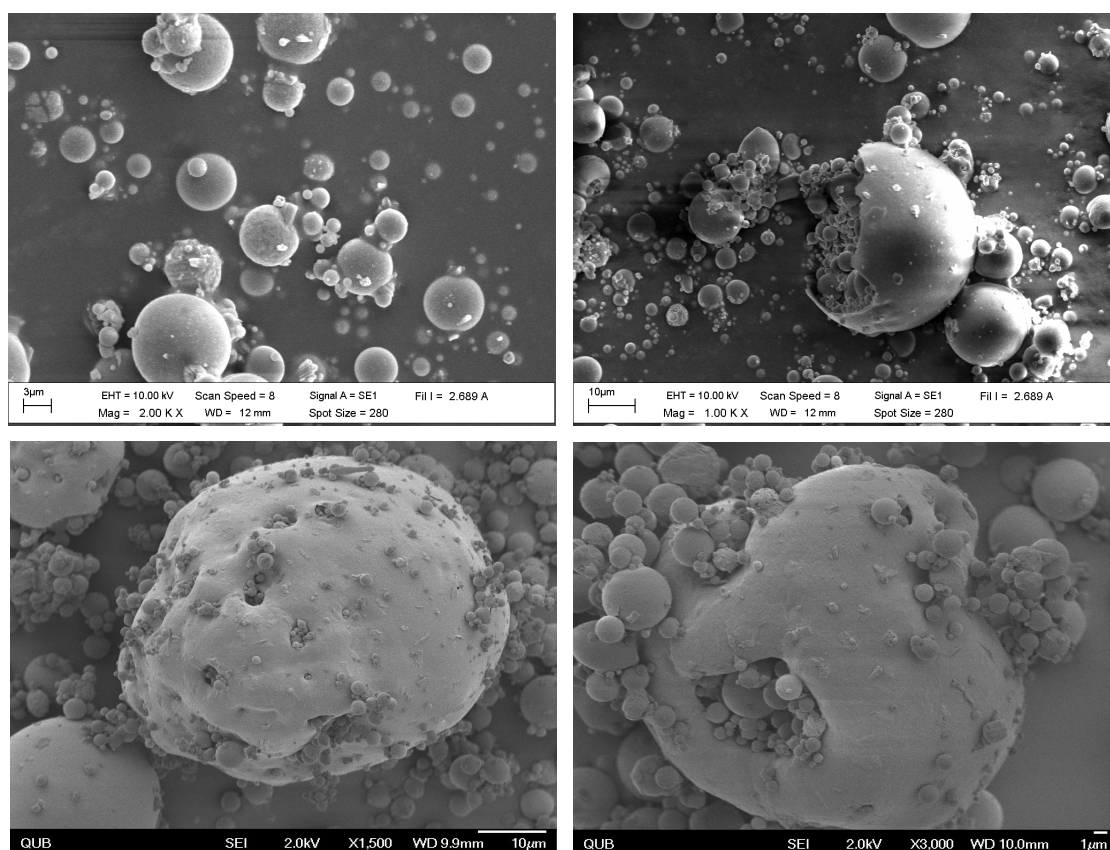


Figure 5.2 SEM micrographs of FA used in the present study

5.1.5 X-Ray Diffractometry (XRD)

By the utilization of XRD technique, the identification of mineralogical constituents and phase proportions of FA could be determined as illustrated in Figure 5.3. Clearly, the commercial silica (Hisil 233) shows amorphous structure whereas the FA consists mainly of crystalline phases as evidenced by the relative small intensity magnitude of peak at 2 Theta of about 26, similar to the quartz as highly crystalline structure [117, 118]. In addition, the XRD results of FA used in the present work are different from those in some works reporting the presence of crystalline (mullite and hematite) in FA [119, 120]. This is not unusual since it is known that the composition of FA depends strongly on the source and processing of FA.

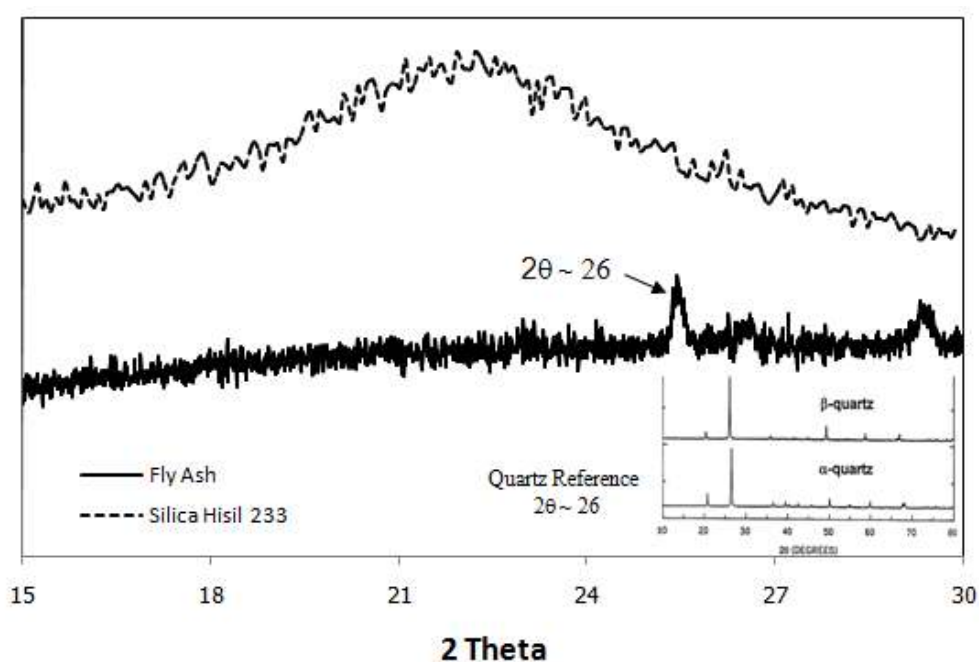


Figure 5.3 XRD results of FA used in the present study, precipitated silica and quartz [122]

5.1.6 Fourier Transform Infrared Spectroscopy (FTIR)

FTIR was employed for examining the functional groups on the FA surfaces as shown in Figure 5.4. The results show that both FA and commercial silica (Hisil 233) surfaces compose of similar functional groups, i.e., silanol and siloxane groups corresponding to wave numbers of approximately $3,400\text{ cm}^{-1}$ (O-H stretching) and $1,100\text{ cm}^{-1}$ (Si-O-Si asymmetric stretching), respectively [121, 122]. Obviously, FA exhibits smaller amount of silanol groups than silica supporting the FA crystalline structure as evidenced by the XRD results (see Figure 5.3). However, the peak approximately $3,400\text{ cm}^{-1}$ might be interfered via just a little moisture (from KBr) supported at the small peak at wave numbers of approximately $1,600\text{--}1,650\text{ cm}^{-1}$ referred to the deformation mode of absorbed molecular water due to O-H stretching [123].

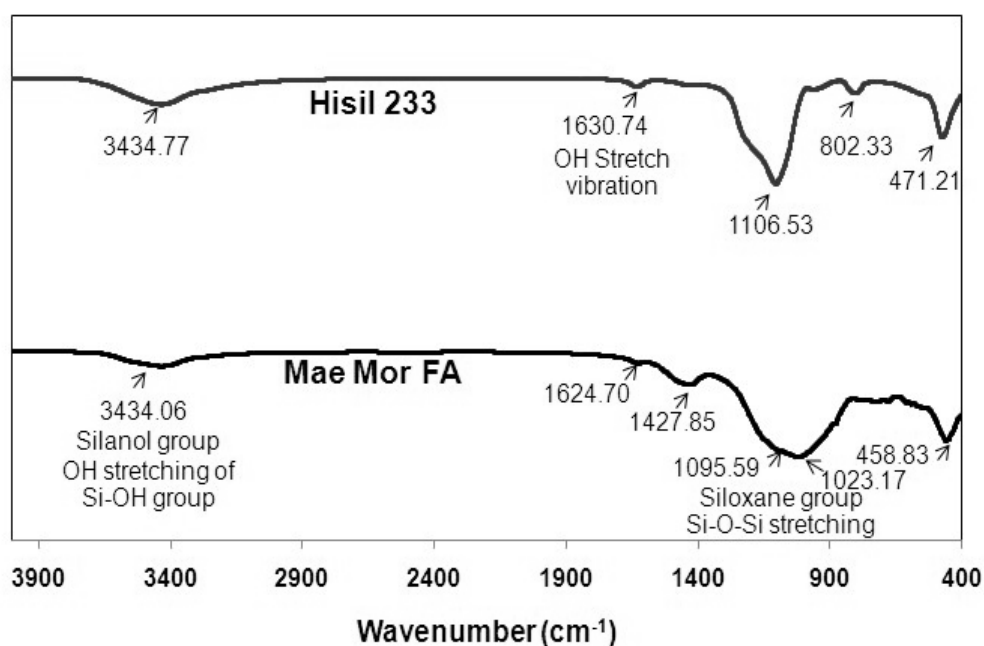


Figure 5.4 FTIR results of FA used in the present study

5.1.7 X-ray Photoelectron Spectroscopy (XPS)

XPS is a quantitative spectroscopic technique capable of measuring the chemical state of the elements existing within a material. Figure 5.5 demonstrates the XPS analysis results of FA particles, where there are strong peaks of iron (Fe) at binding energies of 710.7 and 724.7 eV. The binding energy of Fe 2p in the range of 710.1-711.0 eV corresponds to Fe (III) [124-126].

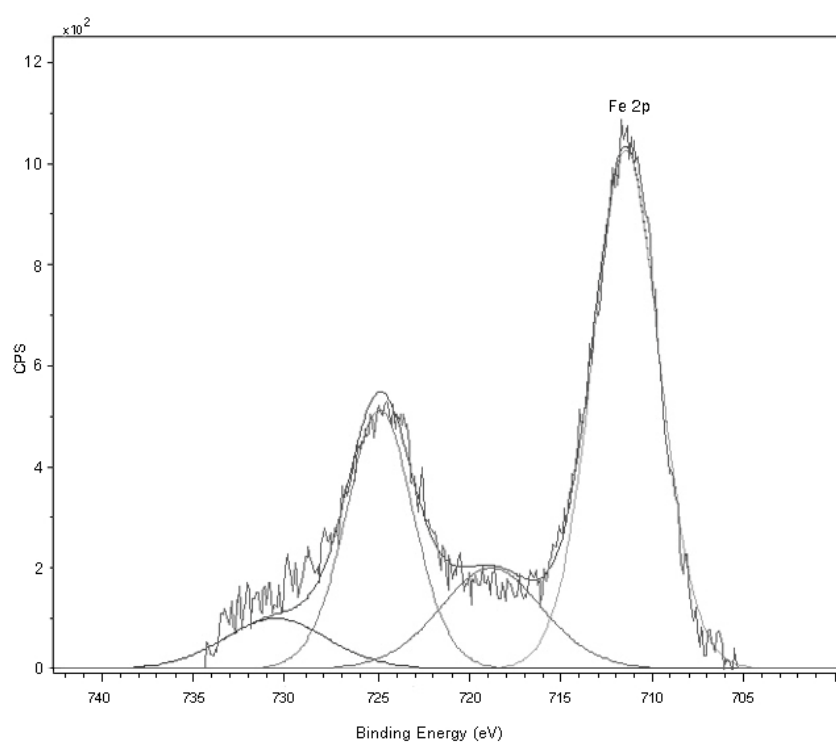


Figure 5.5 XPS results of FA used in the present study

5.2 Effect of FA Loading on Viscoelastic Properties, Storage Stability and Thermal Aging Properties of FA Filled NR Compounds

5.2.1 Viscoelastic Behavior

Viscoelastic behavior of compounds under shear flow were measured using both oscillatory and steady shear rheometers, i.e., Rubber Process Analyzer (RPA2000), parallel-plate rheometer, and rate-controlled capillary rheometer with 3 different die lengths over diameter (L/D) ratios of 10/2, 20/2 and 30/2.

5.2.1.1 Oscillatory Shear Rheometers

A) Time Sweep Test

A time sweep test was firstly performed in order to determine the thermal stability of a test specimen within the test duration of 30 minutes. Evidently from Figure 5.6, there is no significant change in storage modulus (G') of all compounds with increasing test duration up to 30 minutes. This implies that all FA filled NR compounds possess good thermal stability within the test duration to be used for the subsequent strain and frequency sweep tests. In other words, any change observed in those tests would not be caused by the thermal degradation occurring during the test period. Furthermore, it is obvious that the G' increases obviously with the increase in FA loading, which could be due to the hydrodynamic effect and/or filler network formation [127-130].

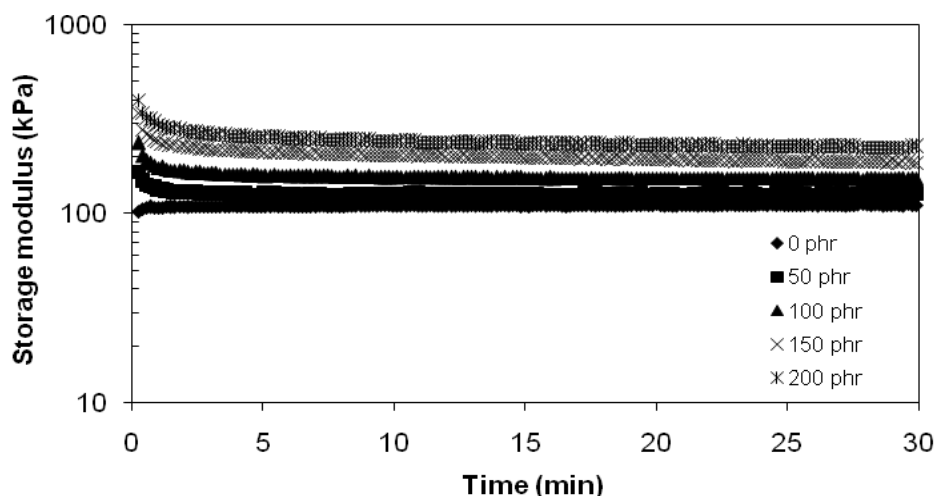


Figure 5.6 Storage modulus (G') as a function of time of FA /NR composites with various FA loadings

B) Strain Sweep Test

To further investigate the viscoelastic responses of FA filled compounds, the results of strain sweep test was performed, and the results obtained, i.e., G' as a function of strain, of FA/NR compounds with various FA loadings at 1 and 100 rad/s are exhibited in Figure 5.7. It can be seen that the strain sweep test measured at 1 and 100 rad/s gives similar result trends. Clearly, G' increases with increasing FA loading similar to the results of time sweep test as shown earlier in Figure 5.6. At high strain, the G' remarkably reduces with strain of deformation. Also, the linear viscoelastic (LVE) region (or the strain-independent region) depends strongly on FA loading. The higher the FA loading, the narrower the LVE region. The observed strain-dependent G' is not unusual as it has been reported previously [127] that the G' of highly silica filled compounds decrease substantially under a large deformation strain, caused mainly by the disruption of filler network associated with a molecular slippage (particularly in the case of uncured compounds). Apart from the broadness of LVE region, the discrepancy in G' at low and high strains (or usually known as the Payne effect) as shown in Figure 5.8, is found to be more apparent with increasing filler loading, which supports clearly the greater magnitude of filler network formation [127-131]. In other words, the strong interaction via H-bond between silanol groups of

silica in FA would cause a formation of filler transient network and thus narrowed the LVE region.

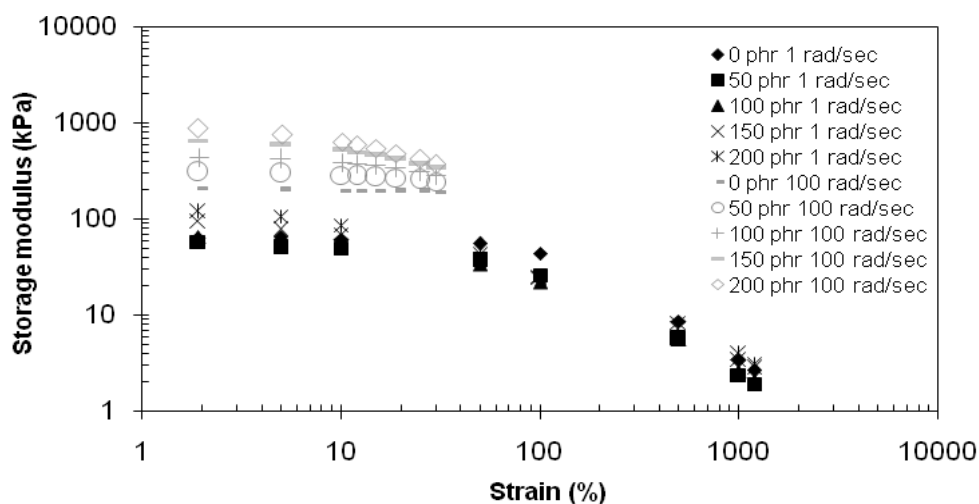


Figure 5.7 Storage modulus (G') as a function of strain amplitude of FA/NR composites with various FA loadings at 1 and 100 rad/sec

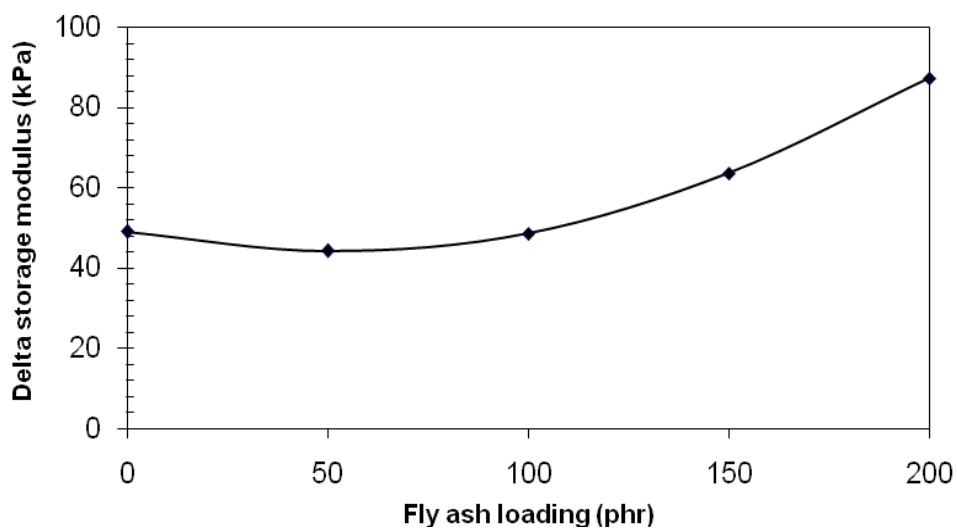


Figure 5.8 Effect of fly ash loadings of FA/NR compounds on the Payne effect

However, in the FA filled system considered in this study, it must be noted that the magnitude of filler network effect might not be as strong as that found in silica filled system reported previously [127-130] because of the relatively low content of silanol group and surface area of FA particles. This means the change in G' with FA loading is dominated by the hydrodynamic over the filler network effects. To clarify the proposed hypothesis, the Guth-Gold expression as illustrated in Equation 5.1 is used to generate the theoretical results of relative modulus with the exclusion of filler network effect as shown in Figure 5.9 [129,132].

$$\text{Relative } G' = \frac{G'_{\text{filled}}}{G'_{\text{unfilled}}} = 1 + 2.5\phi + 14.4\phi^2 \quad (5.1)$$

where G'_{unfilled} and G'_{filled} are elastic moduli of unfilled and filled compounds, respectively. The ϕ is the filler volume fraction.

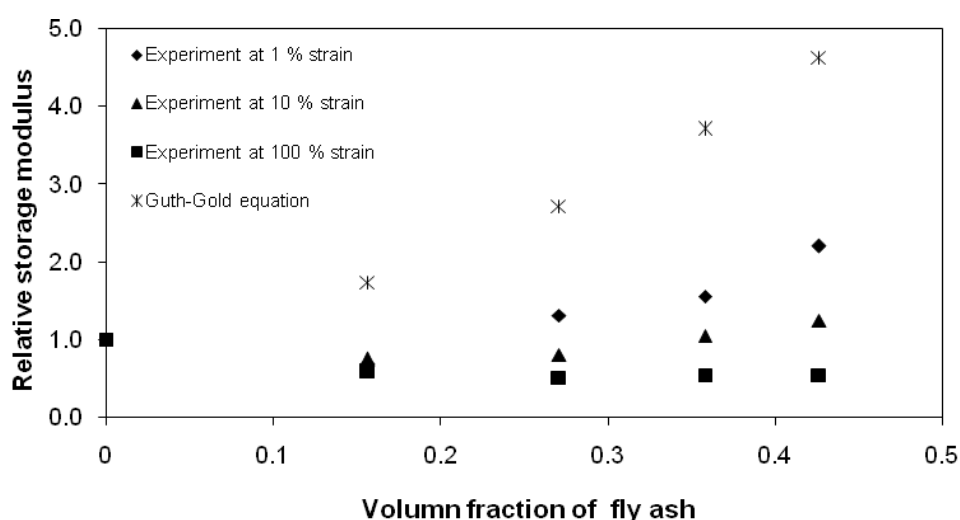


Figure 5.9 Relative storage modulus (relative G') at 1, 10 and 100% strain of NR compounds filled with various FA loadings

In theory, the filled compounds would show the positive deviation relative modulus from the Guth-Gold results, especially, at high filler loading due to the filler network formation as found in silica filled rubber system [129]. Unexpectedly, the experimental data found in this study show the negative deviation of relative modulus from the calculated Guth-Gold data, which is more pronounced at high deformation strain. In general, when strain is applied to the filled rubber compound, the rigid filler in the rubber matrix does not share the deformation of compound. The inclusion of the undeformable filler in the soft rubber matrix causes the local strain in matrix to go beyond the macroscopic strain [133]. Thus, the rubber chains in the highly filled compounds receive greater strain, which leads to the reduction in relative storage modulus via a molecular slippage and/or a disruption of FA-NR interaction. In addition, referred to the spherical shape of FA as shown previously in Figure 5.2, the molecular slippage would be further promoted via a ball-bearing effect.

To support an explanation of the ball bearing effect, the spherically-shaped silica, namely, SIDISTAR as shown in Figure 5.10 was used. It can be noticed from Figure 5.11 that the relative modulus at 1 % strain of NR filled with SIDISTAR is lower than the calculated relative modulus from Guth -Gold equation. This is attributed to the shape of SIDISTAR. The silica SIDISTAR promoted the molecular slippage via the ball-bearing similar to the FA particles.

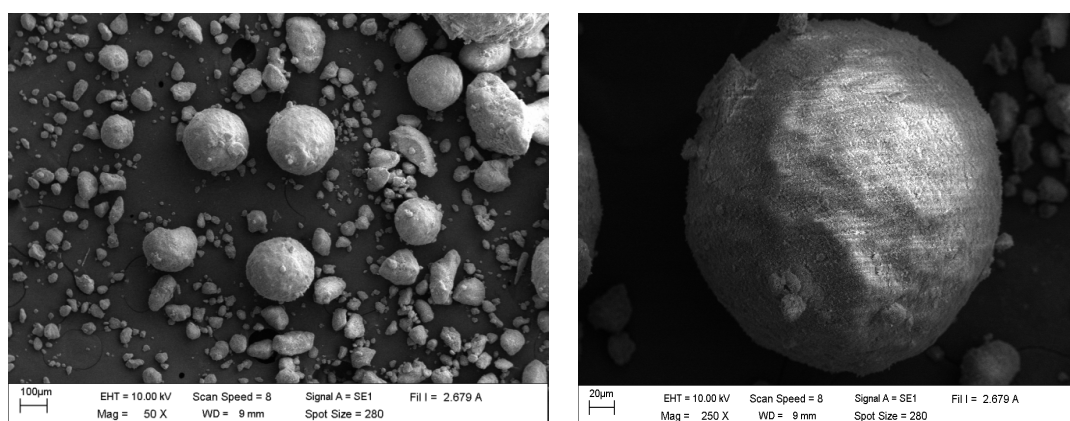


Figure 5.10 SEM micrograph of silica SIDISTAR particles used in the present study

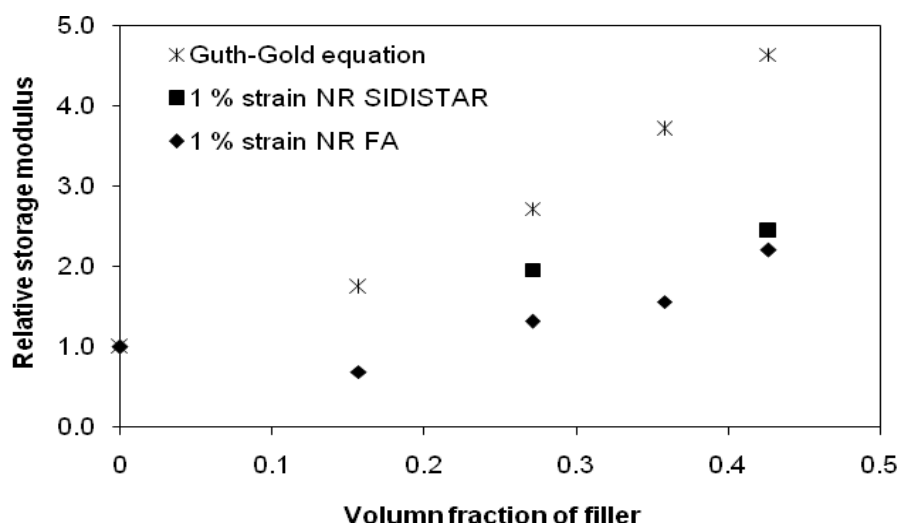


Figure 5.11 Relative storage modulus (relative G') at 1% strain of NR compounds filled with FA and silica SIDISTAR

C) Frequency Sweep Test

Figure 5.12 shows the influence of frequency on the storage modulus (G') of FA/NR compounds with various FA loadings. It is evident that all compounds show an increase in G' or elasticity with increasing frequency which is attributed to the insufficient time for a molecular relaxation at high frequency. Additionally, the magnitude of frequency-dependent responses (as determined from slopes) clearly increases with increasing FA loading. The results unexpectedly imply a decrease in elastic behavior of compounds with increasing FA loading, which is on the contrary to CPE/NR blend filled with precipitated silica (at similar amount of SiO_2 in FA) as shown in Figure 5.13. It has been reported in those work that the strong interaction between polar group of polymer matrix and silanol group of silica could form a tridimensional transient network resulting in a strong elastic response with lowered magnitude of frequency dependent behavior [127, 128]. The unexpected results might be the absence of filler network or the filler network effect might be overridden by some other factors including the ball bearing effect as mentioned earlier.

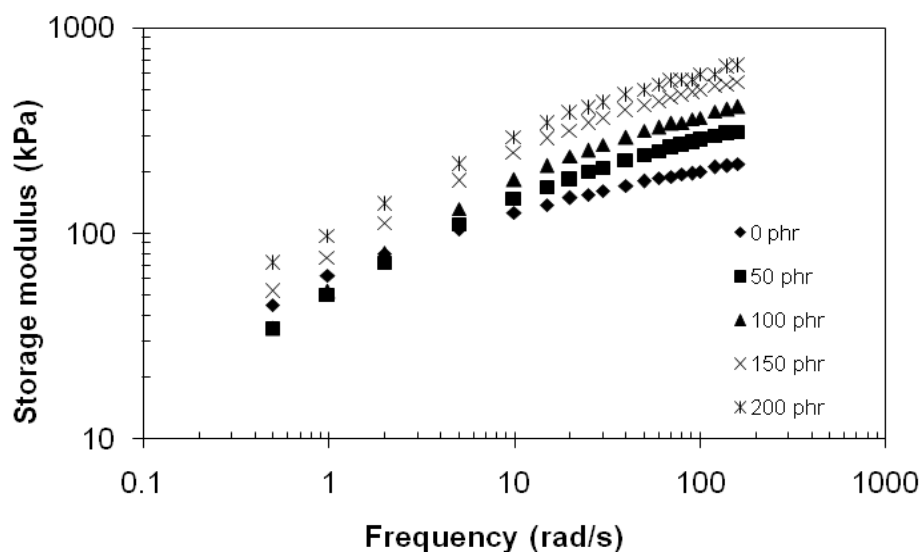


Figure 5.12 Storage modulus (G') as a function of frequency of NR compounds filled with various FA loadings

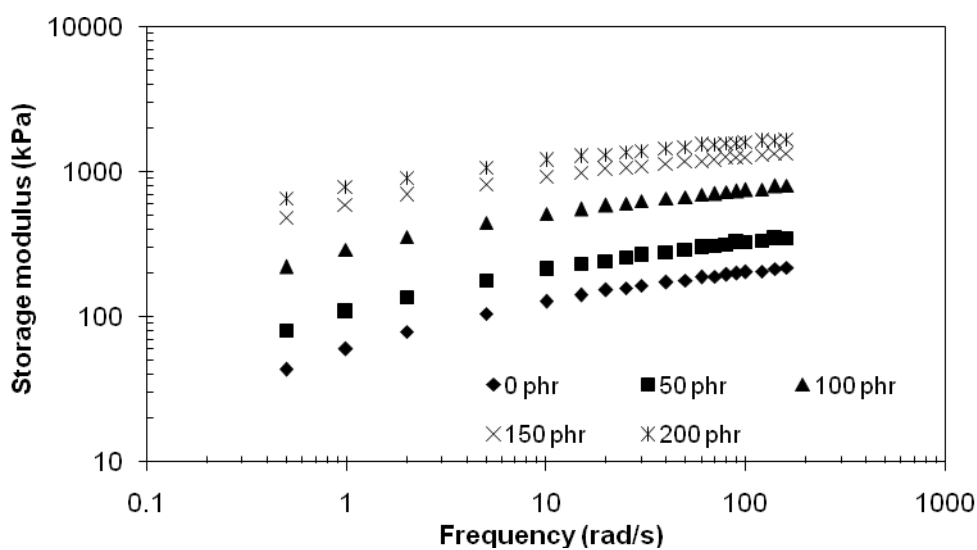


Figure 5.13 Storage modulus (G') as a function of frequency of NR compounds filled with various precipitated silica loadings (equivalent to the total SiO_2 content in FA)

Damping properties of NR compounds with various FA loadings are exhibited in Figure 5.14. Obviously, the reduction in damping factor with increasing frequency could be observed in all compounds indicating a rise in elastic behavior caused by the insufficient time for the molecular relaxation process as discussed in G' results in Figure 5.12. Similar to the case of G' , there is the unexpected increase in damping factor with increasing FA loading. It can be notice that the result of NR filled with FA differs from that of synthetic isoprene rubber (IR) which is free from non-rubber substances as depicted in Figure 5.15. It can be seen that, with increasing FA loading, the increase in damping factor is still noticeable, but the magnitude of discrepancy in damping factor of compounds with different FA loading is much less apparent than that of NR compounds (see Fig. 5.14). This implies the lower viscous response in IR compounds than NR compounds. Thus, it is suggested that the non-rubber substances presented in NR play role in such increase in magnitude of viscous behavior to some extent. By contrast, the result of NR filled with precipitated silica as evidenced in Figure 5.16, there is a decrease in damping of compounds with increasing silica loading via the filler network formation. The unexpected difference in result trends of NR filled with FA from that with silica is believed to be the results of a viscous response promotion given by (i) molecular slippage via the FA ball-bearing effect and (ii) molecular degradation of NR matrix which might be accelerated by the presence of complex substances (fatty acid salts of metals) which are formed via fatty acid in non-rubber component and heavy metals such as iron and copper in FA as illustrated in Table 5.1 [134-136].

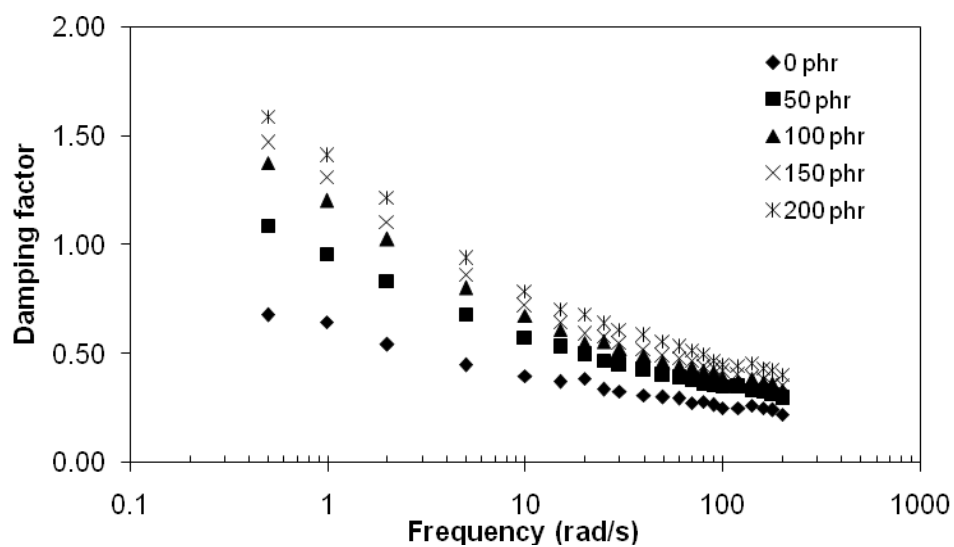


Figure 5.14 Damping factor (tan delta) as a function of frequency of NR compounds filled with various FA loadings

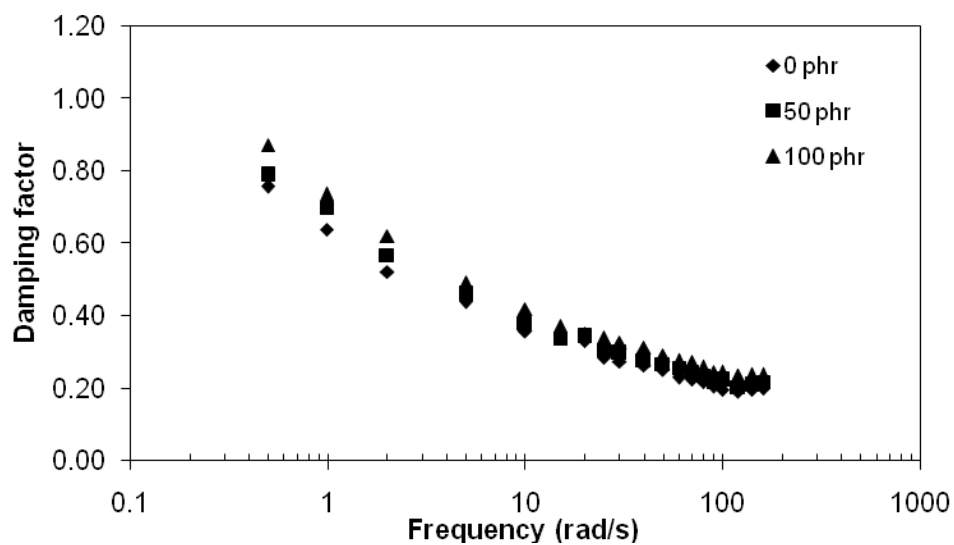


Figure 5.15 Damping factor (tan delta) as a function of frequency of IR compounds filled with various FA loadings

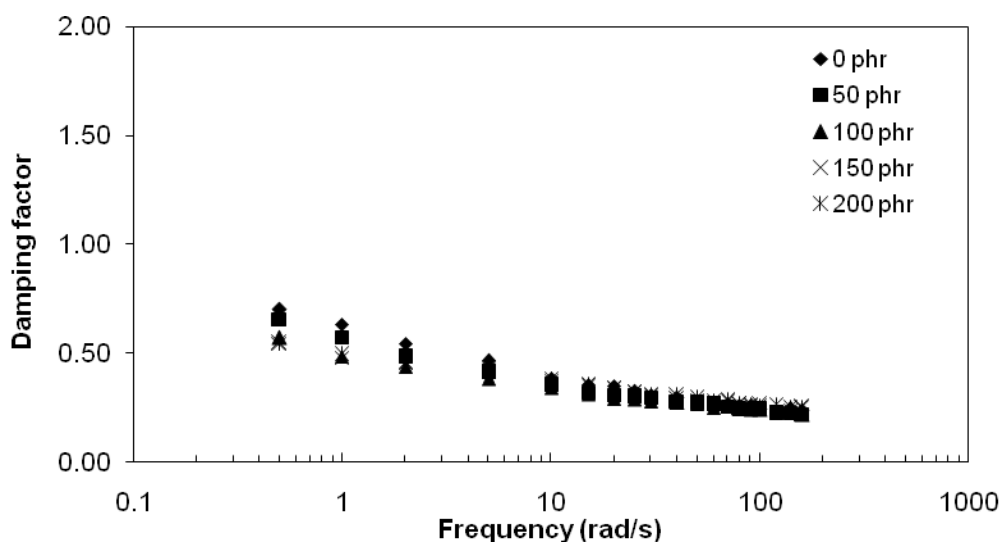


Figure 5.16 Damping factor (tan delta) as a function of frequency of NR compounds filled with various precipitated silica loadings (equivalent to the total SiO₂ content in FA)

Figure 5.17 reveals the processability of the NR compounds with various FA loadings via a complex viscosity as a function of frequency. As expected, the complex viscosity evidently decreases with increasing frequency, indicating the pseudoplastic behavior. Additionally, the increase in complex viscosity with increasing FA loading could be observed. Referred to the results of damping properties shown in Figure 5.14, the highly filled compounds reveal high damping factor particularly at low frequency that means the increase in complex viscosity is caused by the increase in magnitude of viscous response via molecular slippage as discussed earlier.

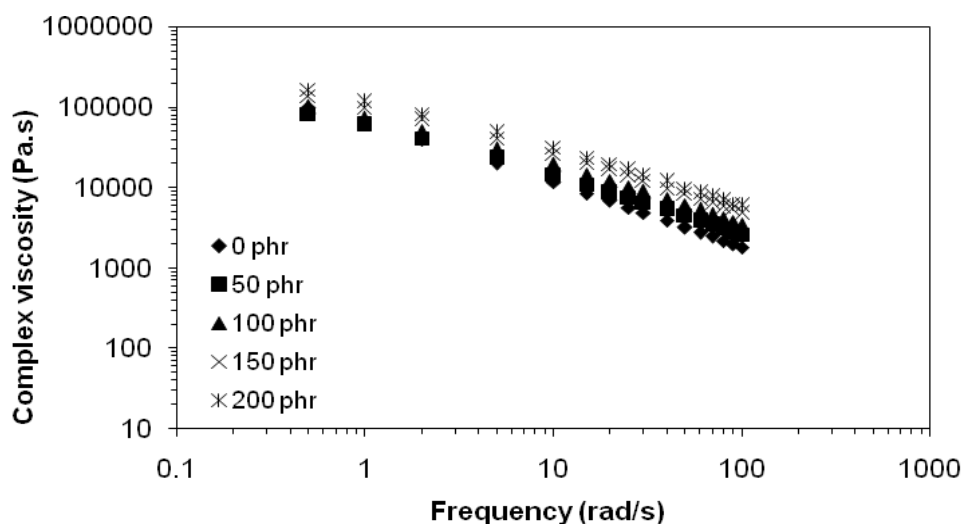


Figure 5.17 Complex viscosity as a function of frequency of NR compounds filled with various FA loadings

For the view of molecular degradation of NR matrix, the molecular weight (MW) of NR matrix as a function of FA loading was measured to validate the proposed explanation by the uses of the rheometers which are RPA2000 and parallel plate rheometer (PP). It can be seen from Figure 5.18 that, both RPA2000 and PP results show similar trend of the normalized MW of NR matrix as a function of FA loading. It is obvious in Figure 5.19 that the MW of both NR and synthetic IR systems decreases with increasing FA loading, and the magnitude of MW reduction is noticeably more pronounced in NR system. Since the main difference in these two systems is the presence of non-rubber substances in NR, it is believed that the non-rubber substances are responsible for the reduction in MW during the storage duration.

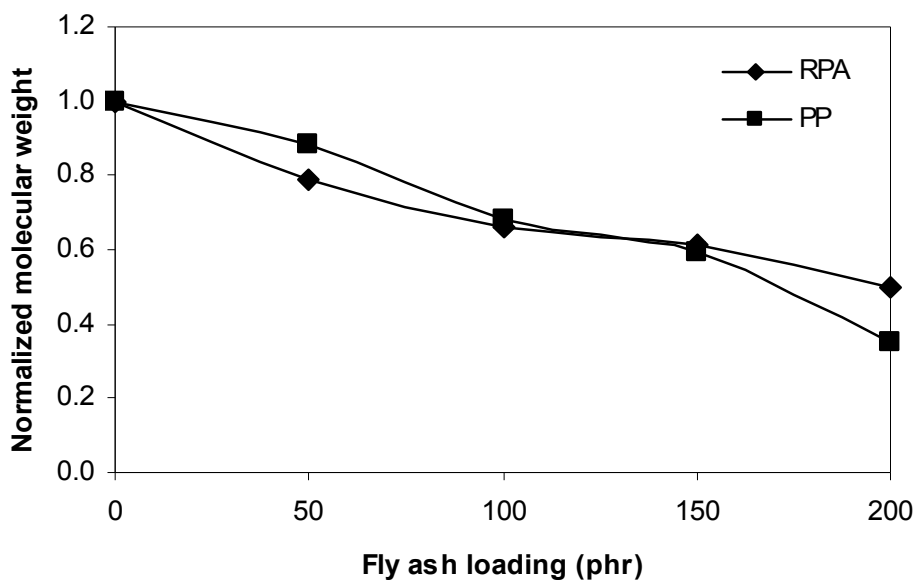


Figure 5.18 Normalized molecular weight (MW) of NR matrix of FA/NR compounds with various FA loadings measured from RPA2000 and PP rheometers

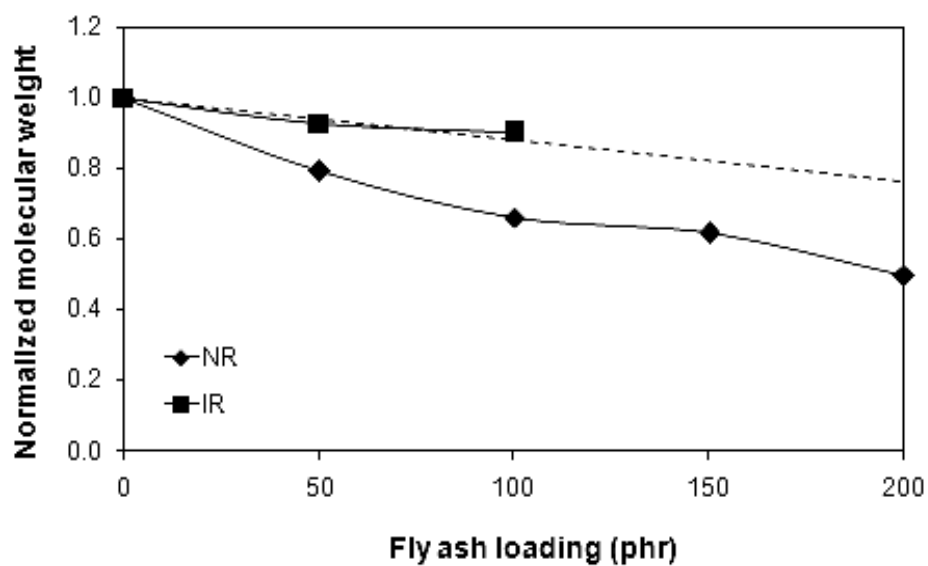


Figure 5.19 Normalized molecular weight (MW) as measured from RPA2000 of IR and NR matrices of FA filled compounds with various FA loadings (dash line as an extrapolation for eye-guidance purpose)

Furthermore, the power law index results of FA/NR and FA/IR as determined from RPA2000 are illustrated in Figure 5.20. The power law index increases with adding FA in NR compounds. Except for unfilled systems, the NR compounds show higher power law index than IR compounds, suggesting that NR compounds is more Newtonian-like as a result of viscous behavior promotion as discussed earlier. Moreover, the result shows that the power law index is independent of FA content in NR and IR compounds implying the domination of the ball bearing effect which could effectively facilitate molecular flow except for low loading of 50 phr

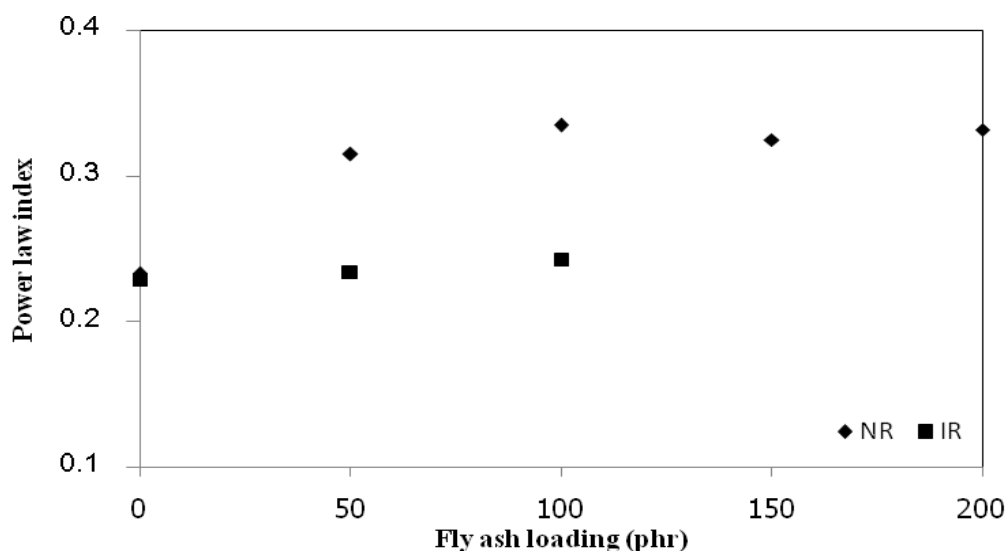


Figure 5.20 Power law index of FA/NR and FA/IR with various FA loading as determined from RPA2000

D) Storage stability and thermal aging properties

For a storage stability investigation of FA-filled NR compounds, the compounds were kept at least 12 weeks at room temperature. The RPA2000 was used for monitoring the viscoelastic behavior of compounds. In the study of thermal aging properties, the compounds were aged in RPA2000 test cavity at 120 °C for 1 hour. The viscoelastic properties of stored or aged compounds were

determined from RPA2000 at 70°C. Results before and after thermal aging were compared.

The results of storage stability are presented in Figure 5.21 demonstrating the normalized storage modulus (G') of FA/NR compounds with various FA loadings. Amazingly, the normalized G' of all compounds with FA particles decreases particularly at the high FA loading. The results suggest relatively poor storage stability of NR compounds filled with FA particles. On the contrary, the normalized G' of synthetic IR compounds with various FA loadings as depicted in Figure 5.22 does not significantly alter with storage duration. The superiority of storage stability found in the synthetic IR is due probably to the absence of non-rubber substances.

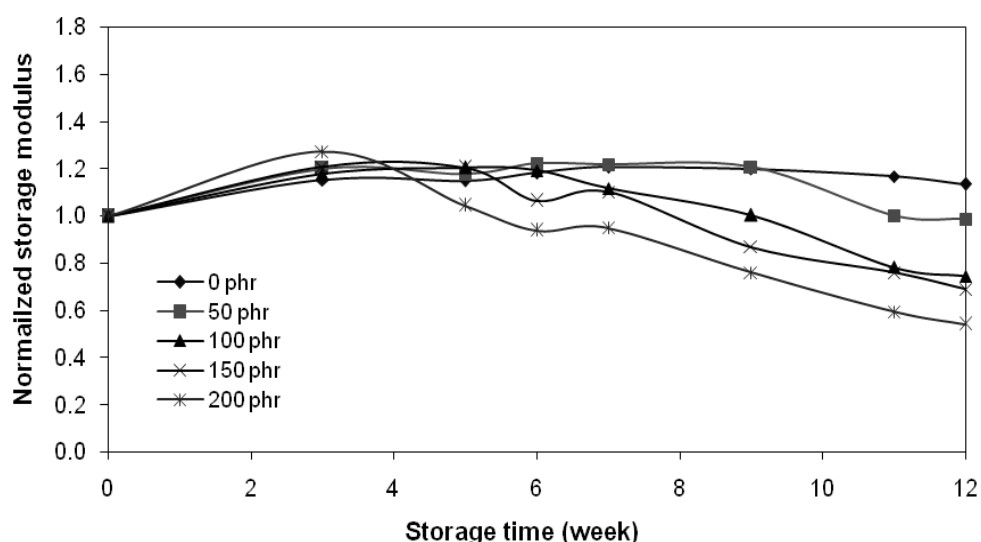


Figure 5.21 Effect of storage time on the storage modulus (G') of FA filled NR compounds

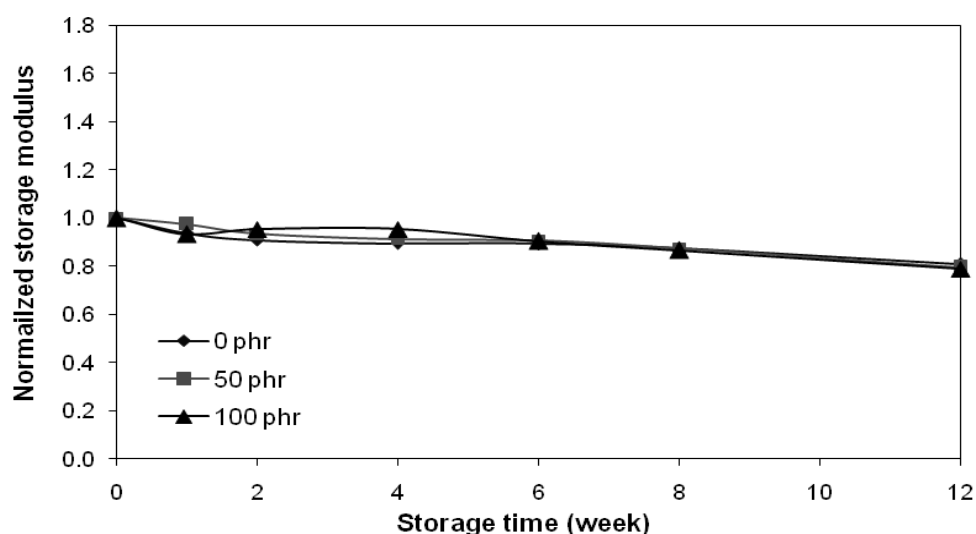


Figure 5.22 Effect of storage time on the storage modulus (G') of FA filled IR compounds

To further investigate the effect of heavy metals in FA, the 8-ppm heavy metals (i.e., manganese, iron and copper) were incorporated, and the results of normalized G' of the IR compounds having FA 0-100 phr were investigated as illustrated in Figures 5.23-5.25. Evidently, the obtained results are exhibited that all IR compounds with heavy metals reveal no significant change in normalized G' after storage time. It is believed that the decrease in normalized G' of FA/NR compounds or molecular degradation of NR matrix is dominated by non-rubber component in NR (See Fig. 5.21). Figure 5.26 illustrates the normalized G' of PSi/NR compounds with various PSi loadings (at similar amount of SiO_2 in FA). The normalized G' does not significantly change with storage duration. The good storage stability found in PSi/NR compounds is probably because of the absence (or very little) of no heavy metals existing in PSi.

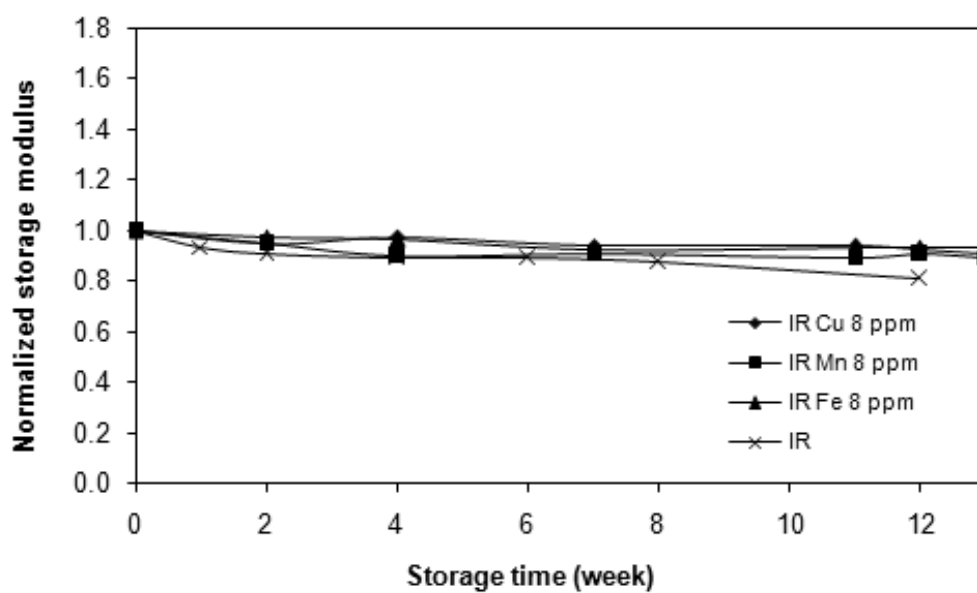


Figure 5.23 Normalized storage modulus of unfilled IR compounds containing different heavy metals

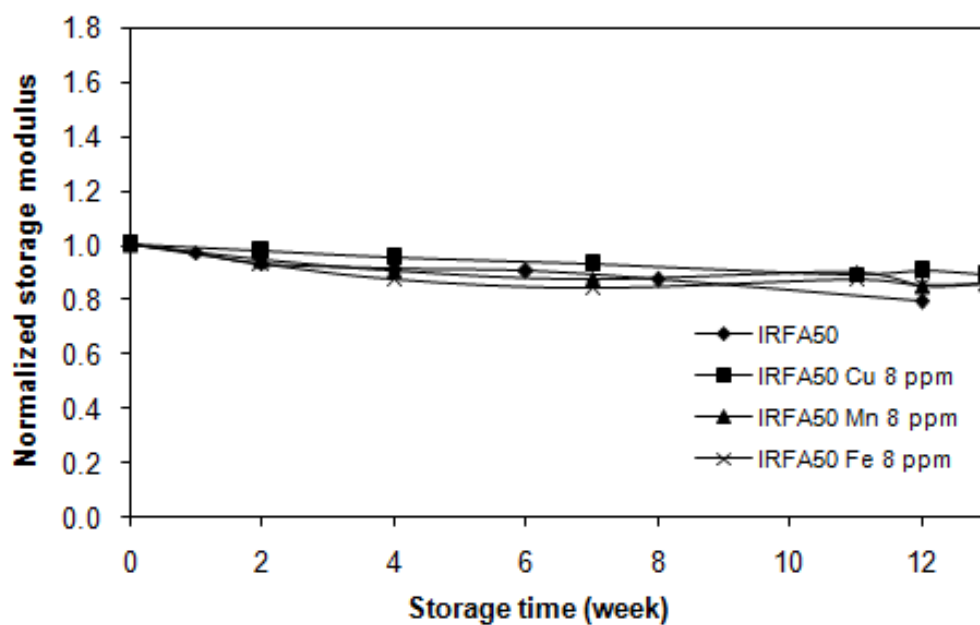


Figure 5.24 Normalized storage modulus of 50 phr FA filled IR containing different heavy metals

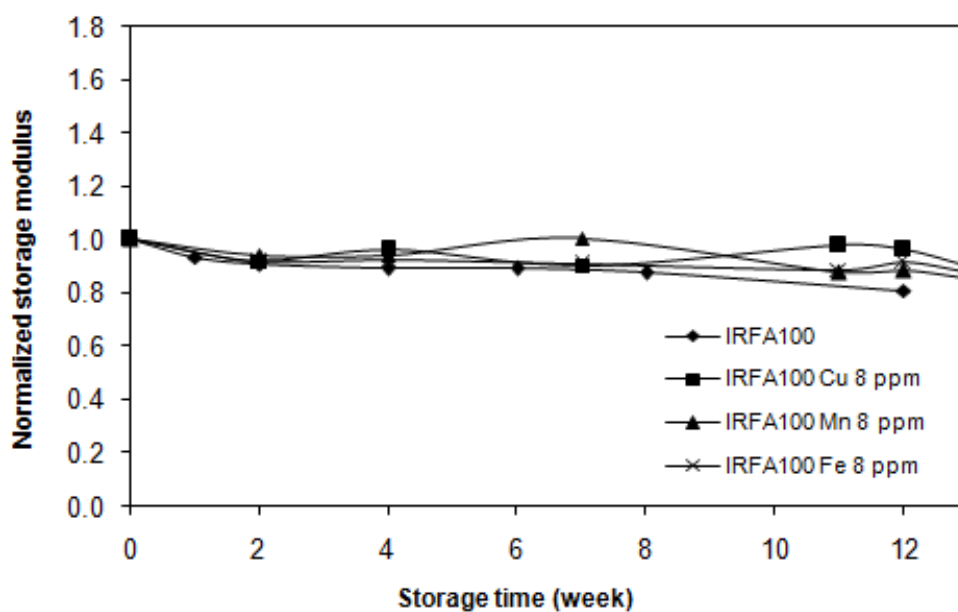


Figure 5.25 Normalized storage modulus of 100 phr FA filled IR containing different heavy metals

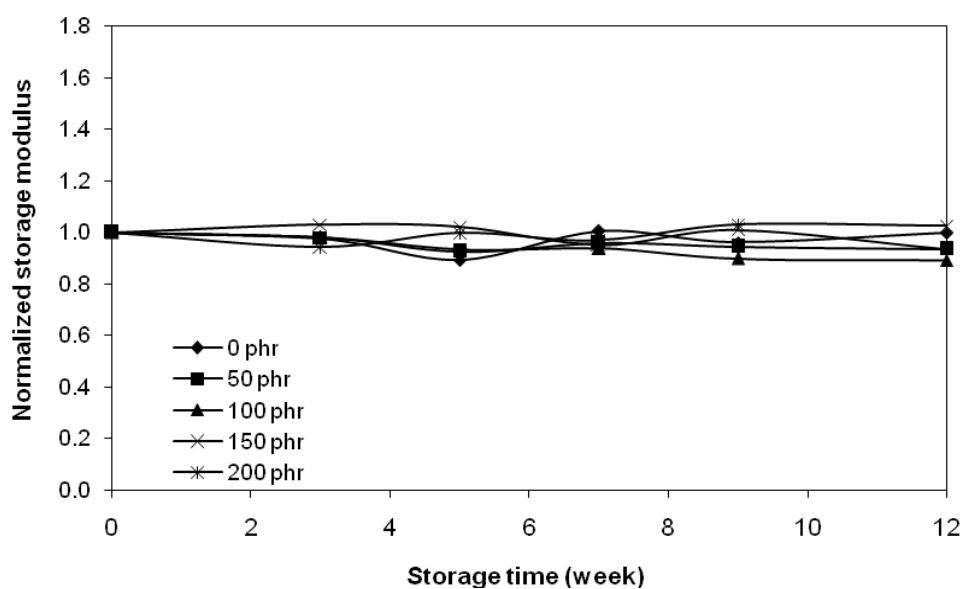


Figure 5.26 Effect of storage time on the storage modulus (G') of precipitated silica filled NR compounds

In other words, the sharp decrease in normalized G' found in NR compounds as shown previously in Figure 5.21 is believed to be the result of molecular degradation of NR molecules. To confirm the proposed statement, the normalized MW in NR matrix as extracted from the NR compounds freshly prepared is compared with that of compounds kept for 14 weeks as shown in Figure 5.27. It is evident that the values of normalized MW of FA filled NR compounds are significantly lower in the compounds with 14-week storage duration. This phenomenon is more pronounced in compounds filled with high FA loading. The results support the promotion of molecular degradation by the presence of FA particles containing heavy metals.

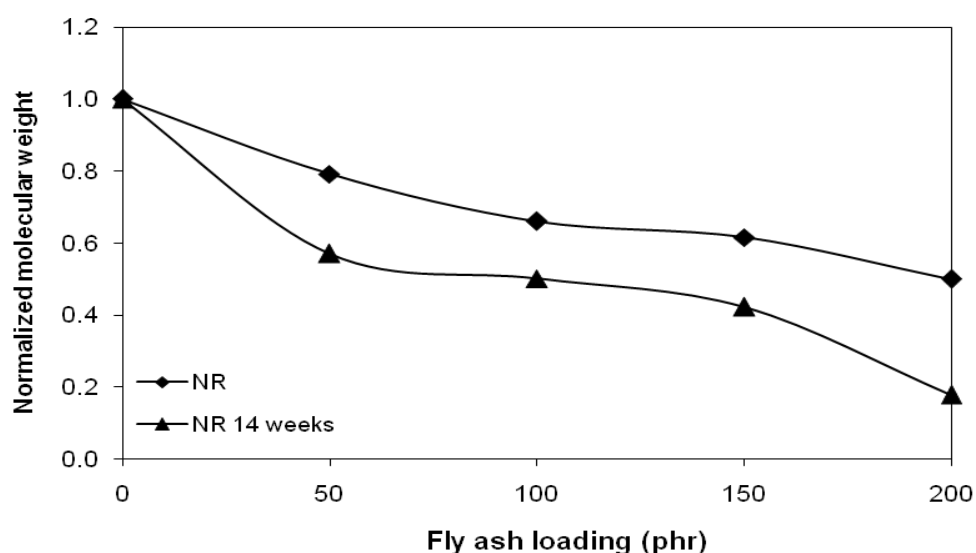


Figure 5.27 Normalized molecular weight of NR matrix of FA/NR compounds before and after 14-week storage

By correlating the change in MW with normalized G' , it is postulated that the storage instability found in FA filled NR system is attributed to the molecular degradation of NR molecules. The presence of heavy metals in both FA particles and non-rubber in NR to achieve the critical contents could accelerate the molecular degradation process. Also, the stearate and oleate salts of copper and manganese in the non-rubber substances are capable of catalyzing the oxidative

degradation of NR [136]. The possible mechanism of oxidative process is demonstrated in Figure 5.28 [137]. According to Figure 5.28, if the proposed mechanism based on radical reaction is true, such storage instability phenomenon shall be suppressed by the addition of radical scavenger.

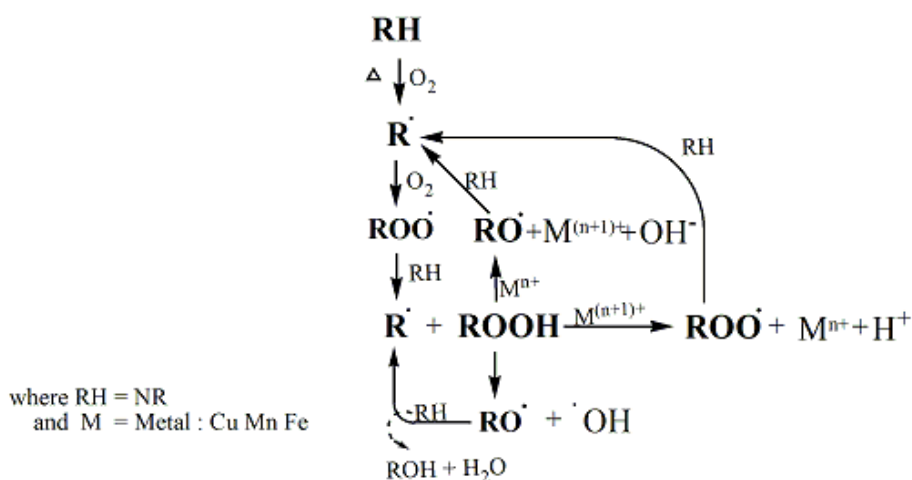


Figure 5.28 The possible mechanism of oxidative process of NR as catalyzed by heavy metals

To further investigate the degradation of NR and synthetic IR systems, the thermal aging test was performed by incubating the test specimens in RPA2000 test cavity at 120 °C for 1 hour. By this means, the acceleration of degradation process is gained. The retention of G' of both FA filled NR and IR compounds measured at 70 °C could then be compared as shown in Figure 5.29. Clearly, the synthetic IR compounds show much superior retention of G' and thus the thermal stability to NR compounds. It seems that the results are in line with the proposed explanation of molecular degradation of NR matrix. However, the thermal aging in RPA2000 performed at high temperature of 120 °C to accelerate the thermal aging process is not applicable to the actual storage conditions usually conducted at room temperature. Thus, the additional experiment on storage stability at room temperature is performed in order to monitor the alteration in viscoelastic properties of FA filled NR compounds as a function of storage duration at room temperature.

It is known that the amine-based antioxidant including N-(1,3-dimethyl)-N'-phenyl-p-phenylenediamine (6-PPD) could donate its reactive hydrogen atom (N-H) to the free radicals, and therefore interrupt the degradation process [139]. Figure 5.30 exhibits the normalized G' of NR compounds with 6-PPD. Evidently, there is no significant decrease in normalized G' with increasing storage duration, which validates the proposed degradation mechanism as illustrated in Figure 5.28.

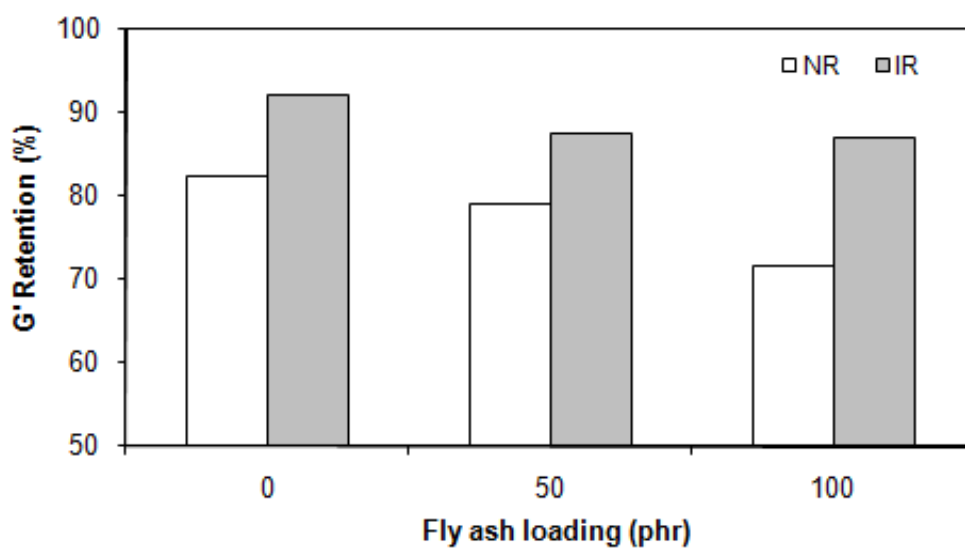


Figure 5.29 Retention of storage modulus (G') in FA filled NR and IR compounds measured from RPA2000

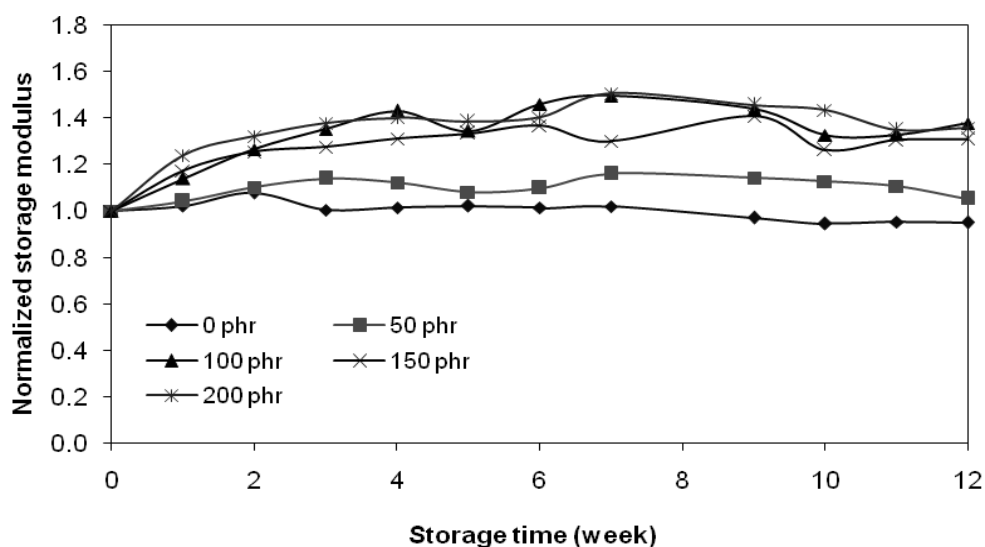


Figure 5.30 Normalized storage modulus (G') of FA/NR compounds stabilized with PPD antioxidant

5.2.1.2 Steady Shear Rheometer

Figure 5.31 shows apparent shear viscosity (η_{app}) of uncured FA/NR with various shear rate ($\dot{\gamma}$). It is evident that the η_{app} decreases with increasing $\dot{\gamma}$, and there is a slight increase in shear viscosity with increasing FA loading at any given shear rate. It is clear that all uncured FA/NR shows pseudoplastic behavior similar to result of complex viscosity (η^*) measured with various frequency from RPA2000.

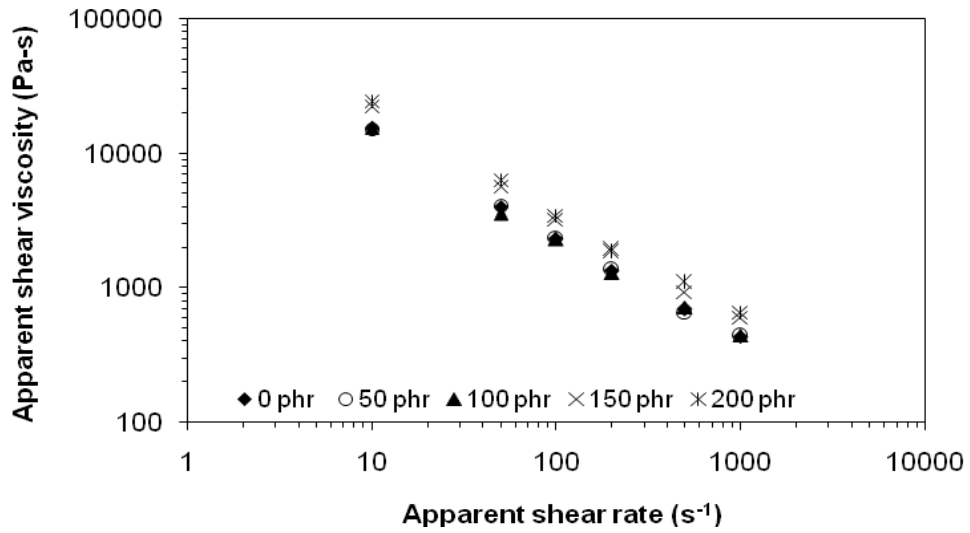


Figure 5.31 Flow curves of uncured FA/NR with FA loading (L/R = 30)

It is well known that the apparent results are the combined responses of elastic and viscous behaviors. In order to eliminate the elastic behavior, the Bagley correction is applied, so that the results obtained is caused mainly by the viscous shear flow. In addition, the Rabinowitsch correction, which is a correction of shear rate, is used to compensate a non-parabolic flow pattern in capillary die. The corrected rheological results of FA/NR is presented in Figure 5.32, which is the plot of true shear viscosity (η_{true}) against corrected shear rate ($\dot{\gamma}_{corr}$). The result shows similar pseudoplastic behavior to that of result before correction. Additionally, the trend of true shear viscosity (excluding elastic behavior) is similar to apparent shear viscosity. The results imply that the elastic behavior does not play strong role on rheological results compared to the viscous component.

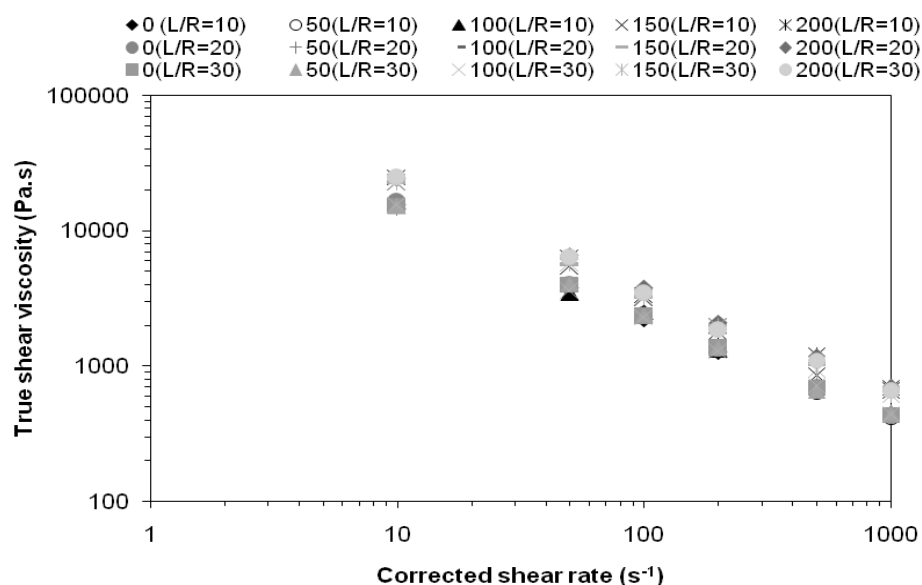


Figure 5.32 Flow curves after corrections of uncured FA/NR with various FA loadings and die geometries

Additionally, the power law index results of FA/NR as determined from capillary rheometer are illustrated in Figure 5.33. The result shows that the power law index is independent of FA content in NR compounds, and increases with adding FA as agreed with the results from RPA2000 because of the dilution effect (see Fig. 5.20)

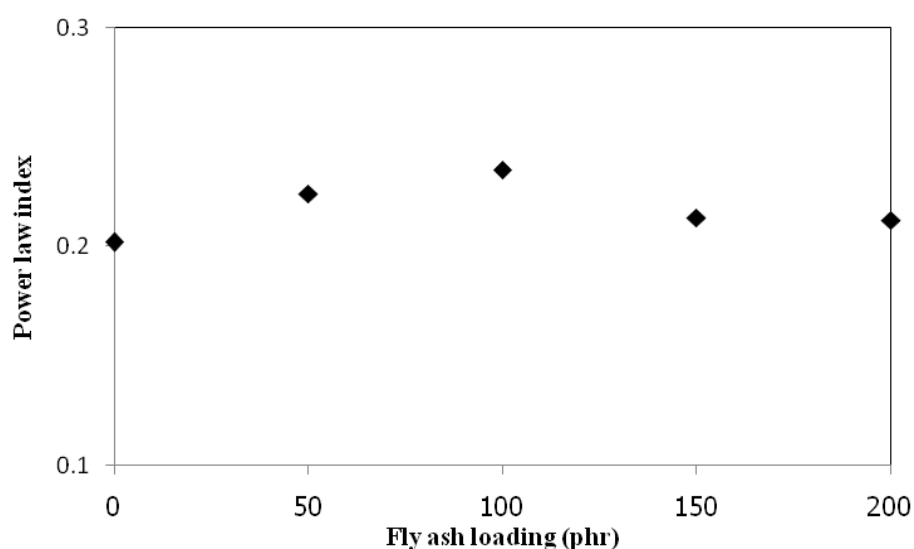


Figure 5.33 Power law index of FA/NR with various FA loadings as determined from capillary rheometer

5.2.1.3 Correlation between Oscillatory and Steady

Shear Flow

In order to correlate the rheological data under oscillatory to those under steady shear flow, the Cox-Merz concept was applied, and the results obtained of NR and IR compounds are illustrated in Figure 5.34. It is obvious that, in the case of unfilled NR and IR compounds, the complex viscosity measured under oscillatory shear flow is slightly higher than apparent shear viscosity measured under steady shear flow, which is due probably to the end effect taking place via elasticity of compounds. This end effect leads to an additional pressure measured. On the contrary, in FA filled compounds, both plots of complex and apparent shear viscosity become superimposed. This is because of the flow promotion via the viscous response as provided by FA.

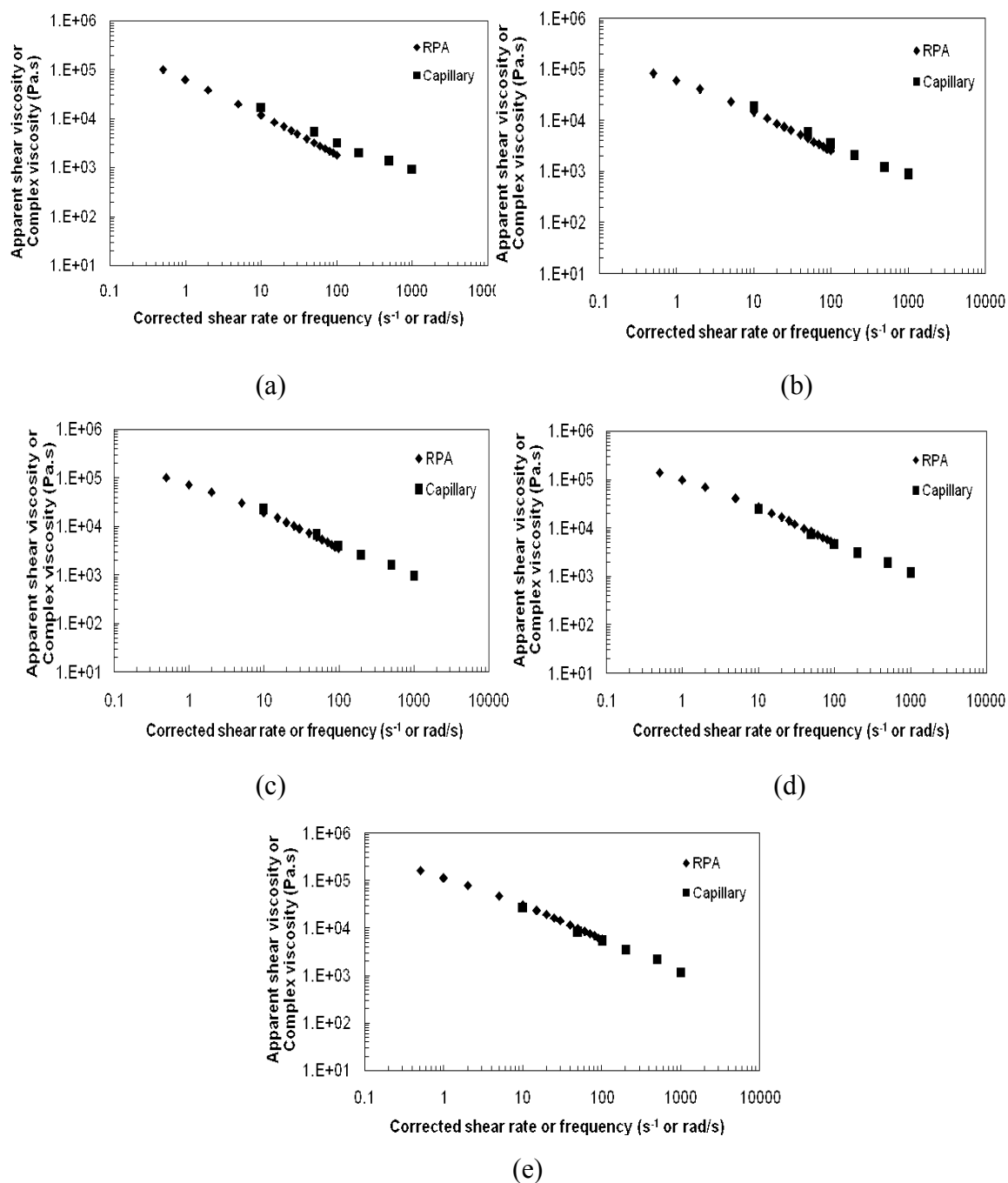
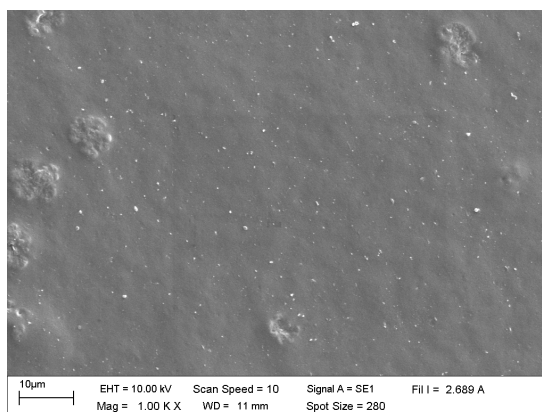


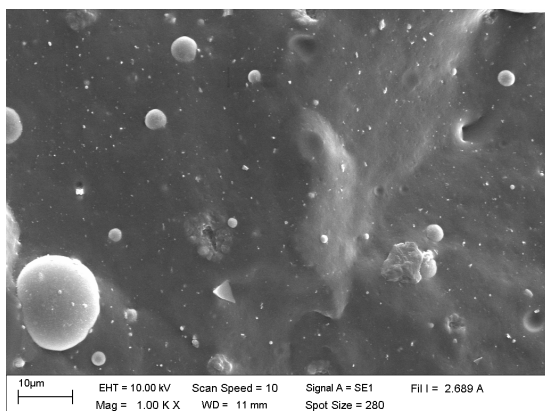
Figure 5.34 Plots of complex viscosity determined from RPA2000 as a function of frequency (◆) and apparent shear viscosity measured from capillary rheometer as a function of shear rate (■) of NR compounds filled with various FA loadings: (a) 0; (b) 50; (c) 100; (d) 150 and (e) 200 phr

5.2.2 Morphological Results

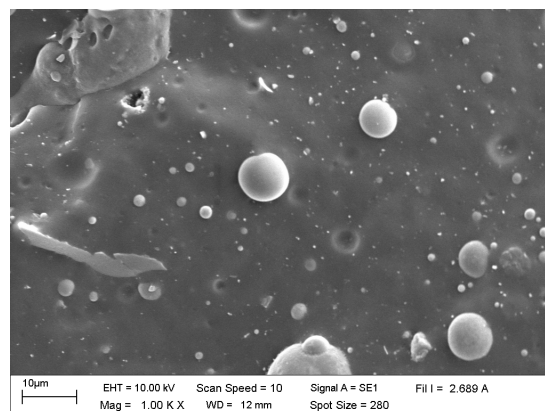
Morphological results of FA filled NR compounds with various FA loadings are illustrated in Figures 5.35. It can be seen that the higher the FA loading, the better the FA distribution. However, interfaces between FA particles and NR matrix are not homogeneity and some FA particles emerged from NR matrix indicating the poor wettability.



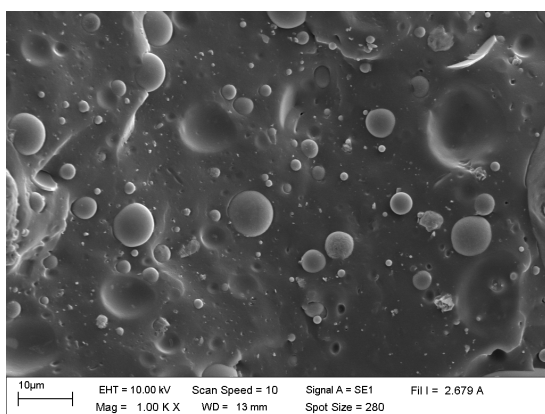
(a)



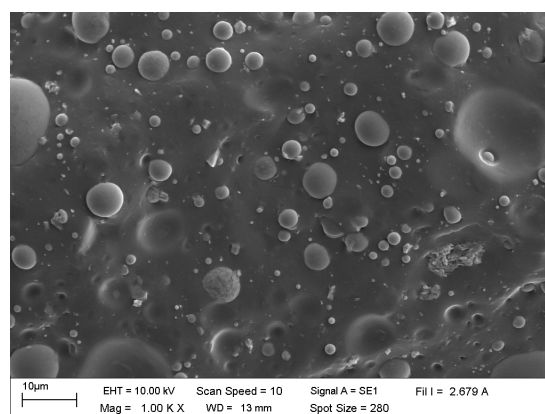
(b)



(c)



(d)



(e)

Figure 5.35 SEM micrographs of FA filled NR with various FA loadings: (a) 0; (b) 50; (c) 100; (d) 150 and (e) 200 phr

5.3 Effect of Silane Coupling Agent on Properties of FA Filled NR Compounds

As mentioned earlier, it is rather difficult for silica to be dispersed throughout the NR matrix and thus the FA because of a silicate structure as a majority in FA particles. Silane coupling agent (SCA) is therefore used to enhance the dispersion of FA and viscoelastic properties of FA/NR compounds. In this part, the attempts to improve viscoelastic properties NR filled with FA was performed by incorporating 3 types of silanes; bis(3-triethoxysilylpropyl) tetrasulfide (Si-69 or TESPT), 3-thiocyanatopropyl triethoxy silane (Si-264 or TCPTS) and vinyltriethoxysilane silane (VTEO) during mixing. The chemical structures of silane coupling agents used in this study are illustrated in Figure 5.36. Among 3 silanes used, it is expected that (i) the sulfur contribution in Si-69 can be promoted the sulfidic linkage formation; (ii) the presence of thiocyanide group (-SCN) in Si-264 might cause a strong interaction by hydrogen bond with silanol groups on FA surface and (iii) the vinyl groups on VTEO might react with NR (activated by peroxide curing system).

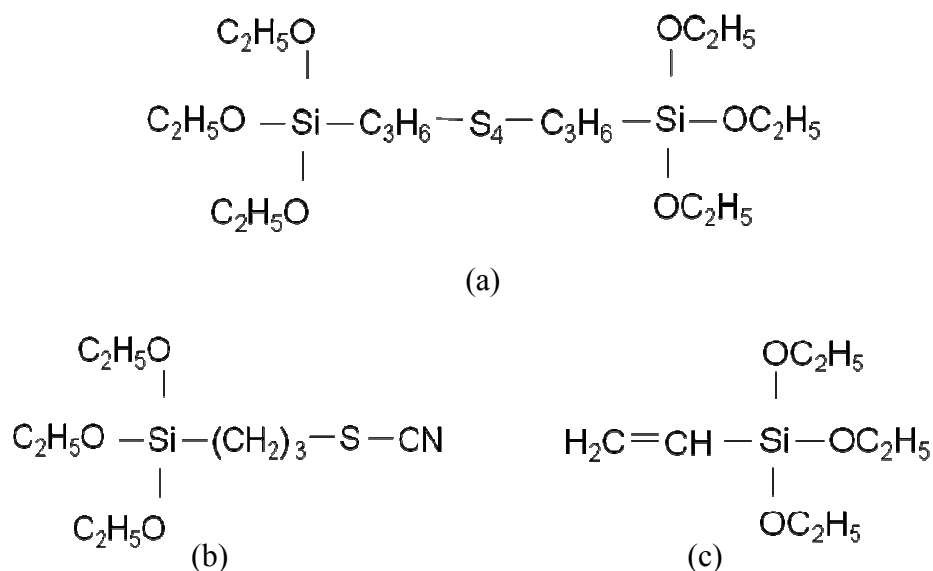


Figure 5.36 Structure of silane coupling: (a) bis(3-triethoxysilylpropyl) tetrasulfide (Si-69 or TESPT), (b) 3-thiocyanatopropyl triethoxy silane (Si-264 or TCPTS), and (c) vinyltriethoxysilane silane (VTEO)

5.3.1 Viscoelastic Behavior

A) Time Sweep Test

Initially, the thermal stability test was performed to determine the time available for performing the test with no thermal degradation related. Results of time sweep test of FA/NR compounds with Si-69, Si-264 and VTEO are shown in Figures 5.37, 5.38, and 5.39, respectively. It is found that the increase in storage modulus or G' is found in compounds with all silanes, which could be explained by the linkage formation between FA and NR. Moreover, all results exhibit good thermal stability. This means the effect of thermal degradation could be excluded from viscoelastic behavior results to be subsequently measured as functions of strain and frequency.

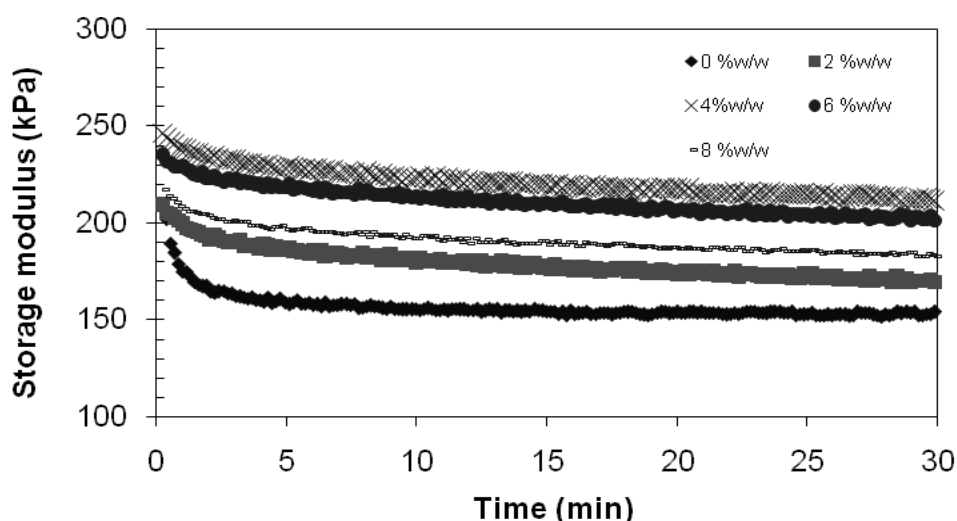


Figure 5.37 Storage modulus (G') as a function of time of 100 phr FA filled NR compounds with various Si-69 loadings

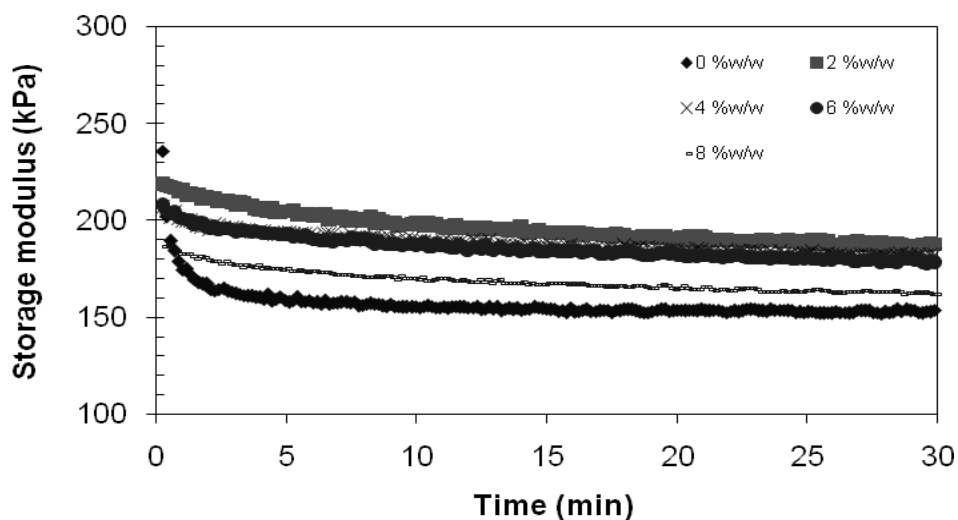


Figure 5.38 Storage modulus (G') as a function of time of 100 phr FA filled NR compounds with various Si-264 loadings

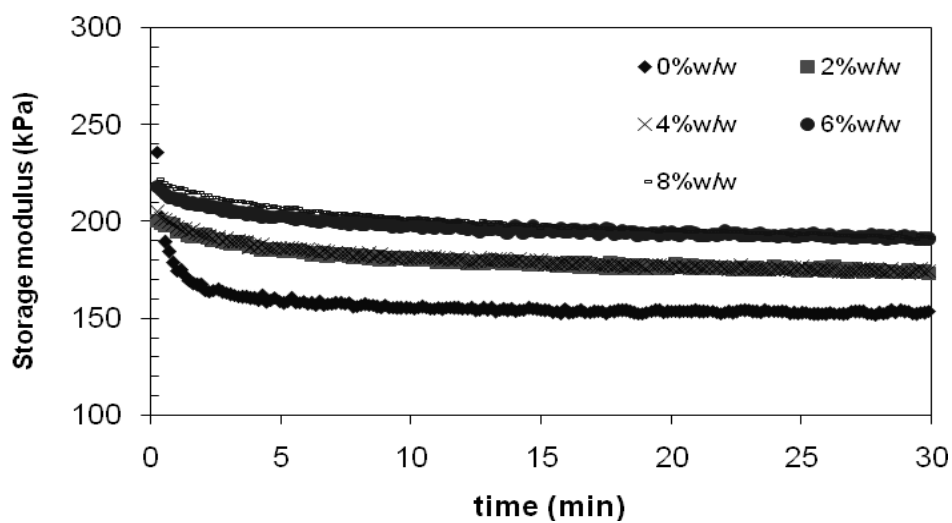


Figure 5.39 Storage modulus (G') as a function of time of 100 phr FA filled NR compounds with various VTEO loadings

B) Strain Sweep Test

Figures 5.40 to 5.42 show G' results as a function of strain deformation in FA/NR compounds with various silane loadings. A decrease in G' at high strain found in all compounds is caused by the disruption of filler transient network in conjunction with the molecular slippage of NR chains (due to the absence of crosslinks). Additionally, it is evident that the G' increases with increasing loading of silanes, which is relatively pronounced in the case of Si-69 silane. It can be explained by the linkage formation between FA and NR. Remarkably, such increase in G' by the addition of silane is in good agreement with the results of time sweep test as discussed previously. The plot of relative storage modulus as a function of silanes loading is shown in Figure 5.43. It is known that the relative G' calculated from Guth-Gold equation excludes the filler network and filler-rubber effects. From the results, it can be seen that the addition of 3 silanes, Si-69, Si-264 and VTEO, to FA/NR compounds still gives the negative deviation of relative modulus from the calculated Guth-Gold data. This is believed to be the results of molecular slippage dominated by a ball-bearing effect as a result of FA spherical shape, unlike the precipitated silica as reported previously [128, 129]. Notably, the addition of 4-8 %w/w of Si-69 shows slightly greater relative G' than the addition of 2 %w/w, especially, 4% of Si-69. This is due to the greater wettability of rubber on FA surfaces and an enhancement in the interaction between rubber-FA interaction in the compounds. Moreover, in the case of 6-8 %w/w of Si-69, the relative G' slightly drops. This is because the domination of plasticizing by formation of flexible polysiloxane from the self-condensation reaction of the Si-69 [140] over the interaction development effects. This proposed explanation is supported by the increase G' of unfilled compound after Si-69 addition as shown in Figure 5.44 [15]. On the other hand, in the case of Si-264 and VTEO silanes, the increase in silane loading shows no significant effect on the relative G' .

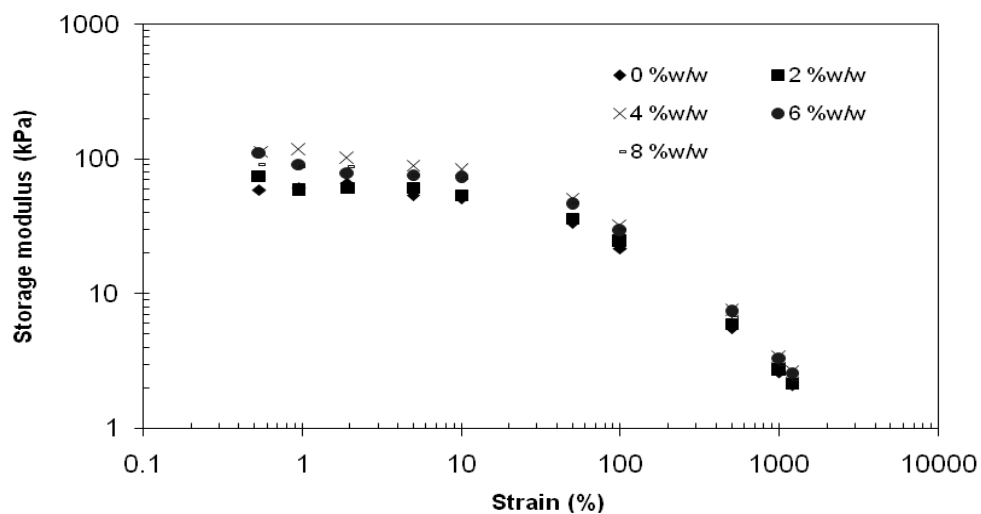


Figure 5.40 Storage modulus (G') as a function of strain amplitude of 100 phr FA filled NR compounds with various Si-69 loadings

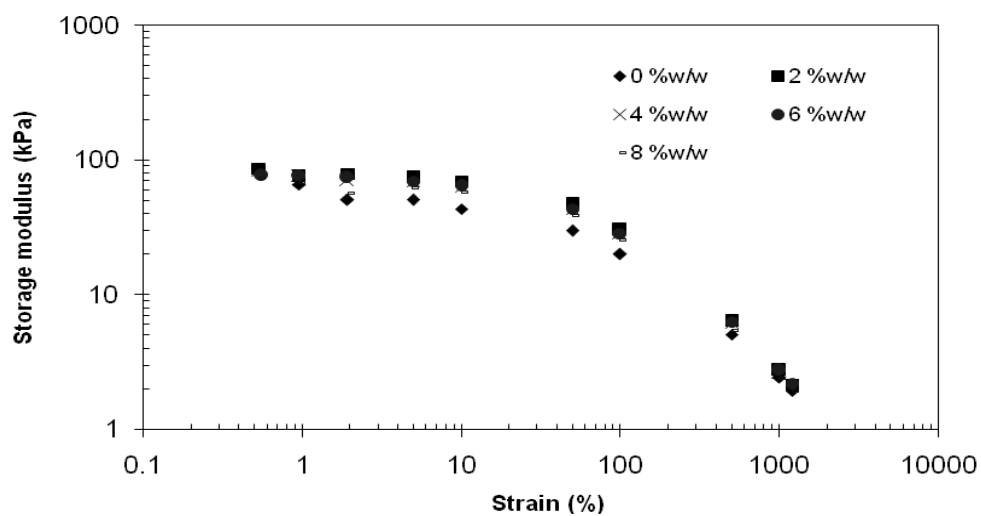


Figure 5.41 Storage modulus (G') as a function of strain amplitude of 100 phr FA filled NR compounds with various Si-264 loadings

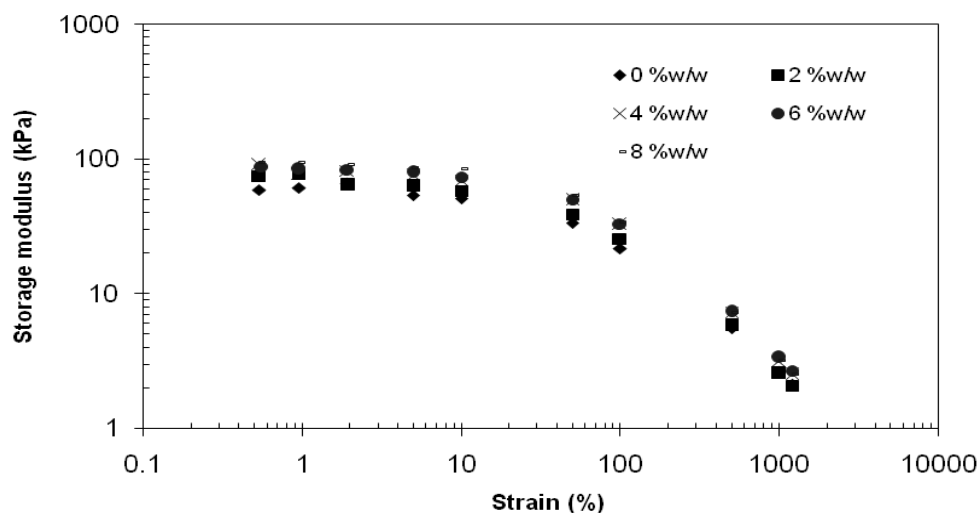


Figure 5.42 Storage modulus (G') as a function of strain amplitude of 100 phr FA filled NR compounds with various VTEO loadings

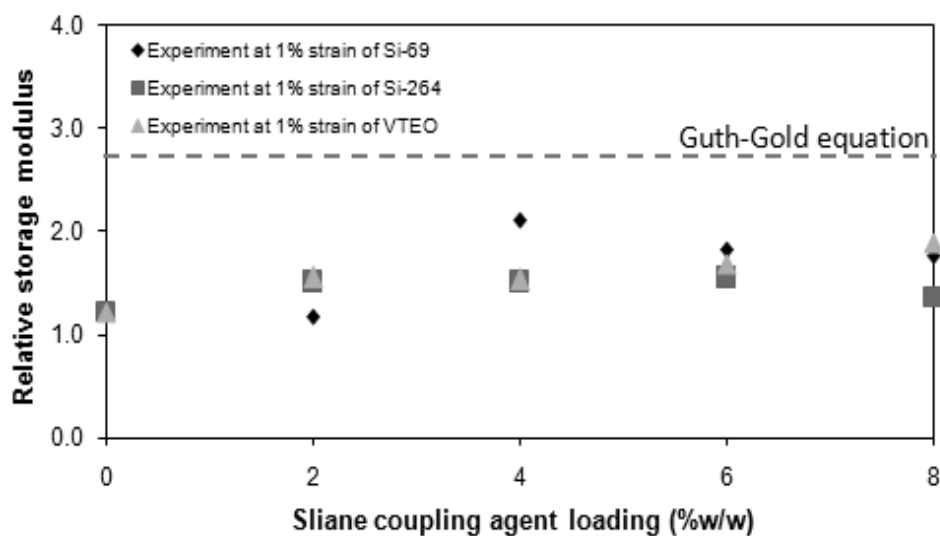


Figure 5.43 Relative storage modulus (relative G') at 1% strain of NR compounds filled with 100 phr of FA as a function of silane coupling agent loading

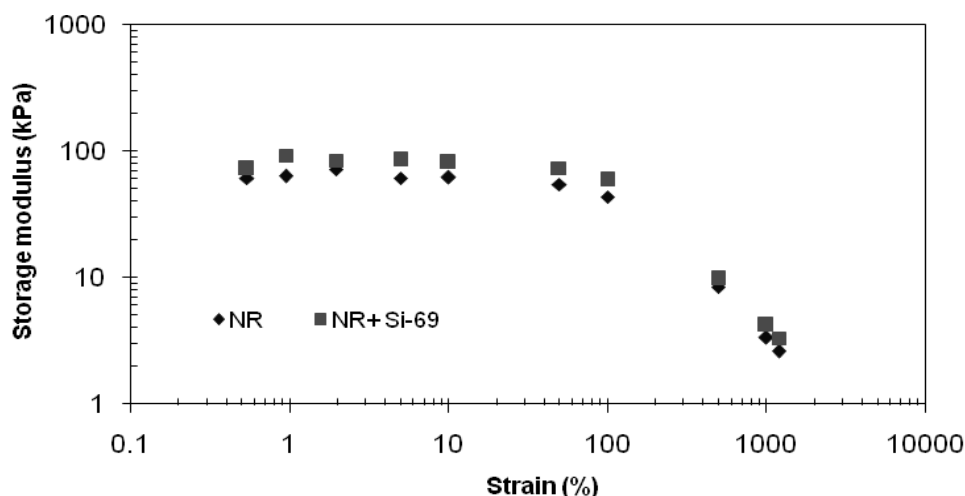


Figure 5.44 Storage modulus (G') as a function of strain amplitude of neat NR and NR with Si-69 compounds

Referred to the Payne concept [141], the enhancement in modulus could be due to various factors as displayed in Figure 5.45. The first one is the crosslink or polymer network following the hydrodynamic effect and the rubber-filler interaction including either physical or chemical interaction and both of all. The last one is the filler-filler interaction which is strain dependent and can be broken down at high strain. Figure 5.46 illustrates the effect of silane coupling agents loadings on the Payne effect of FA/NR compounds. It can be observed that the silane coupling agents do not reduce the Payne effect or $\Delta G'$. This is believed that the small amount of silanol groups on FA surfaces gives low magnitude of FA network formation. Compared with unmodified silica as reported previously, the silane can reduce filler-filler interactions, and thus the reducing Payne effect. Unexpectedly, it is noticed from the results that a magnitude of Payne effect ($\Delta G'$) increases with addition silane. From the Payne concept, the results exclude the crosslink and hydrodynamic effect, therefore, this is suggested that the addition of silanes to FA/NR compounds might help forming linkages between (i) NR and FA, (ii) the FA clusters in the FA/NR compounds, and (iii) the linkages between NR and silane [15].

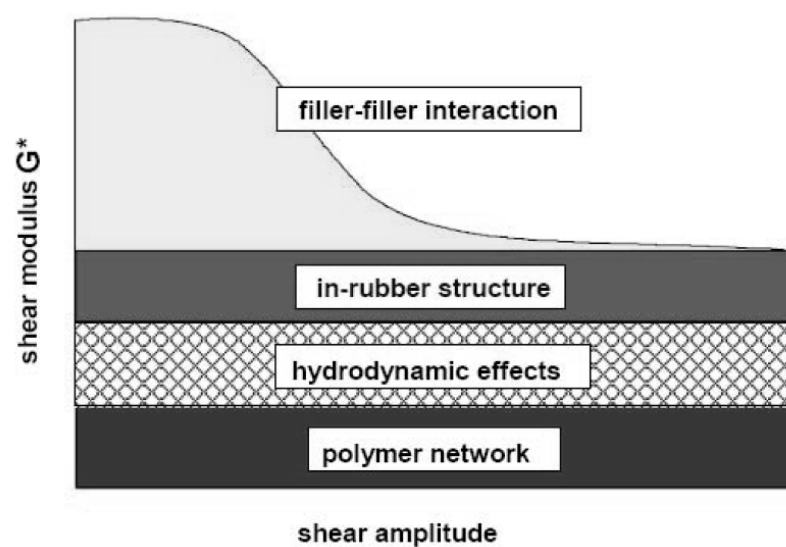


Figure 5.45 Parameters contributing to shear modulus [141]

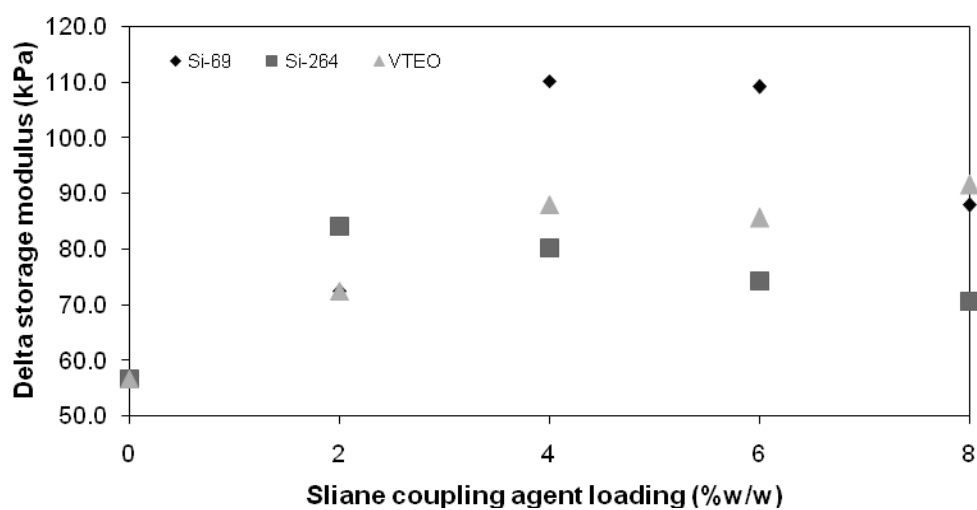


Figure 5.46 Effect of silane coupling agent loadings of 100 phr FA filled NR compounds on the Payne effect

The results of bound rubber content of FA filled NR compounds with various silane loadings are shown in Table 5.2. It can be seen that the bound rubber contents of compounds with silanes are not significantly different from the unmodified system, regardless of silane type. The results suggest poor interaction between FA and rubber molecules. It is noticed that the negative value of bound rubber content is probably due to the experimental error in weighting.

Table 5.2 Bound rubber content of 100 phr FA filled NR compounds with various silane loadings

Silane loading (%w/w)	Bound rubber content (%)		
	Si-69	Si-264	VTEO
0	-0.5004	-0.5004	-0.5004
2	0.0418	-0.0046	-0.0153
4	0.0509	0.0315	-0.0311
6	0.0437	0.0340	0.0375
8	0.0303	0.0288	0.0179

B) Frequency Sweep Test

Results of G' as a function of frequency in FA/NR compounds with 3 silanes coupling agents are illustrated in Figures 5.47 to 5.49. All compounds exhibit strong time dependent behavior, i.e., G' increases with increasing frequency as a result of insufficient time for molecular relaxation at high frequency. It is noticeable that the addition of silane leads to a rise in G' at low frequency. This implies a molecular mobility restriction caused by the interaction development of NR and FA linkages via the coupling reaction provided by (i) silane and (ii) NR and silanes. Moreover, the magnitude of frequency-dependent responses is less pronounced with increasing silane coupling agents. The results imply that an increase in elastic behavior of compounds with the addition of silanes, supporting the interaction development of NR-FA and/or NR-silane.

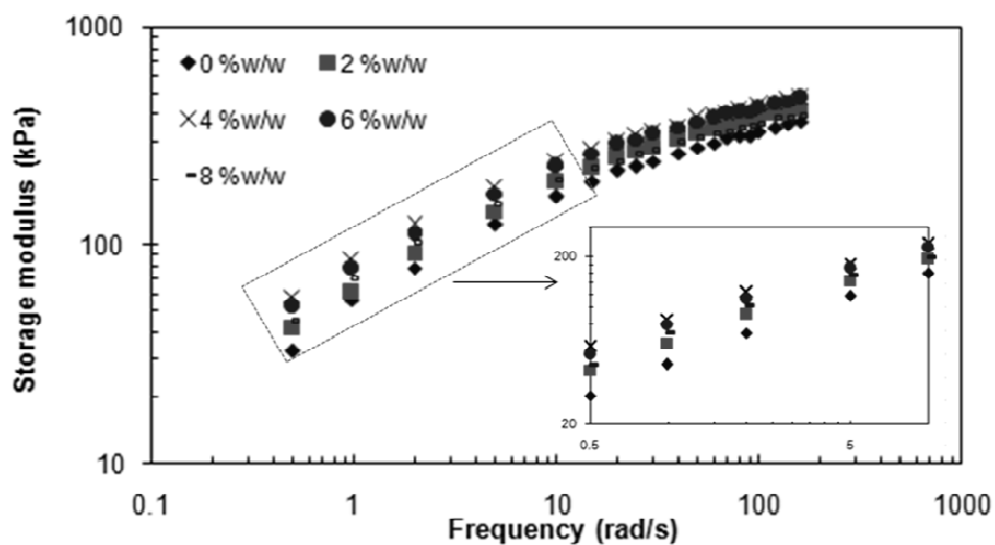


Figure 5.47 Storage modulus (G') as a function of frequency of 100 phr FA filled NR compounds with various Si-69 loadings

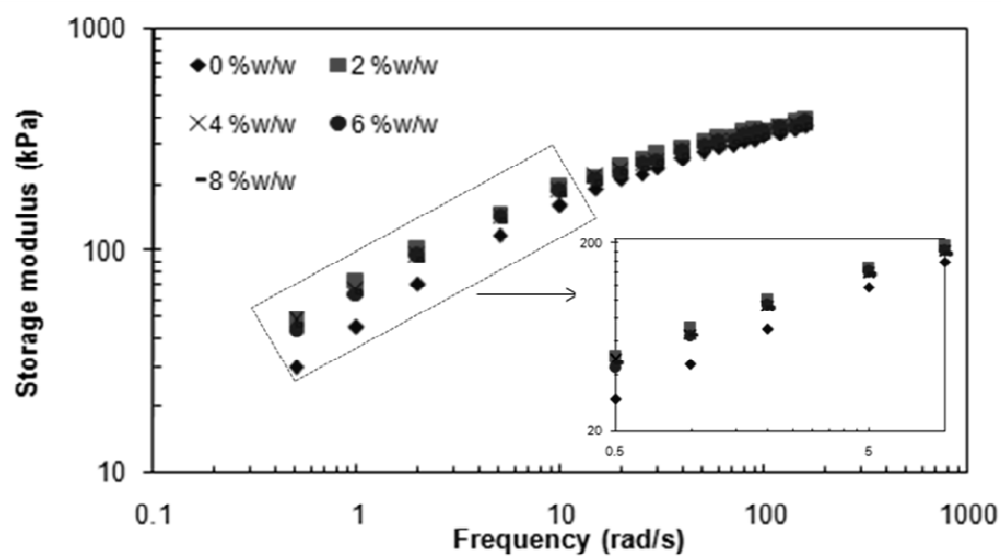


Figure 5.48 Storage modulus (G') as a function of frequency of 100 phr FA filled NR compounds with various Si-264 loadings

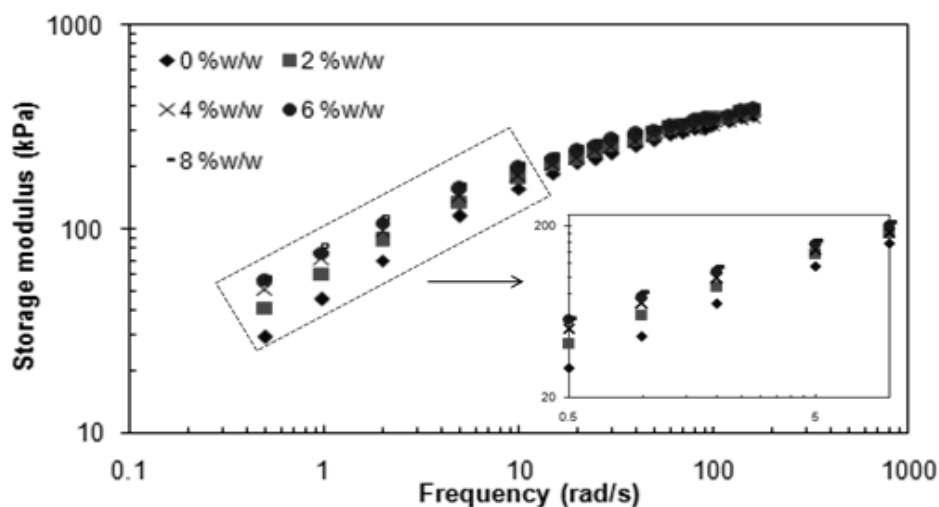


Figure 5.49 Storage modulus (G') as a function of frequency of 100 phr FA filled NR compounds with various VTEO loadings

Figures 5.50 to 5.52 reveal the results of damping factors of FA filled NR compounds with Si-69, Si-264 and VTEO silane coupling agents, respectively. Clearly, with increasing silanes content up to a certain loading, the damping factor decreases. Further increase in silane would lead to a rise in damping factor. The reduction in damping factor is attributed to the interaction development of FA particle with NR molecules and that of NR with silane, giving the elastic contribution of the systems. As silane is excessive, the viscous response would then be dominant at high silane loadings.

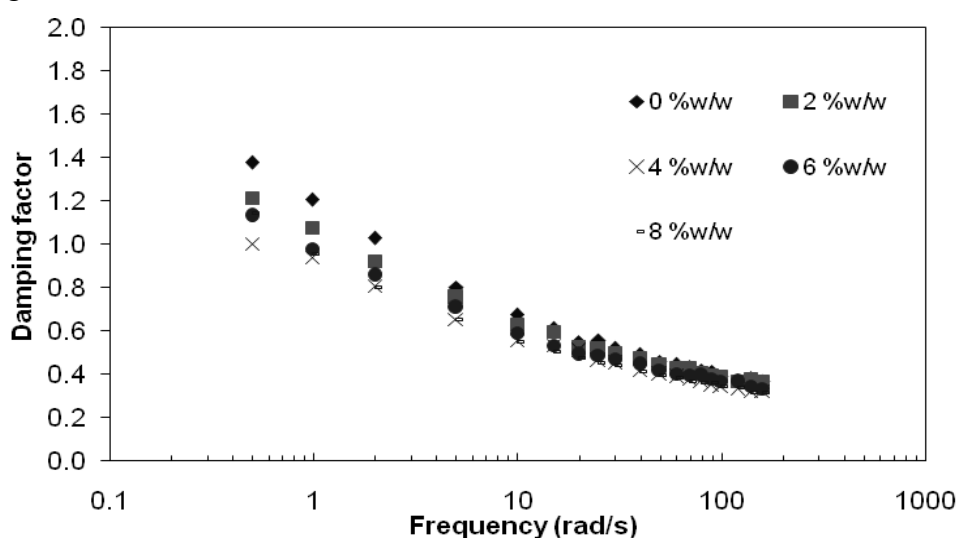


Figure 5.50 Damping factor ($\tan \delta$) as a function of frequency of 100 phr FA filled NR compounds with various Si-69 loadings

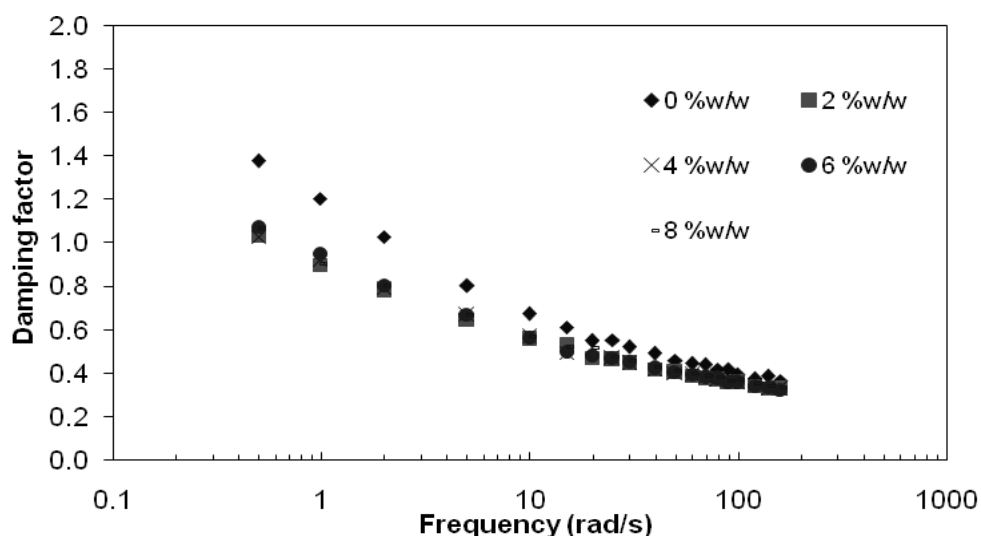


Figure 5.51 Damping factor (tan delta) as a function of frequency of 100 phr FA filled NR compounds with various Si-264 loadings

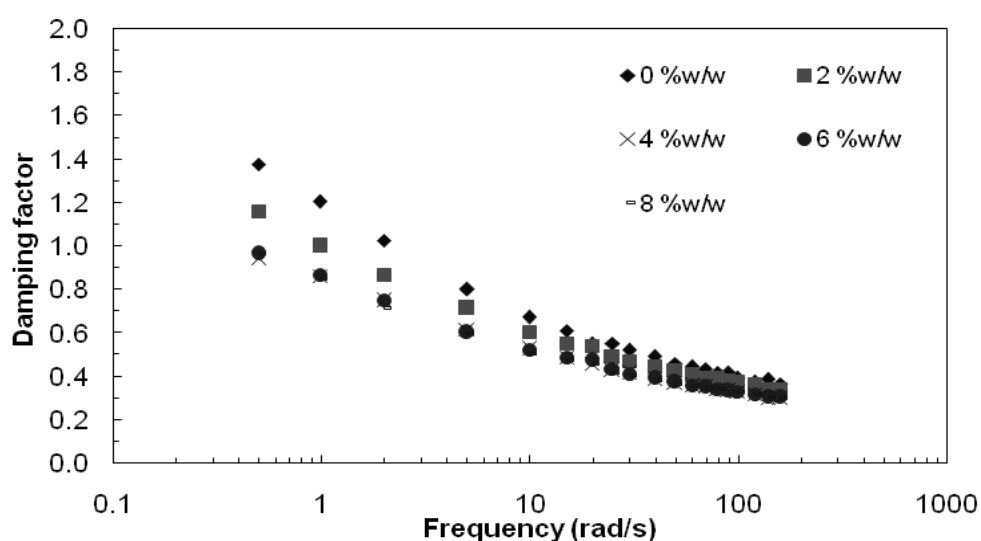


Figure 5.52 Damping factor (tan delta) as a function of frequency of 100 phr FA filled NR compounds with various VTEO loadings

As for the complex viscosity (η^*), it is clear that all compounds with all silane coupling agents exhibit a pseudoplastic behavior (η^* decreases with increasing frequency), as illustrated in Figures 5.53 to 5.55. In addition, as silane loading increases, the η^* slightly increases, it is believed that the development of NR – FA and NR-silane interactions are responsible for the increase in η^* . Figure 5.56 reveals

that the addition of silane coupling agents slightly reduces the power law index due to the rise of elasticity development of NR–FA and NR-silane interactions. The values of power law index with increasing silane loadings reduce and level off at 4 to 8% w/w. It can be noticed that the FA filled NR compounds with Si-69 and Si-264 silanes show the lower power law index than those with VTEO, meaning that the addition of VTEO to the compounds yields the lower degree of shear thinning behavior than Si-69 and Si-264. For Si-69 and Si-264, the power law result is not different, probably suggesting that there is the counter-balance between the size of molecule and the amount of active alkoxy groups on silane backbone.

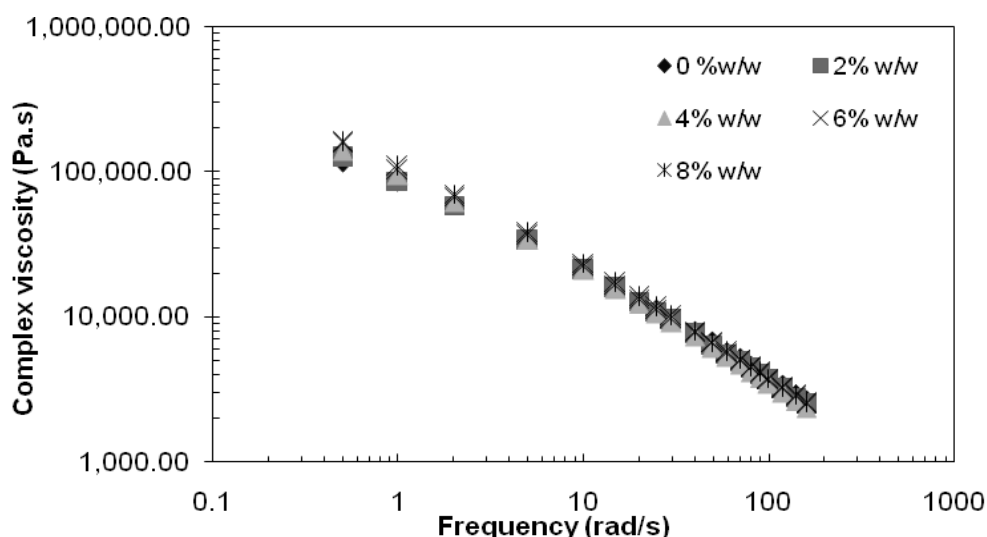


Figure 5.53 Complex viscosity as a function of frequency of 100 phr of FA/NR compound with various Si-69 loadings

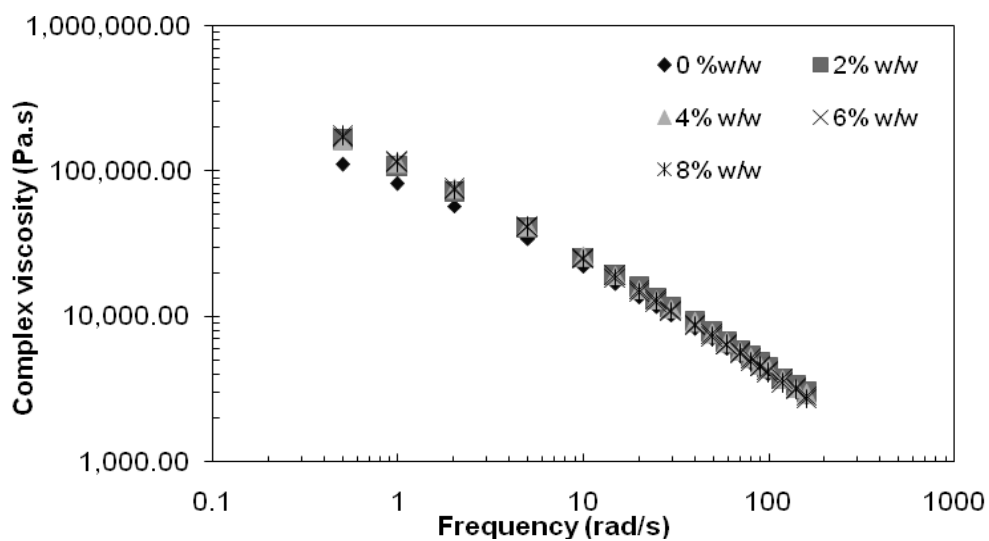


Figure 5.54 Complex viscosity as a function of frequency of 100 phr FA filled NR compounds with various Si-264 loadings

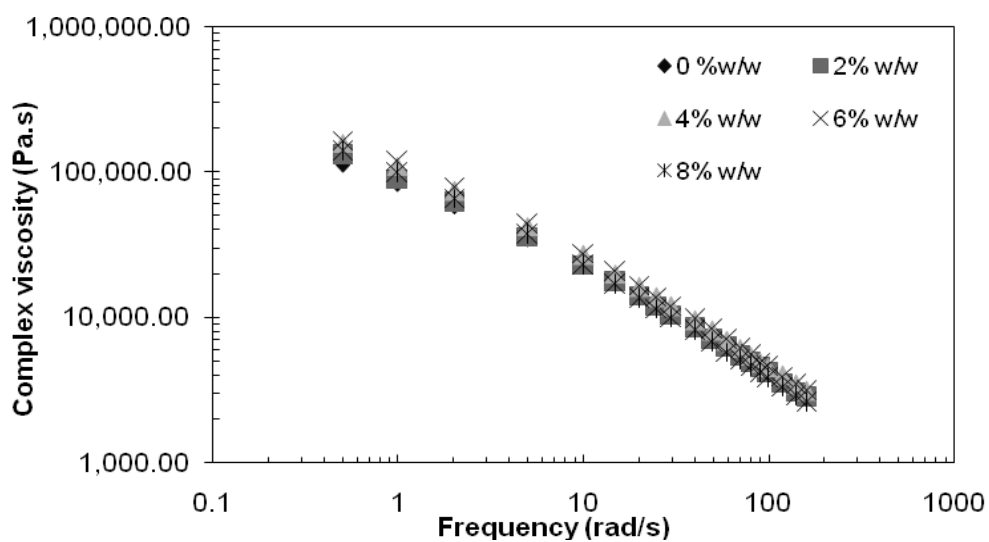


Figure 5.55 Complex viscosity as a function of frequency of 100 phr FA filled NR compounds with various VTEO loadings

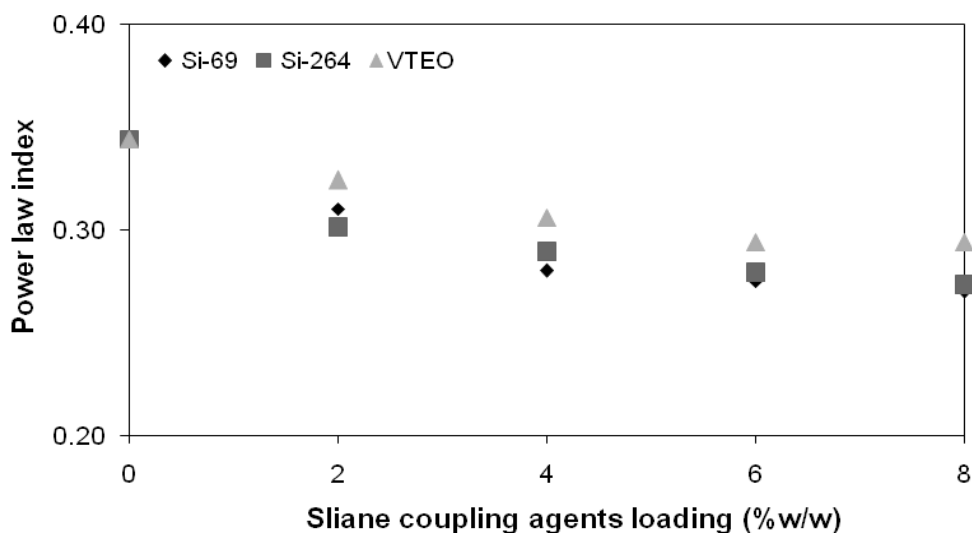
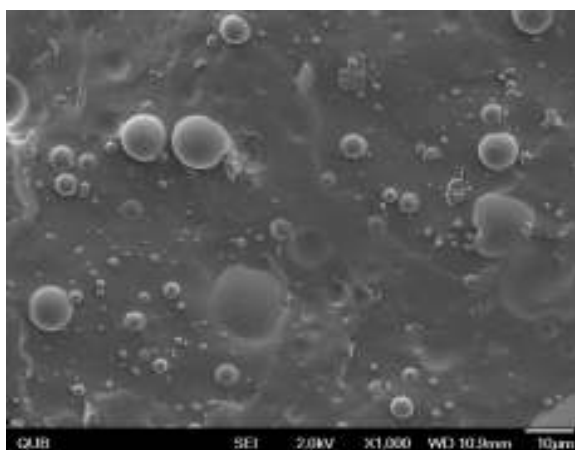


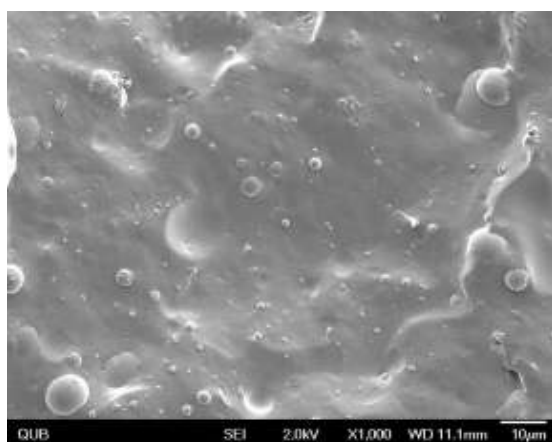
Figure 5.56 The effect of silane coupling agents loadings of FA/NR compounds on Power law index as determined from RPA2000

5.3.2 Morphological Results

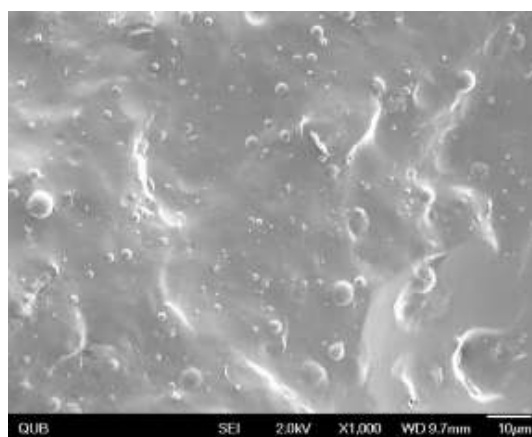
Morphological results of 100 phr FA filled NR compounds modified with various types and loading of silanes are illustrated in Figures 5.57 to 5.59. It can be seen that the boundaries between FA and NR without silane are inhomogeneous indicating poor wettability. Apparently, the silanes treated compounds exhibit superior interfacial adhesion which is evidenced by the embedding of FA particles in the NR matrix supporting the increase in elasticity such as the increased storage modulus and the decreased damping factor.



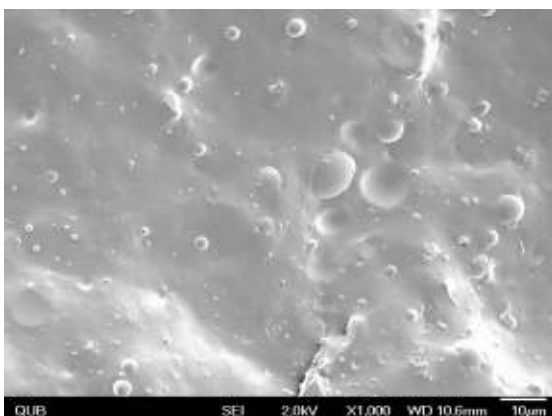
(a) 0 % w/w



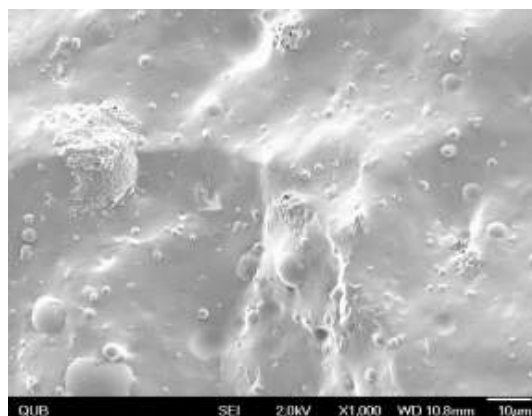
(b) 2 % w/w



(c) 4 % w/w



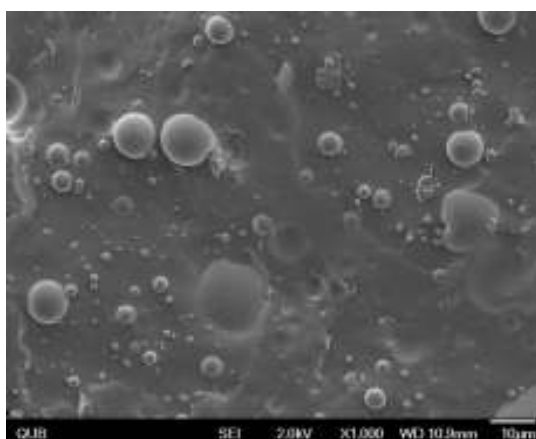
(d) 6 % w/w



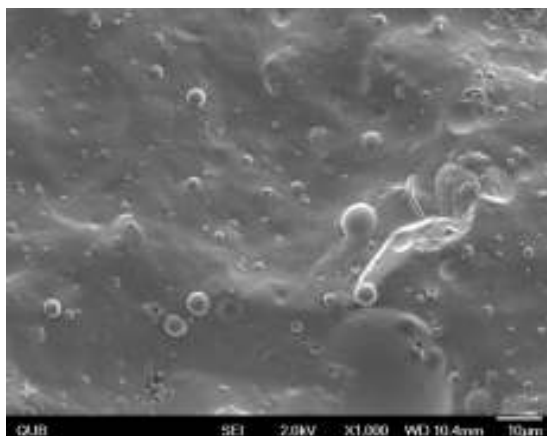
(e) 8 % w/w

Figure 5.57 SEM micrographs of 100 phr FA filled NR with various Si-69 loadings:

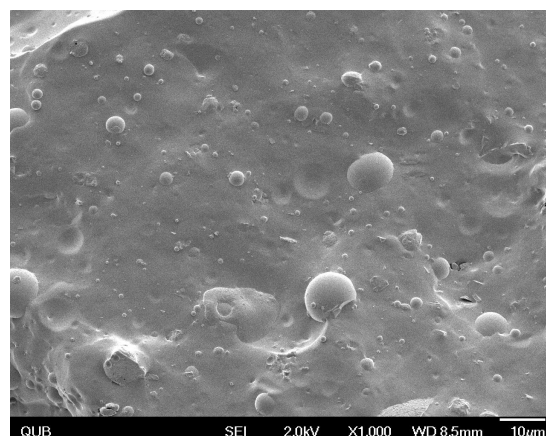
(a) 0; (b) 2; (c) 4; (d) 6 and (e) 8% w/w



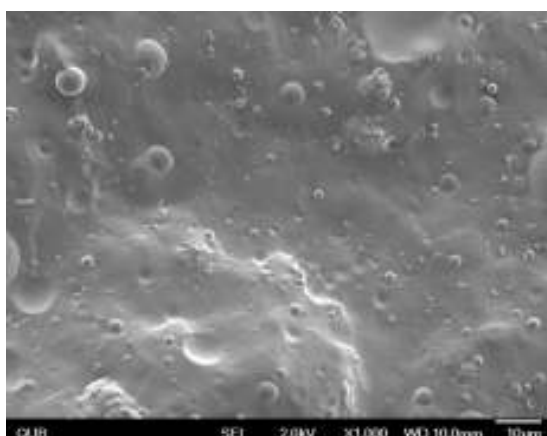
(a) 0 % w/w



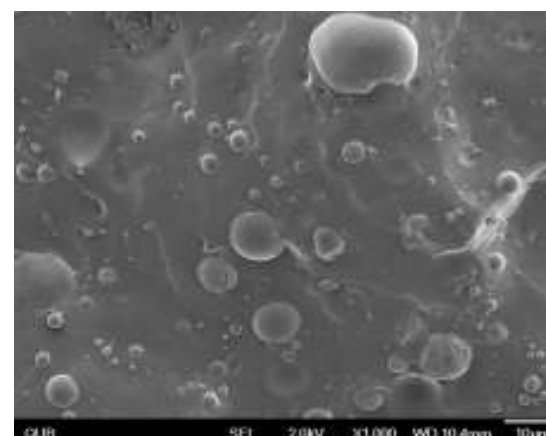
(b) 2 % w/w



(c) 4 % w/w



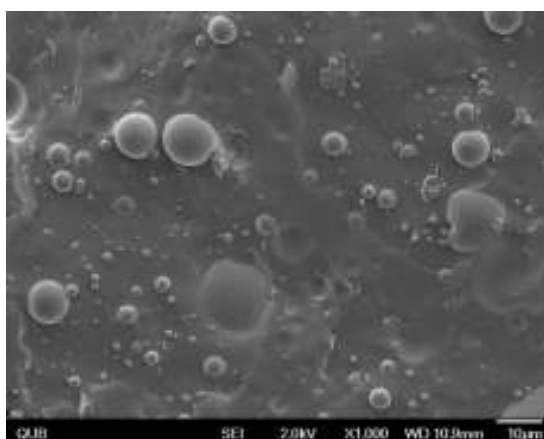
(d) 6 % w/w



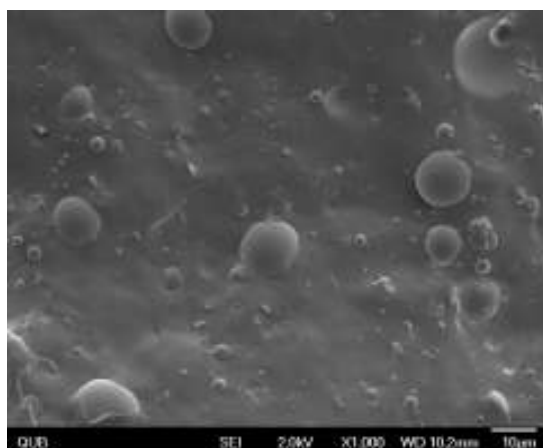
(e) 8 % w/w

Figure 5.58 SEM micrographs of 100 phr FA filled NR with various Si-264 loadings:

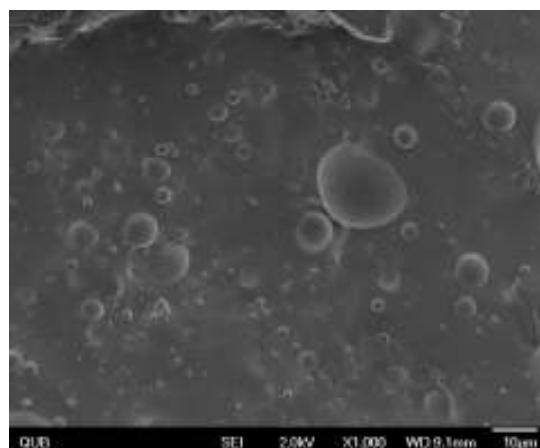
(a) 0; (b) 2; (c) 4; (d) 6 and (e) 8% w/w



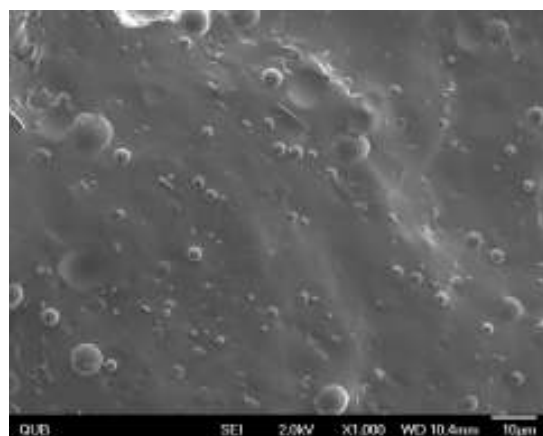
(a) 0 % w/w



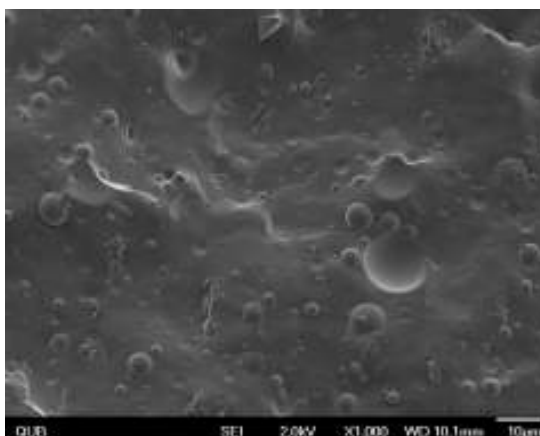
(b) 2 % w/w



(c) 4 % w/w



(d) 6 % w/w



(e) 8 % w/w

Figure 5.59 SEM micrographs of 100 phr FA filled NR with various VTEO loadings:

(a) 0; (b) 2; (c) 4; (d) 6 and (e) 8% w/w

5.3.3 Effect of Silane Treatment Techniques on Viscoelastic Properties of FA Filled NR Compounds

The purpose of this section is to investigate the silane treatment techniques affecting the viscoelastic properties of FA filled NR compounds. The FA loading was kept constant at 100 phr. Since the results of the previous section show the optimum Si-69 silane coupling agent of 4% w/w giving optimum properties for the systems filled with 100 phr FA, it is of interest to focus further on the silane treatment methods: (i) in-situ surface treatment, (ii) pre-surface treatment and (iii) ball-milling with pre-surface treatment. The FTIR technique was used to monitor the change in the surface chemistry of FA particles before and after the silane treatment. Figure 5.60 shows the comparison of FTIR results of FA with and without surface treatment. For the untreated FA, the broad peaks at wave numbers of $1,101\text{ cm}^{-1}$ and $3,434\text{ cm}^{-1}$ representing the Si-O-Si asymmetric stretching and O-H stretching respectively are observed. Upon the treatment with 4% w/w Si-69, new peaks at wave numbers of $2,974\text{--}2,887$ and $1,079\text{ cm}^{-1}$ are noticeable which are in the same region of the stretching vibration of the -CH_2 and -CH_3 groups [142]. The peak of C-O bond at wave numbers of approximately $1,080\text{ cm}^{-1}$ due to the presence of silane can be noticed, but is not sharp because of the interference by the Si-O-Si asymmetric stretching of FA. However, the stretching of alkyl group in Si-69 silane at $2,974\text{--}2,887\text{ cm}^{-1}$ suggests the successful deposition of Si-69 on the FA surfaces.

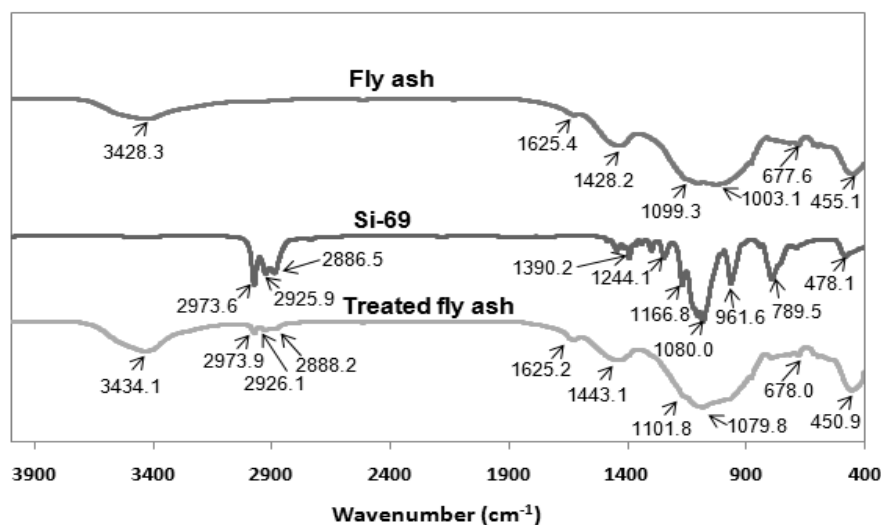


Figure 5.60 FTIR spectra of the untreated FA, the Si-69 silane coupling agent and the Si-69-treated FA in the region of 4000–400 cm^{-1}

For a preparation of milled FA, the FA was ball-milled with various milling times from 0 to 12.5 hours. Particle size of milled FA was then measured with the use of particle size analyzer. Results of particle size distribution, average particle size, specific surface area and morphology of milled FA are displayed in Figures 5.61 to 5.64 respectively. Evidently, the longer the milling time, the smaller the particle size and thus the greater the surface area particularly in the first 5 hours of milling. Beyond 5 hours, only a slight change in particle size of FA is observed. Thus, the 5 hours-milled FA was selected for further study of surface treatment with the procedure as described previously in the experimental section (section 4.4). The morphological results of the pre-surface treated FA and the milled with pre-surface treated FA is shown in Figures 5.65.

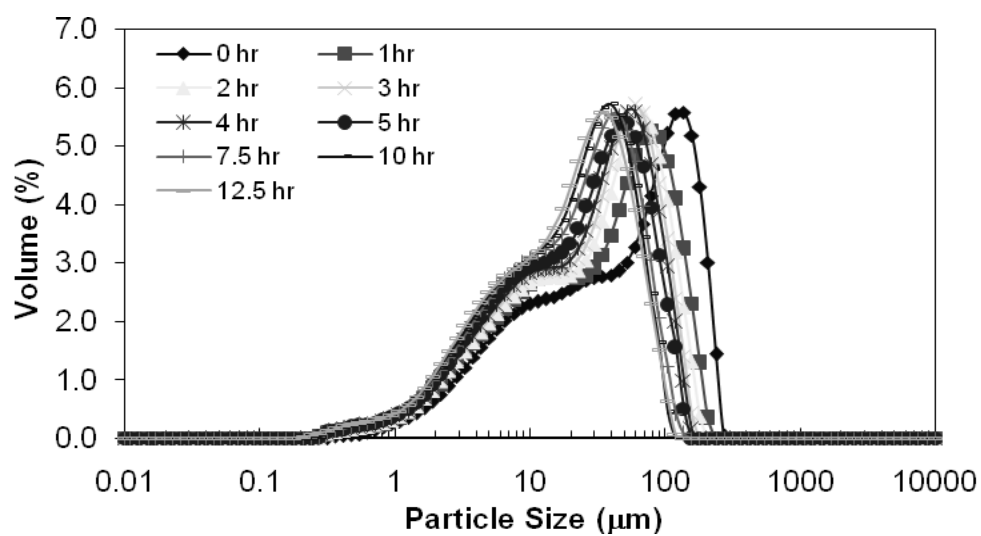


Figure 5.61 Particle size distribution of milled FA with various milling time

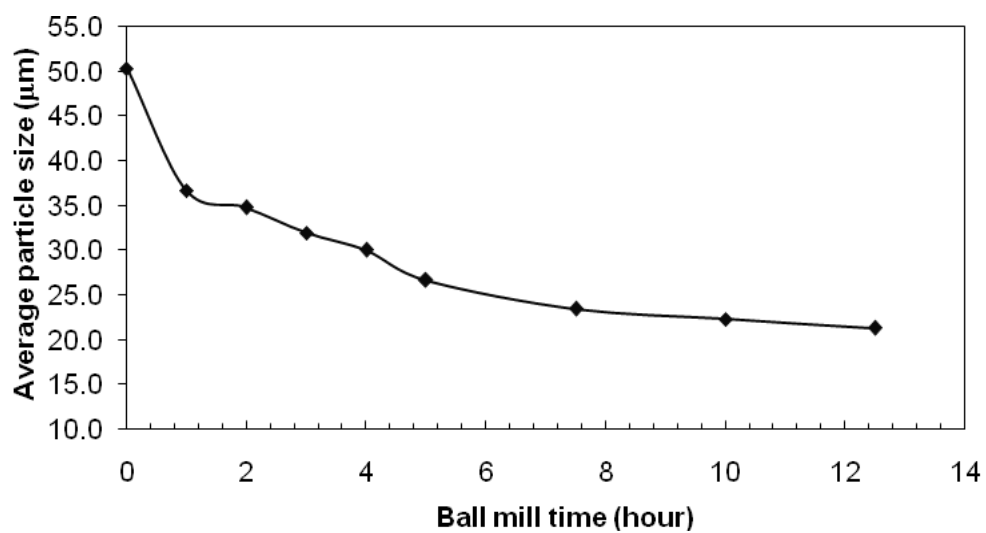


Figure 5.62 Average particle size of milled FA as a function of milling time

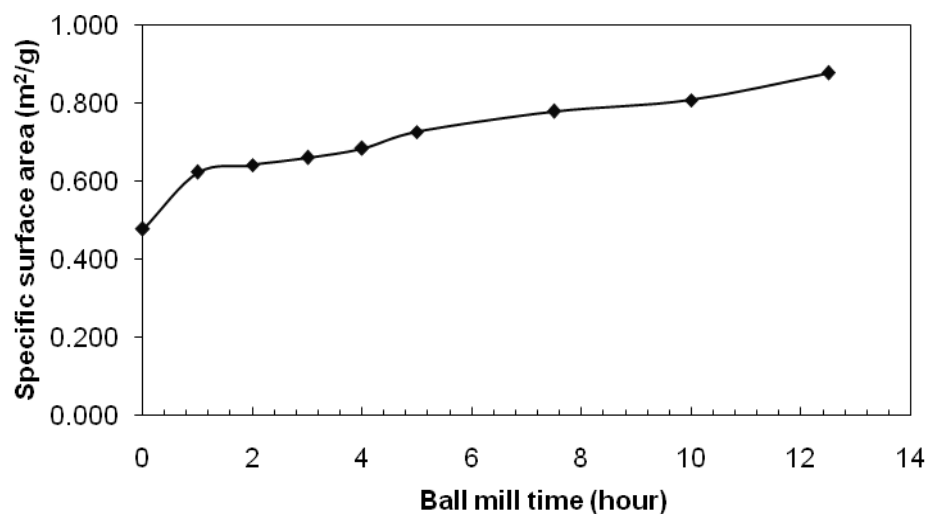


Figure 5.63 Specific surface area of milled FA as a function of milling time

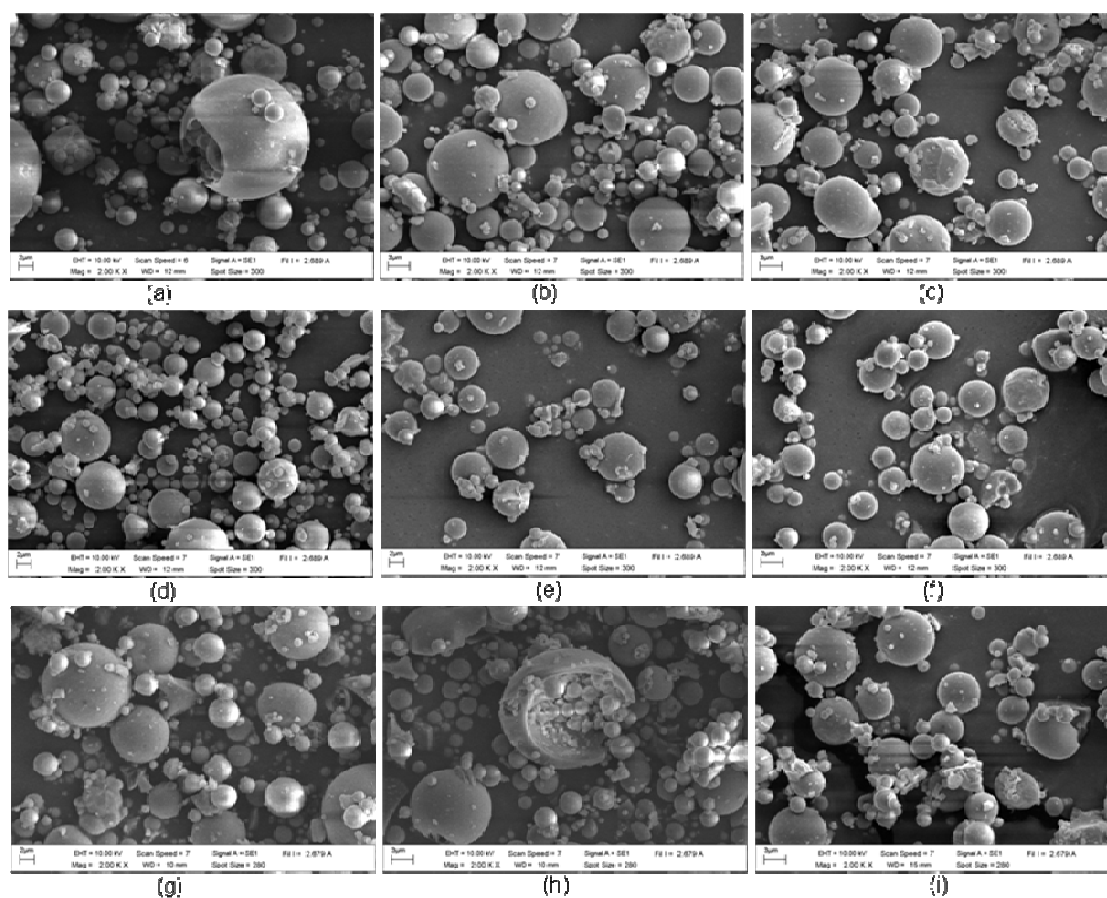


Figure 5.64 SEM micrographs of milled FA with various milling times: (a) 0; (b) 1; (c) 2; (d) 3; (e) 4; (f) 5; (g) 7.5; (h) 10 and (i) 12.5 hours

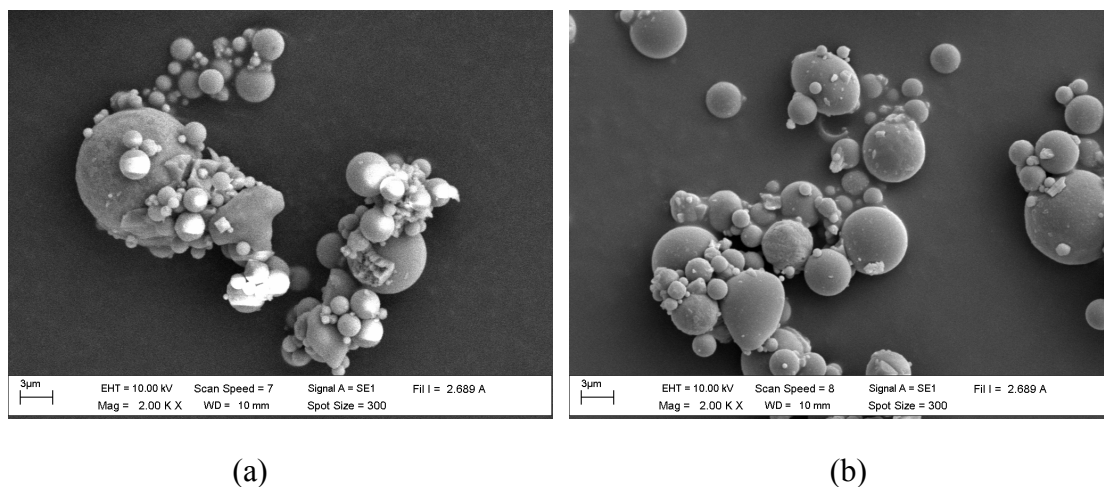


Figure 5.65 SEM micrographs of (a) pre-surface treated FA and (b) the milled with pre-surface treated FA

A) Time Sweep Test

As mentioned previously, the time sweep test was carried out in order to determine the thermal stability of compounds within the test duration of 30 minutes. It can be noticed that the G' is stable throughout the test time of 30 minutes. This implies that any change in viscoelastic results does not cause by the degradation of rubber. Obviously, from Figure 5.66, the in-situ surface treatment technique shows higher G' than other techniques whereas the untreated system demonstrates the lowest G' . It indicates that the enhancement in G' by treating FA surfaces with silane via the development of FA-NR and NR-silane interactions, and probably by the formation of FA cluster which will be discussed in detail later on.

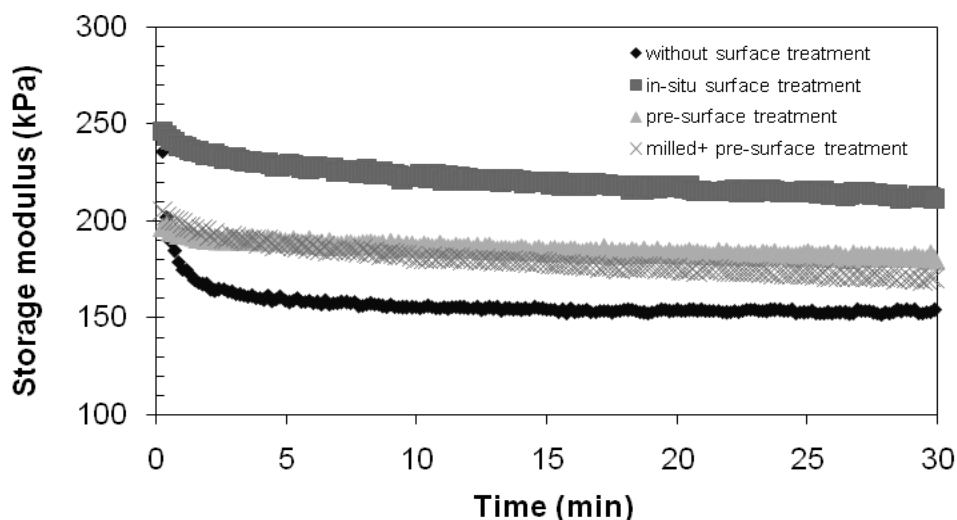


Figure 5.66 Storage modulus (G') as a function of time of 100 phr FA filled NR compounds with various treatment methods

B) Strain Sweep Test

Figure 5.67 shows the G' results as a function of strain amplitude of FA filled NR compounds with various treatment techniques. It can be seen that the in-situ surface treatment method gives slightly higher G' than other methods at low strain which agrees with the results of time sweep test. At high strain, all of treatment methods show no significant difference in the G' , but still greater than the unmodified system. This suggests that the filler-filler network (pseudo network) and rubber-filler interaction can be disrupted at high strain as noticed from the sharp drop in G' . The in-situ surface treatment method shows the higher G' than both pre-surface treatment and milled-pre surface treatment methods. The relatively high G' of in-situ surface treatment method might be caused by: (i) the interaction development between NR and FA; (ii) the formation of NR - silane linkages; and (iii) FA clusters formation in the FA/NR compounds. Obviously, from the results of Payne effect as displayed in Figure 5.68, all 3 treatment methods give higher $\Delta G'$ than that without treatment. This suggests that the rise in $\Delta G'$ in silane treatment due to the interaction improvement between NR and FA together with the formation of NR-silane linkages [143].

Also, both pre-surface treatment and milled- pre surface treatment techniques give lower $\Delta G'$ than the in-situ surface treatment method due probably to a reduction of

FA clusters formation. Figure 5.69 illustrates the experimental values of relative G' compared with calculated values from Guth-Gold equation. It is evident that the experimental values of relative G' of 3 treatment methods at 1 % strain are lower than the calculated ones. This result is caused by the molecular slippage by strain amplification and ball bearing effect as discussed previously [146]. The result of in-situ treatment method remains highest value of relative G' . By applying a large deformation of 100%, the experimental values of relative G' of all compounds decrease because of the disruption of physical interaction. Also, the 3 treatment methods still show greater relative G' at high strain than that without surface treatment. This suggests that the silane treatment with 3 treatment methods gives the combination of chemical and physical interactions. Clearly, the physical interaction dominated in the silane treatment systems agrees with the results of bound rubber content as demonstrated in Table 5.3. Normally, the bound rubber content represents the magnitude of interaction between rubber and filler. From the results, the bound rubber contents of all compounds (both with and without silane treatments) are not significantly different. This means the physical interactions between rubber and FA particle is dominated over the chemical interaction. The proposed explanation can be simplified by the model as illustrated in Figures 5.70 and 5.71. Figure 5.70 shows the FA model, it can be noticed that the FA particles are not only spherical in shape, but also show porous structure consisting of small sized particles. All of treatment FA techniques are illustrated in Figure 5.71. In the case of untreated system (see Fig. 5.71(a)), after being mixed with NR, the small FA particles are released leading to the clusters formation. However, the treatment of FA by the in-situ surface technique (see Fig. 5.71(b)) still shows the cluster formation due to the reduction of silane available for treating the FA surfaces. This is because some amount of silane is soluble in NR matrix contributing to the formation of NR-silane linkages as discussed previously. Interestingly for the case of pre-surface treatment of both FA and milled FA (see Figs. 5.71(c) and (d)), the FA surfaces are treated with silane before being mixed with NR to suppress the cluster formation. It is noticeable that in the pre-surface treatment technique (see Fig. 5.71(c)), the partial amount of FA particles inside the large FA particles are modified by the penetration of silane through the FA pores while the rest are untreated leading to the clusters formation.

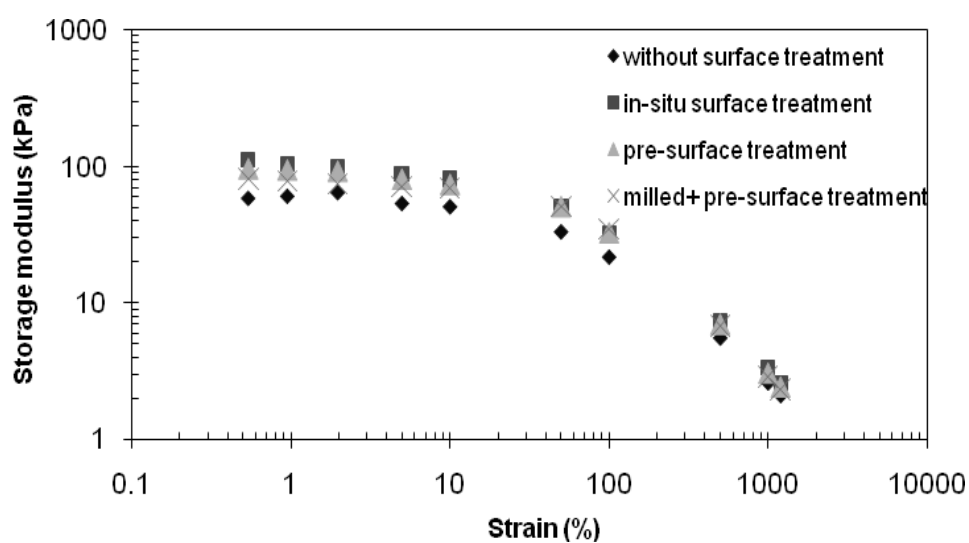


Figure 5.67 Storage modulus (G') as a function of strain amplitude of 100 phr FA filled NR compounds with various treatment methods

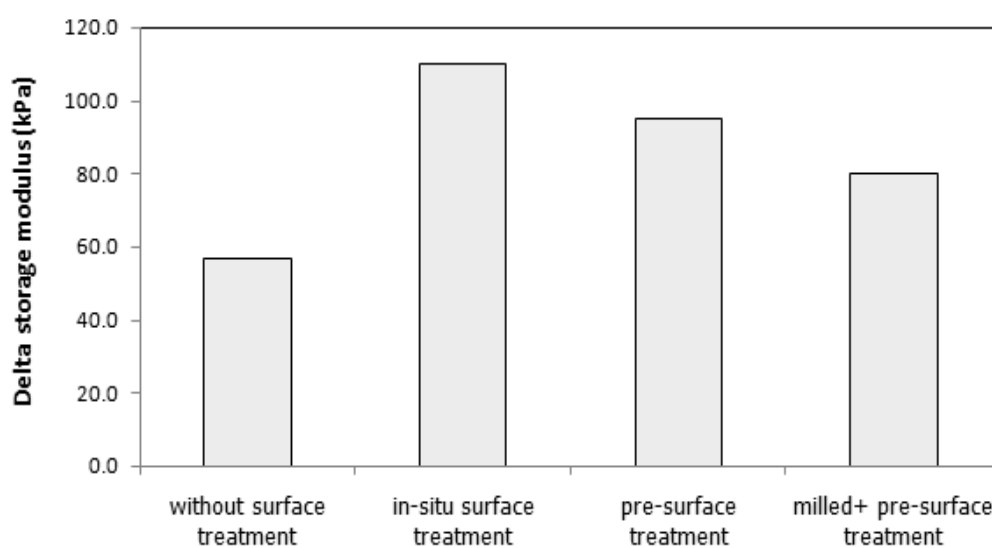


Figure 5.68 The effect of surface treatment methods of 100 phr FA filled NR compounds on the Payne effect

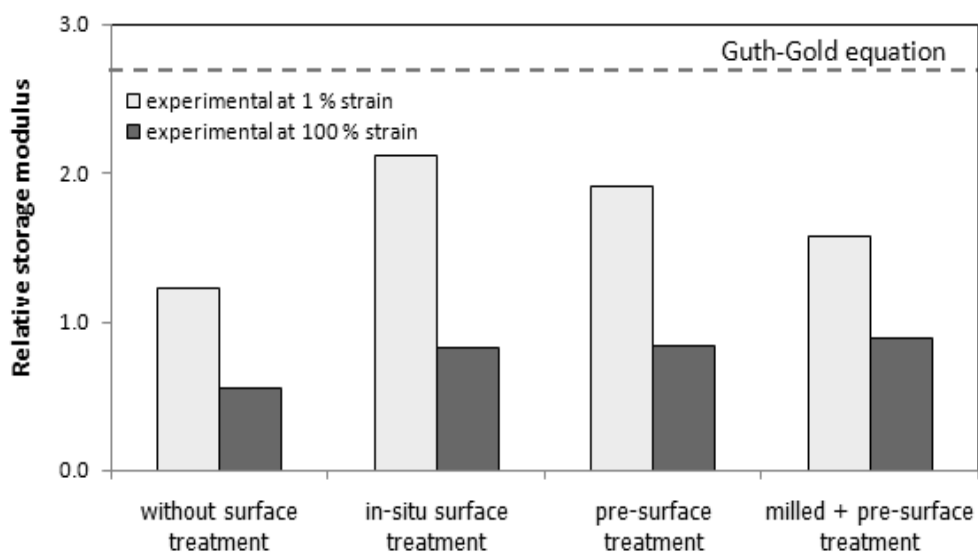


Figure 5.69 Relative storage modulus (relative G') at 1% and 100% strain of 100 phr FA filled NR compounds with various surface treatment methods using 4 % w/w Si-69

Table 5.3 Bound rubber content of 100 phr FA filled NR with various surface treatment methods

Surface treatment methods	Bound rubber content (%)
Without surface treatment	-0.5004
In-situ surface treatment	0.0509
Pre - surface treatment	0.1242
Milled + pre-surface treatment	0.1134

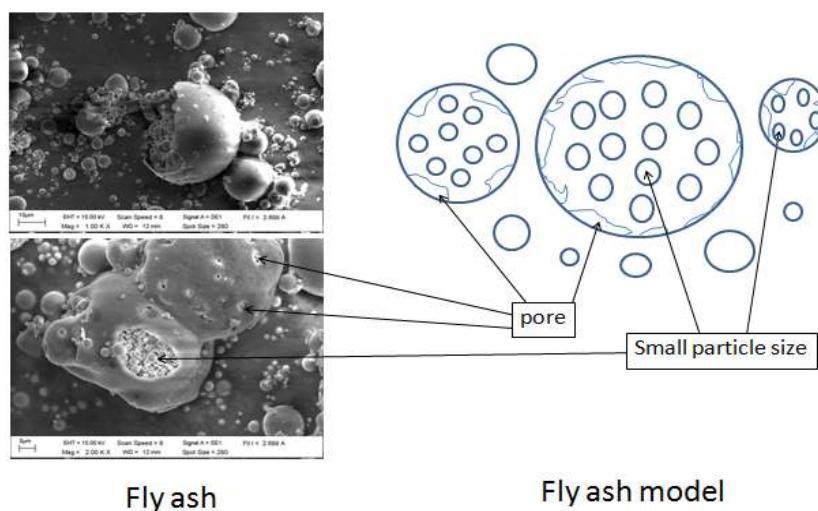


Figure 5.70 Proposed model of fly ash

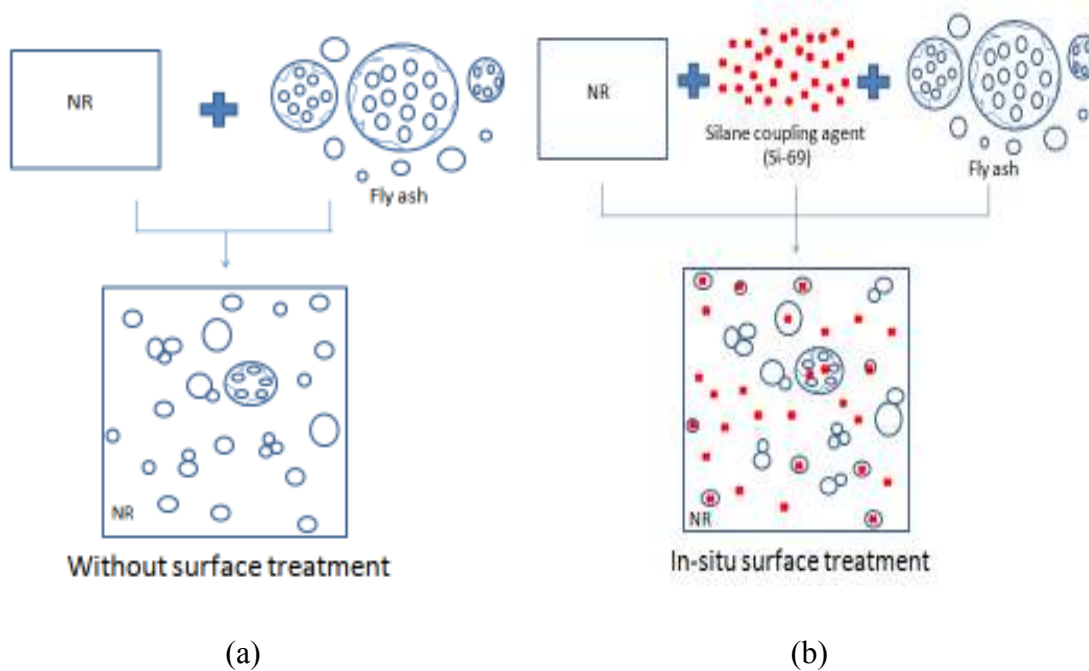


Figure 5.71 Model proposed for explaining the different actions of silane with different treatment techniques: (a) without surface treatment and (b) in-situ surface treatment

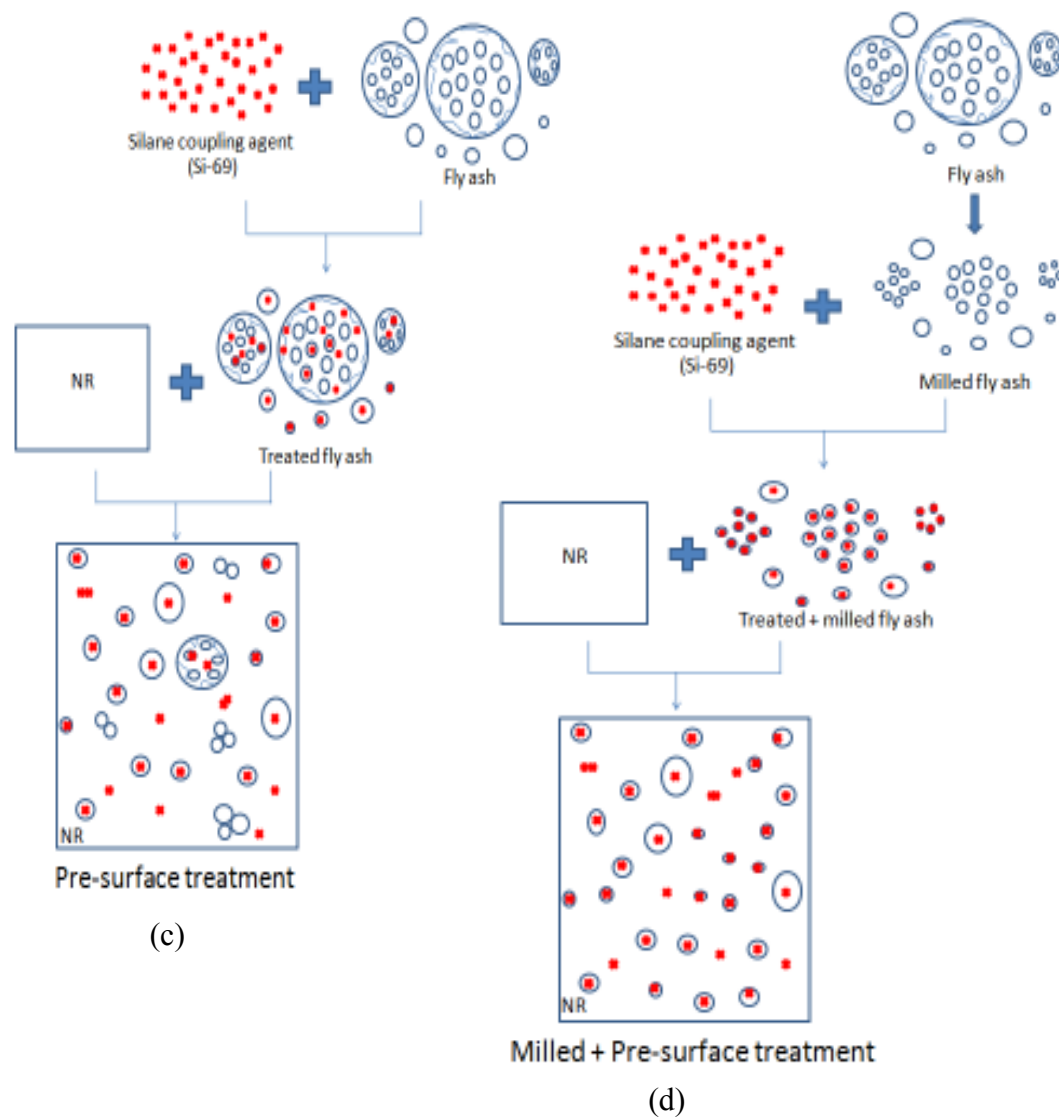


Figure 5.71 Model proposed for explaining the different actions of silane with different treatment techniques: (c) pre-treatment and (d) milled with pre-surface treatment (cont.)

C) Frequency Sweep Test

Figure 5.72 illustrates the influence of silane treatment techniques on frequency-dependent G' of FA filled NR compounds. The G' of all compounds increases with increasing frequency. This is undoubtedly because the insufficient time for molecular relaxation. Furthermore, the 3 treatments methods show the decrease in magnitude of frequency- dependent behavior as observed from the reduction in slope of the plot. The results support the formations of filler-rubber interaction and filler clusters as mentioned previously. Both formations can enhance the elastic contribution. The results agree with the damping factor results as displayed in Figure 5.73. The low values of damping factor of 3 treatment methods are observed compared with the unmodified system.

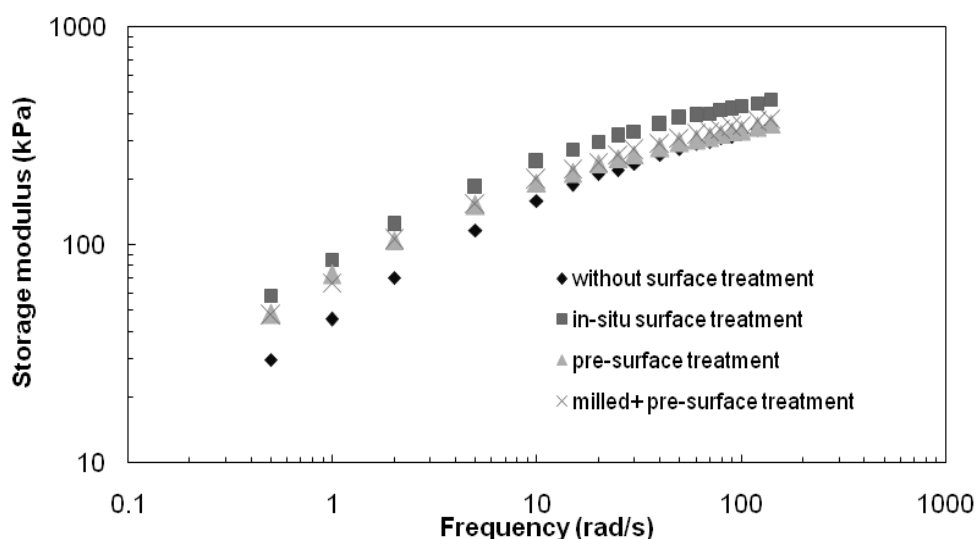


Figure 5.72 Storage modulus (G') as a function of frequency of 100 phr FA filled NR compounds with various treatment methods

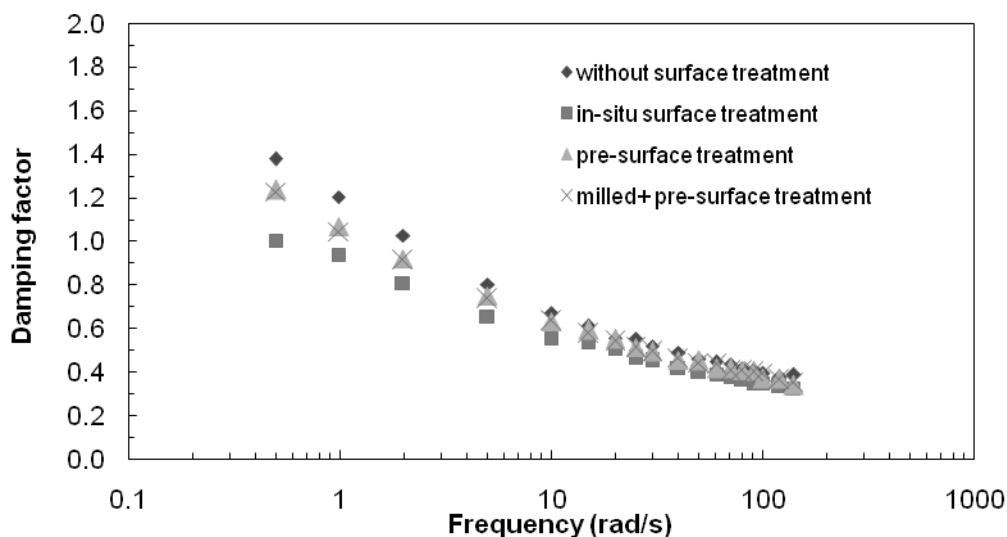


Figure 5.73 Damping factor (tan delta) as a function of frequency of 100 phr FA filled NR compound with various treatment methods

Results of complex viscosity are illustrated in Figure 5.74. The increased magnitude of rubber-filler interaction and filler-filler formation causes the increase in complex viscosity, leading to the negative deviation of power law index from unity as shown in Figure 5.75. The lowest power law index is observed in the system with milled together with pre-surface treatment because of the molecular restriction provided by the rubber-filler interaction resulting from the small FA particles containing inside large FA particles are also treated with silane thus improvement in the rubber-filler interaction.

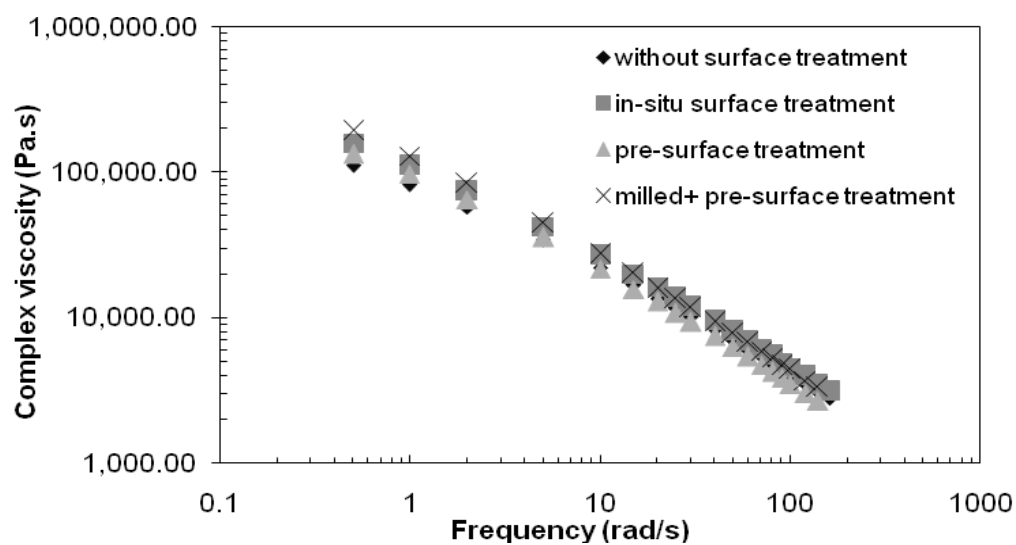


Figure 5.74 Complex viscosity as a function of frequency of 100 phr FA filled NR compounds with various treatment methods

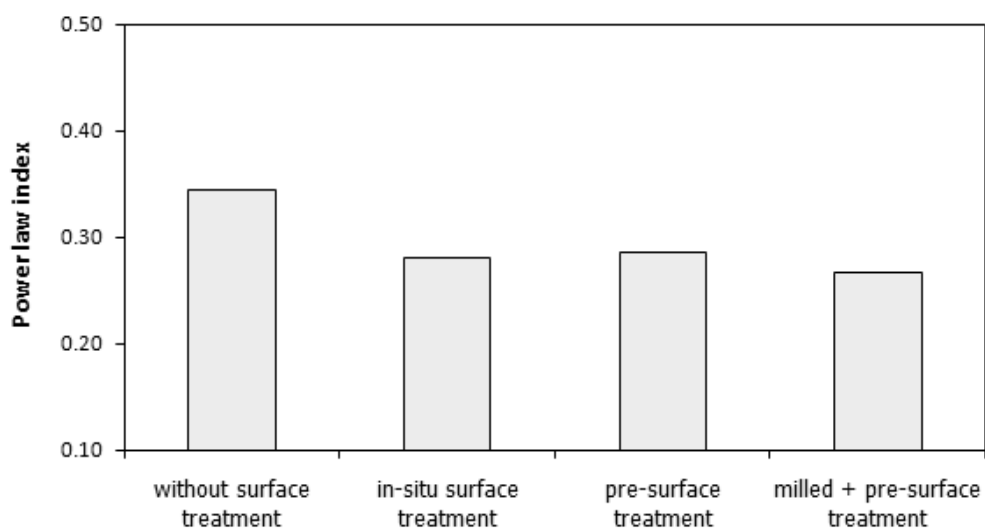


Figure 5.75 Effects of surface treatment methods of 100 phr FA filled NR compounds on power law index as measured from RPA2000

5.3.4 Effect of Silane Treatment Techniques on Morphology of FA Filled NR Compounds

The morphological results of FA filled NR compounds with various silane treatment methods are illustrated in Figure 5.76. The boundaries between FA and NR of unmodified system as exhibited in Figure 5.76(a) imply poor wettability. As illustrated in Figure 5.76 (b) to (d), the silane coupling agents provide adhesion between FA particles and NR matrix and thus the enhancement in wettability. In other words, the silane treatment with any of 3 techniques exhibit a superior interface adhesion between NR and FA particles to that of unmodified system leading to the increase in elasticity as shown in the increased G' , the decreased damping factor and power law index. Nevertheless, the bound rubber content of all compounds is not significantly different.

Referred to Figure 5.71, in the case of in-situ treatment method, few FA particles are poorly wet in rubber matrix. This is because part of silane is soluble in NR matrix and thus less amount of silane available for coupling with FA surfaces. In the system with pre-surface treatment and milled with pre-surface treatment techniques, it is clear that the FA particles embed in the rubber matrix more perfectly than the in-situ treatment method. It is probably due to the fact that almost surface FA particles are coated with silane giving rise to the improvement in wettability of FA particle in NR matrix. Anyway, milled with pre-surface treatment techniques shows greater effectiveness in treatment on FA particles than the pre-surface treatment. This is due to the small FA particles residing inside the large FA particles are coupled more with silane completely. This results in the reduction of cluster formation.

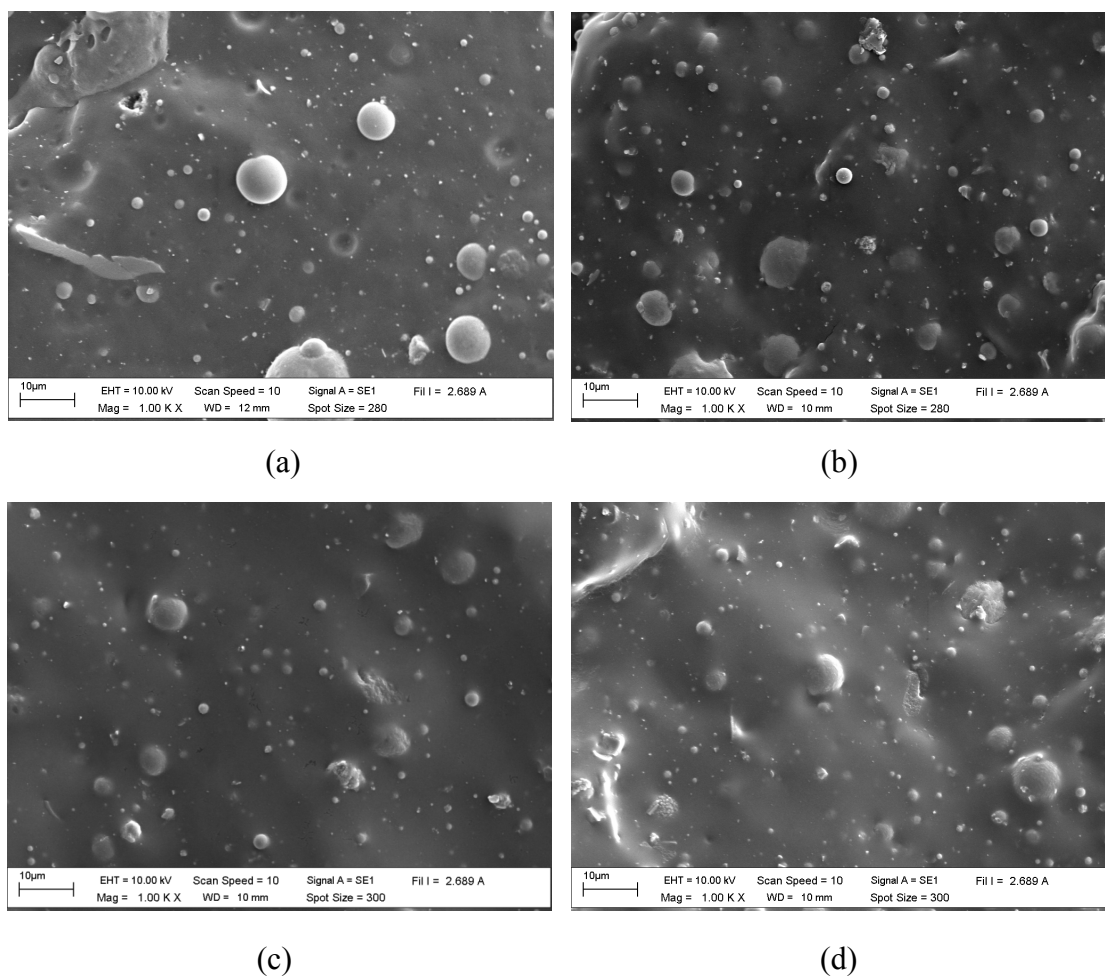


Figure 5.76 SEM micrographs of 100 phr FA filled NR with various treatment methods: (a) without surface treatment; (b) in-situ surface treatment; (c) pre-treatment and (d) milled with pre-surface treatment

5.4 Effects of Curing Systems on Viscoelastic and Mechanical

Properties of FA Filled NR Vulcanizates

This section aims to study the effects of curing systems (i.e., sulfur and peroxide curing systems) on viscoelastic and mechanical properties of FA filled NR composites. For sulfur vulcanization, properties of vulcanizates cured with conventional vulcanization (CV), efficient vulcanization (EV) and semi-efficient vulcanization (semi-EV) will be investigated and compared.

5.4.1 Unmodified FA

5.4.1.1 Cure Characteristics

Cure characteristics of FA filled NR vulcanizates with various curing systems are shown in Figures 5.77 -5.79. Obviously, cure curves of all systems display an increase in maximum torque (M_H) with increasing FA loading. As expected, the cure curves of CV system as illustrated in Figure 5.77(a) show the decrease in torque at long cure time, usually known as cure reversion phenomenon. This is because the CV system provides a large amount of polysulfidic linkages between rubber molecules having relatively low S-S bond strength, and thus poor thermal stability [143]. By contrast, the semi-EV and EV systems offer dominant monosulfidic and disulfidic linkages possessing good thermal stability. Thus, cure curves of these 2 systems show no or little reversion as exhibited in Figures 5.77(b) and (c). For peroxide curing system, the cure behavior differs from that of the sulfur vulcanization, especially in NR filled with 200 phr FA where a substantial magnitude of marching is noticeable (i.e. the rise in measured torque with cure time). Figure 5.78 demonstrates the summarized results of cure reversion as drawn from cure curves shown previously in Figure 5.77. Clearly, the CV and semi-EV systems reveal the increase in magnitude of the reversion with increasing FA loading due probably to the desulfuration and crosslinking breakdown via oxidation process accelerated by heavy metal in FA. On the other hand, the peroxide curing system exhibits the marching phenomena as shown in Figure 5.79 particularly at high FA loading greater than 150 phr. It is possible that the metal oxide in FA might accelerate the oxidation on NR

chains, leading to generated free radicals on NR chains and thus the increased rate of marching.

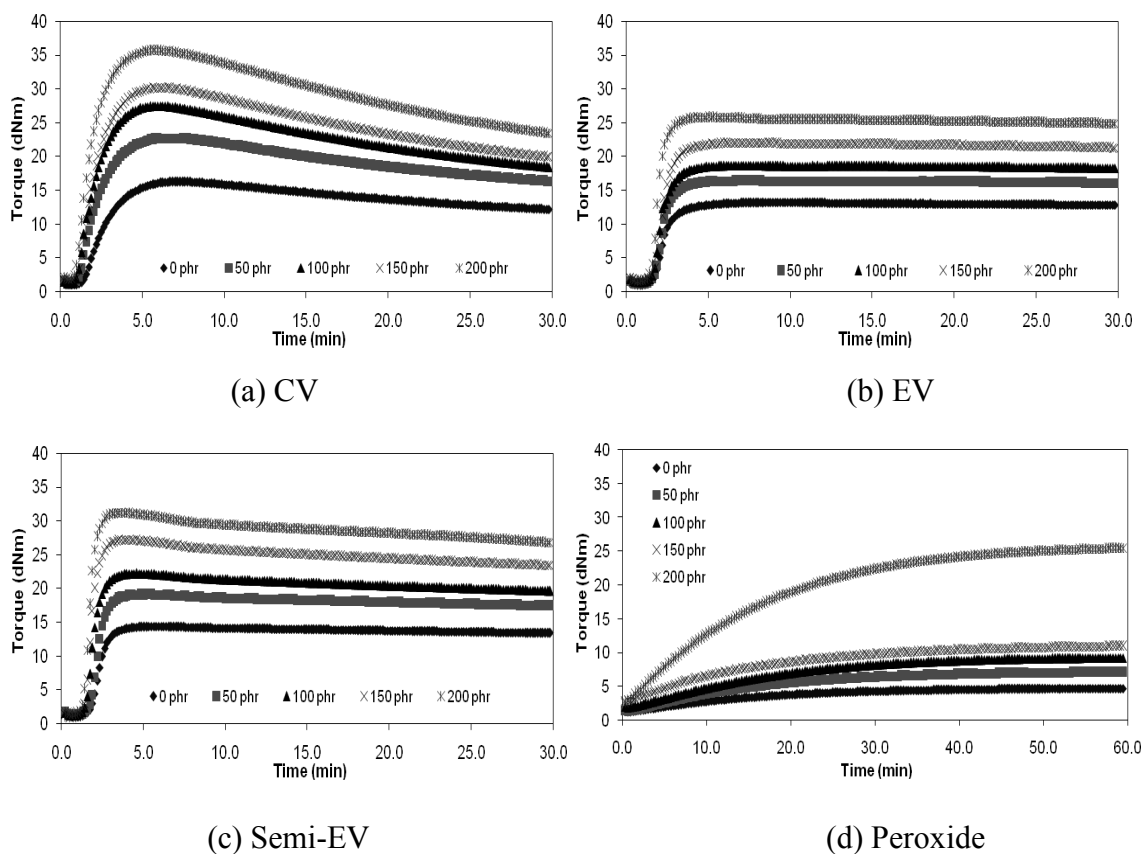


Figure 5.77 Cure curves of FA filled NR vulcanizates with various curing systems:
(a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

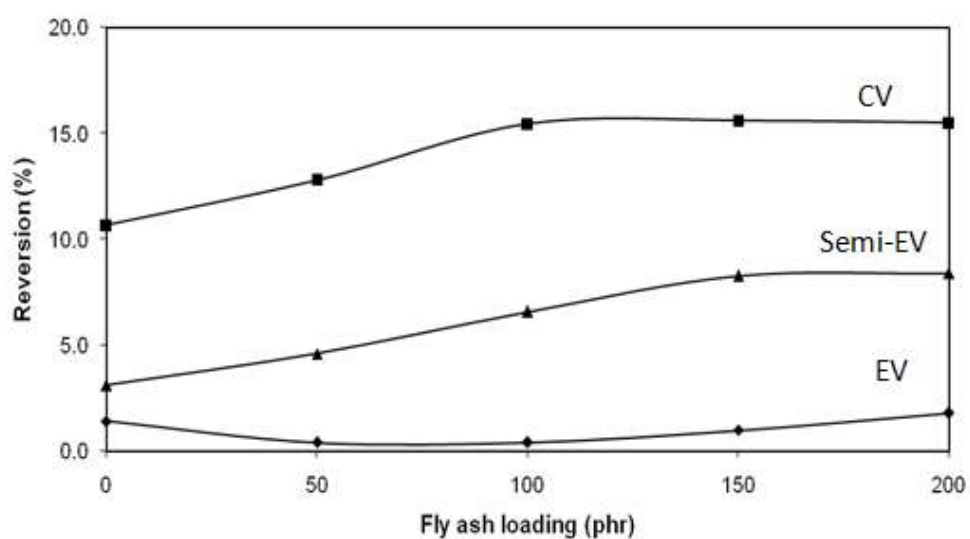


Figure 5.78 Degree of reversion of FA filled NR vulcanizates in sulfur curing systems

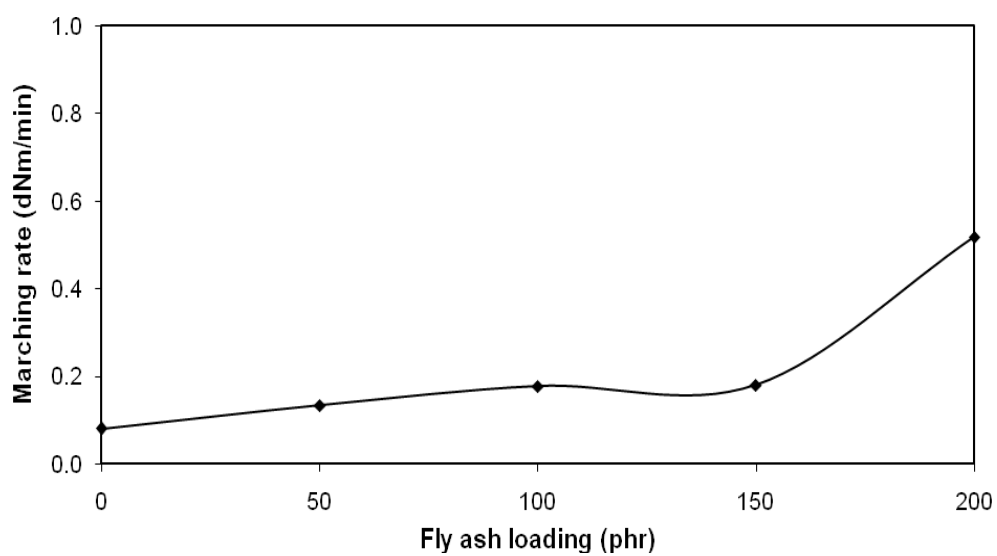
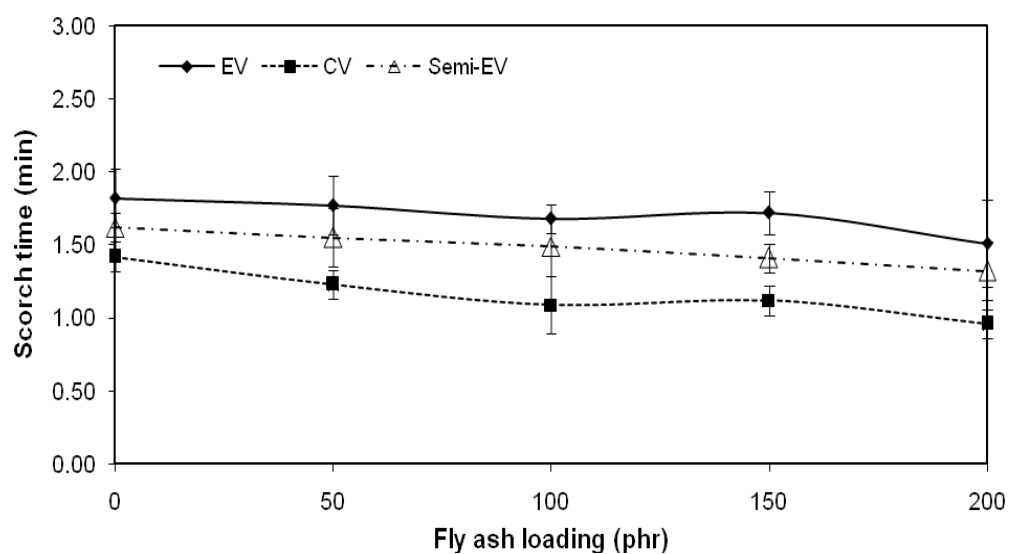


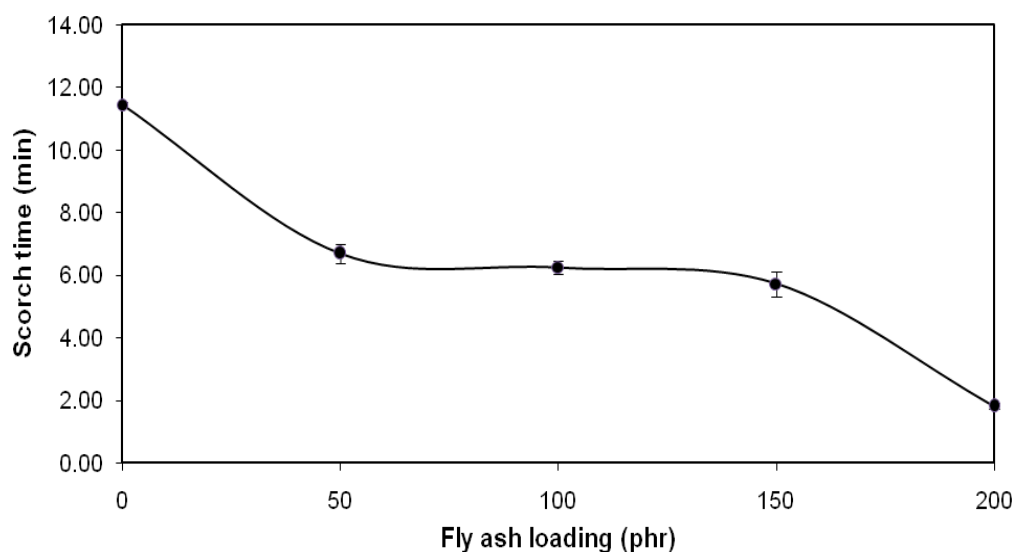
Figure 5.79 Rate of marching of FA filled NR vulcanizates in peroxide curing system

Scorch and cure times of the systems cured with sulfur are shown in Figures 5.80(a) and 5.81(a) which slightly decrease with increasing FA content. It is believed that metal oxides such as Al_2O_3 and MgO existing in FA particles (see Table 5.1) might act as cure activators and thus the cure acceleration phenomenon [3]. Unlike the sulfur curing, the peroxide curing of NR/FA demonstrates significant decrease in scorch time of compounds with no significant change in cure time as observed in Figures 5.80(b) and 5.81(b). The decrease in scorch time may be due probably to the greater thermal history during the mixing process as a result of their higher compound viscosities with higher FA loadings. In addition, the metal salt formed by heavy metal in FA and fatty acid in non-rubber component in NR might initiate a rapid peroxide decomposition [144].

Results of torque difference as an indication of crosslink density of the rubber vulcanizates are shown in Figure 5.82. It is apparent that the torque difference of the vulcanizates of all cure systems increases with increasing FA loading corresponding to the results of crosslink density determined from swelling technique as illustrated in Figure 5.83. This suggests that the FA can accelerate sulfur curing process thus the increase in crosslink density in similar manner to the peroxide curing. In addition, the sulfur cured vulcanizates with CV system have higher crosslink density than Semi-EV and EV vulcanizates, respectively, due to a large amount of free sulfur available for giving the linkages between rubber molecules.

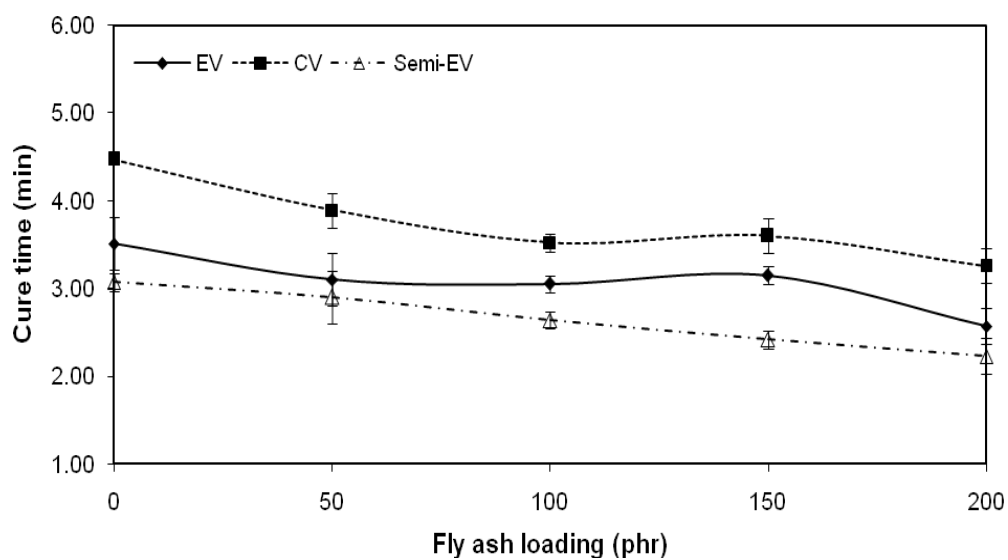


(a)

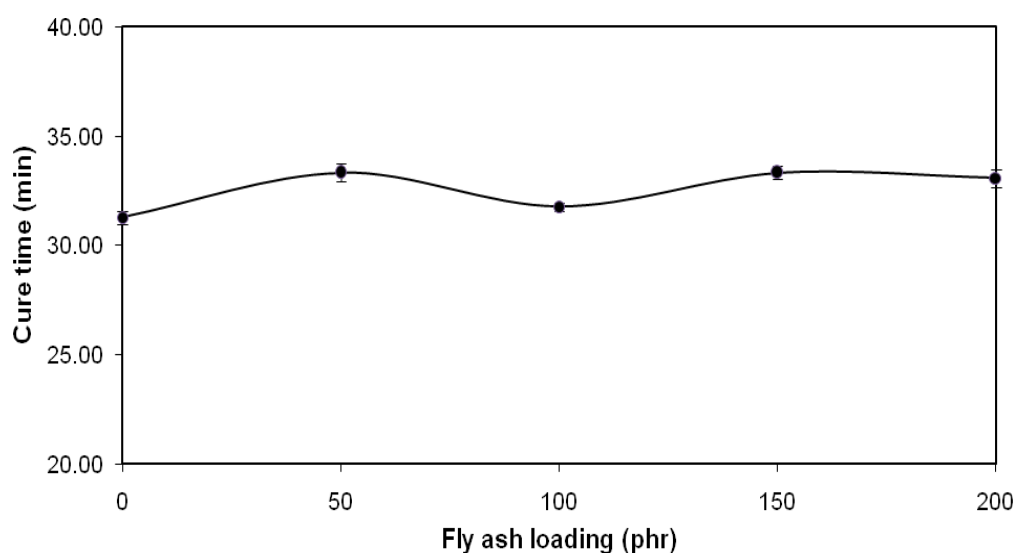


(b)

Figure 5.80 Scorch time of FA filled NR composites with various curing systems:
(a) sulfur and (b) peroxide curing systems

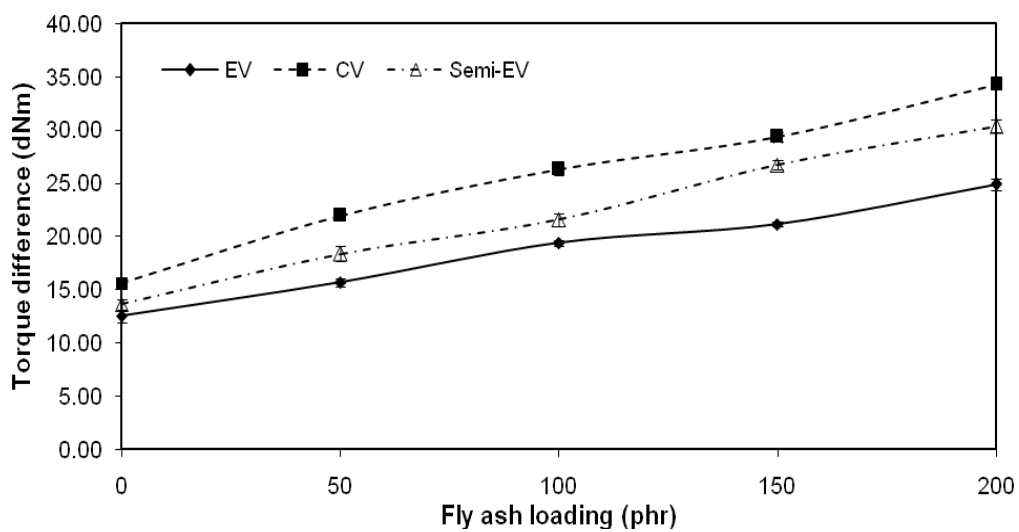


(a)

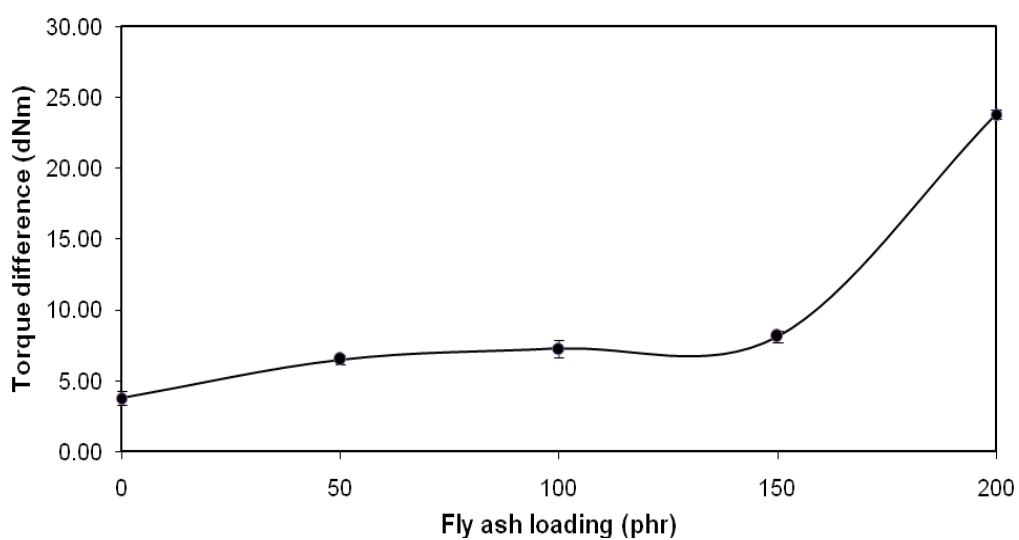


(b)

Figure 5.81 Optimum cure time of FA filled NR composites with various curing systems: (a) sulfur and (b) peroxide curing systems

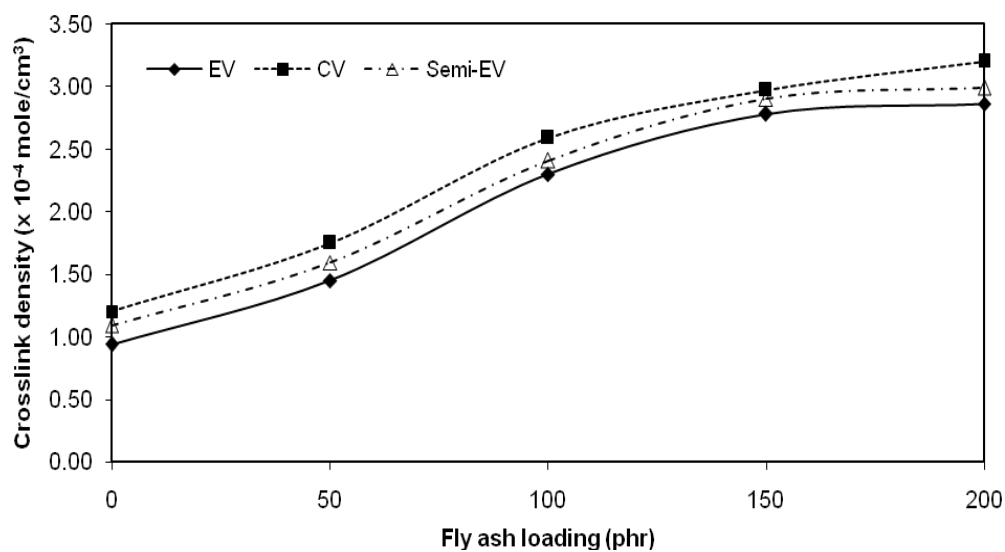


(a)

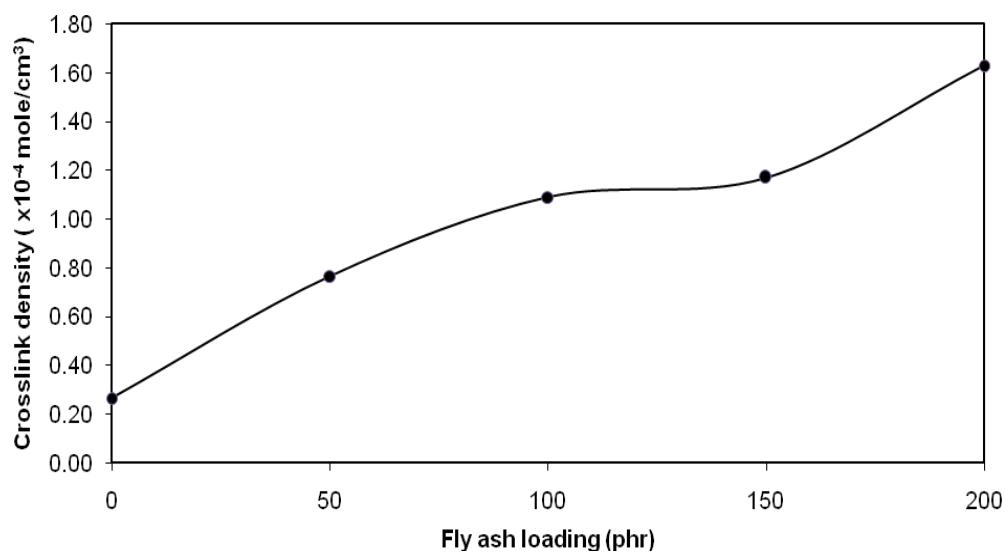


(b)

Figure 5.82 Torque difference of FA filled NR composites with various curing systems: (a) sulfur and (b) peroxide curing systems



(a)



(b)

Figure 5.83 Crosslink density (as determined from Flory-Rehner equation via swelling test) of FA filled NR composites with various curing systems: (a) sulfur and (b) peroxide curing systems

5.4.1.2 Viscoelastic Properties

(A) Time Sweep Test

As mentioned earlier, prior to the rheological tests, the thermal stability of the sample must be determined for the whole test duration with the time sweep test, and the obtained results are shown in Figure 5.84. The G' of all vulcanizates does not significantly change with test duration implying their good thermal stability. Moreover, it is evident that the G' of both sulfur and peroxide curing systems increases with increasing FA loading, due to the reinforcing effect via hydrodynamic effect and crosslink density as discussed previously.

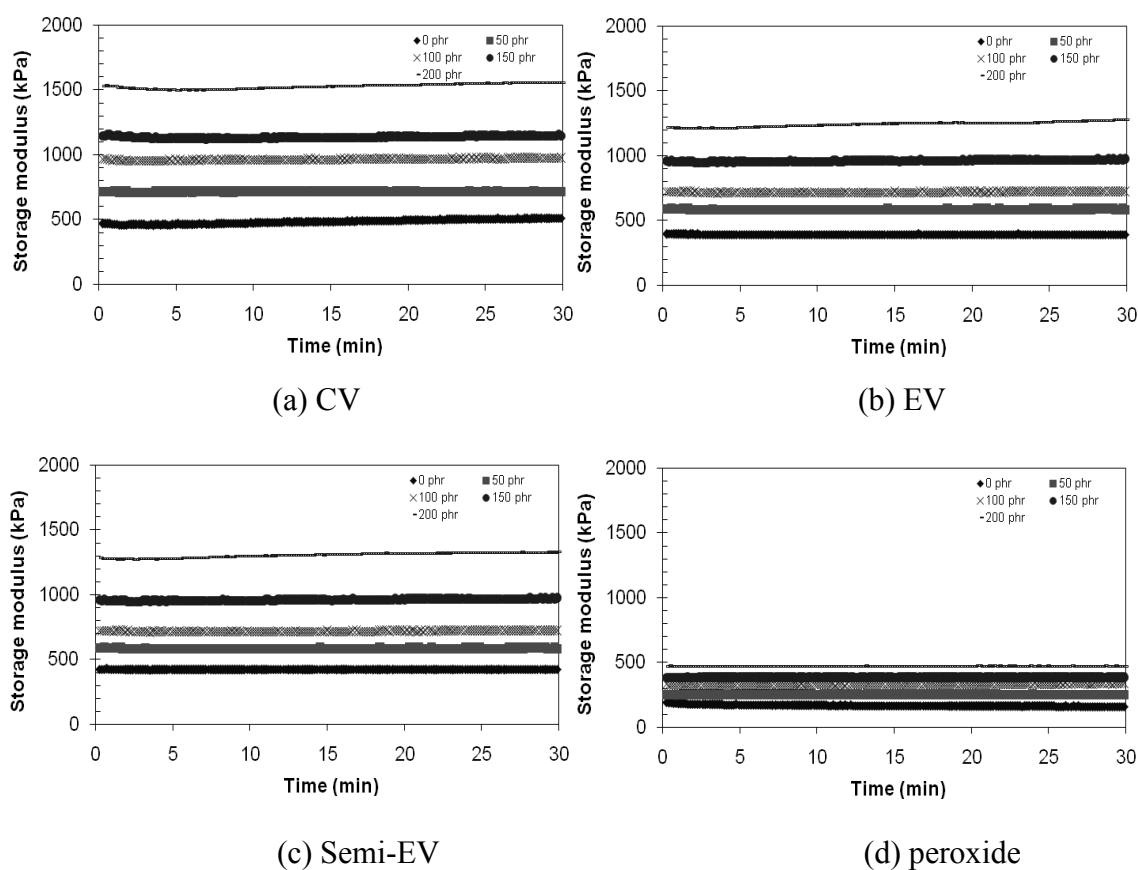


Figure 5.84 Storage modulus (G') as a function of time of cured FA filled NR vulcanizates with various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

(B-1) Uncured Systems

Figure 5.85 exhibits the storage modulus (G') as a function of deformation strain in uncured FA filled NR compounds with various curing systems. The results show that the G' at low strain of compounds in all curing systems increases with increasing FA loading resulting from the (i) hydrodynamic effect, (ii) filler-rubber interaction and (iii) filler-filler interaction. At high FA loading, FA might form FA clusters because of the silanol groups on FA surfaces via H- bonds. Nevertheless, the clusters can be disrupted at high strain as observed from the sharp drop in G' . Additionally, the discrepancies in G' at low and high strains usually known as Payne effect as shown in Figure 5.86 are found to be more apparent with increasing FA loading. Nevertheless, because of a relatively small amount of silanol groups on FA surfaces compared with silica surfaces, the interaction between FA aggregates is relatively weak leading to a small magnitude of Payne effect. Also, it is noticeable that the Payne effect of the systems cured with sulfur is significantly lower than that with peroxide. This means the FA gives relatively poor filler-filler interaction in sulfur curing system. It might be because the silanol groups on FA surfaces might absorb some polar chemicals including ZnO, leading to a decrease in magnitude of FA cluster formation thus a decrease in filler-filler interaction.

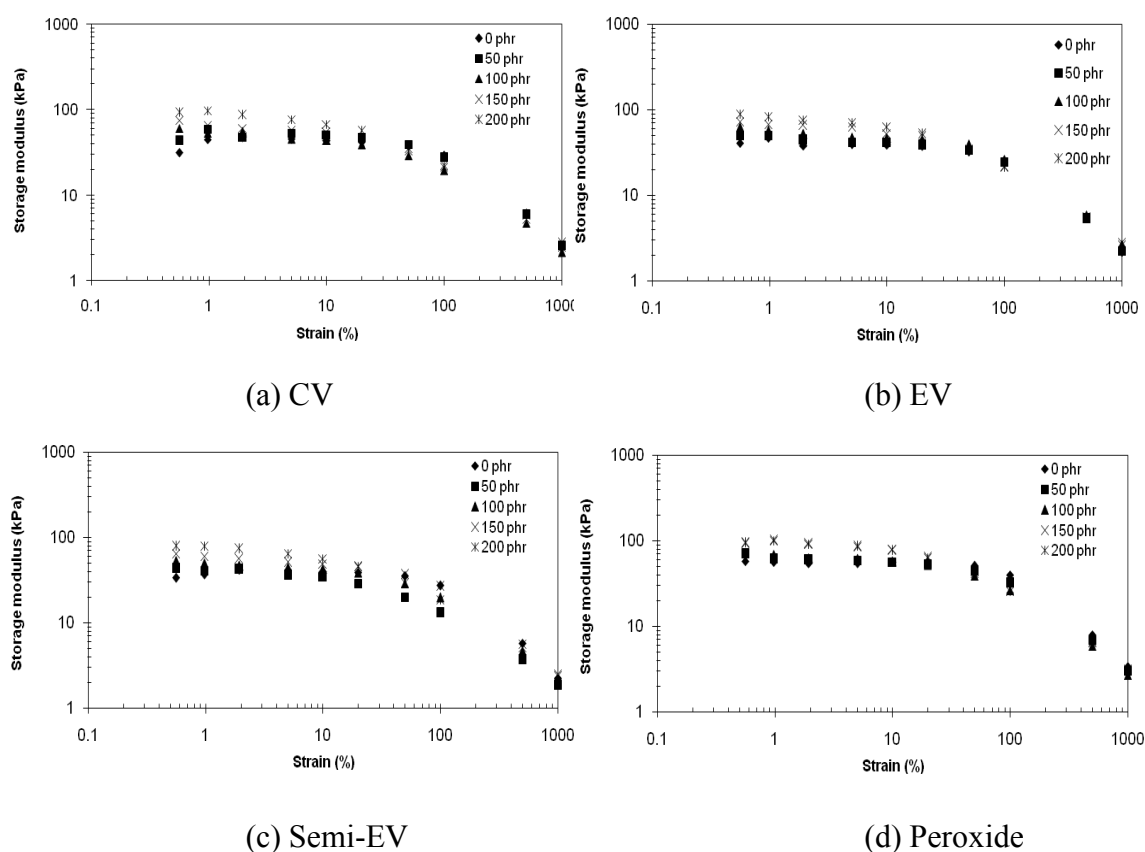


Figure 5.85 Storage modulus (G') as a function of strain amplitude of uncured FA filled NR compounds with various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

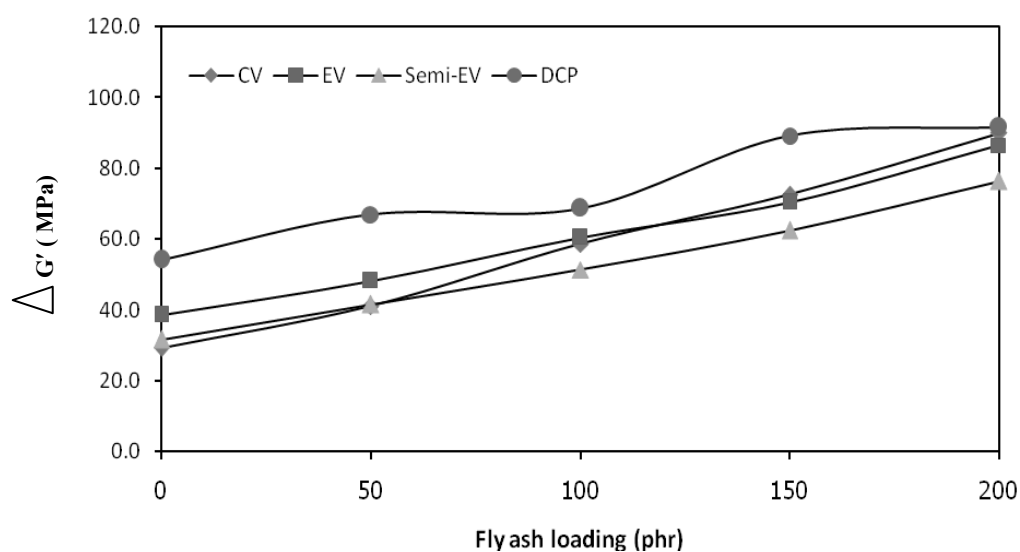


Figure 5.86 Payne effect ($\Delta G'$) of NR filled FA compounds with various curing systems

(B-2) Cured Systems

Figure 5.87 shows G' as a function of deformation strain in cured FA filled NR vulcanizates with various curing systems. It is evident that, for all systems, G' increases with increasing FA content similar to the result trends of uncured compounds. Figure 5.88 shows the plot of relative G' (the ratio of G'_{filled} and G'_{unfilled}) as a function of filler volume fraction in vulcanizates with various curing systems as determined from RPA2000, compared with that calculated from the Guth-Gold equation which excludes the filler-filler and filler-rubber interaction effects. The relative G' slightly increases with increasing FA loading in all curing systems. Nevertheless, the experimental results of relative G' show the negative deviation from that of calculated Guth-Gold data. The results imply that the molecular slippage promoted by the FA ball bearing effect in association with the low magnitude of filler-filler and/or filler-rubber interactions are dominant. At any given volume fraction of FA, the vulcanizates cured with sulfur under the CV system exhibit greatest relative modulus which might be due to their high magnitude of crosslink density, as illustrated previously.

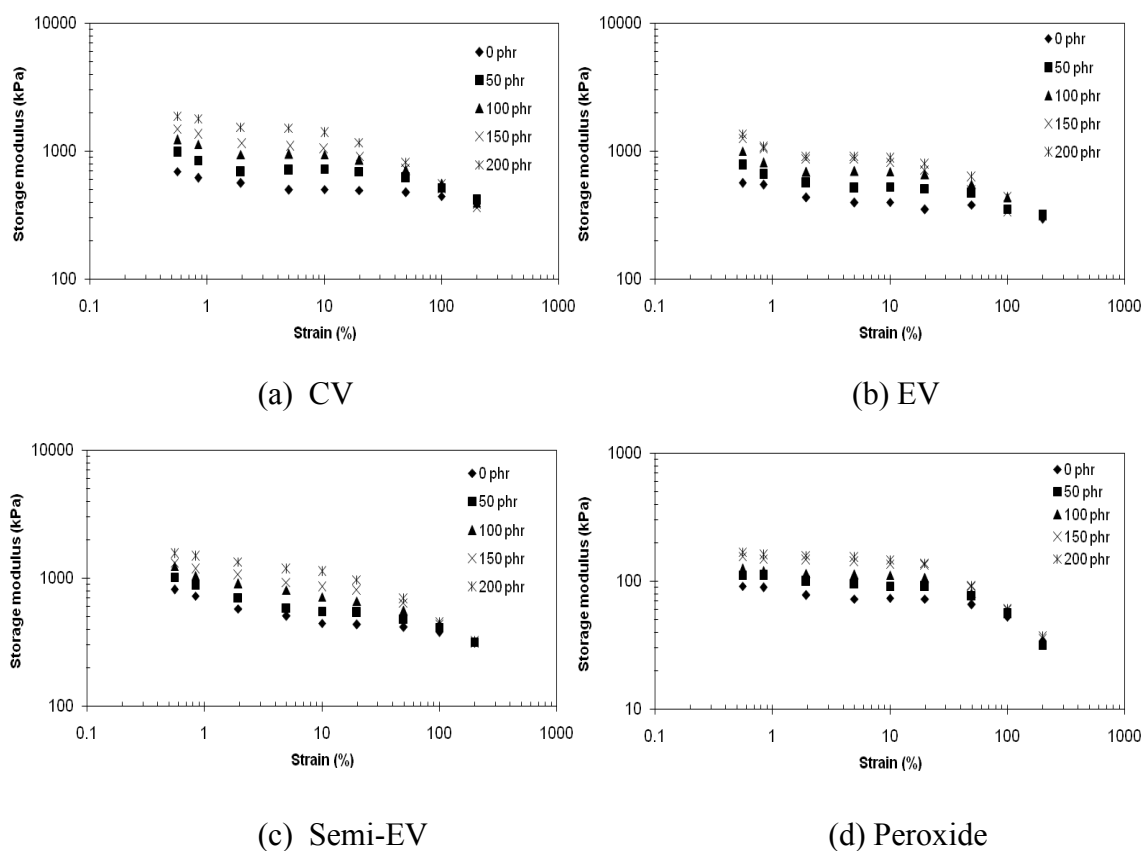


Figure 5.87 Storage modulus (G') as a function of strain amplitude of cured FA filled NR vulcanizates with various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

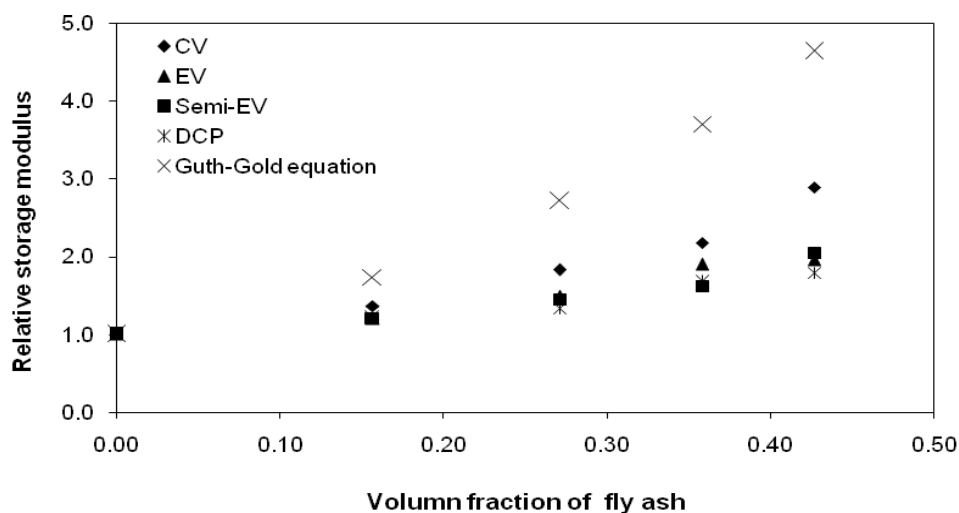


Figure 5.88 Relative storage modulus (relative G') at 1% strain of FA filled NR vulcanizates with various curing systems

(C) Frequency Sweep Test

(C-1) Uncured Systems

Results of G' as a function of frequency in FA/NR compounds with various curing systems are illustrated in Figures 5.89. The G' of all compounds increases with increasing frequency. This means all compounds exhibit strong time-dependent behavior because of the insufficient time for molecule relaxation at high frequency. Furthermore, the G' increases with increasing FA loading due to the hydrodynamic effect. Unexpectedly, both sulfur and peroxides curing systems show the increase in frequency dependence of G' (or the decrease in elastic response) with increasing FA loading as evidenced by the increase in slope of the plots in Figure 5.90. The results can be explained by the FA ball bearing effect and by the small amount of filler network and filler-rubber interaction. This explanation is supported by the result of damping factor, as shown in Figure 5.91. The damping factor increases, i.e., the decrease in elastic contribution with increasing FA loading. In addition, the processability of uncured FA filled NR compounds is investigated in terms of complex viscosity (η^*), as shown in Figure 5.92. The η^* of all compounds decreases with increasing frequency indicating a strong pseudoplastic behavior. Obviously, η^*

increases with increasing FA loading which is in good agreement with those of G' as explained by the hydrodynamic effect. Furthermore, the results of power law index (n) as illustrated in Figure 5.93 appear to be dependent on FA loading and curing systems. Power law index slightly increases with increasing FA loading referring to the increase in viscous response. Compared between sulfur and peroxide curing systems, the results show that the compounds with peroxide reveal the lower power law index than those with sulfur which is in good accordance with the results of G' and damping factor as discussed previously.

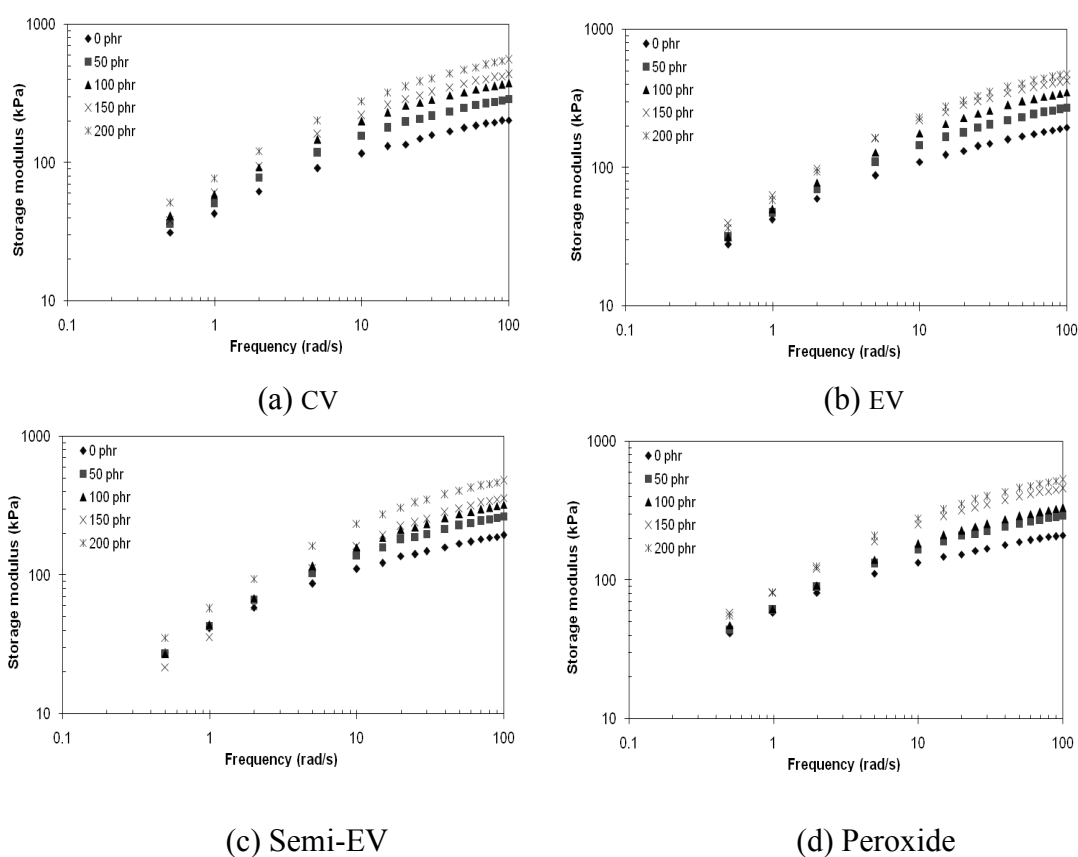


Figure 5.89 Storage modulus (G') as a function of frequency of uncured FA filled NR compounds with various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

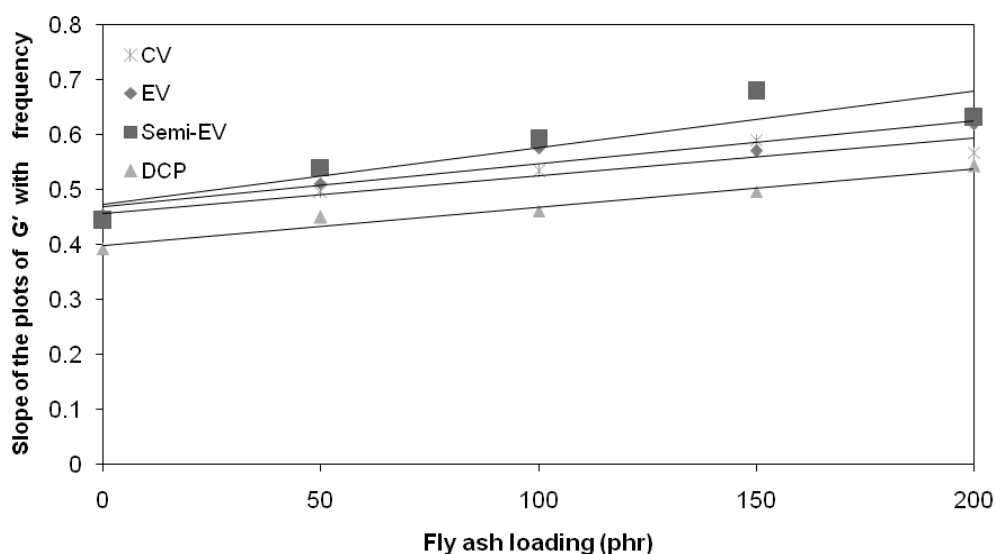


Figure 5.90 Slope calculated from the plots of storage modulus (G') as a function of frequency of uncured FA filled NR compounds with various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing system

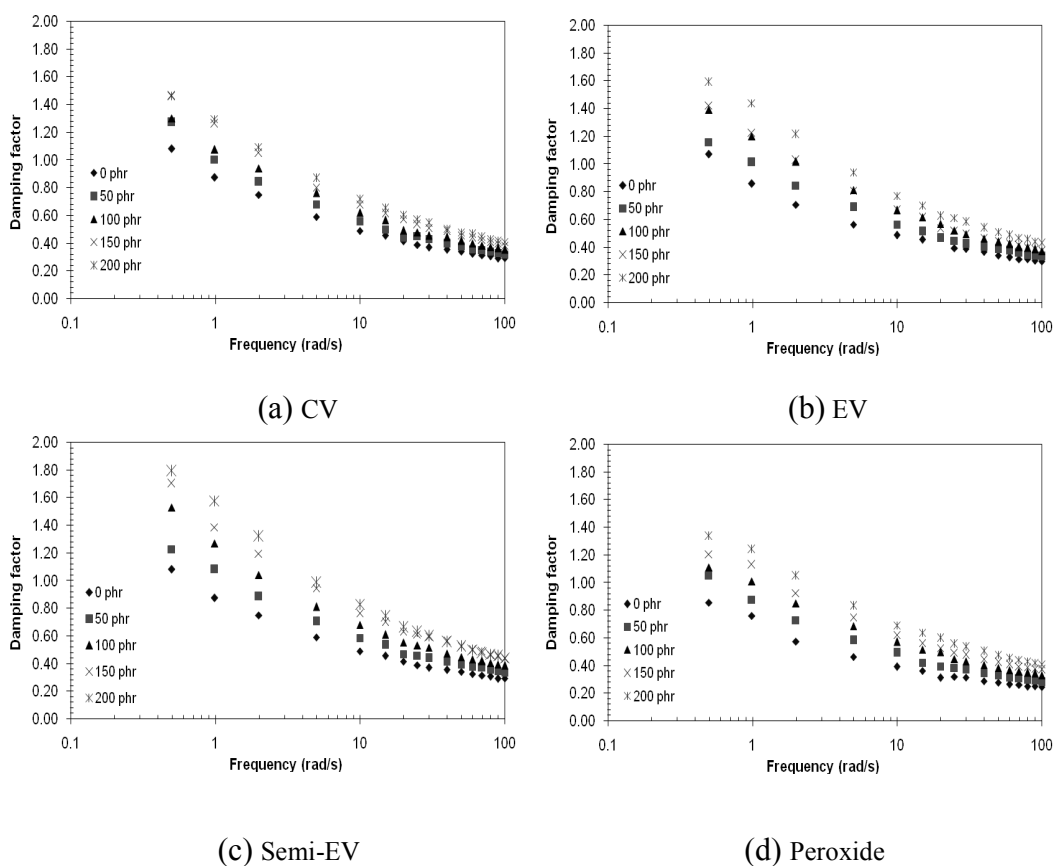


Figure 5.91 Damping factor as a function of frequency of uncured FA filled NR compounds with various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

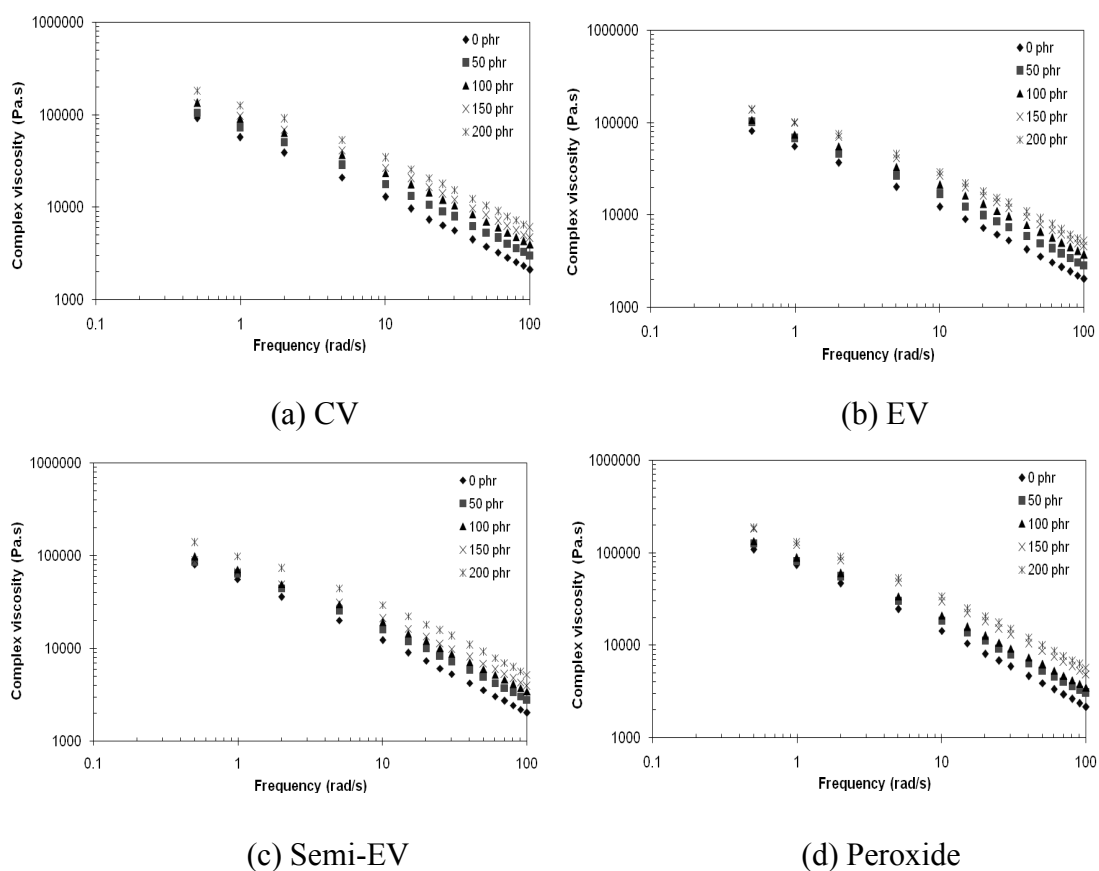


Figure 5.92 Complex viscosity as a function of frequency of uncured FA filled NR compounds with various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

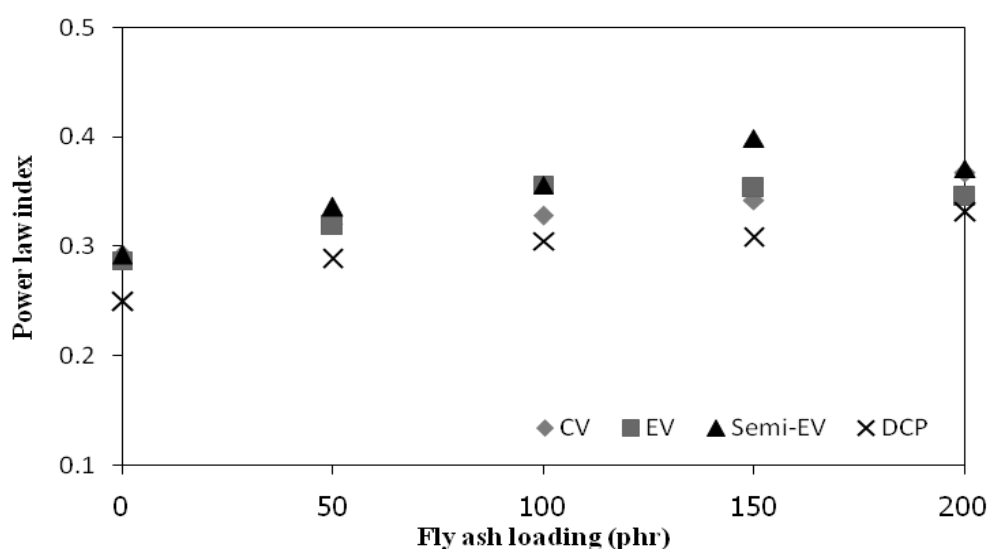
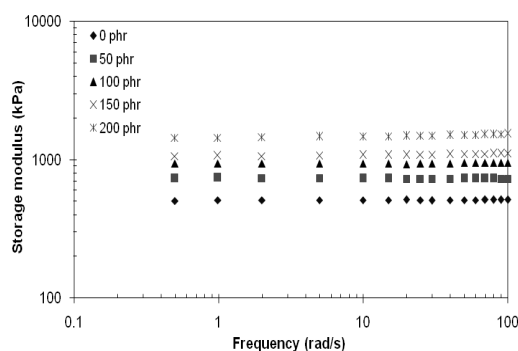


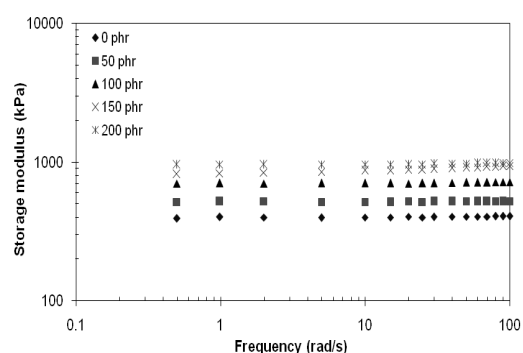
Figure 5.93 Power law index of uncured FA filled NR compounds with various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

(C-2) Cured Systems

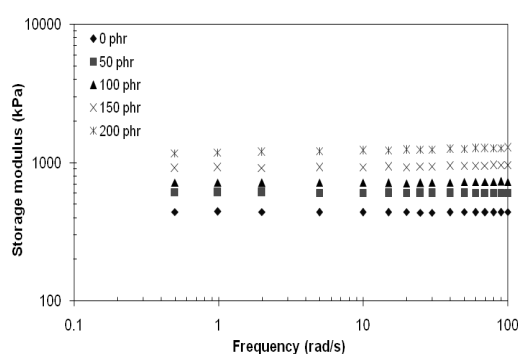
Figure 5.94 shows the influence of frequency on G' of FA/NR vulcanizates with various curing system. It can be seen that, in the sulfur system, G' is independent of frequency in the range of 0.1-100 rad/s which is a result of elastic response domination due to high density of sulfidic linkages from sulfur vulcanization. By contrast, the peroxide systems still demonstrate strong frequency dependence, i.e., the G' increases with increasing frequency indicating the dominantly viscous response. This suggests inferior dynamic properties of the system with peroxide curing system. In addition, in peroxide curing system, the free radicals on the rubber molecules required for crosslinking could be scavenged by 6-PPD as amine-based antioxidant leading to the decrease in crosslink efficiency and thus the reduced elastic component [145]. The G' results are supported by the results of damping factor as shown in Figure 5.95. The damping factor of FA/NR vulcanizates cured with sulfur is not significantly changed with frequency because of the domination of elastic response and resistance to molecular slippage as provided by sulfidic linkages. On the other hand, the damping factor of FA/NR vulcanizates cured with peroxides decreases with increasing frequency which is coincident with the relatively high damping factor. These results suggest the dominant contribution of viscous response which might be attributed to the combined effects of ball-bearing and low crosslink density, as discussed previously.



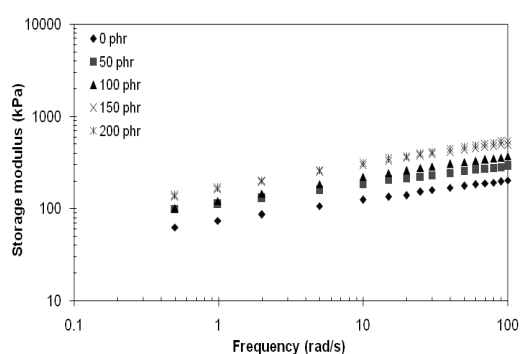
(a) CV



(b) EV



(c) Semi-EV



(d) Peroxide

Figure 5.94 Storage modulus (G') as a function of frequency of FA filled NR vulcanizates with various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

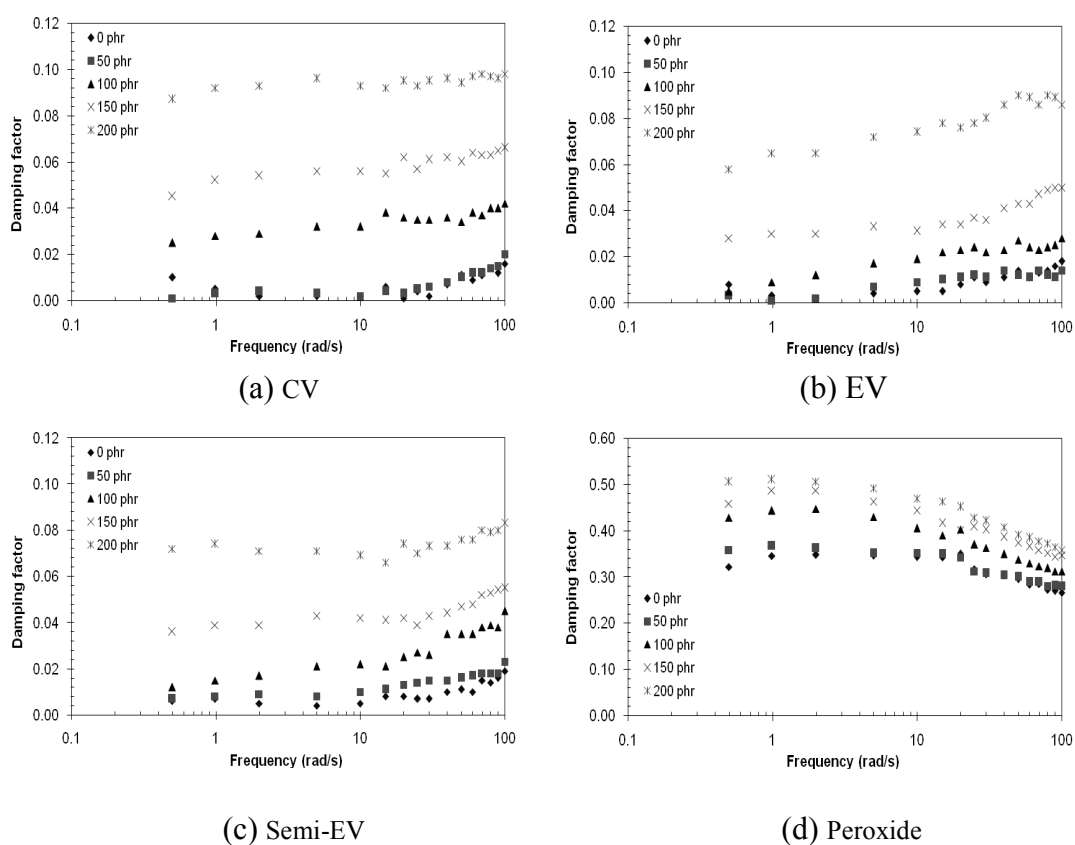


Figure 5.95 Damping factor (tan delta) as a function of frequency of FA filled NR vulcanizates with various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing

(D) Thermal Stability Test

In this study, the vulcanizates were thermally aged in RPA2000 test cavity at 120 °C for 1 hour, and the viscoelastic properties of vulcanizates were measured before and after ageing using RPA2000 at 70°C. Results before and after thermal ageing are reported in the terms of G' retention as illustrated in Figure 5.96. For all curing systems, the G' retention is independent of FA loading. Also, the FA/NR vulcanizates cured with peroxide reveal superior G' retention to those with sulfur indicating the more thermal stability. Among sulfur curing systems, the G' retention of vulcanizates cured with CV, EV and Semi-EV systems is not significantly different. Notably, all curing systems yield the G' retention above 100 % implying the development of additional crosslink network during the ageing treatment or the so-called post curing phenomenon. Compared with sulfur vulcanization, the peroxide-cured specimens exhibit greater magnitude of post-curing effect.

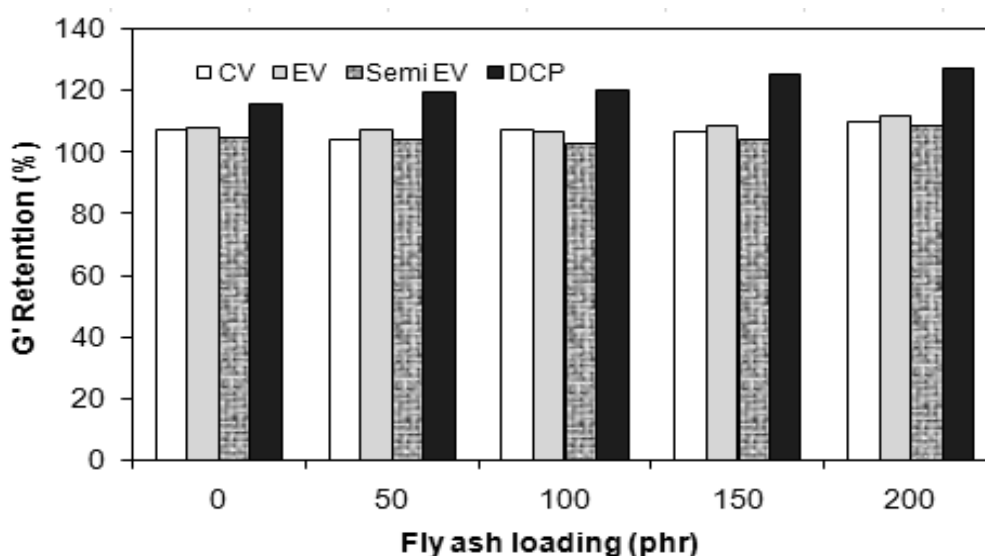


Figure 5.96 Retention of storage modulus (G') of FA filled NR vulcanizates with various curing systems as measured from RPA

5.4.1.3 Tensile Properties

Figure 5.97 represents the plot of M100 with FA loading. It is evident that the M100 of all vulcanizates slightly increases with increasing FA loading. Generally, M100 depends strongly on crosslink density and filler reinforcing effect. For the crosslink density effect, it is known that the higher the crosslink density, the greater the M100. The results of crosslink density as depicted in Figure 5.83 demonstrate the increase in crosslink density with increasing FA loading. Therefore, the increase in M100 is caused partly by the increased crosslink density. In the case of filler reinforcing effect, as discussed previously, the reinforcement of FA should be controlled by the hydrodynamic effect rather than filler-rubber interaction and filler-filler interaction due to the small amount of silanol groups on FA surfaces and the large particle size of FA. This implies that the enhancement in M100 is the combined results of crosslink density and hydrodynamic effect. Compared between 2 curing systems, the FA/NR vulcanizates cured with peroxide yield inferior M100 to those cured with sulfur because of its lower magnitude of crosslink. Elongation at break of all vulcanizates as shown in Figure 5.98 do not significantly change with increasing FA loading.

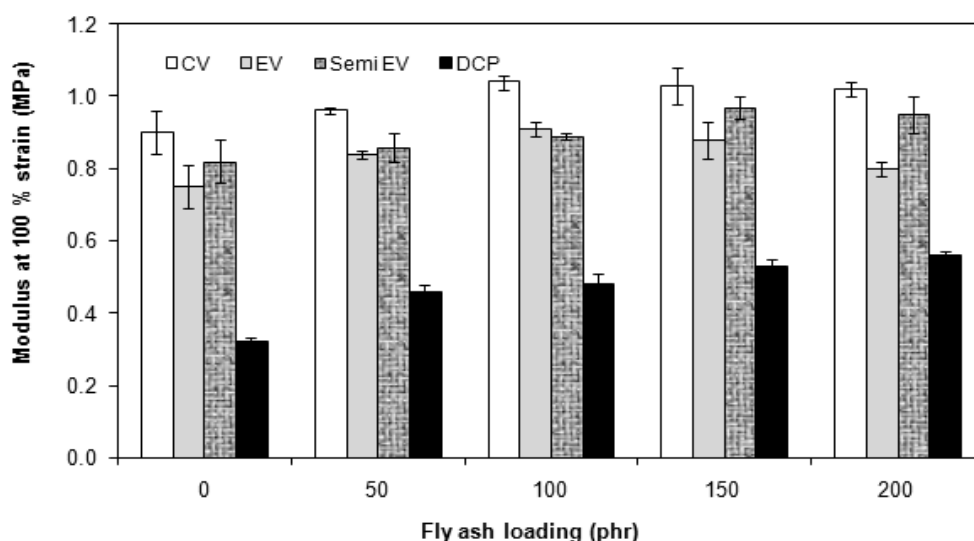


Figure 5.97 Modulus at 100 % strain (M100) of FA filled NR vulcanizates with various curing systems

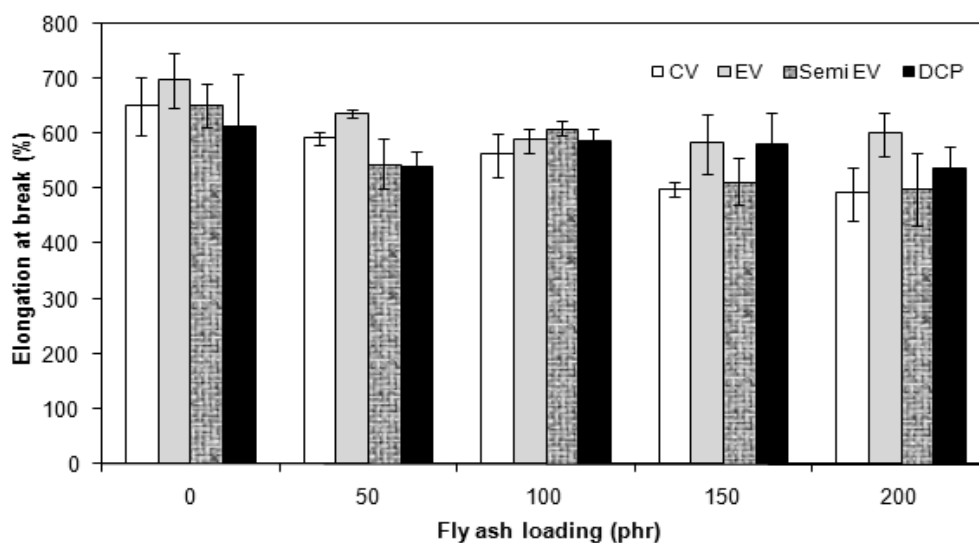


Figure 5.98 Elongation at break of FA filled NR vulcanizates with various curing systems

Figure 5.99 exhibits the tensile strength of the vulcanizates. As expected, the rubber vulcanizates filled with FA show progressive decrease in tensile strength with increasing FA loading. The decreased tensile strength is due to poor filler-rubber interactions as evidenced by the small value of bound rubber content as shown in Table 5.4. Tensile strength in sulfur-cured vulcanizates does not depend strongly on the system of sulfur curing. As expected, the sulfur-cured vulcanizates still yield higher tensile strength than the peroxide-cured vulcanizates. It is known that the polysulfidic bonds are labile, i.e., they could disrupt and slip under the localized high-stress concentrations, thereby relieving the stress, and allow the network as a whole to bear more load [146].

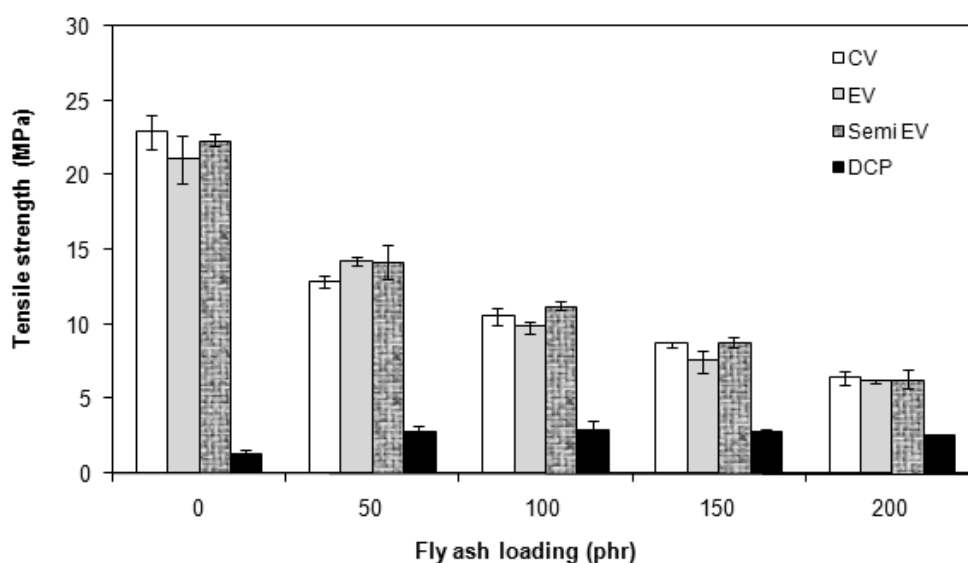


Figure 5.99 Tensile strength of FA filled NR vulcanizates with various curing systems

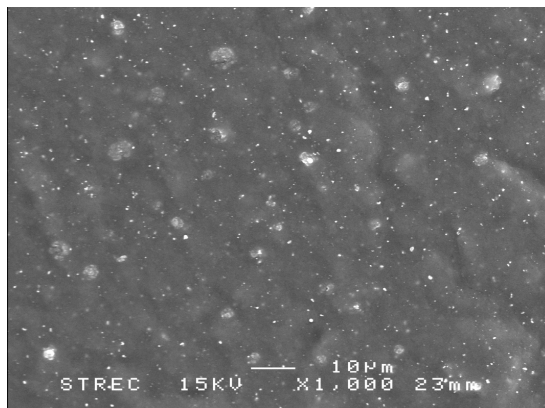
Table 5.4 Bound rubber content of FA filled NR compounds with various curing systems

FA loading (phr)	Bound rubber content (%)			
	CV	EV	Semi-EV	DCP
0	0.0185	0.0277	0.0190	0.0188
50	-0.0103	0.0324	0.0167	-0.0145
100	0.0509	-0.0530	0.0280	0.0142
150	-0.0373	0.0352	-0.0181	0.0388
200	0.0405	0.0269	0.0212	0.0266

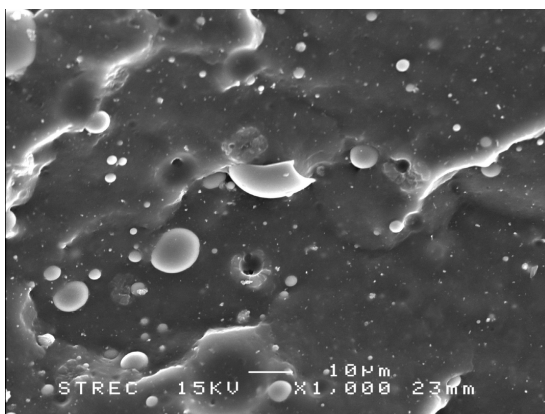
5.4.1.4 Morphological Results

Morphological results of FA filled NR vulcanizates with sulfur and peroxide, are illustrated in Figures 5.100 to 5.103. It can be observed that the interfaces between FA particles and NR matrix are not homogeneous as observed by some voids between FA particles and the rubber matrix, indicating the poor wettability of FA by NR molecules. Also, because the FA particles are porous, the rubber molecules can reside in the FA voids. This means the rubber matrix possesses less continuity and thus the decrease in tensile properties [147]. Compared with peroxide curing system, the dispersion of FA in NR under sulfur system is superior. It is suggested that the reduction in cluster formation and reformation of FA particles

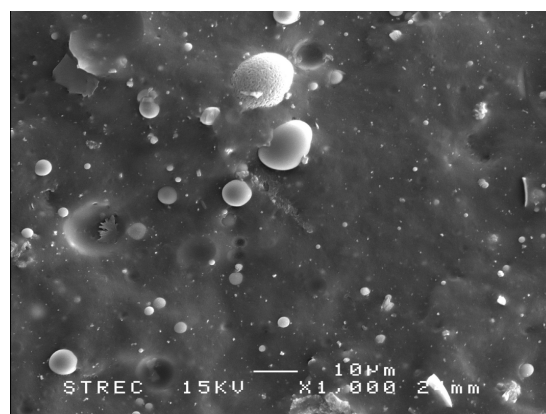
take place via the adsorption of polar ingredients in sulfur curing, e.g. ZnO, on FA surfaces [48].



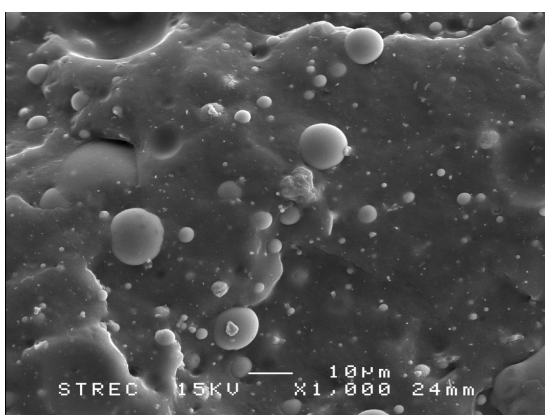
(a)



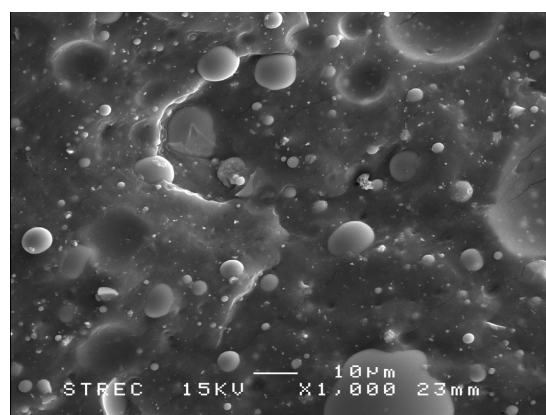
(b)



(c)

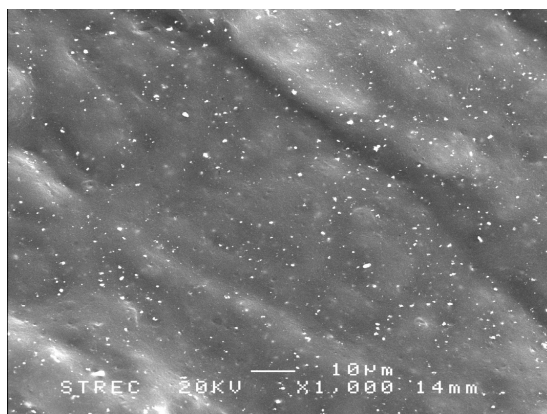


(d)

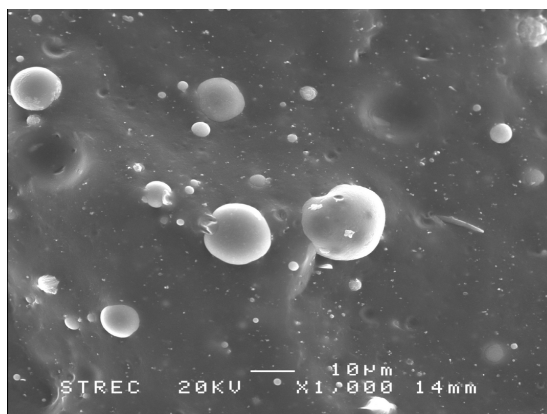


(e)

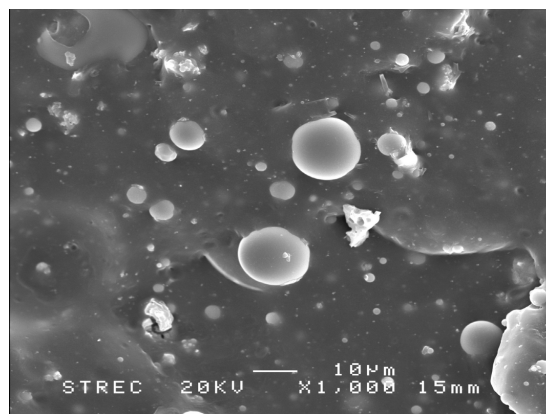
Figure 5.100 SEM micrographs of FA filled NR vulcanizates cured via CV system with various FA loadings: (a) 0; (b) 50; (c) 100; (d) 150 and (e) 200 phr



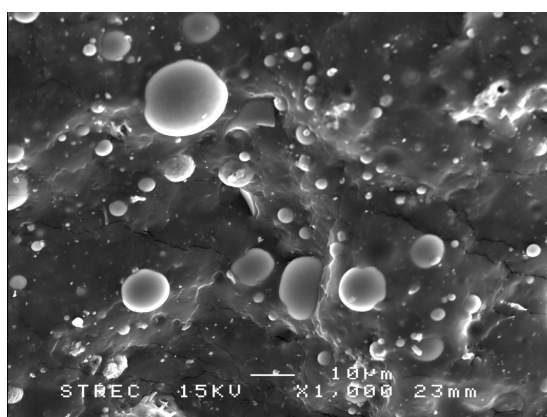
(a)



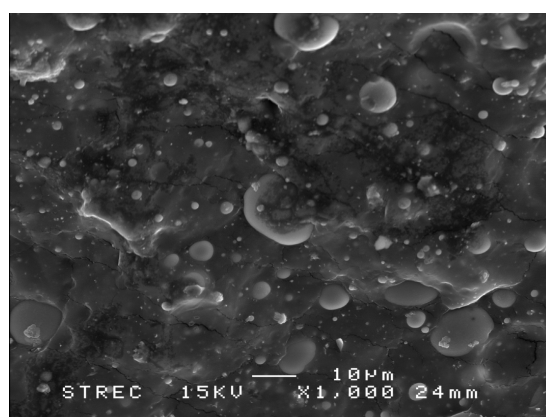
(b)



(c)

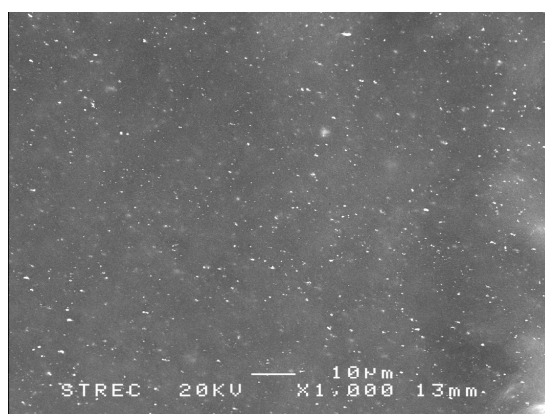


(d)

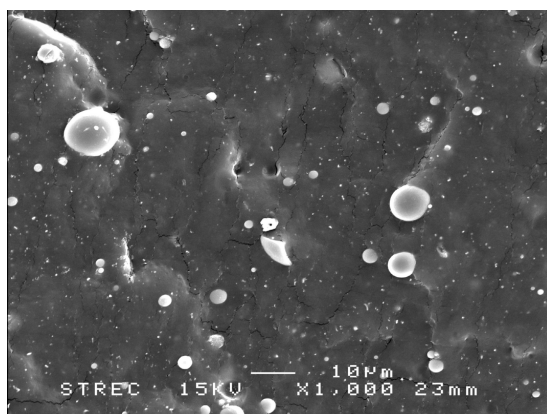


(e)

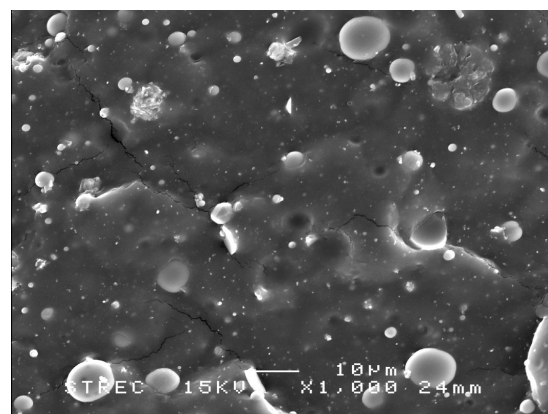
Figure 5.101 SEM micrographs of FA filled NR vulcanizates cured via EV system with various FA loadings: (a) 0; (b) 50; (c) 100; (d) 150 and (e) 200 phr



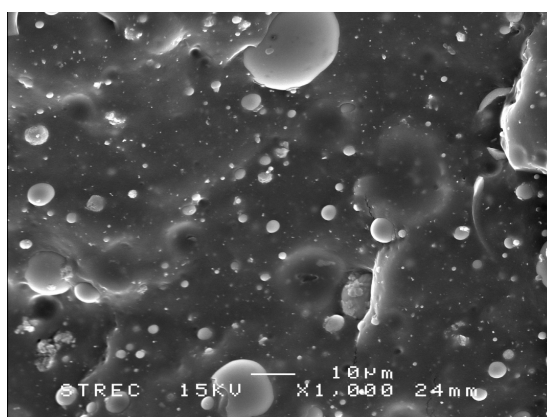
(a)



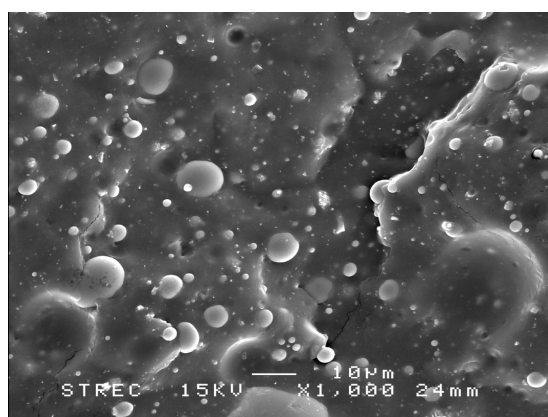
(b)



(c)

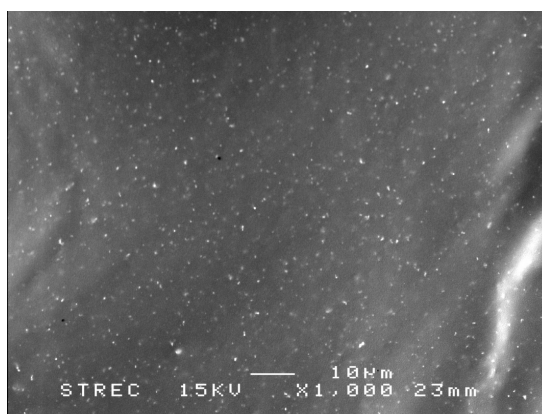


(d)

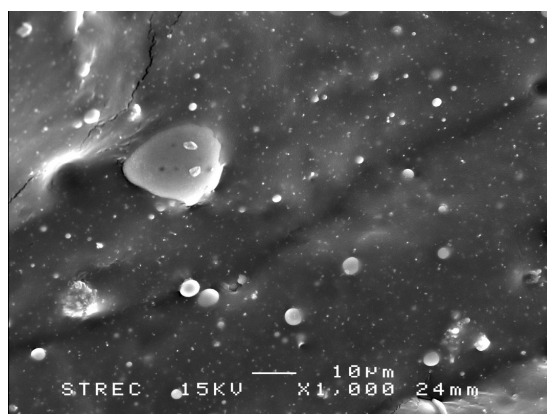


(e)

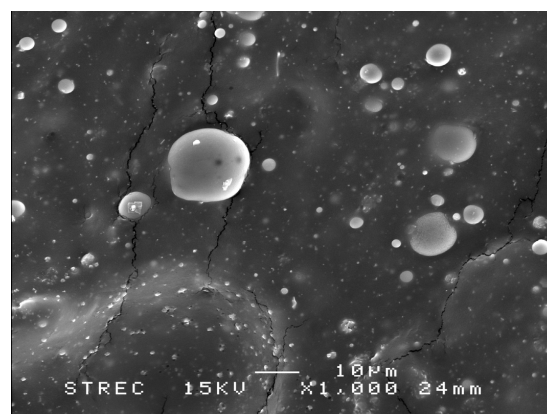
Figure 5.102 SEM micrographs of FA filled NR vulcanizates cured via semi-EV system with various FA loadings: (a) 0; (b) 50; (c) 100; (d) 150 and (e) 200 phr



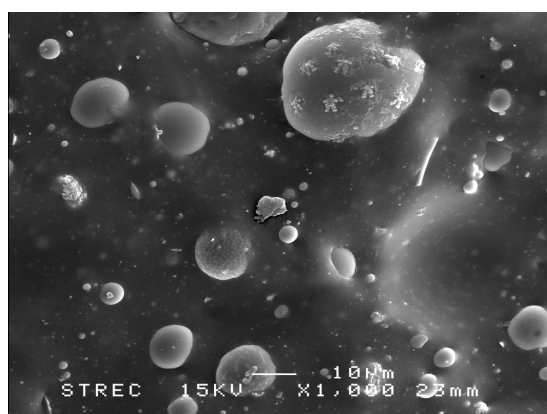
(a)



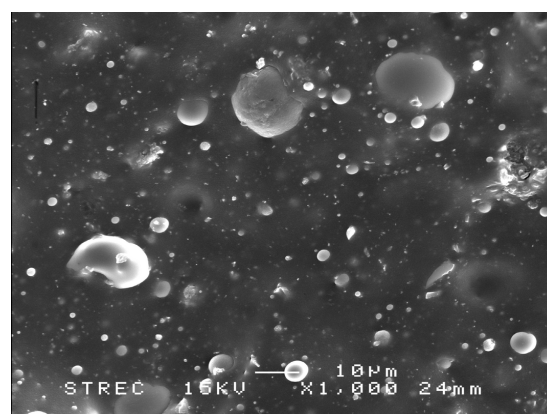
(b)



(c)



(d)



(e)

Figure 5.103 SEM micrographs of FA filled NR vulcanizates cured via peroxide system with various FA loadings: (a) 0; (b) 50; (c) 100; (d) 150 and (e) 200 phr

5.4.2 Silane Surface Treatment

As it is widely known that the reinforcement by PSi in NR is inefficient without the use of coupling agent, it is of interest to investigate whether the silane coupling agent is needed in the case of FA as silica-containing filler. Thus, in this section, the effects of silanes on properties of NR filled with 100 phr FA under sulfur and peroxide crosslinking are investigated, and compared with those without silane system. Similar to the previous section, the silane coupling agents used in the present section are bis(3-triethoxysilylpropyl) tetrasulfide (Si-69 or TESPT), 3-thiocyanatopropyl triethoxy silane (Si-264 or TCPTS) and vinyltriethoxysilane (VTEO) on the advantage of 3 silanes which are (i) the sulfur contribution in Si-69 can be promoted the sulfidic linkage formation, (ii) the presence of thiocyanide group (-SCN) might cause a strong interaction by hydrogen bond with silanol groups on FA surface and (iii) the vinyl groups on VTEO might react with NR activated by peroxide curing system. The FA was initially ball-milled, and subsequently silane treated at a fixed concentration of 4% of silane.

5.4.2.1 Cure Characteristics

Figure 5.104 shows cure curves of the vulcanizates, with and without silane coupling agents. The results suggest that the incorporation of all silanes, Si-69, Si-264 and VTEO, in all curing systems exhibits an increase in maximum torque (M_H). The Si-69 yields the highest magnitude of M_H in all curing systems, especially in sulfur curing system because of the additional sulfur in Si-69 silane. Similar to the previous section, the cure curves of CV system shows cure reversion whereas the semi-EV and EV systems show little or no reversion. For peroxide curing, the cure behavior exhibits substantial magnitude of marching. The degree of reversion and the rate of marching [148, 149] of FA/NR vulcanizates are illustrated in Figures 5.105 and 5.106, respectively. It can be noticed that the Si-69 could reduce the reversion degree in all sulfur curing systems, and the Si-264 slightly decreases the cure reversion in CV and semi-EV systems whereas the VTEO increases the reversion magnitude in sulfur curing. The results suggest that the Si-69 in the sulfur compounds could act as an anti-reversion agent by donating sulfur for compensating the

desulfuration reaction during the cure reversion [150, 151]. Furthermore, the Si-264 and VTEO slightly decrease the rate of marching in peroxide curing system.

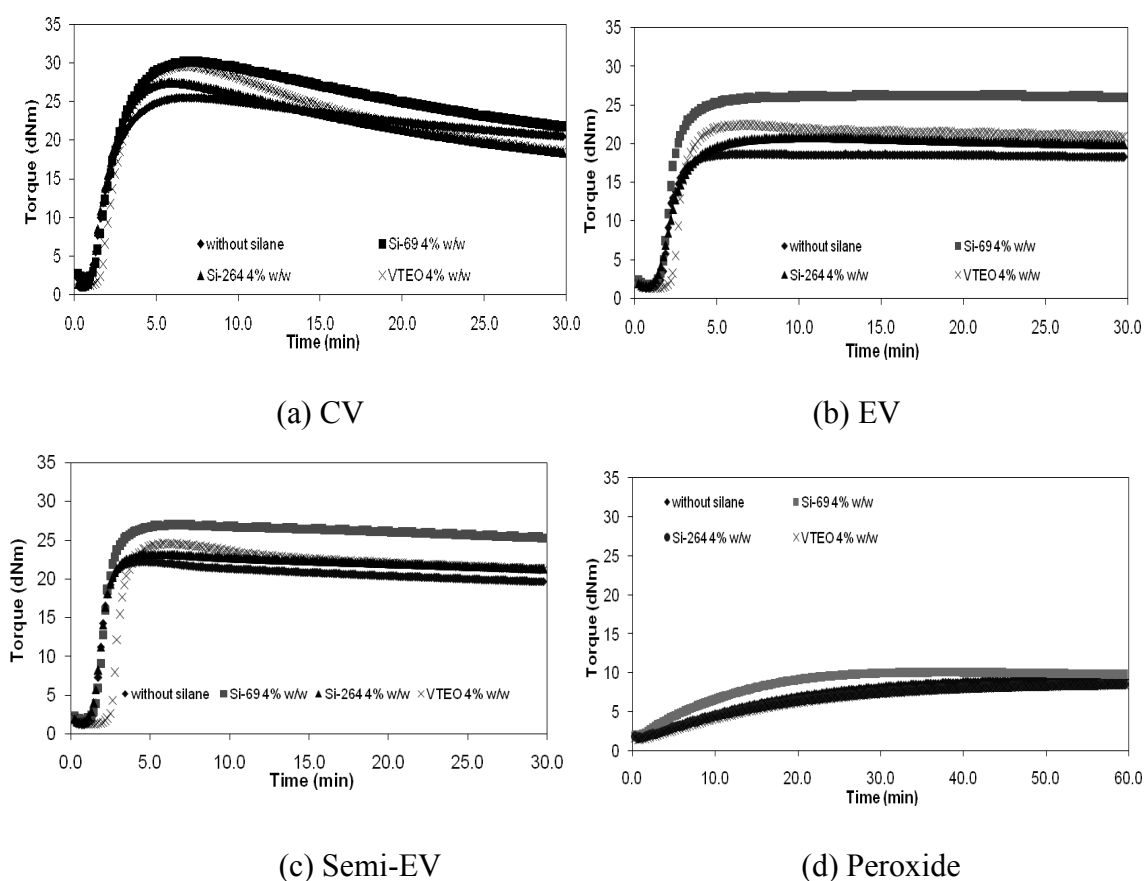


Figure 5.104 Cure curves of FA filled NR vulcanizates with 3 silanes under various curing systems; (a) CV, (b) EV, (c) semi-EV and (d) peroxide curing systems

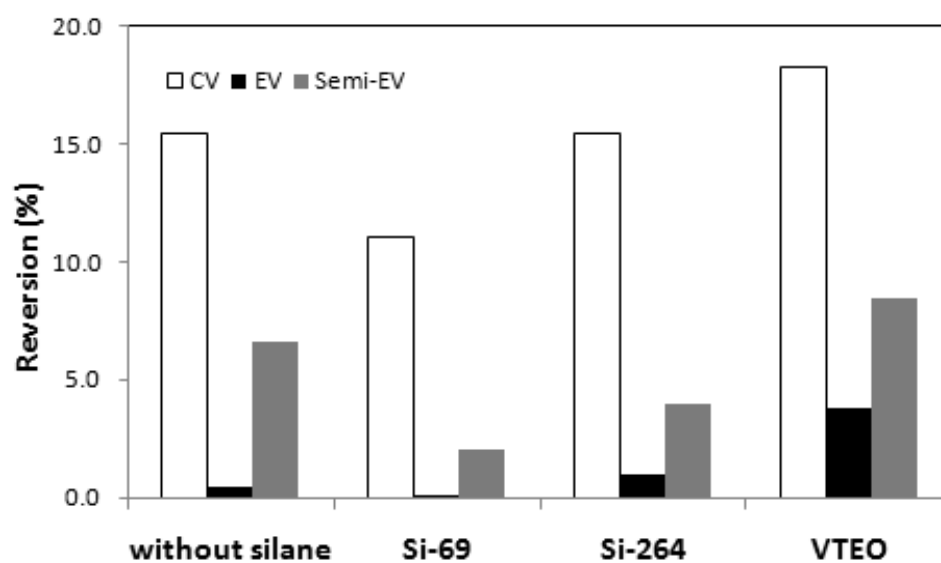


Figure 5.105 Degree of cure reversion in FA filled NR vulcanizates with 3 silanes under sulfur curing systems

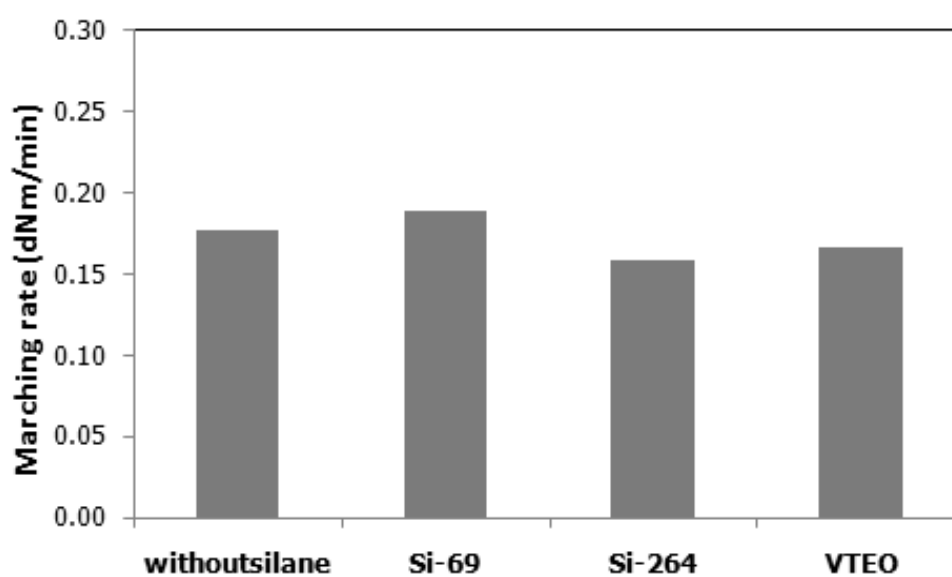
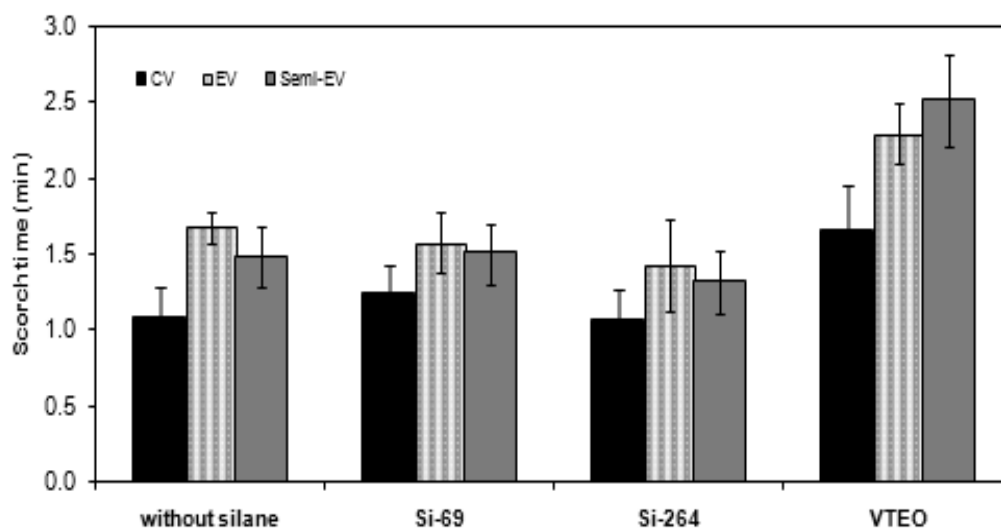


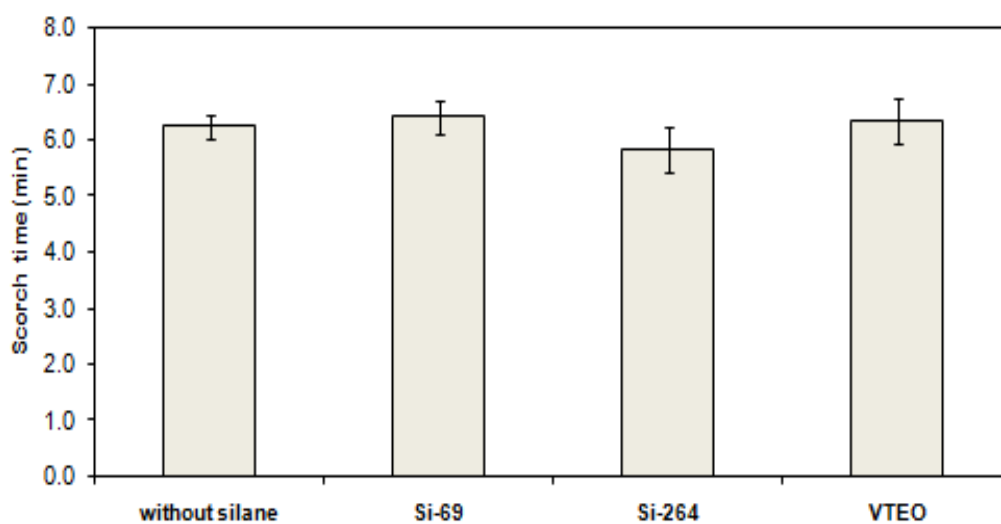
Figure 5.106 Rate of cure marching of FA filled NR vulcanizates with 3 silanes under peroxide curing system

Figures 5.107 and 5.108 show scorch and cure times of the vulcanizates with and without silanes in sulfur and peroxide curing systems. The results demonstrate that the additions of Si-69 and Si-264 in sulfur system do not affect strongly the scorch time of the FA/NR compounds whereas the addition of VTEO gives increased scorch time. The possible explanation is the reaction between the vinyl group on VTEO and sulphur. For peroxide curing system, the silanes give no profound effect on scorch time. Regarding cure times, both sulfur and peroxide curing systems with silanes show longer cure time than those without silane. These silanes probably cause a steric hindrance effect on the formation of crosslink, i.e., the long alkyl chains of Si-69 restrict the linkage formation of NR radicals and silane. By contrast, type of silane gives no significant effect on cure time.

Torque difference (as an indication of crosslink density) of the vulcanizates with and without silane in sulfur and peroxide curing systems is illustrated in Figure 5.109. It is clear that the rise in torque difference of all curing systems is found by the use of Si-69. Nevertheless, the Si-264 shows no significant impact on torque difference in sulfur curing system. The incorporation of VTEO slightly increases torque difference in sulfur curing systems. The results suggest the increase in crosslink density in all curing systems, especially in EV and semi-EV systems as supported by the result of the crosslink density via swelling test as exhibited in Figure 5.110. This is because the polysulfide in Si-69 could offer free sulfur for enhancing the crosslink density. In addition, the incorporations of Si-264 and VTEO show marginal increase in crosslink density.

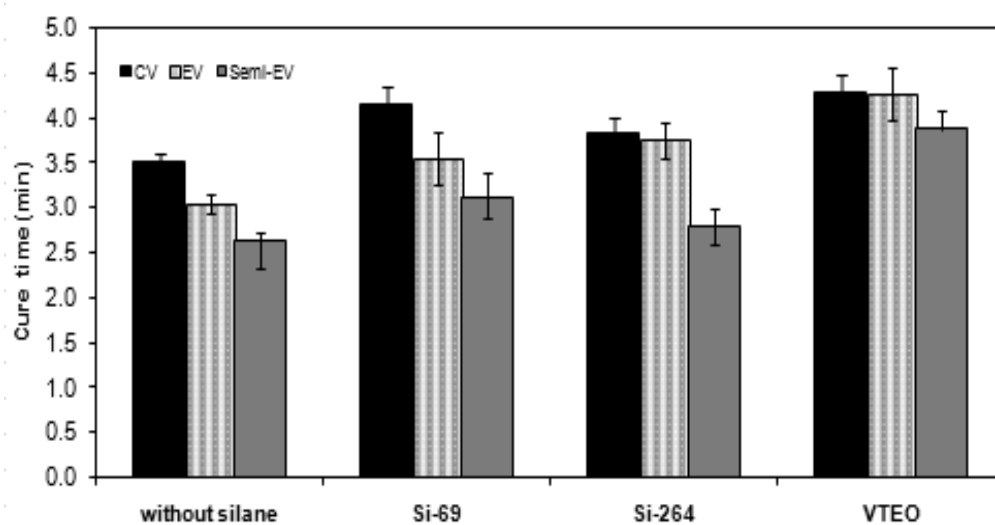


(a)

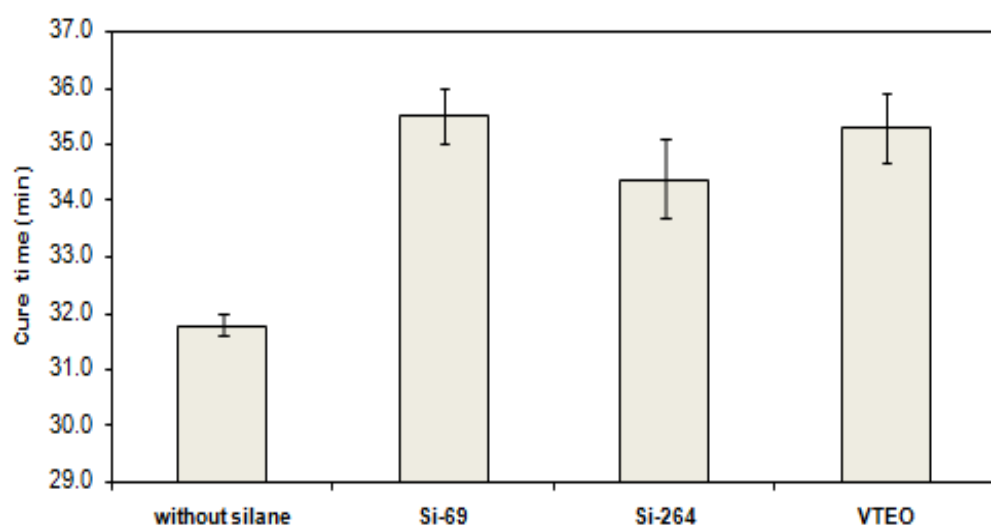


(b)

Figure 5.107 Scorch time of FA filled NR composites with 3 silanes in various curing systems; (a) sulfur and (b) peroxide curing systems

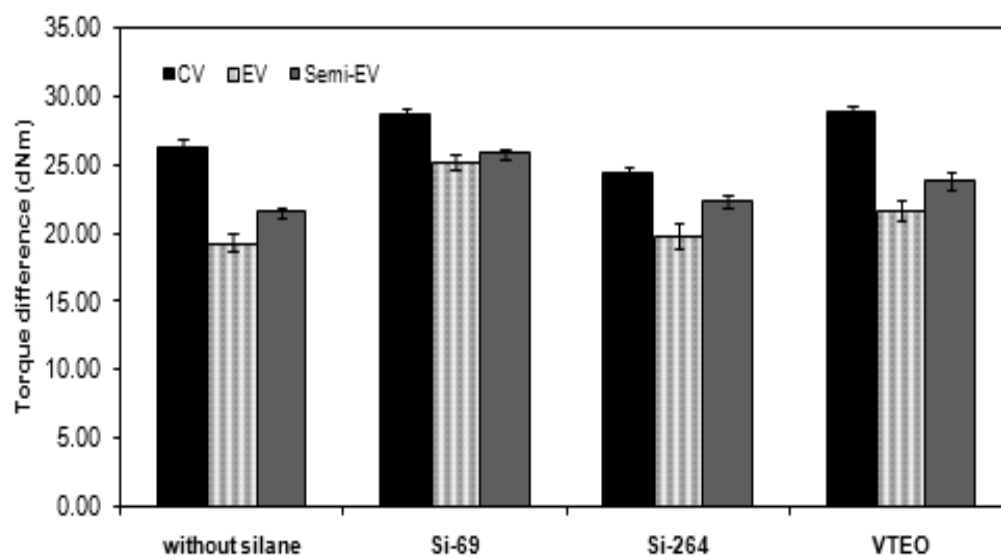


(a)

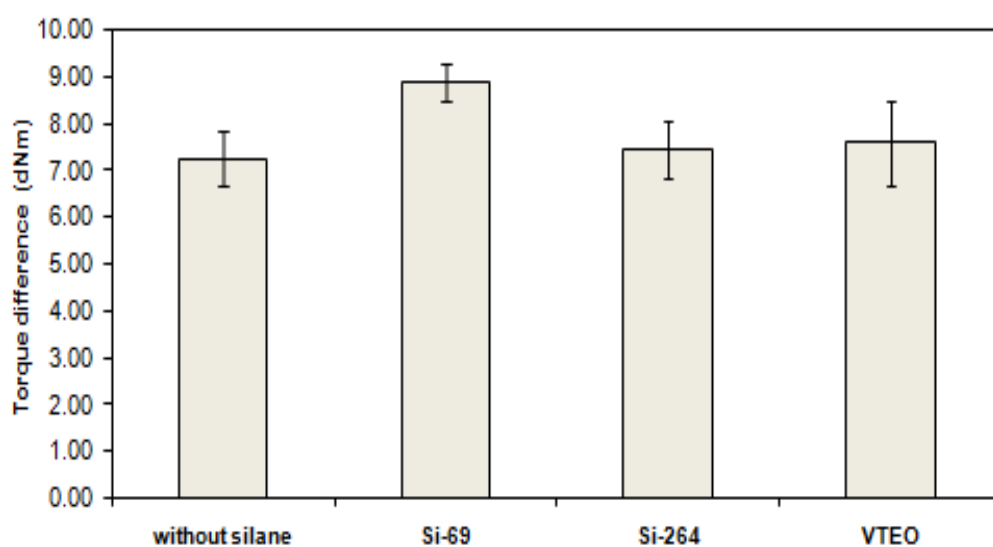


(b)

Figure 5.108 Optimum cure time of FA filled NR composites with 3 silanes in various curing systems; (a) sulfur and (b) peroxide curing systems



(a)



(b)

Figure 5.109 Torque difference of FA filled NR composites with 3 silanes in various curing systems; (a) sulfur and (b) peroxide curing systems

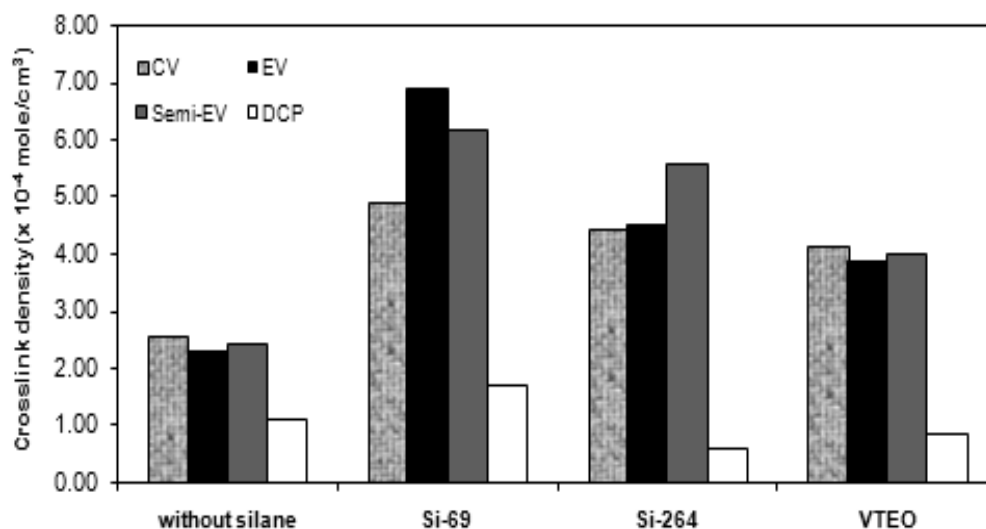


Figure 5.110 Crosslink density (as determined from Flory-Rehner equation via swelling test) of FA filled NR composites with 3 silanes in various curing systems; (a) sulfur and (b) peroxide curing systems

5.4.2.2 Viscoelastic Behavior

(A) Time Sweep Test

As mentioned previously, the thermal stability test must be performed in storage modulus (G') to ensure the absence of thermal degradation during test duration. From Figure 5.111, the G' of all vulcanizates does not significantly change with 30 minutes of test duration. This means all results exhibit good thermal stability. Additionally, it is evident that the G' of test specimens with both sulfur and peroxide curing systems is increased by silane treatment especially Si-69.

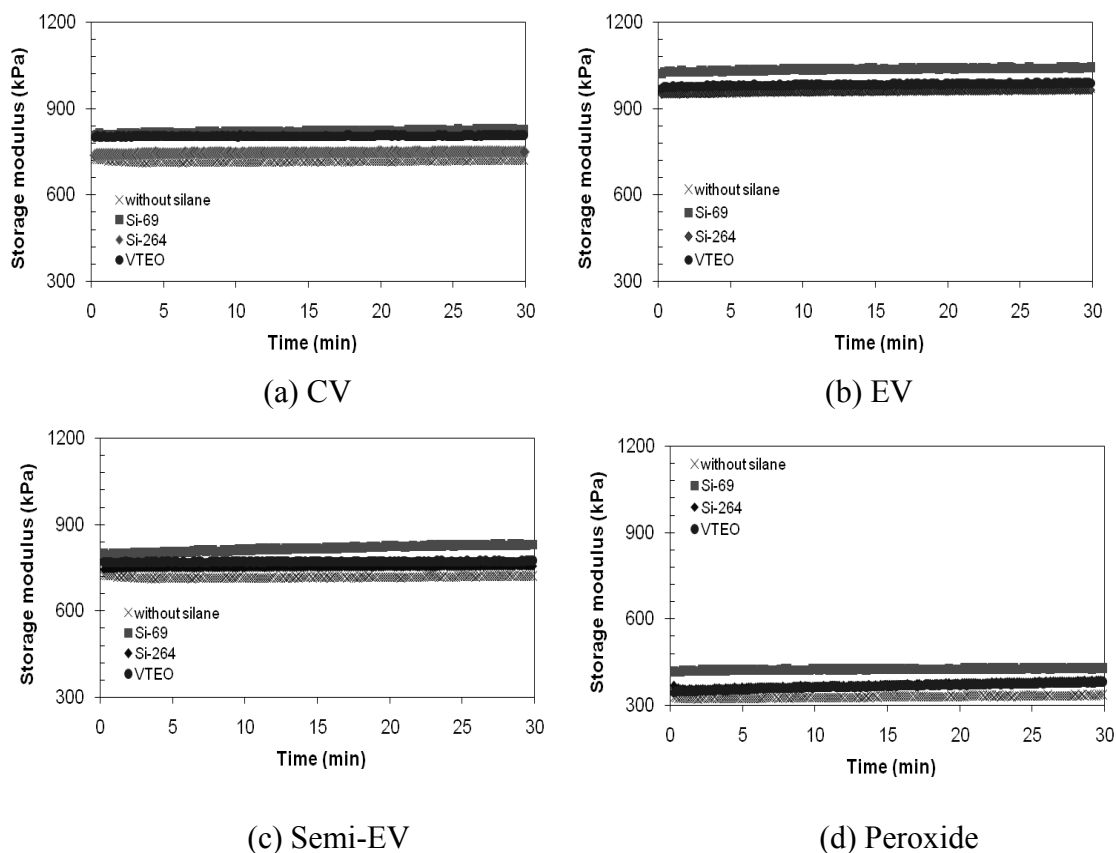


Figure 5.111 Storage modulus (G') as a function of time of cured FA filled NR vulcanizates with 3 silanes in various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

(B) Strain Sweep Test

(B-1) Uncured Systems

Figure 5.112 shows the effect of silane on G' as a function of strain amplitude in FA filled NR compounds with either sulfur or peroxide curing systems. For both curing systems, the G' rises by the use of silanes with the pronounced effect found in the system with Si-69. The decrease in G' with strain amplitude found in all systems is caused by the disruption of filler transient network together with the molecular slippage of NR molecules. To compare the effect of silanes on magnitude of Payne effect, the plots of difference in G' at low and high strain ($\Delta G'$) with and without silanes are shown in Figure 5.113. The $\Delta G'$ increases

with the addition of silanes in all curing systems except for EV curing. The decrease in $\Delta G'$ in the EV curing system is explained by the deactivation of silanol groups on FA surfaces by a large amount of amine accelerator. This explanation is also applicable to results of Payne effect which is lower in sulfur curing system than the peroxide curing, i.e., the existence of amine accelerator in the system with sulfur curing. However, it is still not understood why the Payne effect of the system with silane is greater than those without silane.

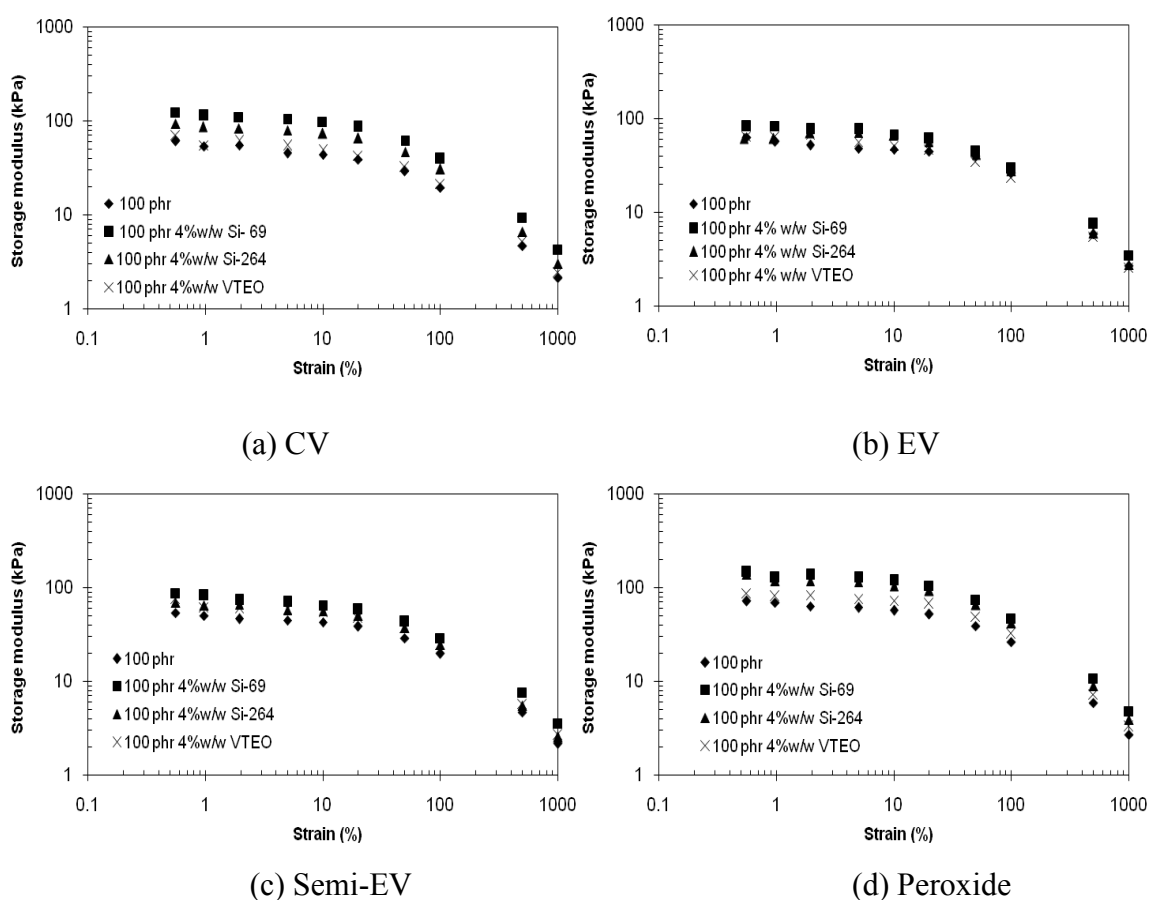


Figure 5.112 Storage modulus (G') as a function of strain amplitude of uncured FA filled NR compounds with 3 silanes in various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing system

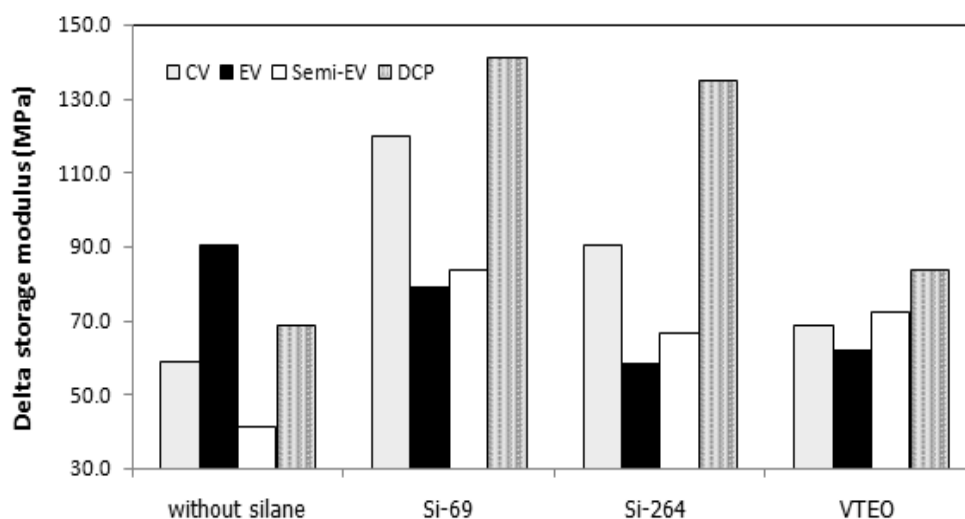


Figure 5.113 Payne effect in FA filled NR with 3 silanes in various curing systems

(B-2) Cured Systems

Figure 5.114 illustrates the G' as a function of strain amplitude of cured FA filled NR specimens with and without silanes. For sulfur curing system, the G' of vulcanizates with and without silanes is not significantly different, except in the EV system, it is noticed that the G' of FA/NR vulcanizate slightly increases by the use of Si-69. The explanation is given by the effect of sulfur contribution provided by Si-69. Unlike sulfur curing system, the peroxide reveals an increase in G' with the addition of silanes. The comparative results of relative G' of all cured specimens as determined from RPA2000 and that calculated from the Guth-Gold equation are given in Figure 5.115. It is evident that, after incorporating silanes, the experimental results of relative G' slightly increases and the magnitude of negative deviation from Guth-Gold results is decreased due to the slightly increased FA-NR interaction via silane. Nevertheless, the experimental results of relative G' of all cured specimens are still lower than calculated results. This is because of the low surface activity and surface area of FA particles together with the ball bearing effect of FA spherical particles as discussed previously.

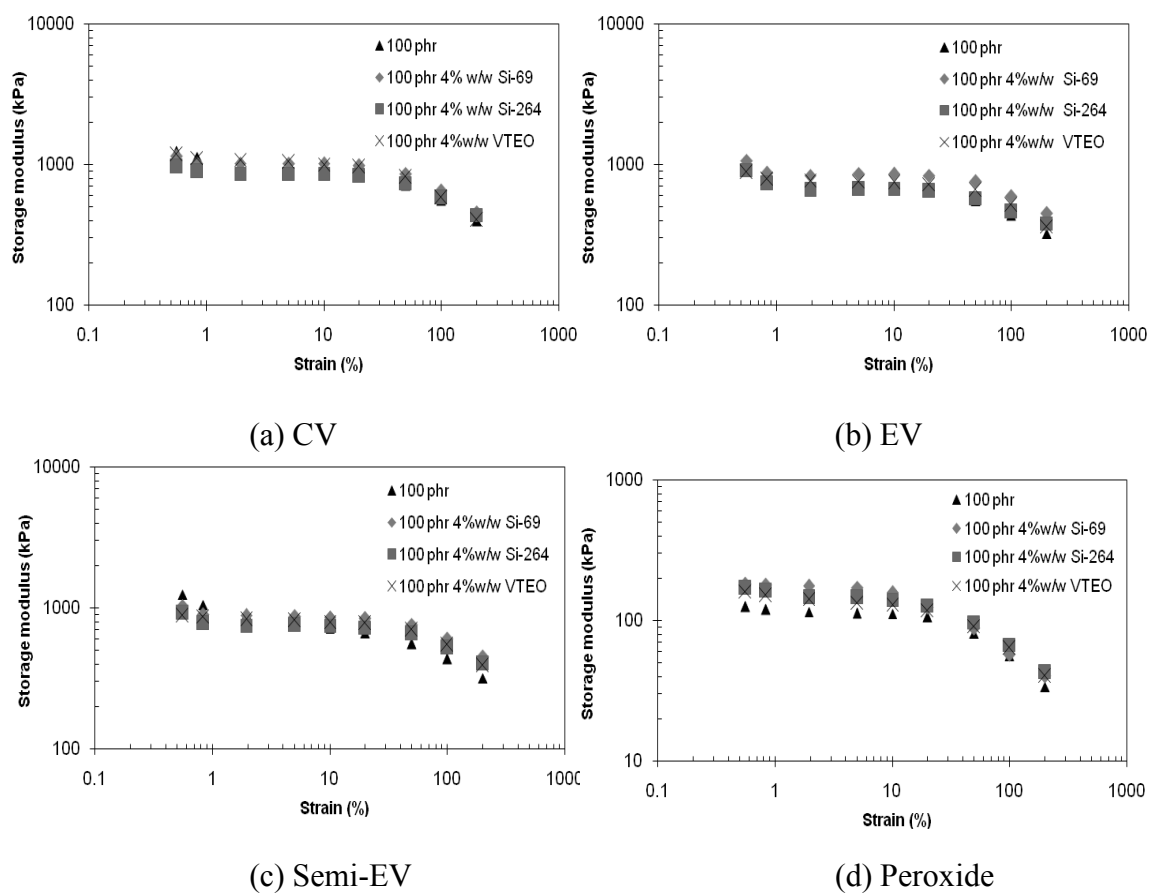


Figure 5.114 Storage modulus (G') as a function of strain amplitude of cured FA filled NR specimens with 3 silanes in various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

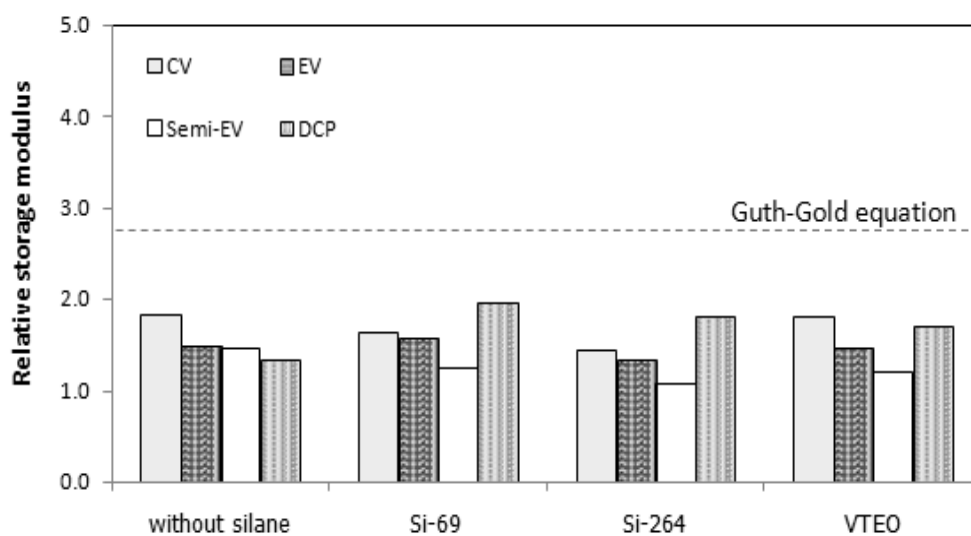


Figure 5.115 Relative storage modulus (relative G') at 1% strain of 100 phr FA filled NR specimens with 3 silanes in various curing systems

(C-1) Uncured Systems

Figure 5.116 shows the frequency dependence of G' for uncured FA filled NR compounds with and without silanes. It is evident that G' of all uncured compounds is frequency-dependent. By comparing the compounds with and without silane, the slope of G' vs. frequency plots as shown in Figure 5.117 decreases with added silane. This might be explained by the formation of interaction between NR and FA via the coupling reaction provided by (i) silane, and (ii) that between NR and silanes as supported by the results of damping factor exhibited in Figure 5.118 where the reduction in damping factor after silane treatment could be observed.

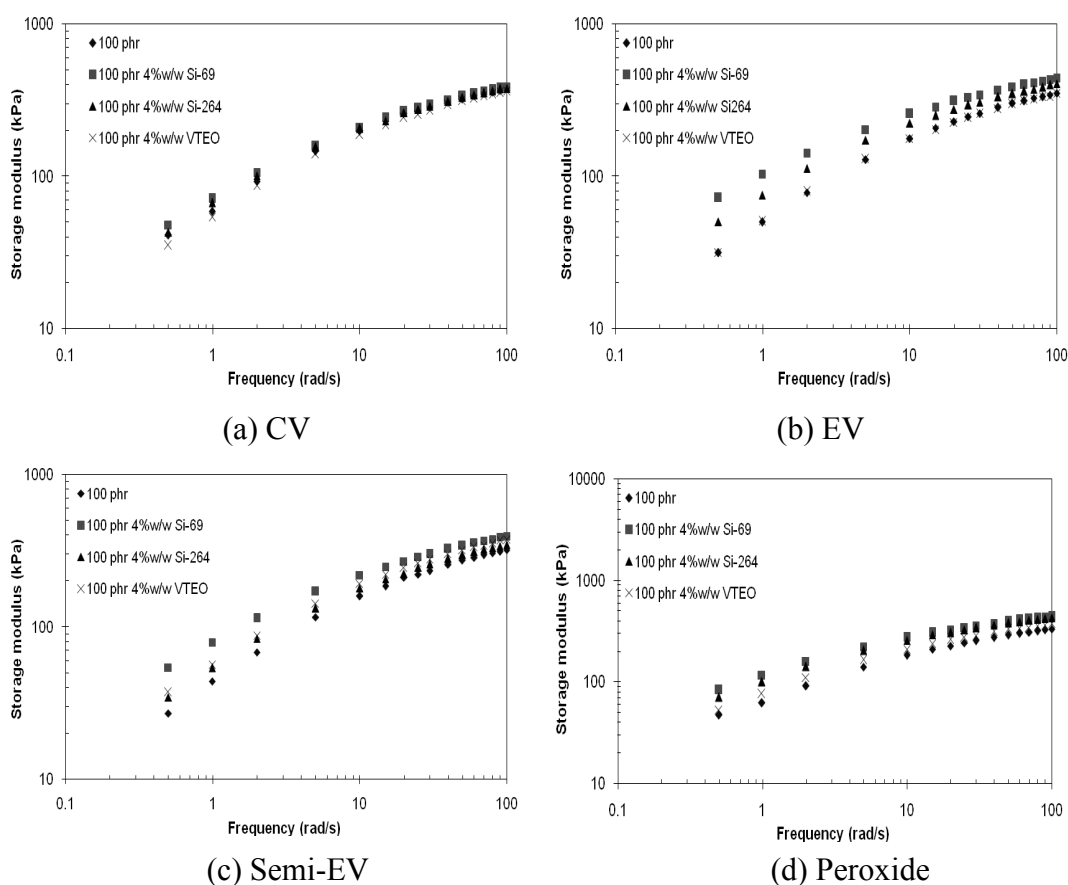


Figure 5.116 Storage modulus (G') as a function of frequency of uncured FA filled NR compounds with 3 silanes in various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

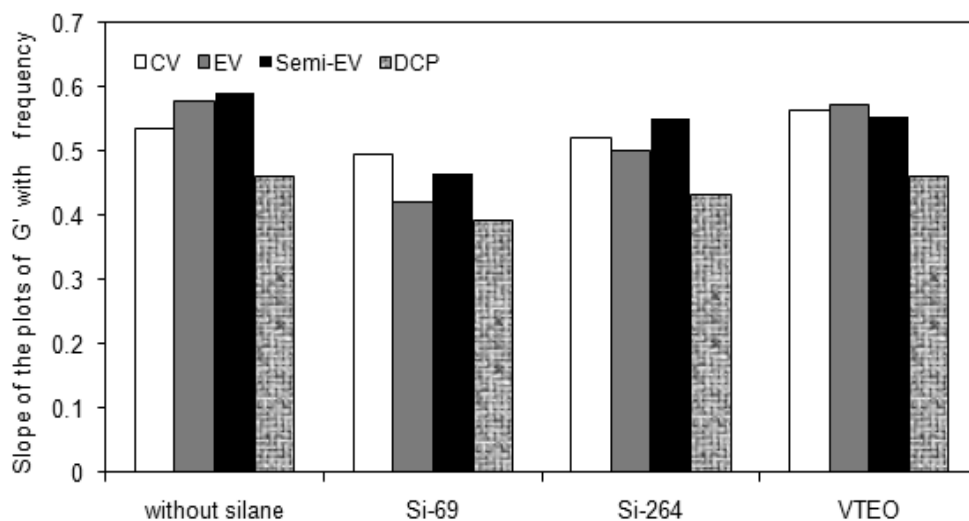


Figure 5.117 Slope calculated from the plots of storage modulus (G') as a function of frequency of uncured FA filled NR compounds with 3 silanes in various curing systems

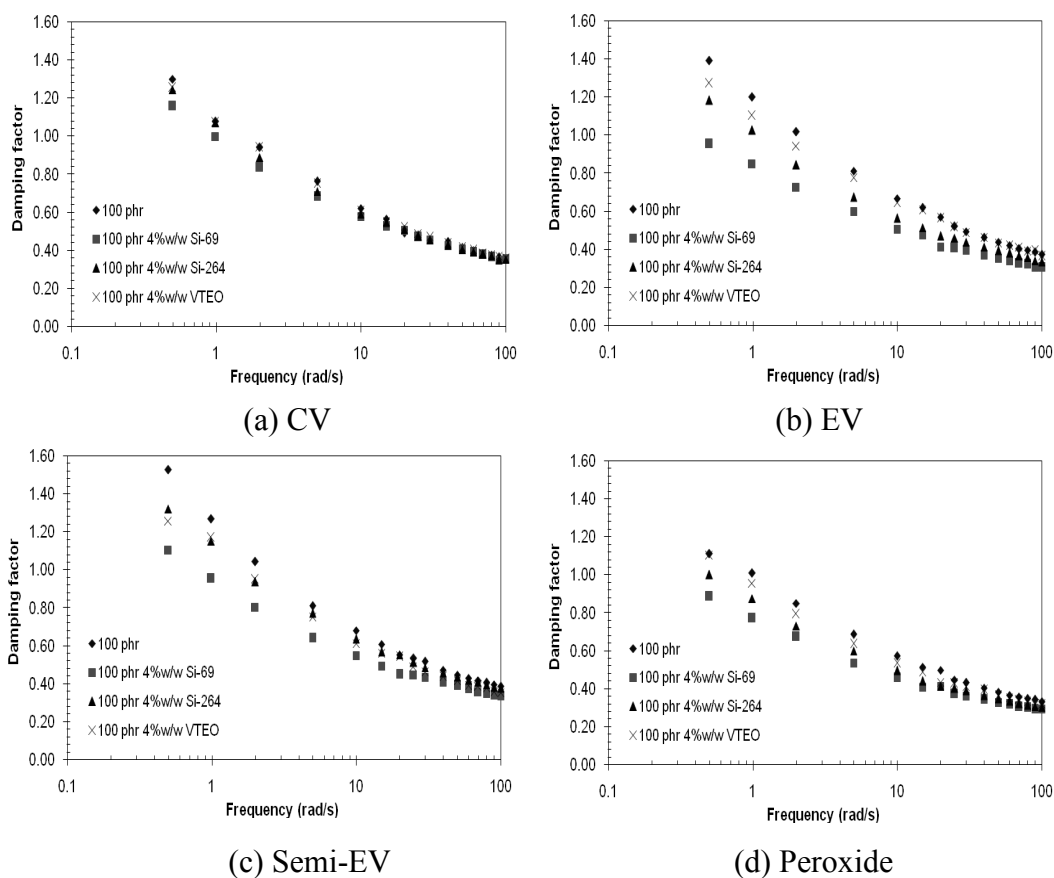


Figure 5.118 Damping factor as a function of frequency of uncured FA filled NR incorporated with 3 silanes in various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

Furthermore, the results of complex viscosity (η^*) as shown in Figure 5.119 still exhibit the pseudoplastic behaviour, and the η^* of compounds in all curing systems increases after the addition of silanes, except for the CV system. The results of η^* agree with the results of power law index as shown in Figure 5.120. The addition of Si-69 decreases power law index in all curing systems whereas the incorporation of other silanes slightly decrease the power law index.

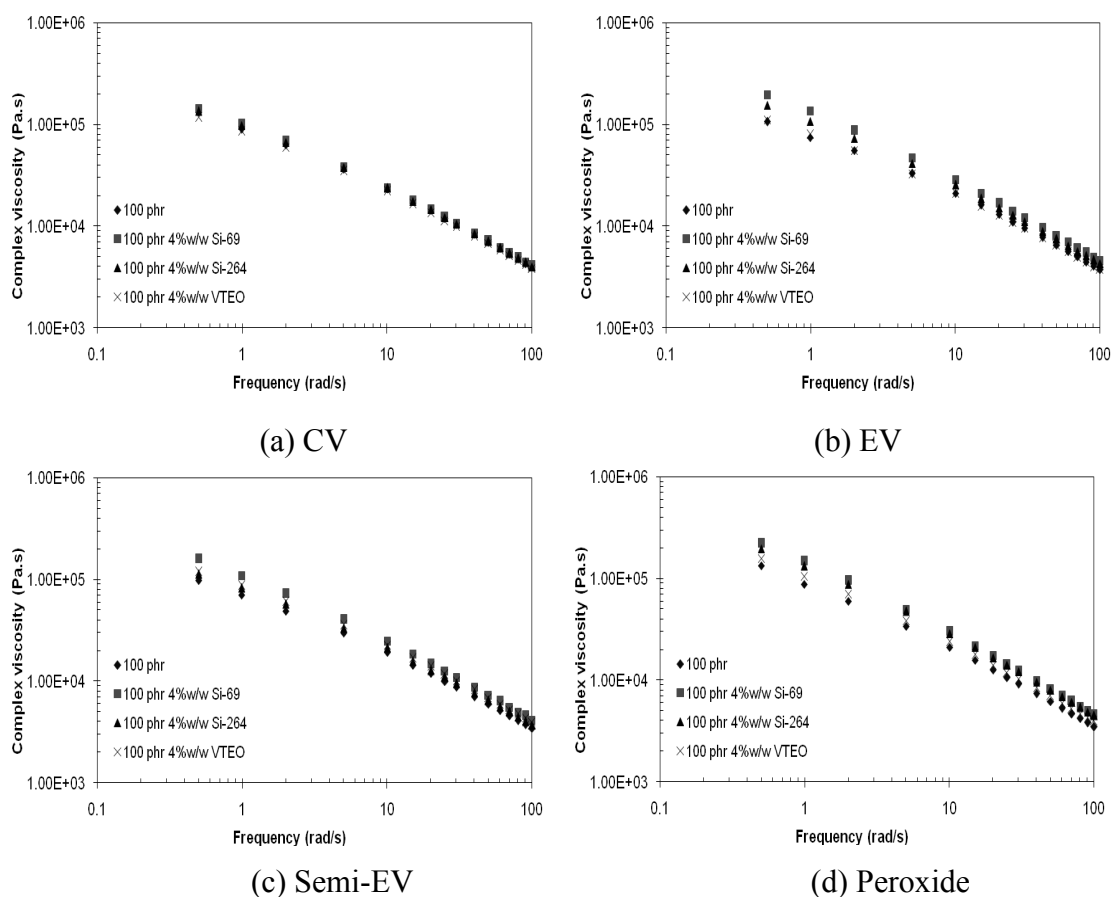


Figure 5.119 Complex viscosity as a function of frequency of uncured FA filled NR compounds with 3 silanes in various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

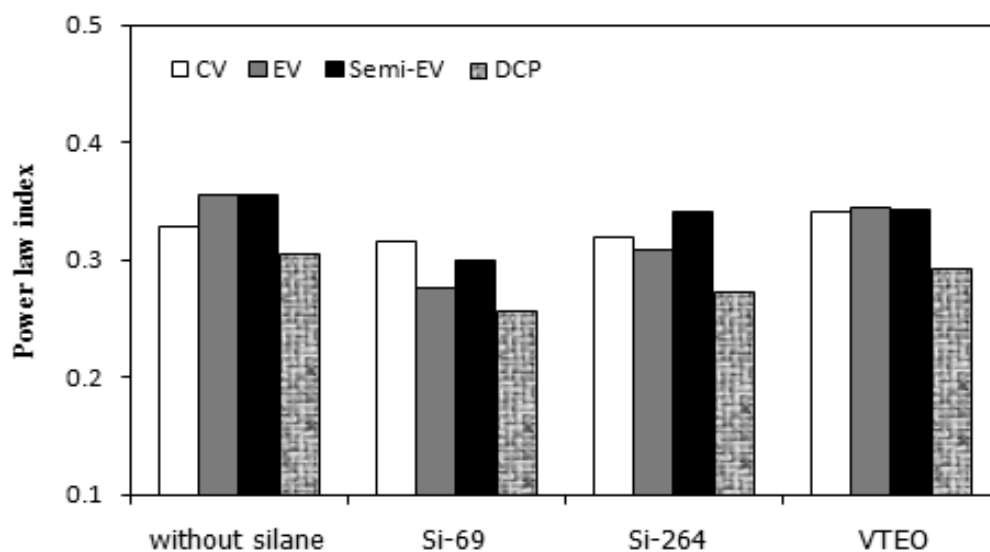


Figure 5.120 Power law index of uncured FA filled NR compounds with 3 silanes in various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

(C-2) Cured Systems

Figure 5.121 shows G' results as a function of angular frequency at 10% strain of cured FA/NR vulcanizates. For sulfur curing system with and without silane, G' is time-independent due to the elastic response domination via sulfur vulcanization. On the other hand, for peroxide curing system with and without silane, the G' is time dependent as noticed from the slight increase in G' with increasing frequency. Moreover, the G' of all curing systems increases after silane treatment. Such increased G' in the vulcanizates with Si-69 which is in-line with the decreased damping factor as illustrated in Figure 5.122.

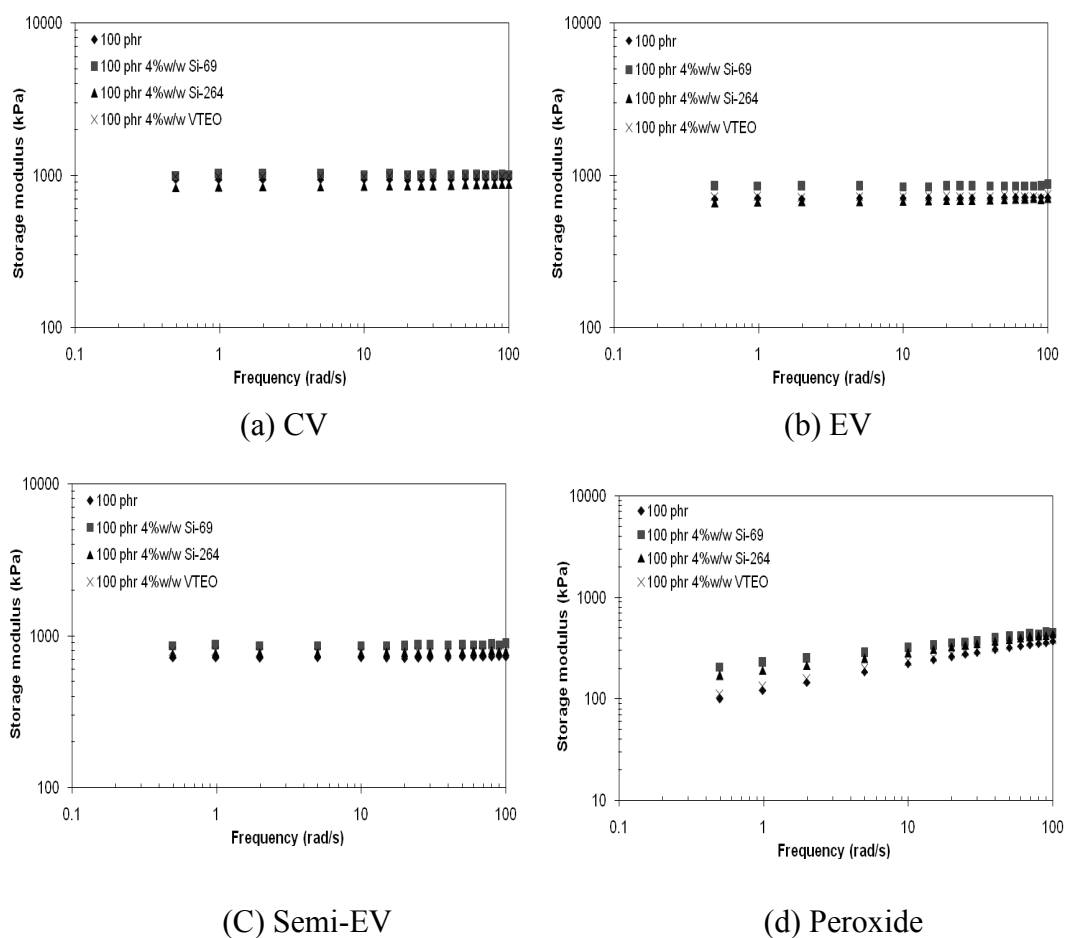
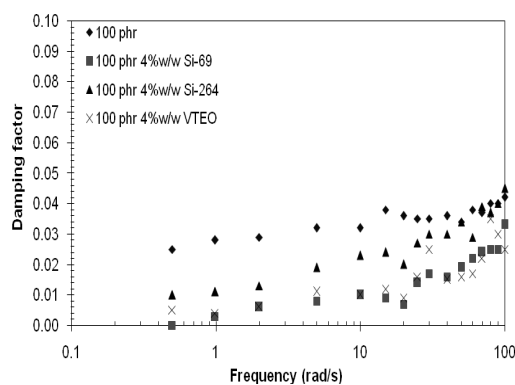
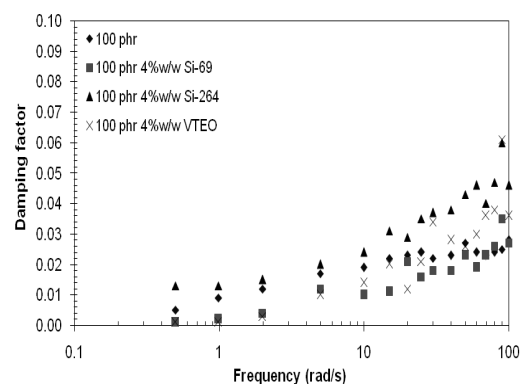


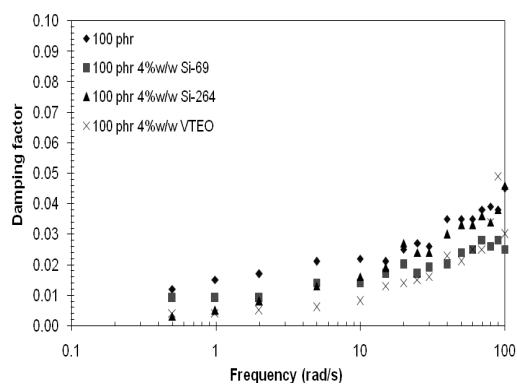
Figure 5.121 Storage modulus (G') as a function of frequency of FA filled NR vulcanizates with 3 different silanes under various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems



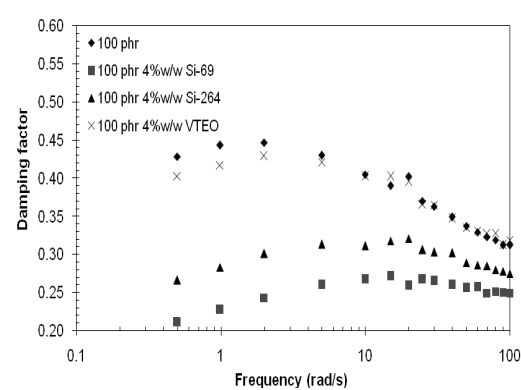
(a) CV



(b) EV



(c) Semi-EV



(d) Peroxide

Figure 5.122 Damping factor (tan delta) as a function of frequency of FA filled NR vulcanizates with 3 different silanes under various curing systems: (a) CV; (b) EV; (c) semi-EV and (d) peroxide curing systems

(D) Thermal Stability Test

Similar to the previous section, the thermal stability test was performed using the RPA2000. For all curing systems, silanes do not affect the thermal stability of FA/NR vulcanizates, as evidenced by the insignificant change in retention modulus shown in Figure 5.123.

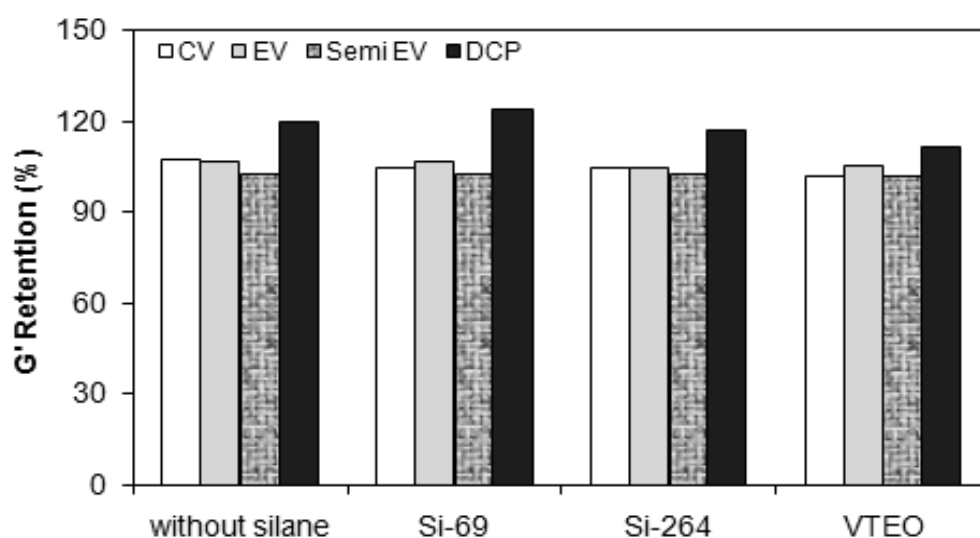


Figure 5.123 Retention of storage modulus (G') of FA filled NR vulcanizates with 3 different silanes under various curing systems as measured from RPA 2000

5.4.2.3 Tensile properties

Figure 5.124 illustrates the effect of silanes on the M100 with various curing systems. The M100 of vulcanizates with sulfur curing systems increases after FA surface treatment with Si-69 and Si-264, which agrees well with the results of crosslink density as shown earlier in Figure 5.109, and thus the lowered elongation at break as displayed in Figure 5.125. Figure 5.126 exhibits tensile strength of the specimens with and without silanes under sulfur and peroxide curing systems. Tensile strength of vulcanizates slightly increases after FA surface treatment with Si-69 in both sulfur and peroxide curing systems due to the free sulfur released from the Si-69 during the curing process. By contrast, other silanes do not effect on tensile strength in sulfur system but decrease in peroxide system because of the small amount

of active silanol groups on FA surfaces and the FA crystalline structure as illustrated in Figures 5.3 and 5.4, respectively. As shown in Table 5.5, the bound rubber content is independent of silane treatment of FA. This implies relatively low rubber-FA interaction, regardless of curatives presented in the compounds.

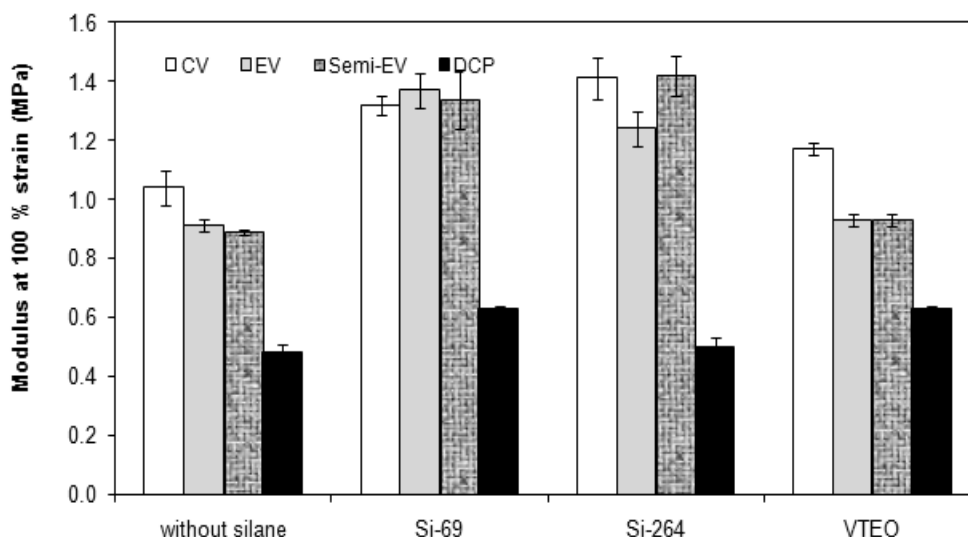


Figure 5.124 Modulus at 100 % strain (M100) of FA filled NR vulcanizates with 3 different silanes in various curing systems

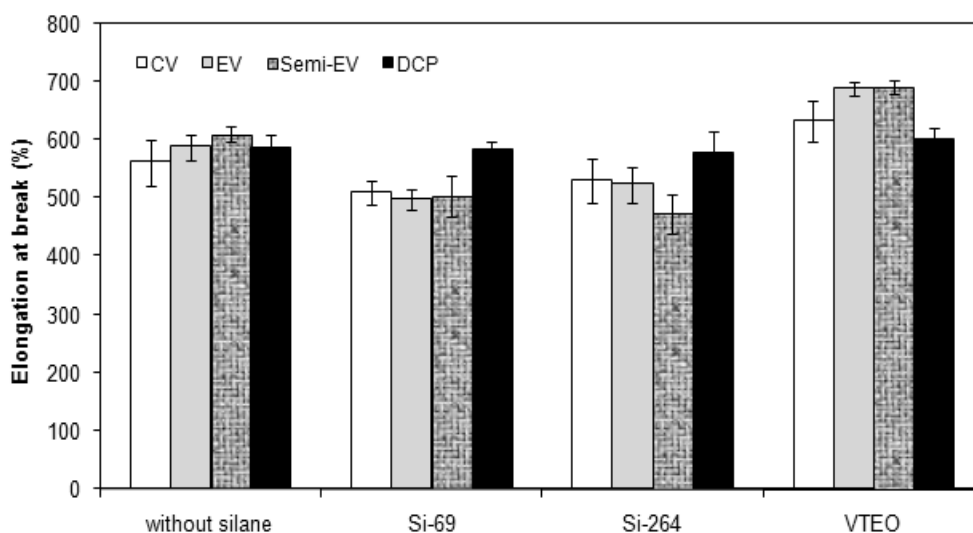


Figure 5.125 Elongation at break of FA filled NR vulcanizates with 3 different silanes in various curing systems

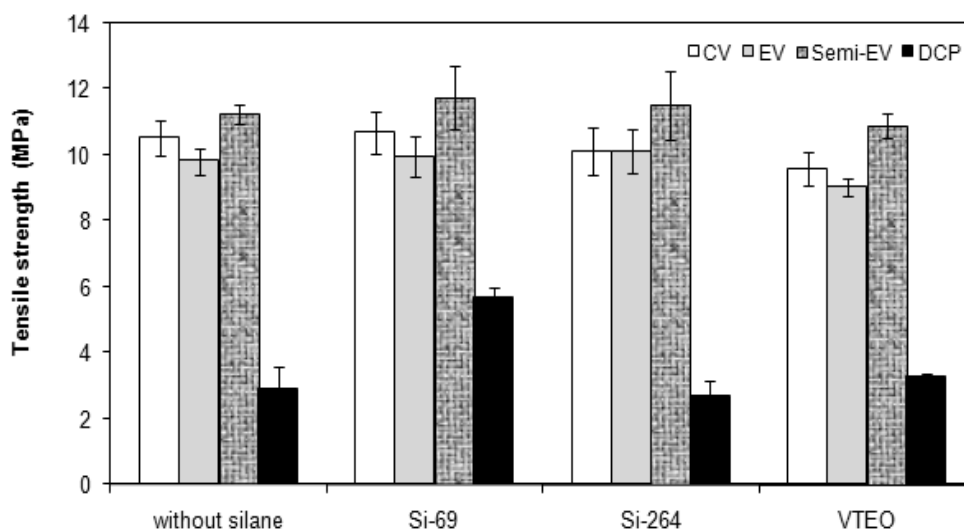


Figure 5.126 Tensile strength of FA filled NR vulcanizates with 3 different silanes in various curing systems

Table 5.5 Bound rubber content of FA filled NR compounds with 3 different silanes in various curing systems

Curing systems	Bound rubber content (%)			
	Without silane	Si-69	Si-264	VTEO
CV	0.0509	0.0983	0.0383	0.0521
EV	-0.0530	0.1144	0.0139	0.0232
Semi-EV	0.0280	0.0678	0.0504	0.0213
DCP	0.0142	0.0809	0.0400	0.0442

By treating FA with silane, in all curing system, the optimum properties are achieved by the use of Si-69. This is because the Si-69 can release free sulfur during curing process. By contrast, other silanes give no enhancement in tensile strength because of the small amount of active silanol groups on FA surfaces resulting from FA crystalline formation structure supported from the FTIR and XRD result as discussed previously.

5.4.2.4 Morphological Results

Figures 5.127 to 5.130 show SEM micrographs of 100 phr FA filled NR vulcanizates with sulfur (i.e., CV, EV and semi-EV) and peroxide curing systems. It can be noticed that the boundaries between FA and NR of untreated FA in all systems suggest poor wettability as evidenced by some voids between FA particles and rubber matrix. For FA surface treated systems, the vulcanizates in all systems exhibit slightly superior phase-continuity and homogeneity the FA-NR interfaces, and the FA particles embed in the rubber matrix. It is due probably to the enhanced wettability of FA particles.

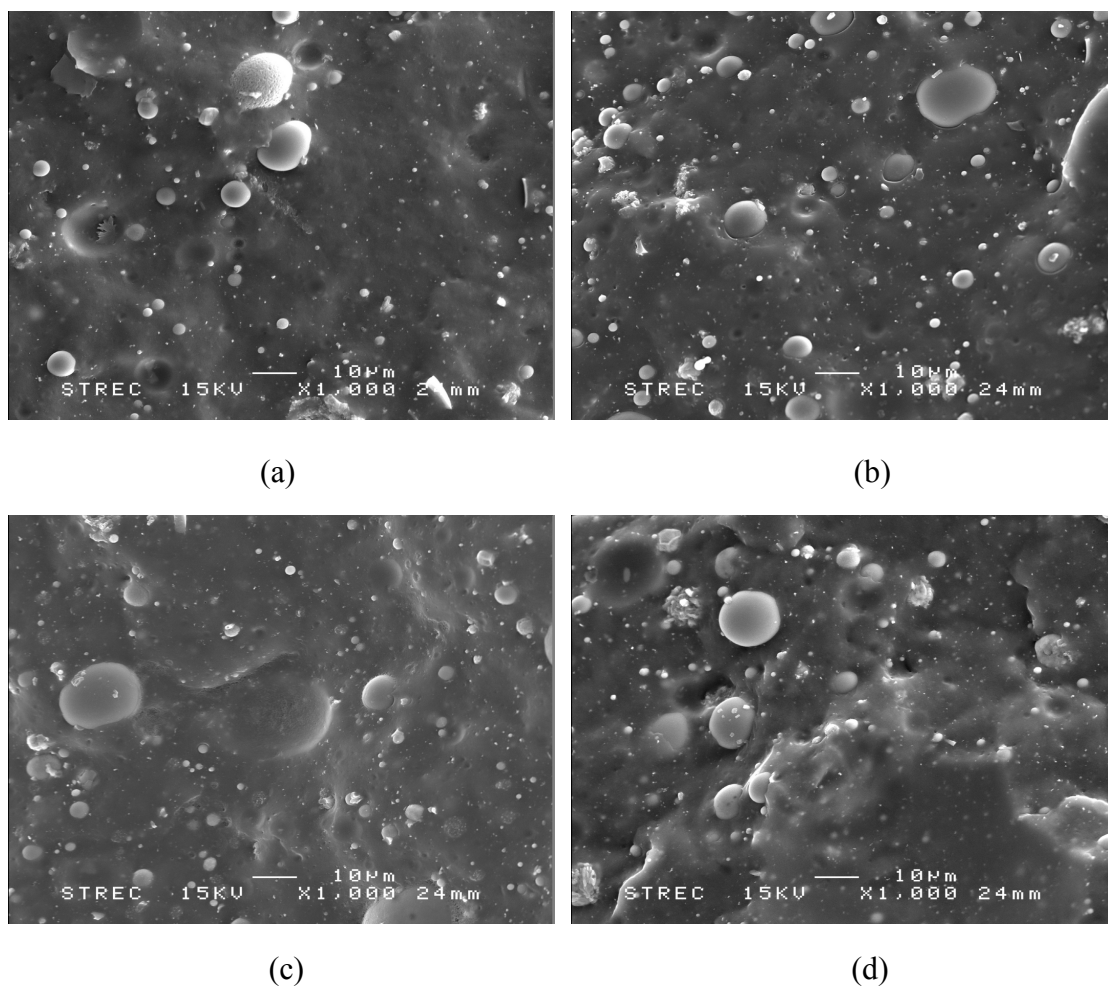


Figure 5.127 SEM micrographs of FA filled NR vulvanizates cured via CV system with 3 silanes : (a) without silane; (b) Si-69; (c) Si-264 and (d) VTEO

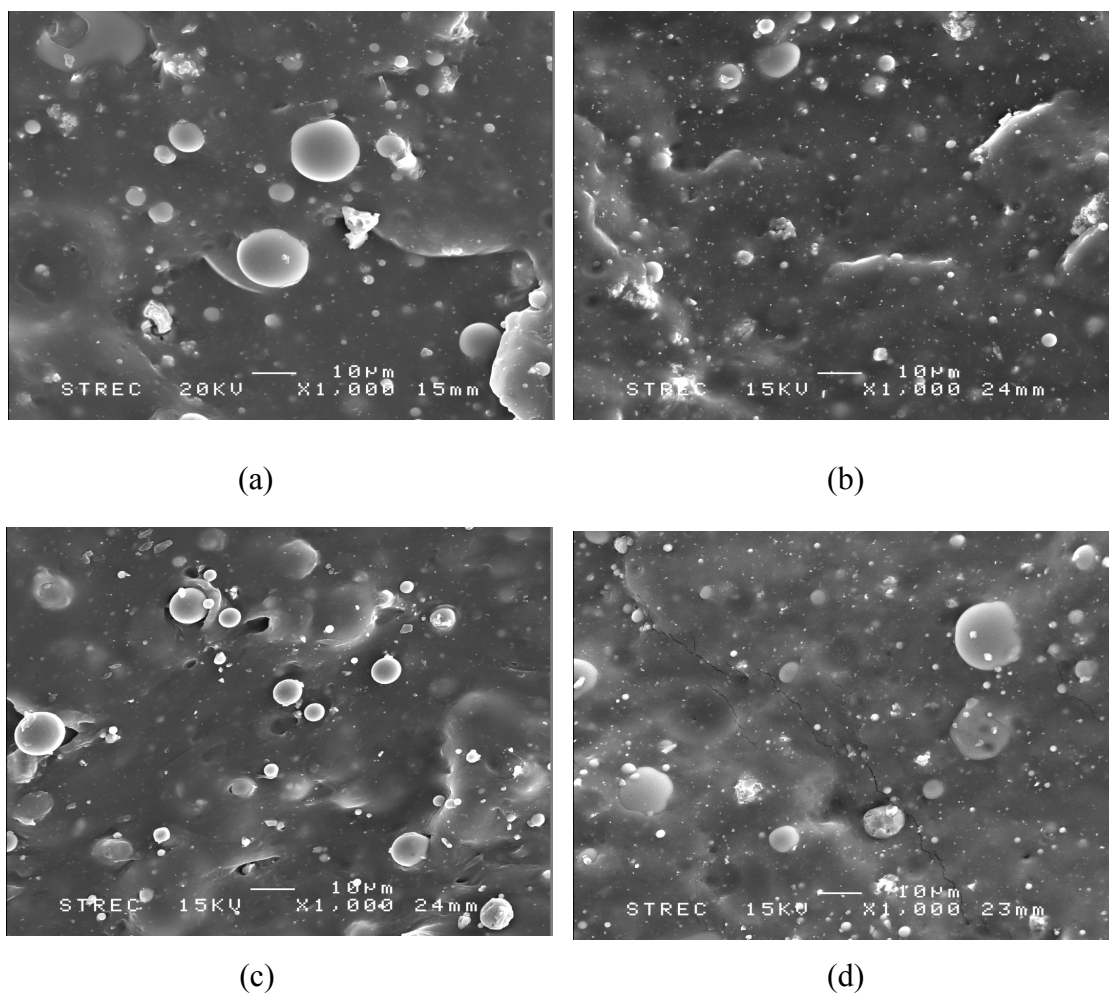
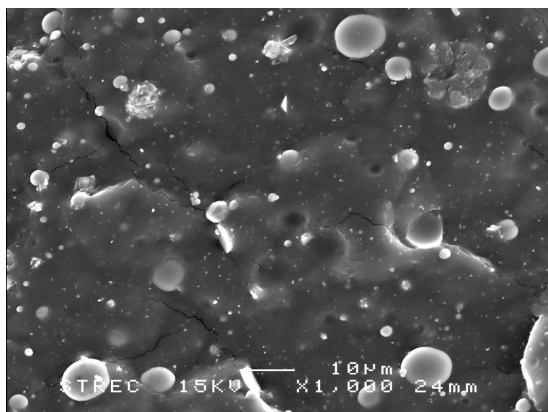
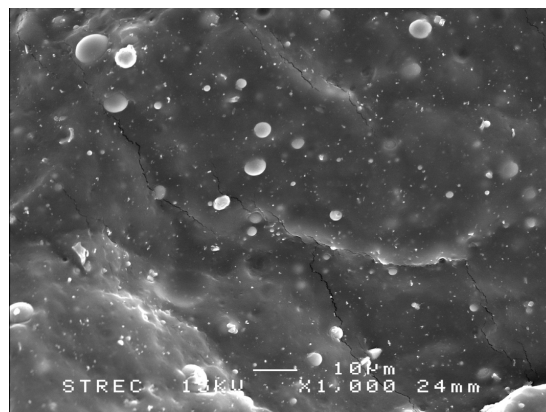


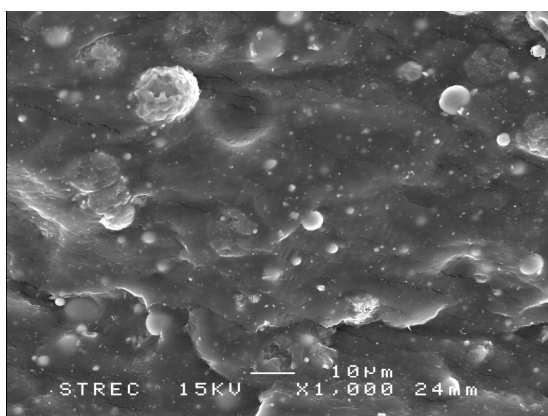
Figure 5.128 SEM micrographs of FA filled NR vulvanizates cured via EV system with 3 silanes : (a) without silane; (b) Si-69; (c) Si-264 and (d) VTEO



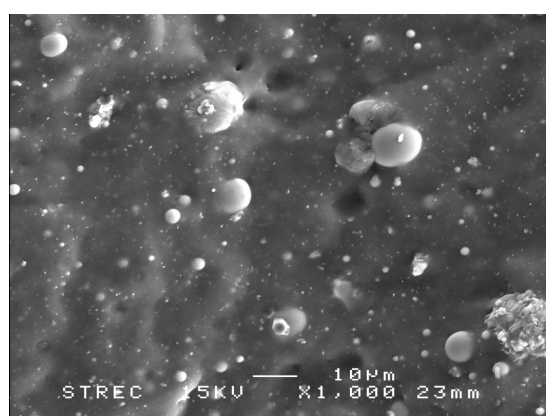
(a)



(b)



(c)



(d)

Figure 5.129 SEM micrographs of FA filled NR vulcanizates cured via semi-EV system with 3 silanes : (a) without silane; (b) Si-69; (c) Si-264 and (d) VTEO

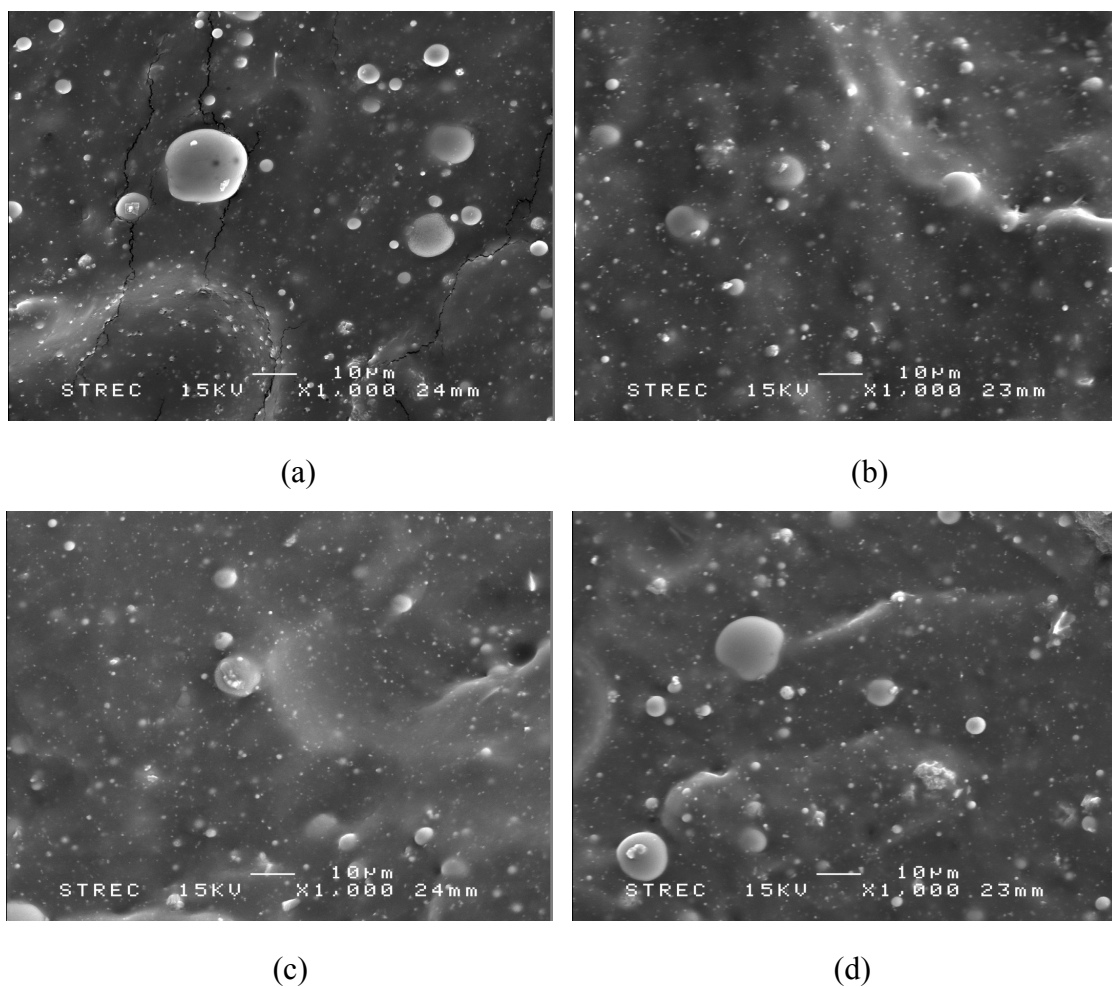


Figure 5.130 SEM micrographs of FA filled NR vulvanizates cured via peroxide system with 3 silanes : (a) without silane; (b) Si-69; (c) Si-264 and (d) VTEO

5.5 Comparison of Viscoelastic and Tensile Properties of NR Filled with Fly Ashes from Mae Moh (Thailand) and Rocktron (UK)

In the present section, the effect of FA origin on viscoelastic properties of FA filled NR composites is focused. The FA used in this part was supplied by Mae Moa power station (Thailand) and RockTron international limited (UK). The loading of FA was kept constant at 100 phr without and with silane coupling agent (i.e., bis(3-triethoxysilylpropyl) tetrasulfide ; Si-69 or TESPT at constant concentration of 4% w/w of FA.

5.5.1 Chemical Compositions of RKFA

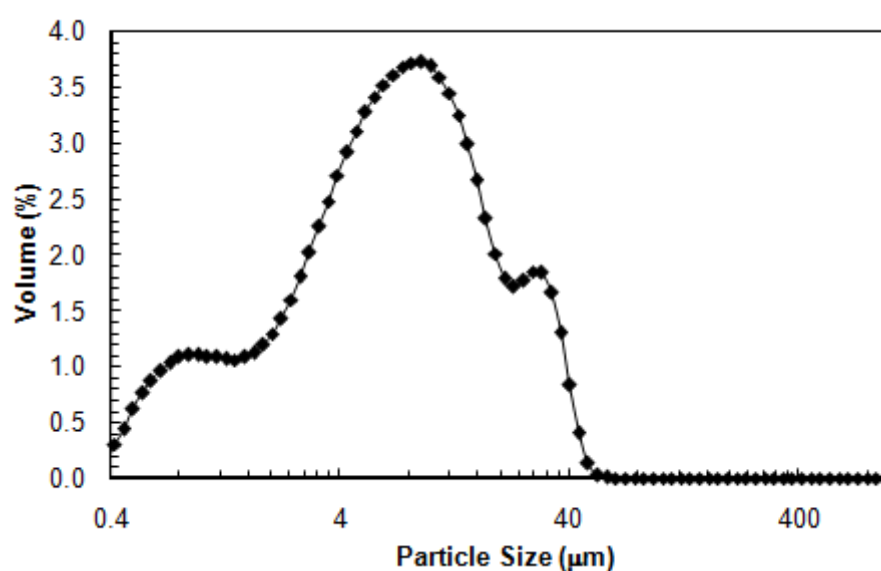
The chemical composition of FA as determined from the X-ray fluorescence spectrometer (XRF) is shown in Table 5.6. The FAs from both sources contain silica (SiO_2) as a major content, and the SiO_2 content is greater in the Rocktron. In this section, the FAs originated from Mae Moa power station and RockTron international limited are denoted as MMFA and RKFA, respectively.

5.5.2 RKFA Particle Size and Its Distribution

The particle size distribution of RKFA is rather broad with the sizes ranging from submicron to 60 microns as illustrated in Figure 5.131. The reported mean particle size of RKFA is of 10.17 microns, which appears to be smaller than that of MMFA as reports earlier in Figure 5.1.

Table 5.6 Chemical compositions of Mae Moh and Rocktron FAs

Composition	Content (%)	
	Mae Moh Fly Ash (MMFA)	Rocktron Fly Ash (RKFA)
SiO ₂	37.55	51.61
Al ₂ O ₃	22.40	22.99
CaO	15.10	4.27
Fe ₂ O ₃	14.50	8.44
K ₂ O	2.97	4.71
SO ₃	2.71	0.46
MgO	2.28	1.92
Na ₂ O	1.34	1.01
TiO ₂	0.46	1.46
BaO	0.13	0.27
SrO	0.12	0.01
MnO	0.10	0.10
ZrO ₂	0.03	0.04
Cr ₂ O ₃	0.02	0.04
CuO	0.02	0.05

**Figure 5.131** Particle size distribution of RKFA

5.5.3 Morphology of RKFA

Figure 5.132 shows SEM micrographs of RKFA particles which are spherical in shape similar to the MMFA. However, the RKFA particles are solid sphere containing no encapsulated sub-spheres, unlike the MMFA ones illustrated previously in Figure 5.2.

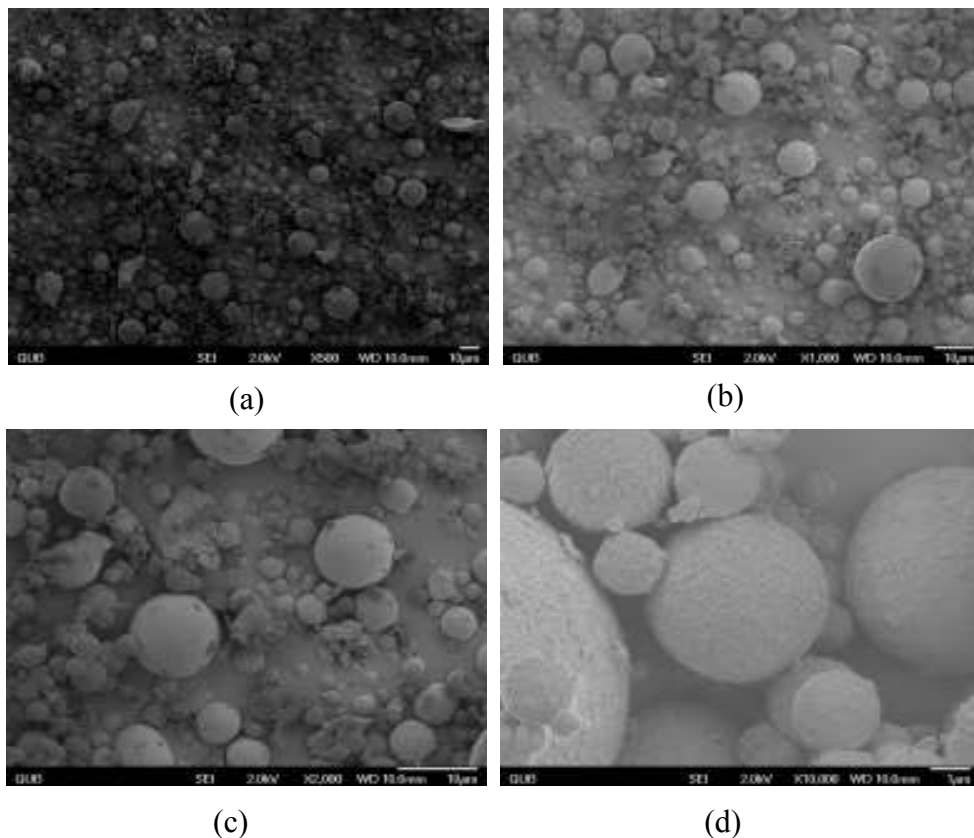


Figure 5.132 SEM micrographs of Rocktron fly ash (RKFA) with various magnification powers : (a) x500; (b) x1,000; (c) x2,000 and (d) x10,000

5.5.4 X-Ray Diffractometry (XRD)

Mineralogical constituents and phase proportions of RKFA determined via X-ray diffractometer are illustrated in Figure 5.133. Obviously, the RKFA consists crystalline phases similar to the MMFA, exhibiting relatively high intensity peak at 2 Theta of about 26 corresponding with quartz (highly crystalline structure) [118, 119]. In addition, the XRD result of RKFA shows a small peak at 2 Theta of 16-17 suggesting the presence of crystalline structure of mullite [120, 121]. It is noticeable that the composition of FAs depends strongly on the origin and processing of FAs.

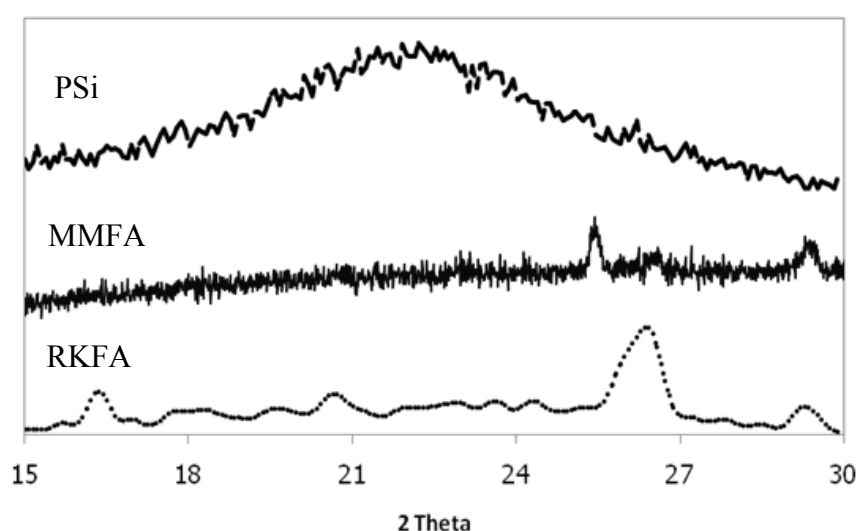


Figure 5.133 Comparison XRD results of precipitated silica (PSi), fly ash from Mae Moh (MMFA) and Rocktron (RKFA)

5.5.5 Cure Characteristics

Cure characteristics of NR filled with different FAs with and without silane (Si-69) are shown in Figures 5.134-5.136. Clearly, cure curves of NR display an increase in torque by incorporating either MMFA or RKFA. A further increase in torque with reduced magnitude of cure marching is found by the use of Si-69 silane. Also, the rate of cure marching is increased by the addition of FA especially for the MMFA as illustrated in Figure 5.135.

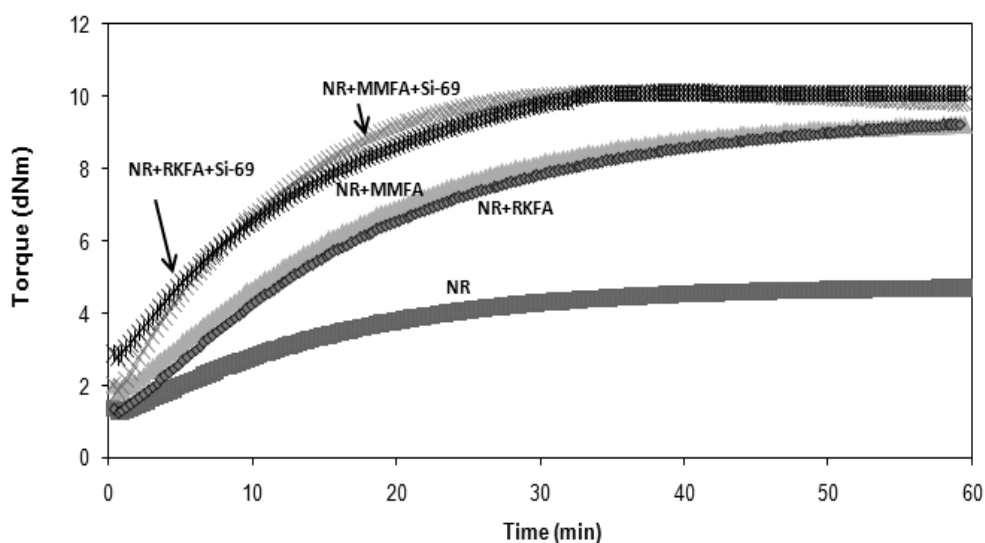


Figure 5.134 Cure curve of NR filled with MMFA or RKFA with and without silane (Si-69) under peroxide curing

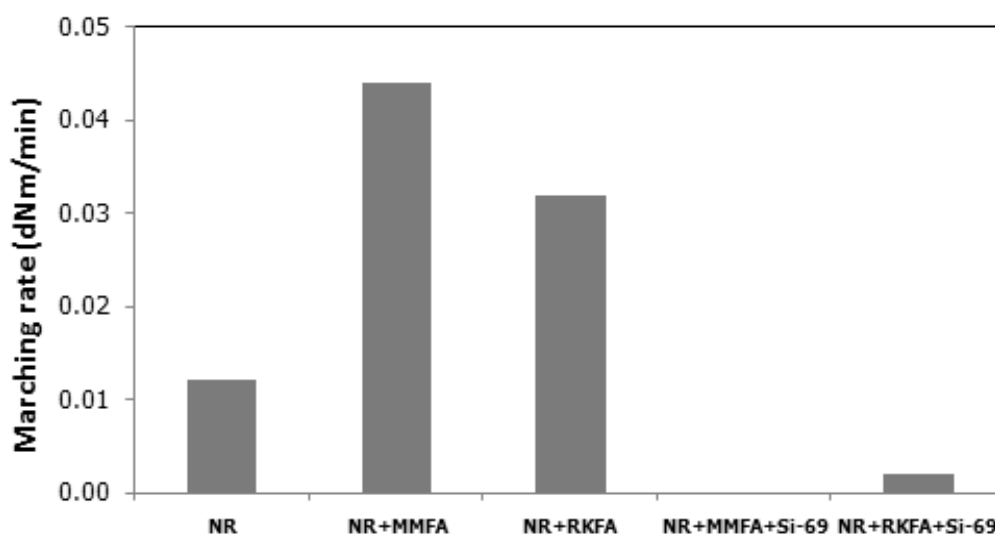


Figure 5.135 Rate of marching of NR filled with MMFA or RKFA with and without silane (Si-69) under peroxide curing

Results of scorch and cure times of NR filled with 2 different sources of FA with and without silane (Si-69) cured with peroxide are shown in Figure 5.136. The systems with RKFA show slightly shorter scorch time than those with MMFA. By contrast, no significant change in cure time is observed. The results of torque difference are illustrated in Figure 5.137. Both FAs yield the compounds with comparable torque difference. In addition, as evidenced in Figure 5.138, the incorporation of silane offers enhancement in crosslink density which is in a good agreement with the increase in torque difference regardless of FA origin.

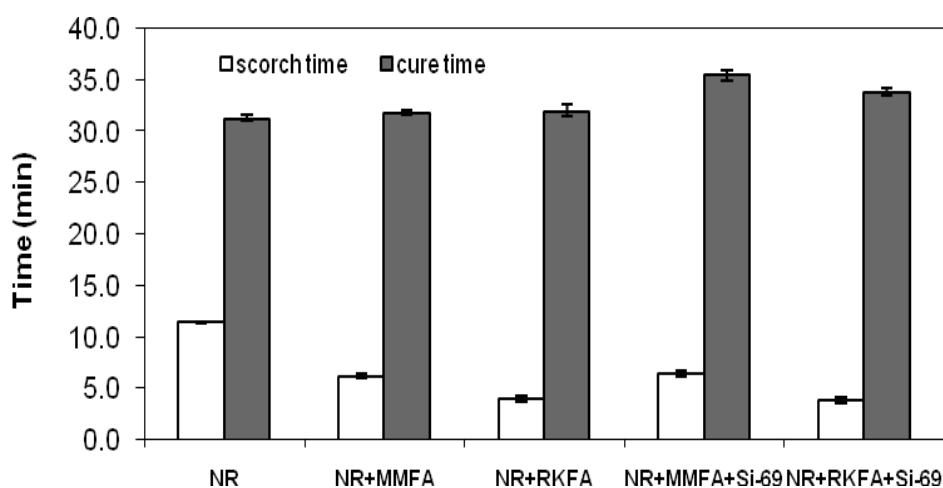


Figure 5.136 Scorch time and cure time of NR filled with MMFA or RKFA with and without silane (Si-69) under peroxide curing

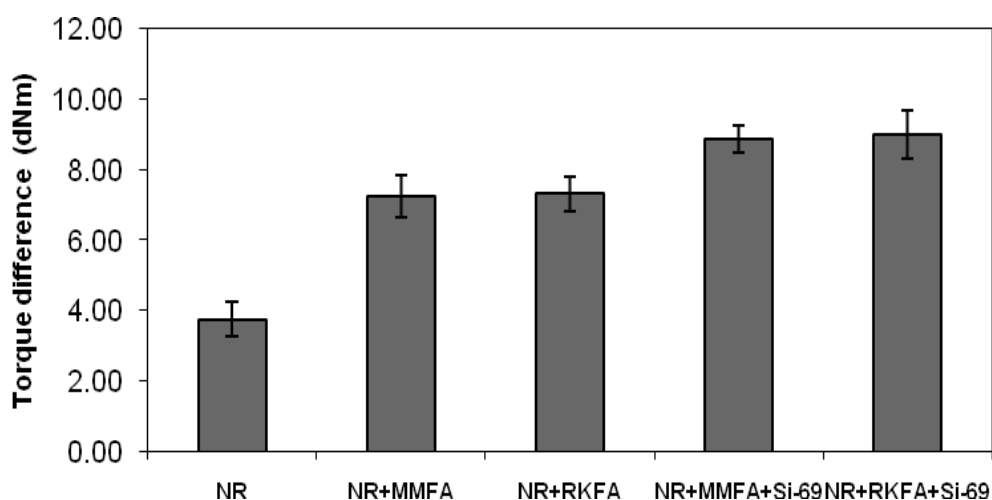


Figure 5.137 Torque difference of NR filled with MMFA or RKFA with and without silane (Si-69) under peroxide curing

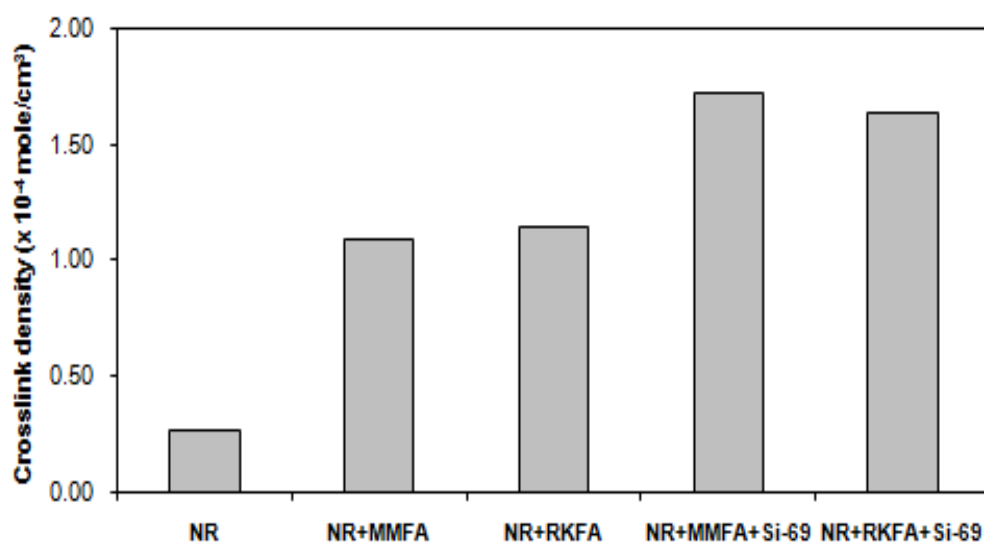


Figure 5.138 Crosslink density (as determined from Flory-Rehner equation via swelling test) of NR filled with MMFA or RKFA with and without silane (Si-69) under peroxide curing

5.5.6 Viscoelastic Properties of Uncured Compounds

(A) Time Sweep Test

It is necessary to know whether the thermal history during the mixing process strongly affect the thermal stability of FA/NR compounds. Consequently, a time sweep test was performed, and the results obtained are illustrated in Figure 5.139. It is clear that no significant change in storage modulus (G') as a function of time is observed. This means all compounds have comparable thermal stability.

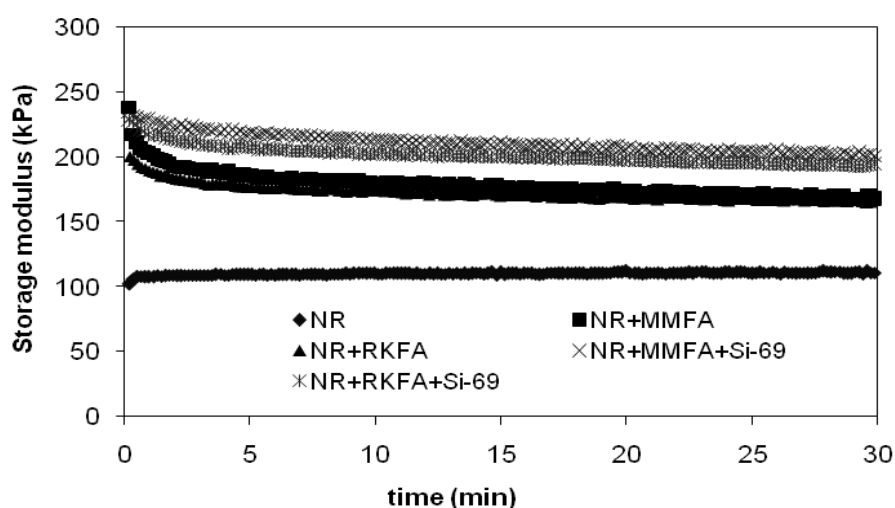


Figure 5.139 Storage modulus (G') as a function of time of uncured NR filled with MMFA or RKFA with and without silane (Si-69) under peroxide curing

(B) Strain Sweep Test

Figure 5.140 displays the plot of G' as a function of strain in uncured NR compounds filled with MMFA and RKFA compounds. The results show that the G' at low strain of compounds slightly increases with the addition of either MMFA or RKFA. After silane treatment (Si-69), the compounds with MMFA or RKFA show further rise in G' . Referred to Figure 5.141, the magnitude of Payne effect of FA filled compounds increases after surface treatment of FA with Si-69.

Unexpectedly, the treated MMFA demonstrates significantly more magnitude of Payne effect than the treated RKFA. The explanation is not know at present.

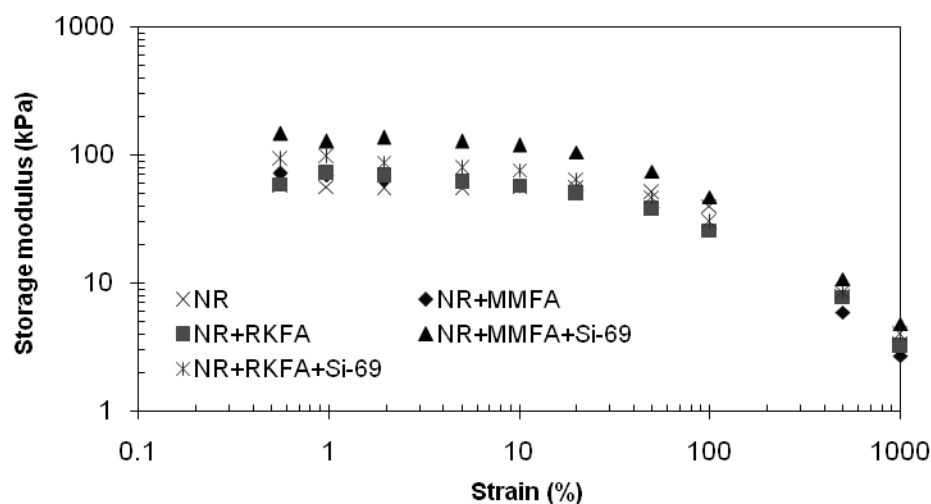


Figure 5.140 Storage modulus (G') as a function of strain amplitude of uncured NR filled with MMFA or RKFA with and without silane (Si-69)

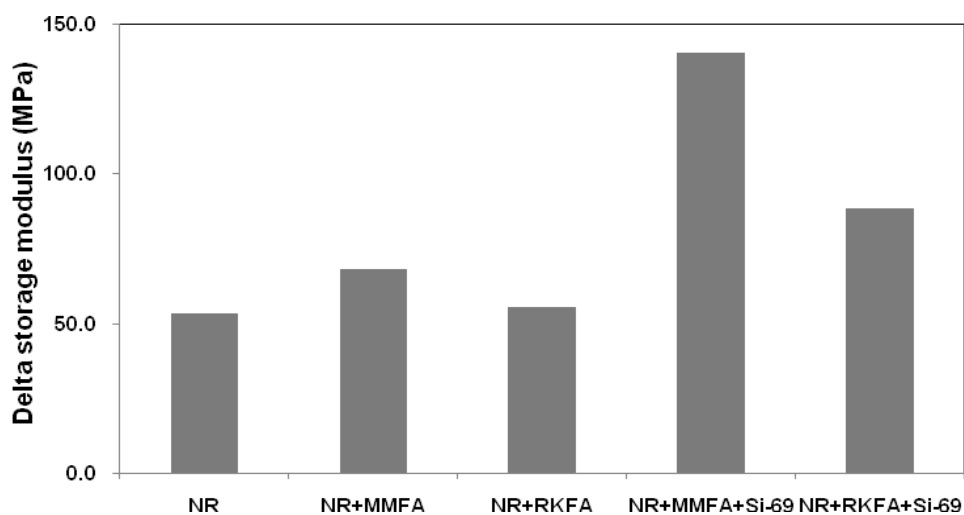


Figure 5.141 Payne effect magnitude of uncured NR filled with MMFA or RKFA with and without silane (Si-69)

(C) Frequency Sweep Test

Figure 5.142 exhibits the frequency dependence of G' in uncured NR compounds filled with MMFA or RKFA with and without Si-69. All compounds show strong frequency dependent G' with greater magnitude of frequency-dependent responses in the filled systems (as determined from slopes of the plots). Also, the damping factor of FA filled compounds show greater magnitude of damping factor than that of unfilled compound as shown in Figure 5.143. Both results imply the domination of viscous response over the elastic response via the ball bearing effect as discussed previously. Notably, the damping factors of compounds with MMFA and RKFA compounds are decreased after silane treatment. This might be explained by the interaction of development NR and FA linkages by the silane coupling reaction. It must be noted that both MMFA and RKFA give comparable viscoelastic behaviour as a function of frequency.

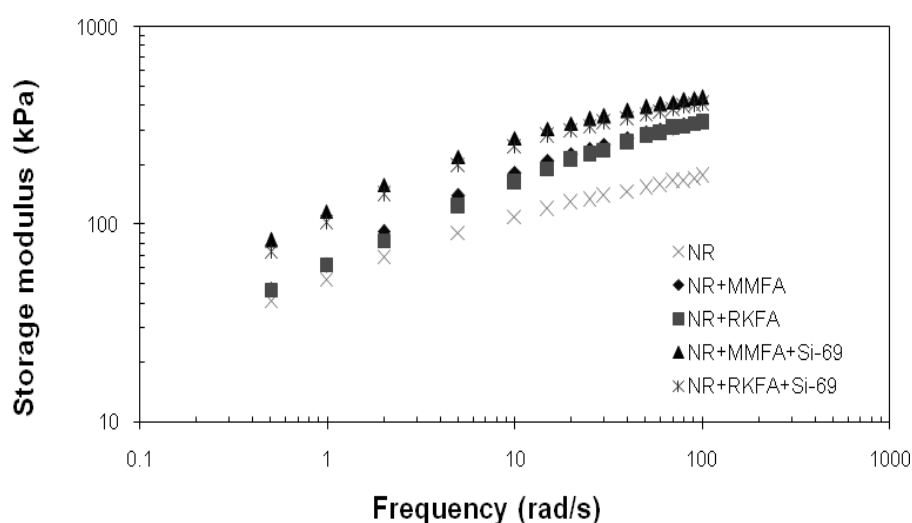


Figure 5.142 Storage modulus (G') as a function of frequency of uncured NR filled with MMFA or RKFA modified with silane (Si-69) under peroxide curing

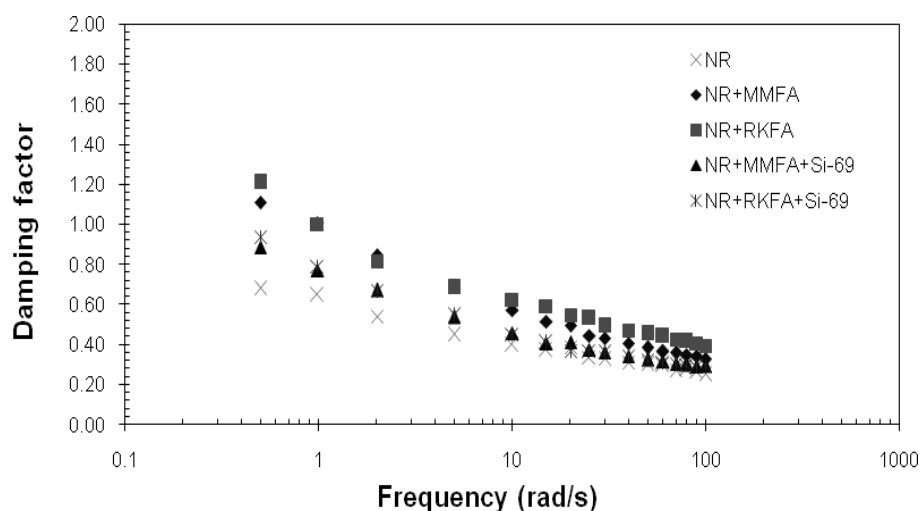


Figure 5.143 Damping factor (tan delta) as a function of frequency of uncured NR filled with MMFA or RKFA modified with silane (Si-69) under peroxide curing

The results of complex viscosity of NR compounds filled with MMFA and RKFA compounds are shown in Figure 5.144. The complex viscosity of MMFA and RKFA compounds gives similar degree of pseudoplastic behaviour. Furthermore, the treatment of MMFA and RKFA with Si-69 yields slight increase in complex viscosity as a result of increased restriction to molecular mobility.

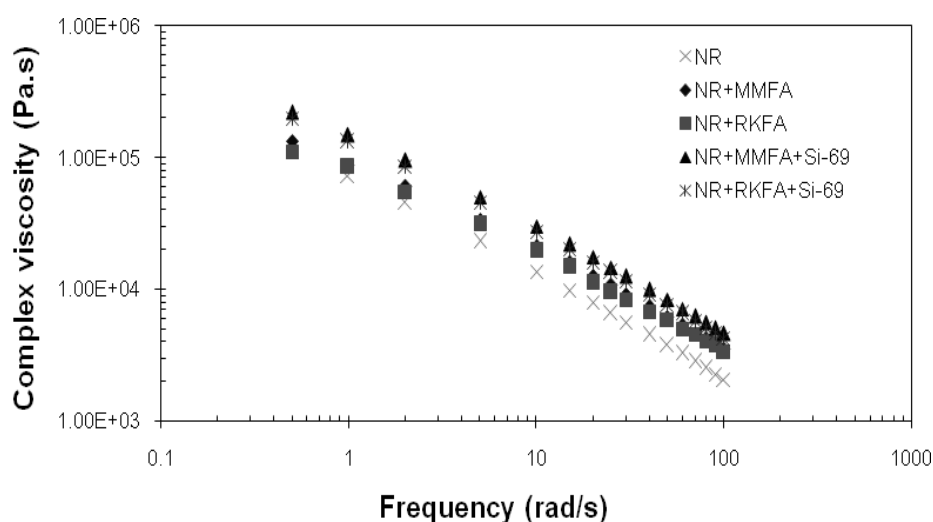


Figure 5.144 Complex viscosity as a function of frequency of uncured NR filled with MMFA or RKFA modified with silane (Si-69) under peroxide curing

5.5.7 Tensile Properties

Figures 5.145 to 5.147 show the modulus at 100 % strain (M100), tensile strength and elongation at break of NR composites with and without Si-69, respectively. The M100 increases with the additions of MMFA and RKFA, and further increases with silane surface treatment of FA particles. The modulus of the NR/FA composites is in line with the elongation at break as shown in Figure 5.146, and the crosslink density as evidenced in Figure 5.138. The tensile strength appears to increase after FA addition, particularly the system with RKFA. The higher tensile strength in the system with RKFA is due probably to the smaller particle size of RKFA.

After silane surface treatment of FA particles, the tensile strength increases in both systems, but more pronounced in the system with MMFA. The enhancement in tensile strength is believed to be caused by the increased crosslink density as a result of free sulfur released from Si-69. However, as illustrated in Table 5.7, there is no significant increase in bound rubber content that means the effect of rubber-FA interaction could probably be disregarded.

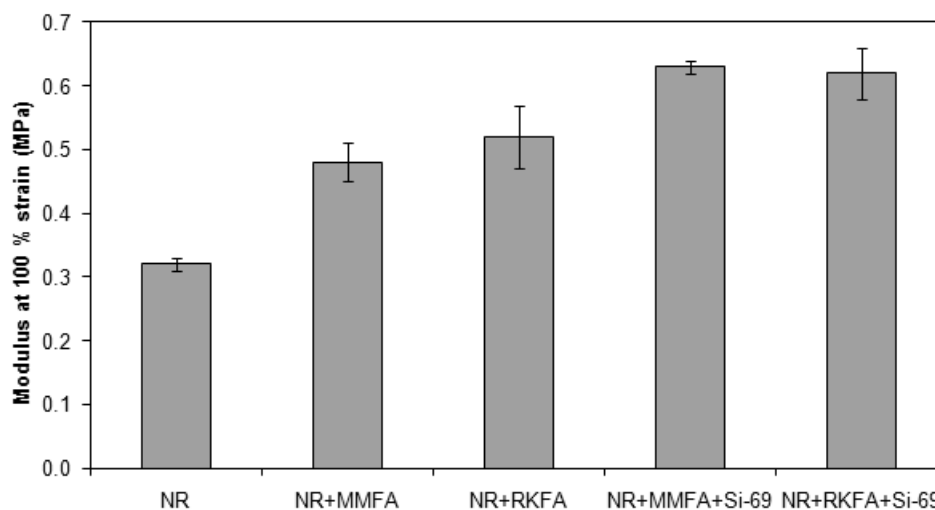


Figure 5.145 Modulus at 100 % strain (M100) of FA filled NR vulcanizates with MMFA or RKFA with and without silane (Si-69) under peroxide curing

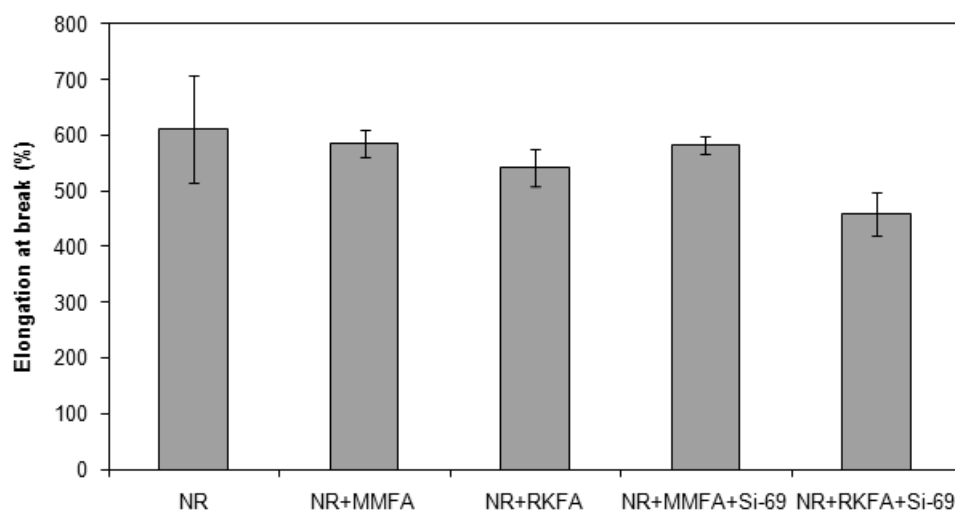


Figure 5.146 Elongation at break of FA filled NR vulcanizates with MMFA or RKFA with and without silane (Si-69) under peroxide curing

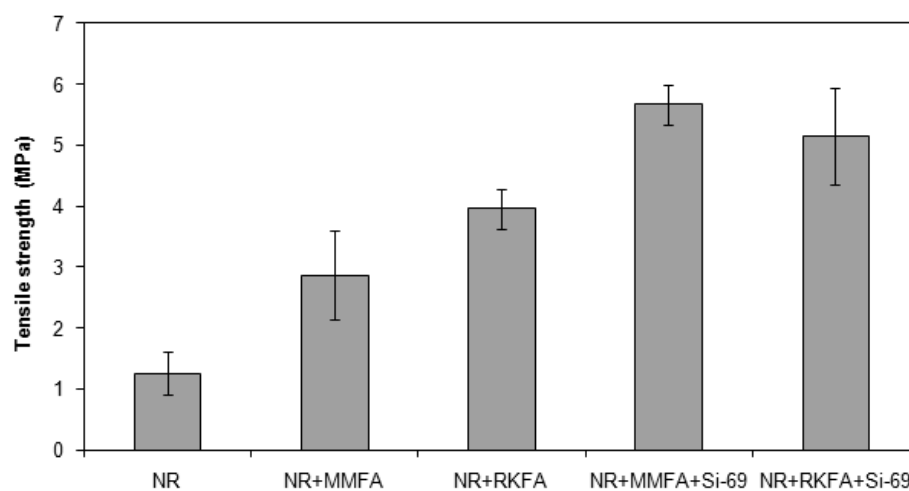


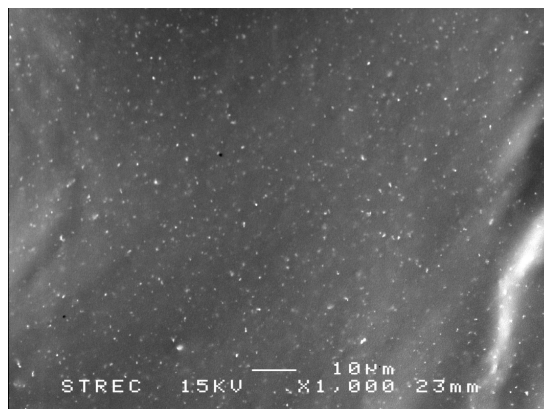
Figure 5.147 Tensile strength of FA filled NR vulcanizates with MMFA or RKFA with and without silane (Si-69) under peroxide curing

Table 5.7 Bound rubber content of FA filled NR vulcanizates with MMFA or RKFA with and without silane (Si-69) under peroxide curing

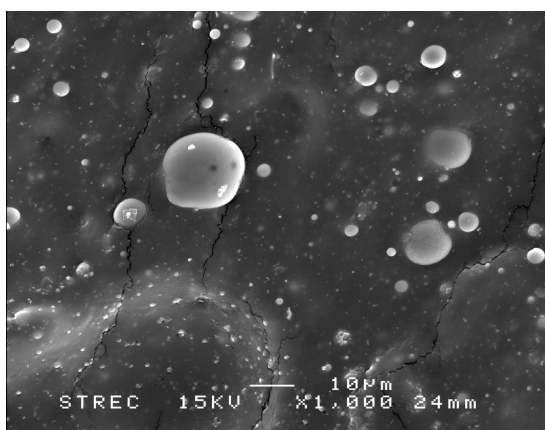
Systems	Bound rubber content (%)
NR	0.0000
NR+MMFA	0.0142
NR+RKFA	-0.0017
NR+MMFA+Si-69	0.0809
NR+RKFA+Si-69	0.0708

5.5.8 Morphological Results

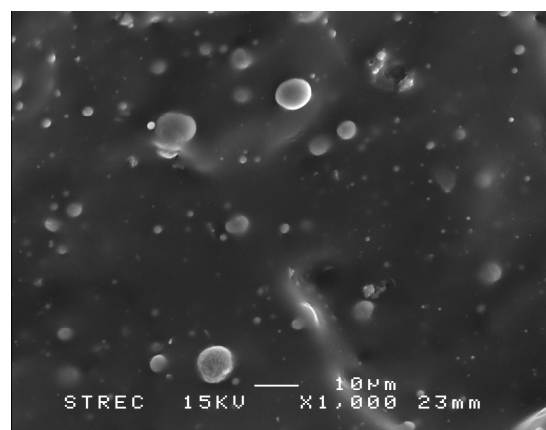
The morphological results of MMFA and RKFA filled NR vulcanizates with and without silane (Si-69) treatment are illustrated in Figure 5.147. There are the interfacial gaps between FA and rubber matrix which seem to disappear by silane surface treatment of FA particles. The marginal enhancement in wettability and interfacial adhesion lead to the slight increase in tensile strength as shown previously.



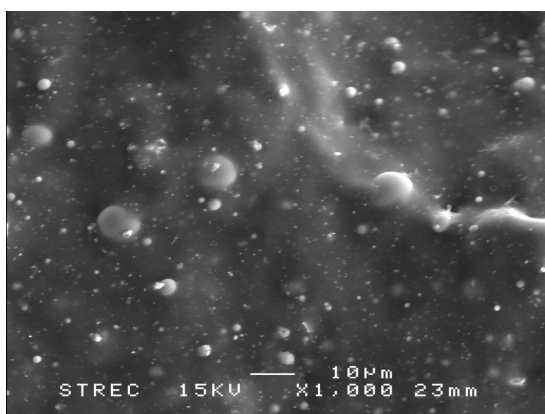
(a) NR



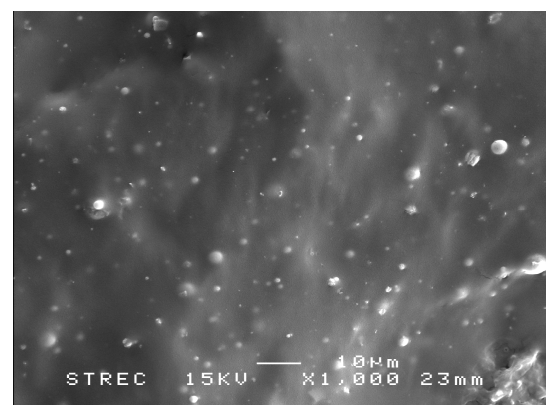
(b) NR+MMFA



(c) NR+RKFA



(d) NR+MMFA+ Si-69



(e) NR+RKFA+ Si-69

Figure 5.148 SEM micrographs of FA filled NR vulcanizates cured via peroxide with MMFA or RKFA with and without silane (Si-69) under peroxide curing: (a) NR; (b) NR+MMFA; (c) NR+RKFA; (d) NR+MMFA+Si-69 and (e) NR+RKFA+Si-69

From the results, it is evident that the properties are gained by the use of MMFA are slightly superior to those of RKFA in the system cured with peroxide. This is due to the difference in characteristics of both FAs, i.e., particle size, chemical composition, mineralogical constituent. By adding Si-69 silane, the G' and tensile strength are enhanced because of the increased crosslink density as a result of free sulfur released from Si-69.

CHAPTER VI

CONCLUSIONS

From the results of the present study, it can be summarized as follows:

The largest amount of composition in FA is of silicate (SiO_2) or silica. The incorporation of FA in NR increases storage modulus and viscosity under both oscillatory and steady shear flow. Moreover, the higher the FA loading, the larger the magnitude of viscous response in the FA-filled NR compounds. This is because the ball-bearing effect of FA associated with molecular degradation induced by the presence of complex substances (fatty acid salts of metals) which are formed via fatty acid in non-rubber component and of metal such as iron and copper in FA particles. With the use of Cox-Merz concept, the results of both oscillatory and steady shear flows can effectively be superimposed in the case of FA-filled compounds.

Storage stability of FA filled NR compounds was investigated via viscoelastic responses. Results of normalized modulus and molecular weight results are in line, i.e., the storage duration of FA filled NR compounds leads to decreases in elastic modulus and molecular weight, particularly in the compounds filled with high FA loading. It is believed that the presence of fatty acid in non-rubber substances together with metal ions in FA particles could catalyze the degradation process of rubber molecules. Such degradation process could effectively be suppressed by the addition of amine-based antioxidant (6-PPD).

By adding 3 types of silanes, Si-69, Si-264 and VTEO, in the selected 100 phr FA filled NR compounds, it is found that the increase in storage modulus or G' is observed in compounds with all silanes, which is pronounced in the case of Si-69 silane especially 4% w/w of Si-69. Regarding the silane treatment techniques, the in-situ surface treatment technique yields the composites with higher G' than other techniques whereas the untreated system demonstrates the lowest G' .

With various curing systems, the cure reversion phenomenon is observed in sulfur system with increasing FA loading which is believed to be the result of heavy metals containing in FA particles accelerating the oxidation process. On the other hand, the peroxide curing system exhibits the marching phenomenon particularly at high FA loading greater than 150 phr. It is possible that the metal oxide in FA might accelerate the oxidation on NR chains, leading to generated free radicals on NR chains and thus the increased rate of marching. Furthermore, the G' in both sulfur and peroxide systems increases with increasing FA loading due to the hydrodynamic effect. Both curing systems show the increase in frequency dependence of G' or the decrease in elastic response with increasing FA loading. The results can be explained by the FA ball bearing effect. Additionally, the sulphur cured NR filled with FA shows progressive decrease in tensile strength with increasing FA loading. The decreased tensile strength is due to poor filler-rubber interaction.

By treating FA with silane, in all curing system, the optimum properties are achieved by the use of Si-69. This is because the Si-69 can release free sulfur during curing process thus improves the co-crosslink with NR. On the other hand, other silanes give no enhancement in tensile strength because of the small amount of active silanol groups on FA surfaces resulting from FA crystalline structure.

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APPENDICES

APPENDIX A

CHARACTERIZATION OF FA AND VISCOELASTIC BEHAVIOR DATA OF FA FILLED NR COMPOUNDS WITH VARIOUS FA LOADINGS

Table A1 Particle size distribution of FA used in the present study

Size (micron)	Volume (%)		Size (micron)	Volume (%)		Size (micron)	Volume (%)	
	Lot1	Lot2		Lot1	Lot2		Lot1	Lot2
0.01	0	0	1.096	0.29	0.38	120.226	4.33	5.55
0.011	0	0	1.259	0.36	0.5	138.038	4.37	5.57
0.013	0	0	1.445	0.43	0.62	158.489	4.2	5.18
0.015	0	0	1.66	0.52	0.75	181.97	3.83	4.3
0.017	0	0	1.905	0.63	0.89	208.93	3.3	3.01
0.02	0	0	2.188	0.75	1.03	239.883	2.71	1.44
0.023	0	0	2.512	0.89	1.17	275.423	2.14	0.04
0.026	0	0	2.884	1.05	1.31	316.228	1.76	0
0.03	0	0	3.311	1.21	1.44	363.078	1.53	0
0.035	0	0	3.802	1.38	1.56	416.869	1.38	0
0.04	0	0	4.365	1.55	1.65	478.63	1.01	0
0.046	0	0	5.0123	1.71	1.73	549.541	0.74	0
0.052	0	0	5.754	1.87	1.79	630.957	0.21	0
0.06	0	0	6.607	2.01	1.84	724.436	0	0
0.069	0	0	7.586	2.13	1.89	831.764	0	0
0.079	0	0	8.71	2.23	1.95	954.993	0	0
0.091	0	0	10	2.29	2.02	1096.478	0	0
0.105	0	0	11.482	2.1	2.34	1258.925	0	0
0.12	0	0	13.183	2.2	2.38	1445.44	0	0
0.138	0	0	15.136	2.29	2.42	2511.886	0	0
0.158	0	0	17.378	2.38	2.48	2884.032	0	0
0.182	0	0	19.953	2.45	2.55	3311.311	0	0
0.209	0	0	22.909	2.49	2.63	3801.894	0	0
0.24	0	0	26.303	2.49	2.7	4365.158	0	0
0.275	0	0	30.2	2.45	2.74	5011.872	0	0
0.316	0	0	34.674	2.4	2.76	5754.399	0	0
0.363	0.04	0.02	39.811	2.36	2.79	6606.934	0	0
0.417	0.05	0.03	45.709	2.36	2.86	7585.776	0	0
0.479	0.06	0.04	52.481	2.46	3.01	8709.636	0	0
0.55	0.07	0.05	60.256	2.66	3.27	10000	0	0
0.631	0.1	0.07	69.183	2.97	3.66			
0.724	0.15	0.07	79.433	3.36	4.15			
0.832	0.19	0.15	91.201	3.76	4.7			
0.955	0.24	0.27	104.713	4.1	5.21			

Table A2 Time sweep results at 70°C of FA /NR compounds with various FA loadings

Time (min)	Storage modulus (kPa)					Time (min)	Storage modulus (kPa)				
	0	50	100	150	200		0	50	100	150	200
0.22	116.57	170.7	238.44	339.81	452.89	7.13	128.08	148.22	183.06	244.11	288.55
0.38	117.32	163.7	217.48	304.74	392.54	7.28	128.61	147.79	182.31	243.05	289.51
0.53	120.83	160.9	210.66	294.3	376.35	7.44	128.72	148.32	182.31	243.47	286.73
0.69	121.26	159.62	206.18	288.97	367.4	7.60	129.14	148.64	182.63	242.3	285.78
0.85	123.81	158.23	204.8	285.35	360.42	7.75	128.93	148.54	181.89	242.62	286.31
1.00	124.45	157.38	202.45	282.05	356.74	7.91	128.29	148.96	181.89	241.98	284.18
1.16	124.88	156.31	200.75	277.33	351.31	8.07	129.14	148.11	181.57	240.92	283.86
1.32	125.73	155.78	197.98	274.36	347.15	8.22	128.93	146.72	180.82	241.02	283.22
1.47	125.2	154.18	197.12	273.2	345.02	8.38	128.61	148.11	181.35	240.92	282.47
1.63	125.41	153.12	196.16	270.43	339.9	8.54	128.4	149.17	181.57	241.48	282.37
1.79	125.84	153.86	195.74	268.94	337.77	8.69	129.04	148.11	181.57	240.81	280.77
1.95	125.84	153.97	192.86	266.6	334.79	8.85	128.61	147.26	181.14	239.85	280.98
2.10	125.2	153.44	193.07	265.32	332.87	9.01	129.78	148.11	181.57	239.53	280.66
2.26	126.16	152.58	192.22	264.35	330.42	9.17	129.14	148.54	180.93	239.03	280.45
2.42	127.33	151.84	191.58	261.91	329.04	9.32	127.97	147.26	181.35	238.15	279.7
2.57	127.69	152.58	192.01	261.89	325.73	9.48	128.29	147.15	181.14	239.43	279.17
2.73	126.59	151.84	191.26	258.71	322.64	9.64	128.4	147.04	179.86	238.68	278.53
2.89	126.16	150.77	190.84	259.46	321.9	9.79	128.93	148.54	179.86	237.4	278.21
3.04	127.22	151.73	190.09	257.33	320.94	9.95	128.72	148.22	179.22	238.15	276.19
3.20	127.97	150.24	189.77	255.83	317.53	10.11	129.04	147.04	180.08	237.29	276.72
3.36	129.08	151.31	188.07	254.88	315.93	10.26	129.04	147.04	180.08	236.97	277.57
3.51	128.08	151.84	189.66	255.41	315.61	10.42	129.36	147.58	179.86	237.29	274.27
3.67	128.61	149.39	189.35	255.51	313.37	10.58	129.36	148.11	179.01	236.23	277.15
3.83	128.08	150.77	187.75	253.6	312.2	10.74	129.36	147.9	179.22	236.02	274.27
3.99	128.4	150.99	187.21	252.64	309.43	10.89	129.36	147.79	179.54	235.48	274.37
4.14	128.29	151.09	188.71	251.57	307.73	11.05	129.78	147.15	179.01	236.23	274.91
4.30	127.22	151.52	187.21	251.47	306.66	11.21	128.93	147.58	179.01	236.34	275.01
4.46	128.08	150.67	185.83	252.32	305.91	11.36	129.14	148.64	178.69	235.7	272.99
4.61	127.65	150.45	185.08	250.51	305.17	11.52	129.78	146.19	178.69	235.59	272.56
4.77	127.65	150.35	185.94	249.55	302.29	11.68	128.08	146.72	179.12	235.27	271.5
4.93	128.29	150.03	186.26	250.19	303.68	11.83	129.36	147.79	179.01	234.42	271.92
5.08	128.72	149.92	185.19	249.76	299.52	11.99	128.93	146.08	179.86	234.63	271.71
5.24	128.08	148.32	184.76	248.7	301.01	12.15	129.46	147.15	178.8	235.27	271.71
5.40	128.61	150.99	184.02	247.63	298.14	12.30	130.21	146.72	178.8	234.52	271.92
5.56	127.54	148.96	184.55	247.31	298.78	12.46	129.68	146.08	179.12	234.52	270.65
5.71	128.29	148.85	185.4	247.42	296.22	12.62	129.36	147.9	178.16	233.88	269.58
5.87	129.14	148.64	183.7	246.88	294.94	12.78	129.99	147.58	178.48	233.35	270.43
6.03	128.72	148.96	183.7	246.56	295.69	12.93	129.46	147.58	180.18	233.88	267.66
6.18	127.22	149.92	183.7	246.14	294.09	13.09	129.36	147.58	178.16	232.71	270.33
6.34	127.97	149.39	183.7	244.43	291.96	13.25	129.99	146.72	177.3	233.46	267.98
6.50	128.61	149.71	182.63	244.01	291.64	13.40	129.78	147.04	177.62	232.39	267.87
6.65	128.93	149.17	183.27	243.47	291.96	13.56	129.68	147.79	178.69	232.39	268.51
6.81	127.86	148.64	182.63	243.37	291.42	13.72	129.99	147.26	177.73	232.82	268.19
6.97	128.61	148.54	182.95	243.15	289.61	13.87	129.78	145.98	177.3	233.03	267.13

Table A2 Time sweep results at 70°C of of FA /NR compounds with various FA loadings (cont.)

Time (min)	Storage modulus (kPa)					Time (min)	Storage modulus (kPa)				
	0	50	100	150	200		0	50	100	150	200
14.03	129.99	146.08	177.62	232.07	267.87	21.09	129.68	144.06	172.3	223.87	255.83
14.19	130.53	147.47	176.56	231.01	266.49	21.25	129.99	144.27	173.36	224.19	255.83
14.35	131.81	146.08	177.3	230.9	266.92	21.41	130.53	144.38	172.08	223.97	256.15
14.50	129.99	147.47	177.62	231.22	266.6	21.57	131.27	144.06	173.47	223.97	257.33
14.66	129.36	146.19	177.73	231.22	264.68	21.72	129.78	143.63	172.4	223.55	256.58
14.82	129.14	145.02	177.73	232.07	267.13	21.88	128.93	144.27	172.4	223.12	255.73
14.97	131.06	147.15	177.41	231.01	265.42	21.09	129.68	144.06	172.3	223.87	255.83
15.13	130.85	145.02	177.41	231.43	265	21.25	129.99	144.27	173.36	224.19	255.83
15.29	129.78	147.58	177.3	230.58	263.4	21.41	130.53	144.38	172.08	223.97	256.15
15.44	130.85	145.76	176.67	229.52	264.47	22.04	130.42	143.31	172.08	222.7	256.26
15.60	129.68	147.04	176.35	230.37	264.47	22.19	129.99	145.34	171.66	222.06	255.94
15.76	129.68	145.13	176.24	229.94	264.04	22.35	130.1	144.91	172.08	222.48	255.83
15.92	129.68	147.04	176.03	230.16	263.72	22.51	131.59	143.85	172.4	223.23	254.45
16.07	129.68	145.02	175.92	229.94	263.4	22.66	131.17	144.91	173.04	223.23	254.45
16.23	130.21	145.76	175.81	228.88	264.04	22.82	130.42	144.59	172.08	221.1	254.88
16.39	130.85	146.08	176.56	230.16	263.61	22.98	130.1	144.7	171.02	222.48	254.13
16.54	129.68	145.13	175.6	228.88	262.55	23.14	131.81	144.38	171.23	222.8	255.09
16.70	131.27	144.59	175.17	228.56	261.8	23.29	131.59	143.85	171.55	222.7	254.45
16.86	130.42	145.13	175.6	229.09	263.61	23.45	130.42	143.63	170.49	221.63	254.88
17.01	130.1	145.76	174.96	228.13	263.19	23.61	130.53	143.95	170.59	222.38	254.34
17.17	130.85	146.4	174.75	228.45	261.48	23.76	130.53	144.59	171.55	222.38	254.66
17.33	130.53	145.13	174.96	228.24	261.16	23.92	130.1	144.59	171.76	222.48	254.02
17.48	130.42	145.02	175.81	228.02	261.8	24.08	131.49	143.95	171.66	222.38	253.6
17.64	130.53	145.02	175.49	227.7	261.27	24.23	131.06	143.85	171.34	221.1	253.38
17.80	129.78	145.45	175.17	227.7	261.16	24.39	130.42	142.99	170.59	220.67	252.74
17.96	129.78	144.91	174.43	227.38	260.42	24.55	130.42	143.53	170.49	220.78	253.38
18.11	131.49	144.7	174.85	227.7	260.84	24.71	130.53	143.21	171.23	220.67	253.6
18.27	130.21	144.59	173.89	227.17	260.52	24.86	131.06	143.95	171.76	220.67	253.06
18.43	130.42	145.02	174.21	226.32	259.35	25.02	132.34	144.06	170.91	219.61	253.06
18.58	130.1	146.4	174.53	227.81	261.16	25.18	131.59	143.95	170.49	220.35	252.53
18.74	130.21	144.38	174.53	226.43	259.14	25.33	130.74	143.95	169.85	219.61	252.74
18.90	130.85	146.51	174.11	226.75	258.71	25.49	130.74	143.63	171.55	219.18	251.68
19.05	130.85	144.06	174.75	226.64	258.92	25.65	132.13	143.53	169.21	219.18	251.15
19.21	130.1	146.4	173.68	226.43	260.1	25.80	130.74	143.31	169.63	220.03	251.57
19.37	129.99	143.63	172.72	226.75	259.14	25.96	131.06	142.99	170.91	219.5	251.15
19.53	130.42	145.45	174.96	226.43	258.39	26.12	130.53	143.53	170.7	218.97	251.68
19.68	130.1	145.34	172.08	225.04	258.07	26.27	130.21	143.53	170.49	219.71	250.93
19.84	130.53	144.59	174.53	225.89	257.65	26.43	131.27	143.31	171.02	218.11	250.51
20.00	130.21	145.66	173.36	226.64	257.33	26.59	131.91	143.21	169.53	218.11	251.31
20.15	129.99	144.59	173.36	224.29	257.33	26.75	131.17	143.31	168.89	218.43	250.4
20.31	130.21	144.27	173.36	224.93	257.01	26.90	130.21	143.63	169.95	219.29	249.76
20.47	129.99	145.45	172.72	224.61	257.33	27.06	131.06	143.21	169.85	217.9	250.93
20.62	129.46	146.08	172.72	224.61	257.86	27.22	131.06	142.99	169.21	218.43	251.15
20.78	130.74	144.7	172.72	224.93	257.65	27.37	130.53	142.89	170.7	217.48	249.76
20.94	130.89	144.91	173.04	224.51	257.65	27.53	130.74	142.99	169.42	218.22	249.76

Table A2 Time sweep results at 70°C of of FA /NR compounds with various FA loadings (cont.)

Time (min)	Storage modulus (kPa)					Time (min)	Storage modulus (kPa)				
	0	50	100	150	200		0	50	100	150	200
27.69	131.06	142.78	169.1	217.05	250.93	29.10	130.53	142.89	168.14	215.98	249.33
27.84	130.21	143.53	168.89	217.9	249.01	29.26	131.17	143.21	168.14	217.05	248.38
28.00	130.21	142.14	169.95	218.86	250.4	29.41	132.55	142.99	168.35	217.37	248.8
28.16	130.74	142.89	169.95	217.79	249.12	29.57	130.85	142.99	169.53	217.05	248.8
28.32	131.27	143.53	169.53	216.3	249.55	29.73	130.85	143.31	169.63	217.16	249.12
28.47	131.59	143.95	169.42	217.9	250.4	29.88	132.23	142.78	167.71	216.3	248.27
28.63	131.81	142.78	169.53	217.79	250.08	29.10	130.53	142.89	168.14	215.98	249.33
28.79	130.85	143.21	168.46	217.05	249.87	29.26	131.17	143.21	168.14	217.05	248.38
28.94	130.74	142.99	168.14	216.3	247.74	29.41	132.55	142.99	168.35	217.37	248.8

Table A3 Strain sweep results at 1 rad/s and 70°C of of FA /NR compounds with various FA loadings

Strain (%)	Storage modulus (kPa)				
	0	50	100	150	200
0.54	60.812	58.85	58.85	111.82	178.51
0.96	63.2	55.439	60.982	103.12	139.71
1.93	71.516	46.014	64.863	92.028	118.08
5.01	60.948	49.441	53.916	76.718	105.91
10.07	61.67	48.241	51.003	68.099	84.911
50	54.177	36.428	33.343	38.615	41.336
100.09	43.11	25.605	21.744	24.296	25.11
499.95	8.4299	5.6761	5.5269	6.9676	8.2468
999.87	3.3345	2.3577	2.5904	3.3355	3.8936
1200.01	2.613	1.8633	2.0874	2.7687	3.1085

Table A4 Strain sweep results at 100 rad/s and 70°C of FA /NR compounds with various FA loadings

Strain (%)	Storage modulus (kPa)				
	0	50	100	150	200
0.5	227.39	340.02	505.78	736.1	1062.6
0.92	212.13	316.45	464.74	695.5	986.33
1.93	206.79	306.02	433.71	651.69	865.95
5.04	199.42	296.7	416.97	581.19	746.24
10.13	194.85	282.46	384.42	514.07	631.38
12.08	194.53	278.63	373.4	489.53	587.4
15.02	196.77	272.22	355.59	454.09	537.52
19	194.24	264.29	334.58	415.93	479.45
24.98	192.18	253.08	306.62	370.16	411.74
30.02	189.52	241.01	283.86	333.84	364.63

Table A5 Delta storage modulus (Payne effect) of FA /NR compounds with various FA loadings

FA loading (phr)	0	50	100	150	200
Delta storage modulus	48.9096	44.1926	48.5155	63.5662	87.1267

Table A6 Relative storage modulus (relative G') at 1, 10 and 100% strain of NR compounds filled with various FA loadings as a function of FA loadings and SIDISTAR

FA loading (phr)	Volume fraction	Relative G'			
		Guth-Gold Equation	Experiment at 1 % strain	Experiment at 10 % strain	Experiment at 100 % strain
0	0.00	1.00	1.00	1.00	1.00
50	0.16	1.74	0.69	0.75	0.59
100	0.27	2.71	1.31	0.81	0.51
150	0.36	3.70	1.56	1.05	0.54
200	0.43	4.63	2.20	1.25	0.55
SIDISTAR 200	0.43	4.63	2.44	1.56	0.74

Table A7 Frequency sweep results of NR compounds filled with various FA loadings

Frequency	Storage modulus (kPa)					Damping factor					Complex viscosity (Pa-s)				
	0	50	100	150	200	0	50	100	150	200	0	50	100	150	200
0.5	44.858	34.65	34.566	52.434	71.896	0.736	1.088	1.244	1.275	1.184	1.11E+05	1.02E+05	1.10E+05	1.69E+05	1.43E+05
0.99	62.525	50.49	52.443	76.505	98.056	0.595	0.948	1.179	1.169	1.126	73120	69939	81513	1.18E+05	98565
2	79.731	72.511	81.123	112.53	138.69	0.526	0.841	0.996	1.025	1.029	45041	47371	57250	80547	69342
5.01	104.97	111.61	131.9	180.3	217.1	0.45	0.683	0.812	0.838	0.879	22999	26999	33943	47005	43371
10	127.1	147.95	182.54	248.37	294.7	0.379	0.577	0.676	0.71	0.76	13590	17082	22037	30457	29468
15	137.86	168.34	213.57	290.38	344.12	0.36	0.516	0.608	0.647	0.707	9769.7	12634	16668	23061	22948
20	149.48	182.91	237.79	319.05	385.59	0.371	0.519	0.545	0.594	0.644	7972.1	10302	13542	18557	19278
25	155.16	199.93	254.81	344.5	409.05	0.329	0.465	0.542	0.58	0.631	6534.5	8822.2	11593	15932	16364
30	161.14	209.62	271.15	365.66	432.05	0.304	0.449	0.52	0.557	0.61	5614.2	7658.7	10185	13950	14400
40	171.56	228.14	294.55	397.7	472.22	0.3	0.43	0.489	0.528	0.578	4477.9	6208.1	8197.4	11241	11805
50	179.8	242.45	317.26	425.39	503.8	0.287	0.407	0.459	0.498	0.541	3740.6	5234.7	6981.1	9503.7	10075
60	186.28	252.47	328.87	442.01	533.38	0.281	0.395	0.452	0.481	0.522	3225.1	4524	6014	8174.5	8888.9
69.99	190.04	262.33	344.04	461.95	554.05	0.268	0.378	0.436	0.464	0.5	2810.9	4006.8	5361.7	7275.4	7915.5
80	194.56	269.46	348	473.44	563.9	0.266	0.373	0.428	0.456	0.505	2516.4	3594.9	4731.7	6505.3	7049.1
90	196.6	278.14	360.38	485.15	563.55	0.257	0.361	0.416	0.446	0.504	2255.7	3286.2	4336.6	5902.3	6261.9
100	199.89	285.81	368.17	502	600.94	0.254	0.351	0.405	0.426	0.476	2062.3	3029.5	3972.6	5456.9	6009.5
120	211.63	298.9	393.85	525.16	600.49	0.237	0.334	0.376	0.415	0.462	1812.3	2625.8	3506.4	4738.6	5004.2
140	214.61	306.17	403.19	535.54	647.6	0.241	0.327	0.379	0.399	0.455	1576.7	2301	3080	4118.8	4625.7
160	218.96	314.13	414.11	551.02	658.34	0.241	0.33	0.362	0.398	0.448	1407.7	2067.6	2752.6	3706.1	4114.6

Table A8 Frequency sweep results of NR compounds filled with various silica loadings

Frequency	Storage modulus (kPa)					Damping factor					Complex viscosity (Pa-s)				
	0	50	100	150	200	0	50	100	150	200	0	50	100	150	200
0.5	43.409	80.67	222.59	483.51	663.92	0.699	0.652	0.568	0.544	0.541	1.05E+05	1.92E+05	5.09E+05	1.10E+06	1.50E+06
0.99	60.254	109.07	292.35	596.06	797.47	0.631	0.57	0.484	0.476	0.492	71617	1.26E+05	3.26E+05	6.64E+05	8.93E+05
2	79.112	137.68	357.95	691.78	911	0.541	0.48	0.437	0.449	0.459	44964	76352	1.95E+05	3.79E+05	5.01E+05
5.01	105.67	178.2	445.82	820.97	1074.9	0.462	0.409	0.38	0.41	0.42	23259	38470	95280	1.77E+05	2.33E+05
10	128.38	213.39	516.07	928.8	1200.5	0.383	0.351	0.338	0.37	0.381	13744	22615	54467	99013	1.28E+05
15	141.21	233.2	556.08	995.18	1277	0.353	0.316	0.308	0.345	0.359	9985.3	16307	38801	70204	90469
20	153.78	241.93	589.96	1039.5	1325.5	0.347	0.307	0.288	0.333	0.341	8137.9	12652	30697	54784	70018
25	158.69	257.27	610.82	1070.6	1369.4	0.321	0.296	0.286	0.315	0.325	6668.5	10732	25417	44900	57605
30	165.15	267.54	632.89	1097.3	1404.4	0.307	0.293	0.277	0.305	0.313	5758	9291.8	21887	38239	49042
40	174.06	279.7	660.09	1138.1	1456.7	0.291	0.277	0.269	0.294	0.303	4531.9	7256	17089	29653	38049
50	179.84	293.45	676.81	1187.8	1492.3	0.282	0.267	0.263	0.281	0.292	3737	6074.1	13995	24677	31086
60	188.12	300.35	698.17	1194.2	1535.5	0.271	0.261	0.246	0.271	0.283	3247.8	5172.9	11981	20622	26597
69.99	191.57	311.55	710.95	1203.1	1561.8	0.262	0.252	0.252	0.273	0.279	2829.3	4590.2	10474	17820	23163
80	197.01	315.81	733.47	1248.8	1579	0.26	0.246	0.241	0.262	0.27	2544.6	4065.7	9432.3	16137	20445
90	200.02	326.19	739.96	1269.3	1581.5	0.26	0.242	0.239	0.259	0.265	2296.2	3729.3	8454.5	14570	18181
100	203.86	330.14	755.43	1262.3	1612.7	0.24	0.237	0.239	0.261	0.259	2096.7	3392.8	7767.2	13047	16662
120	206.69	338.31	759.77	1314.6	1638.3	0.236	0.22	0.229	0.229	0.265	1769.8	2886.6	6494.8	11239	14125
140	214.52	347.28	798.32	1344.6	1648.8	0.248	0.224	0.244	0.236	0.254	1578.5	2542.2	5869.2	9867.7	12150
160	219.26	352.63	806.46	1340.8	1678.2	0.232	0.218	0.214	0.25	0.253	1406.7	2255.6	5154.4	8637.3	10819

Table A9 Power law index of FA/NR and FA/IR with various FA loadings determined from RPA2000

FA loading (phr)	Power law index				
	0	50	100	150	200
NR	0.233	0.315	0.344	0.325	0.332
IR	0.229	0.234	0.243	-	-

Table A10 Storage modulus and normalized storage modulus of FA filled NR compounds with 12-week storage duration

Week	Storage modulus (kPa)					Normalized storage modulus				
	0	50	100	150	200	0	50	100	150	200
0	53.447	40.165	43.307	56.316	66.623	1.000	1.000	1.000	1.000	1.000
3	61.67	48.241	51.003	68.099	84.911	1.154	1.201	1.178	1.209	1.274
5	61.502	47.325	52.206	67.874	69.589	1.151	1.178	1.205	1.205	1.045
6	63.256	49.159	51.716	60.012	62.527	1.184	1.224	1.194	1.066	0.939
7	64.588	48.985	48.453	62.054	63.348	1.208	1.220	1.119	1.102	0.951
9	64.196	48.549	43.488	49.014	50.719	1.201	1.209	1.004	0.870	0.761
11	62.44	40.283	33.99	42.868	39.74	1.168	1.003	0.785	0.761	0.596
12	60.713	39.681	32.364	38.89	36.122	1.136	0.988	0.747	0.691	0.542

Table A11 Storage modulus and normalized storage modulus of FA filled IR compounds with 12-week storage duration

Week	Storage modulus (kPa)			Normalized storage modulus		
	0	50	100	0	50	100
0	75.444	102.82	140.15	1.000	1.000	1.000
1	70.44	100.27	131.26	0.937	0.975	0.934
2	71.93	96.108	127.37	0.909	0.935	0.953
4	72.024	93.883	125.35	0.894	0.913	0.955
6	68.206	93.128	125.28	0.894	0.906	0.904
8	65.348	89.912	122.94	0.877	0.874	0.866
12	59.504	81.909	113.48	0.810	0.797	0.789

Table A12 Storage modulus and normalized storage modulus of FA filled IR compounds containing different heavy metals with 13-week storage duration

Week	Storage modulus (kPa)			Normalized storage modulus		
	IR Cu 8 ppm	IR Mn 8 ppm	IR Fe 8 ppm	IR Cu 8 ppm	IR Mn 8 ppm	IR Fe 8 ppm
0	72.887	72.036	82.996	1.000	1.000	1.000
2	69.386	68.491	81.081	0.952	0.951	0.977
4	70.881	64.833	80.224	0.972	0.900	0.967
7	68.865	65.561	77.036	0.945	0.910	0.928
11	68.609	64.252	77.466	0.941	0.892	0.933
12	67.850	65.652	77.464	0.931	0.911	0.933
13	66.531	64.555	77.568	0.913	0.896	0.935

Table A13 Storage modulus and normalized storage modulus of 50-phr FA filled IR compounds containing different heavy metals with 13-week storage duration

Week	Storage modulus (kPa)			Normalized storage modulus		
	IRFA50 Cu 8 ppm	IRFA50 Mn 8 ppm	IRFA50 Fe 8 ppm	IRFA50 Cu 8 ppm	IRFA50 Mn 8 ppm	IRFA50 Fe 8 ppm
0	82.358	87.039	86.175	1.000	1.000	1.000
2	80.761	82.481	80.549	0.981	0.948	0.935
4	78.737	78.740	75.443	0.956	0.905	0.875
7	76.611	76.399	72.786	0.930	0.878	0.845
11	73.731	78.636	75.440	0.895	0.903	0.875
12	74.691	74.058	73.636	0.907	0.851	0.854
13	73.318	74.810	73.845	0.890	0.859	0.857

Table A14 Storage modulus and normalized storage modulus of 100-phr FA filled IR compounds containing different heavy metals with 13-week storage duration

Week	Storage modulus (kPa)			Normalized storage modulus		
	IRFA100 Cu 8 ppm	IRFA100 Mn 8 ppm	IRFA100 Fe 8 ppm	IRFA100 Cu 8 ppm	IRFA100 Mn 8 ppm	IRFA100 Fe 8 ppm
0	144.810	111.940	144.820	1.000	1.000	1.000
2	133.220	105.550	132.980	0.920	0.943	0.918
4	139.200	105.590	134.260	0.961	0.943	0.927
7	130.800	112.580	131.620	0.903	1.006	0.909
11	141.660	98.364	128.680	0.978	0.879	0.889
12	139.370	99.233	132.500	0.962	0.886	0.915
13	130.130	95.790	126.800	0.899	0.856	0.876

Table A15 Storage modulus and normalized storage modulus of silica filled NR compounds with 12-week storage duration

Week	Storage modulus (kPa)					Normalized storage modulus				
	0	50	100	150	200	0	50	100	150	200
1	61.907	108.550	271.850	439.510	635.600	1.000	1.000	1.000	1.000	1.000
3	60.287	106.300	266.760	452.020	599.270	0.974	0.979	0.981	1.028	0.943
5	55.195	100.370	253.600	448.700	633.430	0.892	0.925	0.933	1.021	0.997
7	62.147	103.930	254.560	418.270	615.570	1.004	0.957	0.936	0.952	0.968
9	59.480	102.500	243.860	443.410	654.060	0.961	0.944	0.897	1.009	1.029
12	61.715	101.510	242.500	411.320	650.720	0.997	0.935	0.892	0.936	1.024

Table A16 Storage modulus and retention of FA filled NR and IR compounds

FA loading (phr)	Storage modulus (kPa)			Retention of storage modulus		
	0	50	100	0	50	100
NR				82.445	79.167	71.734
unaged	42.928	38.146	57.246			
aged	35.392	30.199	41.065			
IR				92.262	87.484	86.992
unaged	72.456	98.957	125.46			
aged	66.849	86.572	109.14			

Table A17 Storage modulus and normalized storage modulus of FA filled NR compounds having 6-PPD with 12-week storage duration

Week	Storage modulus (kPa)					Normalized storage modulus				
	0	50	100	150	200	0	50	100	150	200
0	41.187	43.991	46.457	60.927	74.905	1.000	1.000	1.000	1.000	1.000
1	42.041	45.797	52.883	71.476	92.747	1.021	1.041	1.138	1.173	1.238
2	44.433	48.482	58.711	76.399	98.957	1.079	1.102	1.264	1.254	1.321
3	41.351	50.080	62.760	77.676	103.210	1.004	1.138	1.351	1.275	1.378
4	41.720	49.334	66.475	79.896	104.920	1.013	1.121	1.431	1.311	1.401
5	42.078	47.589	62.460	81.187	103.640	1.022	1.082	1.344	1.333	1.384
6	41.720	48.308	67.743	83.237	105.030	1.013	1.098	1.458	1.366	1.402
7	41.982	51.039	69.483	79.165	112.740	1.019	1.160	1.496	1.299	1.505
9	40.008	50.220	66.768	85.788	109.090	0.971	1.142	1.437	1.408	1.456
10	38.946	49.585	61.629	76.931	107.450	0.946	1.127	1.327	1.263	1.434
11	39.263	48.617	61.694	79.472	101.080	0.953	1.105	1.328	1.304	1.349
12	39.105	46.306	63.984	79.675	101.690	0.949	1.053	1.377	1.308	1.358

Table A18 Rheological results before correction of FA filled NR compounds with various FA loading determined by capillary rheometer with three different die lengths (10, 20 and 30 mm)

Apparent shear rate (s ⁻¹)	Measured pressure drop (x10 ⁵ Pa)														
	0					50					100				
	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm
10	41.00	75.47	101.95	40.78	69.27	101.25	40.68	62.76	101.25	49.18	94.45	139.13	54.77	102.35	152.92
20	57.51	99.25	139.83	59.00	100.35	142.53	64.77	106.54	137.03	74.06	131.63	190.01	81.96	152.22	209.20
50	64.33	113.44	159.02	68.57	117.24	163.22	80.26	120.53	168.32	92.95	157.42	224.39	107.15	189.71	247.38
100	80.96	137.53	192.40	84.76	141.63	193.10	102.65	157.31	205.80	119.54	197.90	272.46	151.23	247.78	301.65
500	128.14	197.90	268.27	124.74	199.70	257.67	160.42	226.67	305.05	186.21	305.45	381.01	221.19	337.80	430.39
1000	185.22	270.87	360.73	174.27	268.66	354.26	190.46	287.33	365.70	230.65	361.11	463.35	238.87	379.75	491.71

Apparent shear rate (s ⁻¹)	Apparent shear stress (Pa)														
	0					50					100				
	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm
10	170415	161170	169915	178910	173163	168750	218390	206898	168750	245875	236133	231883	273865	255873	202065
20	263370	248125	233050	280360	250875	237548	323840	291355	228387	370315	329085	316675	409795	380560	279360
50	320340	283608	265035	342830	293103	272032	401300	351325	280527	464765	393553	373980	535730	474263	321173
100	406295	343828	320673	423790	354073	321840	513245	443278	342995	597700	494753	454107	706145	619440	376145
500	668165	494753	447110	623690	499250	429452	802100	666668	508413	931035	763618	635015	1.11E+06	8.44E+05	493587
1000	926120	677185	601222	871325	671642	590440	952310	718320	609495	1153225	902780	772248	1194365	949365	717308

Apparent shear rate (s ⁻¹)	Apparent shear viscosity (Pa.s)														
	0					50					100				
	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm	10 mm	20 mm	30 mm
10	17052.40	16127.30	17002.40	17902.50	17327.30	16885.80	21853.00	20703.00	16885.80	24603.20	23628.40	23203.20	27404.00	25603.60	20219.40
20	5267.74	4962.82	4661.30	5607.56	5017.82	4751.27	6477.21	5827.47	4568.03	7406.77	6582.12	6333.91	8196.42	7611.69	5587.56
50	3203.60	2836.26	2650.52	3428.52	2931.21	2720.49	4013.26	3513.47	2805.45	4647.95	3935.78	3740.04	5357.64	4742.93	3211.94
100	2031.46	1719.12	1603.35	2118.93	1770.35	1609.19	2566.20	2216.37	1714.96	2988.48	2473.74	2270.52	3550.70	3097.18	1880.71
500	1336.34	989.51	894.23	1247.39	998.51	858.91	1604.21	1333.34	1016.83	1862.08	1527.24	1270.04	2211.90	2038.99	987.18
1000	926.12	677.19	601.22	871.33	671.64	590.44	952.31	718.32	609.50	1153.23	902.78	772.25	1194.37	949.37	717.31

Table A19 Rheological results after correction of FA filled NR compounds with various FA loading determined by capillary rheometer with three different die lengths (10, 20 and 30 mm)

Die length = 10 mm														
Corrected shear rate (s ⁻¹)					True shear stress (Pa)					True shear viscosity (Pa.s)				
0	50	100	150	200	0	50	100	150	200	0	50	100	150	200
20.11	20.31	20.06	21.52	19.77	154975.00	154088.35	165105.00	224386.65	247873.35	15507.42	15418.70	16521.07	22453.03	24803.21
100.62	101.60	100.37	107.64	98.92	187560.00	195010.00	173840.00	270315.00	309795.00	3751.44	3900.45	3477.02	5406.65	6196.30
201.25	203.19	200.74	215.28	197.84	221645.00	242830.00	251300.00	314765.00	335730.00	2216.59	2428.46	2513.16	3147.85	3357.51
402.53	406.41	401.51	430.59	395.70	254815.00	273790.00	263245.00	397700.00	356145.00	1274.06	1368.94	1316.21	1988.48	1780.71
1006.31	1016.02	1003.77	1076.45	989.24	340710.00	323690.00	352100.00	431035.00	605945.00	681.42	647.38	704.20	862.08	1211.90
2012.63	2032.05	2007.55	2152.92	1978.50	426120.00	420000.00	452310.00	653225.00	694365.00	426.12	421.33	452.31	653.23	694.37

Die length = 20 mm														
Corrected shear rate (s ⁻¹)					True shear stress (Pa)					True shear viscosity (Pa.s)				
0	50	100	150	200	0	50	100	150	200	0	50	100	150	200
20.69	18.87	18.28	19.42	18.96	163670.00	148251.68	137755.00	225388.33	242876.68	16377.48	14834.66	13784.32	22553.27	24303.22
103.53	94.39	91.47	97.13	94.86	198125.00	200875.00	191355.00	279085.00	330560.00	3962.75	4017.76	3827.34	5582.06	6611.62
207.05	188.78	182.93	194.26	189.72	233607.50	243102.50	226325.00	318552.50	374262.50	2336.22	2431.18	2263.39	3185.73	3742.86
414.14	377.59	365.89	388.55	379.47	268827.50	279072.50	268277.50	394752.50	419440.00	1344.12	1395.35	1341.37	1973.74	2097.18
1035.32	943.95	914.72	971.37	948.65	344752.50	349250.00	341667.50	513617.50	594490.00	689.51	698.50	683.34	1027.24	1188.99
2070.66	1887.92	1829.45	1942.75	1897.32	427185.00	445000.00	468320.00	652780.00	699365.00	427.19	446.64	468.32	652.78	699.37

Die length = 30 mm														
Corrected shear rate (s ⁻¹)					True shear stress (Pa)					True shear viscosity (Pa.s)				
0	50	100	150	200	0	50	100	150	200	0	50	100	150	200
19.56	20.13	19.27	20.69	20.72	153248.33	152142.78	155988.33	224720.55	246207.78	15334.65	15224.02	15608.82	22486.45	24636.55
97.88	100.69	96.43	103.49	103.67	199716.67	204215.00	178386.67	283341.67	315325.00	3994.59	4084.56	3567.96	5667.20	6306.90
195.76	201.38	192.86	206.98	207.33	231701.67	238698.33	230526.67	323980.00	345626.67	2317.16	2387.14	2305.41	3240.01	3456.49
391.55	402.78	385.74	414.00	414.69	270673.33	271840.00	259661.67	387440.00	369415.00	1353.35	1359.19	1298.30	1937.18	1847.06
978.86	1006.95	964.34	1034.97	1036.72	347110.00	329451.67	358413.33	468348.33	550641.67	694.22	658.91	716.83	936.70	1101.29
1957.73	2013.90	1928.69	2069.96	2073.45	434555.00	440000.00	442828.33	605581.67	652848.33	434.56	440.44	442.83	605.58	652.85

Table A20 Power law index of FA/NR with various FA loading determined from capillary rheometer

FA loading (phr)	Power law index				
	0	50	100	150	200
Before corection	0.271	0.264	0.289	0.267	0.263
After correction	0.222	0.222	0.235	0.213	0.212

Table A21 Molecular weight of masticated NR determined by GPC

	Molecular weight (Daltons)
NR1	1076000
NR2	400162
NR3	352100
NR4	303737
NR5	275325

Table A22 Complex viscosity of masticated NR determined by RPA2000

Frequency (rad/s)	Complex viscosity (Pa.s)				
	NR1	NR2	NR3	NR4	NR5
0.5	2.41E+05	94622	67291	43754	32259
0.99	1.38E+05	64339	48365	32100	30795
2	77180	41668	32264	22649	18691
5.01	34324	21785	17859	13617	11918
10	18686	12851	11100	8780.2	7859.6
15	12903	9535.1	8164.9	6452.8	6072.4
20	10343	7498	6495.2	5478	5291.4
25	8327.2	6263.1	5580.6	4639.8	4278.6
30	7047.4	5332.8	4874.7	4097.7	3757.4
40	5460.7	4249.7	3866.6	3319.6	3098.5
50	4476.7	3564.4	3273.6	2772.1	2611.7
60	3775.5	3066.2	2815.2	2431.5	2268.5
69.99	3270.1	2669.1	2500.2	2179.8	2049.3
80	2919.3	2401.3	2208.1	1956	1849.6
90	2690.1	2151.1	2045.1	1800.3	1686.3
100	2428.9	2007.9	1858	1657.5	1556.4
120	2049.7	1709.3	1593.3	1432.4	1346.3
140	1787.3	1510.7	1421.3	1260.9	1194.2
160	1568.6	1338.2	1264.8	1136.3	1083.7

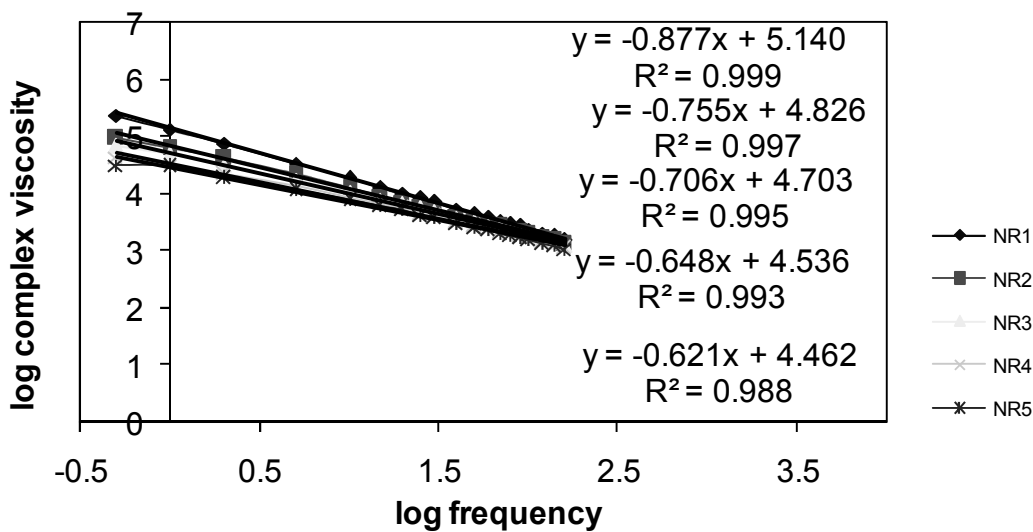


Figure A1 Plot of complex viscosity and frequency of masticated NR
(as followed : $\log \text{ complex viscosity} = \log \text{ zero shear} + a \log \text{ frequency}$)

Table A23 Zero shear viscosity of masticated NR determined by **Figure A1**

	k (log zero shear)	zero shear
NR1	5.1406	138229.3
NR2	4.8265	67065.63
NR3	4.7033	50501
NR4	4.5365	34395.37
NR5	4.4623	28993.46

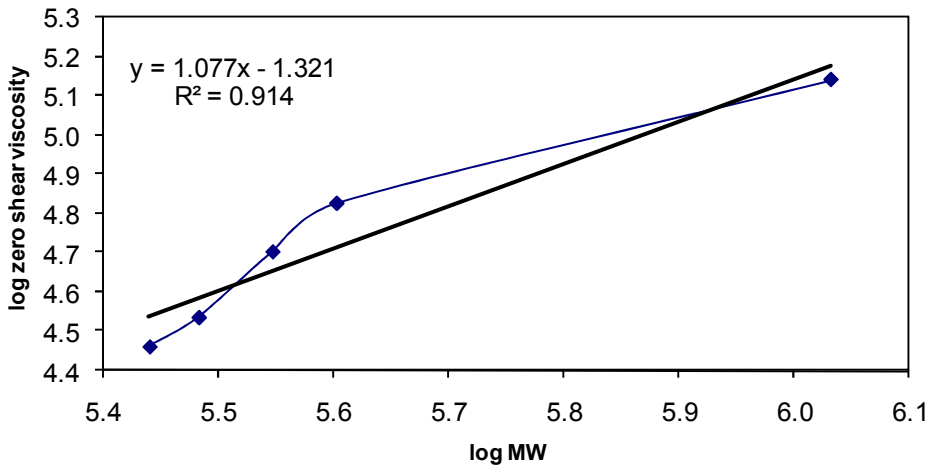


Figure A2 Plot of zero shear viscosity and molecular weight (MW) of masticated NR (as followed: $\log \text{ zero shear} = \log k + a \log \text{ MW}$)

Table A24 Relationship between zero shear viscosity and molecular weight of masticated NR from **Figure A2**

	k (log zero shear viscosity)	zero shear viscosity	log MW	MW (GPC)
NR1	5.1406	138229.3	6.031812	1076000
NR2	4.8265	67065.63	5.602236	400162
NR3	4.7033	50501	5.546666	352100
NR4	4.5365	34395.37	5.482498	303737
NR5	4.4623	28993.46	5.439846	275325

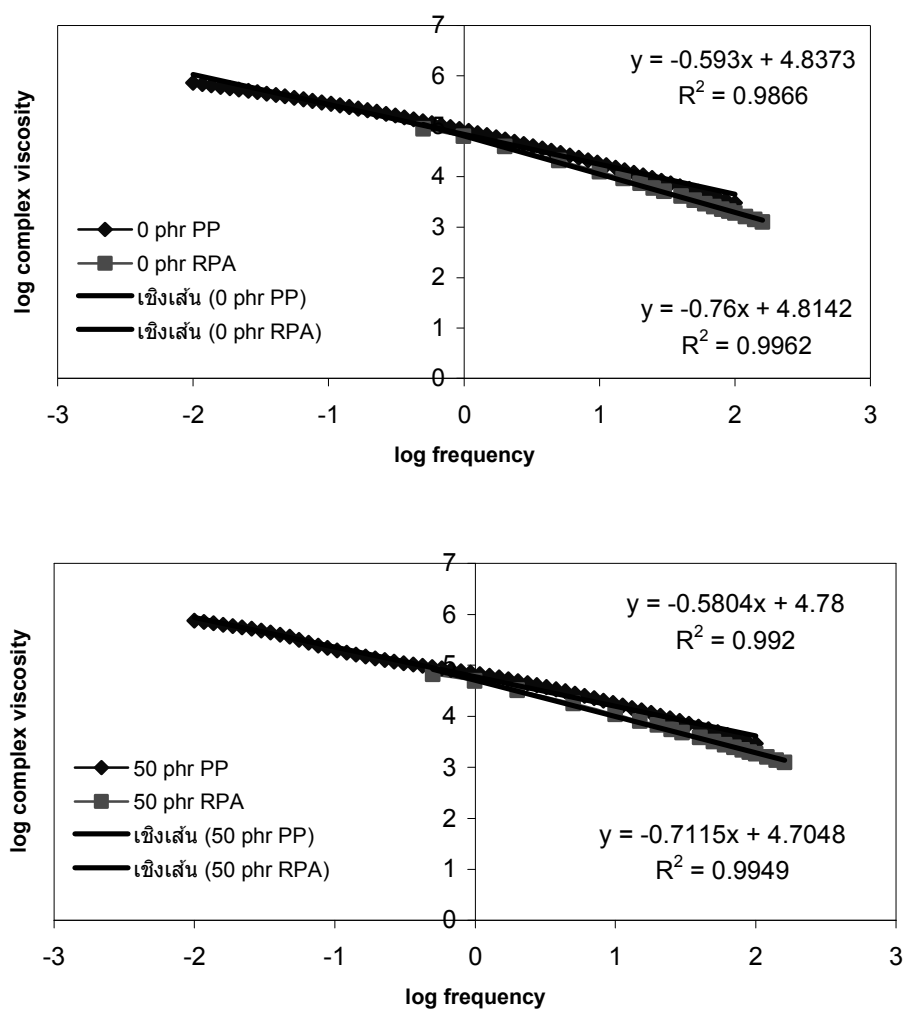


Figure A3 Complex viscosity of NR filled with various FA loadings determined by RPA2000 and PP under frequency sweep test

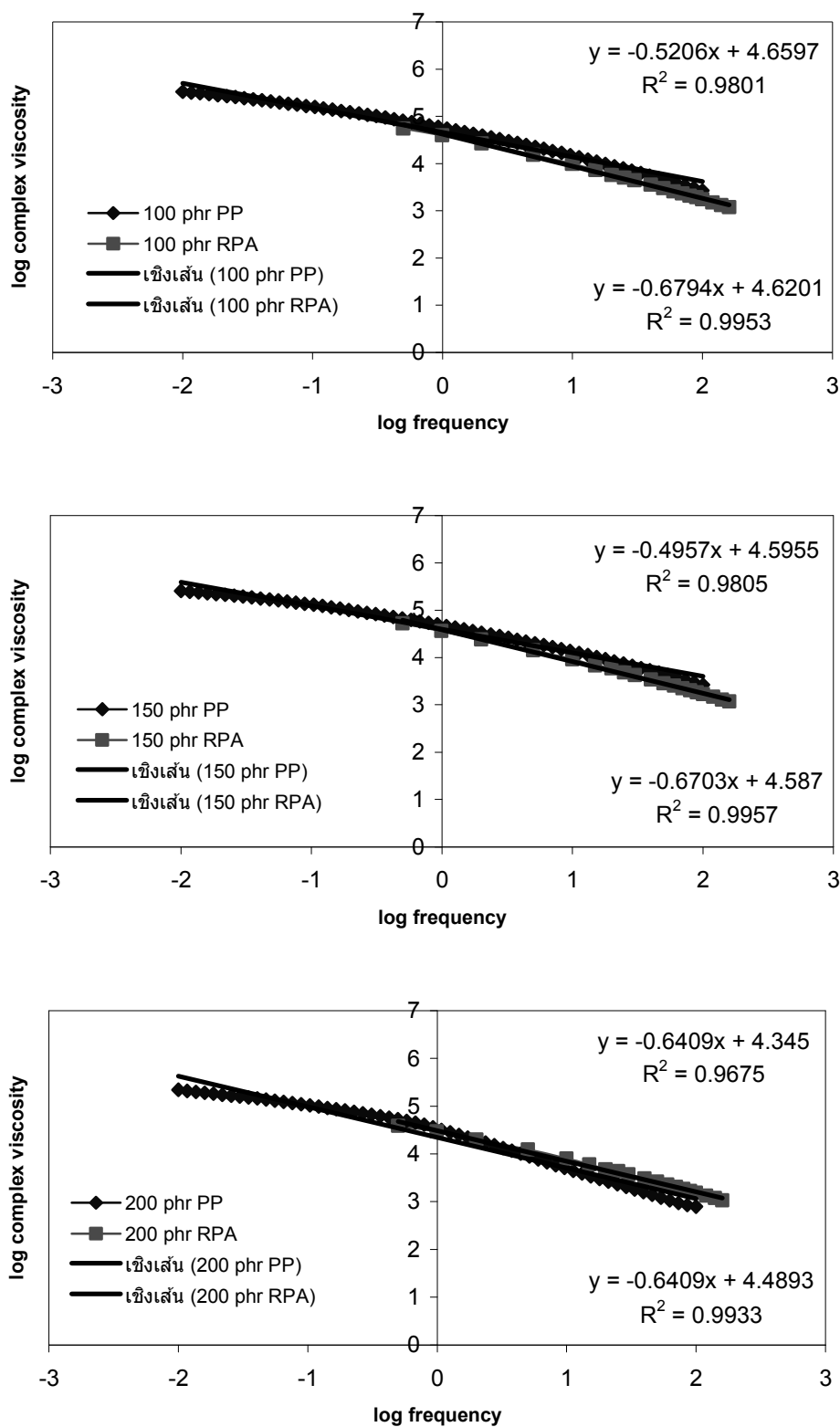


Figure A3 Complex viscosity of NR filled with various FA loadings determined by RPA2000 and PP under frequency sweep test (cont.)

Table A25 Complex viscosity of NR filled with various FA loadings determined by RPA2000 under frequency sweep test

Frequency	Complex viscosity (Pa.s)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.5	89630	66008	55253	53058	38791
0.99	63529	49128	39934	36253	29701
2	39924	32507	26703	24252	19989
5.01	21018	17912	15339	14128	12411
10	12600	10915	9785.9	9115.4	8011.9
15	9179.2	8079.4	7289	6793	6046.1
20	7400.1	6739	5700	5885.9	4704.6
25	5980.3	5491.1	5060.9	4783.9	4316.2
30	5157.2	4783	4408.2	4206.3	3716
40	4097.4	3818.7	3572.2	3402.5	3071.5
50	3391.4	3199.3	2994	2854.4	2602.9
60	2912.4	2737.5	2585.2	2523.8	2253.6
69.99	2527.7	2442.5	2313.5	2233.9	2016.7
80	2267	2187.6	2082.3	2007	1830.1
90	2057.3	1976.2	1922.3	1823.5	1657.8
100	1901.2	1823.3	1749	1681.1	1533.3
120	1611.3	1576.6	1504	1467.8	1324.8
140	1406.5	1381.9	1324	1275.3	1182.9
160	1257.3	1239.2	1194.1	1169.4	1058.7

Table A26 Complex viscosity of NR filled with various FA loadings determined by PP under frequency sweep test

Frequency	Complex viscosity (Pa.s)				
	0 phr	50 phr	100 phr	150 phr	200 phr
100	3.05E+03	2.93E+03	2.70E+03	2.66E+03	7.83E+02
85.55	3.47E+03	3.34E+03	3.05E+03	3.00E+03	8.40E+02
73.18	3.94E+03	3.80E+03	3.45E+03	3.38E+03	9.33E+02
62.61	4.48E+03	4.33E+03	3.90E+03	3.80E+03	1.06E+03
53.56	5.08E+03	4.92E+03	4.40E+03	4.27E+03	1.21E+03
45.82	5.77E+03	5.59E+03	4.96E+03	4.79E+03	1.38E+03
39.19	6.53E+03	6.34E+03	5.58E+03	5.36E+03	1.56E+03
33.53	7.39E+03	7.19E+03	6.27E+03	5.99E+03	1.78E+03
28.68	8.35E+03	8.13E+03	7.03E+03	6.69E+03	2.02E+03
24.54	9.43E+03	9.18E+03	7.88E+03	7.46E+03	2.30E+03
20.99	1.06E+04	1.04E+04	8.82E+03	8.29E+03	2.61E+03
17.96	1.20E+04	1.17E+04	9.85E+03	9.21E+03	2.96E+03
15.36	1.35E+04	1.31E+04	1.10E+04	1.02E+04	3.38E+03
13.14	1.51E+04	1.47E+04	1.22E+04	1.13E+04	3.86E+03
11.24	1.70E+04	1.65E+04	1.36E+04	1.25E+04	4.41E+03
9.617	1.90E+04	1.84E+04	1.51E+04	1.38E+04	5.06E+03
8.227	2.13E+04	2.05E+04	1.68E+04	1.52E+04	5.81E+03
7.038	2.38E+04	2.28E+04	1.85E+04	1.67E+04	6.61E+03
6.021	2.66E+04	2.53E+04	2.05E+04	1.83E+04	7.61E+03
5.151	2.96E+04	2.81E+04	2.26E+04	2.00E+04	8.81E+03
4.406	3.30E+04	3.10E+04	2.49E+04	2.19E+04	1.01E+04
3.769	3.66E+04	3.41E+04	2.74E+04	2.39E+04	1.16E+04
3.225	4.07E+04	3.75E+04	3.00E+04	2.61E+04	1.33E+04
2.759	4.51E+04	4.11E+04	3.29E+04	2.84E+04	1.52E+04
2.36	4.99E+04	4.49E+04	3.60E+04	3.09E+04	1.72E+04
2.019	5.52E+04	4.90E+04	3.93E+04	3.36E+04	1.95E+04
1.727	6.10E+04	5.32E+04	4.29E+04	3.65E+04	2.21E+04
1.477	6.72E+04	5.76E+04	4.67E+04	3.96E+04	2.50E+04
1.264	7.40E+04	6.22E+04	5.07E+04	4.30E+04	2.84E+04
1.081	8.13E+04	6.68E+04	5.51E+04	4.66E+04	3.21E+04
0.9249	8.92E+04	7.16E+04	5.97E+04	5.03E+04	3.62E+04
0.7912	9.77E+04	7.65E+04	6.46E+04	5.43E+04	4.08E+04
0.6769	1.07E+05	8.15E+04	6.98E+04	5.84E+04	4.53E+04
0.579	1.17E+05	8.67E+04	7.54E+04	6.28E+04	4.95E+04
0.4954	1.27E+05	9.19E+04	8.12E+04	6.74E+04	5.37E+04
0.4238	1.39E+05	9.76E+04	8.75E+04	7.22E+04	5.79E+04
0.3625	1.51E+05	1.04E+05	9.40E+04	7.74E+04	6.23E+04
0.3101	1.64E+05	1.10E+05	1.01E+05	8.29E+04	6.68E+04
0.2653	1.79E+05	1.18E+05	1.08E+05	8.87E+04	7.15E+04
0.227	1.94E+05	1.27E+05	1.15E+05	9.49E+04	7.64E+04
0.1941	2.10E+05	1.37E+05	1.22E+05	1.01E+05	8.15E+04
0.1661	2.26E+05	1.49E+05	1.30E+05	1.08E+05	8.68E+04

Table A26 Complex viscosity of NR filled with various FA loadings determined by PP under frequency sweep (cont.)

Frequency	Complex viscosity (Pa.s)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.1421	2.43E+05	1.62E+05	1.39E+05	1.16E+05	9.24E+04
0.1215	2.60E+05	1.78E+05	1.48E+05	1.24E+05	9.83E+04
0.104	2.79E+05	1.96E+05	1.57E+05	1.31E+05	1.04E+05
0.08895	2.99E+05	2.19E+05	1.66E+05	1.38E+05	1.11E+05
0.07609	3.20E+05	2.43E+05	1.76E+05	1.45E+05	1.17E+05
0.0651	3.42E+05	2.77E+05	1.86E+05	1.53E+05	1.24E+05
0.05569	3.65E+05	3.19E+05	1.97E+05	1.62E+05	1.31E+05
0.04764	3.91E+05	3.64E+05	2.07E+05	1.70E+05	1.38E+05
0.04075	4.16E+05	4.02E+05	2.19E+05	1.78E+05	1.46E+05
0.03486	4.43E+05	4.39E+05	2.30E+05	1.87E+05	1.53E+05
0.02982	4.72E+05	4.81E+05	2.43E+05	1.95E+05	1.60E+05
0.02551	5.03E+05	5.25E+05	2.56E+05	2.04E+05	1.67E+05
0.02183	5.37E+05	5.59E+05	2.69E+05	2.12E+05	1.75E+05
0.01867	5.69E+05	5.93E+05	2.81E+05	2.21E+05	1.84E+05
0.01597	6.04E+05	6.28E+05	2.95E+05	2.30E+05	1.92E+05
0.01366	6.44E+05	6.69E+05	3.09E+05	2.38E+05	2.00E+05
0.01169	6.84E+05	7.09E+05	3.20E+05	2.47E+05	2.10E+05
0.01	7.26E+05	7.52E+05	3.34E+05	2.56E+05	2.21E+05

Table A27 Zero shear viscosity of NR filled with various FA loadings determined by RPA2000 and PP

FA loading	PP		RPA	
	log zero shear viscosity	zero shear viscosity	log zero shear viscosity	zero shear viscosity
0	4.8373	68754.3214	4.8142	65192.85
50	4.78	60255.95861	4.7048	50675.73
100	4.6597	45677.25533	4.6201	41696.54
150	4.5955	39400.34276	4.587	38636.7
200	4.345	22130.9471	4.4893	30853.18

Table A28 Molecular weight of NR filled with various FA loadings determined by RPA2000, PP and GPC

FA loading	PP		RPA	
	log MW	MW	log MW	MW
0	5.716513506	520611.1989	5.695071	495531.2
50	5.663324979	460601.1088	5.593521	392212
100	5.55165692	356169.6587	5.514898	327264.1
150	5.492063492	310501.3495	5.484173	304911.2
200	5.259537733	181776.4984	5.393484	247447.9

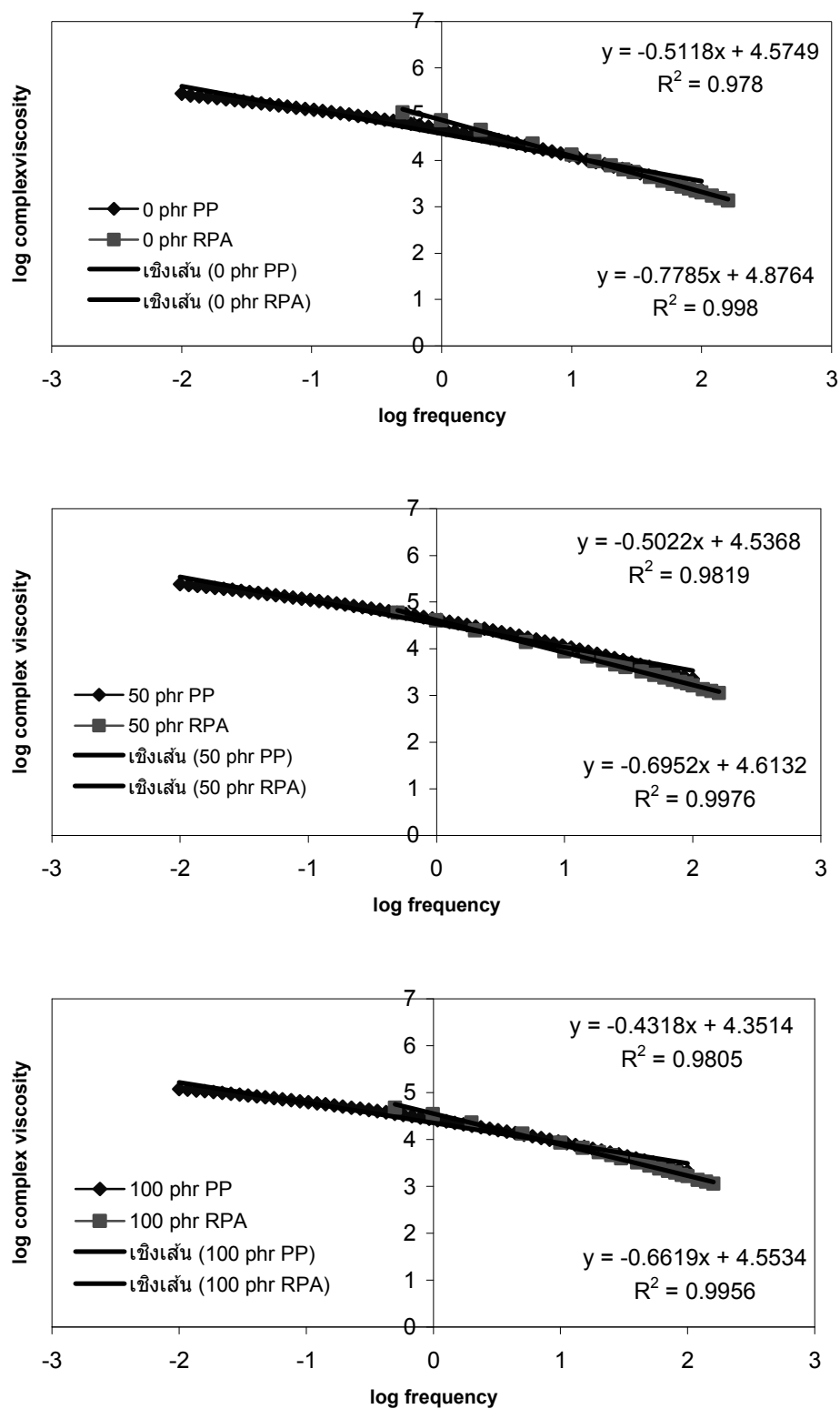


Figure A4 Complex viscosity of NR filled with various FA loadings after 14 weeks determined by RPA2000 and PP under frequency sweep test

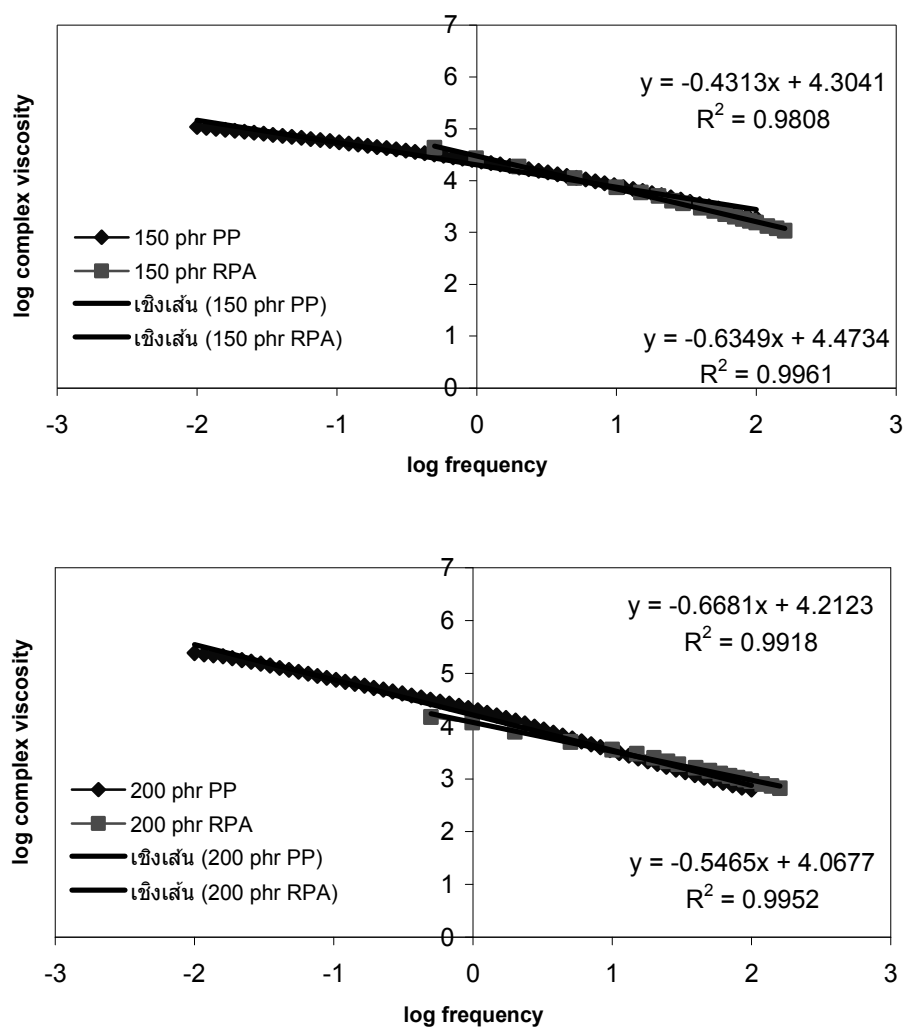


Figure A4 Complex viscosity of NR filled with various FA loading after 14 weeks determined by RPA2000 and PP under frequency sweep test (cont.)

Table A29 Complex viscosity of NR filled with various FA loadings after storage of 14 weeks determined by RPA2000 under frequency sweep test

Frequency	Complex viscosity (Pa.s)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.5	1.10E+05	59098	47469	43017	14887
0.99	74355	40316	34323	26869	11684
2	45697	24763	23007	18657	7773.7
5.01	23079	14004	13326	11213	4970.1
10	13441	8821.3	8542.2	7407.1	3544.9
15	9618.8	6757	6599.8	5876.5	2997.2
20	7864.8	5618.9	5337.3	5040.9	2504.5
25	6402.5	4625.6	4594.5	4081.7	2139
30	5576.5	4025.4	3974.2	3613.9	1883.8
40	4364.9	3246.5	3234.1	2977.2	1622.7
50	3618.7	2750.9	2746	2570.5	1422.2
60	3105.2	2404.8	2389.1	2243.1	1261.1
69.99	2702.1	2144.8	2131.8	1983.7	1138.8
80	2445.5	1924.9	1943.7	1808.7	1042.5
90	2208.3	1799.7	1754.4	1664.9	974.08
100	2038.3	1617.8	1635.1	1542.9	905.33
120	1742.1	1350.1	1373.1	1321.2	802.91
140	1513.4	1236.6	1255.8	1190	736.33
160	1352	1116.2	1136.8	1083.3	666.36

Table A30 Complex viscosity of NR filled with various FA loadings after storage of 14 weeks determined by PP under frequency sweep test

Frequency	Complex viscosity (Pa.s)				
	0 phr	50 phr	100 phr	150 phr	200 phr
100	2.90E+03	1.90E+03	2.09E+03	1.89E+03	6.28E+02
85.55	3.31E+03	2.10E+03	2.34E+03	2.11E+03	6.85E+02
73.18	3.78E+03	2.34E+03	2.60E+03	2.34E+03	7.56E+02
62.61	4.31E+03	2.60E+03	2.90E+03	2.61E+03	8.38E+02
53.56	4.92E+03	2.91E+03	3.21E+03	2.89E+03	9.40E+02
45.82	5.60E+03	3.25E+03	3.56E+03	3.20E+03	1.06E+03
39.19	6.38E+03	3.63E+03	3.94E+03	3.53E+03	1.19E+03
33.53	7.26E+03	4.05E+03	4.36E+03	3.90E+03	1.34E+03
28.68	8.25E+03	4.52E+03	4.80E+03	4.30E+03	1.51E+03
24.54	9.37E+03	5.03E+03	5.29E+03	4.74E+03	1.69E+03
20.99	1.06E+04	5.61E+03	5.82E+03	5.21E+03	1.91E+03
17.96	1.21E+04	6.25E+03	6.38E+03	5.72E+03	2.15E+03
15.36	1.37E+04	6.95E+03	6.99E+03	6.27E+03	2.43E+03
13.14	1.55E+04	7.72E+03	7.65E+03	6.86E+03	2.75E+03
11.24	1.75E+04	8.58E+03	8.35E+03	7.50E+03	3.11E+03
9.617	1.98E+04	9.53E+03	9.11E+03	8.18E+03	3.52E+03
8.227	2.24E+04	1.06E+04	9.91E+03	8.90E+03	3.99E+03
7.038	2.52E+04	1.17E+04	1.08E+04	9.68E+03	4.52E+03
6.021	2.85E+04	1.29E+04	1.17E+04	1.05E+04	5.13E+03
5.151	3.21E+04	1.43E+04	1.27E+04	1.14E+04	5.83E+03
4.406	3.61E+04	1.57E+04	1.37E+04	1.23E+04	6.64E+03
3.769	4.05E+04	1.73E+04	1.48E+04	1.33E+04	7.59E+03
3.225	4.55E+04	1.90E+04	1.59E+04	1.44E+04	8.68E+03
2.759	5.10E+04	2.08E+04	1.72E+04	1.55E+04	9.85E+03
2.36	5.72E+04	2.29E+04	1.84E+04	1.66E+04	1.12E+04
2.019	6.39E+04	2.51E+04	1.98E+04	1.79E+04	1.27E+04
1.727	7.14E+04	2.76E+04	2.12E+04	1.92E+04	1.43E+04
1.477	7.97E+04	3.02E+04	2.27E+04	2.05E+04	1.60E+04
1.264	8.88E+04	3.30E+04	2.43E+04	2.19E+04	1.79E+04
1.081	9.89E+04	3.61E+04	2.60E+04	2.34E+04	1.99E+04
0.9249	1.10E+05	3.94E+04	2.78E+04	2.50E+04	2.20E+04
0.7912	1.22E+05	4.29E+04	2.97E+04	2.66E+04	2.43E+04
0.6769	1.35E+05	4.67E+04	3.16E+04	2.84E+04	2.67E+04
0.579	1.49E+05	5.07E+04	3.36E+04	3.01E+04	2.93E+04
0.4954	1.65E+05	5.50E+04	3.57E+04	3.20E+04	3.20E+04
0.4238	1.81E+05	5.94E+04	3.79E+04	3.39E+04	3.50E+04
0.3625	1.99E+05	6.41E+04	4.02E+04	3.59E+04	3.81E+04
0.3101	2.19E+05	6.91E+04	4.26E+04	3.80E+04	4.15E+04
0.2653	2.39E+05	7.42E+04	4.51E+04	4.02E+04	4.51E+04
0.227	2.61E+05	7.98E+04	4.76E+04	4.24E+04	4.91E+04
0.1941	2.85E+05	8.58E+04	5.03E+04	4.48E+04	5.33E+04
0.1661	3.10E+05	9.20E+04	5.31E+04	4.72E+04	5.81E+04

Table A30 Complex viscosity of NR filled with various FA loadings after storage of 14 weeks determined by PP under frequency sweep (cont.)

Frequency	Complex viscosity (Pa.s)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.1421	3.37E+05	9.87E+04	5.61E+04	4.97E+04	6.33E+04
0.1215	3.65E+05	1.06E+05	5.90E+04	5.24E+04	6.89E+04
0.104	3.95E+05	1.14E+05	6.22E+04	5.51E+04	7.53E+04
0.08895	4.25E+05	1.22E+05	6.53E+04	5.79E+04	8.21E+04
0.07609	4.58E+05	1.30E+05	6.86E+04	6.08E+04	8.95E+04
0.0651	4.94E+05	1.39E+05	7.20E+04	6.38E+04	9.80E+04
0.05569	5.30E+05	1.49E+05	7.55E+04	6.70E+04	1.07E+05
0.04764	5.68E+05	1.59E+05	7.89E+04	7.02E+04	1.17E+05
0.04075	6.08E+05	1.70E+05	8.25E+04	7.35E+04	1.28E+05
0.03486	6.50E+05	1.81E+05	8.63E+04	7.70E+04	1.40E+05
0.02982	6.93E+05	1.93E+05	9.01E+04	8.06E+04	1.53E+05
0.02551	7.37E+05	2.06E+05	9.40E+04	8.45E+04	1.66E+05
0.02183	7.84E+05	2.20E+05	9.82E+04	8.81E+04	1.80E+05
0.01867	8.37E+05	2.33E+05	1.03E+05	9.18E+04	1.94E+05
0.01597	8.92E+05	2.49E+05	1.07E+05	9.62E+04	2.11E+05
0.01366	9.45E+05	2.64E+05	1.11E+05	1.01E+05	2.22E+05
0.01169	1.00E+06	2.79E+05	1.15E+05	1.05E+05	2.32E+05
0.01	1.07E+06	2.96E+05	1.19E+05	1.09E+05	2.42E+05

Table A31 Zero shear viscosity of NR filled with various FA loadings after storage of 14 weeks determined by RPA2000 and PP

FA loading	PP		RPA	
	log zero shear viscosity	zero shear viscosity	log zero shear viscosity	zero shear viscosity
0	4.9196	83099.8041	4.8764	75231.55
50	4.4995	31586.39048	4.6132	41039.31
100	4.3514	22459.49567	4.5534	35760.21
150	4.3014	20017.04659	4.4734	29744.04
200	4.2123	16304.21899	4.0677	11686.92

Table A32 Molecular weight of NR filled with various FA loadings after storage of 14 weeks determined by RPA2000, PP and GPC

FA loading	PP		RPA	
	log MW	MW	log MW	MW
0	5.792908196	620737.8054	5.752808	565988.9
50	5.402951824	252901.7439	5.508493	322473.1
100	5.265478511	184280.1305	5.452984	283781.7
150	5.219066184	165602.2313	5.378725	239179.9
200	5.136359417	136886.1211	5.002135	100492.8

Table A33 Normalized molecular weight (MW) of NR matrix of FA/NR compounds with various FA loadings

FA loading	MW		Normalized MW	
	PP	RPA	PP	RPA
0	520611.2	495531.2	1	1
50	460601.1	392212	0.884731	0.791498
100	356169.7	327264.1	0.684138	0.660431
150	310501.3	304911.2	0.596417	0.615322
200	181776.5	247447.9	0.34916	0.499359

Table A34 Normalized molecular weight of NR matrix of FA/NR compounds before and after 14-week storage duration

FA loading	MW		Normalized MW	
	PP	RPA	PP	RPA
0	620737.8	565988.9	1	1
50	252901.7	322473.1	0.569752	0.407421
100	184280.1	283781.7	0.501391	0.296873
150	165602.2	239179.9	0.422587	0.266783
200	136886.1	100492.8	0.177553	0.220522

Table A35 Molecular weight of masticated IR determined by GPC

	Molecular weight (Daltons)
IR1	991419
IR2	409872
IR3	354392
IR4	323987
IR5	279200

Table A36 Complex viscosity of masticated IR determined by RPA2000

	Complex viscosity (Pa.s)				
Frequency	IR1	IR2	IR3	IR4	IR5
0.5	2.25E+05	95778	69787	52294	44155
0.99	1.36E+05	70973	50477	36166	34824
2	77875	43321	34430	27861	25040
5.01	36124	23023	19080	16549	14927
10	19941	13676	11814	10481	9670.1
15	14016	9970.3	8635	7841.5	7205.6
20	11006	7653.5	6670.2	6294.3	6132.3
25	8855.3	6593.2	5885.9	5335.6	5037.2
30	7513.3	5637.7	5098.1	4627	4386.5
40	5815.6	4467	4035.1	3739.5	3544
50	4702.1	3712.3	3404.2	3130.4	2946.5
60	4061.8	3159.2	2881	2716	2574.5
69.99	3515.4	2779.3	2543.5	2351	2264.2
80	3104.1	2513.4	2263.2	2149.3	2047.2
90	2749.6	2214.6	2101.4	1922.3	1879.1
100	2580.6	2055.5	1921.8	1793.1	1741.5
120	2097.6	1768.6	1644.3	1571.3	1495
140	1862.2	1545.3	1444.1	1352	1322.5
160	1643.4	1374.1	1291.7	1223.3	1177

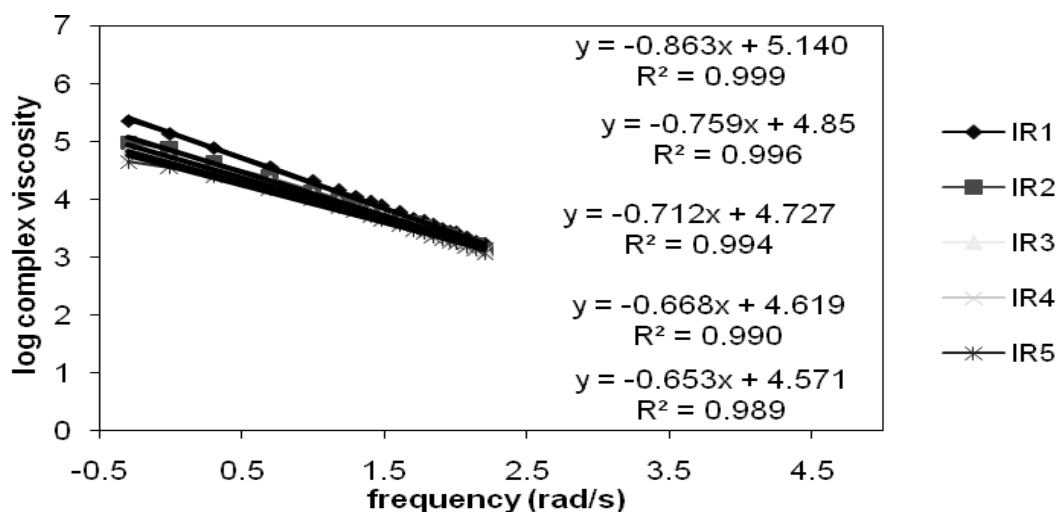


Figure A5 Plot of complex viscosity and frequency of masticated IR
(as followed : $\log \text{ complex viscosity} = \log \text{ zero shear} + a \log \text{ frequency}$)

Table A37 Zero shear viscosity of masticated IR determined by **Figure A5**

	k (log zero shear viscosity)	zero shear viscosity
IR1	5.1408	138292.9
IR2	4.85	70794.58
IR3	4.7279	53444.13
IR4	4.6195	41638.97
IR5	4.5717	37299.24

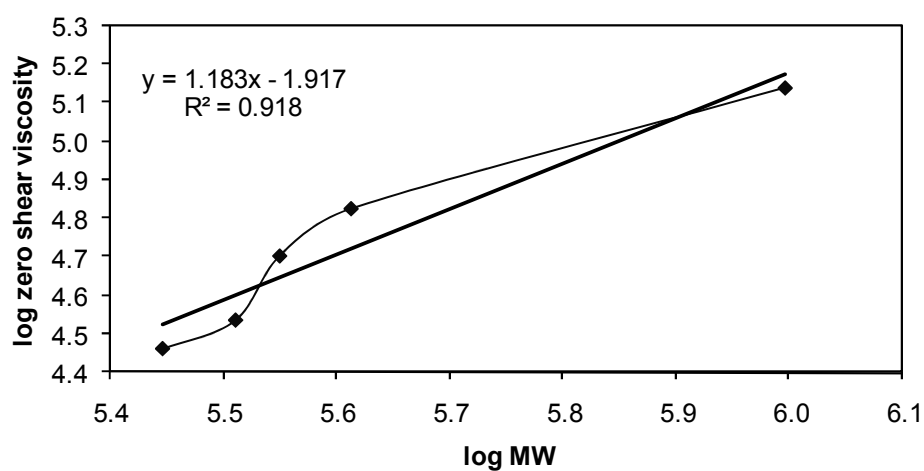


Figure A6 Plot of zero shear viscosity and molecular weight (MW) of masticated IR
(as followed: $\log \text{ zero shear} = \log k + a \log \text{ MW}$)

Table A38 Relationship between zero shear viscosity and molecular weight of masticated IR **Figure A6**

	k (log zero shear viscosity)	zero shear viscosity	log MW	MW (GPC)
IR1	5.1408	138292.9	5.996257	991419
IR2	4.85	70794.58	5.612648	409872
IR3	4.7279	53444.13	5.549484	354392
IR4	4.6195	41638.97	5.510528	323987
IR5	4.5717	37299.24	5.445915	279200

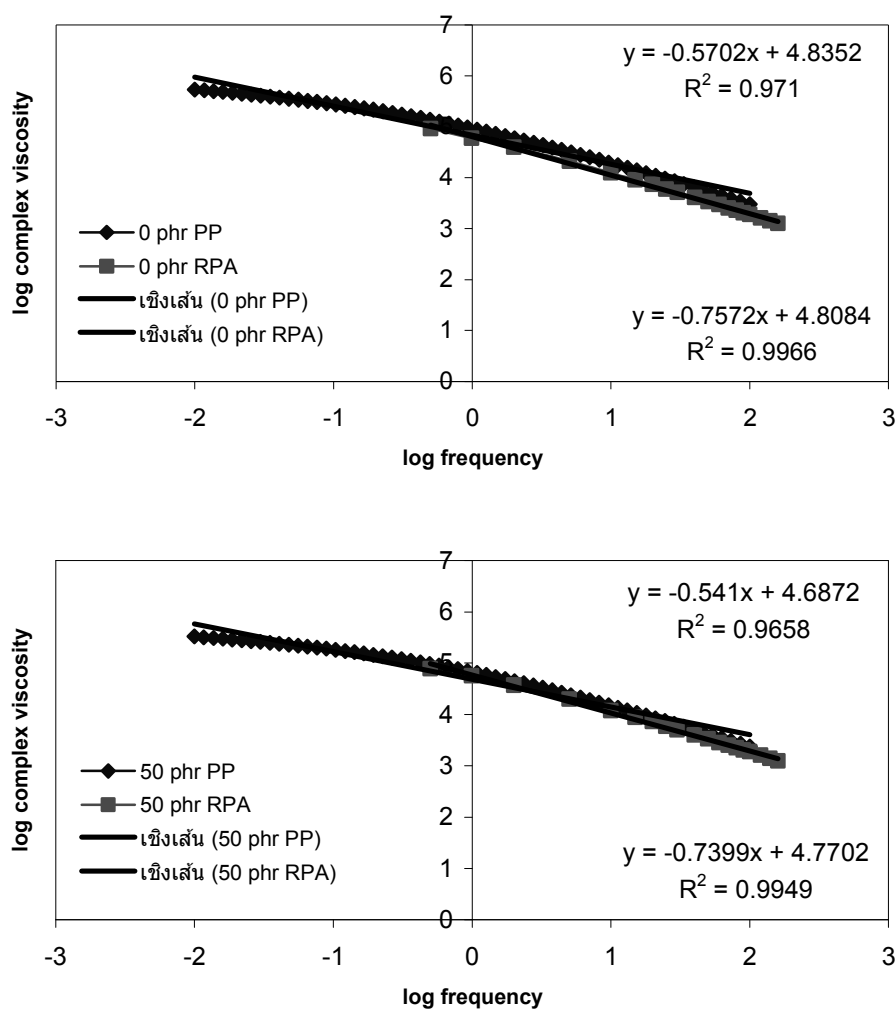


Figure A7 Complex viscosity of IR filled with various FA loadings determined by RPA2000 and PP under frequency sweep test

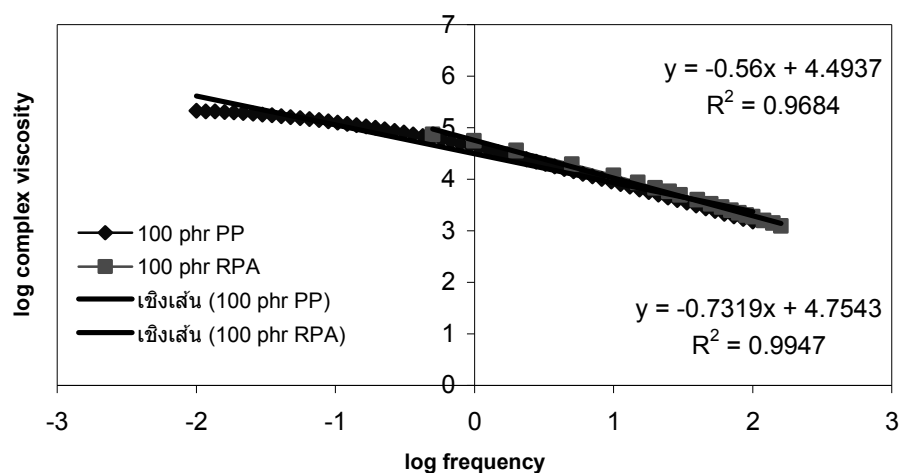


Figure A7 Complex viscosity of IR filled with various FA loadings determined by RPA2000 under frequency sweep test (cont.)

Table A39 Complex viscosity of IR filled with various FA loadings determined by RPA2000 under frequency sweep test

Frequency	Complex viscosity (Pa.s)		
	0 phr	50 phr	100 phr
0.5	92249	77961	74090
0.99	59401	56868	55082
2	39873	37003	35884
5.01	20876	19880	19600
10	12409	11964	11888
15	9047.5	8793.6	8672
20	7375.5	7271.4	6823.8
25	5929.4	5864.5	5825.6
30	5168.4	4995.1	4992.6
40	4071.6	4000.8	3979
50	3372.7	3333.9	3337.1
60	2943.7	2808.1	2881.7
69.99	2510.5	2504.1	2503
80	2291.1	2229.6	2236.9
90	2040.3	2017.9	2010.8
100	1883.5	1876.7	1872.6
120	1618.2	1616.1	1571.1
140	1421.2	1388	1397.8
160	1258.3	1238	1242.7

Table A40 Complex viscosity of IR filled with various FA loadings determined by PP under frequency sweep test

Frequency	Complex viscosity (Pa.s)			Frequency	Complex viscosity (Pa.s)		
	0 phr	50 phr	100 phr		0 phr	50 phr	100 phr
100	2.99E+03	2.37E+03	1.50E+03	0.9249	9.73E+04	7.02E+04	4.47E+04
85.55	3.41E+03	2.70E+03	1.68E+03	0.7912	1.07E+05	7.63E+04	4.90E+04
73.18	3.90E+03	3.07E+03	1.90E+03	0.6769	1.17E+05	8.28E+04	5.35E+04
62.61	4.45E+03	3.50E+03	2.14E+03	0.579	1.27E+05	8.96E+04	5.83E+04
53.56	5.07E+03	3.98E+03	2.42E+03	0.4954	1.38E+05	9.68E+04	6.33E+04
45.82	5.78E+03	4.53E+03	2.74E+03	0.4238	1.50E+05	1.04E+05	6.87E+04
39.19	6.57E+03	5.14E+03	3.10E+03	0.3625	1.62E+05	1.12E+05	7.43E+04
33.53	7.47E+03	5.83E+03	3.50E+03	0.3101	1.75E+05	1.20E+05	8.03E+04
28.68	8.48E+03	6.60E+03	3.95E+03	0.2653	1.88E+05	1.28E+05	8.65E+04
24.54	9.62E+03	7.47E+03	4.46E+03	0.227	2.02E+05	1.37E+05	9.28E+04
20.99	1.09E+04	8.45E+03	5.03E+03	0.1941	2.16E+05	1.45E+05	9.92E+04
17.96	1.23E+04	9.53E+03	5.67E+03	0.1661	2.30E+05	1.54E+05	1.06E+05
15.36	1.39E+04	1.08E+04	6.39E+03	0.1421	2.45E+05	1.63E+05	1.12E+05
13.14	1.57E+04	1.21E+04	7.19E+03	0.1215	2.60E+05	1.73E+05	1.19E+05
11.24	1.77E+04	1.36E+04	8.08E+03	0.104	2.76E+05	1.82E+05	1.26E+05
9.617	2.00E+04	1.53E+04	9.08E+03	0.08895	2.91E+05	1.91E+05	1.33E+05
8.227	2.24E+04	1.71E+04	1.02E+04	0.07609	3.07E+05	2.01E+05	1.40E+05
7.038	2.52E+04	1.92E+04	1.14E+04	0.0651	3.23E+05	2.11E+05	1.47E+05
6.021	2.83E+04	2.15E+04	1.28E+04	0.05569	3.39E+05	2.21E+05	1.54E+05
5.151	3.16E+04	2.40E+04	1.43E+04	0.04764	3.57E+05	2.30E+05	1.61E+05
4.406	3.53E+04	2.67E+04	1.60E+04	0.04075	3.73E+05	2.40E+05	1.67E+05
3.769	3.94E+04	2.97E+04	1.78E+04	0.03486	3.90E+05	2.50E+05	1.74E+05
3.225	4.39E+04	3.29E+04	1.99E+04	0.02982	4.07E+05	2.60E+05	1.81E+05
2.759	4.88E+04	3.65E+04	2.21E+04	0.02551	4.24E+05	2.70E+05	1.87E+05
2.36	5.42E+04	4.03E+04	2.46E+04	0.02183	4.41E+05	2.79E+05	1.94E+05
2.019	6.00E+04	4.45E+04	2.73E+04	0.01867	4.60E+05	2.89E+05	1.98E+05
1.727	6.64E+04	4.89E+04	3.03E+04	0.01597	4.80E+05	3.01E+05	2.03E+05
1.477	7.32E+04	5.37E+04	3.35E+04	0.01366	4.97E+05	3.10E+05	2.08E+05
1.264	8.07E+04	5.89E+04	3.70E+04	0.01169	5.16E+05	3.21E+05	2.09E+05
1.081	8.87E+04	6.44E+04	4.07E+04	0.01	5.37E+05	3.32E+05	2.13E+05

Table A41 Zero shear viscosity of IR filled with various FA loadings determined by RPA2000 and PP

FA loading	PP		RPA	
	log zero shear viscosity	zero shear viscosity	log zero shear viscosity	zero shear viscosity
0	4.8352	68422.66728	4.8084	64327.99
50	4.6872	48663.12561	4.7702	58911.49
100	4.4937	31167.35875	4.7543	56793.68

Table A42 Molecular weight of IR filled with various FA loadings determined by RPA2000, PP and GPC

FA loading	PP		RPA	
	log MW	MW	log MW	MW
0	5.708453085	511037.8712	5.708453085	511037.8712
50	5.583347422	383131.1135	5.583347422	383131.1135
100	5.41978022	262893.7248	5.41978022	262893.7248

Table A43 Normalized molecular weight (MW) of IR matrix of FA/NR compounds with various FA loadings

FA loading	MW		Normalized MW	
	PP	RPA	PP	RPA
0	511037.9	485063.7	1	1
50	383131.1	450306.3	0.749712	0.928345
100	262893.7	436583.8	0.514431	0.900055

APPENDIX B

VISCOELASTIC BEHAVIOR DATA OF 100 PHR FA FILLED NR COMPOUNDS WITH VARIOUS SILANES

Table B1 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-69 loadings

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
0.22	235.89	209.06	246.56	234.86	217.01
0.38	202	205.43	243.69	231.87	214.13
0.53	189.35	204.58	241.59	231.23	212.63
0.69	184.34	202.45	241.16	229.3	210.28
0.85	178.8	201.17	238.36	229.09	208.47
1.00	174.75	199.04	238.36	228.77	207.14
1.16	174.96	197.55	237.61	227.7	205.65
1.32	172.08	196.91	236.23	226.11	205.75
1.47	169.42	196.16	235.59	225.25	205.12
1.63	168.03	195.53	235.59	224.61	204.8
1.79	167.29	193.93	235.27	225.68	204.26
1.95	167.25	192.97	233.46	223.12	203.62
2.10	164.62	191.58	233.78	223.23	202.56
2.26	163.88	192.97	234.1	224.51	201.81
2.42	164.62	191.8	232.82	222.7	201.6
2.57	164.91	190.73	231.65	221.31	200.75
2.73	163.56	189.66	232.29	223.44	200.75
2.89	163.24	189.66	232.39	221.63	201.07
3.04	162.39	190.73	232.07	222.16	200.53
3.20	161.96	190.41	231.01	220.78	200.32
3.36	161.43	188.07	230.26	221.84	200.75
3.51	161	187.75	231.33	220.25	198.72
3.67	161.11	188.28	231.33	221.1	198.3
3.83	160.36	188.6	229.84	219.5	198.62
3.99	160.36	186.68	229.52	220.78	197.66
4.14	159.3	188.39	229.94	218.54	197.23
4.30	161.64	187.75	230.69	220.57	196.91
4.46	158.55	186.15	229.3	218.54	197.87
4.61	159.62	187.21	227.7	220.03	198.19
4.77	160.58	187.53	228.02	218.65	198.3
4.93	158.98	185.83	228.56	218.43	196.8
5.08	157.81	186.57	228.77	219.61	195.84

Table B1 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-69 loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
5.24	158.76	185.4	228.13	217.37	196.91
5.40	160.36	185.08	228.56	217.58	196.27
5.56	158.23	184.87	227.06	218.43	197.12
5.71	158.13	185.51	227.7	218.11	197.34
5.87	158.76	183.8	226.64	217.37	195.84
6.03	157.49	185.94	227.7	216.41	194.99
6.18	157.91	184.12	227.49	218.54	195.42
6.34	157.06	185.19	227.17	215.98	195.21
6.50	158.55	182.95	226.96	217.48	194.99
6.65	156.85	184.02	227.17	217.37	195.21
6.81	157.49	183.48	227.06	215.24	194.46
6.97	157.7	183.7	225.25	216.73	194.46
7.13	157.91	184.02	225.36	216.09	194.14
7.28	157.81	181.89	225.57	215.77	194.35
7.44	157.49	183.48	226	216.09	194.46
7.60	158.13	182.74	225.57	214.6	195.42
7.75	155.99	182.63	224.29	216.84	194.03
7.91	156.31	183.8	224.51	215.02	194.99
8.07	156.74	181.89	226.11	216.41	192.54
8.22	157.17	181.67	224.83	215.02	194.99
8.38	156.74	183.27	223.76	214.7	194.14
8.54	157.06	181.67	223.97	214.7	193.07
8.69	155.99	180.93	224.93	214.6	194.14
8.85	155.78	183.27	224.61	214.6	192.97
9.01	156.85	183.06	223.97	214.17	194.03
9.17	155.67	180.5	224.51	213.53	192.33
9.32	155.67	180.5	223.12	215.02	192.01
9.48	155.35	180.82	221.42	213.21	194.25
9.64	155.25	182.21	223.23	213.53	192.54
9.79	155.25	180.93	222.7	213.21	192.22
9.95	155.67	179.54	223.44	213.85	192.01
10.11	155.03	181.25	223.55	212.57	193.07

Table B1 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-69 loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
10.26	155.57	180.29	223.23	213.85	193.39
10.42	155.99	180.61	222.06	213.11	192.54
10.58	155.78	180.18	222.38	212.79	192.97
10.74	154.93	180.61	223.76	212.89	191.16
10.89	155.03	180.5	222.38	211.72	190.84
11.05	155.67	179.12	220.78	213.64	192.33
11.21	154.72	179.01	222.8	210.98	191.58
11.36	155.57	180.29	221.63	213.21	191.16
11.52	155.67	179.01	221.74	210.98	191.8
11.68	155.35	179.44	222.38	213.21	190.2
11.83	155.35	179.76	220.78	211.08	192.22
11.99	154.93	178.69	221.63	212.89	189.45
12.15	155.03	179.76	222.06	211.19	191.9
12.30	155.35	178.05	220.57	211.83	189.77
12.46	153.97	179.76	220.25	210.98	190.2
12.62	154.5	177.41	220.35	211.51	191.58
12.78	155.25	179.44	222.06	209.59	190.2
12.93	154.5	178.37	220.99	211.4	189.88
13.09	155.35	177.94	220.25	210.98	190.94
13.25	153.54	179.76	219.71	210.02	191.16
13.40	154.5	177.41	220.25	211.51	189.45
13.56	154.61	177.94	220.99	211.19	189.66
13.72	154.18	179.12	220.99	209.7	188.81
13.87	154.72	177.73	220.67	210.98	189.13
14.03	153.86	176.98	220.35	210.02	189.13
14.19	154.5	176.88	218.97	208.95	189.45
14.35	154.29	178.48	220.03	210.02	189.35
14.50	155.35	178.37	220.78	210.66	189.35
14.66	155.03	177.3	219.58	208.95	190.52
14.82	154.61	176.98	219.61	210.02	190.41
14.97	154.29	178.16	219.93	210.66	188.81
15.13	154.18	176.24	218.65	210.02	189.66
15.29	154.29	177.73	218.97	209.27	190.41
15.44	153.12	176.88	219.71	209.7	188.39
15.60	153.86	176.03	217.9	210.34	188.81
15.76	152.37	177.3	218.97	209.38	190.09

Table B1 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-69 loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
15.92	154.93	176.98	218.97	208.21	189.66
16.07	152.8	175.6	218.86	208.52	187.96
16.23	153.22	176.03	218.65	208.84	188.28
16.39	153.65	177.41	218.11	209.38	189.66
16.54	154.18	175.49	217.79	210.12	189.35
16.70	153.12	176.24	218.97	207.78	189.45
16.86	153.12	176.98	218.97	208.84	188.81
17.01	153.54	174.85	218.65	208.21	188.6
17.17	153.65	176.03	218.54	209.06	187.96
17.33	153.65	175.49	218.43	207.57	188.6
17.48	152.58	175.28	218.11	208.95	189.03
17.64	153.65	176.88	217.58	206.93	189.13
17.80	153.44	176.03	217.79	208.95	189.03
17.96	154.18	175.81	217.79	207.57	187.96
18.11	153.97	174.85	217.48	207.25	188.6
18.27	154.18	174.75	215.98	208.95	188.28
18.43	153.54	175.92	218.11	206.82	188.28
18.58	152.9	175.92	217.58	207.46	187.53
18.74	152.58	175.6	215.98	207.46	187.75
18.90	152.37	174.21	217.79	205.65	188.28
19.05	153.12	174.43	216.84	208.63	187.32
19.21	153.65	174.11	216.52	206.71	186.68
19.37	153.97	175.17	217.79	207.14	187.32
19.53	154.18	175.49	217.48	207.78	186.89
19.68	153.22	174.21	217.12	207.89	187
19.84	153.44	173.79	217.16	206.18	187.21
20.00	153.86	173.68	217.48	206.5	186.89
20.15	153.65	174.75	217.05	207.14	186.68
20.31	153.44	175.49	216.73	206.71	187
20.47	153.86	173.47	217.48	205.75	186.68
20.62	153.86	173.79	215.98	207.46	186.68
20.78	153.44	174.53	215.45	206.82	186.47
20.94	153.97	174.85	215.98	204.58	186.15
21.09	153.54	173.89	215.34	205.75	186.26
21.25	153.54	173.04	215.45	206.39	187.21
21.41	153.65	174.43	215.34	205.01	187.53
21.57	153.65	172.83	215.34	206.18	186.89
21.72	153.12	173.47	215.24	206.18	187.21
21.88	152.48	174.11	215.98	204.69	186.15

Table B1 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-69 loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
22.04	153.12	172.62	215.45	205.33	185.94
22.19	153.54	174.53	215.66	205.86	186.99
22.35	152.9	172.08	214.7	204.37	187.21
22.51	153.54	173.04	214.28	206.71	185.19
22.66	154.29	173.15	215.34	206.39	185.51
22.82	154.18	173.15	215.77	204.69	186.47
22.98	154.29	173.36	214.7	204.05	186.47
23.14	153.44	172.62	214.92	205.65	186.15
23.29	153.44	172.08	215.02	204.8	184.76
23.45	153.22	173.68	215.45	203.94	185.62
23.61	153.97	172.08	215.66	205.75	185.83
23.76	153.86	173.36	214.92	204.05	185.83
23.92	153.86	172.83	214.6	205.01	186.67
24.08	153.65	171.98	214.7	205.12	185.08
24.23	153.54	172.62	214.7	203.3	185.4
24.39	153.86	171.55	214.39	204.8	185.4
24.55	152.37	172.83	215.24	204.8	185.62
24.71	152.9	171.23	215.45	203.62	185.83
24.86	152.48	173.15	214.92	204.69	185.51
25.02	152.8	171.76	214.6	203.73	185.83
25.18	153.22	173.04	214.28	203.3	185.94
25.33	152.37	171.66	214.6	204.69	185.19
25.49	152.16	171.76	213.85	204.26	183.8
25.65	152.16	172.3	215.02	203.3	184.44
25.80	152.48	171.76	214.6	204.05	184.44
25.96	152.16	171.34	213.21	205.33	184.76
26.12	153.12	172.83	213.64	202.66	185.19
26.27	152.58	170.91	213.32	203.2	184.55
26.43	152.48	171.55	212.25	204.26	184.34
26.59	152.05	171.66	213.11	203.52	185.4
26.75	152.37	170.7	214.17	202.98	183.38
26.90	153.86	171.55	214.39	203.2	185.19
27.06	153.54	171.34	214.17	204.05	183.06
27.22	153.44	170.17	214.28	203.62	183.7
27.37	153.44	172.72	213.53	202.13	184.02
27.53	152.48	169.85	213.32	202.56	184.12
27.69	152.37	171.34	213.53	203.94	184.02
27.84	151.73	170.91	213.11	203.52	183.38
28.00	152.9	170.91	212.89	203.2	184.12

Table B1 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-69 loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
28.16	152.37	171.66	212.25	201.6	184.76
28.32	152.9	169.63	212.89	201.92	183.48
28.47	153.97	171.98	213.21	202.24	183.7
28.63	153.22	169.1	211.4	203.62	183.7
28.79	153.97	171.66	212.15	202.98	184.76
28.94	153.44	169.53	212.15	201.17	184.34
29.10	153.86	170.27	212.15	202.24	182.95
29.26	152.37	170.17	212.47	203.3	182.95
29.41	152.16	170.27	212.57	201.92	182.63
29.57	153.12	170.27	212.25	201.81	183.7
29.73	152.9	169.95	212.79	202.98	183.06
29.88	153.86	169.42	211.08	201.17	182.95

Table B2 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-264 loadings

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
0.22	235.89	218.54	206.65	207.72	186.57
0.38	202	218.19	204.83	205.12	186.89
0.53	189.35	217.76	201.81	203.73	185.62
0.69	184.34	216.84	204.09	203.94	184.12
0.85	178.8	215.24	201.17	201.92	182.63
1.00	174.75	216.3	199.68	201.07	182.95
1.16	174.96	213.11	199.47	200.43	182.74
1.32	172.08	214.39	199.47	200.43	181.25
1.47	169.42	212.89	199.25	197.87	180.18
1.63	168.03	212.89	197.34	198.19	181.25
1.79	167.29	211.08	196.8	198.3	180.61
1.95	167.25	212.57	197.23	196.48	179.44
2.10	164.62	212.04	195.53	196.27	178.69
2.26	163.88	210.98	197.66	197.34	178.8
2.42	164.62	210.02	197.12	195.1	177.62
2.57	164.91	209.59	195.74	196.27	178.69
2.73	163.56	209.59	195.1	196.27	177.62

Table B2 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-264 loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
2.89	163.24	208.95	196.59	195.53	176.88
3.04	162.39	207.99	195.84	193.93	176.88
3.20	161.96	208.95	194.67	194.46	176.03
3.36	161.43	208.52	194.46	194.46	176.88
3.51	161	207.99	195.53	194.99	176.35
3.67	161.11	208.21	194.03	194.35	175.81
3.83	160.36	206.39	194.03	194.67	176.98
3.99	160.36	205.75	194.78	194.03	175.28
4.14	159.3	206.71	194.67	193.29	175.6
4.30	161.64	205.01	193.39	193.39	174.85
4.46	158.55	205.86	193.61	193.39	175.17
4.61	159.62	204.26	192.54	193.39	175.17
4.77	160.58	204.26	192.97	193.07	174.53
4.93	158.98	204.8	194.14	192.54	175.17
5.08	157.81	204.83	194.35	192.65	174.11
5.24	158.76	204.58	193.39	191.58	173.68
5.40	160.36	202.88	192.22	192.22	174.75
5.56	158.23	203.52	192.65	191.58	173.89
5.71	158.13	203.2	193.39	191.9	173.36
5.87	158.76	202.66	193.07	190.84	173.79
6.03	157.49	202.45	191.8	190.41	173.68
6.18	157.91	202.88	191.16	190.41	173.36
6.34	157.06	202.13	191.16	189.66	173.36
6.50	158.55	203.73	192.54	189.45	173.04
6.65	156.85	200.11	192.54	189.77	172.83
6.81	157.49	202.45	192.54	189.13	171.76
6.97	157.7	200.85	191.16	190.52	171.98
7.13	157.91	202.45	190.94	190.52	171.66
7.28	157.81	199.79	190.84	189.88	171.66
7.44	157.49	201.81	192.54	190.73	171.55
7.60	158.13	199.47	190.73	190.09	171.23
7.75	155.99	200.32	190.73	190.2	171.02
7.91	156.31	199.79	190.84	190.2	170.91
8.07	156.74	199.79	189.66	190.09	170.49
8.22	157.17	201.39	190.73	189.13	170.49
8.38	156.74	198.72	191.48	187.96	171.02
8.54	157.06	200.32	190.09	188.71	170.7

Table B2 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-264 loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
8.69	155.99	199.04	190.2	188.39	171.02
8.85	155.78	199.25	190.94	187.75	171.34
9.01	156.85	198.72	190.73	187.64	170.49
9.17	155.67	199.36	189.88	187.64	169.63
9.32	155.67	197.34	189.45	187.96	171.34
9.48	155.35	197.87	189.13	187.32	169.85
9.64	155.25	197.98	190.2	187.96	170.27
9.79	155.25	198.62	190.09	187.53	169.95
9.95	155.67	198.4	189.88	187	169.53
10.11	155.03	197.98	189.88	187.96	170.59
10.26	155.57	197.12	189.66	188.28	168.57
10.42	155.99	198.19	187.96	186.26	168.78
10.58	155.78	197.23	189.66	186.57	169.42
10.74	154.93	197.55	189.13	187.53	169.95
10.89	155.03	197.23	188.39	187.21	170.17
11.05	155.67	196.91	189.77	187.21	169.85
11.21	154.72	196.27	187.64	186.26	169.63
11.36	155.57	197.23	189.13	186.15	168.89
11.52	155.67	197.12	189.13	186.15	169.1
11.68	155.35	196.16	188.07	186.89	169.21
11.83	155.35	196.16	187.53	186.57	168.78
11.99	154.93	195.74	188.39	184.87	168.46
12.15	155.03	195.53	189.35	186.68	168.35
12.30	155.35	195.74	189.03	185.62	169.42
12.46	153.97	195.53	188.71	185.51	168.03
12.62	154.5	195.74	188.07	186.89	169.21
12.78	155.25	195.21	189.35	185.19	167.71
12.93	154.5	195.53	187.75	184.76	168.78
13.09	155.35	195.53	186.89	186.15	169.42
13.25	153.54	196.16	187.64	185.62	167.5
13.40	154.5	195.1	187.75	185.51	167.29
13.56	154.61	194.99	187.32	184.55	167.29
13.72	154.18	195.1	187.21	185.51	167.82
13.87	154.72	194.78	187.32	185.83	167.82
14.03	153.86	196.48	186.89	184.02	168.35
14.19	154.5	193.93	186.68	184.55	166.76
14.35	154.29	193.39	186.89	184.87	167.29
14.50	155.35	194.67	186.57	185.08	167.08

Table B2 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-264 loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
14.66	155.03	194.46	186.57	184.12	166.76
14.82	154.61	194.14	187.32	184.02	167.08
14.97	154.29	194.35	187.32	184.02	167.5
15.13	154.18	193.71	187	184.44	167.5
15.29	154.29	193.61	186.68	184.34	167.71
15.44	153.12	194.35	185.94	183.7	167.5
15.60	153.86	193.71	186.47	183.7	167.5
15.76	152.37	194.14	187.32	183.8	166.76
15.92	154.93	194.14	187	184.02	167.5
16.07	152.8	192.01	185.4	184.34	167.08
16.23	153.22	194.14	185.51	184.76	167.5
16.39	153.65	192.33	185.94	183.38	166.65
16.54	154.18	192.65	185.4	184.12	166.01
16.70	153.12	192.86	185.62	184.87	166.44
16.86	153.12	193.07	185.08	183.8	166.01
17.01	153.54	192.86	184.87	182.95	166.22
17.17	153.65	192.01	185.51	182.95	165.69
17.33	153.65	192.22	186.15	183.06	166.01
17.48	152.58	191.58	186.15	182.95	165.69
17.64	153.65	192.65	185.83	183.06	166.33
17.80	153.44	192.22	185.51	184.12	166.22
17.96	154.18	192.01	187	183.8	165.9
18.11	153.97	191.58	186.15	183.7	165.9
18.27	154.18	190.94	185.51	183.8	166.22
18.43	153.54	191.8	185.19	183.48	166.01
18.58	152.9	191.16	186.47	182.74	166.22
18.74	152.58	192.22	186.15	182.21	165.69
18.90	152.37	191.8	185.62	182.21	165.16
19.05	153.12	191.48	185.94	182.31	165.16
19.21	153.65	190.73	186.15	182.63	166.44
19.37	153.97	192.33	185.19	183.48	166.65
19.53	154.18	192.22	184.76	183.27	166.33
19.68	153.22	191.16	185.51	182.95	164.94
19.84	153.44	190.52	185.19	183.27	164.62
20.00	153.86	190.52	184.44	181.99	164.84

Table B2 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-264 loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
20.15	153.65	191.58	184.02	181.99	166.01
20.31	153.44	189.66	184.87	181.14	165.26
20.47	153.86	191.26	183.48	181.25	164.62
20.62	153.86	190.41	184.12	182.21	164.52
20.78	153.44	189.13	185.08	181.25	164.31
20.94	153.97	191.48	185.19	182.21	165.58
21.09	153.54	190.73	184.02	182.21	165.58
21.25	153.54	190.52	184.55	182.72	164.09
21.41	153.65	189.45	184.02	181.67	163.88
21.57	153.65	189.66	184.12	181.14	164.62
21.72	153.12	190.41	184.12	180.61	164.09
21.88	152.48	190.41	184.34	181.25	163.88
22.04	153.12	189.77	183.48	181.35	163.77
22.19	153.54	189.88	183.48	181.99	164.94
22.35	152.9	189.77	183.8	181.89	164.62
22.51	153.54	189.77	183.8	181.35	164.94
22.66	154.29	189.77	184.02	180.61	164.84
22.82	154.18	189.13	184.55	180.5	164.84
22.98	154.29	188.81	183.7	180.93	164.09
23.14	153.44	189.03	184.02	180.82	163.24
23.29	153.44	189.35	183.06	180.93	163.24
23.45	153.22	189.35	182.74	180.5	163.13
23.61	153.97	189.88	183.7	180.08	163.24
23.76	153.86	189.66	183.48	180.5	163.13
23.92	153.86	188.71	183.8	180.93	163.45
24.08	153.65	188.71	182.63	180.93	163.13
24.23	153.54	189.45	182.31	180.5	163.24
24.39	153.86	188.71	182.63	180.18	163.56
24.55	152.37	189.13	182.95	180.29	163.03
24.71	152.9	188.28	182.74	180.5	163.24
24.86	152.48	189.77	182.42	180.18	163.77
25.02	152.8	187.75	183.38	180.61	163.88
25.18	153.22	189.35	183.7	180.29	164.2
25.33	152.37	187.75	182.31	179.94	164.2
25.49	152.16	189.35	182.74	180.29	163.77
25.65	152.16	188.81	182.95	179.76	163.56
25.80	152.48	189.66	183.48	179.86	163.13
25.96	152.16	187.96	182.95	179.22	162.49
26.12	153.12	187	182.21	180.5	163.03

Table B2 Time sweep results at 70 °C of 100 phr FA /NR compounds with various Si-264 loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
26.27	152.58	187.75	181.89	180.61	162.71
26.43	152.48	188.28	181.67	180.29	163.03
26.59	152.05	188.6	181.67	180.61	163.24
26.75	152.37	187.21	181.57	179.01	162.81
26.90	153.86	188.81	182.21	179.01	163.13
27.06	153.54	186.89	181.99	179.76	163.56
27.22	153.44	188.28	181.89	179.76	163.56
27.37	153.44	187.32	181.35	179.76	162.81
27.53	152.48	188.39	180.61	179.86	163.24
27.69	152.37	186.57	181.25	180.18	163.13
27.84	151.73	189.03	181.14	178.37	163.13
28.00	152.9	186.89	182.21	179.22	163.13
28.16	152.37	186.68	181.67	179.01	163.24
28.32	152.9	187.96	181.35	178.69	163.56
28.47	153.97	187.64	181.99	178.48	163.13
28.63	153.22	187.96	182.42	180.08	162.07
28.79	153.97	186.68	181.89	179.86	162.17
28.94	153.44	186.26	181.89	179.86	162.39
29.10	153.86	186.26	180.93	179.22	162.81
29.26	152.37	187.21	181.89	179.86	162.49
29.41	152.16	187.75	180.61	178.16	163.13
29.57	153.12	187.21	181.35	177.62	162.71
29.73	152.9	187.53	181.89	178.69	162.07
29.88	153.86	187.75	181.99	178.05	162.07

Table B3 Time sweep results at 70 °C of 100 phr FA /NR compounds with various VTEO loadings

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
0.22	235.89	200.43	204.51	218.19	221.63
0.38	202	198.93	201.92	216.09	219.93
0.53	189.35	197.98	201.17	214.6	218.65
0.69	184.34	198.3	199.79	212.57	218.11
0.85	178.8	197.78	198.72	212.47	216.84
1.00	174.75	194.99	197.34	210.98	217.58
1.16	174.96	196.06	196.16	211.08	216.52
1.32	172.08	194.67	195.74	209.7	214.92
1.47	169.42	192.86	194.67	209.27	215.77
1.63	168.03	192.01	195.1	208.95	215.24
1.79	167.29	193.07	194.35	208.84	214.92
1.95	167.25	191.9	193.61	208.52	213.96
2.10	164.62	192.54	192.86	208.31	213.64
2.26	163.88	191.26	192.01	207.14	212.57
2.42	164.62	190.52	190.73	207.25	212.15
2.57	164.91	190.09	191.26	206.5	212.04
2.73	163.56	189.77	189.45	205.75	211.4
2.89	163.24	188.71	190.84	207.25	211.19
3.04	162.39	188.6	189.88	204.8	210.44
3.20	161.96	188.28	189.88	204.69	210.76
3.36	161.43	188.71	189.35	205.12	210.34
3.51	161	188.28	189.77	205.12	210.12
3.67	161.11	188.28	188.71	203.73	209.59
3.83	160.36	187.32	186.57	203.94	208.95
3.99	160.36	187.21	188.6	203.52	208.52
4.14	159.3	186.68	186.26	204.05	208.21
4.30	161.64	185.51	187.53	202.56	208.52
4.46	158.55	185.83	186.68	202.88	207.57
4.61	159.62	185.4	185.62	202.45	208.21
4.77	160.58	185.08	185.94	202.45	207.57
4.93	158.98	185.4	186.47	203.2	206.93
5.08	157.81	184.87	186.15	202.98	207.25
5.24	158.76	184.55	186.57	202.24	206.5
5.40	160.36	184.44	186.57	201.49	207.25
5.56	158.23	184.44	185.94	201.17	206.5
5.71	158.13	184.44	185.94	201.6	205.65
5.87	158.76	184.12	185.19	201.6	205.33
6.03	157.49	184.12	185.08	201.17	205.12
6.18	157.91	182.95	184.44	200.53	205.65

Table B3 Time sweep results at 70 °C of 100 phr FA /NR compounds with various VTEO loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
6.34	157.06	182.63	184.76	200.53	205.75
6.50	158.55	183.48	183.48	200	205.12
6.65	156.85	183.06	184.34	200.11	205.01
6.81	157.49	182.31	184.76	201.07	204.37
6.97	157.7	182.42	183.38	200.11	203.73
7.13	157.91	182.63	184.02	199.79	204.37
7.28	157.81	182.74	183.48	199.36	204.26
7.44	157.49	182.63	183.38	201.17	203.52
7.60	158.13	182.21	183.27	198.72	203.62
7.75	155.99	181.67	182.21	200.11	204.26
7.91	156.31	181.57	182.42	198.93	203.73
8.07	156.74	181.89	181.99	198.72	202.98
8.22	157.17	181.35	182.21	198.72	203.73
8.38	156.74	180.82	183.06	199.68	202.88
8.54	157.06	180.82	181.67	199.36	202.24
8.69	155.99	181.25	182.21	198.4	202.66
8.85	155.78	180.29	183.38	198.4	202.24
9.01	156.85	180.61	182.31	198.72	202.13
9.17	155.67	180.29	181.67	199.04	202.13
9.32	155.67	180.5	182.21	197.12	202.24
9.48	155.35	180.5	180.93	199.79	201.49
9.64	155.25	180.61	180.61	197.12	201.07
9.79	155.25	180.5	180.61	197.98	201.49
9.95	155.67	180.18	180.82	198.4	201.39
10.11	155.03	180.29	181.57	197.87	201.6
10.26	155.57	179.86	182.21	198.93	201.49
10.42	155.99	179.86	181.57	196.59	201.39
10.58	155.78	180.29	181.35	198.72	200.75
10.74	154.93	180.08	181.67	197.23	200.85
10.89	155.03	180.08	181.35	196.8	200.75
11.05	155.67	179.01	180.5	198.3	200.75
11.21	154.72	179.01	179.86	197.87	200.43
11.36	155.57	178.8	180.82	196.48	199.79
11.52	155.67	179.22	180.18	196.48	200.11
11.68	155.35	179.12	180.29	197.98	200
11.83	155.35	179.54	180.5	197.55	200.11
11.99	154.93	179.44	180.08	196.48	200.11
12.15	155.03	178.48	179.01	196.27	199.68
12.30	155.35	178.8	179.01	197.23	198.93

Table B3 Time sweep results at 70 °C of 100 phr FA /NR compounds with various VTEO loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
12.46	153.97	178.69	179.54	196.06	198.4
12.62	154.5	178.37	180.61	194.78	200.43
12.78	155.25	178.16	180.29	197.23	199.47
12.93	154.5	178.69	179.22	195.84	198.4
13.09	155.35	179.44	179.54	195.74	199.47
13.25	153.54	179.12	180.08	196.8	199.47
13.40	154.5	179.12	180.08	195.84	199.04
13.56	154.61	178.48	180.08	196.16	197.87
13.72	154.18	178.69	179.54	194.03	198.3
13.87	154.72	178.37	179.54	196.27	198.3
14.03	153.86	178.8	179.54	197.12	198.72
14.19	154.5	178.16	179.86	196.59	198.3
14.35	154.29	178.05	179.76	194.03	197.66
14.50	155.35	177.94	178.69	195.53	198.19
14.66	155.03	178.05	179.12	197.12	197.98
14.82	154.61	177.73	179.44	194.99	196.91
14.97	154.29	178.05	179.44	194.99	198.19
15.13	154.18	178.16	177.73	196.06	197.55
15.29	154.29	178.37	177.62	196.06	197.98
15.44	153.12	177.41	178.48	194.46	197.34
15.60	153.86	176.98	177.94	195.42	196.91
15.76	152.37	177.41	178.05	195.74	197.87
15.92	154.93	177.94	178.16	194.67	196.91
16.07	152.8	178.16	178.16	194.99	197.55
16.23	153.22	176.88	177.73	196.27	197.23
16.39	153.65	176.88	178.05	195.21	196.8
16.54	154.18	176.24	178.48	194.14	196.48
16.70	153.12	178.16	178.16	195.42	197.12
16.86	153.12	177.41	178.37	195.42	197.12
17.01	153.54	176.56	177.94	195.21	196.59
17.17	153.65	176.67	177.3	193.71	196.48
17.33	153.65	177.94	177.3	194.46	196.59
17.48	152.58	175.49	177.94	195.84	196.16
17.64	153.65	178.05	177.62	195.21	196.48
17.80	153.44	176.03	177.09	193.71	196.91
17.96	154.18	177.41	177.62	194.14	196.27
18.11	153.97	175.92	177.3	195.53	196.48
18.27	154.18	177.09	176.67	194.14	196.8
18.43	153.54	175.6	176.98	194.67	196.06

Table B3 Time sweep results at 70 °C of 100 phr FA /NR compounds with various VTEO loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
18.58	152.9	177.73	177.73	194.67	195.74
18.74	152.58	175.81	177.62	193.07	196.16
18.90	152.37	176.88	176.98	194.35	195.74
19.05	153.12	176.35	176.03	193.61	195.74
19.21	153.65	175.81	176.88	194.14	195.21
19.37	153.97	176.98	176.88	194.35	194.99
19.53	154.18	176.67	177.62	192.86	195.53
19.68	153.22	175.6	177.62	193.93	195.42
19.84	153.44	175.6	176.35	193.71	195.74
20.00	153.86	176.56	176.35	193.93	195.42
20.15	153.65	177.41	176.98	193.71	195.74
20.31	153.44	176.88	175.92	192.97	195.53
20.47	153.86	176.88	176.24	194.14	194.67
20.62	153.86	176.67	176.35	194.78	194.67
20.78	153.44	176.88	175.92	193.93	194.99
20.94	153.97	175.81	176.98	194.35	195.21
21.09	153.54	176.03	176.67	192.97	195.1
21.25	153.54	175.92	175.6	193.07	194.78
21.41	153.65	176.24	176.24	192.86	194.99
21.57	153.65	175.92	176.98	193.39	194.78
21.72	153.12	176.56	176.35	192.65	194.67
21.88	152.48	175.81	176.98	193.61	195.1
22.04	153.12	175.49	175.81	194.46	194.78
22.19	153.54	175.81	174.96	193.61	194.99
22.35	152.9	175.17	175.81	194.14	194.46
22.51	153.54	175.81	176.56	194.14	194.46
22.66	154.29	175.6	176.88	193.07	194.78
22.82	154.18	175.28	175.92	192.54	194.35
22.98	154.29	175.17	176.03	193.07	194.14
23.14	153.44	174.96	175.6	192.86	194.35
23.29	153.44	175.49	175.92	193.61	193.93
23.45	153.22	175.28	175.81	193.29	194.03
23.61	153.97	175.28	174.96	193.07	193.61
23.76	153.86	174.85	175.17	192.86	194.03
23.92	153.86	176.24	174.85	192.97	194.78
24.08	153.65	176.03	175.49	192.22	193.93
24.23	153.54	175.92	175.17	192.97	194.03
24.39	153.86	175.6	175.28	193.07	193.61

Table B3 Time sweep results at 70 °C of 100 phr FA /NR compounds with various VTEO loadings (cont.)

Time (min)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
24.55	152.37	175.81	174.75	193.71	192.54
24.71	152.9	175.28	175.49	191.9	193.93
24.86	152.48	174.53	175.81	192.01	193.61
25.02	152.8	174.75	176.24	192.65	193.39
25.18	153.22	174.53	175.17	192.86	194.03
25.33	152.37	174.96	174.96	193.71	193.93
25.49	152.16	175.92	175.28	193.29	194.03
25.65	152.16	175.28	174.75	193.07	193.29
25.80	152.48	175.49	174.96	193.07	193.29
25.96	152.16	174.43	174.85	192.22	193.39
26.12	153.12	174.75	175.17	192.65	193.29
26.27	152.58	176.24	174.96	191.8	193.07
26.43	152.48	174.96	173.79	192.33	192.86
26.59	152.05	174.43	173.89	192.01	192.54
26.75	152.37	174.11	173.68	192.97	193.07
26.90	153.86	174.96	174.43	191.26	192.22
27.06	153.54	176.03	174.75	191.58	192.86
27.22	153.44	174.75	174.85	192.22	192.22
27.37	153.44	173.89	174.96	192.22	192.54
27.53	152.48	174.75	174.85	192.22	192.54
27.69	152.37	175.28	175.17	192.54	192.97
27.84	151.73	175.28	174.53	191.58	192.97
28.00	152.9	174.21	174.21	191.9	192.33
28.16	152.37	173.79	173.89	191.9	192.01
28.32	152.9	173.79	174.11	191.16	191.8
28.47	153.97	173.68	174.96	191.9	192.86
28.63	153.22	174.11	174.43	191.8	193.93
28.79	153.97	173.89	174.11	192.22	192.01
28.94	153.44	174.11	174.11	192.01	191.26
29.10	153.86	175.17	173.36	191.8	190.94
29.26	152.37	174.43	174.11	192.33	191.16
29.41	152.16	173.89	175.17	190.94	192.97
29.57	153.12	174.53	174.96	190.73	192.33
29.73	152.9	173.79	173.79	190.84	192.01
29.88	153.86	173.36	174.43	190.84	191.8

Table B4 Strain sweep results at 70 °C of 100 phr FA /NR compounds with various Si-69 loadings

Strain (%)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
0.54	58.85	74.544	112.85	111.82	90.237
0.95	60.982	58.504	105.42	90.919	87.593
1.97	64.863	61.313	100.9	78.723	86.963
4.99	53.916	60.905	89.079	76.292	76.075
10.02	51.003	53.916	82.153	72.669	71.936
49.98	33.343	36.064	50.79	45.989	46.97
100.15	21.744	24.319	32.253	29.121	30.007
499.97	5.5269	5.9407	7.4543	7.3478	6.5706
999.91	2.5904	2.695	3.3494	3.3078	2.9439
1199.98	2.0874	2.1372	2.5953	2.5882	2.3061

Table B5 Strain sweep results at 70 °C of 100 phr FA /NR compounds with various Si-264 loadings

Strain (%)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
0.54	58.85	86.314	82.39	76.505	72.582
0.96	60.982	75.396	75.396	77.614	67.635
1.93	64.863	78.707	69.853	73.753	56.691
5.01	53.916	74.155	69.239	69.239	61.588
10.03	51.003	69.714	63.542	63.984	57.166
49.98	33.343	46.735	43.406	43.418	38.765
100.08	21.744	30.227	28.506	28.447	25.253
499.95	5.5269	6.3523	6.099	6.3166	5.4329
999.94	2.5904	2.7558	2.6342	2.7528	2.3876
1200.01	2.0874	2.1568	2.0944	2.1879	1.9033

Table B6 Strain sweep results at 70 °C of 100 phr FA /NR compounds with various VTEO loadings

Strain (%)	Storage modulus (kPa)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
0.54	58.85	74.544	90.598	88.275	94.16
0.96	60.982	77.614	76.505	84.267	94.246
1.93	64.863	63.846	82.049	83.11	90.365
5.01	53.916	62.866	75.013	79.489	85.694
10.03	51.003	57.698	69.512	73.636	83.954
49.98	33.343	38.797	49.673	50.229	53.317
100.08	21.744	25.623	32.694	32.793	33.968
499.95	5.5269	5.7745	7.2473	7.3433	7.2168
999.94	2.5904	2.5446	3.1551	3.3472	3.1817
1200.01	2.0874	2.0634	2.5037	2.6611	2.5027

Table B7 Relative storage modulus (relative G') at 1 % strain of 100 phr FA /NR compounds with various silane loadings

Silane	Relative G'				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
Guth-Gold Equation	2.71	2.71	2.71	2.71	2.71
Si-69	1.22	1.17	2.11	1.82	1.76
Si-264	1.22	1.51	1.51	1.56	1.36
VTEO	1.22	1.56	1.53	1.69	1.89

Table B8 Strain sweep results at 70 °C of NR with and without Si-69 compounds

Strain (%)	NR	Strain (%)	NR+ Si-69
	Storage modulus (kPa)		Storage modulus (kPa)
0.54	0.067	0.54	72.582
0.96	0.113	0.96	92.028
1.93	0.295	1.93	83.712
5.01	0.66	4.99	84.84
10.07	1.345	10.04	81.181
50	6.034	49.97	73.024
100.09	10.22	100.09	58.677
499.95	19.22	499.95	9.7427
999.87	23.87	999.89	4.1958
1200.01	24.92	1199.97	3.2961

Table B9 Delta storage modulus (Payne effect) of 100 phr FA /NR compounds with various silane loadings

Silane	Delta G'				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
Si-69	56.7626	72.4068	110.2547	109.2318	87.9309
Si-264	56.7626	84.1572	80.2956	74.3171	70.6787
VTEO	56.7626	72.4806	88.0943	85.6139	91.6573

Table B10 Frequency sweep results of 100 phr FA /NR compounds with various Si-69 loadings

Frequency rad/s	Storage modulus (kPa)					Damping factor					Complex viscosity (Pa-s)				
	0	2	4	6	8	0	2	4	6	8	0	2	4	6	8
	%w/w	%w/w	%w/w	%w/w	%w/w	%w/w	%w/w	%w/w	%w/w	%w/w	% w/w	%w/w	%w/w	%w/w	%w/w
0.5	29.74	41.55	58.50	52.39	45.47	1.376	1.214	1.003	1.129	1.145	1.10E+05	1.25E+05	1.39E+05	1.57E+05	1.59E+05
0.99	45.47	60.41	85.40	78.04	70.82	1.203	1.076	0.937	0.98	0.952	81513	85653	93700	1.02E+05	1.08E+05
2	70.35	90.79	123.46	111.84	102.67	1.026	0.92	0.809	0.856	0.799	57250	58092	61647	67307	70222
5.01	116.59	142.74	182.91	172.48	155.31	0.801	0.753	0.654	0.708	0.652	33943	33419	33841	36344	37784
10	158.50	194.10	239.63	229.52	199.16	0.673	0.631	0.555	0.59	0.551	22037	20991	20812	22477	23345
15	187.76	223.54	271.68	263.25	224.30	0.611	0.595	0.532	0.532	0.506	16668	15828	15500	16521	17131
20	210.58	250.21	294.52	289.92	242.55	0.55	0.519	0.507	0.49	0.475	13542	12669	12183	13263	13670
25	220.34	265.39	313.23	306.33	262.79	0.553	0.522	0.462	0.487	0.452	11593	10805	10463	11257	11580
30	235.33	278.73	328.56	322.27	274.44	0.52	0.5	0.451	0.472	0.442	10185	9422.7	9085.2	9775.5	10039
40	257.42	302.35	354.98	347.33	296.52	0.49	0.469	0.419	0.445	0.415	8197.4	7571.2	7251.5	7801.9	8001.2
50	273.74	319.94	380.21	369.47	311.68	0.459	0.449	0.4	0.42	0.398	6981.1	6353.7	6069.1	6563.8	6658.1
60	289.40	339.97	390.50	381.46	323.31	0.444	0.423	0.387	0.403	0.385	6014	5548	5231.2	5625.3	5736.3
69.99	295.03	346.56	399.32	398.81	332.64	0.437	0.422	0.378	0.396	0.369	5361.7	4871.7	4640.4	4960.8	5087.7
80	308.78	360.76	413.50	405.32	344.04	0.418	0.408	0.365	0.393	0.361	4731.7	4362	4140.2	4449.8	4531.1
90	311.18	376.98	417.81	410.15	348.01	0.414	0.387	0.348	0.381	0.362	4336.6	3978.2	3750	4037.2	4105
100	323.24	381.91	431.80	423.60	359.94	0.392	0.39	0.347	0.369	0.347	3972.6	3657	3439	3678.4	3751
120	335.71	397.67	444.98	444.10	378.65	0.373	0.369	0.332	0.365	0.336	3506.4	3149.8	2971	3177.9	3227
140	350.02	402.76	459.95	455.97	383.60	0.386	0.37	0.323	0.347	0.315	3080	2764.7	2606.1	2789.5	2844.8
160	362.48	419.86	472.98	469.00	392.34	0.365	0.367	0.321	0.334	0.318	2752.6	2482.3	2331	2491.8	2542.6

Table B11 Frequency sweep results of 100 phr FA /NR compounds with various Si-264 loadings

Frequency rad/s	Storage modulus (kPa)					Damping factor					Complex viscosity (Pa-s)				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w	0 % w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
0.5	29.744	49.675	47.988	43.542	46.148	1.376	1.031	1.029	1.074	1.066	1.10E+05	1.63E+05	1.61E+05	1.72E+05	1.73E+05
0.99	45.473	71.271	65.884	64.062	64.522	1.203	0.898	0.921	0.95	0.905	81513	1.06E+05	1.13E+05	1.15E+05	1.12E+05
2	70.348	100.32	94.172	93.746	91.129	1.026	0.778	0.793	0.806	0.802	57250	70921	75161	74929	74616
5.01	116.59	148.26	141.33	141.33	136.42	0.801	0.647	0.666	0.668	0.671	33943	39996	41314	41134	40681
10	158.5	192.72	183.98	183.98	177.39	0.673	0.555	0.567	0.563	0.57	22037	25348	25679	25416	24958
15	187.76	215.41	212.96	211.73	200.54	0.611	0.527	0.495	0.498	0.524	16668	18992	19172	18998	18327
20	210.58	245	227.22	224.92	222.77	0.55	0.469	0.514	0.483	0.514	13542	15704	15313	15262	14678
25	220.34	254.51	244.69	243.93	235.34	0.553	0.461	0.467	0.471	0.476	11593	13048	12955	12775	12395
30	235.33	267.54	256.35	256.35	246.07	0.52	0.443	0.445	0.452	0.458	10185	11336	11263	11039	10772
40	257.42	288.24	276.89	276.12	266.47	0.49	0.418	0.419	0.421	0.43	8197.4	9137	9093.8	8818	8621.6
50	273.74	304.49	295.11	292.84	283.28	0.459	0.402	0.399	0.407	0.412	6981.1	7706.7	7579.9	7439.4	7099
60	289.4	317.52	305.41	305.25	292.84	0.444	0.387	0.391	0.393	0.399	6014	6610.2	6562.9	6371.6	6286.7
69.99	295.03	329.02	315.68	315.51	305.25	0.437	0.375	0.38	0.379	0.387	5361.7	5877.9	5763.7	5592.2	5490.7
80	308.78	337.51	324.73	324.73	313.23	0.418	0.368	0.37	0.373	0.38	4731.7	5257.2	5151.4	5029.1	4896
90	311.18	344.81	332.24	331.83	321.48	0.414	0.359	0.368	0.36	0.364	4336.6	4774.4	4621.2	4525.1	4463
100	323.24	351.25	340.81	340.89	328.1	0.392	0.357	0.359	0.356	0.371	3972.6	4383.5	4360.3	4245.1	4024.3
120	335.71	365.81	351.62	353.8	339.9	0.373	0.343	0.346	0.342	0.352	3506.4	3777.1	3674.9	3658.9	3414.3
140	350.02	378.38	363.82	364.43	353.59	0.386	0.328	0.326	0.335	0.338	3080	3313.1	3203.7	3156	3093.8
160	362.48	386.67	372.87	374.4	362.71	0.365	0.326	0.326	0.324	0.335	2752.6	2978.5	2923.5	2825.7	2724.3

Table B12 Frequency sweep results of 100 phr FA /NR compounds with various VTEO loadings

Frequency rad/s	Storage modulus (kPa)					Damping factor					Complex viscosity (Pa-s)				
	0	2	4	6	8	0	2	4	6	8	0	2	4	6	8
	%w/w	%w/w	%w/w	%w/w	%w/w	%w/w	%w/w	%w/w	%w/w	%w/w	% w/w	%w/w	%w/w	%w/w	%w/w
0.5	29.744	41.078	50.901	56.727	57.647	1.376	1.16	0.94	0.965	0.963	1.10E+05	1.30E+05	1.57E+05	1.65E+05	1.38E+05
0.99	45.473	60.254	70.656	76.659	81.882	1.203	1	0.861	0.867	0.85	81513	89201	1.10E+05	1.18E+05	98303
2	70.348	87.72	98.716	107.66	114.22	1.026	0.869	0.748	0.751	0.716	57250	61695	73595	79401	65710
5.01	116.59	136.11	144.27	155.93	162.52	0.801	0.714	0.615	0.601	0.595	33943	35695	42211	43657	37038
10	158.5	179.69	184.29	199.31	208.51	0.673	0.604	0.525	0.522	0.504	22037	22951	26645	27409	22739
15	187.76	207.9	208.78	222.77	231.97	0.611	0.551	0.489	0.487	0.476	16668	17348	19882	20518	16762
20	210.58	222.78	221.54	239.79	248.83	0.55	0.542	0.458	0.473	0.455	13542	14094	16143	16506	13427
25	220.34	242.24	240.58	258.03	267.08	0.553	0.493	0.427	0.435	0.418	11593	11975	13631	13804	11535
30	235.33	255.73	251.64	271.22	279.29	0.52	0.471	0.416	0.412	0.404	10185	10387	11880	12012	10001
40	257.42	276.58	270.38	290.69	299.26	0.49	0.446	0.389	0.391	0.379	8197.4	8348.5	9503.3	9622.8	8024.4
50	273.74	292.12	284.56	307.56	312.61	0.459	0.428	0.371	0.373	0.366	6981.1	7014	8015.1	8190.5	6707.9
60	289.4	308.63	295.75	317.52	324.27	0.444	0.404	0.356	0.361	0.356	6014	6151.7	6854.2	6978.4	5773.2
69.99	295.03	317.21	306.48	327.64	336.75	0.437	0.394	0.351	0.351	0.344	5361.7	5374.3	6129	6098.7	5065.5
80	308.78	325.61	313.38	336.75	343.28	0.418	0.385	0.342	0.343	0.339	4731.7	4870.6	5444.7	5502.9	4573
90	311.18	335.36	320.31	344.58	351.01	0.414	0.374	0.332	0.334	0.328	4336.6	4491.8	4876.7	4915.1	4112.6
100	323.24	343.2	326.57	349.17	356.36	0.392	0.368	0.33	0.331	0.328	3972.6	4099.1	4515	4570	3810.6
120	335.71	356.06	339.89	363.48	369.37	0.373	0.356	0.317	0.317	0.315	3506.4	3532.2	3939.2	3907.5	3328.6
140	350.02	366.27	349.34	373.79	380.84	0.386	0.342	0.301	0.303	0.306	3080	3067.5	3447	3452.5	2872.8
160	362.48	376.85	357.49	381.61	389.73	0.365	0.333	0.297	0.303	0.299	2752.6	2795.2	3090.8	3104.9	2573.4

Table B13 Power law index of 100 phr FA /NR compounds with various silanes loadings determined from RPA2000

Silane	Power law index				
	0 %w/w	2 %w/w	4 %w/w	6 %w/w	8 %w/w
Si-69	0.344	0.31	0.28	0.275	0.27
Si-264	0.344	0.301	0.289	0.279	0.273
VTEO	0.344	0.324	0.306	0.294	0.294

Table B14 Particle size distribution of FA with various milling times

Size (micron)	Volume (%)								
	0 hr	1hr	2 hr	3 hr	4 hr	5 hr	7.5 hr	10 hr	12.5 hr
0.01	0	0	0	0	0	0	0	0	0
0.011	0	0	0	0	0	0	0	0	0
0.013	0	0	0	0	0	0	0	0	0
0.015	0	0	0	0	0	0	0	0	0
0.017	0	0	0	0	0	0	0	0	0
0.02	0	0	0	0	0	0	0	0	0
0.023	0	0	0	0	0	0	0	0	0
0.026	0	0	0	0	0	0	0	0	0
0.03	0	0	0	0	0	0	0	0	0
0.035	0	0	0	0	0	0	0	0	0
0.04	0	0	0	0	0	0	0	0	0
0.046	0	0	0	0	0	0	0	0	0
0.052	0	0	0	0	0	0	0	0	0
0.06	0	0	0	0	0	0	0	0	0
0.069	0	0	0	0	0	0	0	0	0
0.079	0	0	0	0	0	0	0	0	0
0.091	0	0	0	0	0	0	0	0	0
0.105	0	0	0	0	0	0	0	0	0
0.12	0	0	0	0	0	0	0	0	0
0.138	0	0	0	0	0	0	0	0	0
0.158	0	0	0	0	0	0	0	0	0
0.182	0	0	0	0	0	0	0	0	0
0.209	0	0	0	0	0	0	0	0	0.02
0.24	0	0	0	0	0	0	0	0	0.06
0.275	0	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.09
0.316	0	0.08	0.08	0.08	0.09	0.1	0.11	0.11	0.13
0.363	0.04	0.1	0.11	0.1	0.12	0.13	0.14	0.15	0.17
0.417	0.05	0.12	0.13	0.13	0.15	0.16	0.17	0.19	0.21
0.479	0.06	0.15	0.16	0.16	0.17	0.19	0.2	0.22	0.24
0.55	0.07	0.17	0.18	0.18	0.2	0.21	0.23	0.25	0.27
0.631	0.1	0.19	0.2	0.2	0.22	0.24	0.26	0.27	0.29
0.724	0.15	0.21	0.23	0.23	0.24	0.26	0.29	0.3	0.32
0.832	0.19	0.25	0.26	0.27	0.28	0.3	0.33	0.34	0.35

Table B14 Particle size distribution of FA with various milling times (cont.)

[illegible]

Table B14 Particle size distribution of FA with various milling times (cont.)

Size (micron)	Volome (%)								
	0 hr	1hr	2 hr	3 hr	4 hr	5 hr	7.5 hr	10 hr	12.5 hr
416.869	0	0	0	0	0	0	0	0	0
478.63	0	0	0	0	0	0	0	0	0
549.541	0	0	0	0	0	0	0	0	0
630.957	0	0	0	0	0	0	0	0	0
724.436	0	0	0	0	0	0	0	0	0
831.764	0	0	0	0	0	0	0	0	0
954.993	0	0	0	0	0	0	0	0	0
1096.478	0	0	0	0	0	0	0	0	0
1258.925	0	0	0	0	0	0	0	0	0
1445.44	0	0	0	0	0	0	0	0	0
1695.587	0	0	0	0	0	0	0	0	0
1905.461	0	0	0	0	0	0	0	0	0
2187.762	0	0	0	0	0	0	0	0	0
2511.886	0	0	0	0	0	0	0	0	0
2884.032	0	0	0	0	0	0	0	0	0
3311.311	0	0	0	0	0	0	0	0	0
3801.894	0	0	0	0	0	0	0	0	0
4365.158	0	0	0	0	0	0	0	0	0
5011.872	0	0	0	0	0	0	0	0	0
5754.399	0	0	0	0	0	0	0	0	0
6606.934	0	0	0	0	0	0	0	0	0
7585.776	0	0	0	0	0	0	0	0	0
8709.636	0	0	0	0	0	0	0	0	0
10000	0	0	0	0	0	0	0	0	0

Table B15 Average particle size distribution and specific surface area of milled FA
with various milling times

Milling time	0 hr	1 hr	2 hr	3 hr	4 hr	5 hr	7.5 hr	10 hr	12.5 hr
Average particle size (micron)	50.311	36.692	34.782	31.942	30.003	26.635	23.437	22.249	21.296
Specific surface area (m ² /g)	0.478	0.624	0.642	0.661	0.685	0.727	0.779	0.808	0.878

Table B16 Time sweep results at 70 °C of 100 phr FA /NR compounds with various treatment methods

Time (min)	Storage modulus (kPa)			
	Without	In-situ	Pre-surface	Milled + Pre-surface
0.22	235.89	246.56	196.82	205.26
0.38	202	243.69	197.03	202.88
0.53	189.35	241.59	194.58	200.53
0.69	184.34	241.16	194.15	199.79
0.85	178.8	238.36	192.86	200.02
1.00	174.75	238.36	192.87	198.42
1.16	174.96	237.61	192.86	196.27
1.32	172.08	236.23	192.54	196.48
1.47	169.42	235.59	191.9	196.06
1.63	168.03	235.59	190.84	194.14
1.79	167.29	235.27	191.9	195.21
1.95	167.25	233.46	190.09	194.35
2.10	164.62	233.78	191.26	192.01
2.26	163.88	234.1	190.73	194.03
2.42	164.62	232.82	189.88	192.01
2.57	164.91	231.65	189.77	190.52
2.73	163.56	232.29	190.52	190.84
2.89	163.24	232.39	191.58	191.8
3.04	162.39	232.07	190.41	191.26
3.20	161.96	231.01	189.45	190.84
3.36	161.43	230.26	190.09	189.66
3.51	161	231.33	190.84	189.13
3.67	161.11	231.33	191.26	189.66
3.83	160.36	229.84	190.2	189.88
3.99	160.36	229.52	189.45	188.07
4.14	159.3	229.94	189.13	189.03
4.30	161.64	230.69	190.41	189.66
4.46	158.55	229.3	189.88	187.75
4.61	159.62	227.7	189.13	186.68
4.77	160.58	228.02	190.19	187
4.93	158.98	228.56	189.35	187.96
5.08	157.81	228.77	188.71	188.07
5.24	158.76	228.13	189.66	186.57
5.40	160.36	228.56	188.6	185.94
5.56	158.23	227.06	188.81	187
5.71	158.13	227.7	187.64	186.89
5.87	158.76	226.64	189.66	185.08
6.03	157.49	227.7	187.75	187
6.18	157.91	227.49	187.75	185.08

Table B16 Time sweep results at 70 °C of 100 phr FA /NR compounds with various treatment methods (cont.)

Time (min)	Storage modulus (kPa)			
	Without	In-situ	Pre-surface	Milled + Pre-surface
6.34	157.06	227.17	188.81	184.87
6.50	158.55	226.96	186.89	186.68
6.65	156.85	227.17	189.45	184.02
6.81	157.49	227.06	187.32	184.76
6.97	157.7	225.25	187.53	185.51
7.13	157.91	225.36	187.32	184.87
7.28	157.81	225.57	187.64	184.34
7.44	157.49	226	187.75	184.34
7.60	158.13	225.57	187.53	183.7
7.75	155.99	224.29	187.32	183.38
7.91	156.31	224.51	187.75	185.08
8.07	156.74	226.11	187.75	183.06
8.22	157.17	224.83	188.71	182.42
8.38	156.74	223.76	188.28	183.8
8.54	157.06	223.97	187.32	183.38
8.69	155.99	224.93	187.96	181.99
8.85	155.78	224.61	185.51	182.74
9.01	156.85	223.97	188.6	183.06
9.17	155.67	224.51	186.15	182.42
9.32	155.67	223.12	187.64	181.99
9.48	155.35	221.42	186.15	182.95
9.64	155.25	223.23	187	181.67
9.79	155.25	222.7	187	181.25
9.95	155.67	223.44	186.15	180.29
10.11	155.03	223.55	185.94	180.93
10.26	155.57	223.23	187	180.18
10.42	155.99	222.06	186.68	180.18
10.58	155.78	222.38	185.83	181.35
10.74	154.93	223.76	186.15	180.18
10.89	155.03	222.38	188.07	180.5
11.05	155.67	220.78	185.83	180.5
11.21	154.72	222.8	186.15	180.82
11.36	155.57	221.63	185.19	180.08
11.52	155.67	221.74	186.47	180.5
11.68	155.35	222.38	185.83	180.5
11.83	155.35	220.78	185.83	179.22
11.99	154.93	221.63	186.47	180.29
12.15	155.03	222.06	185.4	180.08
12.30	155.35	220.57	186.68	179.01

Table B16 Time sweep results at 70°C of 100 phr FA /NR compounds with various treatment methods (cont.)

Time (min)	Storage modulus (kPa)			
	Without	In-situ	Pre-surface	Milled + Pre-surface
12.46	153.97	220.25	184.76	179.44
12.62	154.5	220.35	186.89	179.76
12.78	155.25	222.06	186.68	179.54
12.93	154.5	220.99	185.51	178.48
13.09	155.35	220.25	184.44	180.08
13.25	153.54	219.71	185.4	179.54
13.40	154.5	220.25	185.4	178.69
13.56	154.61	220.99	184.34	179.54
13.72	154.18	220.99	184.76	179.01
13.87	154.72	220.67	185.62	178.37
14.03	153.86	220.35	186.15	178.48
14.19	154.5	218.97	185.62	178.8
14.35	154.29	220.03	184.12	178.16
14.50	155.35	220.78	184.34	178.16
14.66	155.03	219.58	185.08	178.37
14.82	154.61	219.61	184.87	178.37
14.97	154.29	219.93	183.48	177.73
15.13	154.18	218.65	184.34	177.62
15.29	154.29	218.97	185.4	177.41
15.44	153.12	219.71	183.7	177.62
15.60	153.86	217.9	184.34	177.62
15.76	152.37	218.97	184.12	177.09
15.92	154.93	218.97	184.44	177.09
16.07	152.8	218.86	184.44	176.98
16.23	153.22	218.65	183.27	176.67
16.39	153.65	218.11	183.27	176.98
16.54	154.18	217.79	183.7	175.92
16.70	153.12	218.97	184.76	176.67
16.86	153.12	218.97	184.76	176.56
17.01	153.54	218.65	182.74	175.49
17.17	153.65	218.54	185.94	176.24
17.33	153.65	218.43	182.63	175.81
17.48	152.58	218.11	184.76	176.24
17.64	153.65	217.58	182.42	175.17
17.80	153.44	217.79	183.8	174.85
17.96	154.18	217.79	184.12	175.81
18.11	153.97	217.48	182.63	175.6
18.27	154.18	215.98	184.87	175.6
18.43	153.54	218.11	184.12	175.6

Table B16 Time sweep results at 70 °C of 100 phr FA /NR compounds with various treatment methods (cont.)

Time (min)	Storage modulus (kPa)			
	Without	In-situ	Pre-surface	Milled + Pre-surface
18.58	152.9	217.58	182.74	175.28
18.74	152.58	215.98	184.44	176.03
18.90	152.37	217.79	182.95	176.03
19.05	153.12	216.84	183.38	175.81
19.21	153.65	216.52	184.34	174.53
19.37	153.97	217.79	182.74	175.49
19.53	154.18	217.48	182.95	174.53
19.68	153.22	217.12	183.7	175.28
19.84	153.44	217.16	182.74	175.49
20.00	153.86	217.48	183.38	175.17
20.15	153.65	217.05	182.63	174.21
20.31	153.44	216.73	183.48	174.21
20.47	153.86	217.48	183.7	174.11
20.62	153.86	215.98	182.63	174.43
20.78	153.44	215.45	183.06	173.47
20.94	153.97	215.98	182.95	174.75
21.09	153.54	215.34	182.42	174.75
21.25	153.54	215.45	182.31	173.79
21.41	153.65	215.34	182.31	174.43
21.57	153.65	215.34	183.7	174.53
21.72	153.12	215.24	181.89	173.89
21.88	152.48	215.98	184.12	173.15
22.04	153.12	215.45	181.99	173.68
22.19	153.54	215.66	182.31	173.36
22.35	152.9	214.7	183.38	173.47
22.51	153.54	214.28	182.31	173.47
22.66	154.29	215.34	181.89	173.68
22.82	154.18	215.77	182.42	173.68
22.98	154.29	214.7	182.95	173.36
23.14	153.44	214.92	181.57	173.36
23.29	153.44	215.02	183.38	172.62
23.45	153.22	215.45	182.42	173.47
23.61	153.97	215.66	181.99	172.62
23.76	153.86	214.92	183.06	172.3
23.92	153.86	214.6	182.21	172.83
24.08	153.65	214.7	182.31	172.08
24.23	153.54	214.7	182.95	172.3
24.39	153.86	214.39	181.35	171.76
24.55	152.37	215.24	182.21	172.62

Table B16 Time sweep results at 70 °C of 100 phr FA /NR compounds with various treatment methods at 4%w/w Si-69 (cont.)

Time (min)	Storage modulus (kPa)			
	Without	In-situ	Pre-surface	Milled + Pre-surface
24.71	152.9	215.45	181.67	172.08
24.86	152.48	214.92	182.42	171.76
25.02	152.8	214.6	183.06	171.98
25.18	153.22	214.28	182.74	171.98
25.33	152.37	214.6	181.14	171.23
25.49	152.16	213.85	181.67	172.3
25.65	152.16	215.02	181.99	172.62
25.80	152.48	214.6	182.42	170.59
25.96	152.16	213.21	180.82	170.59
26.12	153.12	213.64	182.42	171.23
26.27	152.58	213.32	181.67	170.59
26.43	152.48	212.25	181.57	171.23
26.59	152.05	213.11	180.18	171.02
26.75	152.37	214.17	182.63	171.34
26.90	153.86	214.39	181.35	170.7
27.06	153.54	214.17	181.89	171.34
27.22	153.44	214.28	181.25	170.91
27.37	153.44	213.53	181.89	170.91
27.53	152.48	213.32	180.82	171.23
27.69	152.37	213.53	181.89	171.23
27.84	151.73	213.11	181.14	169.85
28.00	152.9	212.89	181.35	170.91
28.16	152.37	212.25	181.14	170.7
28.32	152.9	212.89	181.14	170.49
28.47	153.97	213.21	180.93	169.63
28.63	153.22	211.4	181.25	170.59
28.79	153.97	212.15	180.82	170.17
28.94	153.44	212.15	180.82	169.85
29.10	153.86	212.15	181.99	169.53
29.26	152.37	212.47	180.61	169.85
29.41	152.16	212.57	180.93	168.57
29.57	153.12	212.25	179.76	169.21
29.73	152.9	212.79	182.63	170.91
29.88	153.86	211.08	179.86	169.85

Table B17 Strain sweep results at 70 °C of 100 phr FA /NR compounds with various treatment methods at 4%w/w Si-69

Strain (%)	Storage modulus (kPa)			
	Without	In-situ	Pre-surface	Milled + Pre-surface
0.54	58.85	112.85	97.572	82.049
0.95	60.982	105.42	95.572	78.467
1.97	64.863	100.9	93.568	75.35
4.99	53.916	89.079	81.648	71.376
10.02	51.003	82.153	74.902	69.589
49.98	33.343	50.79	51.338	51.188
100.15	21.744	32.253	32.87	34.766
499.97	5.5269	7.4543	6.9605	6.7691
999.91	2.5904	3.3494	3.028	2.8423
1199.98	2.0874	2.5953	2.3934	2.2821

Table B18 Delta storage modulus (Payne effect) of 100 phr FA /NR compounds with various treatment methods at 4%w/w Si-69

Delta G'	Without	In-situ	Pre-surface	Milled + Pre-surface
	56.7626	110.2547	95.1786	79.7669

Table B19 Relative storage modulus (relative G') at 1 % strain of 100 phr FA /NR compounds with various treatment methods at 4%w/w Si-69

	Relative G'			
	Without	In-situ	Pre-surface	Milled + Pre-surface
Guth-Gold Equation	2.714408112			
experimental at 1 % strain	1.222207	2.112837	1.915462471	1.572642549
experimental at 100 % strain	0.555729	0.8243157	0.840084852	0.888542439

Table B20 Frequency sweep results of 100 phr FA /NR compounds with various treatment methods at 4%/w/w Si-69

Frequency rad/s	Storage modulus (kPa)				Damping factor				Complex viscosity (Pa-s)			
	Without	In-situ	Pre-surface	Milled + Pre-surface	Without	In-situ	Pre-surface	Milled + Pre-surface	Without	In-situ	Pre-surface	Milled + Pre-surface
0.5	29.744	58.495	48.181	47.375	1.376	1.003	1.226	1.234	1.10E+05	1.39E+05	1.32E+05	1.90E+05
0.99	45.473	85.398	73.119	66.98	1.203	0.937	1.025	1.067	81513	93700	96688	1.29E+05
2	70.348	123.46	104.63	104.85	1.026	0.809	0.912	0.92	57250	61647	64442	83231
5.01	116.59	182.91	151.77	155.02	0.801	0.654	0.734	0.75	33943	33841	35309	44516
10	158.5	239.63	192.18	197.87	0.673	0.555	0.635	0.63	22037	20812	21489	27055
15	187.76	271.68	212.65	222.3	0.611	0.532	0.574	0.592	16668	15500	15609	20020
20	210.58	294.52	235.5	238.71	0.55	0.507	0.54	0.551	13542	12183	12876	15732
25	220.34	313.23	247.64	258.65	0.553	0.462	0.511	0.512	11593	10463	10717	13372
30	235.33	328.56	258.28	270.14	0.52	0.451	0.491	0.491	10185	9085.2	9241.5	11563
40	257.42	354.98	278.12	291.15	0.49	0.419	0.465	0.455	8197.4	7251.5	7420.8	9241.1
50	273.74	380.21	293.27	307.4	0.459	0.4	0.442	0.452	6981.1	6069.1	6232.1	7762.9
60	289.4	390.5	302.95	317.52	0.444	0.387	0.432	0.416	6014	5231.2	5342	6680.7
69.99	295.03	399.32	310.46	329.63	0.437	0.378	0.403	0.416	5361.7	4640.4	4681.4	5874.7
80	308.78	413.5	321.23	339.05	0.418	0.365	0.404	0.406	4731.7	4140.2	4221.4	5283.6
90	311.18	417.81	329.83	345.27	0.414	0.348	0.399	0.404	4336.6	3750	3838.3	4661.4
100	323.24	431.8	331.17	353.55	0.392	0.347	0.396	0.37	3972.6	3439	3473.8	4379.5
120	335.71	444.98	346.65	364.62	0.373	0.332	0.364	0.368	3506.4	2971	3009	3672.6
140	350.02	459.95	357.49	378.22	0.386	0.323	0.353	0.338	3080	2606.1	2659.9	3283.7
160	29.744	58.495	48.181	47.375	0.365	0.321	1.226	1.234	2752.6	1.39E+05	1.32E+05	1.90E+05

Table B21 Power law index of 100 phr FA /NR compounds with various treatment methods at 4%w/w Si-69 determined from RPA2000

Power law index	Without	In-situ	Pre-surface	Milled + Pre-surface
	0.344	0.28	0.286	0.267

APPENDIX C

VISCOELASTIC AND MECHANICAL PROPERTIES DATA OF UNMODIFIED FA FILLED NR VULCANIZATES WITH VARIOUS CURING SYSTEMS

Table C1 Cure behavior results of FA /NR compounds with various FA loadings in
CV systems

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.243	1.334	1.529	1.421	1.695	2.204
0.393	1.168	1.32	1.205	1.453	1.895
0.54	1.109	1.239	1.123	1.346	1.827
0.69	1.067	1.232	1.166	1.359	1.977
0.84	1.068	1.333	1.364	1.538	2.472
0.99	1.105	1.6	1.886	1.998	3.86
1.14	1.207	2.221	3.319	3.195	6.741
1.29	1.405	3.538	5.832	5.477	10.43
1.44	1.801	5.45	8.507	8.202	13.87
1.59	2.563	7.373	10.88	10.73	16.88
1.74	3.67	9.066	12	13.02	19.47
1.89	4.861	10.56	13.9	15.09	21.71
2.04	5.96	11.9	15.59	16.95	23.64
2.19	6.97	13.1	17.09	18.61	25.31
2.34	7.901	14.17	18.4	20.08	26.73
2.49	8.757	15.12	19.57	21.36	27.95
2.64	9.54	15.98	20.59	22.48	28.99
2.79	10.23	16.74	21.48	23.48	29.91
2.94	10.86	17.44	22.28	24.36	30.71
3.09	11.43	18.07	22.99	25.12	31.41
3.24	11.94	18.63	23.63	25.8	32.02
3.39	12.41	19.13	24.19	26.42	32.56
3.54	12.83	19.59	24.68	26.96	33.03
3.69	13.2	20.01	25.12	27.43	33.46
3.84	13.55	20.38	25.5	27.84	33.82
3.99	13.87	20.71	25.85	28.22	34.16
4.14	14.17	21	26.13	28.55	34.45
4.29	14.43	21.26	26.39	28.83	34.68
4.44	14.66	21.5	26.6	29.09	34.91
4.59	14.88	21.7	26.79	29.31	35.07

Table C1 Cure behavior results of FA /NR compounds with various FA loadings in CV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
4.74	15.07	21.88	26.96	29.5	35.24
4.89	15.25	22.03	27.1	29.64	35.36
5.04	15.41	22.16	27.2	29.78	35.46
5.19	15.55	22.28	27.29	29.89	35.55
5.34	15.67	22.38	27.35	29.98	35.61
5.49	15.78	22.46	27.4	30.05	35.65
5.64	15.89	22.52	27.43	30.11	35.65
5.79	15.97	22.58	27.45	30.14	35.67
5.94	16.04	22.61	27.46	30.16	35.65
6.09	16.11	22.64	27.44	30.16	35.66
6.24	16.13	22.67	27.43	30.15	35.62
6.39	16.19	22.68	27.41	30.14	35.6
6.54	16.22	22.69	27.37	30.12	35.57
6.69	16.25	22.69	27.33	30.09	35.52
6.84	16.25	22.67	27.28	30.05	35.47
6.99	16.27	22.68	27.23	30	35.41
7.14	16.27	22.66	27.18	29.95	35.36
7.29	16.27	22.64	27.11	29.88	35.3
7.44	16.27	22.61	27.06	29.83	35.23
7.59	16.25	22.59	27	29.77	35.15
7.74	16.24	22.57	26.94	29.7	35.07
7.89	16.24	22.54	26.87	29.63	35
8.04	16.21	22.49	26.8	29.56	34.92
8.19	16.19	22.45	26.72	29.49	34.83
8.34	16.17	22.42	26.67	29.42	34.75
8.49	16.14	22.38	26.61	29.34	34.66
8.64	16.13	22.33	26.54	29.25	34.57
8.79	16.09	22.28	26.45	29.18	34.48
8.94	16.07	22.24	26.39	29.1	34.39
9.09	16.04	22.19	26.3	29.03	34.29
9.24	16.01	22.15	26.25	28.94	34.21
9.39	15.98	22.1	26.16	28.87	34.12
9.54	15.95	22.04	26.09	28.78	34.01
9.69	15.92	22	26.02	28.69	33.93
9.84	15.89	21.94	25.94	28.62	33.82
9.99	15.84	21.89	25.87	28.52	33.73
10.14	15.81	21.84	25.78	28.44	33.63
10.29	15.78	21.77	25.72	28.36	33.55
10.44	15.73	21.72	25.63	28.27	33.44

Table C1 Cure behavior results of FA /NR compounds with various FA loadings in CV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
10.59	15.7	21.66	25.56	28.18	33.35
10.74	15.66	21.61	25.48	28.1	33.25
10.89	15.63	21.56	25.41	28.01	33.14
11.04	15.6	21.48	25.34	27.93	33.04
11.19	15.56	21.42	25.25	27.85	32.94
11.34	15.52	21.36	25.18	27.76	32.85
11.49	15.49	21.3	25.11	27.68	32.75
11.64	15.46	21.25	25.03	27.58	32.66
11.79	15.41	21.18	24.94	27.51	32.57
11.94	15.38	21.12	24.89	27.42	32.47
12.09	15.34	21.06	24.8	27.32	32.34
12.24	15.3	21	24.72	27.25	32.25
12.39	15.28	20.95	24.65	27.16	32.14
12.54	15.24	20.88	24.57	27.07	32.06
12.69	15.2	20.82	24.52	26.98	31.96
12.84	15.17	20.76	24.44	26.9	31.86
12.99	15.13	20.7	24.36	26.83	31.76
13.14	15.1	20.64	24.29	26.73	31.66
13.29	15.06	20.58	24.21	26.65	31.56
13.44	15.03	20.53	24.15	26.56	31.46
13.59	15	20.47	24.07	26.48	31.37
13.74	14.96	20.42	24.01	26.4	31.28
13.89	14.92	20.36	23.92	26.3	31.19
14.04	14.88	20.31	23.86	26.22	31.09
14.19	14.86	20.26	23.79	26.14	30.97
14.34	14.82	20.2	23.72	26.06	30.88
14.49	14.77	20.14	23.65	25.98	30.79
14.64	14.75	20.08	23.58	25.9	30.7
14.79	14.71	20.04	23.51	25.82	30.6
14.94	14.69	19.99	23.45	25.75	30.51
15.09	14.65	19.94	23.39	25.66	30.42
15.24	14.61	19.88	23.31	25.59	30.31
15.39	14.58	19.84	23.24	25.51	30.24
15.54	14.54	19.77	23.18	25.43	30.14
15.69	14.52	19.74	23.11	25.35	30.05
15.84	14.48	19.69	23.04	25.26	29.95
15.99	14.45	19.64	22.97	25.19	29.86
16.14	14.42	19.59	22.92	25.12	29.76
16.29	14.38	19.54	22.84	25.04	29.67

Table C1 Cure behavior results of FA /NR compounds with various FA loadings in CV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
16.44	14.35	19.5	22.78	24.96	29.59
16.59	14.31	19.45	22.7	24.89	29.48
16.74	14.28	19.39	22.66	24.81	29.4
16.89	14.25	19.35	22.58	24.74	29.32
17.04	14.22	19.31	22.53	24.67	29.24
17.19	14.19	19.26	22.47	24.6	29.12
17.34	14.16	19.21	22.39	24.52	29.04
17.49	14.13	19.17	22.33	24.43	28.95
17.64	14.09	19.13	22.27	24.37	28.87
17.79	14.06	19.08	22.2	24.29	28.79
17.94	14.03	19.02	22.13	24.22	28.69
18.09	13.99	18.98	22.08	24.15	28.59
18.24	13.97	18.94	22.02	24.08	28.52
18.39	13.94	18.91	21.96	24	28.43
18.54	13.91	18.86	21.91	23.94	28.36
18.69	13.88	18.81	21.84	23.87	28.26
18.84	13.85	18.77	21.79	23.79	28.19
18.99	13.83	18.72	21.73	23.74	28.09
19.14	13.79	18.69	21.66	23.66	28.02
19.29	13.78	18.64	21.61	23.59	27.94
19.44	13.74	18.6	21.55	23.52	27.86
19.59	13.72	18.56	21.5	23.46	27.78
19.74	13.68	18.52	21.44	23.39	27.69
19.89	13.66	18.47	21.38	23.31	27.62
20.04	13.62	18.43	21.32	23.26	27.54
20.19	13.6	18.39	21.27	23.19	27.47
20.34	13.58	18.35	21.21	23.13	27.39
20.49	13.55	18.31	21.16	23.07	27.31
20.64	13.52	18.27	21.1	23.01	27.23
20.79	13.49	18.24	21.06	22.94	27.15
20.94	13.47	18.2	20.99	22.88	27.08
21.09	13.44	18.15	20.94	22.82	27.01
21.24	13.42	18.13	20.88	22.75	26.93
21.39	13.39	18.08	20.83	22.7	26.86
21.54	13.37	18.04	20.79	22.63	26.8
21.69	13.34	18	20.73	22.57	26.72
21.84	13.31	17.96	20.69	22.51	26.64
21.99	13.28	17.94	20.62	22.45	26.57
22.14	13.27	17.89	20.58	22.4	26.49

Table C1 Cure behavior results of FA /NR compounds with various FA loadings in CV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
22.29	13.23	17.86	20.52	22.33	26.43
22.44	13.21	17.81	20.49	22.27	26.36
22.59	13.18	17.79	20.43	22.22	26.3
22.74	13.15	17.75	20.39	22.16	26.22
22.89	13.14	17.71	20.34	22.11	26.14
23.04	13.11	17.68	20.29	22.05	26.08
23.19	13.09	17.64	20.25	22	26.01
23.34	13.06	17.61	20.19	21.95	25.94
23.49	13.03	17.56	20.14	21.89	25.87
23.64	13.02	17.53	20.1	21.82	25.8
23.79	12.99	17.5	20.05	21.78	25.73
23.94	12.96	17.45	20.01	21.73	25.67
24.09	12.94	17.43	19.95	21.66	25.6
24.24	12.91	17.39	19.9	21.62	25.54
24.39	12.9	17.36	19.86	21.56	25.47
24.54	12.87	17.32	19.83	21.52	25.41
24.69	12.85	17.3	19.77	21.45	25.34
24.84	12.83	17.26	19.73	21.4	25.28
24.99	12.81	17.23	19.68	21.35	25.21
25.14	12.78	17.2	19.64	21.3	25.15
25.29	12.76	17.17	19.6	21.25	25.09
25.44	12.73	17.14	19.56	21.21	25.03
25.59	12.72	17.1	19.51	21.15	24.97
25.74	12.68	17.08	19.47	21.1	24.91
25.89	12.66	17.05	19.42	21.05	24.85
26.04	12.64	17.02	19.37	21.01	24.79
26.19	12.63	16.98	19.34	20.95	24.73
26.34	12.6	16.96	19.29	20.91	24.67
26.49	12.59	16.93	19.25	20.86	24.62
26.64	12.57	16.91	19.21	20.81	24.54
26.79	12.55	16.86	19.17	20.76	24.49
26.94	12.53	16.84	19.13	20.73	24.44
27.09	12.5	16.81	19.09	20.67	24.39
27.24	12.49	16.78	19.05	20.62	24.32
27.39	12.48	16.76	19.01	20.57	24.27
27.54	12.45	16.73	18.97	20.54	24.21
27.69	12.44	16.7	18.93	20.49	24.16
27.84	12.42	16.68	18.9	20.44	24.1
27.99	12.39	16.64	18.86	20.4	24.05

Table C1 Cure behavior results of FA /NR compounds with various FA loadings in CV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
28.14	12.37	16.62	18.82	20.35	23.99
28.29	12.36	16.59	18.78	20.31	23.93
28.44	12.34	16.56	18.74	20.27	23.89
28.59	12.32	16.54	18.71	20.22	23.83
28.74	12.3	16.51	18.67	20.18	23.79
28.89	12.28	16.49	18.63	20.15	23.73
29.04	12.26	16.45	18.6	20.1	23.67
29.19	12.25	16.43	18.55	20.05	23.63
29.34	12.23	16.41	18.52	20.02	23.55
29.49	12.22	16.39	18.48	19.99	23.51
29.64	12.19	16.36	18.45	19.95	23.47
29.79	12.18	16.33	18.42	19.89	23.41

Table C2 Cure behavior results of FA /NR compounds with various FA loadings in EV systems

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.243	1.258	1.563	1.863	2.075	2.133
0.393	1.124	1.333	1.585	1.772	1.823
0.54	1.052	1.219	1.444	1.621	1.669
0.69	1.016	1.17	1.379	1.559	1.608
0.84	1.011	1.153	1.37	1.557	1.645
0.99	1.027	1.188	1.396	1.592	1.713
1.14	1.078	1.23	1.465	1.654	1.857
1.29	1.171	1.343	1.612	1.782	2.127
1.44	1.344	1.534	1.871	2.005	2.717
1.59	1.667	1.889	2.385	2.439	4.165
1.74	2.257	2.573	3.49	3.327	7.714
1.89	3.331	3.899	5.793	5.197	12.92
2.04	5.026	6.155	9.121	8.125	17.32
2.19	6.913	8.726	12.3	11.24	20.05
2.34	8.418	10.79	13	13.83	21.68
2.49	9.458	12.2	13.74	15.76	22.74
2.64	10.16	13.16	14.83	17.17	23.52
2.79	10.67	13.85	15.61	18.22	24.09
2.94	11.06	14.36	16.22	19.01	24.51
3.09	11.38	14.76	16.68	19.61	24.83

Table C2 Cure behavior results of FA /NR compounds with various FA loadings in EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
3.24	11.62	15.07	17.03	20.06	25.07
3.39	11.83	15.31	17.33	20.4	25.25
3.54	12	15.5	17.56	20.69	25.4
3.69	12.14	15.65	17.76	20.9	25.51
3.84	12.28	15.78	17.92	21.08	25.58
3.99	12.39	15.87	18.03	21.21	25.65
4.14	12.48	15.96	18.15	21.33	25.69
4.29	12.55	16.02	18.23	21.44	25.72
4.44	12.62	16.07	18.3	21.51	25.76
4.59	12.68	16.13	18.36	21.59	25.78
4.74	12.74	16.16	18.4	21.66	25.78
4.89	12.78	16.21	18.45	21.71	25.8
5.04	12.82	16.24	18.49	21.77	25.81
5.19	12.87	16.26	18.52	21.81	25.8
5.34	12.9	16.28	18.53	21.85	25.81
5.49	12.94	16.3	18.56	21.88	25.8
5.64	12.96	16.31	18.57	21.9	25.8
5.79	12.99	16.32	18.59	21.93	25.79
5.94	13.02	16.33	18.6	21.95	25.79
6.09	13.04	16.34	18.62	21.96	25.77
6.24	13.05	16.35	18.62	21.98	25.78
6.39	13.07	16.36	18.63	21.98	25.75
6.54	13.09	16.36	18.63	22	25.75
6.69	13.11	16.37	18.64	22.01	25.72
6.84	13.12	16.38	18.64	22.02	25.71
6.99	13.14	16.37	18.63	22.03	25.69
7.14	13.14	16.37	18.64	22.03	25.67
7.29	13.14	16.37	18.63	22.02	25.66
7.44	13.15	16.38	18.64	22.02	25.64
7.59	13.17	16.38	18.64	22.02	25.62
7.74	13.17	16.37	18.63	22	25.6
7.89	13.18	16.38	18.63	22.01	25.58
8.04	13.18	16.37	18.62	22	25.56
8.19	13.19	16.37	18.62	21.99	25.54
8.34	13.18	16.35	18.63	21.99	25.53
8.49	13.19	16.35	18.61	21.98	25.51
8.64	13.19	16.34	18.6	21.97	25.5
8.79	13.19	16.34	18.6	21.96	25.49
8.94	13.19	16.34	18.6	21.94	25.48

Table C2 Cure behavior results of FA /NR compounds with various FA loadings in EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
9.09	13.19	16.33	18.59	21.94	25.47
9.24	13.19	16.32	18.59	21.93	25.47
9.39	13.2	16.32	18.57	21.92	25.45
9.54	13.19	16.32	18.57	21.92	25.46
9.69	13.2	16.31	18.56	21.92	25.45
9.84	13.2	16.31	18.56	21.91	25.46
9.99	13.19	16.32	18.56	21.91	25.45
10.14	13.19	16.31	18.56	21.9	25.45
10.29	13.19	16.31	18.56	21.91	25.45
10.44	13.19	16.31	18.56	21.88	25.45
10.59	13.19	16.32	18.55	21.88	25.44
10.74	13.18	16.31	18.55	21.88	25.43
10.89	13.18	16.31	18.56	21.89	25.44
11.04	13.17	16.31	18.55	21.89	25.43
11.19	13.17	16.31	18.56	21.88	25.44
11.34	13.17	16.31	18.57	21.87	25.44
11.49	13.16	16.31	18.56	21.88	25.44
11.64	13.15	16.31	18.57	21.89	25.44
11.79	13.14	16.31	18.56	21.87	25.43
11.94	13.14	16.32	18.56	21.88	25.43
12.09	13.14	16.32	18.56	21.87	25.43
12.24	13.13	16.32	18.57	21.88	25.43
12.39	13.12	16.32	18.57	21.87	25.44
12.54	13.12	16.32	18.57	21.88	25.43
12.69	13.12	16.32	18.57	21.88	25.43
12.84	13.11	16.32	18.59	21.87	25.43
12.99	13.1	16.32	18.58	21.87	25.43
13.14	13.09	16.32	18.57	21.87	25.43
13.29	13.09	16.33	18.58	21.87	25.42
13.44	13.08	16.32	18.57	21.86	25.43
13.59	13.08	16.32	18.59	21.86	25.43
13.74	13.07	16.32	18.57	21.86	25.42
13.89	13.06	16.33	18.58	21.84	25.42
14.04	13.07	16.32	18.58	21.85	25.42
14.19	13.06	16.33	18.58	21.85	25.41
14.34	13.06	16.32	18.59	21.85	25.4
14.49	13.05	16.32	18.58	21.84	25.39
14.64	13.05	16.33	18.57	21.84	25.4
14.79	13.05	16.32	18.58	21.83	25.39

Table C2 Cure behavior results of FA /NR compounds with various FA loadings in EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
14.94	13.04	16.33	18.58	21.83	25.39
15.09	13.03	16.32	18.57	21.83	25.38
15.24	13.04	16.32	18.59	21.83	25.38
15.39	13.03	16.32	18.58	21.82	25.37
15.54	13.03	16.31	18.58	21.82	25.37
15.69	13.03	16.32	18.58	21.81	25.37
15.84	13.01	16.33	18.58	21.8	25.36
15.99	13.02	16.32	18.57	21.8	25.36
16.14	13.02	16.32	18.56	21.81	25.35
16.29	13.03	16.32	18.57	21.8	25.35
16.44	13.02	16.32	18.56	21.79	25.36
16.59	13.02	16.31	18.56	21.79	25.34
16.74	13.02	16.3	18.56	21.78	25.34
16.89	13.01	16.31	18.56	21.77	25.33
17.04	13.02	16.3	18.56	21.77	25.32
17.19	13.02	16.3	18.55	21.76	25.32
17.34	13.01	16.3	18.55	21.77	25.32
17.49	13	16.31	18.55	21.76	25.32
17.64	13.01	16.31	18.55	21.76	25.32
17.79	13	16.29	18.55	21.75	25.31
17.94	13.01	16.3	18.55	21.74	25.31
18.09	13.01	16.28	18.55	21.74	25.3
18.24	13	16.29	18.55	21.74	25.3
18.39	13	16.29	18.54	21.74	25.29
18.54	13.02	16.3	18.53	21.73	25.29
18.69	13	16.29	18.54	21.73	25.28
18.84	12.99	16.29	18.53	21.71	25.28
18.99	13	16.28	18.53	21.71	25.28
19.14	12.99	16.29	18.53	21.71	25.27
19.29	12.98	16.27	18.53	21.71	25.26
19.44	12.98	16.28	18.52	21.71	25.25
19.59	12.99	16.27	18.52	21.7	25.26
19.74	12.98	16.27	18.52	21.69	25.25
19.89	12.97	16.26	18.51	21.68	25.25
20.04	12.98	16.26	18.5	21.68	25.24
20.19	12.97	16.26	18.5	21.67	25.23
20.34	12.98	16.26	18.5	21.66	25.23
20.49	12.97	16.27	18.5	21.67	25.23
20.64	12.96	16.26	18.5	21.65	25.22

Table C2 Cure behavior results of FA /NR compounds with various FA loadings in EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
20.79	12.96	16.26	18.5	21.65	25.22
20.94	12.97	16.26	18.5	21.64	25.21
21.09	12.96	16.25	18.5	21.64	25.2
21.24	12.96	16.24	18.49	21.64	25.2
21.39	12.96	16.25	18.49	21.63	25.19
21.54	12.96	16.25	18.49	21.62	25.18
21.69	12.95	16.23	18.48	21.61	25.18
21.84	12.95	16.23	18.49	21.61	25.18
21.99	12.94	16.24	18.46	21.6	25.16
22.14	12.95	16.23	18.45	21.59	25.15
22.29	12.95	16.23	18.46	21.6	25.15
22.44	12.94	16.22	18.46	21.59	25.15
22.59	12.94	16.21	18.45	21.59	25.14
22.74	12.94	16.22	18.45	21.58	25.14
22.89	12.93	16.21	18.45	21.58	25.13
23.04	12.93	16.21	18.45	21.57	25.12
23.19	12.93	16.21	18.44	21.56	25.1
23.34	12.93	16.21	18.44	21.57	25.11
23.49	12.91	16.21	18.43	21.56	25.1
23.64	12.92	16.21	18.43	21.55	25.09
23.79	12.92	16.2	18.42	21.53	25.09
23.94	12.92	16.2	18.42	21.54	25.09
24.09	12.91	16.2	18.42	21.53	25.08
24.24	12.9	16.19	18.41	21.53	25.07
24.39	12.91	16.19	18.41	21.52	25.07
24.54	12.91	16.18	18.4	21.51	25.06
24.69	12.91	16.19	18.4	21.51	25.06
24.84	12.9	16.17	18.39	21.5	25.05
24.99	12.9	16.17	18.39	21.49	25.04
25.14	12.9	16.17	18.38	21.49	25.04
25.29	12.89	16.16	18.37	21.48	25.03
25.44	12.88	16.17	18.38	21.48	25.03
25.59	12.89	16.16	18.38	21.47	25.02
25.74	12.88	16.15	18.37	21.47	25.02
25.89	12.88	16.15	18.37	21.46	25.01
26.04	12.88	16.16	18.37	21.45	25
26.19	12.87	16.16	18.37	21.45	24.99
26.34	12.87	16.15	18.35	21.44	24.98
26.49	12.86	16.15	18.35	21.44	24.98

Table C2 Cure behavior results of FA /NR compounds with various FA loadings in EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
26.64	12.86	16.14	18.34	21.43	24.97
26.79	12.86	16.13	18.35	21.43	24.97
26.94	12.85	16.13	18.34	21.42	24.95
27.09	12.86	16.13	18.33	21.41	24.96
27.24	12.85	16.13	18.33	21.41	24.95
27.39	12.85	16.12	18.33	21.41	24.94
27.54	12.84	16.11	18.32	21.39	24.95
27.69	12.84	16.12	18.32	21.39	24.92
27.84	12.84	16.11	18.31	21.38	24.93
27.99	12.84	16.1	18.31	21.37	24.92
28.14	12.84	16.1	18.31	21.36	24.91
28.29	12.83	16.09	18.3	21.35	24.9
28.44	12.84	16.09	18.28	21.34	24.9
28.59	12.83	16.08	18.28	21.33	24.9
28.74	12.83	16.09	18.28	21.32	24.9
28.89	12.82	16.09	18.28	21.31	24.88
29.04	12.82	16.08	18.27	21.3	24.88
29.19	12.82	16.08	18.26	21.29	24.87
29.34	12.81	16.07	18.27	21.29	24.86
29.49	12.82	16.08	18.26	21.28	24.86
29.64	12.81	16.07	18.26	21.27	24.86
29.79	12.8	16.07	18.26	21.26	24.85

Table C3 Cure behavior results of FA /NR compounds with various FA loadings in semi-EV systems

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.243	1.386	1.654	1.58	1.252	1.821
0.393	1.221	1.426	1.336	1.066	1.538
0.54	1.147	1.317	1.227	0.974	1.411
0.69	1.106	1.257	1.187	0.939	1.397
0.84	1.088	1.244	1.202	0.994	1.47
0.99	1.097	1.272	1.272	1.086	1.629
1.14	1.144	1.332	1.42	1.289	1.946

Table C3 Cure behavior results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
1.29	1.232	1.444	1.707	1.729	2.753
1.44	1.373	1.651	2.377	2.984	5.226
1.59	1.648	2.042	4.016	6.45	10.92
1.74	2.131	2.823	7.329	11.87	17.69
1.89	2.974	4.337	11.21	16.68	22.69
2.04	4.403	6.872	14.26	19.99	25.75
2.19	6.351	9.958	16.45	22.24	27.63
2.34	8.314	12.61	18.04	23.8	28.85
2.49	9.916	14.52	19.19	24.85	29.68
2.64	11.11	15.82	20.01	25.6	30.23
2.79	11.97	16.73	20.6	26.11	30.59
2.94	12.59	17.35	21.03	26.46	30.85
3.09	13.02	17.81	21.34	26.73	30.99
3.24	13.34	18.14	21.59	26.9	31.1
3.39	13.57	18.4	21.76	27.02	31.16
3.54	13.75	18.58	21.9	27.11	31.19
3.69	13.89	18.72	21.99	27.15	31.2
3.84	13.99	18.82	22.06	27.18	31.19
3.99	14.09	18.9	22.1	27.19	31.18
4.14	14.14	18.96	22.14	27.19	31.14
4.29	14.19	18.99	22.16	27.19	31.1
4.44	14.23	19.02	22.18	27.16	31.05
4.59	14.28	19.05	22.19	27.13	31.01
4.74	14.3	19.07	22.18	27.09	30.97
4.89	14.32	19.08	22.17	27.08	30.92
5.04	14.33	19.07	22.16	27.04	30.87
5.19	14.34	19.08	22.14	27.01	30.82
5.34	14.35	19.08	22.13	26.96	30.79
5.49	14.35	19.08	22.1	26.92	30.72
5.64	14.36	19.07	22.07	26.87	30.66
5.79	14.36	19.05	22.04	26.81	30.6
5.94	14.36	19.04	22.02	26.77	30.55
6.09	14.36	19.03	21.99	26.73	30.5
6.24	14.35	19.02	21.97	26.68	30.43
6.39	14.35	18.99	21.93	26.62	30.36
6.54	14.34	18.98	21.9	26.55	30.29
6.69	14.34	18.96	21.86	26.5	30.22
6.84	14.33	18.94	21.82	26.44	30.16
6.99	14.32	18.93	21.79	26.39	30.1

Table C3 Cure behavior results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
7.14	14.31	18.9	21.75	26.34	30.04
7.29	14.3	18.89	21.73	26.28	29.98
7.44	14.3	18.86	21.68	26.21	29.93
7.59	14.28	18.83	21.65	26.14	29.87
7.74	14.27	18.82	21.62	26.11	29.82
7.89	14.26	18.8	21.59	26.06	29.78
8.04	14.24	18.77	21.56	26.02	29.73
8.19	14.24	18.75	21.53	26	29.68
8.34	14.23	18.73	21.51	25.98	29.67
8.49	14.21	18.71	21.48	25.95	29.63
8.64	14.2	18.68	21.46	25.92	29.62
8.79	14.2	18.67	21.45	25.89	29.59
8.94	14.18	18.65	21.42	25.86	29.56
9.09	14.17	18.64	21.41	25.84	29.53
9.24	14.16	18.62	21.39	25.81	29.5
9.39	14.16	18.61	21.37	25.79	29.49
9.54	14.15	18.6	21.35	25.76	29.46
9.69	14.15	18.58	21.33	25.74	29.43
9.84	14.15	18.57	21.32	25.72	29.41
9.99	14.14	18.56	21.3	25.68	29.38
10.14	14.14	18.55	21.29	25.66	29.37
10.29	14.13	18.54	21.26	25.65	29.34
10.44	14.12	18.53	21.26	25.63	29.33
10.59	14.11	18.53	21.24	25.6	29.31
10.74	14.11	18.51	21.23	25.58	29.29
10.89	14.1	18.51	21.21	25.56	29.26
11.04	14.1	18.5	21.19	25.54	29.24
11.19	14.09	18.49	21.18	25.54	29.23
11.34	14.09	18.48	21.18	25.51	29.21
11.49	14.08	18.48	21.16	25.49	29.19
11.64	14.08	18.46	21.14	25.47	29.16
11.79	14.08	18.44	21.12	25.43	29.14
11.94	14.07	18.43	21.11	25.42	29.12
12.09	14.05	18.43	21.09	25.4	29.1
12.24	14.05	18.43	21.09	25.39	29.09
12.39	14.05	18.41	21.07	25.36	29.07
12.54	14.04	18.41	21.06	25.34	29.06
12.69	14.04	18.39	21.04	25.31	29.03
12.84	14.03	18.38	21.02	25.31	28.99

Table C3 Cure behavior results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
12.99	14.03	18.38	21.01	25.28	28.98
13.14	14.02	18.37	21	25.27	28.95
13.29	14.02	18.37	20.99	25.25	28.94
13.44	14.01	18.35	20.97	25.23	28.93
13.59	14.01	18.35	20.96	25.21	28.91
13.74	14	18.33	20.95	25.19	28.89
13.89	13.99	18.33	20.91	25.17	28.87
14.04	13.99	18.31	20.91	25.15	28.84
14.19	13.99	18.31	20.89	25.14	28.82
14.34	13.97	18.3	20.87	25.11	28.81
14.49	13.97	18.3	20.86	25.09	28.8
14.64	13.97	18.29	20.85	25.07	28.79
14.79	13.96	18.28	20.84	25.05	28.75
14.94	13.96	18.28	20.82	25.04	28.74
15.09	13.95	18.26	20.81	25.02	28.7
15.24	13.94	18.26	20.79	25	28.7
15.39	13.94	18.25	20.78	24.97	28.69
15.54	13.93	18.25	20.77	24.96	28.66
15.69	13.92	18.24	20.77	24.94	28.65
15.84	13.92	18.22	20.75	24.92	28.64
15.99	13.92	18.22	20.74	24.9	28.61
16.14	13.91	18.21	20.73	24.89	28.59
16.29	13.91	18.2	20.7	24.86	28.57
16.44	13.9	18.19	20.7	24.84	28.56
16.59	13.9	18.17	20.68	24.83	28.55
16.74	13.88	18.16	20.67	24.8	28.52
16.89	13.88	18.15	20.65	24.79	28.51
17.04	13.87	18.16	20.64	24.78	28.49
17.19	13.87	18.14	20.63	24.76	28.47
17.34	13.86	18.13	20.61	24.74	28.45
17.49	13.86	18.12	20.6	24.73	28.45
17.64	13.85	18.11	20.58	24.71	28.41
17.79	13.84	18.11	20.58	24.69	28.4
17.94	13.84	18.09	20.55	24.69	28.4
18.09	13.83	18.09	20.54	24.66	28.37
18.24	13.83	18.07	20.52	24.63	28.35
18.39	13.82	18.06	20.51	24.62	28.34
18.54	13.81	18.06	20.5	24.61	28.34
18.69	13.8	18.04	20.49	24.59	28.32

Table C3 Cure behavior results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
18.84	13.8	18.03	20.47	24.56	28.3
18.99	13.79	18.02	20.46	24.54	28.29
19.14	13.79	18.02	20.44	24.52	28.26
19.29	13.79	18.01	20.43	24.51	28.22
19.44	13.78	18.01	20.42	24.49	28.21
19.59	13.78	17.99	20.41	24.49	28.2
19.74	13.77	17.98	20.39	24.47	28.19
19.89	13.76	17.98	20.39	24.44	28.16
20.04	13.76	17.96	20.37	24.42	28.15
20.19	13.76	17.96	20.36	24.4	28.13
20.34	13.75	17.94	20.34	24.39	28.11
20.49	13.74	17.94	20.34	24.37	28.09
20.64	13.74	17.93	20.32	24.35	28.07
20.79	13.73	17.91	20.31	24.35	28.06
20.94	13.72	17.91	20.3	24.32	28.05
21.09	13.72	17.9	20.29	24.3	28.03
21.24	13.71	17.89	20.27	24.29	28
21.39	13.7	17.89	20.27	24.26	28
21.54	13.7	17.88	20.27	24.26	27.98
21.69	13.69	17.86	20.25	24.24	27.95
21.84	13.69	17.85	20.23	24.22	27.94
21.99	13.68	17.85	20.23	24.21	27.92
22.14	13.68	17.84	20.2	24.18	27.91
22.29	13.67	17.84	20.19	24.16	27.89
22.44	13.67	17.83	20.18	24.15	27.87
22.59	13.66	17.82	20.17	24.14	27.85
22.74	13.66	17.81	20.17	24.12	27.82
22.89	13.65	17.8	20.16	24.11	27.8
23.04	13.64	17.79	20.14	24.09	27.79
18.84	13.8	18.03	20.47	24.56	28.3
18.99	13.79	18.02	20.46	24.54	28.29
19.14	13.79	18.02	20.44	24.52	28.26
19.29	13.79	18.01	20.43	24.51	28.22
19.44	13.78	18.01	20.42	24.49	28.21
19.59	13.78	17.99	20.41	24.49	28.2
19.74	13.77	17.98	20.39	24.47	28.19
19.89	13.76	17.98	20.39	24.44	28.16
20.04	13.76	17.96	20.37	24.42	28.15
20.19	13.76	17.96	20.36	24.4	28.13

Table C3 Cure behavior results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
20.34	13.75	17.94	20.34	24.39	28.11
20.49	13.74	17.94	20.34	24.37	28.09
20.64	13.74	17.93	20.32	24.35	28.07
20.79	13.73	17.91	20.31	24.35	28.06
20.94	13.72	17.91	20.3	24.32	28.05
21.09	13.72	17.9	20.29	24.3	28.03
21.24	13.71	17.89	20.27	24.29	28
21.39	13.7	17.89	20.27	24.26	28
21.54	13.7	17.88	20.27	24.26	27.98
21.69	13.69	17.86	20.25	24.24	27.95
21.84	13.69	17.85	20.23	24.22	27.94
21.99	13.68	17.85	20.23	24.21	27.92
22.14	13.68	17.84	20.2	24.18	27.91
22.29	13.67	17.84	20.19	24.16	27.89
22.44	13.67	17.83	20.18	24.15	27.87
22.59	13.66	17.82	20.17	24.14	27.85
22.74	13.66	17.81	20.17	24.12	27.82
22.89	13.65	17.8	20.16	24.11	27.8
23.04	13.64	17.79	20.14	24.09	27.79
23.19	13.63	17.79	20.13	24.07	27.78
23.34	13.63	17.78	20.11	24.05	27.77
23.49	13.63	17.77	20.1	24.04	27.75
23.64	13.61	17.76	20.09	24.02	27.71
23.79	13.61	17.75	20.07	24	27.7
23.94	13.61	17.74	20.07	23.99	27.68
24.09	13.6	17.73	20.05	23.99	27.66
24.24	13.6	17.73	20.05	23.97	27.65
24.39	13.59	17.72	20.03	23.95	27.62
24.54	13.58	17.72	20.03	23.94	27.6
24.69	13.58	17.7	20.01	23.92	27.58
24.84	13.58	17.69	19.99	23.91	27.56
24.99	13.56	17.68	19.99	23.89	27.54
25.14	13.56	17.67	19.98	23.87	27.52
25.29	13.55	17.67	19.97	23.86	27.5
25.44	13.55	17.65	19.95	23.85	27.48
25.59	13.54	17.65	19.94	23.83	27.47
25.74	13.54	17.64	19.93	23.82	27.44
25.89	13.54	17.63	19.91	23.8	27.42
26.04	13.52	17.62	19.9	23.79	27.39

Table C3 Cure behavior results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
26.19	13.52	17.62	19.89	23.77	27.38
26.34	13.52	17.61	19.88	23.75	27.35
26.49	13.51	17.6	19.87	23.73	27.31
26.64	13.5	17.59	19.87	23.72	27.29
26.79	13.5	17.59	19.84	23.7	27.27
26.94	13.49	17.57	19.82	23.69	27.25
27.09	13.49	17.57	19.82	23.67	27.23
27.24	13.49	17.56	19.81	23.66	27.19
27.39	13.49	17.55	19.81	23.64	27.17
27.54	13.47	17.54	19.79	23.63	27.15
27.69	13.47	17.54	19.78	23.62	27.12
27.84	13.46	17.53	19.77	23.6	27.1
27.99	13.46	17.51	19.76	23.58	27.09
28.14	13.45	17.51	19.74	23.57	27.06
28.29	13.44	17.49	19.73	23.55	27.03
28.44	13.44	17.5	19.72	23.53	27.01
28.59	13.43	17.49	19.71	23.53	26.98
28.74	13.43	17.48	19.7	23.52	26.95
28.89	13.43	17.46	19.69	23.5	26.94
29.04	13.42	17.45	19.69	23.49	26.9
29.19	13.41	17.45	19.67	23.47	26.89
29.34	13.4	17.44	19.66	23.46	26.86
29.49	13.4	17.43	19.65	23.45	26.83
29.64	13.39	17.43	19.64	23.44	26.8
29.79	13.39	17.41	19.64	23.42	26.78

Table C4 Cure behavior results of FA /NR compounds with various FA loadings in peroxide systems

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.383	1.37	1.425	2.003	2.868	2.62
0.68	1.261	1.295	1.827	2.781	2.793
0.98	1.27	1.359	1.887	2.9	3.138
1.28	1.317	1.433	1.95	3.018	3.478
1.58	1.361	1.517	2.027	3.151	3.838
1.88	1.418	1.598	2.114	3.295	4.204

Table C4 Cure behavior results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
2.18	1.464	1.688	2.205	3.448	4.575
2.48	1.516	1.787	2.302	3.591	4.947
2.78	1.571	1.885	2.4	3.746	5.302
3.08	1.626	1.978	2.497	3.888	5.669
3.38	1.686	2.068	2.6	4.024	6.029
3.68	1.741	2.166	2.7	4.169	6.371
3.98	1.793	2.264	2.798	4.31	6.712
4.28	1.849	2.358	2.895	4.443	7.057
4.58	1.896	2.444	2.991	4.573	7.39
4.88	1.957	2.538	3.096	4.71	7.718
5.18	2.006	2.633	3.193	4.839	8.044
5.48	2.063	2.723	3.293	4.956	8.364
5.78	2.106	2.816	3.39	5.076	8.685
6.08	2.16	2.903	3.488	5.193	8.986
6.38	2.21	2.982	3.589	5.308	9.305
6.68	2.261	3.073	3.687	5.419	9.603
6.98	2.308	3.147	3.777	5.541	9.896
7.28	2.363	3.237	3.871	5.648	10.19
7.58	2.413	3.306	3.964	5.75	10.48
7.88	2.466	3.383	4.054	5.86	10.76
8.18	2.508	3.473	4.147	5.966	11.04
8.48	2.544	3.549	4.238	6.056	11.32
8.78	2.587	3.621	4.333	6.143	11.59
9.08	2.636	3.69	4.422	6.24	11.86
9.38	2.676	3.756	4.501	6.33	12.12
9.68	2.722	3.82	4.591	6.425	12.37
9.98	2.762	3.898	4.667	6.51	12.62
10.28	2.801	3.973	4.757	6.586	12.88
10.58	2.852	4.033	4.85	6.671	13.12
10.88	2.885	4.097	4.929	6.763	13.35
11.18	2.922	4.156	5.002	6.837	13.58
11.48	2.964	4.22	5.084	6.918	13.82
11.78	2.993	4.274	5.166	6.988	14.03
12.08	3.034	4.332	5.243	7.07	14.25
12.38	3.072	4.398	5.32	7.142	14.48
12.68	3.103	4.449	5.397	7.213	14.69
12.98	3.144	4.507	5.469	7.284	14.9
10.28	2.801	3.973	4.757	6.586	12.88
10.58	2.852	4.033	4.85	6.671	13.12

Table C4 Cure behavior results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
10.88	2.885	4.097	4.929	6.763	13.35
11.18	2.922	4.156	5.002	6.837	13.58
11.48	2.964	4.22	5.084	6.918	13.82
11.78	2.993	4.274	5.166	6.988	14.03
12.08	3.034	4.332	5.243	7.07	14.25
12.38	3.072	4.398	5.32	7.142	14.48
12.68	3.103	4.449	5.397	7.213	14.69
12.98	3.144	4.507	5.469	7.284	14.9
13.28	3.172	4.563	5.553	7.355	15.11
13.58	3.21	4.62	5.622	7.426	15.31
13.88	3.24	4.671	5.702	7.491	15.51
14.18	3.267	4.72	5.773	7.561	15.7
14.48	3.3	4.775	5.832	7.615	15.9
14.78	3.335	4.83	5.908	7.685	16.1
15.08	3.361	4.875	5.969	7.739	16.29
15.38	3.39	4.918	6.041	7.801	16.47
15.68	3.423	4.964	6.104	7.861	16.65
15.98	3.441	5.005	6.162	7.921	16.83
16.28	3.474	5.048	6.228	7.978	17
16.58	3.494	5.084	6.285	8.032	17.18
16.88	3.53	5.133	6.341	8.086	17.35
17.18	3.549	5.184	6.405	8.143	17.51
17.48	3.583	5.232	6.467	8.196	17.67
17.78	3.603	5.269	6.519	8.245	17.83
18.08	3.64	5.308	6.583	8.296	17.99
18.38	3.651	5.356	6.629	8.351	18.15
18.68	3.68	5.389	6.69	8.4	18.29
18.98	3.705	5.423	6.739	8.454	18.44
19.28	3.728	5.464	6.796	8.492	18.59
19.58	3.749	5.496	6.852	8.537	18.73
19.88	3.764	5.537	6.901	8.586	18.88
20.18	3.788	5.564	6.952	8.633	19.01
20.48	3.807	5.598	7.009	8.676	19.14
20.78	3.827	5.634	7.054	8.713	19.29
21.08	3.848	5.659	7.098	8.759	19.4
21.38	3.864	5.691	7.146	8.8	19.53
21.68	3.88	5.727	7.187	8.85	19.66
21.98	3.902	5.753	7.228	8.888	19.79
22.28	3.922	5.796	7.276	8.932	19.91

Table C4 Cure behavior results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
22.58	3.935	5.823	7.317	8.974	20.01
22.88	3.952	5.845	7.36	9.01	20.13
23.18	3.972	5.873	7.405	9.041	20.25
23.48	3.988	5.899	7.434	9.082	20.37
23.78	4.006	5.929	7.487	9.118	20.47
24.08	4.028	5.949	7.521	9.147	20.57
24.38	4.044	5.976	7.555	9.19	20.69
24.68	4.057	6.003	7.604	9.229	20.78
24.98	4.069	6.025	7.642	9.256	20.89
25.28	4.084	6.055	7.675	9.301	20.98
25.58	4.101	6.074	7.701	9.325	21.08
25.88	4.114	6.1	7.743	9.365	21.17
26.18	4.128	6.123	7.775	9.399	21.27
26.48	4.15	6.145	7.803	9.428	21.36
26.78	4.158	6.168	7.841	9.455	21.47
27.08	4.169	6.189	7.877	9.488	21.56
27.38	4.179	6.211	7.906	9.512	21.64
27.68	4.194	6.229	7.937	9.546	21.73
27.98	4.208	6.244	7.96	9.579	21.81
28.28	4.217	6.274	7.996	9.609	21.89
28.58	4.224	6.29	8.029	9.629	21.98
28.88	4.236	6.298	8.044	9.667	22.06
29.18	4.248	6.325	8.081	9.69	22.13
29.48	4.264	6.343	8.098	9.723	22.22
29.78	4.269	6.362	8.129	9.741	22.3
30.08	4.278	6.383	8.163	9.775	22.36
30.38	4.284	6.402	8.19	9.792	22.45
30.68	4.295	6.41	8.215	9.826	22.52
30.98	4.31	6.427	8.231	9.85	22.59
31.28	4.319	6.45	8.256	9.868	22.66
31.58	4.324	6.467	8.286	9.898	22.74
31.88	4.335	6.475	8.312	9.919	22.78
32.18	4.338	6.49	8.331	9.937	22.85
32.48	4.352	6.514	8.349	9.96	22.92
32.78	4.361	6.52	8.378	9.985	22.98
33.08	4.372	6.533	8.393	10.01	23.06
33.38	4.38	6.545	8.418	10.02	23.12
33.68	4.383	6.568	8.438	10.05	23.17
33.98	4.394	6.582	8.448	10.07	23.22

Table C4 Cure behavior results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
34.28	4.396	6.588	8.473	10.08	23.28
34.58	4.405	6.597	8.498	10.11	23.32
34.88	4.41	6.609	8.512	10.13	23.39
35.18	4.425	6.63	8.528	10.15	23.44
35.48	4.43	6.638	8.549	10.17	23.5
35.78	4.432	6.65	8.569	10.19	23.54
36.08	4.445	6.664	8.58	10.19	23.6
36.38	4.451	6.67	8.601	10.23	23.63
36.68	4.454	6.682	8.621	10.25	23.67
36.98	4.462	6.693	8.632	10.26	23.72
37.28	4.471	6.704	8.648	10.29	23.77
37.58	4.475	6.715	8.67	10.29	23.81
37.88	4.489	6.72	8.68	10.31	23.86
38.18	4.491	6.73	8.698	10.33	23.91
38.48	4.493	6.747	8.71	10.34	23.94
38.78	4.5	6.747	8.73	10.35	23.98
39.08	4.499	6.763	8.74	10.38	24.03
39.38	4.512	6.771	8.752	10.38	24.06
39.68	4.515	6.784	8.766	10.4	24.1
39.98	4.517	6.786	8.782	10.42	24.14
40.28	4.514	6.794	8.789	10.42	24.19
40.58	4.527	6.802	8.802	10.44	24.22
40.88	4.529	6.816	8.83	10.46	24.26
41.18	4.543	6.822	8.841	10.47	24.29
41.48	4.539	6.832	8.842	10.48	24.32
41.78	4.545	6.831	8.85	10.48	24.36
42.08	4.546	6.847	8.862	10.51	24.39
42.38	4.552	6.852	8.884	10.52	24.43
42.68	4.56	6.854	8.89	10.53	24.45
42.98	4.568	6.871	8.904	10.54	24.49
43.28	4.571	6.879	8.912	10.55	24.52
43.58	4.571	6.884	8.915	10.56	24.55
43.88	4.577	6.89	8.928	10.58	24.58
44.18	4.576	6.891	8.932	10.59	24.61
44.48	4.577	6.897	8.947	10.6	24.63
44.78	4.585	6.917	8.952	10.6	24.66
45.08	4.595	6.923	8.963	10.62	24.68
45.38	4.596	6.92	8.981	10.63	24.7
45.68	4.601	6.923	8.983	10.63	24.73

Table C4 Cure behavior results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
45.98	4.6	6.935	8.996	10.64	24.75
46.28	4.609	6.939	8.997	10.65	24.77
46.58	4.617	6.942	9.009	10.66	24.79
46.88	4.607	6.948	9.019	10.68	24.82
47.18	4.614	6.954	9.025	10.68	24.83
47.48	4.616	6.952	9.032	10.69	24.86
47.78	4.623	6.958	9.042	10.7	24.88
48.08	4.621	6.978	9.043	10.71	24.9
48.38	4.626	6.975	9.053	10.71	24.92
48.68	4.624	6.979	9.064	10.73	24.94
48.98	4.633	6.984	9.068	10.74	24.97
49.28	4.634	6.989	9.072	10.75	24.98
49.58	4.634	6.991	9.081	10.75	25
49.88	4.637	6.998	9.084	10.75	25.01
50.18	4.633	6.994	9.088	10.77	25.04
50.48	4.643	7.004	9.093	10.77	25.06
50.78	4.648	7.007	9.097	10.78	25.08
51.08	4.649	7.015	9.104	10.79	25.08
51.38	4.648	7.021	9.106	10.79	25.1
51.68	4.649	7.018	9.111	10.8	25.11
51.98	4.638	7.026	9.114	10.8	25.13
52.28	4.648	7.028	9.132	10.81	25.15
52.58	4.649	7.033	9.133	10.82	25.17
52.88	4.659	7.036	9.133	10.83	25.19
53.18	4.663	7.034	9.143	10.83	25.19
53.48	4.658	7.043	9.144	10.83	25.21
53.78	4.659	7.043	9.146	10.84	25.22
54.08	4.661	7.044	9.149	10.86	25.23
54.38	4.663	7.05	9.149	10.86	25.25
54.68	4.668	7.041	9.159	10.87	25.26
54.98	4.672	7.049	9.163	10.87	25.27
55.28	4.675	7.058	9.165	10.87	25.3
55.58	4.673	7.051	9.169	10.88	25.31
55.88	4.674	7.053	9.174	10.89	25.32
56.18	4.676	7.06	9.182	10.88	25.33
56.48	4.678	7.064	9.186	10.88	25.34
56.78	4.674	7.061	9.187	10.9	25.36
57.08	4.679	7.064	9.191	10.9	25.36
57.38	4.679	7.063	9.19	10.9	25.36

Table C4 Cure behavior results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Torque (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
57.68	4.685	7.067	9.199	10.92	25.38
57.98	4.685	7.071	9.206	10.91	25.39
58.28	4.684	7.076	9.198	10.92	25.4
58.58	4.685	7.074	9.203	10.93	25.4
58.88	4.685	7.075	9.206	10.93	25.41
59.18	4.684	7.075	9.208	10.94	25.43
59.48	4.688	7.084	9.211	10.94	25.43

Table C5 Degree of reversion of FA filled NR vulcanizates in sulfur systems

Sulfur systems	Degree of reversion				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	10.66	12.82	15.45	15.62	15.51
EV	1.39	0.39	0.41	0.98	1.78
Semi-EV	3.09	4.60	6.57	8.27	8.39

Table C6 Marching rate of FA /NR compounds in peroxide systems

Peroxide systems	Marching rate (dNm/min)				
	0 phr	50 phr	100 phr	150 phr	200 phr
	0.08	0.133	0.177	0.18	0.518

Table C7 Scorch time of FA /NR compounds in sulfur and peroxide systems

Curing systems	Scorch time (min)				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	1.42 $\pm$ 0.10	1.23 $\pm$ 0.10	1.09 $\pm$ 0.20	1.12 $\pm$ 0.10	0.96 $\pm$ 0.10
EV	1.87 $\pm$ 0.20	1.77 $\pm$ 0.10	1.68 $\pm$ 0.10	1.72 $\pm$ 0.15	1.51 $\pm$ 0.30
Semi-EV	1.62 $\pm$ 0.10	1.55 $\pm$ 0.20	1.49 $\pm$ 0.20	1.41 $\pm$ 0.10	1.32 $\pm$ 0.20
DCP	11.45 $\pm$ 0.10	6.69 $\pm$ 0.30	6.25 $\pm$ 0.20	5.73 $\pm$ 0.40	1.82 $\pm$ 0.10

Table C8 Cure time of FA /NR compounds in sulfur and peroxide systems

Curing systems	Cure time (min)				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	4.47 $\pm$ 0.05	3.89 $\pm$ 0.20	3.52 $\pm$ 0.10	3.6 $\pm$ 0.20	3.26 $\pm$ 0.20
EV	3.51 $\pm$ 0.30	3.1 $\pm$ 0.30	3.05 $\pm$ 0.10	3.15 $\pm$ 0.10	2.57 $\pm$ 0.20
Semi-EV	3.07 $\pm$ 0.10	2.9 $\pm$ 0.30	2.64 $\pm$ 0.10	2.42 $\pm$ 0.10	2.23 $\pm$ 0.20
DCP	31.28 $\pm$ 0.30	33.34 $\pm$ 0.40	31.78 $\pm$ 0.20	33.32 $\pm$ 0.30	33.08 $\pm$ 0.40

Table C9 Torque difference of FA /NR compounds in sulfur and peroxide systems

Curing systems	Torque difference (dNm)				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	15.58 $\pm$ 0.50	21.96 $\pm$ 0.40	26.34 $\pm$ 0.60	29.37 $\pm$ 0.40	34.29 $\pm$ 0.30
EV	12.54 $\pm$ 0.50	15.7 $\pm$ 0.70	19.39 $\pm$ 0.60	21.16 $\pm$ 0.40	24.89 $\pm$ 0.60
Semi-EV	13.64 $\pm$ 0.60	18.35 $\pm$ 0.40	21.56 $\pm$ 0.30	26.73 $\pm$ 0.20	30.34 $\pm$ 0.50
DCP	3.78 $\pm$ 0.50	6.50 $\pm$ 0.30	7.26 $\pm$ 0.60	8.14 $\pm$ 0.40	23.81 $\pm$ 0.30

Table C10 Crosslink density of FA /NR compounds in sulfur and peroxide systems

Curing systems	Crosslink density ($\times 10^{-4}$ mole/dm ³)				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	1.2	1.75	2.59	2.97	3.2
EV	0.94	1.45	2.3	2.78	2.86
Semi-EV	1.09	1.59	2.41	2.9	2.99
DCP	0.264	0.766	1.09	1.17	1.63

Table C11 Time sweep results of FA /NR compounds with various FA loadings in CV systems

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.22	466.9	716.46	967.43	1145.2	1530.1
0.38	467.4	716.04	964.33	1148	1537
0.53	466.9	713.69	965.61	1149.2	1532.3
0.69	465.7	714.33	962.18	1149.4	1531.6
0.85	462.8	713.59	954.62	1145.1	1531.2
1.00	464.7	711.99	957.06	1144.6	1527.4
1.16	463	711.14	958.45	1144.2	1518.1
1.32	457.8	709.86	957.59	1143.7	1522
1.47	458.2	709.86	953.86	1140.7	1517.4
1.63	458.2	710.92	955.14	1141.2	1516.3
1.79	457.9	710.92	950.68	1140.5	1515
1.95	462.3	710.39	953.65	1136.9	1512.4
2.10	459.3	709.33	953.12	1136.3	1513.2
2.26	458.9	709.43	952.69	1132.8	1510.6
2.42	460.2	709.86	953.86	1132.9	1509.3
2.57	459.4	709.75	952.8	1132.7	1508.2
2.73	458.7	709.75	955.78	1130.7	1506.8
2.89	459.1	709.64	955.78	1130.7	1507.4
3.04	457.6	709.64	954.08	1131	1506.3
3.20	457.3	710.39	953.12	1128.8	1504
3.36	459.4	709.11	952.59	1127.3	1505.7
3.51	454.2	708.37	953.44	1127	1506.4
3.67	458.9	711.56	952.27	1128.9	1502.4
3.83	459.7	709.86	953.65	1127	1501.4
3.99	458	710.71	955.78	1125.6	1501.9
4.14	458.6	709.33	952.59	1126.1	1501.9
4.30	459.3	711.56	955.14	1125.6	1500.1
4.46	458.3	709.64	953.65	1125.2	1499
4.61	458	709.11	956	1122.8	1500.3
4.77	461.2	711.56	955.57	1122.8	1500.8
4.93	460	710.5	956	1122.8	1499.6
5.08	465.8	709.75	953.65	1122	1499.6
5.24	461.5	711.88	957.38	1122.4	1501
5.40	461.2	709.86	953.12	1123.6	1500.5
5.56	460.8	710.5	955.78	1122.8	1500.8
5.71	460.8	711.88	955.25	1123.5	1497.5
5.87	462.3	710.71	955.78	1122.8	1500.7
6.03	461.1	709.75	956	1123.5	1500.1
6.18	462.8	710.5	958.66	1122.5	1499.6

Table C11 Time sweep results of FA /NR compounds with various FA loadings in CV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
6.34	461.8	711.46	956.21	1122.5	1500.4
6.50	468.3	711.78	954.82	1123.6	1500.4
6.65	462.3	711.24	956	1123.3	1499
6.81	467.9	711.24	958.98	1122.4	1500.4
6.97	464.9	711.14	957.06	1121.5	1502.4
7.13	464.2	710.71	957.06	1122.9	1502.4
7.28	464.2	710.92	958.66	1123.8	1503.7
7.44	464.4	711.14	958.34	1122.5	1503.5
7.60	465.8	711.24	959.09	1123.1	1503.6
7.75	466	711.24	954.62	1124.3	1505
7.91	467.5	712.2	957.91	1124.7	1505
8.07	467.2	710.39	959.09	1125	1505.8
8.22	468.1	710.39	957.06	1123.8	1504.2
8.38	470.6	710.71	958.13	1123.9	1506
8.54	468.7	710.92	956.64	1125	1506.1
8.69	469.7	710.39	957.59	1124.9	1505.4
8.85	469	711.56	958.34	1124.6	1507.9
9.01	470.6	712.84	960.47	1127.4	1506.4
9.17	470.8	710.71	958.45	1127	1508.6
9.32	468.6	711.88	958.66	1126.3	1508.3
9.48	466.9	711.56	957.91	1126.3	1509.9
9.64	470.4	711.14	959.41	1126.3	1509.3
9.79	472.5	710.92	959.83	1127.4	1511
9.95	471.1	712.63	960.26	1126.3	1511.1
10.11	473.2	711.78	958.66	1126.1	1511.8
10.26	472.8	712.84	959.83	1126.5	1513.1
10.42	473.2	712.2	959.41	1126.8	1512.4
10.58	475.1	711.78	958.02	1128.5	1513.8
10.74	475.7	712.84	960.26	1128.8	1513.5
10.89	473.8	710.5	959.51	1128.8	1514.9
11.05	476.3	711.99	960.58	1127.1	1515.6
11.21	475.4	710.92	961.54	1127.4	1516.3

Table C11 Time sweep results of FA /NR compounds with various FA loadings in CV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
11.36	477.1	712.84	961.64	1128.8	1515.3
11.52	475.3	711.56	960.26	1126.8	1516.8
11.68	475	711.24	962.41	1128.9	1515.3
11.83	475	711.88	960.58	1129.1	1517
11.99	479.5	711.56	960.79	1130.7	1517.9
12.15	478.5	712.63	961.22	1130	1517.5
12.30	478.9	712.31	961.22	1130	1517.4
12.46	478.3	710.71	959.83	1129.5	1519.6
12.62	478.6	710.92	960.47	1130.6	1519.9
12.78	479.5	712.95	961.32	1131	1520.7
12.93	480.9	711.14	959.73	1131.7	1520.9
13.09	478.5	711.99	959.51	1131.6	1522.1
13.25	477.5	713.59	962.18	1132.3	1523.1
13.40	478.8	713.37	962.28	1131.7	1521.6
13.56	478.6	713.59	963.03	1132	1522.9
13.72	482.6	712.63	963.03	1132.4	1524.1
13.87	480	711.46	962.28	1132.4	1524.2
14.03	479.3	713.37	962.71	1132	1525.6
14.19	480.3	710.82	961.86	1132.4	1526.1
14.35	481.4	713.05	962.28	1133.5	1524.2
14.50	480.7	712.63	962.18	1133.5	1524.9
14.66	482.4	712.31	962.39	1132.8	1527.8
14.82	480.4	712.2	961.54	1133.5	1527.4
14.97	482.8	712.84	962.18	1131.8	1525.9
15.13	480.4	712.2	961.64	1132.9	1528.5
15.29	483.9	710.82	961.11	1132.9	1529.3
15.44	483.5	712.2	961.54	1132.3	1528.5
15.60	483.1	712.84	962.92	1132.1	1528.4
15.76	483.9	712.63	961.54	1132.7	1529.1
15.92	484.2	713.69	962.6	1133.7	1530.3
16.07	485.3	714.12	961.96	1132.9	1531.4
16.23	484.7	712.95	964.41	1133.5	1529.9
16.39	485.7	712.31	963.77	1133.8	1530.5
16.54	484.2	713.37	965.91	1133.4	1530.1
16.70	487.4	713.27	964.52	1134.5	1532.7
16.86	488.8	712.95	964.31	1133.9	1532.3

Table C11 Time sweep results of FA /NR compounds with various FA loadings in CV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
17.01	486.4	713.37	965.59	1134.2	1533.3
17.17	490.2	713.05	965.48	1134.5	1533.5
17.33	487.3	712.31	965.37	1133.5	1534.2
17.48	489.9	713.91	964.31	1134.3	1534.5
17.64	489.2	713.05	964.31	1133.2	1535.4
17.80	489.2	712.84	965.37	1133.8	1535.4
17.96	489.8	712.2	965.37	1133.8	1534.5
18.11	490.3	712.31	965.05	1135.6	1534.6
18.27	489.8	712.95	964.41	1134.9	1534
18.43	489.9	712.31	965.16	1136	1536.2
18.58	490.8	713.69	966.12	1133.8	1536
18.74	488.1	714.01	965.16	1136.7	1535.9
18.90	490.8	714.12	965.8	1135.2	1535.6
19.05	489.8	713.37	964.09	1136.6	1535.5
19.21	491.4	712.31	965.16	1135.6	1535.5
19.37	490.6	712.63	964.31	1135.3	1538.3
19.53	490.5	714.33	965.37	1137.7	1537.7
19.68	492	712.95	965.37	1136	1536.7
19.84	493.2	714.97	964.52	1135	1538.7
20.00	494.9	712.95	965.05	1136.9	1537.7
20.15	493	712.95	966.54	1137.3	1538.1
20.31	494.6	713.37	965.59	1137	1538.3
20.47	493.1	712.95	966.22	1138.5	1538.4
20.62	493.1	714.65	965.8	1137.1	1539.1
20.78	495.1	713.37	965.37	1138	1540.4
20.94	493.2	714.44	966.54	1138.5	1540.1
21.09	497.2	714.44	965.91	1138	1540.1
21.25	495.2	714.44	967.5	1137.7	1543.7
21.41	495.3	714.01	967.18	1138.4	1544.7
21.57	494.8	713.91	967.18	1137.8	1543.4
21.72	496.2	714.76	966.54	1138	1543.3
21.88	496.4	714.44	966.12	1138.7	1544.2
22.04	499.9	714.97	966.65	1139.5	1542.7
22.19	497.8	714.65	965.59	1139.2	1544.1
22.35	497.3	716.97	966.97	1138.5	1544.8
22.51	499.8	713.27	968.04	1140.2	1545.4
22.66	498.8	713.59	969	1139.6	1543.6
22.82	499.9	715.08	969.42	1139.2	1544.7
22.98	501	713.59	968.78	1139.5	1547.3

Table C11 Time sweep results of FA /NR compounds with various FA loadings in CV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
23.14	500.4	715.4	968.25	1140.1	1546.8
23.29	500.4	714.01	968.57	1138.5	1545.9
23.45	500.6	715.08	969.32	1140.7	1548.3
23.61	501.2	713.59	968.25	1140.9	1547.2
23.76	501	714.33	970.74	1142	1548.6
23.92	501.2	712.52	968.25	1143.5	1549.4
24.08	501.5	714.76	969.74	1141.7	1551.9
24.23	501.6	714.76	969	1142.7	1549.8
24.39	501.5	713.59	969.74	1141.3	1551.8
24.55	501	715.08	968.78	1142.8	1551.2
24.71	501.9	715.19	969.32	1142.8	1551.4
24.86	502.6	713.69	970.06	1143.3	1550.1
25.02	503.3	712.95	970.49	1144.1	1553
25.18	501.9	714.65	969.74	1144.8	1551.2
25.33	502.3	714.76	969.85	1143.4	1550.8
25.49	503.6	715.19	968.25	1142.8	1552.3
25.65	502.7	714.33	969.42	1143.7	1550
25.80	505.4	712.95	968.78	1143.7	1552.2
25.96	502.8	714.12	968.68	1147.4	1551.8
26.12	504.1	715.19	968.78	1144.4	1553.2
26.27	504.1	713.91	968.68	1144.5	1553.7
26.43	502.7	714.12	970.7	1142.3	1552.7
26.59	502.7	715.72	971.55	1142.7	1554.8
26.75	503.7	715.51	972.19	1142.8	1553.3
26.90	503	715.08	972.51	1144.9	1554.6
27.06	505.1	715.51	971.13	1143.4	1555.5
27.22	504.5	716.46	970.38	1144.1	1555.1
27.37	504.8	716.14	971.87	1145.7	1555.4
27.53	506	715.51	971.77	1144.1	1557
27.69	507.6	715.08	970.91	1144.9	1557
27.84	504.2	714.44	971.77	1144.2	1555.9
28.00	506.9	715.4	971.77	1143.7	1556.8
28.16	506.3	713.05	971.23	1144.2	1557
28.32	507.4	714.12	970.91	1145.6	1555.9
28.47	507.9	714.76	972.83	1145.6	1557.5
28.63	506.9	715.4	972.62	1146.2	1558.9
28.79	507.7	715.82	972.62	1146.9	1557.2
28.94	507.6	715.51	973.04	1145.9	1558.2
29.10	507.6	714.76	972.3	1146.5	1558.7

Table C11 Time sweep results of FA /NR compounds with various FA loadings in CV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
29.26	508.1	715.82	973.36	1144.4	1557.6
29.41	510.6	714.65	971.87	1147.7	1558.3
29.57	508.6	714.97	972.3	1145.7	1558.9
29.73	509.5	715.08	972.19	1149.1	1559
29.88	510.1	715.72	970.81	1147.4	1558.6

Table C12 Time sweep results of FA /NR compounds with various FA loadings in EV systems

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.22	394.67	589.41	723.71	961.44	1216
0.38	395.1	588.35	721.91	954.3	1217
0.53	395.31	587.07	721.47	957.91	1217.1
0.69	393.71	589.67	721.9	954.93	1213.9
0.85	393.39	588.92	722.11	954.4	1214.9
1.00	394.35	587.32	721.79	952.59	1212.5
1.16	393.61	588.53	716.6	951.2	1212.2
1.32	392.65	585.4	718.91	950.67	1211
1.47	393.18	586.15	717.32	952.05	1212.5
1.63	390.71	585.4	719.02	950.24	1211.3
1.79	392.97	583.91	717.1	950.99	1213.2
1.95	392.86	582.63	716.78	949.92	1212.6
2.10	391.8	582.53	717.53	948.75	1212.9
2.26	393.42	583.06	716.04	947.79	1214.6
2.42	391.16	584.02	715.08	947.47	1211.4
2.57	391.05	582.53	715.19	947.79	1211.3
2.73	390.84	583.59	713.59	948.54	1213.4
2.89	391.05	581.99	714.97	948.86	1213.2
3.04	391.26	581.99	712.63	949.82	1215.1
3.20	391.05	579.76	713.91	950.56	1212.4
3.36	390.09	580.72	713.27	950.56	1214.2
3.51	390.09	580.82	710.18	948.43	1214.5
3.67	391.29	581.99	713.05	949.5	1213.9
3.83	390.52	583.06	711.14	950.46	1211.7
3.99	391.26	583.27	713.37	950.56	1212.6
4.14	391.05	582.63	712.63	948.86	1213.5

Table C12 Time sweep results of FA /NR compounds with various FA loadings in EV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
4.30	390.84	581.89	713.05	950.56	1213.2
4.46	391.16	583.38	714.12	954.18	1213.7
4.61	391.48	583.7	713.27	948.75	1213.9
4.77	390.84	582.53	712.84	950.67	1214.6
4.93	391.05	582.85	713.37	950.46	1215.7
5.08	390.52	582.85	713.59	951.63	1214.7
5.24	390.84	582.21	713.59	951.52	1217.4
5.40	390.73	582.63	713.05	953.86	1216.3
5.56	391.05	582.95	713.69	954.18	1221
5.71	391.48	583.27	713.37	952.59	1220
5.87	390.73	583.27	714.76	954.18	1221.3
6.03	390.2	583.7	712.63	953.65	1220.3
6.18	391.05	582.85	714.12	953.44	1221.8
6.34	391.05	583.59	711.56	953.76	1222.7
6.50	391.48	583.38	715.19	953.12	1223
6.65	391.48	582.31	715.72	953.76	1224.6
6.81	391.26	582.53	713.59	953.65	1224.3
6.97	391.58	581.25	712.95	953.76	1225.4
7.13	390.2	583.59	714.33	954.4	1224.5
7.28	389.35	583.91	713.27	954.18	1227.3
7.44	391.26	583.91	713.05	954.08	1226
7.60	390.84	582.95	714.12	954.18	1227.4
7.75	391.16	583.59	713.59	954.72	1227
7.91	391.05	583.27	714.12	953.76	1228.1
8.07	390.2	584.44	714.33	953.86	1229.4
8.22	391.48	583.7	713.91	957.6	1229.1
8.38	390.73	582.95	714.44	954.72	1233.8
8.54	390.41	583.27	714.65	951.85	1229.5
8.69	391.8	582.63	715.4	954.82	1231.7
8.85	390.2	583.7	712.63	955.14	1231
9.01	390.73	582.85	714.44	954.08	1232.8
9.17	390.84	583.27	712.84	953.44	1232.4
9.32	391.9	581.89	714.76	954.5	1233.8
9.48	391.16	582.21	713.27	955.46	1233.5
9.64	390.41	583.06	716.14	955.46	1233
9.79	390.52	583.38	712.84	954.82	1234.4
9.95	390.52	582.53	715.08	954.5	1234.5
10.11	390.84	583.38	713.91	954.82	1234.2
10.26	391.9	583.59	713.37	955.25	1235.6

Table C12 Time sweep results of FA /NR compounds with various FA loadings in EV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
10.42	390.09	583.27	713.59	955.57	1236.2
10.58	391.26	583.27	713.37	954.93	1237.1
10.74	390.84	583.38	714.44	955.46	1236.4
10.89	391.16	582.31	713.59	955.78	1239.1
11.05	391.48	581.89	715.19	956.21	1236.6
11.21	391.26	582.53	713.69	955.89	1238.5
11.36	390.73	582.95	714.01	954.5	1239
11.52	390.84	583.7	714.44	955.14	1238.5
11.68	390.41	582.85	714.01	954.5	1240.1
11.83	390.73	581.25	712.31	956.21	1240.2
11.99	390.84	581.78	715.82	957.38	1239.8
12.15	390.41	582.53	713.69	957.7	1244.9
12.30	391.05	581.99	715.19	955.89	1241.6
12.46	391.9	580.93	714.33	956.95	1239.9
12.62	391.8	583.06	715.82	956.53	1242.3
12.78	391.16	583.06	714.12	956.53	1241.3
12.93	390.2	583.38	712.95	957.38	1241.3
13.09	390.41	582.53	716.78	956.53	1243.7
13.25	391.9	582.95	714.33	958.02	1243.1
13.40	390.84	583.27	714.97	957.06	1243.1
13.56	390.73	583.59	715.51	956.32	1243.4
13.72	389.98	583.38	715.08	958.02	1244.9
13.87	390.84	582.31	715.4	958.02	1244
14.03	390.84	582.63	716.78	957.38	1245.1
14.19	391.16	584.12	716.57	957.38	1247.8
14.35	391.16	583.06	715.51	958.34	1245.5
14.50	391.58	584.66	715.72	958.02	1247.8
14.66	390.52	583.27	714.33	958.02	1243.7
14.82	390.84	581.78	716.04	958.02	1248.8
14.97	392.12	583.59	717.53	959.73	1246.7
15.13	391.9	582.31	716.57	958.98	1250.2
15.29	391.26	582.95	715.72	955.36	1248.8
15.44	390.84	583.06	713.52	959.09	1250.1
15.60	391.16	583.7	716.04	959.41	1249.7
15.76	390.52	582.63	716.78	959.09	1251.3
15.92	391.16	582.95	714.97	958.66	1249.9
16.07	391.48	583.91	715.4	958.45	1250.9
16.23	391.48	583.59	715.82	958.98	1250.9
16.39	391.48	583.59	715.4	959.83	1251.6

Table C12 Time sweep results of FA /NR compounds with various FA loadings in EV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
16.54	392.36	583.38	716.46	960.15	1250.4
16.70	390.2	583.38	717.64	963.15	1250.8
16.86	391.48	584.02	719.53	960.04	1249.9
17.01	390.84	583.27	714.97	959.73	1251.3
17.17	391.05	583.91	716.57	960.26	1252.5
17.33	391.16	583.38	716.78	959.09	1253.1
17.48	391.05	582.63	715.4	960.15	1253.1
17.64	390.09	583.7	715.19	959.51	1253.4
17.80	389.98	584.02	714.97	960.15	1254.8
17.96	391.16	582.85	715.72	960.15	1253.6
18.11	391.05	582.31	715.82	959.19	1253.1
18.27	390.2	584.12	716.89	959.51	1254.1
18.43	391.48	586.29	717.32	960.79	1253.4
18.58	391.16	583.59	718.28	960.15	1254.1
18.74	391.05	583.38	717.85	960.47	1259.9
18.90	391.05	583.91	717.96	960.47	1260
19.05	391.48	583.7	716.78	960.79	1254
19.21	390.84	583.91	717.64	960.9	1255.1
19.37	391.58	584.12	717.32	965.08	1251.9
19.53	391.05	583.91	717.1	961.86	1252.9
19.68	390.52	584.02	717.1	962.18	1250.5
19.84	391.05	583.59	717.32	964.09	1250.2
20.00	390.73	584.12	716.57	960.9	1249
20.15	391.48	584.02	717.1	962.6	1250.5
20.31	391.16	584.76	716.89	962.71	1249.3
20.47	390.52	585.72	717.21	962.71	1251.2
20.62	391.16	585.19	717.1	963.45	1250.6
20.78	391.8	584.34	713.84	963.03	1250.9
20.94	391.48	585.51	716.46	963.45	1252.6
21.09	390.09	584.44	717.1	962.92	1249.4
21.25	389.35	584.44	716.78	964.84	1249.3
21.41	391.26	584.02	717.85	964.84	1251.4
21.57	390.84	584.34	717.1	963.77	1251.2
21.72	391.05	584.76	717.53	963.24	1253.1
21.88	389.77	584.34	716.89	964.31	1250.4
22.04	391.16	583.27	716.46	964.52	1252.2
22.19	391.26	584.44	717.21	964.73	1252.5
22.35	390.41	584.02	717.85	964.84	1251.9
22.51	390.2	584.34	718.28	965.48	1249.7

Table C12 Time sweep results of FA /NR compounds with various FA loadings in EV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
22.66	391.16	583.06	718.91	965.05	1250.6
22.82	391.48	582.95	719.32	964.52	1251.5
22.98	392.22	583.38	718.38	965.05	1251.2
23.14	389.45	583.59	720.28	964.41	1251.7
23.29	391.05	583.59	719.66	963.77	1251.9
23.45	390.73	583.59	717.96	963.77	1252.6
23.61	391.16	583.91	717.53	965.37	1253.7
23.76	390.84	582.85	718.6	964.84	1252.7
23.92	390.73	584.66	719.98	965.37	1255.4
24.08	390.2	584.02	717.21	964.84	1254.3
24.23	391.05	584.66	717.96	965.05	1259
24.39	390.84	584.12	717.64	966.44	1258
24.55	390.52	585.08	718.17	965.48	1259.3
24.71	390.73	584.02	719.34	965.8	1258.3
24.86	390.09	585.08	719.23	965.59	1259.8
25.02	390.73	583.91	718.28	966.22	1260.7
25.18	390.73	584.12	718.6	966.97	1261
25.33	391.05	584.12	719.98	967.29	1262.6
25.49	391.05	582.95	718.38	966.86	1262.3
25.65	391.58	584.44	718.28	967.5	1263.4
25.80	391.26	583.91	719.45	966.86	1262.5
25.96	390.73	584.02	717.32	967.93	1265.3
26.12	390.84	583.91	719.45	966.54	1264
26.27	391.26	584.12	718.28	967.61	1265.4
26.43	391.48	584.34	719.02	967.5	1265
26.59	391.26	584.12	720.3	966.86	1266.1
26.75	391.58	585.4	718.91	968.04	1267.4
26.90	390.73	584.76	718.6	967.93	1267.1
27.06	390.84	584.12	718.38	967.93	1271.8
27.22	391.16	585.08	719.23	966.65	1267.5
27.37	390.2	584.34	719.45	968.04	1269.7
27.53	390.52	585.4	718.38	967.93	1269
27.69	390.2	584.44	718.7	968.04	1270.8
27.84	390.41	585.19	718.91	968.25	1270.4
28.00	390.41	585.4	718.91	967.29	1271.8
28.16	391.26	586.15	718.6	970.74	1271.5
28.32	391.48	585.72	721.24	969	1271
28.47	391.58	586.15	718.38	968.68	1272.4
28.63	391.9	584.76	718.91	968.57	1272.5

Table C12 Time sweep results of FA /NR compounds with various FA loadings in EV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
28.79	391.48	584.76	719.45	969.32	1272.2
28.94	391.48	585.08	718.91	968.68	1273.6
29.10	391.26	584.66	719.34	969.42	1274.2
29.26	391.16	584.12	719.77	969	1275.1
29.41	391.05	584.34	718.28	968.36	1274.4
29.57	391.05	584.34	719.23	969.42	1277.1
29.73	391.58	584.66	719.34	969.1	1274.6
29.88	391.48	584.44	719.23	971.87	1276.5

Table C13 Time sweep results of FA /NR compounds with various FA loadings in semi-EV systems

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.22	424.71	589.41	723.71	961.44	1284
0.38	424.18	588.35	721.91	954.3	1278
0.53	422.9	587.07	721.47	957.91	1279.1
0.69	425.36	589.67	721.9	954.93	1275.9
0.85	423.23	588.92	722.11	954.4	1276.9
1.00	424.08	587.32	721.79	952.59	1274.5
1.16	423.02	588.53	716.6	951.2	1274.2
1.32	423.44	585.4	718.91	950.67	1273
1.47	422.38	586.15	717.32	952.05	1274.5
1.63	422.48	585.4	719.02	950.24	1273.3
1.79	421.63	583.91	717.1	950.99	1275.2
1.95	421.31	582.63	716.78	949.92	1274.6
2.10	420.57	582.53	717.53	948.75	1274.9
2.26	420.35	583.06	716.04	947.79	1276.6
2.42	420.35	584.02	715.08	947.47	1273.4
2.57	420.67	582.53	715.19	947.79	1273.3
2.73	420.35	583.59	713.59	948.54	1275.4
2.89	419.93	581.99	714.97	948.86	1275.2
3.04	419.82	581.99	712.63	949.82	1277.1
3.20	418.97	579.76	713.91	950.56	1274.4
3.36	419.29	580.72	713.27	950.56	1276.2
3.51	418.54	580.82	710.18	948.43	1276.5
3.67	419.18	581.99	713.05	949.5	1275.9

Table C13 Time sweep results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
3.83	419.18	583.06	711.14	950.46	1273.7
3.99	418.75	583.27	713.37	950.56	1274.6
4.14	419.18	582.63	712.63	948.86	1275.5
4.30	420.35	581.89	713.05	950.56	1275.2
4.46	419.5	583.38	714.12	954.18	1275.7
4.61	420.03	583.7	713.27	948.75	1275.9
4.77	421.42	582.53	712.84	950.67	1276.6
4.93	420.89	582.85	713.37	950.46	1277.7
5.08	420.35	582.85	713.59	951.63	1276.7
5.24	420.03	582.21	713.59	951.52	1279.4
5.40	420.35	582.63	713.05	953.86	1278.3
5.56	419.93	582.95	713.69	954.18	1283
5.71	420.99	583.27	713.37	952.59	1282
5.87	420.89	583.27	714.76	954.18	1283.3
6.03	420.03	583.7	712.63	953.65	1282.3
6.18	420.67	582.85	714.12	953.44	1283.8
6.34	420.25	583.59	711.56	953.76	1284.7
6.50	421.31	583.38	715.19	953.12	1285
6.65	420.57	582.31	715.72	953.76	1286.6
6.81	420.03	582.53	713.59	953.65	1286.3
6.97	420.67	581.25	712.95	953.76	1287.4
7.13	421.1	583.59	714.33	954.4	1286.5
7.28	421.74	583.91	713.27	954.18	1289.3
7.44	420.89	583.91	713.05	954.08	1288
7.60	420.57	582.95	714.12	954.18	1289.4
7.75	420.67	583.59	713.59	954.72	1289
7.91	419.61	583.27	714.12	953.76	1290.1
8.07	420.03	584.44	714.33	953.86	1291.4
8.22	420.89	583.7	713.91	957.6	1291.1
8.38	420.99	582.95	714.44	954.72	1295.8
8.54	420.03	583.27	714.65	951.85	1291.5
8.69	420.25	582.63	715.4	954.82	1293.7
8.85	420.99	583.7	712.63	955.14	1293
9.01	420.89	582.85	714.44	954.08	1294.8
9.17	420.25	583.27	712.84	953.44	1294.4
9.32	420.25	581.89	714.76	954.5	1295.8
9.48	420.25	582.21	713.27	955.46	1295.5
9.64	420.03	583.06	716.14	955.46	1295
9.79	421.74	583.38	712.84	954.82	1296.4

Table C13 Time sweep results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
9.95	421.63	582.53	715.08	954.5	1296.5
10.11	420.99	583.38	713.91	954.82	1296.2
10.26	419.29	583.59	713.37	955.25	1297.6
10.42	420.35	583.27	713.59	955.57	1298.2
10.58	421.74	583.27	713.37	954.93	1299.1
10.74	421.1	583.38	714.44	955.46	1298.4
10.89	420.99	582.31	713.59	955.78	1301.1
11.05	421.31	581.89	715.19	956.21	1298.6
11.21	421.42	582.53	713.69	955.89	1300.5
11.36	421.42	582.95	714.01	954.5	1301
11.52	421.95	583.7	714.44	955.14	1300.5
11.68	421.95	582.85	714.01	954.5	1302.1
11.83	422.48	581.25	712.31	956.21	1302.2
11.99	421.74	581.78	715.82	957.38	1301.8
12.15	421.95	582.53	713.69	957.7	1306.9
12.30	421.95	581.99	715.19	955.89	1303.6
12.46	421.1	580.93	714.33	956.95	1301.9
12.62	421.74	583.06	715.82	956.53	1304.3
12.78	420.57	583.06	714.12	956.53	1303.3
12.93	422.16	583.38	712.95	957.38	1303.3
13.09	420.67	582.53	716.78	956.53	1305.7
13.25	421.95	582.95	714.33	958.02	1305.1
13.40	421.1	583.27	714.97	957.06	1305.1
13.56	420.99	583.59	715.51	956.32	1305.4
13.72	420.67	583.38	715.08	958.02	1306.9
13.87	420.57	582.31	715.4	958.02	1306
14.03	420.67	582.63	716.78	957.38	1307.1
14.19	421.42	584.12	716.57	957.38	1309.8
14.35	421.95	583.06	715.51	958.34	1307.5
14.50	421.63	584.66	715.72	958.02	1309.8
14.66	422.16	583.27	714.33	958.02	1305.7
14.82	421.74	581.78	716.04	958.02	1310.8
14.97	421.31	583.59	717.53	959.73	1308.7
15.13	422.48	582.31	716.57	958.98	1312.2
15.29	422.38	582.95	715.72	955.36	1310.8
15.44	422.48	583.06	713.52	959.09	1312.1
15.60	420.89	583.7	716.04	959.41	1311.7
15.76	421.42	582.63	716.78	959.09	1313.3
15.92	421.31	582.95	714.97	958.66	1311.9

Table C13 Time sweep results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
16.07	420.99	583.91	715.4	958.45	1312.9
16.23	420.99	583.59	715.82	958.98	1312.9
16.39	421.42	583.59	715.4	959.83	1313.6
16.54	421.42	583.38	716.46	960.15	1312.4
16.70	421.95	583.38	717.64	963.15	1312.8
16.86	422.38	584.02	719.53	960.04	1311.9
17.01	421.95	583.27	714.97	959.73	1313.3
17.17	422.16	583.91	716.57	960.26	1314.5
17.33	421.42	583.38	716.78	959.09	1315.1
17.48	420.57	582.63	715.4	960.15	1315.1
17.64	422.16	583.7	715.19	959.51	1315.4
17.80	422.38	584.02	714.97	960.15	1316.8
17.96	420.99	582.85	715.72	960.15	1315.6
18.11	421.95	582.31	715.82	959.19	1315.1
18.27	421.1	584.12	716.89	959.51	1316.1
18.43	420.99	586.29	717.32	960.79	1315.4
18.58	421.63	583.59	718.28	960.15	1316.1
18.74	422.48	583.38	717.85	960.47	1321.9
18.90	421.1	583.91	717.96	960.47	1315.6
19.05	421.63	583.7	716.78	960.79	1317.9
19.21	420.99	583.91	717.64	960.9	1318.6
19.37	421.42	584.12	717.32	965.08	1317.5
19.53	422.7	583.91	717.1	961.86	1317.4
19.68	422.48	584.02	717.1	962.18	1318.9
19.84	422.38	583.59	717.32	964.09	1318.3
20.00	422.16	584.12	716.57	960.9	1318.2
20.15	422.38	584.02	717.1	962.6	1318.5
20.31	422.38	584.76	716.89	962.71	1318.5
20.47	420.67	585.72	717.21	962.71	1318.9
20.62	422.16	585.19	717.1	963.45	1319.2
20.78	421.63	584.34	713.84	963.03	1318.6
20.94	421.74	585.51	716.46	963.45	1317.9
21.09	422.48	584.44	717.1	962.92	1319.2
21.25	421.74	584.44	716.78	964.84	1318.3
21.41	420.99	584.02	717.85	964.84	1318.6
21.57	421.42	584.34	717.1	963.77	1319.9
21.72	421.42	584.76	717.53	963.24	1319.1
21.88	420.99	584.34	716.89	964.31	1318.3
22.04	420.99	583.27	716.46	964.52	1321

Table C13 Time sweep results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
22.19	421.63	584.44	717.21	964.73	1319.6
22.35	422.16	584.02	717.85	964.84	1321
22.51	421.95	584.34	718.28	965.48	1319.7
22.66	422.16	583.06	718.91	965.05	1321.8
22.82	422.16	582.95	719.32	964.52	1320.3
22.98	421.95	583.38	718.38	965.05	1322.8
23.14	421.31	583.59	720.28	964.41	1320.7
23.29	421.63	583.59	719.66	963.77	1320.9
23.45	421.95	583.59	717.96	963.77	1322.3
23.61	421.95	583.91	717.53	965.37	1323.5
23.76	422.38	582.85	718.6	964.84	1322.9
23.92	421.95	584.66	719.98	965.37	1321
24.08	422.38	584.02	717.21	964.84	1322
24.23	421.95	584.66	717.96	965.05	1322.1
24.39	421.95	584.12	717.64	966.44	1322.3
24.55	422.06	585.08	718.17	965.48	1322.4
24.71	422.38	584.02	719.34	965.8	1322.8
24.86	422.7	585.08	719.23	965.59	1322.3
25.02	422.48	583.91	718.28	966.22	1322.8
25.18	421.74	584.12	718.6	966.97	1322.8
25.33	420.89	584.12	719.98	967.29	1323.8
25.49	422.48	582.95	718.38	966.86	1323.4
25.65	421.1	584.44	718.28	967.5	1323.5
25.80	423.02	583.91	719.45	966.86	1324.6
25.96	421.42	584.02	717.32	967.93	1323.9
26.12	421.1	583.91	719.45	966.54	1324.1
26.27	421.63	584.12	718.28	967.61	1323.9
26.43	421.31	584.34	719.02	967.5	1323.4
26.59	421.63	584.12	720.3	966.86	1322.8
26.75	421.63	585.4	718.91	968.04	1323.6
26.90	422.8	584.76	718.6	967.93	1325.5
27.06	422.16	584.12	718.38	967.93	1325.2
27.22	422.06	585.08	719.23	966.65	1324.6
27.37	422.06	584.34	719.45	968.04	1326.8
27.53	421.42	585.4	718.38	967.93	1327.7
27.69	422.16	584.44	718.7	968.04	1324.6
27.84	421.95	585.19	718.91	968.25	1324.9
28.00	422.8	585.4	718.91	967.29	1326.6
28.16	423.12	586.15	718.6	970.74	1325.6

Table C13 Time sweep results of FA /NR compounds with various FA loadings in semi-EV systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
28.32	423.02	585.72	721.24	969	1325.7
28.47	422.06	586.15	718.38	968.68	1326.6
28.63	422.48	584.76	718.91	968.57	1326.1
28.79	421.63	584.76	719.45	969.32	1325.6
28.94	421.74	585.08	718.91	968.68	1326.4
29.10	421.1	584.66	719.34	969.42	1325.9
29.26	421.63	584.12	719.77	969	1327.7
29.41	422.48	584.34	718.28	968.36	1325.5
29.57	423.44	584.34	719.23	969.42	1328.1
29.73	422.7	584.66	719.34	969.1	1326.6
29.88	422.7	584.44	719.23	971.87	1327.7

Table C14 Time sweep results of FA /NR compounds with various FA loadings in peroxide systems

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.22	192.01	250.8	326.17	384.67	471.07
0.38	189.13	251.49	324.35	385.1	471.75
0.53	187.63	250.42	323.39	385.31	471.64
0.69	185.28	251.28	322.54	383.71	470.36
0.85	183.47	249.55	322.54	383.39	470.57
1.00	182.14	249.12	321.58	384.35	469.05
1.16	180.65	249.44	321.9	383.61	470.33
1.32	180.75	248.48	322.32	382.65	470.33
1.47	180.12	248.27	322.22	383.18	470.01
1.63	179.8	248.27	322	380.71	470.01
1.79	179.26	247.73	321.79	382.97	469.58
1.95	178.62	249.12	319.52	382.86	469.58
2.10	177.56	248.69	322	381.8	468.62
2.26	176.81	248.48	323.12	383.42	469.69
2.42	176.6	247.95	322.48	381.16	469.69
2.57	175.75	248.05	324.19	381.05	469.58
2.73	175.75	248.37	323.76	380.84	469.05
2.89	176.07	249.36	323.6	381.05	468.83
3.04	175.53	248.05	324.83	381.26	468.62
3.20	175.32	250.32	323.39	381.05	468.51

Table C14 Time sweep results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
3.36	175.75	248.93	322.54	380.09	469.05
3.51	173.72	246.88	322.54	380.09	468.94
3.67	173.3	247.95	322.54	381.29	469.93
3.83	173.62	249.01	323.07	380.52	468.2
3.99	172.66	249.01	323.92	381.26	468.62
4.14	172.23	250.32	323.39	381.05	468.83
4.30	171.91	250.21	323.39	380.84	468.3
4.46	172.87	249.33	322.86	381.16	469.18
4.61	173.19	249.68	324.19	381.48	468.94
4.77	173.3	247.73	324.03	380.84	467.45
4.93	171.8	248.69	323.28	381.05	468.83
5.08	170.84	248.37	324.03	380.52	470.01
5.24	171.91	249.89	323.28	380.84	469.69
5.40	171.27	247.63	324.19	380.73	468.94
5.56	172.12	248.27	323.71	381.05	467.98
5.71	172.34	249.01	323.28	381.48	467.88
5.87	170.84	247.63	324.19	380.73	468.2
6.03	169.99	248.8	323.71	380.2	468.3
6.18	170.42	248.27	323.39	381.05	469.05
6.34	170.21	249.01	323.39	381.05	469.37
6.50	169.99	250.64	324.67	381.48	469.37
6.65	170.21	249.86	325.2	381.48	468.62
6.81	169.46	248.69	324.67	381.26	469.05
6.97	169.46	249.86	324.99	381.58	469.26
7.13	169.14	248.69	325.09	380.2	468.51
7.28	169.35	248.27	324.35	379.35	469.58
7.44	169.46	248.48	323.92	381.26	468.94
7.60	170.42	248.37	324.35	380.84	469.9
7.75	169.03	249.12	324.93	381.16	470.01
7.91	169.99	248.05	324.35	381.05	469.37
8.07	167.54	247.95	325.09	380.2	468.2
8.22	169.99	247.2	323.92	381.48	469.69
8.38	169.14	247.2	325.73	380.73	469.9
8.54	168.07	247.41	325.41	380.41	467.98
8.69	169.14	247.95	325.09	381.8	468.51
8.85	167.97	247.41	324.67	380.2	470.01
9.01	169.03	246.99	326.27	380.73	469.58
9.17	167.33	248.05	325.73	380.84	469.37
9.32	167.01	250.42	325.09	381.9	470.43

Table C14 Time sweep results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
9.48	169.25	248.8	325.73	381.16	469.26
9.64	167.54	248.8	325.41	380.41	469.58
9.79	167.22	249.44	326.16	380.52	469.26
9.95	167.01	249.01	326.27	380.52	468.83
10.11	168.07	250.74	326.05	380.84	469.58
10.26	168.39	249.12	326.27	381.9	469.69
10.42	167.54	248.37	326.05	380.09	469.37
10.58	167.97	248.05	326.05	381.26	468.94
10.74	166.16	248.69	326.59	380.84	469.05
10.89	165.84	250	326.27	381.16	469.58
11.05	167.33	249.44	326.48	381.48	470.33
11.21	166.58	249.55	325.52	381.26	469.9
11.36	166.16	248.37	326.16	380.73	469.58
11.52	166.8	249.55	326.59	380.84	469.58
11.68	165.2	249.76	326.27	380.41	469.9
11.83	167.22	248.48	324.83	380.73	471.39
11.99	164.45	248.8	326.8	380.84	469.37
12.15	166.9	249.12	326.59	380.41	470.11
12.30	164.77	249.76	326.91	381.05	469.69
12.46	165.2	248.69	327.23	381.9	469.69
12.62	166.58	247.4	326.91	381.8	470.65
12.78	165.2	250.18	326.8	381.16	469.9
12.93	164.88	249.12	327.54	380.2	470.43
13.09	165.94	250.96	326.8	380.41	472.39
13.25	166.16	248.8	326.91	381.9	470.75
13.40	164.45	248.8	326.8	380.84	469.69
13.56	164.66	248.48	327.65	380.73	470.01
13.72	163.81	249.55	327.23	379.98	469.26
13.87	164.13	248.27	327.65	380.84	469.37
14.03	164.13	248.69	327.33	380.84	469.05
14.19	164.45	249.55	327.82	381.16	470.65
14.35	164.35	248.8	326.8	381.16	469.9
14.50	164.35	249.01	327.54	381.58	470.43
14.66	165.52	248.05	328.18	380.52	469.26
14.82	165.41	248.27	328.18	380.84	470.65
14.97	163.81	248.05	327.54	382.12	470.65
15.13	164.66	248.27	328.18	381.9	471.18
15.29	165.41	249.44	328.4	381.26	469.37
15.44	163.39	249.25	328.61	380.84	469.69

Table C14 Time sweep results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
15.60	163.81	248.69	328.18	381.16	470.97
15.76	165.09	249.55	328.4	380.52	470.97
15.92	164.66	249.55	327.65	381.16	471.07
16.07	162.96	248.69	328.4	381.48	471.39
16.23	163.28	249.01	327.97	381.48	470.33
16.39	164.66	247.95	328.18	381.48	470.65
16.54	164.35	249.01	328.18	382.36	470.43
16.70	164.45	249.33	328.4	380.2	470.43
16.86	163.81	248.69	328.29	381.48	469.58
17.01	163.6	248.69	329.46	380.84	469.69
17.17	162.96	248.05	327.86	381.05	471.64
17.33	163.6	248.27	328.18	381.16	470.33
17.48	164.03	248.27	327.54	381.05	470.33
17.64	164.13	248.8	328.61	380.09	470.65
17.80	164.03	250.08	328.61	379.98	470.65
17.96	162.96	248.69	329.68	381.16	469.9
18.11	163.6	247.95	328.72	381.05	470.11
18.27	163.28	249.44	329.36	380.2	471
18.43	163.28	248.8	328.61	381.48	469.9
18.58	162.53	247.41	329.78	381.16	470.01
18.74	162.75	249.01	329.25	381.05	470.33
18.90	163.28	247.73	329.04	381.05	471.18
19.05	162.32	249.12	329.78	381.48	471.5
19.21	161.68	248.57	330	380.84	471.5
19.37	162.32	248.05	330.32	381.58	471.18
19.53	161.89	248.05	329.46	381.05	471.5
19.68	162	247.61	328.29	380.52	470.01
19.84	162.21	250.08	329.68	381.05	470.43
20.00	161.89	249.12	329.78	380.73	471.18
20.15	161.68	247.95	329.68	381.48	471.18
20.31	162	250.21	329.78	381.16	471.82
20.47	161.68	250.08	330.53	380.52	472.24
20.62	161.68	249.76	330.32	381.16	471.39
20.78	161.47	248.69	330.74	381.8	471.43
20.94	161.15	248.8	330.42	381.48	470.43
21.09	161.26	248.48	331.06	380.09	471.82
21.25	162.21	249.44	330.32	379.35	471.18
21.41	162.53	250.4	330.32	381.26	472.14
21.57	161.89	248.37	330.42	380.84	471.18

Table C14 Time sweep results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
21.72	162.21	249.55	329.68	381.05	472.82
21.88	161.15	251.81	331.24	379.77	471.5
22.04	160.94	249.33	330.32	381.16	471.5
22.19	161.99	249.86	330.53	381.26	471.71
22.35	162.21	250.61	330	380.41	471.82
22.51	160.19	249.76	331.06	380.2	471.18
22.66	160.51	249.44	330.74	381.16	471.82
22.82	161.47	251.06	330.1	381.48	471.71
22.98	161.47	250.18	331.49	382.22	470.97
23.14	161.15	250.5	331.59	379.45	471.5
23.29	159.76	250.93	331.17	381.05	470.97
23.45	160.62	250.18	331.17	380.73	470.97
23.61	160.83	249.33	329.78	381.16	470.72
23.76	160.83	249.44	331.81	380.84	472.14
23.92	161.67	248.88	330	380.73	471.5
24.08	160.08	250.4	332.84	380.2	471.39
24.23	160.4	250.61	332.13	381.05	472.24
24.39	160.4	250.61	331.38	380.84	471.5
24.55	160.62	249.86	330.74	380.52	471.5
24.71	160.83	250.08	331.17	380.73	471.71
24.86	160.51	249.63	331.06	380.09	471.82
25.02	160.83	249.55	330.85	380.73	471.39
25.18	160.94	251.06	331.81	380.73	471.5
25.33	160.19	250.4	331.91	381.05	471.18
25.49	158.8	249.55	331.59	381.05	470.75
25.65	159.44	249.44	332.23	381.58	470.97
25.80	159.44	250.82	332.55	381.26	470.11
25.96	159.76	250.08	332.13	380.73	471.07
26.12	160.19	250.93	333.8	380.84	470.97
26.27	159.55	250.4	332.13	381.26	471.07
26.43	159.34	249.86	331.91	381.48	471.96
26.59	160.4	251.14	332.45	381.26	471.18
26.75	158.38	250.82	332.98	381.58	471.07
26.90	160.19	250.4	332.55	380.73	470.43
27.06	158.06	251.46	332.98	380.84	469.05
27.22	158.7	250.61	332.87	381.16	470.01
27.37	159.02	250.5	332.98	380.2	470.33
27.53	159.12	251.46	331.91	380.52	471.18
27.69	159.02	251.25	331.59	380.2	471.18

Table C14 Time sweep results of FA /NR compounds with various FA loadings in peroxide systems (cont.)

Time (min)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
27.84	158.38	250.5	332.13	380.41	470.11
28.00	159.12	250.82	333.62	380.41	469.9
28.16	159.76	250.82	333.19	381.26	471.18
28.32	158.48	251.14	332.55	381.48	471.71
28.47	158.7	250.5	333.3	381.58	470.65
28.63	158.7	251.68	332.55	381.9	471.82
28.79	159.76	249.86	333.72	381.48	470.33
28.94	159.34	251.14	333.51	381.48	471.07
29.10	157.95	250.08	332.13	381.26	470.43
29.26	157.95	251.14	333.19	381.16	472.14
29.41	157.63	251.25	333.19	381.05	471.18
29.57	158.7	250.5	333.62	381.05	471.5
29.73	158.06	252.74	333.62	381.58	471.71
29.88	157.95	251.46	334.36	381.48	470.75
27.84	158.38	250.5	332.13	380.41	470.11
28.00	159.12	250.82	333.62	380.41	469.9
28.16	159.76	250.82	333.19	381.26	471.18
28.32	158.48	251.14	332.55	381.48	471.71
28.47	158.7	250.5	333.3	381.58	470.65
28.63	158.7	251.68	332.55	381.9	471.82
28.79	159.76	249.86	333.72	381.48	470.33
28.94	159.34	251.14	333.51	381.48	471.07
29.10	157.95	250.08	332.13	381.26	470.43
29.26	157.95	251.14	333.19	381.16	472.14
29.41	157.63	251.25	333.19	381.05	471.18
29.57	158.7	250.5	333.62	381.05	471.5
29.73	158.06	252.74	333.62	381.58	471.71
29.88	157.95	251.46	334.36	381.48	470.75

Table C15 Strain sweep results of uncured FA /NR compounds with various FA loadings in CV systems

Strain (%)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.56	31.195	43.157	60.399	74.544	92.199
0.98	44.351	58.765	53.221	65.418	97.271
1.95	46.784	47.677	54.489	60.544	86.412
5.02	44.841	52.998	44.877	57.965	76.078
10.04	43.367	50.79	43.534	53.458	66.809
19.95	41.75	47.689	38.52	45.871	56.043
49.94	37.421	38.904	28.882	32.584	34.903
100.02	29.327	27.279	19.366	21.347	22.028
499.97	6.1931	5.8812	4.6687	5.1852	5.8106
999.94	2.3855	2.5147	2.1305	2.4399	2.7569
1199.98	1.9024	2.0581	1.7547	2.0366	2.2858

Table C16 Strain sweep results of uncured FA /NR compounds with various FA loadings in EV systems

Strain (%)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.56	40.266	49.964	62.612	72.505	88.702
0.98	46.568	49.937	56.505	67.614	82.702
1.95	37.822	43.939	51.516	65.129	75.404
5.02	38.998	41.716	47.743	62.615	70.538
10.04	38.465	41.424	46.544	52.172	62.508
19.95	37.29	38.324	44.18	48.218	51.664
50.02	32.386	33.663	39.757	33.343	33.718
100.12	25.466	24.393	26.309	21.79	20.952
499.98	5.4837	5.277	5.8811	5.4519	5.6248
999.98	2.1516	2.2372	2.6801	2.6278	2.7921
1199.95	1.6232	1.7842	2.1204	2.0803	2.1799

Table C17 Strain sweep results of uncured FA /NR compounds with various FA loadings in semi-EV systems

Strain (%)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.56	33.348	43.093	53.157	64.425	78.467
0.98	36.589	40.438	49.895	59.853	78.755
1.95	41.579	41.83	46.014	55.558	73.203
5.02	40.817	35.688	44.664	50.579	63.932
10.04	39.851	34.262	42.562	48.842	55.727
19.95	38.815	28.689	38.172	45.016	45.604
49.94	34.783	19.724	28.812	36.556	39.522
100.02	27.058	12.991	19.676	26.366	18.508
499.97	5.7594	3.6527	4.6514	5.6123	5.1019
999.94	2.2222	1.8336	2.1496	2.3908	2.4997
1199.98	1.7361	1.5378	1.7734	1.9238	2.0599

Table C18 Strain sweep results of uncured FA /NR compounds with various FA loadings in peroxide systems

Strain (%)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.56	56.889	69.429	70.961	91.62	94.16
0.98	56.254	60.372	68.697	100.88	99.789
1.95	53.939	59.537	62.646	92.028	90.816
5.02	54.28	58.391	60.433	84.649	85.694
10.04	55.65	55.747	56.686	78.53	76.185
19.95	55.786	53.142	51.378	63.433	63.149
49.94	50.598	44.37	38.797	43.056	41.422
100.02	39.559	31.962	26.053	27.297	25.623
499.97	7.8599	6.8566	5.8256	6.3806	6.4873
999.94	3.3931	3.0013	2.662	2.962	3.0366
1199.98	2.6948	2.4281	2.244	2.429	2.4448

Table C19 Strain sweep results of cured FA /NR compounds with various FA loadings in CV systems

Strain (%)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.56	686.91	970.83	1222.2	1481.5	1878.2
0.84	616.29	842.97	1127.7	1342.4	1778
1.95	561.55	682.47	938.33	1137.5	1531.6
5.02	495	706.9	944.52	1086.5	1509.8
10.04	498.35	723.02	933.83	1036	1383.1
19.95	491.58	691.75	845.51	909.74	1157
49.94	473.43	614.37	706.86	715.86	812.73
100.02	441.39	516.53	553.31	509.22	543.43
200.04	384.07	413.03	394.18	362.93	385.46

Table C20 Strain sweep results of cured FA /NR compounds with various FA loadings in EV systems

Strain (%)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.56	559.23	771.26	996.96	1249.6	1357.4
0.84	544.06	652.54	810.14	1034.8	1065.6
1.95	432.63	559.63	684.98	868.74	905.74
5.02	392.47	512.63	695.97	870.66	907.9
10.04	395.6	521.97	691.95	815.91	873.59
19.95	348.63	509.11	652.59	710.15	786.87
49.94	377.76	464.9	549.09	486.8	624.49
100.02	349.37	350.14	432.67	333.83	442.95
200.04	294.05	312.73	320.44	312.53	311.18

Table C21 Strain sweep results of cured FA /NR compounds with various FA loadings in semi-EV systems

Strain (%)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.56	809	1011	1232.6	1310.2	1540.5
0.84	721.42	874.82	1048.06	1172.4	1475.9
1.95	572.11	702.36	899.05	1053.7	1336.5
5.02	503.4	584.74	800.9	920.54	1173.5
10.04	440.09	548.69	713.91	847.26	1115.6
19.95	431.54	533.87	658.04	800.02	962.18
49.94	411.03	478.9	550.4	636.03	684.07
100.02	377	409.44	430.84	436.04	445.15
200.04	309.71	316.53	315.88	309.69	318.42

Table C22 Strain sweep results of cured FA /NR compounds with various FA loadings in peroxide systems

Strain (%)	Storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
0.56	90.63	110.63	124.78	155.65	167.67
0.84	88.801	109.81	119.01	148.65	159.33
1.95	77.465	98.72	114.78	145.09	157.48
5.02	71.919	94.76	112.04	140.5	151.62
10.04	73.526	90.92	110.92	135.5	145.87
19.95	72.445	90.1	104.96	132.18	137.31
49.94	65.295	77.087	80.385	90.889	92.468
100.02	52.439	55.242	55.395	58.728	60.748
200.04	33.72	31.544	33.542	35.545	36.482

Table C23 Delta storage modulus of FA /NR compounds with various FA loadings in sulfur and peroxide systems (Payne effect)

Curing systems	Delta G'				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	29.2926	41.0989	58.6443	72.5074	89.9132
EV	38.6428	48.1798	60.4916	70.4247	86.5221
Semi-EV	31.6119	41.5552	51.3836	62.5012	76.4071
DCP	54.1942	67.0009	68.717	89.191	91.7152

Table C24 Relative storage modulus (relative G') at 1 % strain of FA /NR compounds with various FA loadings in sulfur and peroxide systems

Curing systems	Relative G'				
	0 phr	50 phr	100 phr	150 phr	200 phr
Guth-Gold Equation					
	1.00	1.74	2.71	3.70	4.63
CV	1.00	1.37	1.83	2.18	2.89
EV	1.00	1.20	1.49	1.90	1.96
Semi-EV	1.00	1.21	1.45	1.63	2.05
DCP	1.00	1.24	1.34	1.67	1.79

Table C25 Frequency sweep results of uncured FA /NR compounds with various FA loadings in CV systems

Frequency rad/s	Storage modulus (kPa)					Damping factor					Complex viscosity (Pa-s)				
	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr
0.5	31.198	35.828	41.075	37.709	51.467	1.08	1.27	1.298	1.461	1.456	91371	1.04E+05	1.34E+05	1.33E+05	1.81E+05
0.99	42.486	50.595	58.304	59.98	76.968	0.873	1.003	1.075	1.26	1.295	56696	72032	90408	96994	1.27E+05
2	61.733	77.598	92.579	94.977	120.8	0.748	0.841	0.94	1.047	1.092	38545	50693	63522	68750	89439
5.01	90.57	117.46	145.75	161.36	201.74	0.589	0.678	0.761	0.801	0.868	20997	28346	36596	41297	53359
10	115.6	155.78	198.2	218.94	277.08	0.487	0.557	0.619	0.679	0.714	12859	17833	23313	26456	34042
15	130.89	177.69	229.82	259.63	321.84	0.454	0.5	0.565	0.613	0.65	9586.5	13250	17605	20307	25599
20	134.92	195.33	258.34	287.16	352.02	0.414	0.441	0.49	0.573	0.604	7299.7	10674	14381	16550	20561
25	149.02	206.06	269.84	306.17	382.53	0.389	0.45	0.477	0.534	0.572	6395.9	9040.1	11959	13887	17632
30	158.22	216.18	284.82	323.04	404.49	0.368	0.43	0.454	0.509	0.544	5619.9	7844.7	10426	12083	15347
40	168.34	233.2	305.25	349.56	441.21	0.354	0.396	0.444	0.486	0.505	4464.5	6270.8	8349.1	9717.3	12357
50	177.59	248.37	321.81	371.79	468.84	0.339	0.377	0.416	0.457	0.478	3750.2	5307.6	6970.2	8175.9	10391
60	184.29	260.86	336.53	387.43	485.7	0.325	0.356	0.398	0.437	0.464	3229.6	4614.1	6035.7	7046	8923.6
69.99	190.73	266.09	348.18	400.31	506.31	0.313	0.351	0.382	0.426	0.444	2855	4029.3	5325.4	6216.2	7913.4
80	194.78	273.28	356.67	411.2	530.44	0.306	0.339	0.372	0.408	0.429	2546.6	3607.4	4757.7	5551.4	7215.7
90	200.08	280.57	364.01	420.24	541.4	0.29	0.329	0.363	0.398	0.418	2314.6	3281.9	4302.6	5025.2	6520.7
100	202.43	287.89	372.89	431.99	551.33	0.293	0.318	0.353	0.382	0.41	2109.2	3020.6	3954.9	4623.9	5959.2

Table C26 Frequency sweep results of uncured FA /NR compounds with various FA loadings in EV systems

Frequency rad/s	Storage modulus (kPa)				Damping factor				Complex viscosity (Pa-s)						
	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr
0.5	27.749	31.419	31.312	39.882	36.623	1.068	1.155	1.389	1.42	1.589	80761	1.00E+05	1.07E+05	1.38E+05	1.37E+05
0.99	41.94	46.796	49.821	62.132	57.687	0.857	1.01	1.199	1.226	1.436	55524	66852	74414	98827	1.01E+05
2	59.693	69.873	77.739	97.877	93.506	0.705	0.845	1.016	1.027	1.212	36522	45746	55404	70141	73442
5.01	87.368	108.74	128.18	160.91	163.39	0.563	0.688	0.811	0.805	0.94	20031	26368	32970	41259	44804
10	109.45	144.7	175.9	219.18	231.06	0.485	0.564	0.667	0.666	0.77	12164	16610	21145	26329	29161
15	123.27	164.53	206.01	251.64	271.92	0.454	0.513	0.617	0.612	0.699	9027.5	12329	16146	19671	22128
20	131.24	177.54	228.14	284.82	306.48	0.471	0.465	0.568	0.537	0.623	7252.3	9790.8	13117	16165	18056
25	142.1	194.18	244.42	301.72	327.99	0.394	0.445	0.52	0.519	0.61	6109.2	8501.6	11020	13601	15372
30	148.26	203.15	258.19	317.67	347.19	0.389	0.423	0.493	0.492	0.581	5302	7350.9	9594	11802	13386
40	158.68	219.24	281.9	344.89	377.93	0.366	0.405	0.461	0.467	0.539	4224.4	5912.9	7759.9	9514.7	10731
50	168.37	230.28	299.58	363.97	404.19	0.341	0.388	0.438	0.445	0.509	3557.9	4939.8	6539.6	7967.8	9068.3
60	174.32	240.55	311.69	378.85	423	0.327	0.374	0.421	0.422	0.486	3056.7	4280.3	5635.2	6852	7838.1
69.99	181.37	249.14	324.3	393.72	439.17	0.313	0.361	0.401	0.411	0.468	2715	3784.5	4992.3	6081.1	6927.3
80	185.67	257.42	331.11	403.07	449.53	0.313	0.345	0.396	0.399	0.454	2432	3404.5	4452.6	5425.7	6171.3
90	190.27	261.95	341.58	413.04	460.72	0.304	0.34	0.385	0.388	0.441	2209.6	3074.3	4067.5	4922.7	5594.2
100	194.25	269.84	348.95	423.85	473.16	0.297	0.331	0.371	0.376	0.427	2026.3	2842.7	3721.8	4528.7	5144.4

Table C27 Frequency sweep results of uncured FA /NR compounds with various FA loadings in semi-EV systems

Frequency rad/s	Storage modulus (kPa)					Damping factor					Complex viscosity (Pa-s)				
	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr
0.5	27.391	27.177	26.919	21.546	35.249	1.075	1.225	1.526	1.7	1.79	79994	85517	97719	87885	1.38E+05
0.99	41.325	42.861	43.695	35.702	56.995	0.881	1.079	1.267	1.385	1.576	55362	63377	70883	66976	97889
2	58.033	66.305	68.073	59.572	93.472	0.721	0.883	1.041	1.193	1.32	35778	44233	49116	49333	72750
5.01	86.437	103.44	115.76	112.35	162.11	0.587	0.704	0.808	0.943	0.985	20024	25271	29729	31502	44512
10	110.06	138.39	158.39	161.48	233.22	0.484	0.583	0.679	0.765	0.827	12226	16017	19143	20957	29364
15	121.82	157.61	184.75	194.95	273.82	0.462	0.534	0.608	0.697	0.741	8947.4	11915	14420	16178	22253
20	136.61	179.38	210.2	223.24	305.87	0.383	0.467	0.551	0.632	0.661	7312.9	9896.9	11997	13378	18093
25	140.59	187.35	220.01	238.1	330.91	0.396	0.457	0.532	0.61	0.628	6049.1	8239.6	9971.3	11247	15507
30	147.64	197.56	232.58	254.56	348.34	0.384	0.437	0.515	0.595	0.597	5271.8	7186.2	8721.5	9881	13511
40	158.37	213.85	255.58	281.49	378.54	0.359	0.412	0.471	0.56	0.554	4206.8	5782.4	7062	8044.1	10846
50	167.73	226.76	272.75	302.18	404.3	0.338	0.393	0.447	0.524	0.522	3541	4872.1	5974.5	6817.3	9128.4
60	173.29	236.87	284.29	317.67	422.08	0.321	0.377	0.43	0.497	0.5	3033	4218.4	5157.4	5917.9	7855.9
69.99	180.45	244.26	296.21	331.01	438.03	0.316	0.37	0.416	0.485	0.478	2703.8	3720.7	4583.7	5242.5	6954.6
80	184.59	251.75	304.18	340.98	452.3	0.314	0.355	0.405	0.463	0.468	2418.6	3339.5	4102.1	4705.5	6231
90	188.12	257.63	312.46	350.02	463.78	0.307	0.344	0.394	0.448	0.454	2186.8	3027.4	3731.7	4271	5646.7
100	193.57	264.32	320.13	357.54	477.97	0.294	0.334	0.384	0.433	0.441	2017.9	2786.9	3429.4	3908.1	5208.3

Table C28 Frequency sweep results of uncured FA /NR compounds with various FA loadings in peroxide systems

Frequency rad/s	Storage modulus (kPa)					Damping factor					Complex viscosity (Pa-s)				
	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr
0.5	41.159	43.717	47.165	58.055	55.245	0.855	1.051	1.11	1.205	1.334	1.08E+05	1.26E+05	1.33E+05	1.81E+05	1.83E+05
0.99	57.954	60.714	61.882	80.491	81.871	0.759	0.876	1.007	1.128	1.238	73143	81143	88296	1.22E+05	1.31E+05
2	80.499	88.512	91.343	121.24	125.09	0.573	0.727	0.848	0.924	1.048	46376	54710	59869	82514	90598
5.01	111.27	131.06	139.54	189.33	205.51	0.463	0.581	0.686	0.749	0.83	24496	30287	33803	47247	53367
10	132.85	165.48	182.1	250.45	277.7	0.39	0.491	0.571	0.62	0.691	14260	18435	20965	29460	33747
15	146.57	189.11	210.35	285.9	318.44	0.36	0.419	0.514	0.563	0.637	10388	13674	15771	21875	25183
20	152.86	208.93	226.29	317.98	353.8	0.31	0.392	0.497	0.526	0.597	8000.9	11219	12634	17963	20605
25	162.36	215.1	242.57	333	377.46	0.318	0.383	0.445	0.495	0.554	6816.3	9214.1	10623	14867	17265
30	168.34	224.14	253.13	348.95	396.78	0.313	0.373	0.431	0.476	0.533	5879.9	7973.1	9186	12880	14985
40	178	240.71	273.06	376.85	430	0.288	0.345	0.404	0.446	0.502	4630.2	6364.5	7361.2	10316	12026
50	187.2	252.93	288.97	398.47	454.43	0.277	0.33	0.383	0.422	0.478	3884.5	5326.9	6188.9	8649.6	10072
60	193.33	262.94	298.66	415.71	475.28	0.262	0.313	0.365	0.407	0.457	3330.5	4591.8	5298.5	7479.5	8708
69.99	198.02	271.06	307.86	428	491.29	0.26	0.307	0.357	0.393	0.441	2923.1	4051.2	4670.3	6570.9	7670.9
80	203.71	278.21	316.29	435.88	503.19	0.247	0.297	0.347	0.388	0.429	2622.8	3627.6	4185.2	5843.8	6844.6
90	204.98	283.33	324.46	445.23	515.87	0.25	0.291	0.342	0.377	0.418	2347.7	3278.9	3810.1	5286.6	6212.4
100	208.82	291.43	330.86	454.59	527.09	0.243	0.28	0.329	0.368	0.409	2149	3026.4	3483.6	4843.4	5694.1

Table C29 Slope calculated from the plots of storage modulus (G') as a function of frequency of uncured FA /NR compounds with various FA loadings in sulfur and peroxide systems

Curing systems	Slope				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	0.443	0.496	0.533	0.591	0.568
EV	0.455	0.511	0.576	0.571	0.62
Semi-EV	0.443	0.541	0.592	0.679	0.632
DCP	0.392	0.45	0.461	0.496	0.544

Table C30 Power law index of uncured FA /NR compounds with various FA loadings in sulfur and peroxide systems

Curing systems	Power law index				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	0.292	0.32	0.328	0.342	0.367
EV	0.287	0.319	0.356	0.353	0.345
Semi-EV	0.292	0.336	0.356	0.399	0.371
DCP	0.25	0.289	0.305	0.309	0.332

Table C31 Frequency sweep results of cured FA /NR compounds with various FA loadings in CV systems

Frequency rad/s	Storage modulus (kPa)					Damping factor				
	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr
0.5	500.37	728.76	932.66	1059.7	1436.3	0.01	0.001	0.025	0.045	0.087
0.99	508.45	735.83	932.29	1060.7	1435.7	0.005	0.003	0.028	0.052	0.092
2	508.96	734.17	932.01	1057.7	1447.5	0.002	0.004	0.029	0.054	0.093
5.01	507.88	730.49	932.5	1070.2	1458.8	0.002	0.003	0.032	0.056	0.096
10	506.71	727.34	932.32	1076.6	1472	0.002	0.002	0.032	0.056	0.093
15	509.01	726.11	931.71	1073.7	1471.5	0.006	0.004	0.038	0.055	0.092
20	511.62	723.04	924.96	1077.2	1478.4	0.001	0.003	0.036	0.062	0.095
25	506.25	723.96	931.86	1080.6	1483.4	0.004	0.005	0.035	0.057	0.093
30	505.49	723.2	933.24	1083	1488.1	0.002	0.006	0.035	0.061	0.095
40	507.02	725.65	938.15	1087	1494.7	0.007	0.008	0.036	0.062	0.096
50	509.32	726.88	944.95	1095	1509.2	0.011	0.01	0.034	0.06	0.094
60	507.79	727.64	944.13	1096.2	1507	0.009	0.012	0.038	0.064	0.097
69.99	510.19	726.11	945.51	1098.2	1514.3	0.011	0.012	0.037	0.063	0.098
80	509.93	727.18	944.89	1103	1516.9	0.014	0.014	0.04	0.063	0.097
90	510.96	724.88	949.56	1103.3	1522	0.012	0.015	0.04	0.065	0.096
100	511.31	726.03	947.65	1106.6	1531	0.016	0.02	0.042	0.066	0.098

Table C32 Frequency sweep results of cured FA /NR compounds with various FA loadings in EV systems

Frequency rad/s	Storage modulus (kPa)					Damping factor				
	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr
0.5	391.34	515.69	697.05	811.74	950.64	0.008	0.003	0.005	0.028	0.058
0.99	400.46	518.25	701.86	826.41	955.36	0.003	0.001	0.009	0.03	0.065
2	398.47	518.91	698.97	836.64	954.42	0.002	0.002	0.012	0.03	0.065
5.01	396.17	513	701.45	848.78	956.23	0.004	0.007	0.017	0.033	0.072
10	397.83	514.99	700.51	863.79	959.17	0.005	0.009	0.019	0.031	0.074
15	396.94	511.16	702.5	870.99	955.34	0.005	0.01	0.022	0.034	0.078
20	401.69	519.59	695.91	879.58	953.96	0.008	0.011	0.023	0.034	0.076
25	397.22	514.07	701.43	881.57	960.86	0.011	0.012	0.024	0.037	0.078
30	399.7	516.07	702.19	886.33	963.45	0.009	0.011	0.022	0.036	0.08
40	401.84	517.56	705.26	894.76	966.99	0.011	0.014	0.023	0.041	0.086
50	402.92	518.52	709.09	905.03	970.52	0.014	0.012	0.027	0.043	0.09
60	403.88	517.6	711.54	913.62	975.28	0.011	0.011	0.024	0.043	0.089
69.99	402.3	518.06	711.39	919.6	974.04	0.013	0.014	0.023	0.047	0.086
80	404.14	518.83	712.62	922.36	976.19	0.014	0.012	0.024	0.049	0.096
90	404.14	517.94	715.38	930.97	979.89	0.016	0.011	0.025	0.05	0.098
100	405.37	519.75	715.07	934.04	979.71	0.018	0.014	0.028	0.05	0.098

Table C33 Frequency sweep results of cured FA /NR compounds with various FA loadings in semi-EV systems

Frequency rad/s	Storage modulus (kPa)					Damping factor				
	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr
0.5	439.79	608.71	717.08	908.33	1166.6	0.006	0.007	0.012	0.036	0.072
0.99	440.98	607.16	716.55	917.76	1182.4	0.007	0.008	0.015	0.039	0.074
2	437.5	605.31	710.69	911.36	1186.3	0.005	0.009	0.017	0.039	0.071
5.01	436.03	602.06	713.28	918.37	1204.3	0.004	0.008	0.021	0.043	0.071
10	436.34	601.31	712.46	922.97	1218.3	0.005	0.01	0.022	0.042	0.069
15	435.27	597.94	711.19	928.8	1225.8	0.008	0.011	0.021	0.041	0.066
20	437.57	601.31	707.87	923.89	1232.5	0.008	0.013	0.025	0.042	0.074
25	434.65	599.14	711.65	931.86	1236.3	0.007	0.014	0.027	0.039	0.07
30	434.5	599.47	714	936.31	1237.7	0.007	0.015	0.026	0.043	0.073
40	434.96	599.93	717.16	939.99	1246	0.01	0.015	0.035	0.044	0.073
50	437.11	598.73	721.82	948.17	1251	0.011	0.016	0.035	0.047	0.076
60	435.73	601.77	726.03	948.33	1261.3	0.01	0.017	0.035	0.048	0.076
69.99	436.65	604.51	722.36	949.03	1262.4	0.015	0.018	0.038	0.052	0.08
80	435.57	601.31	725.8	951.03	1266.1	0.014	0.018	0.039	0.053	0.079
90	435.73	600.85	728.34	954.71	1269	0.016	0.018	0.038	0.054	0.08
100	437.22	602.38	723.36	956.55	1275.4	0.019	0.023	0.045	0.055	0.083

Table C34 Frequency sweep results of cured FA /NR compounds with various FA loadings in peroxide systems

Frequency rad/s	Storage modulus (kPa)					Damping factor				
	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr
0.5	62.444	98.493	100.03	134.83	138.7	0.321	0.358	0.428	0.458	0.505
0.99	73.592	110.95	121.43	161.26	166.35	0.345	0.367	0.443	0.487	0.51
2	86.471	129.19	145.5	192.72	201.06	0.348	0.362	0.447	0.487	0.506
5.01	106	157.77	184.41	248.87	260.24	0.346	0.351	0.43	0.463	0.492
10	124.34	182.51	220.3	297.74	314.61	0.343	0.352	0.405	0.444	0.469
15	134.15	203.25	243.34	331.16	346.8	0.342	0.349	0.39	0.416	0.463
20	138.91	214.18	258.65	355.39	369.19	0.349	0.342	0.403	0.401	0.451
25	152.09	220.79	274.53	371.62	396.97	0.317	0.312	0.37	0.408	0.428
30	158.39	229.82	285.52	387.44	413.65	0.306	0.308	0.363	0.402	0.421
40	167.45	243.62	304.95	412.42	442.32	0.303	0.303	0.349	0.386	0.407
50	175.39	255.48	319.67	435.27	469.17	0.295	0.3	0.337	0.374	0.391
60	182.66	263.09	332.39	452.29	486.99	0.283	0.289	0.329	0.365	0.384
69.99	186.65	271.46	341.13	467.16	502.57	0.285	0.289	0.323	0.358	0.377
80	191.34	276.99	350.48	478.81	516.07	0.272	0.28	0.319	0.352	0.372
90	196.03	281.8	357.95	490	529.56	0.27	0.281	0.312	0.344	0.364
100	200.94	289.46	367.47	496.9	540.29	0.266	0.279	0.312	0.347	0.358

Table C35 Retention of storage modulus (G') of FA filled NR vulcanizates with various FA loadings in sulfur and peroxide systems as measured from RPA 2000

Curing systems	Unaged storage modulus (kPa)					Aged storage modulus (kPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr	0 phr	50 phr	100 phr	150 phr	200 phr
CV	485.8	708.36	918.8	1036.3	1346.8	522.49	735.19	984.15	1105.7	1477.5
EV	392.5	512.75	681.65	853.58	837.1	423.54	549.19	726.00	925.65	934.47
Semi-EV	442.4	608.54	732.54	913.81	1136.3	463.93	636.12	755.24	953.91	1237
DCP	75.024	109.63	121.79	149.01	185.5	86.827	131.27	145.87	186.42	236.34
Retention storage modulus										
Curing systems	0 phr		50 phr		100 phr		150 phr		200 phr	
CV	107.55		103.79		107.11		106.70		109.70	
EV	107.91		107.11		106.51		108.44		111.63	
Semi-EV	104.87		104.53		103.10		104.39		108.86	
DCP	115.73		119.74		119.77		125.11		127.41	

Table C36 Modulus at 100 % strain (M100) of FA /NR composites with various FA loadings in sulfur and peroxide systems

Curing systems	M100(MPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	0.90±0.06	0.96±0.02	1.04±0.06	1.03±0.03	1.02±0.04
EV	0.75±0.06	0.84±0.01	0.91±0.02	0.88±0.05	0.80±0.02
Semi-EV	0.82±0.06	0.86±0.04	0.89±0.01	0.97±0.03	0.95±0.05
DCP	0.32±0.01	0.46±0.02	0.48±0.03	0.53±0.02	0.56±0.01

Table C37 Elongation at break of FA /NR composites with various FA loadings in sulfur and peroxide systems

Curing systems	Elongation at break (%)				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	650.76±53.8	591.59±12.70	561.31±38.53	499.62±12.27	490.97±48.19
EV	698.35±49.61	636.3±7.71	588.27±21.83	582.07±54.38	599.4±39.47
Semi-EV	652.03±38.2	546.35±45.39	610.95±13.46	514.21±42.54	500.98±66.04
DCP	612.15±96.87	537.82±29.91	585.63±25.34	581.17±57.03	537.35±40.0

Table C38 Tensile strength of FA /NR composites with various FA loadings in sulfur and peroxide systems

Curing systems	Tensile strength (MPa)				
	0 phr	50 phr	100 phr	150 phr	200 phr
CV	22.9±1.1	12.88±0.35	10.53±0.52	8.66±0.17	6.42±0.47
EV	21.1±1.60	14.22±0.29	9.82±0.39	7.51±0.71	6.22±0.12
Semi-EV	22.38±0.45	14.21±1.13	11.26±0.30	8.77±0.35	6.32±0.61
DCP	1.26±0.36	2.71±0.47	2.86±0.73	2.79±0.15	2.55±0.10

APPENDIX D

VISCOELASTIC AND MECHANICAL PROPERTIES DATA OF TREATED FA FILLED NR VULCANIZATES WITH VARIOUS CURING SYSTEMS

Table D1 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV system

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
0.243	2.123	2.558	1.421	1.756
0.393	1.832	2.222	1.205	1.451
0.54	1.742	2.066	1.123	1.329
0.69	1.815	2.017	1.166	1.261
0.84	2.148	2.119	1.364	1.261
0.99	2.773	2.41	1.886	1.292
1.14	3.86	2.975	3.319	1.366
1.29	5.551	4.032	5.832	1.504
1.44	7.715	5.758	8.507	1.775
1.59	9.935	7.945	10.88	2.429
1.74	11.9	10.14	12	3.981
1.89	13.55	12.19	13.9	6.552
2.04	14.96	14.07	15.59	9.241
2.19	16.18	15.78	17.09	11.63
2.34	17.25	17.33	18.4	13.74
2.49	18.19	18.73	19.57	15.57
2.64	19.03	19.96	20.59	17.19
2.79	19.75	21.05	21.48	18.61
2.94	20.4	22.05	22.28	19.88
3.09	20.98	22.92	22.99	21.01
3.24	21.5	23.72	23.63	22.01
3.39	21.96	24.45	24.19	22.91
3.54	22.38	25.09	24.68	23.73
3.69	22.76	25.69	25.12	24.45
3.84	23.09	26.2	25.5	25.11
3.99	23.39	26.71	25.85	25.69
4.14	23.67	27.13	26.13	26.22
4.29	23.93	27.52	26.39	26.69
4.44	24.14	27.87	26.6	27.12
4.59	24.35	28.18	26.79	27.49

Table D1 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
4.74	24.52	28.46	26.96	27.82
4.89	24.69	28.71	27.1	28.13
5.04	24.82	28.93	27.2	28.37
5.19	24.95	29.13	27.29	28.61
5.34	25.04	29.3	27.35	28.8
5.49	25.14	29.45	27.4	28.97
5.64	25.21	29.58	27.43	29.11
5.79	25.29	29.69	27.44	29.23
5.94	25.35	29.79	27.45	29.32
6.09	25.39	29.86	27.44	29.4
6.24	25.43	29.92	27.43	29.45
6.39	25.46	29.98	27.41	29.49
6.54	25.48	30.02	27.37	29.52
6.69	25.5	30.05	27.33	29.53
6.84	25.5	30.07	27.28	29.54
6.99	25.5	30.07	27.23	29.52
7.14	25.5	30.07	27.18	29.5
7.29	25.49	30.08	27.11	29.48
7.44	25.48	30.06	27.06	29.43
7.59	25.46	30.05	27	29.38
7.74	25.45	30.02	26.94	29.33
7.89	25.43	30.01	26.87	29.27
8.04	25.39	29.96	26.8	29.2
8.19	25.37	29.93	26.72	29.13
8.34	25.34	29.88	26.67	29.06
8.49	25.31	29.83	26.61	28.98
8.64	25.27	29.8	26.54	28.9
8.79	25.25	29.74	26.45	28.81
8.94	25.21	29.68	26.39	28.73
9.09	25.18	29.65	26.3	28.64
9.24	25.15	29.58	26.25	28.55
9.39	25.11	29.55	26.16	28.45
9.54	25.06	29.49	26.09	28.35
9.69	25.03	29.42	26.02	28.24
9.84	24.99	29.38	25.94	28.14
9.99	24.95	29.3	25.87	28.05
10.14	24.91	29.25	25.78	27.93
10.29	24.87	29.17	25.72	27.83
10.44	24.83	29.1	25.63	27.72

Table D1 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
10.59	24.79	29.04	25.56	27.62
10.74	24.75	28.99	25.48	27.5
10.89	24.7	28.91	25.41	27.4
11.04	24.65	28.85	25.34	27.28
11.34	24.57	28.72	25.18	27.05
11.49	24.53	28.65	25.11	26.94
11.64	24.49	28.59	25.03	26.83
11.79	24.44	28.52	24.94	26.71
11.94	24.41	28.44	24.89	26.6
12.09	24.36	28.38	24.8	26.48
12.24	24.32	28.31	24.72	26.37
12.39	24.29	28.24	24.65	26.26
12.54	24.24	28.17	24.57	26.14
12.69	24.2	28.1	24.52	26.02
12.84	24.15	28.04	24.44	25.91
12.99	24.11	27.96	24.36	25.8
13.14	24.08	27.89	24.29	25.68
13.29	24.04	27.81	24.21	25.57
13.44	24	27.75	24.15	25.46
13.59	23.96	27.67	24.07	25.35
13.74	23.91	27.6	24.01	25.26
13.89	23.86	27.53	23.92	25.15
14.04	23.83	27.46	23.86	25.04
14.19	23.78	27.4	23.79	24.94
14.34	23.75	27.33	23.72	24.84
14.49	23.7	27.26	23.65	24.74
14.64	23.67	27.19	23.58	24.65
14.79	23.63	27.12	23.51	24.55
14.94	23.59	27.06	23.45	24.46
15.09	23.55	26.98	23.39	24.37
15.24	23.5	26.93	23.31	24.27
15.39	23.47	26.86	23.24	24.18
15.54	23.43	26.78	23.18	24.09
15.69	23.39	26.72	23.11	24
15.84	23.34	26.65	23.04	23.92
15.99	23.31	26.6	22.97	23.82
16.14	23.27	26.53	22.92	23.75
16.29	23.22	26.45	22.84	23.66
16.44	23.19	26.38	22.78	23.57

Table D1 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
16.59	23.16	26.32	22.7	23.48
16.74	23.12	26.27	22.66	23.4
16.89	23.09	26.19	22.58	23.32
17.04	23.04	26.12	22.53	23.25
17.19	23	26.07	22.47	23.15
17.34	22.97	26	22.39	23.08
17.49	22.93	25.93	22.33	22.99
17.64	22.9	25.86	22.27	22.92
17.79	22.86	25.79	22.2	22.85
17.94	22.83	25.73	22.13	22.77
18.09	22.79	25.68	22.08	22.69
18.24	22.74	25.62	22.02	22.61
18.39	22.72	25.54	21.96	22.54
18.54	22.67	25.48	21.91	22.47
18.69	22.66	25.42	21.84	22.4
18.84	22.62	25.36	21.79	22.33
18.99	22.57	25.3	21.73	22.25
19.14	22.54	25.22	21.66	22.19
19.29	22.51	25.18	21.61	22.11
19.44	22.47	25.11	21.55	22.04
19.59	22.45	25.05	21.5	21.98
19.74	22.41	25.01	21.44	21.9
19.89	22.37	24.93	21.38	21.84
20.04	22.34	24.87	21.32	21.77
20.19	22.32	24.82	21.27	21.71
20.34	22.27	24.77	21.21	21.64
20.49	22.24	24.7	21.16	21.57
20.64	22.21	24.64	21.1	21.52
20.79	22.17	24.58	21.06	21.44
20.94	22.15	24.53	20.99	21.39
21.09	22.1	24.45	20.94	21.32
21.24	22.08	24.4	20.88	21.26
21.39	22.04	24.35	20.83	21.19
21.54	22	24.31	20.79	21.13
21.69	21.96	24.24	20.73	21.08
21.84	21.94	24.18	20.69	21.02
21.99	21.91	24.13	20.62	20.95
22.14	21.88	24.07	20.58	20.89
22.29	21.85	24.02	20.52	20.84

Table D1 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
22.44	21.81	23.97	20.49	20.78
22.59	21.79	23.91	20.43	20.72
22.74	21.76	23.86	20.39	20.67
22.89	21.74	23.81	20.34	20.61
23.04	21.7	23.75	20.29	20.55
23.19	21.67	23.71	20.25	20.5
23.34	21.65	23.65	20.19	20.44
23.49	21.62	23.6	20.14	20.39
23.64	21.59	23.54	20.1	20.32
23.79	21.56	23.49	20.05	20.28
23.94	21.52	23.44	20.01	20.22
24.09	21.5	23.39	19.95	20.17
24.24	21.46	23.35	19.9	20.13
24.39	21.44	23.3	19.86	20.06
24.54	21.41	23.24	19.83	20.01
24.69	21.39	23.2	19.77	19.96
24.84	21.36	23.14	19.73	19.91
24.99	21.32	23.09	19.68	19.86
25.14	21.28	23.04	19.64	19.81
25.29	21.27	23	19.6	19.75
25.44	21.24	22.94	19.56	19.7
25.59	21.2	22.91	19.51	19.65
25.74	21.18	22.86	19.47	19.61
25.89	21.14	22.81	19.42	19.56
26.04	21.13	22.77	19.37	19.51
26.19	21.09	22.73	19.34	19.47
26.34	21.07	22.67	19.29	19.43
26.49	21.05	22.63	19.25	19.38
26.64	21.02	22.59	19.21	19.34
26.79	21	22.54	19.17	19.3
26.94	20.97	22.5	19.13	19.24
27.09	20.95	22.46	19.09	19.2
27.24	20.92	22.41	19.05	19.16
27.39	20.9	22.36	19.01	19.11
27.54	20.87	22.33	18.97	19.08
27.69	20.84	22.28	18.93	19.03
27.84	20.81	22.23	18.9	18.99

Table D1 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
27.99	20.8	22.18	18.86	18.95
28.14	20.76	22.15	18.82	18.91
28.29	20.74	22.11	18.78	18.88
28.44	20.72	22.06	18.74	18.82
28.59	20.69	22.03	18.71	18.78
28.74	20.67	21.98	18.67	18.74
28.89	20.65	21.94	18.63	18.7
29.04	20.62	21.89	18.6	18.67
29.19	20.6	21.86	18.55	18.63
29.34	20.57	21.82	18.52	18.59
29.49	20.56	21.78	18.48	18.56
29.64	20.52	21.74	18.45	18.52
29.79	20.5	21.7	18.42	18.47

Table D2 Cure behavior of FA /NR compounds with 4% w/w of 3 types of silanes in EV system

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
0.243	1.863	2.212	1.946	1.955
0.393	1.585	1.878	1.671	1.614
0.54	1.444	1.708	1.536	1.462
0.69	1.379	1.636	1.529	1.361
0.84	1.37	1.635	1.619	1.319
0.99	1.396	1.699	1.784	1.307
1.14	1.465	1.814	2.058	1.289
1.29	1.612	2.049	2.503	1.292
1.44	1.871	2.486	3.177	1.319
1.59	2.385	3.294	4.145	1.364
1.74	3.49	4.794	5.402	1.449
1.89	5.793	7.445	6.89	1.595
2.04	9.121	11	8.52	1.829
2.19	12.3	14.42	10.12	2.291
2.34	13	17.01	11.56	3.274

Table D2 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in EV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
2.49	13.74	18.81	12.8	5.576
2.64	14.83	20.11	13.85	9.419
2.79	15.61	21.07	14.72	12.89
2.94	16.22	21.84	15.45	15.23
3.09	16.68	22.43	16.08	16.78
3.24	17.03	22.93	16.6	17.9
3.39	17.33	23.34	17.07	18.74
3.54	17.56	23.67	17.46	19.42
3.69	17.76	23.96	17.81	19.94
3.84	17.92	24.19	18.11	20.38
3.99	18.03	24.4	18.38	20.74
4.14	18.15	24.57	18.62	21.03
4.29	18.23	24.72	18.84	21.27
4.44	18.3	24.88	19.03	21.48
4.59	18.36	25	19.19	21.64
4.74	18.4	25.09	19.34	21.79
4.89	18.45	25.19	19.49	21.91
5.04	18.49	25.26	19.61	22.01
5.19	18.52	25.34	19.72	22.09
5.34	18.53	25.41	19.82	22.16
5.49	18.56	25.49	19.93	22.21
5.64	18.57	25.54	20.01	22.25
5.79	18.59	25.58	20.08	22.27
5.94	18.6	25.63	20.15	22.3
6.09	18.62	25.69	20.21	22.32
6.24	18.62	25.7	20.27	22.33
6.39	18.63	25.74	20.32	22.36
6.54	18.63	25.78	20.37	22.35
6.69	18.64	25.8	20.41	22.36
6.84	18.64	25.84	20.46	22.36
6.99	18.63	25.86	20.49	22.36
7.14	18.64	25.88	20.52	22.34
7.29	18.63	25.89	20.55	22.34
7.44	18.64	25.91	20.58	22.33
7.59	18.64	25.95	20.6	22.31
7.74	18.63	25.95	20.62	22.3
7.89	18.63	25.97	20.64	22.29

Table D2 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in EV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
8.04	18.62	25.98	20.67	22.26
8.19	18.62	25.99	20.68	22.24
8.34	18.63	26.01	20.7	22.23
8.49	18.61	26.01	20.72	22.2
8.64	18.6	26.01	20.72	22.2
8.79	18.6	26.02	20.74	22.16
8.94	18.6	26.02	20.75	22.15
9.09	18.59	26.04	20.76	22.12
9.24	18.59	26.03	20.77	22.09
9.39	18.57	26.05	20.78	22.06
9.54	18.57	26.06	20.78	22.04
9.69	18.56	26.05	20.79	22.01
9.84	18.56	26.05	20.79	21.98
9.99	18.56	26.06	20.79	21.95
10.14	18.56	26.06	20.79	21.93
10.29	18.56	26.06	20.8	21.9
10.44	18.56	26.06	20.8	21.88
10.59	18.55	26.07	20.8	21.86
10.74	18.55	26.07	20.8	21.82
10.89	18.56	26.09	20.8	21.8
11.04	18.55	26.08	20.81	21.79
11.19	18.56	26.08	20.8	21.77
11.34	18.57	26.1	20.8	21.75
11.49	18.56	26.1	20.8	21.74
11.64	18.57	26.11	20.8	21.73
11.79	18.56	26.11	20.79	21.72
11.94	18.56	26.12	20.79	21.71
12.09	18.56	26.13	20.78	21.7
12.24	18.57	26.13	20.77	21.7
12.39	18.57	26.13	20.77	21.68
12.54	18.57	26.14	20.76	21.67
12.69	18.57	26.14	20.76	21.67
12.84	18.59	26.14	20.74	21.66
12.99	18.58	26.16	20.74	21.66
13.14	18.57	26.16	20.74	21.65
13.29	18.58	26.16	20.73	21.64
13.44	18.57	26.17	20.72	21.63

Table D2 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in EV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
13.59	18.59	26.17	20.72	21.63
13.74	18.57	26.18	20.69	21.62
13.89	18.58	26.18	20.69	21.61
14.04	18.58	26.19	20.68	21.61
14.19	18.58	26.19	20.67	21.61
14.34	18.59	26.2	20.67	21.6
14.49	18.58	26.2	20.65	21.6
14.64	18.57	26.2	20.66	21.6
14.79	18.58	26.21	20.64	21.59
14.94	18.58	26.22	20.64	21.58
15.09	18.57	26.22	20.62	21.57
15.24	18.59	26.21	20.61	21.56
15.39	18.58	26.23	20.61	21.56
15.54	18.58	26.23	20.6	21.55
15.69	18.58	26.23	20.59	21.55
15.84	18.58	26.23	20.58	21.56
15.99	18.57	26.23	20.57	21.54
16.14	18.56	26.24	20.55	21.54
16.29	18.57	26.23	20.54	21.53
16.44	18.56	26.25	20.55	21.53
16.59	18.56	26.24	20.54	21.52
16.74	18.56	26.24	20.51	21.52
16.89	18.56	26.24	20.51	21.51
17.04	18.56	26.23	20.5	21.49
17.19	18.55	26.24	20.49	21.49
17.34	18.55	26.24	20.48	21.49
17.49	18.55	26.24	20.48	21.48
17.64	18.55	26.24	20.47	21.48
17.79	18.55	26.24	20.47	21.48
17.94	18.55	26.24	20.44	21.47
18.09	18.55	26.24	20.43	21.46
18.24	18.55	26.25	20.43	21.47
18.39	18.54	26.25	20.43	21.45
18.54	18.53	26.25	20.42	21.44
18.69	18.54	26.25	20.41	21.44
18.84	18.53	26.25	20.39	21.44
18.99	18.53	26.25	20.38	21.44

Table D2 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in EV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
19.14	18.53	26.25	20.38	21.42
19.29	18.53	26.24	20.37	21.42
19.44	18.52	26.25	20.36	21.41
19.59	18.52	26.24	20.35	21.4
19.74	18.52	26.24	20.35	21.4
19.89	18.51	26.25	20.33	21.39
20.04	18.5	26.24	20.33	21.39
20.19	18.5	26.24	20.33	21.37
20.34	18.5	26.24	20.31	21.37
20.49	18.5	26.24	20.31	21.37
20.64	18.5	26.24	20.3	21.37
20.79	18.5	26.24	20.3	21.35
20.94	18.5	26.23	20.29	21.35
21.09	18.5	26.23	20.28	21.33
21.24	18.49	26.21	20.28	21.33
21.39	18.49	26.22	20.26	21.32
21.54	18.49	26.21	20.25	21.32
21.69	18.48	26.21	20.25	21.31
21.84	18.49	26.19	20.24	21.3
21.99	18.46	26.2	20.24	21.31
22.14	18.45	26.2	20.24	21.29
22.29	18.46	26.21	20.23	21.29
22.44	18.46	26.18	20.22	21.28
22.59	18.45	26.2	20.2	21.27
22.74	18.45	26.2	20.2	21.27
22.89	18.45	26.2	20.19	21.26
23.04	18.45	26.18	20.19	21.25
23.19	18.44	26.18	20.18	21.25
23.34	18.44	26.18	20.18	21.25
23.49	18.43	26.18	20.17	21.23
23.64	18.43	26.17	20.15	21.23
23.79	18.42	26.15	20.15	21.22
23.94	18.42	26.16	20.14	21.2
24.09	18.42	26.16	20.14	21.2
24.24	18.41	26.15	20.14	21.2
24.39	18.41	26.15	20.13	21.19
24.54	18.4	26.16	20.11	21.18

Table D2 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in EV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
24.69	18.4	26.15	20.13	21.17
24.84	18.39	26.15	20.1	21.17
24.99	18.39	26.16	20.1	21.15
25.14	18.38	26.14	20.09	21.15
25.29	18.37	26.15	20.09	21.14
25.44	18.38	26.14	20.08	21.13
25.59	18.38	26.13	20.07	21.13
25.74	18.37	26.12	20.07	21.12
25.89	18.37	26.12	20.06	21.11
26.04	18.37	26.12	20.05	21.11
26.19	18.37	26.12	20.05	21.1
26.34	18.35	26.12	20.04	21.1
26.49	18.35	26.11	20.04	21.09
26.64	18.34	26.1	20.03	21.08
26.79	18.35	26.1	20.03	21.07
26.94	18.34	26.1	20.02	21.07
27.09	18.33	26.1	20	21.06
27.24	18.33	26.09	20.01	21.05
27.39	18.33	26.09	20	21.04
27.54	18.32	26.08	19.98	21.03
27.69	18.32	26.08	19.98	21.03
27.84	18.31	26.07	19.98	21.02
27.99	18.31	26.07	19.98	21.02
28.14	18.31	26.07	19.97	21.01
28.29	18.3	26.06	19.95	21.01
28.44	18.28	26.05	19.96	20.99
28.59	18.28	26.04	19.94	20.99
28.74	18.28	26.04	19.94	20.98
28.89	18.28	26.04	19.93	20.98
29.04	18.27	26.04	19.92	20.97
29.19	18.26	26.04	19.92	20.97
29.34	18.27	26.03	19.91	20.96
29.49	18.26	26.02	19.9	20.95
29.64	18.26	26.02	19.9	20.95
29.79	18.26	26.01	19.89	20.95

Table D3 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in semi-EV system

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
0.243	1.58	2.357	1.846	1.949
0.393	1.336	1.998	1.562	1.606
0.54	1.227	1.843	1.452	1.462
0.69	1.187	1.758	1.446	1.372
0.84	1.202	1.756	1.561	1.32
0.99	1.272	1.828	1.793	1.295
1.14	1.42	1.983	2.187	1.277
1.29	1.707	2.3	2.866	1.269
1.44	2.377	2.873	3.975	1.269
1.59	4.016	3.95	5.724	1.287
1.74	7.329	5.951	8.225	1.318
1.89	11.21	9.108	11.21	1.382
2.04	14.26	12.77	14.04	1.485
2.19	16.45	16	16.35	1.667
2.34	18.04	18.54	18.06	1.99
2.49	19.19	20.42	19.29	2.68
2.64	20.01	21.82	20.18	4.35
2.79	20.6	22.86	20.85	7.894
2.94	21.03	23.65	21.35	12.16
3.09	21.34	24.27	21.74	15.4
3.24	21.59	24.75	22.05	17.66
3.39	21.76	25.13	22.3	19.26
3.54	21.9	25.45	22.49	20.43
3.69	21.99	25.71	22.64	21.28
3.84	22.06	25.91	22.76	21.94
3.99	22.1	26.09	22.86	22.46
4.14	22.14	26.23	22.94	22.89
4.29	22.16	26.35	23	23.22
4.44	22.18	26.45	23.05	23.49
4.59	22.19	26.54	23.1	23.72
4.74	22.18	26.62	23.13	23.91
4.89	22.17	26.67	23.14	24.05
5.04	22.16	26.72	23.16	24.18
5.19	22.14	26.75	23.17	24.27
5.34	22.13	26.79	23.18	24.33
5.49	22.1	26.82	23.18	24.39
5.64	22.07	26.85	23.17	24.44
5.79	22.04	26.86	23.17	24.47
5.94	22.02	26.88	23.16	24.49

Table D3 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in semi-EV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
6.09	21.99	26.9	23.15	24.5
6.24	21.97	26.9	23.15	24.5
6.39	21.93	26.91	23.12	24.5
6.54	21.9	26.91	23.12	24.5
6.69	21.86	26.92	23.1	24.49
6.84	21.82	26.91	23.08	24.45
6.99	21.79	26.91	23.07	24.43
7.14	21.75	26.91	23.05	24.4
7.29	21.73	26.91	23.02	24.38
7.44	21.68	26.9	23.01	24.34
7.59	21.65	26.89	22.99	24.32
7.74	21.62	26.88	22.97	24.26
7.89	21.59	26.88	22.96	24.24
8.04	21.56	26.86	22.94	24.2
8.19	21.53	26.85	22.91	24.15
8.34	21.51	26.83	22.89	24.11
8.49	21.48	26.82	22.88	24.07
8.64	21.46	26.81	22.85	24.03
8.79	21.45	26.8	22.83	23.99
8.94	21.42	26.79	22.83	23.94
9.09	21.41	26.77	22.8	23.9
9.24	21.39	26.77	22.79	23.84
9.39	21.37	26.75	22.77	23.8
9.54	21.35	26.73	22.75	23.75
9.69	21.33	26.72	22.74	23.71
9.84	21.32	26.71	22.72	23.65
9.99	21.3	26.7	22.71	23.6
10.14	21.29	26.69	22.7	23.54
10.29	21.26	26.68	22.68	23.5
10.44	21.26	26.67	22.67	23.44
10.59	21.24	26.67	22.65	23.38
10.74	21.23	26.65	22.64	23.34
10.89	21.21	26.65	22.62	23.29
11.04	21.19	26.64	22.62	23.25
11.19	21.18	26.63	22.61	23.21
11.34	21.18	26.62	22.58	23.17
11.49	21.16	26.62	22.57	23.12
11.64	21.14	26.61	22.56	23.09
11.79	21.12	26.6	22.54	23.05

Table D3 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in semi-EV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
11.94	21.11	26.59	22.53	23.02
12.09	21.09	26.59	22.52	22.98
12.24	21.09	26.58	22.51	22.95
12.39	21.07	26.57	22.5	22.93
12.54	21.06	26.56	22.49	22.9
12.69	21.04	26.55	22.48	22.87
12.84	21.02	26.55	22.47	22.84
12.99	21.01	26.54	22.46	22.82
13.14	21	26.53	22.44	22.79
13.29	20.99	26.52	22.43	22.78
13.44	20.97	26.51	22.43	22.75
13.59	20.96	26.5	22.42	22.73
13.74	20.95	26.49	22.41	22.7
13.89	20.91	26.48	22.39	22.69
14.04	20.91	26.48	22.38	22.67
14.19	20.89	26.45	22.38	22.64
14.34	20.87	26.44	22.37	22.62
14.49	20.86	26.44	22.36	22.6
14.64	20.85	26.43	22.35	22.59
14.79	20.84	26.43	22.33	22.57
14.94	20.82	26.42	22.33	22.55
15.09	20.81	26.41	22.31	22.53
15.24	20.79	26.4	22.3	22.52
15.39	20.78	26.39	22.28	22.49
15.54	20.77	26.37	22.27	22.48
15.69	20.77	26.36	22.27	22.47
15.84	20.75	26.36	22.25	22.44
15.99	20.74	26.35	22.25	22.43
16.14	20.73	26.33	22.24	22.41
16.29	20.7	26.32	22.23	22.39
16.44	20.7	26.31	22.21	22.38
16.59	20.68	26.3	22.2	22.36
16.74	20.67	26.3	22.19	22.35
16.89	20.65	26.29	22.18	22.33
17.04	20.64	26.27	22.17	22.32
17.19	20.63	26.25	22.16	22.3
17.34	20.61	26.25	22.15	22.29
17.49	20.6	26.24	22.14	22.28
17.64	20.58	26.22	22.13	22.25

Table D3 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in semi-EV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
17.79	20.58	26.21	22.12	22.23
17.94	20.55	26.2	22.11	22.22
18.09	20.54	26.2	22.1	22.2
18.24	20.52	26.19	22.09	22.18
18.39	20.51	26.16	22.07	22.17
18.54	20.5	26.16	22.07	22.15
18.69	20.49	26.15	22.05	22.14
18.84	20.47	26.14	22.05	22.13
18.99	20.46	26.12	22.04	22.11
19.14	20.44	26.12	22.03	22.1
19.29	20.43	26.1	22.01	22.09
19.44	20.42	26.09	22	22.06
19.59	20.41	26.07	21.99	22.05
19.74	20.39	26.07	21.98	22.03
19.89	20.39	26.05	21.97	22.03
20.04	20.37	26.06	21.96	22.01
20.19	20.36	26.03	21.95	22
20.34	20.34	26.02	21.94	21.98
20.49	20.34	26.01	21.92	21.97
20.64	20.32	26.01	21.91	21.95
20.79	20.31	26	21.91	21.94
20.94	20.3	25.97	21.89	21.93
21.09	20.29	25.96	21.9	21.91
21.24	20.27	25.96	21.88	21.91
21.39	20.27	25.95	21.87	21.89
21.54	20.27	25.94	21.86	21.87
21.69	20.25	25.92	21.85	21.85
21.84	20.23	25.9	21.84	21.85
21.99	20.23	25.9	21.84	21.83
22.14	20.2	25.88	21.82	21.81
22.29	20.19	25.87	21.81	21.8
22.44	20.18	25.87	21.79	21.79
22.59	20.17	25.85	21.79	21.77
22.74	20.17	25.84	21.78	21.76
22.89	20.16	25.82	21.77	21.75
23.04	20.14	25.81	21.76	21.74
23.19	20.13	25.8	21.75	21.72
23.34	20.11	25.79	21.73	21.71
23.49	20.1	25.77	21.72	21.7

Table D3 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in semi-EV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
23.64	20.09	25.76	21.71	21.68
23.79	20.07	25.74	21.7	21.67
23.94	20.07	25.73	21.69	21.66
24.09	20.05	25.73	21.67	21.64
24.24	20.05	25.71	21.66	21.63
24.39	20.03	25.69	21.66	21.62
24.54	20.03	25.68	21.65	21.6
24.69	20.01	25.67	21.64	21.59
24.84	19.99	25.66	21.62	21.58
24.99	19.99	25.65	21.62	21.56
25.14	19.98	25.64	21.6	21.55
25.29	19.97	25.63	21.59	21.54
25.44	19.95	25.61	21.59	21.53
25.59	19.94	25.6	21.58	21.52
25.74	19.93	25.59	21.56	21.51
25.89	19.91	25.58	21.56	21.5
26.04	19.9	25.56	21.55	21.48
26.19	19.89	25.55	21.54	21.47
26.34	19.88	25.54	21.53	21.44
26.49	19.87	25.53	21.51	21.44
26.64	19.87	25.52	21.51	21.43
26.79	19.84	25.49	21.5	21.41
26.94	19.82	25.49	21.49	21.39
27.09	19.82	25.48	21.47	21.39
27.24	19.81	25.47	21.47	21.38
27.39	19.81	25.47	21.46	21.37
27.54	19.79	25.45	21.45	21.36
27.69	19.78	25.43	21.44	21.34
27.84	19.77	25.43	21.43	21.34
27.99	19.76	25.42	21.42	21.32
28.14	19.74	25.4	21.4	21.31
28.29	19.73	25.39	21.4	21.3
28.44	19.72	25.38	21.39	21.27
28.59	19.71	25.37	21.37	21.27
28.74	19.7	25.35	21.37	21.26
28.89	19.69	25.35	21.35	21.25
29.04	19.69	25.33	21.34	21.24
29.19	19.67	25.32	21.33	21.22
29.34	19.66	25.31	21.32	21.2

Table D3 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in semi-EV system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
29.49	19.65	25.3	21.3	21.2
29.64	19.64	25.29	21.3	21.19
29.79	19.64	25.28	21.29	21.17

Table D4 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
0.383	2.003	2.009	1.782	1.65
0.68	1.827	1.851	1.611	1.468
0.98	1.887	1.97	1.653	1.499
1.28	1.95	2.138	1.714	1.547
1.58	2.027	2.331	1.791	1.626
1.88	2.114	2.538	1.889	1.692
2.18	2.205	2.745	1.985	1.78
2.48	2.302	2.949	2.086	1.865
2.78	2.4	3.156	2.181	1.953
3.08	2.497	3.353	2.289	2.043
3.38	2.6	3.538	2.392	2.138
3.68	2.7	3.719	2.487	2.23
3.98	2.798	3.896	2.592	2.325
4.28	2.895	4.072	2.691	2.421
4.58	2.991	4.234	2.795	2.517
4.88	3.096	4.395	2.892	2.606
5.18	3.193	4.548	3	2.704
5.48	3.293	4.698	3.09	2.799
5.78	3.39	4.847	3.196	2.896
6.08	3.488	4.992	3.282	2.99
6.38	3.589	5.126	3.381	3.078
6.68	3.687	5.273	3.478	3.17
6.98	3.777	5.406	3.565	3.264
7.28	3.871	5.54	3.658	3.351
7.58	3.964	5.672	3.752	3.439
7.88	4.054	5.802	3.832	3.527
8.18	4.147	5.92	3.922	3.619

Table D4 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
8.48	4.238	6.044	4.004	3.706
8.78	4.333	6.16	4.092	3.788
9.08	4.422	6.273	4.172	3.875
9.38	4.501	6.388	4.262	3.949
9.68	4.591	6.502	4.336	4.04
9.98	4.667	6.607	4.424	4.113
10.28	4.757	6.715	4.497	4.185
10.58	4.85	6.822	4.573	4.267
10.88	4.929	6.92	4.652	4.345
11.18	5.002	7.026	4.721	4.415
11.48	5.084	7.116	4.795	4.496
11.78	5.166	7.211	4.862	4.575
12.08	5.243	7.311	4.93	4.641
12.38	5.32	7.409	4.995	4.709
12.68	5.397	7.502	5.067	4.788
12.98	5.469	7.586	5.133	4.86
13.28	5.553	7.68	5.2	4.923
13.58	5.622	7.756	5.263	4.99
13.88	5.702	7.845	5.32	5.059
14.18	5.773	7.921	5.386	5.125
14.48	5.832	7.999	5.45	5.195
14.78	5.908	8.084	5.503	5.247
15.08	5.969	8.155	5.563	5.306
15.38	6.041	8.226	5.628	5.372
15.68	6.104	8.304	5.683	5.437
15.98	6.162	8.371	5.732	5.496
16.28	6.228	8.442	5.79	5.553
16.58	6.285	8.5	5.85	5.605
16.88	6.341	8.563	5.893	5.668
17.18	6.405	8.626	5.948	5.716
17.48	6.467	8.69	5.999	5.784
17.78	6.519	8.75	6.039	5.835
18.08	6.583	8.802	6.094	5.89
18.38	6.629	8.865	6.139	5.939
18.68	6.69	8.914	6.185	5.986
18.98	6.739	8.969	6.238	6.036
19.28	6.796	9.017	6.283	6.084
19.58	6.852	9.068	6.323	6.13
19.88	6.901	9.114	6.362	6.173

Table D4 Cure behaviour of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
20.18	6.952	9.157	6.414	6.225
20.48	7.009	9.199	6.457	6.268
20.78	7.054	9.251	6.492	6.312
21.08	7.098	9.286	6.543	6.36
21.38	7.146	9.327	6.573	6.398
21.68	7.187	9.367	6.614	6.441
21.98	7.228	9.404	6.65	6.487
22.28	7.276	9.439	6.688	6.525
22.58	7.317	9.478	6.728	6.561
22.88	7.36	9.504	6.759	6.6
23.18	7.405	9.54	6.79	6.638
23.48	7.434	9.569	6.828	6.67
23.78	7.487	9.595	6.866	6.715
24.08	7.521	9.619	6.895	6.75
24.38	7.555	9.651	6.929	6.785
24.68	7.604	9.67	6.962	6.817
24.98	7.642	9.693	6.999	6.853
25.28	7.675	9.727	7.03	6.894
25.58	7.701	9.737	7.054	6.923
25.88	7.743	9.773	7.088	6.948
26.18	7.775	9.782	7.112	6.981
26.48	7.803	9.805	7.149	7.015
26.78	7.841	9.822	7.172	7.039
27.08	7.877	9.837	7.198	7.076
27.38	7.906	9.855	7.228	7.105
27.68	7.937	9.871	7.255	7.141
27.98	7.96	9.89	7.283	7.165
28.28	7.996	9.904	7.308	7.201
28.58	8.029	9.916	7.332	7.222
28.88	8.044	9.934	7.354	7.256
29.18	8.081	9.947	7.373	7.273
29.48	8.098	9.954	7.404	7.296
29.78	8.129	9.964	7.429	7.322
30.08	8.163	9.973	7.444	7.355
30.38	8.19	9.976	7.479	7.377
30.68	8.215	10	7.509	7.396
30.98	8.231	10	7.522	7.419
31.28	8.256	10.01	7.544	7.445
31.58	8.286	10.02	7.571	7.468

Table D4 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
31.88	8.312	10.02	7.58	7.492
32.18	8.331	10.03	7.61	7.508
32.48	8.349	10.04	7.627	7.541
32.78	8.378	10.04	7.64	7.561
33.08	8.393	10.04	7.664	7.592
33.38	8.418	10.05	7.683	7.605
33.68	8.438	10.06	7.696	7.625
33.98	8.448	10.06	7.719	7.631
34.28	8.473	10.06	7.735	7.654
34.58	8.498	10.06	7.754	7.669
34.88	8.512	10.07	7.768	7.699
35.18	8.528	10.07	7.79	7.723
35.48	8.549	10.08	7.804	7.736
35.78	8.569	10.07	7.818	7.754
36.08	8.58	10.08	7.836	7.769
36.38	8.601	10.08	7.85	7.785
36.68	8.621	10.09	7.859	7.804
36.98	8.632	10.08	7.881	7.823
37.28	8.648	10.08	7.89	7.834
37.58	8.67	10.08	7.909	7.849
37.88	8.68	10.08	7.92	7.869
38.18	8.698	10.08	7.929	7.879
38.48	8.71	10.08	7.955	7.898
38.78	8.73	10.08	7.957	7.908
39.08	8.74	10.08	7.974	7.917
39.38	8.752	10.08	7.985	7.945
39.68	8.766	10.08	8.008	7.955
39.98	8.782	10.07	8.019	7.961
40.28	8.789	10.07	8.027	7.985
40.58	8.802	10.07	8.035	7.993
40.88	8.83	10.06	8.038	8.011
41.18	8.841	10.06	8.056	8.017
41.48	8.842	10.07	8.072	8.027
41.78	8.85	10.06	8.089	8.038
42.08	8.862	10.06	8.098	8.059
42.38	8.884	10.05	8.107	8.073
42.68	8.89	10.06	8.115	8.075
42.98	8.904	10.05	8.136	8.083
43.28	8.912	10.05	8.139	8.091

Table D4 Cure behaviour of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
43.58	8.915	10.04	8.148	8.114
43.88	8.928	10.04	8.157	8.122
44.18	8.932	10.04	8.173	8.135
44.48	8.947	10.03	8.172	8.139
44.78	8.952	10.03	8.184	8.147
45.08	8.963	10.02	8.196	8.161
45.38	8.981	10.02	8.205	8.169
45.68	8.983	10.01	8.208	8.179
45.98	8.996	10.01	8.22	8.188
46.28	8.997	10.01	8.228	8.195
46.58	9.009	10.01	8.239	8.207
46.88	9.019	10	8.247	8.217
47.18	9.025	9.993	8.256	8.235
47.48	9.032	9.996	8.264	8.235
47.78	9.042	9.989	8.267	8.242
48.08	9.043	9.992	8.273	8.25
48.38	9.053	9.985	8.277	8.253
48.68	9.064	9.972	8.279	8.261
48.98	9.068	9.971	8.297	8.267
49.28	9.072	9.973	8.297	8.277
49.58	9.081	9.967	8.302	8.286
49.88	9.084	9.967	8.308	8.293
50.18	9.088	9.966	8.323	8.307
50.48	9.093	9.958	8.328	8.306
50.78	9.097	9.954	8.331	8.314
51.08	9.104	9.936	8.33	8.318
51.38	9.106	9.945	8.343	8.323
51.68	9.111	9.937	8.35	8.339
51.98	9.114	9.935	8.352	8.334
52.28	9.132	9.922	8.366	8.344
52.58	9.133	9.916	8.366	8.354
52.88	9.133	9.915	8.376	8.356
53.18	9.143	9.9	8.38	8.36
53.48	9.144	9.903	8.386	8.366
53.78	9.146	9.9	8.392	8.375
54.08	9.149	9.895	8.404	8.377
54.38	9.149	9.887	8.403	8.387
54.68	9.159	9.886	8.408	8.391
54.98	9.163	9.88	8.414	8.397

Table D4 Cure behavior of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system (cont.)

Time (min)	Torque (dNm)			
	without	Si-69	Si-264	VTEO
55.28	9.165	9.866	8.417	8.405
55.58	9.169	9.873	8.421	8.408
55.88	9.174	9.865	8.432	8.422
56.18	9.182	9.865	8.434	8.423
56.48	9.186	9.854	8.438	8.426
56.78	9.187	9.855	8.445	8.433
57.08	9.191	9.848	8.442	8.429
57.38	9.19	9.847	8.441	8.438
57.68	9.199	9.838	8.445	8.437
57.98	9.206	9.837	8.453	8.441
58.28	9.198	9.83	8.46	8.447
58.58	9.203	9.824	8.457	8.454
58.88	9.206	9.821	8.461	8.461
59.18	9.208	9.815	8.466	8.464
59.48	9.211	9.814	8.474	8.47
55.28	9.165	9.866	8.417	8.405
55.58	9.169	9.873	8.421	8.408
55.88	9.174	9.865	8.432	8.422
56.18	9.182	9.865	8.434	8.423
56.48	9.186	9.854	8.438	8.426
56.78	9.187	9.855	8.445	8.433
57.08	9.191	9.848	8.442	8.429
57.38	9.19	9.847	8.441	8.438
57.68	9.199	9.838	8.445	8.437
57.98	9.206	9.837	8.453	8.441
58.28	9.198	9.83	8.46	8.447
58.58	9.203	9.824	8.457	8.454
58.88	9.206	9.821	8.461	8.461
59.18	9.208	9.815	8.466	8.464
59.48	9.211	9.814	8.474	8.47

Table D5 Degree of reversion of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in sulfur systems

Sulfur systems	Degree of reversion			
	without	Si-69	Si-264	VTEO
CV	15.45	11.05	15.42	18.28
EV	0.41	0.12	0.99	3.75
Semi-EV	6.57	2.03	4.00	8.48

Table D6 Marching rate of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in peroxide system

Peroxide system	Marching rate			
	without	Si-69	Si-264	VTEO
	0.18	0.19	0.16	0.17

Table D7 Scorch time of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in sulfur and peroxide systems

Curing systems	Scorch time (min)			
	without	Si-69	Si-264	VTEO
CV	1.09 $\pm$ 0.20	1.24 $\pm$ 0.20	1.07 $\pm$ 0.20	1.66 $\pm$ 0.30
EV	1.68 $\pm$ 0.10	1.58 $\pm$ 0.20	1.43 $\pm$ 0.30	2.30 $\pm$ 0.20
Semi-EV	1.49 $\pm$ 0.20	1.51 $\pm$ 0.20	1.32 $\pm$ 0.20	2.52 $\pm$ 0.30
DCP	6.25 $\pm$ 0.20	6.43 $\pm$ 0.30	5.84 $\pm$ 0.40	6.36 $\pm$ 0.40

Table D8 Cure time of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in sulfur and peroxide systems

Curing systems	Cure time (min)			
	without	Si-69	Si-264	VTEO
CV	3.52 $\pm$ 0.10	4.15 $\pm$ 0.20	3.82 $\pm$ 0.20	4.28 $\pm$ 0.20
EV	3.05 $\pm$ 0.10	3.56 $\pm$ 0.30	3.76 $\pm$ 0.20	4.28 $\pm$ 0.30
Semi-EV	2.64 $\pm$ 0.10	3.10 $\pm$ 0.30	2.80 $\pm$ 0.20	3.88 $\pm$ 0.20
DCP	31.78 $\pm$ 0.20	35.49 $\pm$ 0.50	34.37 $\pm$ 0.70	35.28 $\pm$ 0.60

Table D9 Torque difference of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in sulfur and peroxide systems

Curing systems	Torque difference (dNm)			
	without	Si-69	Si-264	VTEO
CV	26.34±0.60	28.73±0.50	24.44±0.50	28.9±0.40
EV	19.39±0.60	25.28±0.50	19.93±0.90	21.7±0.70
Semi-EV	21.56±0.30	25.83±0.40	22.38±0.60	23.85±0.50
DCP	7.26±0.60	8.88±0.40	7.45±0.60	7.61±0.90

Table D10 Crosslink density of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in sulfur and peroxide systems

Curing systems	Crosslink density ($\times 10^{-4}$ mole/dm ³)			
	without	Si-69	Si-264	VTEO
CV	2.59	4.93	4.43	4.13
EV	2.3	6.88	4.52	3.86
Semi-EV	2.41	6.19	5.57	3.98
DCP	1.09	1.72	0.6	0.84

Table D11 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in CV system

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.22	723.71	807.1	736.36	801.46
0.38	721.91	810	740.55	802.14
0.53	721.47	810.9	740.23	800.01
0.69	721.9	810.2	740.01	801.71
0.85	722.11	809.5	740.33	801.82
1.00	721.79	810.4	739.69	801.07
1.16	716.6	809.4	740.33	800.75
1.32	718.91	810.5	739.91	803.84
1.47	717.32	809.1	740.65	801.18
1.63	719.02	809	741.4	801.18
1.79	717.1	809	740.76	802.56
1.95	716.78	810.6	741.08	802.24
2.10	717.53	810.8	741.29	801.71
2.26	716.04	811.1	741.72	802.88
2.42	715.08	815.2	740.97	800.97

Table D11 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in CV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
2.57	715.19	811.5	740.55	802.88
2.73	713.59	811.6	740.55	800.97
2.89	714.97	809.8	741.29	802.03
3.04	712.63	811.5	740.55	802.88
3.20	713.91	812.6	742.14	802.56
3.36	713.27	812.7	740.55	802.14
3.51	710.18	813.2	743.42	802.24
3.67	713.05	814.7	742.36	802.14
3.83	711.14	813.2	743.53	802.78
3.99	713.37	815.1	742.36	802.24
4.14	712.63	813.7	743.74	804.2
4.30	713.05	813.6	743.95	802.24
4.46	714.12	814.6	745.34	802.24
4.61	713.27	815.2	743.1	802.78
4.77	712.84	814.6	743.95	802.78
4.93	713.37	815.9	743.21	802.88
5.08	713.59	815.1	744.59	802.46
5.24	713.59	814.6	744.27	803.52
5.40	713.05	814.6	745.02	803.52
5.56	713.69	815.5	744.59	802.24
5.71	713.37	815.9	744.49	803.31
5.87	714.76	816.1	744.49	803.95
6.03	712.63	816.1	744.91	803.63
6.18	714.12	816.4	744.59	802.03
6.34	711.56	815.9	744.49	804.27
6.50	715.19	816.5	745.23	802.56
6.65	715.72	816.4	744.91	803.95
6.81	713.59	816.4	744.91	802.46
6.97	712.95	816.4	745.34	803.2
7.13	714.33	815.8	746.09	803.63
7.28	713.27	815.9	746.09	802.56
7.44	713.05	817.5	745.02	803.95
7.60	714.12	817	744.27	803.31
7.75	713.59	816.8	745.66	803.31
7.91	714.12	816.8	744.81	803.2
8.07	714.33	816.8	746.94	803.1
8.22	713.91	816.8	745.55	804.59

Table D11 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in CV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
8.38	714.44	816.8	746.94	802.46
8.54	714.65	817.2	746.41	804.27
8.69	715.4	815.8	745.87	802.14
8.85	712.63	815.9	746.73	803.84
9.01	714.44	815.5	745.87	802.24
9.17	712.84	816.2	747.15	802.56
9.32	714.76	817.9	745.02	804.59
9.48	713.27	817	746.62	802.24
9.64	716.14	816.9	746.73	803.95
9.79	712.84	817.5	744.81	802.56
9.95	715.08	817	748	804.38
10.11	713.91	816.9	744.91	801.71
10.26	713.37	817	744.81	805.33
10.42	713.59	818.2	746.62	803.31
10.58	713.37	818.6	747.36	804.27
10.74	714.44	817.5	744.91	802.56
10.89	713.59	817.9	745.98	803.95
11.05	715.19	818	746.3	803.2
11.21	713.69	817.6	746.94	802.56
11.36	714.01	816.4	746.3	803.84
11.52	714.44	816.9	745.87	804.16
11.68	714.01	818	746.62	803.52
11.83	712.31	817.8	745.34	804.27
11.99	715.82	817.3	745.23	804.69
12.15	713.69	818.6	746.3	804.59
12.30	715.19	817.2	747.36	804.69
12.46	714.33	817.5	746.3	804.38
12.62	715.82	817.9	745.98	803.95
12.78	714.12	819	745.98	803.84
12.93	712.95	817.9	746.3	804.27
13.09	716.78	818	745.55	803.95
13.25	714.33	818	745.66	802.78
13.40	714.97	819.1	745.34	803.52
13.56	715.51	818	746.73	803.63
13.72	715.08	817.9	744.91	804.16
13.87	715.4	818.6	745.34	804.59
14.03	716.78	818.9	745.66	804.38

Table D11 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in CV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
14.19	716.57	819	745.66	804.38
14.35	715.51	818.9	747.15	803.95
14.50	715.72	818.9	746.73	804.69
14.66	714.33	819.6	745.66	803.63
14.82	716.04	820.1	745.23	804.38
14.97	717.53	819.7	744.91	804.27
15.13	716.57	818.7	745.55	804.59
15.29	715.72	820	746.41	804.38
15.44	713.52	818.7	746.41	803.95
15.60	716.04	819	745.66	804.27
15.76	716.78	820.2	746.94	803.52
15.92	714.97	820.4	748	803.52
16.07	715.4	820.4	747.04	803.63
16.23	715.82	820.7	746.41	802.46
16.39	715.4	820.2	745.34	804.27
16.54	716.46	819.7	747.36	804.59
16.70	717.64	820.8	746.94	804.16
16.86	719.53	820.2	747.04	803.84
17.01	714.97	820.7	745.87	804.38
17.17	716.57	820.8	746.94	804.59
17.33	716.78	821	747.04	803.95
17.48	715.4	821.1	746.62	804.69
17.64	715.19	820.7	746.94	805.01
17.80	714.97	820.8	746.62	805.33
17.96	715.72	821	746.41	803.95
18.11	715.82	820.5	746.73	803.95
18.27	716.89	820.4	746.3	803.63
18.43	717.32	822.3	746.94	804.59
18.58	718.28	821.1	747.47	804.69
18.74	717.85	821.4	746.73	804.16
18.90	717.96	820.1	747.04	804.38
19.05	716.78	820.5	747.36	804.38
19.21	717.64	820.8	747.36	804.38
19.37	717.32	820.8	747.04	804.69
19.53	717.1	820.4	747.15	804.38
19.68	717.1	821.6	747.79	805.23
19.84	717.32	820.8	748.11	805.01

Table D11 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in CV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
20.00	716.57	821.6	748.11	805.23
20.15	717.1	820.5	747.79	805.01
20.31	716.89	821.6	747.36	805.23
20.47	717.21	821.6	746.62	805.01
20.62	717.1	820.1	747.15	804.91
20.78	713.84	821.8	748.75	804.59
20.94	716.46	822.2	747.47	805.33
21.09	717.1	822.3	748.75	805.01
21.25	716.78	822.6	747.68	804.59
21.41	717.85	821.8	748	805.01
21.57	717.1	822.2	748	805.33
21.72	717.53	821.8	746.73	804.69
21.88	716.89	823.4	746.3	804.27
22.04	716.46	823.3	747.68	804.38
22.19	717.21	823.4	750.52	804.59
22.35	717.85	823.6	749.07	804.38
22.51	718.28	824.2	748.43	803.84
22.66	718.91	823.3	749.6	804.27
22.82	719.32	824.6	748	804.38
22.98	718.38	823.3	747.15	804.59
23.14	720.28	823.4	747.47	804.38
23.29	719.66	823.9	748	804.38
23.45	717.96	823.3	747.47	804.16
23.61	717.53	825	749.18	804.38
23.76	718.6	824.3	748.43	804.59
23.92	719.98	822.9	748.86	805.23
24.08	717.21	823.6	748.54	805.01
24.23	717.96	823.6	749.18	804.69
24.39	717.64	824.3	749.6	805.65
24.55	718.17	825.3	748.54	804.38
24.71	719.34	824.7	747.36	804.69
24.86	719.23	825.4	749.18	803.84
25.02	718.28	825.4	748.11	804.59
25.18	718.6	824.7	748.43	804.69
25.33	719.98	826.3	749.07	803.84
25.49	718.38	825.3	748.75	803.95
25.65	718.28	824.3	749.07	804.27

Table D11 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in CV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
25.80	719.45	824.3	749.07	805.23
25.96	717.32	824.7	749.5	805.65
26.12	719.45	824.6	748.54	805.44
26.27	718.28	824.3	748.75	805.23
26.43	719.02	824.7	748.54	804.69
26.59	720.3	825.7	748.75	804.69
26.75	718.91	826	749.07	805.44
26.90	718.6	825.4	750.13	804.69
27.06	718.38	826	749.07	805.23
27.22	719.23	826	750.24	804.91
27.37	719.45	825.7	749.5	805.01
27.53	718.38	826.3	749.5	805.33
27.69	718.7	825.7	749.18	805.01
27.84	718.91	825.7	750.13	805.97
28.00	718.91	825.4	748.75	805.33
28.16	718.6	825	749.07	805.23
28.32	721.24	826	750.56	805.76
28.47	718.38	827.4	750.24	805.44
28.63	718.91	826.1	750.13	805.76
28.79	719.45	825.3	750.35	805.01
28.94	718.91	826.3	750.67	805.23
29.10	719.34	826.6	750.99	806.29
29.26	719.77	826.9	749.6	806.51
29.41	718.28	827.5	750.35	805.97
29.57	719.23	827.4	749.6	805.44
29.73	719.34	827.8	749.82	805.76
29.88	719.23	827.8	749.6	805.44

Table D12 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in EV system

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.22	967.43	1022.2	950.27	970.17
0.38	964.33	1027.2	951.36	972.73
0.53	965.61	1027	951.79	976.35
0.69	962.18	1026.3	951.79	974.32
0.85	954.62	1026.6	952.11	974.86
1.00	957.06	1027.4	951.57	975.92
1.16	958.45	1027.9	951.04	974.54
1.32	957.59	1028.2	951.36	976.77
1.47	953.86	1028	951.04	976.35
1.63	955.14	1028.8	952.96	975.71
1.79	950.68	1027.9	952.53	974.54
1.95	953.65	1027.7	953.49	977.84
2.10	953.12	1028	952.96	976.35
2.26	952.69	1027.3	954.66	977.31
2.42	953.86	1027	954.02	977.73
2.57	952.8	1028	952.85	977.41
2.73	955.78	1027.4	952.96	976.77
2.89	955.78	1029.3	953.6	976.77
3.04	954.08	1029.5	954.66	977.73
3.20	953.12	1028.5	953.17	977.84
3.36	952.59	1028	954.24	978.37
3.51	953.44	1028.5	953.6	979.65
3.67	952.27	1029.3	954.56	977.52
3.83	953.65	1029.8	954.98	978.91
3.99	955.78	1029.4	955.73	979.54
4.14	952.59	1028.8	956.37	979.54
4.30	955.14	1029	955.84	977.41
4.46	953.65	1028.5	955.84	978.05
4.61	956	1029.4	956.05	977.09
4.77	955.57	1029.7	956.9	978.59
4.93	956	1030.2	956.16	978.48
5.08	953.65	1030.5	956.79	979.65
5.24	957.38	1030.6	957.75	979.44
5.40	953.12	1030.5	957.75	977.84
5.56	955.78	1032.2	957.75	978.59
5.71	955.25	1031.4	958.61	980.29
5.87	955.78	1032.3	957.75	978.8

Table D12 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in EV (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
6.03	956	1032	957.97	980.72
6.18	958.66	1030.6	958.82	979.97
6.34	956.21	1030.9	957.86	979.65
6.50	954.82	1030.8	957.43	979.54
6.65	956	1031.6	958.29	980.93
6.81	958.98	1031.1	958.5	980.5
6.97	957.06	1032.3	957.86	980.18
7.13	957.06	1033.6	957.54	980.18
7.28	958.66	1031.4	957.54	980.72
7.44	958.34	1031.6	959.03	979.86
7.60	959.09	1031.9	958.5	980.61
7.75	954.62	1033.2	958.5	979.44
7.91	957.91	1032.6	958.18	978.8
8.07	959.09	1034.3	958.5	980.29
8.22	957.06	1032.9	958.61	980.29
8.38	958.13	1034.4	959.57	981.36
8.54	956.64	1034.4	958.61	980.72
8.69	957.59	1034.6	959.88	980.61
8.85	958.34	1034.3	958.93	980.61
9.01	960.47	1034.7	959.25	982.42
9.17	958.45	1034.6	959.03	980.29
9.32	958.66	1034.8	959.25	978.91
9.48	957.91	1034.8	959.03	981.68
9.64	959.41	1034.8	958.61	981.57
9.79	959.83	1035.9	958.18	981.36
9.95	960.26	1034.4	960.1	981.57
10.11	958.66	1034.7	959.67	980.93
10.26	959.83	1034.4	958.18	981.68
10.42	959.41	1035.2	957.97	982.1
10.58	958.02	1035.5	959.03	981.57
10.74	960.26	1034.4	959.25	979.97
10.89	959.51	1034.8	959.25	982
11.05	960.58	1036.2	959.25	981.04
11.21	961.54	1035.2	958.29	981.25
11.36	961.64	1034	959.03	982.32
11.52	960.26	1035.5	959.67	982.42
11.68	962.41	1035.8	959.57	983.06

Table D12 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in EV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
11.83	960.58	1036.1	959.99	982.32
11.99	960.79	1034.7	959.99	982.32
12.15	961.22	1035.1	958.61	982.1
12.30	961.22	1036.6	959.25	980.93
12.46	959.83	1036.1	958.93	982
12.62	960.47	1035.5	959.67	982.63
12.78	961.32	1035.7	959.88	981.57
12.93	959.73	1036.1	959.67	981.36
13.09	959.51	1035.1	960.31	983.17
13.25	962.18	1035.5	960.31	981.04
13.40	962.28	1035.9	959.88	983.38
13.56	963.03	1036.2	960.74	982.42
13.72	963.03	1036.6	960.31	982.85
13.87	962.28	1036.2	959.57	982.32
14.03	962.71	1037.5	959.57	983.81
14.19	961.86	1035.7	959.67	983.49
14.35	962.28	1037	961.06	982.74
14.50	962.18	1037.6	960.31	981.25
14.66	962.39	1037.5	958.61	982.32
14.82	961.54	1038	959.67	982
14.97	962.18	1037.3	959.99	981.78
15.13	961.64	1036.9	960.31	982.74
15.29	961.11	1036.1	960.74	982.63
15.44	961.54	1035.8	959.99	982.1
15.60	962.92	1035.7	961.38	980.72
15.76	961.54	1036.2	958.82	982.42
15.92	962.6	1036.2	961.06	982.42
16.07	961.96	1036.2	960.95	982.74
16.23	964.41	1036.6	961.16	984.13
16.39	963.77	1036.2	961.7	984.13
16.54	965.91	1036.8	959.57	983.81
16.70	964.52	1035.8	960.42	983.91
16.86	964.31	1037.9	960.42	983.38
17.01	965.59	1037.2	961.06	983.17
17.17	965.48	1036.2	960.74	983.91
17.33	965.37	1037.8	961.48	983.49
17.48	964.31	1038	961.48	984.87

Table D12 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in EV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
17.64	964.31	1037.5	960.42	983.7
17.80	965.37	1036.9	961.16	984.55
17.96	965.37	1037	961.7	984.87
18.11	965.05	1038	962.23	983.49
18.27	964.41	1039.1	961.16	985.51
18.43	965.16	1037.9	961.16	984.98
18.58	966.12	1038.7	961.16	985.19
18.74	965.16	1037.9	962.12	985.51
18.90	965.8	1037.9	961.7	984.45
19.05	964.09	1038.6	962.23	984.23
19.21	965.16	1037.6	962.23	984.77
19.37	964.31	1036.8	962.12	982.85
19.53	965.37	1038.4	961.48	983.49
19.68	965.37	1039.3	961.48	983.49
19.84	964.52	1038.4	961.06	984.13
20.00	965.05	1038	961.16	983.91
20.15	966.54	1038.3	961.8	985.51
20.31	965.59	1037.6	962.02	984.98
20.47	966.22	1037.2	962.12	986.26
20.62	965.8	1037.9	963.19	984.87
20.78	965.37	1039.1	962.76	984.45
20.94	966.54	1038.7	962.23	984.77
21.09	965.91	1038.4	962.87	984.77
21.25	967.5	1039	963.08	984.77
21.41	967.18	1039	962.87	984.98
21.57	967.18	1039.4	962.44	985.83
21.72	966.54	1038.3	962.23	985.3
21.88	966.12	1039	962.02	986.26
22.04	966.65	1039.6	962.44	984.23
22.19	965.59	1039	962.23	983.91
22.35	966.97	1038.4	962.76	984.23
22.51	968.04	1038.7	962.76	984.55
22.66	969	1038.7	962.76	985.19
22.82	969.42	1039.1	962.76	984.87
22.98	968.78	1039.3	963.83	984.45
23.14	968.25	1039.4	963.19	984.77
23.29	968.57	1039.4	963.51	986.04

Table D12 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in EV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
23.45	969.32	1038.9	963.51	985.19
23.61	968.25	1039.4	962.55	985.51
23.76	970.74	1040.8	963.51	986.04
23.92	968.25	1039.6	963.08	986.36
24.08	969.74	1039.6	962.55	986.58
24.23	969	1040.7	963.61	987.11
24.39	969.74	1038.7	962.87	986.58
24.55	968.78	1039	964.25	986.36
24.71	969.32	1039.4	964.25	986.26
24.86	970.06	1039.4	963.83	985.19
25.02	970.49	1039	964.25	986.68
25.18	969.74	1039	963.61	986.04
25.33	969.85	1040.1	963.61	986.68
25.49	968.25	1040.5	963.93	986.9
25.65	969.42	1040.7	963.08	985.3
25.80	968.78	1040.4	963.08	985.83
25.96	968.68	1040.4	963.93	986.68
26.12	968.78	1039.4	964.15	987
26.27	968.68	1039.4	963.19	987.96
26.43	970.7	1039.1	963.93	986.36
26.59	971.55	1039.4	963.29	986.68
26.75	972.19	1039.3	963.83	987.96
26.90	972.51	1039.4	963.93	987.75
27.06	971.13	1039.6	964.57	986.36
27.22	970.38	1039.7	963.61	986.04
27.37	971.87	1039.4	963.83	987.32
27.53	971.77	1040.5	964.25	988.07
27.69	970.91	1040.5	964.89	987.96
27.84	971.77	1040.8	964.25	986.9
28.00	971.77	1041.9	964.68	986.36
28.16	971.23	1041.2	965.43	987.32
28.32	970.91	1041.5	964.57	987.75
28.47	972.83	1041.6	965.43	988.18
28.63	972.62	1040.5	966.06	987
28.79	972.62	1041.6	964.15	987
28.94	973.04	1041.8	964.57	987
29.10	972.3	1040.5	963.93	987.64

Table D12 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in EV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
29.26	973.36	1042.3	964.68	988.39
29.41	971.87	1042.1	964.25	987
29.57	972.3	1042.1	964.68	987
29.73	972.19	1042.6	964.68	987.32
29.88	970.81	1041.6	964.57	988.71

Table D13 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in semi-EV system

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.22	723.71	796.3	744.16	766.38
0.38	721.91	791.9	746.63	766.7
0.53	721.47	792.4	747.27	766.92
0.69	721.9	794.5	747.06	766.7
0.85	722.11	795.8	748.02	767.45
1.00	721.79	796.7	748.02	767.77
1.16	716.6	796.2	748.55	768.62
1.32	718.91	795.5	748.55	768.2
1.47	717.32	797.4	748.34	766.7
1.63	719.02	796.2	749.19	766.7
1.79	717.1	795.8	748.12	768.94
1.95	716.78	796.7	749.83	766.49
2.10	717.53	795.6	748.55	766.38
2.26	716.04	796.5	749.83	768.2
2.42	715.08	799.8	748.44	768.83
2.57	715.19	800.2	749.51	766.81
2.73	713.59	799.5	749.4	767.98
2.89	714.97	800.6	749.51	767.98
3.04	712.63	798	749.93	767.98
3.20	713.91	799	750.25	768.94
3.36	713.27	799.1	750.47	767.98
3.51	710.18	799	750.47	769.26
3.67	713.05	798.8	750.68	767.56
3.83	711.14	799.1	750.57	769.69

Table D13 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in semi-EV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
3.99	713.37	798.8	750.15	767.77
4.14	712.63	798	749.93	767.56
4.30	713.05	801.7	750.47	769.05
4.46	714.12	802.9	750.47	768.83
4.61	713.27	803.6	750.57	769.5
4.77	712.84	803	751.75	769.26
4.93	713.37	802.5	751	768.2
5.08	713.59	801.6	749.83	769.9
5.24	713.59	802.9	750.25	768.94
5.40	713.05	803.4	750.89	768.3
5.56	713.69	802.9	750.89	768.83
5.71	713.37	802.7	751.32	769.26
5.87	714.76	802.7	750.57	767.88
6.03	712.63	803.3	751	768.3
6.18	714.12	802.5	750.25	768.83
6.34	711.56	803.3	750.89	768.83
6.50	715.19	805.1	749.19	769.05
6.65	715.72	807.3	750.15	768.83
6.81	713.59	808.1	749.93	769.05
6.97	712.95	808.1	749.93	768.94
7.13	714.33	807.9	750.25	768.3
7.28	713.27	807.6	751.21	768.83
7.44	713.05	808.6	750.89	768.83
7.60	714.12	808.6	751.64	769.05
7.75	713.59	808.7	750.57	769.58
7.91	714.12	806.5	750.25	769.37
8.07	714.33	807.6	749.62	769.05
8.22	713.91	807.2	751	768.62
8.38	714.44	807.2	751.21	768.94
8.54	714.65	808.3	750.68	769.26
8.69	715.4	807.9	751.32	769.26
8.85	712.63	806.2	751.53	768.94
9.01	714.44	807.8	751.32	768.83
9.17	712.84	808.6	750.57	768.94
9.32	714.76	808.7	750.47	769.37
9.48	713.27	810.4	750.68	769.37
9.64	716.14	810.8	751.21	769.9

Table D13 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in semi-EV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
9.79	712.84	809.8	750.89	769.37
9.95	715.08	812.6	750.89	769.26
10.11	713.91	811.6	751.21	768.83
10.26	713.37	810.2	751.32	769.69
10.42	713.59	810.1	751.21	768.94
10.58	713.37	811.6	751.53	768.51
10.74	714.44	812.5	751.53	769.05
10.89	713.59	813.4	751.21	769.05
11.05	715.19	814	752.07	768.62
11.21	713.69	812.8	751.96	769.26
11.36	714.01	814	751	768.62
11.52	714.44	812.6	751.64	769.69
11.68	714.01	812.3	751.64	769.58
11.83	712.31	812.5	751.21	769.9
11.99	715.82	814	752.07	769.37
12.15	713.69	814.4	751.64	769.26
12.30	715.19	812.8	751.32	770.11
12.46	714.33	812.8	752.39	770.11
12.62	715.82	813.4	752.07	769.9
12.78	714.12	814.4	751.96	770.01
12.93	712.95	813.6	752.28	768.94
13.09	716.78	814.3	751.64	768.3
13.25	714.33	815.3	751.53	770.33
13.40	714.97	814.4	752.07	770.75
13.56	715.51	814.8	752.07	769.58
13.72	715.08	814.3	752.28	769.9
13.87	715.4	814.3	751.75	769.05
14.03	716.78	814.8	752.07	770.97
14.19	716.57	815	752.6	770.11
14.35	715.51	814.7	753.77	770.97
14.50	715.72	815.5	752.71	769.9
14.66	714.33	814.8	753.02	770.11
14.82	716.04	815.8	752.39	769.37
14.97	717.53	813.9	751.96	769.26
15.13	716.57	815.1	751.96	769.37
15.29	715.72	815.7	751.75	770.43
15.44	713.52	816.5	753.13	770.43

Table D13 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in semi-EV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
15.60	716.04	816.9	751.64	768.94
15.76	716.78	815.5	752.39	769.37
15.92	714.97	816.8	752.81	770.01
16.07	715.4	817.4	752.6	770.43
16.23	715.82	816.9	753.13	770.33
16.39	715.4	816.6	753.13	770.33
16.54	716.46	816.8	753.02	770.97
16.70	717.64	817.6	753.88	770.11
16.86	719.53	818.3	752.6	770.65
17.01	714.97	816.8	752.6	771.07
17.17	716.57	818.5	753.13	770.33
17.33	716.78	818	752.81	770.33
17.48	715.4	820.1	752.81	769.58
17.64	715.19	820.7	752.6	769.58
17.80	714.97	819.6	753.77	769.9
17.96	715.72	817.2	753.88	770.65
18.11	715.82	819.6	753.66	770.75
18.27	716.89	821.1	753.45	770.43
18.43	717.32	821.8	753.34	770.65
18.58	718.28	821.4	754.09	769.37
18.74	717.85	820.7	752.07	770.01
18.90	717.96	820.4	753.77	769.58
19.05	716.78	820	752.6	769.37
19.21	717.64	821.2	752.39	769.69
19.37	717.32	822.4	752.81	769.69
19.53	717.1	821.2	752.71	770.01
19.68	717.1	822.9	754.84	770.43
19.84	717.32	823.6	754.52	770.33
20.00	716.57	823.6	754.84	769.9
20.15	717.1	825	754.2	771.18
20.31	716.89	823.3	753.88	770.01
20.47	717.21	824.2	753.45	770.43
20.62	717.1	824.7	754.09	769.69
20.78	713.84	824.4	753.34	770.75
20.94	716.46	824.6	753.45	770.01
21.09	717.1	822.2	754.41	770.11
21.25	716.78	823.9	754.09	771.07

Table D13 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in semi-EV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
21.41	717.85	824.6	753.66	771.18
21.57	717.1	823	754.09	770.33
21.72	717.53	825.6	754.73	770.43
21.88	716.89	824	753.88	770.01
22.04	716.46	824.2	753.88	770.11
22.19	717.21	824	754.2	770.33
22.35	717.85	823.8	754.09	770.01
22.51	718.28	824.9	753.88	770.65
22.66	718.91	824.6	754.52	770.01
22.82	719.32	826.2	753.02	771.07
22.98	718.38	824.9	752.81	771.07
23.14	720.28	825.4	753.88	770.75
23.29	719.66	825.3	754.52	772.03
23.45	717.96	826.8	754.94	771.39
23.61	717.53	825.6	755.26	772.14
23.76	718.6	826.4	755.8	771.39
23.92	719.98	826	755.16	771.18
24.08	717.21	826.8	754.73	770.33
24.23	717.96	825.7	754.41	770.01
24.39	717.64	825.4	754.84	770.75
24.55	718.17	826.1	754.41	770.65
24.71	719.34	827	754.41	771.07
24.86	719.23	826.1	754.2	769.9
25.02	718.28	825.7	754.41	770.97
25.18	718.6	825.7	754.84	771.39
25.33	719.98	827	755.26	772.24
25.49	718.38	826.4	754.41	771.18
25.65	718.28	826.4	755.16	770.75
25.80	719.45	827.2	755.26	770.65
25.96	717.32	826.5	754.52	771.71
26.12	719.45	827.9	756.01	771.18
26.27	718.28	826.7	754.73	772.46
26.43	719.02	826.1	754.41	771.71
26.59	720.3	826.7	756.22	772.03
26.75	718.91	828.1	756.01	771.18
26.90	718.6	828.6	755.58	771.71
27.06	718.38	827.8	755.26	771.39

Table D13 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in semi-EV system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
27.22	719.23	829.2	756.65	772.24
27.37	719.45	829.2	756.33	771.82
27.53	718.38	828.1	756.86	772.14
27.69	718.7	829.3	756.97	771.82
27.84	718.91	828.2	755.9	770.11
28.00	718.91	827.9	757.07	771.71
28.16	718.6	828.6	755.9	771.71
28.32	721.24	827.8	756.22	771.39
28.47	718.38	828.8	755.48	771.82
28.63	718.91	828.6	756.01	771.18
28.79	719.45	828.6	758.03	771.18
28.94	718.91	829.6	756.86	771.18
29.10	719.34	828.3	756.65	773.14
29.26	719.77	830.2	758.03	771.39
29.41	718.28	828.6	756.22	772.78
29.57	719.23	830.2	756.33	772.56
29.73	719.34	829.9	755.48	772.78
29.88	719.23	831.4	756.54	772.78

Table D14 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in peroxide system

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.22	326.17	413.9	367.14	344.9
0.38	324.35	415.42	358.34	345.7
0.53	323.39	416.16	358.09	346.6
0.69	322.54	416.38	356.81	348.7
0.85	322.54	416.91	354.64	345.2
1.00	321.58	417.23	356.27	347.7
1.16	321.9	417.76	353.79	346.2
1.32	322.32	417.55	354.11	348.7
1.47	322.22	417.55	355.39	346.6
1.63	322	419.25	353.79	345.2
1.79	321.79	418.83	354.43	347.8
1.95	319.52	419.36	356.13	348.8
2.10	322	418.93	355.92	350.8

Table D14 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in peroxide system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
2.26	323.12	419.89	354.86	349.2
2.42	322.48	419.89	355.71	349.2
2.57	324.19	419.57	355.71	350.2
2.73	323.76	419.57	356.56	348.3
2.89	323.6	420.32	356.88	349.9
3.04	324.83	419.04	357.31	350.9
3.20	323.39	419.89	359.12	351.3
3.36	322.54	418.83	358.27	351.5
3.51	322.54	419.04	356.99	351.5
3.67	322.54	419.89	356.88	349.8
3.83	323.07	419.04	357.84	350.1
3.99	323.92	420	359.44	350.2
4.14	323.39	420.32	359.12	352
4.30	323.39	420.74	358.9	351.6
4.46	322.86	420.43	360.08	351.6
4.61	324.19	419.89	359.12	352.9
4.77	324.03	420.11	359.65	351.9
4.93	323.28	420.32	360.4	352.4
5.08	324.03	420.96	359.97	353.4
5.24	323.28	420.74	362.15	353
5.40	324.19	421.06	361.04	354
5.56	323.71	421.17	360.4	355.1
5.71	323.28	420.43	359.44	355
5.87	324.19	421.17	362.31	356.1
6.03	323.71	421.49	360.18	356.2
6.18	323.39	421.7	360.72	357.6
6.34	323.39	420.96	360.5	358.3
6.50	324.67	420.74	361.04	355.9
6.65	325.2	421.7	360.82	358.4
6.81	324.67	422.13	360.72	358.3
6.97	324.99	421.06	360.82	357.7
7.13	325.09	421.49	361.14	357.2
7.28	324.35	421.38	360.82	358
7.44	323.92	422.13	362.53	357.7
7.60	324.35	420	362.85	359.1
7.75	324.93	422.56	363.59	358.6
7.91	324.35	423.3	362.21	359.7

Table D14 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in peroxide system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
8.07	325.09	422.77	362.85	358.4
8.22	323.92	422.24	362.85	357.4
8.38	325.73	421.81	362.85	359.3
8.54	325.41	422.45	363.27	360.5
8.69	325.09	422.24	363.38	359.4
8.85	324.67	422.13	365.08	360.3
9.01	326.27	422.45	366.96	359.1
9.17	325.73	421.7	365.08	359
9.32	325.09	421.38	368.24	360.1
9.48	325.73	422.45	363.91	360.9
9.64	325.41	422.02	364.76	360.1
9.79	326.16	422.88	365.3	359
9.95	326.27	423.83	364.45	360.3
10.11	326.05	423.3	364.98	360.9
10.26	326.27	422.24	365.08	360.5
10.42	326.05	423.3	365.83	361.1
10.58	326.05	422.77	366.15	360.3
10.74	326.59	422.77	363.91	361.2
10.89	326.27	422.77	365.83	360.6
11.05	326.48	422.45	366.47	361.9
11.21	325.52	422.45	366.04	363.2
11.36	326.16	422.77	366.47	363.2
11.52	326.59	422.45	367.22	362.6
11.68	326.27	422.02	367.11	363
11.83	324.83	423.62	365.72	361.4
11.99	326.8	423.2	366.15	361.5
12.15	326.59	422.56	365.4	363
12.30	326.91	422.56	366.79	362.7
12.46	327.23	422.77	366.9	363.5
12.62	326.91	422.24	368.71	362.3
12.78	326.8	423.2	367.64	362.7
12.93	327.54	423.09	367.11	363.7
13.09	326.8	424.9	368.92	363
13.25	326.91	424.69	366.79	365.1
13.40	326.8	423.52	368.6	363.5
13.56	327.65	421.7	367.85	362.6
13.72	327.23	423.52	369.77	363.5

Table D14 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in peroxide system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
13.87	327.65	423.62	367.85	365.9
14.03	327.33	424.26	368.17	365.8
14.19	327.82	423.83	371.37	365.8
14.35	326.8	422.88	368.28	364.6
14.50	327.54	424.15	369.24	364.1
14.66	328.18	422.77	370.41	364.8
14.82	328.18	423.94	369.35	366.1
14.97	327.54	423.2	369.67	365
15.13	328.18	423.2	369.24	365.5
15.29	328.4	423.52	369.03	365.9
15.44	328.61	423.83	368.92	366.1
15.60	328.18	423.83	369.67	365.9
15.76	328.4	424.15	371.69	366.4
15.92	327.65	423.3	370.31	367
16.07	328.4	424.58	370.73	367
16.23	327.97	424.15	369.99	366.2
16.39	328.18	424.58	372.76	367
16.54	328.18	423.94	371.69	367.2
16.70	328.4	423.94	370.84	367.9
16.86	328.29	423.83	371.8	368.2
17.01	329.46	424.26	372.44	365.4
17.17	327.86	424.15	372.44	368.2
17.33	328.18	424.15	370.73	367.6
17.48	327.54	424.9	371.8	368.3
17.64	328.61	424.58	372.54	369.3
17.80	328.61	424.26	373.18	369.6
17.96	329.68	424.37	372.22	368.3
18.11	328.72	424.9	373.93	367.9
18.27	329.36	425.22	372.54	368.6
18.43	328.61	424.37	373.18	369.7
18.58	329.78	424.37	374.03	368.7
18.74	329.25	425.65	373.18	369.1
18.90	329.04	425.01	373.61	370.8
19.05	329.78	425.22	374.99	370
19.21	330	423.62	375.31	368.9
19.37	330.32	424.37	374.67	368
19.53	329.46	424.37	374.57	369.7

Table D14 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in peroxide system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
19.68	328.29	424.37	376.38	369.7
19.84	329.68	424.9	376.49	369.9
20.00	329.78	424.26	374.25	368.9
20.15	329.68	424.58	375.74	369.9
20.31	329.78	425.75	375.74	369.4
20.47	330.53	424.9	375.95	368
20.62	330.32	424.15	375.42	371.2
20.78	330.74	426.07	375.42	369.6
20.94	330.42	424.58	376.06	369.4
21.09	331.06	424.15	375.42	371
21.25	330.32	425.75	376.38	371.1
21.41	330.32	425.22	377.02	371.4
21.57	330.42	425.75	377.44	370.8
21.72	329.68	425.01	377.44	370.5
21.88	331.24	423.62	377.87	372.1
22.04	330.32	427.14	377.02	371.9
22.19	330.53	425.75	378.51	372.3
22.35	330	424.69	377.44	372.1
22.51	331.06	426.29	378.19	372.5
22.66	330.74	424.58	377.12	372.3
22.82	330.1	425.65	378.94	371.5
22.98	331.49	425.97	377.12	373.1
23.14	331.59	426.5	379.36	374
23.29	331.17	426.5	378.51	373.2
23.45	331.17	424.37	378.08	372.2
23.61	329.78	424.26	379.58	371.9
23.76	331.81	426.39	378.08	374.2
23.92	330	425.22	378.83	374.4
24.08	332.84	424.58	378.08	373.3
24.23	332.13	424.69	379.9	373.2
24.39	331.38	426.29	380.64	373.6
24.55	330.74	426.07	379.36	373.6
24.71	331.17	426.5	380	374.4
24.86	331.06	425.75	380.21	374.4
25.02	330.85	426.29	380	373.9
25.18	331.81	425.65	382.77	373.7
25.33	331.91	425.65	378.94	373.9

Table D14 Time sweep results of 100 phr-FA filled NR vulcanizates with 4% w/w of 3 types of silanes in peroxide system (cont.)

Time (min)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
25.49	331.59	426.29	380.96	376.4
25.65	332.23	426.71	380.75	375.3
25.80	332.55	426.5	380.21	374.7
25.96	332.13	426.39	381.28	375.3
26.12	333.8	426.5	380.75	376.5
26.27	332.13	425.65	382.03	375.7
26.43	331.91	426.39	381.07	375.3
26.59	332.45	426.39	382.45	375.3
26.75	332.98	426.07	382.45	375.4
26.90	332.55	427.14	381.39	376.8
27.06	332.98	426.07	381.28	376.5
27.22	332.87	427.35	383.52	377.4
27.37	332.98	427.46	382.45	377.2
27.53	331.91	427.14	381.71	377.4
27.69	331.59	426.71	382.56	377.5
27.84	332.13	426.39	383.41	377.1
28.00	333.62	426.07	382.77	376.8
28.16	333.19	426.5	385.22	377.2
28.32	332.55	426.07	381.39	376.4
28.47	333.3	426.39	382.45	378.3
28.63	332.55	427.35	384.16	378.5
28.79	333.72	427.56	382.56	377.1
28.94	333.51	426.39	382.77	379
29.10	332.13	425.43	384.48	377.2
29.26	333.19	425.97	383.62	377.9
29.41	333.19	427.14	384.26	378.1
29.57	333.62	426.5	384.69	379
29.73	333.62	427.78	384.58	380.4
29.88	334.36	426.71	386.29	379.2

Table D15 Strain sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV system

Strain (%)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.56	60.399	122.81	92.612	70.302
0.98	53.221	112.57	85.375	54.33
1.95	54.489	109.77	83.158	60.544
5.02	44.877	103.78	79.063	54.28
10.04	43.534	96.593	72.877	48.135
19.95	38.52	85.559	64.896	42.627
49.94	28.882	61.012	46.671	31.797
100.02	19.366	39.287	30.478	21.376
499.97	4.6687	9.2576	6.5983	5.2044
999.94	2.1305	4.2163	2.9917	2.4378
1199.98	1.7547	3.0426	2.3249	1.9638

Table D16 Strain sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in EV system

Strain (%)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.56	62.612	81.43	60.399	64.133
0.98	56.505	82.899	60.754	64.309
1.95	51.516	76.505	69.9	66.598
5.02	47.743	76.292	70.094	56.047
10.04	46.544	67.649	65.393	51.287
19.95	44.18	60.605	56.161	46.255
49.94	39.757	43.897	41.166	34.678
100.02	26.309	29.035	27.478	23.312
499.97	5.8811	7.4481	5.9216	5.4927
999.94	2.6801	3.4637	2.7379	2.5489
1199.98	2.1204	2.5606	2.146	2.0421

Table D17 Strain sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in Semi-EV system

Strain (%)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.56	53.157	86.372	68.585	74.492
0.98	49.895	82.131	64.309	60.982
1.95	46.014	73.179	65.337	59.443
5.02	44.664	69.667	57.486	58.178
10.04	42.562	64.571	55.805	54.449
19.95	38.172	58.007	49.576	47.889
49.94	28.812	42.885	36.801	36.822
100.02	19.676	28.572	24.496	24.386
499.97	4.6514	7.5718	5.4903	5.6957
999.94	2.1496	3.5457	2.5317	2.646
1199.98	1.7734	2.5783	1.9851	2.1248

Table D18 Strain sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system

Strain (%)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.56	70.961	144.96	138.13	86.572
0.98	68.697	126.4	117.53	79.832
1.95	62.646	135.4	117.53	82.56
5.02	60.433	126.37	114.44	75.65
10.04	56.686	117.95	103.25	72.036
19.95	51.378	104.41	91.337	66.522
49.94	38.797	73.922	64.686	48.728
100.02	26.053	46.814	41.389	31.724
499.97	5.8256	10.639	8.8419	7.1796
999.94	2.662	4.7615	3.8523	3.2682
1199.98	2.244	3.3629	2.8602	2.5775

Table D19 Strain sweep results of cured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV system

Strain (%)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.56	1222.2	1141.4	964.37	1169.2
0.84	1127.7	1005.5	889.22	1114.4
1.95	938.33	1014.8	857.5	1079.8
5.02	944.52	1012.9	857.32	1037.8
10.04	933.83	1007.2	854.73	1000.8
19.95	845.51	972.14	826.21	959.54
49.94	706.86	855.92	734.04	812.24
100.02	553.31	643.76	576.02	579.57
200.04	394.18	457.24	434.32	401.32

Table D20 Strain sweep results of cured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in EV system

Strain (%)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.56	996.96	1059.8	910.89	875.78
0.84	810.14	861.62	726.15	793.9
1.95	684.98	831.74	654.66	768.77
5.02	695.97	856.06	666.87	758.06
10.04	691.95	852.82	669.31	741.39
19.95	652.59	827.81	646.43	721.43
49.94	549.09	751.28	572.64	656.42
100.02	432.67	587.11	460.96	511.23
200.04	320.44	447.25	379.96	361.35

Table D21 Strain sweep results of cured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in Semi-EV system

Strain (%)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.56	1232.6	1028.6	918.06	877.9
0.84	1048.06	889.22	775.09	868.33
1.95	899.05	878.73	747.64	825.51
5.02	800.9	864.53	748.7	813.51
10.04	713.91	856.73	742.98	794.06
19.95	658.04	836.86	722.91	771.97
49.94	550.4	757.53	652.56	694.88
100.02	430.84	599.03	521.34	544.63
200.04	315.88	445.91	399.09	389.87

Table D22 Strain sweep results of cured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system

Strain (%)	Storage modulus (kPa)			
	without	Si-69	Si-264	VTEO
0.56	124.78	179.54	169.9	160.86
0.84	119.01	176.95	161.65	150.5
1.95	114.78	174.75	145.62	142.24
5.02	112.04	170.81	145.23	133.83
10.04	110.92	157	140.17	129.2
19.95	104.96	123.41	127.28	118.48
49.94	80.385	87.503	96.484	90.801
100.02	55.395	57.047	66.801	63.333
200.04	33.542	40.047	42.449	40.157

Table D23 Delta storage modulus of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in sulfur and peroxide systems (Payne effect)

Curing systems	Delta G'			
	without	Si-69	Si-264	VTEO
CV	58.64	119.77	90.29	68.34
EV	90.49	78.87	58.25	62.09
Semi-EV	41.38	83.79	66.60	72.37
DCP	68.72	141.60	135.27	83.99

Table D24 Relative storage modulus (relative G') at 1 % strain of of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in sulfur and peroxide system

Curing systems	Relative G'			
	without	Si-69	Si-264	VTEO
Guth-Gold Equation	2.71	2.71	2.71	2.71
CV	1.83	1.63	1.44	1.81
EV	1.49	1.58	1.33	1.46
Semi-EV	1.45	1.23	1.07	1.20
DCP	1.34	1.97	1.82	1.69

Table D25 Frequency sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV system

Frequency rad/s	Storage modulus (kPa)				Damping factor			
	without	Si-69	Si-264	VTEO	without	Si-69	Si-264	VTEO
0.5	41.075	47.607	42.672	34.969	1.298	1.157	1.243	1.254
0.99	58.304	71.128	66.98	54.121	1.075	0.996	1.069	1.071
2	92.579	105.56	100.26	86.84	0.94	0.833	0.886	0.94
5.01	145.75	159.8	155.17	139.44	0.761	0.681	0.708	0.749
10	198.2	208.51	205.14	187.81	0.619	0.577	0.589	0.611
15	229.82	241.32	230.44	216.33	0.565	0.524	0.546	0.552
20	258.34	265.7	260.03	240.23	0.49	0.504	0.509	0.526
25	269.84	278.73	273.61	255.27	0.477	0.469	0.474	0.488
30	284.82	292.38	287.62	270.53	0.454	0.454	0.454	0.47
40	305.25	315.99	309.85	293.14	0.444	0.425	0.427	0.436
50	321.81	335.21	328.41	311.71	0.416	0.402	0.404	0.414
60	336.53	346.96	341.66	323.62	0.398	0.388	0.39	0.401
69.99	348.18	361.17	351.86	335.82	0.382	0.375	0.38	0.387
80	356.67	368.42	361.37	343.66	0.372	0.369	0.368	0.377
90	364.01	379.15	373.43	351.37	0.363	0.349	0.35	0.369
100	372.89	385.6	377.16	361.52	0.353	0.35	0.352	0.355
Complex viscosity (Pa-s)								
Frequency rad/s	without	Si-69	Si-264	VTEO				
0.5	1.34E +05	1.45E +05	1.35E+05	1.17E+05				
0.99	90408	1.01E+05	98546	85381				
2	63522	68692	66975	59575				

Table D25 Frequency sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV system (cont.)

Frequency rad/s	Complex viscosity (Pa-s)			
	without	Si-69	Si-264	VTEO
5.01	36596	38631	37978	34802
10	23313	24073	23805	22012
15	17605	18169	17510	16479
20	14381	14874	14587	13571
25	11959	12314	12115	11365
30	10426	10703	10529	9961.9
40	8349.1	8581.5	8422.4	7994.8
50	6970.2	7226	7084.1	6745.5
60	6035.7	6203	6111.3	5811.5
69.99	5325.4	5511.2	5377.6	5143.8
80	4757.7	4908.6	4813.3	4590.7
90	4302.6	4461.9	4396	4161
100	3954.9	4085.7	3999.1	3836.8

Table D26 Frequency sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in EV system

Frequency rad/s	Storage modulus (kPa)				Damping factor			
	without	Si-69	Si-264	VTEO	without	Si-69	Si-264	VTEO
0.5	31.312	71.933	50.24	31.583	1.389	0.953	1.182	1.271
0.99	49.821	103.29	74.819	50.543	1.199	0.848	1.025	1.101
2	77.739	141.26	111.9	79.43	1.016	0.725	0.843	0.943
5.01	128.18	202.1	171.56	129.04	0.811	0.599	0.673	0.776
10	175.9	255.73	223.08	175.85	0.667	0.505	0.562	0.646
15	206.01	284.86	249.14	201.97	0.617	0.475	0.514	0.607
20	228.14	310.93	273.36	225.08	0.568	0.411	0.469	0.563
25	244.42	327.79	293.14	240.84	0.52	0.406	0.458	0.515
30	258.19	341.13	305.41	255.79	0.493	0.393	0.438	0.487
40	281.9	366.4	331.22	278.06	0.461	0.367	0.411	0.464
50	299.58	384.83	347.72	296.48	0.438	0.353	0.392	0.434
60	311.69	397.09	360.91	310.48	0.421	0.339	0.38	0.419
69.99	324.3	410.43	371.82	320.28	0.401	0.325	0.366	0.411
80	331.11	418.94	382.83	331.73	0.396	0.32	0.356	0.391
90	341.58	428.46	397.8	333.34	0.385	0.307	0.341	0.392
100	348.95	439.6	405.14	350.35	0.371	0.304	0.333	0.363

Table D26 Frequency sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in EV system (cont.)

Frequency rad/s	Complex viscosity (Pa-s)			
	without	Si-69	Si-264	VTEO
0.5	1.07E+05	1.98E+05	1.55E+05	1.12E+05
0.99	74414	1.36E+05	1.08E+05	79383
2	55404	87251	73155	55674
5.01	32970	47070	41311	32635
10	21145	28652	25582	20934
15	16146	21028	18679	15754
20	13117	16805	15095	12913
25	11020	14155	12897	10839
30	9594	12218	11114	9483.1
40	7759.9	9757.5	8953.1	7663.1
50	6539.6	8160.3	7469.1	6463.5
60	5635.2	6988.4	6433.8	5609.3
69.99	4992.3	6165.6	5657.1	4946.7
80	4452.6	5498.2	5080.4	4452.8
90	4067.5	4980.6	4669.7	3978.5
100	3721.8	4594.1	4269.7	3727.8

Table D27 Frequency sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in Semi-EV system

Frequency (rad/s)	Storage modulus (kPa)				Damping factor			
	without	Si-69	Si-264	VTEO	without	Si-69	Si-264	VTEO
0.5	26.919	53.95	34.29	37.103	1.526	1.102	1.32	1.256
0.99	43.695	78.963	53.769	56.38	1.267	0.953	1.151	1.169
2	68.073	113.44	83.769	87.742	1.041	0.801	0.935	0.949
5.01	115.76	169.82	132.03	142.37	0.808	0.639	0.77	0.752
10	158.39	218.63	179.07	192.57	0.679	0.548	0.635	0.612
15	184.75	248.22	205.29	219.86	0.608	0.493	0.567	0.561
20	210.2	268.3	222.62	244.54	0.551	0.448	0.552	0.542
25	220.01	287.01	244.88	261.87	0.532	0.446	0.511	0.489
30	232.58	300.04	257.57	276.95	0.515	0.433	0.483	0.462
40	255.58	324.73	279.65	301.72	0.471	0.405	0.457	0.436
50	272.75	340.3	297.11	318.59	0.447	0.388	0.436	0.415
60	284.29	357.02	310.01	332.34	0.43	0.372	0.419	0.39
69.99	296.21	366.73	323.57	342.28	0.416	0.357	0.405	0.387
80	304.18	375.47	330.25	354.01	0.405	0.347	0.394	0.376
90	312.46	388.23	337.39	371.17	0.394	0.338	0.38	0.355
100	320.13	393.87	345.58	374.25	0.384	0.334	0.376	0.353

Table D27 Frequency sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in Semi-EV system (cont.)

Frequency rad/s	Complex viscosity (Pa-s)			
	without	Si-69	Si-264	VTEO
0.5	97719	1.60E+05	1.13E+05	1.19E+05
0.99	70883	1.10E+05	82426	87181
2	49116	72673	57349	60481
5.01	29729	40253	33288	35584
10	19143	24925	21214	22578
15	14420	18454	15736	16813
20	11997	14699	12715	13908
25	9971.3	12572	11000	11661
30	8721.5	10897	9535.3	10170
40	7062	8756.9	7686.8	8227.7
50	5974.5	7300.9	6482.9	6897.8
60	5157.4	6348.7	5601.4	5944.8
69.99	4583.7	5563.4	4988.1	5243
80	4102.1	4969	4437.4	4728.4
90	3731.7	4553.2	4010.5	4376.9
100	3429.4	4152.7	3692.4	3969.1

Table D28 Frequency sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system

Frequency (rad/s)	Storage modulus (kPa)				Damping factor			
	without	Si-69	Si-264	VTEO	without	Si-69	Si-264	VTEO
0.5	47.165	83.363	70.269	53.248	1.11	0.885	1	1.103
0.99	61.882	116.84	99.441	75.583	1.007	0.771	0.875	0.955
2	91.343	158.43	141.75	110.22	0.848	0.672	0.731	0.791
5.01	139.54	220.74	203.81	163.63	0.686	0.536	0.598	0.635
10	182.1	273.06	257.42	210.35	0.571	0.456	0.495	0.532
15	210.35	304.03	290.23	238.7	0.514	0.407	0.439	0.487
20	226.29	324.88	305.87	259.11	0.497	0.411	0.413	0.434
25	242.57	343.51	325.95	275.05	0.445	0.372	0.401	0.426
30	253.13	354.47	340.67	286.4	0.431	0.362	0.388	0.411
40	273.06	375.93	362.4	310.16	0.404	0.342	0.363	0.393
50	288.97	394.33	379	328.39	0.383	0.327	0.346	0.363
60	298.66	407.26	392.2	336.99	0.365	0.316	0.334	0.352
69.99	307.86	416.87	405.72	344.89	0.357	0.304	0.32	0.344
80	316.29	425.06	412.06	356.67	0.347	0.299	0.313	0.33
90	324.46	436.14	420.93	364.09	0.342	0.291	0.305	0.313
100	330.86	439.03	428	375	0.329	0.294	0.302	0.317

Table D28 Frequency sweep results of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system (cont.)

Frequency rad/s	Complex viscosity (Pa-s)			
	without	Si-69	Si-264	VTEO
0.5	1.33E+05	2.21E+05	1.98E+05	1.58E+05
0.99	88296	1.48E+05	1.33E+05	1.05E+05
2	59869	95413	87796	70242
5.01	33803	50025	47445	38723
10	20965	30014	28720	23825
15	15771	21892	21137	17701
20	12634	17561	16545	14120
25	10623	14660	14050	11960
30	9186	12565	12179	10321
40	7361.2	9931.3	9638.6	8330.6
50	6188.9	8296	8021	6986.7
60	5298.5	7118.2	6891.4	5953.3
69.99	4670.3	6225.1	6086.6	5211.1
80	4185.2	5546.2	5397.5	4695.5
90	3810.1	5046.7	4889.9	4239.2
100	3483.6	4575.7	4470.9	3933.6

Table D29 Slope calculated from the plots of storage modulus (G') as a function of frequency of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide system

Curing systems	Slope			
	without	Si-69	Si-264	VTEO
CV	0.533	0.493	0.521	0.564
EV	0.576	0.42	0.499	0.572
Semi-EV	0.592	0.467	0.55	0.553
DCP	0.461	0.394	0.434	0.461

Table D30 Power law index of uncured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in sulfur and peroxide systems

Curing systems	Power law index			
	without	Si-69	Si-264	VTEO
CV	0.328	0.315	0.32	0.34
EV	0.356	0.276	0.308	0.345
Semi-EV	0.356	0.3	0.34	0.343
DCP	0.305	0.257	0.273	0.293

Table D31 Frequency sweep results of cured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in CV systems

Frequency (rad/s)	Storage modulus (kPa)				Damping factor			
	without	Si-69	Si-264	VTEO	without	Si-69	Si-264	VTEO
0.5	932.66	996.81	830.39	981.99	0.025	0	0.01	0.005
0.99	932.29	1013.4	834.38	983.75	0.028	0.003	0.011	0.004
2	932.01	1012.4	836.64	978.53	0.029	0.006	0.013	0.006
5.01	932.5	1010.7	840.02	984.56	0.032	0.008	0.019	0.011
10	932.32	1008.5	843.09	980.62	0.032	0.01	0.023	0.01
15	931.71	1010.7	847.69	975.86	0.038	0.009	0.024	0.012
20	924.96	1008.1	848.46	975.4	0.036	0.007	0.02	0.009
25	931.86	1006.5	848.46	980.43	0.035	0.014	0.027	0.016
30	933.24	1009.6	850.6	984.43	0.035	0.017	0.03	0.025
40	938.15	1007	855.05	980.74	0.036	0.016	0.03	0.015
50	944.95	1015.5	861.8	990.26	0.034	0.019	0.034	0.016
60	944.13	1016.8	862.26	976.89	0.038	0.022	0.029	0.017
69.99	945.51	1009	861.8	976.28	0.037	0.024	0.039	0.022
80	944.89	1008.6	867.16	984.32	0.04	0.025	0.037	0.035
90	949.56	1018.4	869.82	985.14	0.04	0.025	0.04	0.03
100	947.65	1008.3	865.89	993.04	0.042	0.033	0.045	0.025

Table D32 Frequency sweep results of cured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in EV system

Frequency (rad/s)	Storage modulus (kPa)				Damping factor			
	without	Si-69	Si-264	VTEO	without	Si-69	Si-264	VTEO
0.5	697.05	841.71	657.2	721.27	0.005	0.001	0.013	0.001
0.99	701.86	845.54	663.81	720.49	0.009	0.002	0.013	0.001
2	698.97	843.55	667.77	717.05	0.012	0.004	0.015	0.003
5.01	701.45	842.33	669.31	721.95	0.017	0.012	0.02	0.01
10	700.51	840.64	672.14	721.42	0.019	0.01	0.024	0.014
15	702.5	840.48	674.87	720.65	0.022	0.011	0.031	0.02
20	695.91	842.63	681.95	718.66	0.023	0.021	0.029	0.012
25	701.43	840.94	679.65	721.27	0.024	0.016	0.035	0.021
30	702.19	841.86	679.96	724.03	0.022	0.018	0.037	0.034
40	705.26	847.09	683.33	721.97	0.023	0.018	0.038	0.028
50	709.09	850.14	688.85	727.18	0.027	0.023	0.043	0.026
60	711.54	850.6	692.84	726.72	0.024	0.019	0.046	0.03
69.99	711.39	848.3	692.38	721.23	0.023	0.023	0.04	0.036
80	712.62	849.36	700.4	721.92	0.024	0.026	0.047	0.038
90	715.38	840.95	687.18	708.09	0.025	0.035	0.06	0.061
100	715.07	862.07	701.3	736.88	0.028	0.027	0.046	0.036

Table D33 Frequency sweep results cured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in semi-EV systems

Frequency (rad/s)	Storage modulus (kPa)				Damping factor			
	without	Si-69	Si-264	VTEO	without	Si-69	Si-264	VTEO
0.5	717.08	860.08	758.15	782.02	0.012	0.009	0.003	0.004
0.99	716.55	865.58	771.02	783.07	0.015	0.009	0.005	0.004
2	710.69	861.38	764.44	780.66	0.017	0.009	0.008	0.005
5.01	713.28	858.61	763.52	780.14	0.021	0.014	0.013	0.006
10	712.46	860.42	767.2	775.78	0.022	0.014	0.016	0.008
15	711.19	861.18	772.1	774.42	0.021	0.017	0.019	0.013
20	707.87	854.13	774.1	771.66	0.025	0.02	0.027	0.014
25	711.65	862.41	771.49	774.86	0.027	0.017	0.024	0.015
30	714	862.26	771.8	778.26	0.026	0.019	0.024	0.016
40	717.16	867.98	778.39	775.94	0.035	0.02	0.03	0.023
50	721.82	862.06	776.68	785.57	0.035	0.024	0.033	0.021
60	726.03	867.57	783.6	782.38	0.035	0.025	0.033	0.025
69.99	722.36	866.8	776.66	788.3	0.038	0.028	0.036	0.025
80	725.8	872.68	790.97	781.24	0.039	0.026	0.034	0.034
90	728.34	869.66	788.14	763.4	0.038	0.028	0.038	0.049
100	723.36	886.38	784.29	785.79	0.045	0.025	0.046	0.03

Table D34 Frequency sweep results of cured 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in peroxide systems

Frequency (rad/s)	Storage modulus (kPa)				Damping factor			
	without	Si-69	Si-264	VTEO	without	Si-69	Si-264	VTEO
0.5	100.03	205.77	169.91	111.97	0.428	0.211	0.266	0.403
0.99	121.43	226.76	190.27	131.41	0.443	0.227	0.283	0.417
2	145.5	249.68	213.39	155.15	0.447	0.243	0.301	0.429
5.01	184.41	282.62	249.03	196.11	0.43	0.26	0.313	0.421
10	220.3	316.16	283.79	231.67	0.405	0.267	0.311	0.402
15	243.34	333.37	307.09	255.17	0.39	0.271	0.318	0.402
20	258.65	357.54	323.96	274.74	0.403	0.259	0.321	0.395
25	274.53	364.28	336.38	289.62	0.37	0.267	0.306	0.366
30	285.52	375.47	348.18	300.19	0.363	0.265	0.303	0.365
40	304.95	392.8	367.78	318.26	0.349	0.26	0.302	0.347
50	319.67	408.9	384.06	337.24	0.337	0.256	0.289	0.335
60	332.39	420.01	395.89	352.97	0.329	0.257	0.286	0.331
69.99	341.13	431.84	407.67	363.09	0.323	0.249	0.285	0.328
80	350.48	437.57	415.64	370.26	0.319	0.251	0.28	0.328
90	357.95	447.38	421.47	373.76	0.312	0.25	0.278	0.312
100	367.47	452.76	435.68	379.5	0.312	0.249	0.275	0.319

Table D35 Retention of storage modulus (G') of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in sulfur and peroxide systems as measured from RPA 2000

Curing systems	Unaged storage modulus (kPa)				Aged storage modulus (kPa)			
	without	Si-69	Si-264	VTEO	without	Si-69	Si-264	VTEO
CV	918.8	920	746.24	919.9	984.15	960.78	778.08	935.7
EV	681.65	858.67	749.62	915.49	726.00	918.59	786.35	960.52
Semi-EV	732.54	1043.4	884.33	919.44	755.24	1072.1	906.92	940.83
DCP	121.79	192.43	198.76	199.51	145.87	238.49	232.82	221.94
Retention of storage modulus								
Curing systems	without		Si-69	Si-264	VTEO			
CV	107.11		104.43	104.27	101.72			
EV	106.51		106.98	104.90	104.92			
Semi-EV	103.10		102.75	102.55	102.33			
DCP	119.77		123.94	117.14	111.24			

Table D36 Modulus at 100 % strain (M100) of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in sulfur and peroxide systems

Curing systems	M100 (MPa)			
	without	Si-69	Si-264	VTEO
CV	1.04±0.06	1.32±0.03	1.41±0.07	1.17±0.02
EV	0.91±0.02	1.37±0.06	1.24±0.06	0.93±0.02
Semi-EV	0.89±0.01	1.34±0.10	1.42±0.07	0.93±0.02
DCP	0.48±0.03	0.63±0.01	0.50±0.03	0.63±0.01

Table D37 Elongation at break of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in sulfur and peroxide systems

Curing systems	Elongation at break (%)			
	without	Si-69	Si-264	VTEO
CV	561.31±38.53	509.14±21.13	529.85±38.60	632.8±34.82
EV	588.27±21.83	498.78±18.10	523.08±30.61	688.42±12.01
Semi-EV	610.95±13.46	504.8±34.65	473.77±34.27	692.18±12.22
DCP	585.63±25.34	582.53±15.84	578.12±36.47	600.48±19.82

Table D38 Tensile strength of 100 phr FA /NR compounds with 4% w/w of 3 types of silanes in sulfur and peroxide systems

Curing systems	Tensile strength (MPa)			
	without	Si-69	Si-264	VTEO
CV	10.53±0.52	10.7±0.65	10.12±0.74	9.58±0.52
EV	9.82±0.39	9.96±0.63	10.12±0.65	9.01±0.27
Semi-EV	11.26±0.30	11.75±0.96	11.52±1.06	10.9±0.35
DCP	2.86±0.73	5.67±0.33	2.66±0.47	3.25±0.13

APPENDIX E

VISCOELASTIC AND MECHANICAL PROPERTIES DATA OF NR FILLED WITH FA FROM MAE MOH (MMFA) AND ROCKTRON (RK) IN PEROXIDE SYSTEM

Table E1 Particle size distribution of RKFA

Size (micron)	Volume (%)	Size (micron)	Volume (%)	Size (micron)	Volume (%)
0.375	0.17	8.147	3.72	176.8	0
0.412	0.302	8.944	3.73	194.2	0
0.452	0.442	9.819	3.69	213.2	0
0.496	0.621	10.78	3.59	234.1	0
0.545	0.759	11.83	3.45	256.8	0
0.598	0.869	12.99	3.25	282.1	0
0.657	0.958	14.26	2.99	309.6	0
0.721	1.03	15.65	2.67	339.8	0
0.791	1.08	17.18	2.32	373.1	0
0.869	1.1	18.86	2.01	409.6	0
0.953	1.1	20.7	1.79	449.7	0
1.047	1.09	22.73	1.72	493.6	0
1.149	1.08	24.95	1.77	541.9	0
1.261	1.07	27.38	1.85	594.9	0
1.385	1.06	30.07	1.84	653	0
1.52	1.08	33	1.66	716.9	0
1.669	1.12	36.24	1.3	786.9	0
1.832	1.19	39.77	0.836	863.9	0
2.01	1.29	43.66	0.405	948.2	0
2.207	1.43	47.93	0.131		
2.423	1.6	52.63	0.021		
2.66	1.8	57.77	0.001		
2.92	2.02	63.41	0		
3.206	2.25	69.62	0		
3.519	2.48	76.43	0		
3.862	2.71	83.9	0		
4.241	2.92	92.09	0		
4.656	3.11	101.1	0		
5.111	3.28	111	0		
5.611	3.41	121.8	0		
6.158	3.52	133.7	0		
6.761	3.61	146.8	0		
7.421	3.68	161.2	0		

Table E2 Cure behavior results of NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system

Time (min)	Torque (dNm)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
0.383	1.37	2.003	1.341	2.009	2.868
0.68	1.261	1.827	1.263	1.851	2.781
0.98	1.27	1.887	1.313	1.97	2.9
1.28	1.317	1.95	1.395	2.138	3.018
1.58	1.361	2.027	1.478	2.331	3.151
1.88	1.418	2.114	1.564	2.538	3.295
2.18	1.464	2.205	1.657	2.745	3.448
2.48	1.516	2.302	1.761	2.949	3.591
2.78	1.571	2.4	1.862	3.156	3.746
3.08	1.626	2.497	1.967	3.353	3.888
3.38	1.686	2.6	2.07	3.538	4.024
3.68	1.741	2.7	2.174	3.719	4.169
3.98	1.793	2.798	2.279	3.896	4.31
4.28	1.849	2.895	2.38	4.072	4.443
4.58	1.896	2.991	2.485	4.234	4.573
4.88	1.957	3.096	2.582	4.395	4.71
5.18	2.006	3.193	2.696	4.548	4.839
5.48	2.063	3.293	2.795	4.698	4.956
5.78	2.106	3.39	2.9	4.847	5.076
6.08	2.16	3.488	3.004	4.992	5.193
6.38	2.21	3.589	3.096	5.126	5.308
6.68	2.261	3.687	3.21	5.273	5.419
6.98	2.308	3.777	3.304	5.406	5.541
7.28	2.363	3.871	3.408	5.54	5.648
7.58	2.413	3.964	3.507	5.672	5.75
7.88	2.466	4.054	3.603	5.802	5.86
8.18	2.508	4.147	3.688	5.92	5.966
8.48	2.544	4.238	3.787	6.044	6.056
8.78	2.587	4.333	3.876	6.16	6.143
9.08	2.636	4.422	3.967	6.273	6.24
9.38	2.676	4.501	4.057	6.388	6.33
9.68	2.722	4.591	4.15	6.502	6.425
9.98	2.762	4.667	4.239	6.607	6.51
10.28	2.801	4.757	4.337	6.715	6.586
10.58	2.852	4.85	4.416	6.822	6.671
10.88	2.885	4.929	4.505	6.92	6.763
11.18	2.922	5.002	4.588	7.026	6.837

Table E2 Cure behavior results of NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system (cont.)

Time (min)	Torque (dNm)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
11.48	2.964	5.084	4.666	7.116	6.918
11.78	2.993	5.166	4.749	7.211	6.988
12.08	3.034	5.243	4.827	7.311	7.07
12.38	3.072	5.32	4.906	7.409	7.142
12.68	3.103	5.397	4.99	7.502	7.213
12.98	3.144	5.469	5.061	7.586	7.284
13.28	3.172	5.553	5.142	7.68	7.355
13.58	3.21	5.622	5.213	7.756	7.426
13.88	3.24	5.702	5.289	7.845	7.491
14.18	3.267	5.773	5.351	7.921	7.561
14.48	3.3	5.832	5.432	7.999	7.615
14.78	3.335	5.908	5.504	8.084	7.685
15.08	3.361	5.969	5.571	8.155	7.739
15.38	3.39	6.041	5.638	8.226	7.801
15.68	3.423	6.104	5.707	8.304	7.861
15.98	3.441	6.162	5.772	8.371	7.921
16.28	3.474	6.228	5.834	8.442	7.978
16.58	3.494	6.285	5.892	8.5	8.032
16.88	3.53	6.341	5.968	8.563	8.086
17.18	3.549	6.405	6.018	8.626	8.143
17.48	3.583	6.467	6.085	8.69	8.196
17.78	3.603	6.519	6.14	8.75	8.245
18.08	3.64	6.583	6.196	8.802	8.296
18.38	3.651	6.629	6.249	8.865	8.351
18.68	3.68	6.69	6.316	8.914	8.4
18.98	3.705	6.739	6.365	8.969	8.454
19.28	3.728	6.796	6.416	9.017	8.492
19.58	3.749	6.852	6.47	9.068	8.537
19.88	3.764	6.901	6.526	9.114	8.586
20.18	3.788	6.952	6.569	9.157	8.633
20.48	3.807	7.009	6.627	9.199	8.676
20.78	3.827	7.054	6.658	9.251	8.713
21.08	3.848	7.098	6.715	9.286	8.759
21.38	3.864	7.146	6.772	9.327	8.8
21.68	3.88	7.187	6.81	9.367	8.85
21.98	3.902	7.228	6.852	9.404	8.888
22.28	3.922	7.276	6.896	9.439	8.932
22.58	3.935	7.317	6.945	9.478	8.974

Table E2 Cure behavior results of NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system (cont.)

Time (min)	Torque (dNm)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
22.88	3.952	7.36	6.994	9.504	9.01
23.18	3.972	7.405	7.033	9.54	9.041
23.48	3.988	7.434	7.075	9.569	9.082
23.78	4.006	7.487	7.121	9.595	9.118
24.08	4.028	7.521	7.16	9.619	9.147
24.38	4.044	7.555	7.204	9.651	9.19
24.68	4.057	7.604	7.235	9.67	9.229
24.98	4.069	7.642	7.281	9.693	9.256
25.28	4.084	7.675	7.313	9.727	9.301
25.58	4.101	7.701	7.351	9.737	9.325
25.88	4.114	7.743	7.393	9.773	9.365
26.18	4.128	7.775	7.425	9.782	9.399
26.48	4.15	7.803	7.463	9.805	9.428
26.78	4.158	7.841	7.486	9.822	9.455
27.08	4.169	7.877	7.523	9.837	9.488
27.38	4.179	7.906	7.567	9.855	9.512
27.68	4.194	7.937	7.592	9.871	9.546
27.98	4.208	7.96	7.63	9.89	9.579
28.28	4.217	7.996	7.661	9.904	9.609
28.58	4.224	8.029	7.692	9.916	9.629
28.88	4.236	8.044	7.723	9.934	9.667
29.18	4.248	8.081	7.75	9.947	9.69
29.48	4.264	8.098	7.787	9.954	9.723
29.78	4.269	8.129	7.812	9.964	9.741
30.08	4.278	8.163	7.841	9.973	9.775
30.38	4.284	8.19	7.871	9.976	9.792
30.68	4.295	8.215	7.899	10	9.826
30.98	4.31	8.231	7.925	10	9.85
31.28	4.319	8.256	7.951	10.01	9.868
31.58	4.324	8.286	7.983	10.02	9.868
31.88	4.335	8.312	8.005	10.02	9.868
32.18	4.338	8.331	8.031	10.03	9.937
32.48	4.352	8.349	8.057	10.04	9.96
32.78	4.361	8.378	8.082	10.04	9.985
33.08	4.372	8.393	8.104	10.04	10.01
33.38	4.38	8.418	8.131	10.05	10.02
33.68	4.383	8.438	8.155	10.06	10.05
33.98	4.394	8.448	8.173	10.06	10.06

Table E2 Cure behavior results of NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system (cont.)

Time (min)	Torque (dNm)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
34.28	4.396	8.473	8.198	10.06	10.06
34.58	4.405	8.498	8.226	10.06	10.06
34.88	4.41	8.512	8.241	10.07	10.07
35.18	4.425	8.528	8.266	10.07	10.07
35.48	4.43	8.549	8.281	10.08	10.07
35.78	4.432	8.569	8.301	10.07	10.07
36.08	4.445	8.58	8.326	10.08	10.07
36.38	4.451	8.601	8.345	10.08	10.07
36.68	4.454	8.621	8.364	10.09	10.08
36.98	4.462	8.632	8.384	10.08	10.08
37.28	4.471	8.648	8.408	10.08	10.08
37.58	4.475	8.67	8.426	10.08	10.08
37.88	4.489	8.68	8.451	10.08	10.09
38.18	4.491	8.698	8.461	10.08	10.09
38.48	4.493	8.71	8.481	10.08	10.09
38.78	4.5	8.73	8.504	10.08	10.09
39.08	4.499	8.74	8.514	10.08	10.09
39.38	4.512	8.752	8.533	10.08	10.1
39.68	4.515	8.766	8.552	10.08	10.1
39.98	4.517	8.782	8.574	10.07	10.1
40.28	4.514	8.789	8.58	10.07	10.1
40.58	4.527	8.802	8.6	10.07	10.1
40.88	4.529	8.83	8.618	10.06	10.09
41.18	4.543	8.841	8.634	10.06	10.09
41.48	4.539	8.842	8.649	10.07	10.09
41.78	4.545	8.85	8.664	10.06	10.09
42.08	4.546	8.862	8.679	10.06	10.08
42.38	4.552	8.884	8.69	10.05	10.08
42.68	4.56	8.89	8.706	10.06	10.08
42.98	4.568	8.904	8.722	10.05	10.08
43.28	4.571	8.912	8.731	10.05	10.08
43.58	4.571	8.915	8.741	10.04	10.08
43.88	4.577	8.928	8.756	10.04	10.08
44.18	4.576	8.932	8.773	10.04	10.07
44.48	4.577	8.947	8.786	10.03	10.07
44.78	4.585	8.952	8.796	10.03	10.07
45.08	4.595	8.963	8.808	10.02	10.07
45.38	4.596	8.981	8.823	10.02	10.07

Table E2 Cure behavior results of NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system (cont.)

Time (min)	Torque (dNm)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
45.68	4.601	8.983	8.84	10.01	10.07
45.98	4.6	8.996	8.848	10.01	10.07
46.28	4.609	8.997	8.857	10	10.07
46.58	4.617	9.009	8.87	9.993	10.06
46.88	4.607	9.019	8.886	9.996	10.06
47.18	4.614	9.025	8.893	9.989	10.07
47.48	4.616	9.032	8.901	9.992	10.07
47.78	4.623	9.042	8.918	9.985	10.07
48.08	4.621	9.043	8.931	9.972	10.06
48.38	4.626	9.053	8.945	9.971	10.06
48.68	4.624	9.064	8.951	9.973	10.06
48.98	4.633	9.068	8.961	9.967	10.07
49.28	4.634	9.072	8.965	9.967	10.07
49.58	4.634	9.081	8.982	9.966	10.07
49.88	4.637	9.084	8.996	9.958	10.07
50.18	4.633	9.088	8.995	9.954	10.07
50.48	4.643	9.093	9.012	9.936	10.07
50.78	4.648	9.097	9.019	9.945	10.07
51.08	4.649	9.104	9.032	9.937	10.06
51.38	4.648	9.106	9.035	9.935	10.06
51.68	4.649	9.111	9.051	9.922	10.07
51.98	4.638	9.114	9.064	9.916	10.07
52.28	4.648	9.132	9.055	9.915	10.07
52.58	4.649	9.133	9.072	9.9	10.07
52.88	4.659	9.133	9.075	9.903	10.07
53.18	4.663	9.143	9.082	9.9	10.07
53.48	4.658	9.144	9.088	9.895	10.07
53.78	4.659	9.146	9.109	9.887	10.08
54.08	4.661	9.149	9.114	9.886	10.08
54.38	4.663	9.149	9.125	9.88	10.08
54.68	4.668	9.159	9.132	9.866	10.08
54.98	4.672	9.163	9.139	9.873	10.07
55.28	4.675	9.165	9.138	9.865	10.07
55.58	4.673	9.169	9.148	9.865	10.08
55.88	4.674	9.174	9.159	9.854	10.07
56.18	4.676	9.182	9.159	9.855	10.07
56.48	4.678	9.186	9.182	9.848	10.07
56.78	4.674	9.187	9.182	9.847	10.08

Table E2 Cure behavior results of NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system (cont.)

Time (min)	Torque (dNm)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
57.08	4.679	9.191	9.192	9.848	10.07
57.38	4.679	9.19	9.186	9.847	10.08
57.68	4.685	9.199	9.196	9.838	10.08
57.98	4.685	9.206	9.206	9.837	10.07
58.28	4.684	9.198	9.217	9.83	10.07
58.58	4.685	9.203	9.226	9.824	10.07
58.88	4.685	9.206	9.231	9.821	10.07
59.18	4.684	9.208	9.235	9.815	10.07
59.48	4.688	9.211	9.238	9.814	10.07

Table E3 Marching rate of NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system

	Marching rate (dNm/min)
NR	0.012
NR+MMFA	0.044
NR+RKFA	0.032
NR+MMFA Si-69	0
NR+RKFA Si-69	0.002

Table E4 Scorch time, cure time, torque difference and crosslink density of NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system

	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
Scorch time (min)	11.45±0.10	6.25±0.20	4.01±0.30	6.43±0.30	3.86±0.30
Cure time (min)	31.28±0.30	31.78±0.20	32.01±0.60	35.49±0.50	33.84±0.40
Torque difference (dNm)	3.78±0.50	7.26±0.60	7.32±0.40	8.88±0.40	9.00±0.70
Crosslink density (x10 ⁻⁴ mole/dm ³)	0.27	1.09	1.14	1.72	1.64

Table E5 Time sweep results of uncured NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system

Time (min)	Storage modulus (kPa)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
0.22	102.25	238.44	200.63	234.86	228.01
0.38	106.04	217.48	197.9	231.87	226.12
0.53	107.41	210.66	194.62	231.23	224.41
0.69	108.15	206.18	192.39	229.3	220.88
0.85	107.73	204.8	191.43	229.09	220.14
1.00	107.74	202.45	189.94	228.77	217.68
1.16	107.74	200.75	188.98	227.7	216.57
1.32	109.11	197.98	187.7	226.11	216.89
1.47	108.15	197.12	185.67	225.25	216.03
1.63	108.36	196.16	185.67	224.61	214.97
1.79	108.9	195.74	185.25	225.68	213.58
1.95	108.9	192.86	184.5	223.12	213.37
2.10	108.36	193.07	183.22	223.23	212.84
2.26	108.68	192.22	182.8	224.51	212.2
2.42	108.9	191.58	182.16	222.7	211.45
2.57	108.79	192.01	182.05	221.31	210.71
2.73	108.79	191.26	181.73	223.44	210.49
2.89	108.9	190.84	181.09	221.63	211.45
3.04	109.11	190.09	180.99	222.16	210.81
3.20	109.11	189.77	179.6	220.78	209.43
3.36	108.79	188.07	178.96	221.84	209.43
3.51	109.11	189.66	179.28	220.25	209.43
3.67	109.43	189.35	179.17	221.1	208.58
3.83	109.75	187.75	179.28	219.5	208.26
3.99	108.9	187.21	178.85	220.78	208.04
4.14	109.22	188.71	179.17	218.54	206.76
4.30	109.86	187.21	178.85	220.57	206.44
4.46	109.43	185.83	178.11	218.54	208.04
4.61	109.43	185.08	177.26	220.03	207.94
4.77	109.43	185.94	177.58	218.65	207.94
4.93	109.54	186.26	177.15	218.43	207.51
5.08	109.43	185.19	176.83	219.61	207.19
5.24	109.11	184.76	177.04	217.37	206.87
5.40	109.75	184.02	176.83	217.58	205.81
5.56	109.43	184.55	176.19	218.43	205.81
5.71	109.22	185.4	176.19	218.11	207.3
5.87	109.75	183.7	175.98	217.37	206.55
6.03	109.22	183.7	175.76	216.41	206.55
6.18	109.54	183.7	176.72	218.54	205.91

Table E5 Time sweep results of uncured NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system (cont.)

Time (min)	Storage modulus (kPa)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
6.34	109.54	183.7	176.83	215.98	205.7
6.50	109.54	182.63	176.4	217.48	205.38
6.65	109.54	183.27	175.76	217.37	204.85
6.81	109.43	182.63	175.45	215.24	204.74
6.97	109.22	182.95	175.02	216.73	204.74
7.13	109.86	183.06	174.91	216.09	204.74
7.28	109.86	182.31	175.02	215.77	205.06
7.44	109.54	182.31	175.02	216.09	204.74
7.60	109.54	182.63	174.91	214.6	204.85
7.75	109.96	181.89	174.27	216.84	203.67
7.91	109.43	181.89	174.38	215.02	204.85
8.07	109.11	181.57	175.76	216.41	204.74
8.22	109.86	180.82	175.45	215.02	203.03
8.38	109.96	181.35	175.66	214.7	203.35
8.54	109.96	181.57	174.06	214.7	204.42
8.69	109.86	181.57	174.06	214.6	204.42
8.85	110.18	181.14	173.53	214.6	203.99
9.01	109.96	181.57	174.27	214.17	203.25
9.17	110.5	180.93	175.45	213.53	203.67
9.32	110.18	181.35	173.95	215.02	201.97
9.48	110.6	181.14	173.53	213.21	202.5
9.64	110.28	179.86	173.95	213.53	202.72
9.79	110.6	179.86	174.27	213.21	201.97
9.95	110.18	179.22	174.38	213.85	202.5
10.11	109.96	180.08	174.38	212.57	203.03
10.26	109.75	180.08	173.95	213.85	203.25
10.42	110.18	179.86	173.63	213.11	201.65
10.58	110.6	179.01	173.95	212.79	201.86
10.74	110.5	179.22	173.21	212.89	202.5
10.89	110.18	179.54	173.21	211.72	203.03
11.05	110.18	179.01	172.78	213.64	202.29
11.21	110.18	179.01	171.93	210.98	201.22
11.36	110.5	178.69	173.21	213.21	201.54
11.52	109.86	178.69	172.46	210.98	202.61
11.68	110.18	179.12	172.14	213.21	201.54
11.83	109.96	179.01	172.89	211.08	201.12
11.99	109.96	179.86	173.53	212.89	201.65
12.15	110.18	178.8	171.18	211.19	202.18
12.30	109.75	178.8	172.57	211.83	201.97

Table E5 Time sweep results of uncured NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system (cont.)

Time (min)	Storage modulus (kPa)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
12.46	110.28	179.12	172.89	210.98	200.16
12.62	109.86	178.16	172.89	211.51	200.48
12.78	109.96	178.48	171.08	209.59	201.86
12.93	110.92	180.18	171.18	211.4	201.54
13.09	109.75	178.16	171.5	210.98	200.9
13.25	111.03	177.3	171.4	210.02	200.9
13.40	109.75	177.62	171.5	211.51	200.9
13.56	110.18	178.69	171.5	211.19	200.9
13.72	109.96	177.73	171.08	209.7	200.16
13.87	110.18	177.3	170.76	210.98	200.48
14.03	111.03	177.62	171.82	210.02	199.84
14.19	110.6	176.56	171.93	208.95	200.9
14.35	109.86	177.3	171.82	210.02	200.58
14.50	110.28	177.62	172.78	210.66	200.05
14.66	111.24	177.73	172.46	208.95	200.37
14.82	109.11	177.73	171.18	210.02	199.84
14.97	111.24	177.41	170.01	210.66	200.48
15.13	109.54	177.41	170.33	210.02	200.8
15.29	110.28	177.3	170.33	209.27	200.37
15.44	110.28	176.67	170.33	209.7	200.37
15.60	110.5	176.35	170.44	210.34	199.52
15.76	109.96	176.24	170.33	209.38	200.16
15.92	110.5	176.03	170.86	208.21	199.84
16.07	110.6	175.92	171.08	208.52	198.67
16.23	110.5	175.81	170.76	208.84	199.73
16.39	109.96	176.56	169.58	209.38	199.31
16.54	109.54	175.6	170.65	210.12	200.05
16.70	109.96	175.17	170.65	207.78	199.41
16.86	110.92	175.6	170.65	208.84	199.84
17.01	111.03	174.96	169.27	208.21	198.99
17.17	110.82	174.75	170.44	209.06	198.03
17.33	110.5	174.96	169.8	207.57	198.67
17.48	110.5	175.81	168.52	208.95	198.77
17.64	110.18	175.49	171.4	206.93	197.92
17.80	110.92	175.17	168.73	208.95	198.67
17.96	110.28	174.43	169.69	207.57	198.67
18.11	110.18	174.85	170.12	207.25	198.67
18.27	110.28	173.89	170.01	208.95	198.67
18.43	109.75	174.21	169.05	206.82	198.77

Table E5 Time sweep results of uncured NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system (cont.)

Time (min)	Storage modulus (kPa)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
18.58	110.28	174.53	168.95	207.46	198.77
18.74	110.18	174.53	168.31	207.46	198.35
18.90	110.5	174.11	167.99	205.65	198.35
19.05	110.28	174.75	168.95	208.63	198.35
19.21	110.6	173.68	169.69	206.71	197.92
19.37	111.03	172.72	170.44	207.14	198.67
19.53	111.56	174.96	169.27	207.78	198.35
19.68	111.24	172.08	169.8	207.89	197.39
19.84	111.24	174.53	169.27	206.18	197.71
20.00	111.99	173.36	169.37	206.5	197.92
20.15	111.03	173.36	168.52	207.14	197.28
20.31	110.18	173.36	167.88	206.71	197.39
20.47	110.5	172.72	169.37	205.75	196.96
20.62	109.75	172.72	170.01	207.46	197.71
20.78	110.18	172.72	168.2	206.82	196.96
20.94	110.6	173.04	168.63	204.58	196.54
21.09	110.6	172.3	169.05	205.75	196.85
21.25	110.5	173.36	167.24	206.39	196.64
21.41	110.82	172.08	169.27	205.01	197.28
21.57	109.96	173.47	169.27	206.18	197.17
21.72	110.6	172.4	168.95	206.18	197.17
21.88	110.5	172.4	169.37	204.69	196.54
22.04	110.92	172.08	168.52	205.33	196.64
22.19	110.18	171.66	169.27	205.86	196.54
22.35	110.5	172.08	168.73	204.37	196.54
22.51	111.24	172.4	167.99	206.71	195.9
22.66	110.5	173.04	168.52	206.39	195.9
22.82	111.88	172.08	168.73	204.69	196.11
22.98	111.24	171.02	167.56	204.05	196.32
23.14	111.24	171.23	167.24	205.65	196.22
23.29	110.82	171.55	167.88	204.8	195.79
23.45	110.5	170.49	168.31	203.94	195.9
23.61	110.92	170.59	168.31	205.75	196.22
23.76	110.5	171.55	167.99	204.05	195.15
23.92	111.35	171.76	167.13	205.01	195.58
24.08	111.35	171.66	166.6	205.12	195.9
24.23	110.6	171.34	167.56	203.3	195.58
24.39	111.35	170.59	168.31	204.8	195.04
24.55	111.24	170.49	168.31	204.8	195.15

Table E5 Time sweep results of uncured NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system (cont.)

Time (min)	Storage modulus (kPa)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
24.71	111.56	171.23	167.56	203.62	195.26
24.86	111.03	171.76	167.56	204.69	196.22
25.02	110.5	170.91	168.2	203.73	195.58
25.18	110.6	170.49	166.81	203.3	195.26
25.33	111.24	169.85	166.92	204.69	195.47
25.49	110.92	171.55	167.13	204.26	194.72
25.65	111.03	169.21	167.24	203.3	194.4
25.80	111.88	169.63	166.81	204.05	194.83
25.96	110.82	170.91	167.56	205.33	194.08
26.12	109.96	170.7	167.13	202.66	194.51
26.27	111.67	170.49	166.49	203.2	195.04
26.43	111.03	171.02	167.24	204.26	193.98
26.59	111.03	169.53	168.52	203.52	194.19
26.75	110.5	168.89	167.56	202.98	194.08
26.90	111.03	169.95	167.24	203.2	193.66
27.06	110.6	169.85	166.39	204.05	194.72
27.22	110.18	169.21	166.81	203.62	194.4
27.37	110.6	170.7	167.45	202.13	194.51
27.53	110.6	169.42	167.13	202.56	193.98
27.69	111.56	169.1	166.81	203.94	193.34
27.84	112.09	168.89	165.86	203.52	193.34
28.00	111.67	169.95	166.49	203.2	193.34
28.16	111.24	169.95	166.81	201.6	194.19
28.32	111.56	169.53	166.39	201.92	194.51
28.47	111.56	169.42	166.39	202.24	193.98
28.63	110.18	169.53	167.24	203.62	193.66
28.79	110.5	168.46	166.18	202.98	193.45
28.94	110.5	168.14	167.56	201.17	193.34
29.10	111.67	168.14	167.45	202.24	192.7
29.26	111.24	168.14	167.13	203.3	193.13
29.41	109.96	168.35	166.07	201.92	193.13
29.57	112.09	169.53	165.11	201.81	193.34
29.73	110.82	169.63	165.43	202.98	193.66
29.88	110.6	167.71	166.92	201.17	194.08

Table E6 Strainsweep results of uncured NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system

Strain (%)	Storage modulus (kPa)				
	NR	NR+MMFA	NR+RKFA	NR+MMFA Si-69	NR+RKFA Si-69
0.56	56.889	70.961	58.319	144.96	91.8467
0.98	56.254	68.697	70.982	126.4	96.128
1.95	53.939	62.646	67.905	135.4	86.35
5.02	54.28	60.433	60.605	126.37	78.146
10.04	55.65	56.686	57.361	117.95	75.921
19.95	55.786	51.378	49.112	104.41	64.001
49.94	50.598	38.797	37.31	73.922	46.683
100.02	39.559	26.053	25.66	46.814	29.579
499.97	7.8599	5.8256	7.7456	10.639	8.4505
999.94	3.3931	2.662	3.1731	4.7615	3.9094
1199.98	2.6948	2.244	2.7459	3.3629	3.5084

Table E7 Delta storage of uncured NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system (Payne effect)

	Delta G'
NR	53.50
NR+MMFA	68.30
NR+RKFA	55.57
NR+MMFA Si-69	140.20
NR+RKFA Si-69	88.34

Table E8 Frequency sweep results of uncured NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system

Frequency rad/s	Storage modulus (kPa)					Damping factor					Complex viscosity (Pa-s)				
	NR	NR+ MMFA	NR+ RKFA	NR+ MMFA Si-69	NR+ RKFA Si-69	NR	NR+ MMFA	NR+ RKFA	NR+ MMFA Si-69	NR+ RKFA Si-69	NR	NR+ MMFA	NR+ RKFA	NR+ MMFA Si-69	NR+ RKFA Si-69
0.5	41.159	47.165	46.524	83.363	73.035	0.855	1.11	1.208	0.885	0.937	1.08E+05	1.33E+05	1.11+05	2.21E+05	1.99E+05
0.99	57.954	61.882	62.4	116.84	103.03	0.759	1.007	1	0.771	0.789	73143	88296	84673	1.48E+05	1.32E+05
2	80.499	91.343	82.441	158.43	141.77	0.573	0.848	0.816	0.672	0.669	46376	59869	54920	95413	85274
5.01	111.27	139.54	122.98	220.74	200.67	0.463	0.686	0.684	0.536	0.543	24496	33803	30968	50025	45626
10	132.85	182.1	162.58	273.06	250.3	0.39	0.571	0.622	0.456	0.45	14260	20965	19581	30014	27447
15	146.57	210.35	190.8	304.03	280.26	0.36	0.514	0.591	0.407	0.41	10388	15771	14770	21892	20201
20	152.86	226.29	212.11	324.88	299.58	0.31	0.497	0.546	0.411	0.367	8000.9	12634	11123	17561	15954
25	162.36	242.57	226.06	343.51	313.99	0.318	0.445	0.53	0.372	0.366	6816.3	10623	9515.8	14660	13377
30	168.34	253.13	238.78	354.47	327.33	0.313	0.431	0.492	0.362	0.358	5879.9	9186	8352.5	12565	11588
40	178	273.06	259.02	375.93	345.73	0.288	0.404	0.469	0.342	0.341	4630.2	7361.2	6751.2	9931.3	9132.6
50	187.2	288.97	281.05	394.33	362.14	0.277	0.383	0.454	0.327	0.322	3884.5	6188.9	5834.6	8296	7607.8
60	193.33	298.66	289.3	407.26	373.94	0.262	0.365	0.441	0.316	0.31	3330.5	5298.5	4982.9	7118.2	6525.1
69.99	198.02	307.86	308.37	416.87	384.83	0.26	0.357	0.424	0.304	0.302	2923.1	4670.3	4533.4	6225.1	5743.2
80	203.71	316.29	313.55	425.06	393.41	0.247	0.347	0.414	0.299	0.296	2622.8	4185.2	4039.1	5546.2	5129.1
90	204.98	324.46	325.09	436.14	399.7	0.25	0.342	0.406	0.291	0.29	2347.7	3810.1	3710.1	5046.7	4624.2
100	208.82	330.86	328.32	439.03	407.52	0.243	0.329	0.392	0.294	0.288	2149	3483.6	3368	4575.7	4241.1

Table E9 Tensile properties of NR filled with 2 sources of 100 phr FA with and without silane (Si-69) in peroxide system

	M100 (MPa)	Elongation at break (%)	Tensile strength (MPa)
NR	0.32 $\pm$ 0.01	612.15 $\pm$ 96.87	1.26 $\pm$ 0.36
NR+MMFA	0.48 $\pm$ 0.03	585.63 $\pm$ 25.34	2.86 $\pm$ 0.73
NR+RKFA	0.52 $\pm$ 0.05	542.48 $\pm$ 33.10	3.96 $\pm$ 0.31
NR+MMFA Si-69	0.63 $\pm$ 0.01	582.53 $\pm$ 15.84	5.67 $\pm$ 0.33
NR+RKFA Si-69	0.62 $\pm$ 0.04	579.84 $\pm$ 38.89	5.14 $\pm$ 0.79

APPENDIX F

PUBLICATION

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Viscoelastic Properties of Fly Ash-Filled Natural Rubber Compounds: Effect of Fly Ash Loading

Thanunya Saowapark,¹ Narongrit Sombatsompop,² Chakrit Sirisinha^{1,3}

¹Department of Chemistry, Faculty of Science, Mahidol University, Bangkok 10400, Thailand

²Polymer Processing and Flow (P-PROF) Group, School of Energy, Environment, and Materials, King Mongkut's University of Technology Thonburi (KMUTT), Bangmod, Thungkru, Bangkok 10140 Thailand

³Research and Development Center for Thai Rubber Industry (RDCTRI), Faculty of Science, Mahidol University, Salaya Campus, Salaya, Nakhon Pathom 73170, Thailand

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ABSTRACT: Fly ash (FA) as a by-product of power station plants is known to consist of silicon dioxide similar to precipitated silica. The use of FA as filler in natural rubber (NR) was of interest to reinforce and/or reduce product cost. In this article, viscoelastic properties of FA-filled NR composites with various FA loadings were investigated with the utilization of two different modes of shear flow, namely, oscillatory and steady shear flow. It is found that the addition of FA to NR increases storage modulus (G') and shear viscosity under both oscillatory and steady shear flow. Moreover, the oscillatory test results exhibit the unexpected increase in magnitude of viscous response with increasing FA loading in FA-filled NR compounds. The explanation is proposed in terms of the ball-bearing effect of FA with spherical shape associated with the

occurrence of molecular degradation induced by inorganic constituents particularly manganese, iron, and copper in nonrubber component of NR as well as the small amount of heavy metals including iron, copper in FA. An isoprene rubber (IR) containing no nonrubber component was used to validate the proposed explanation. In addition, with the use of Cox-Merz concept, the results of both complex viscosity under oscillatory shear flow and apparent shear viscosity under steady shear flow can effectively be superimposed in the case of FA-filled compounds, supporting the promotion of viscous response by FA. © 2009 Wiley Periodicals, Inc. *J Appl Polym Sci* 112: 2552–2558, 2009

Keywords: fly ash; natural rubber; viscoelastic properties; filler; rheology

INTRODUCTION

Fillers are added to rubber for a variety of purposes, of which the most important are enhancements in reinforcement, processability and material costs.¹ Typically, fillers used in rubber industry are classified into reinforcing and nonreinforcing fillers. In some cases, fillers are divided into black and non-black types. There are basically two categories of reinforcing fillers—carbon black and mineral or 'white' fillers such as fumed and precipitated silica.² In rubber industry, because of its fine particle size (high specific surface area), such silica is widely used as nonblack reinforcing filler to improve mechanical properties of vulcanizates, particularly hardness and resistances to tension, tear and abra-

sion. Although, silica is still well accepted among rubber technologists, the use of some other fillers from the natural resources as alternative reinforcing fillers in NR has been carried out to replace silica in rubber compounds.³ Such fillers include clay, lignin, black rice husk ash (BRHA) and white rice husk ash (WRHA), and cellulose fiber.

Ash residues are wastes of coal-fired power plants; these include fly ash (FA) and bottom ash.^{4,5} FA is a relatively inexpensive by-product, and its usages have the benefit of decreasing environmental problems. Furthermore, FA has been used in industry as a consequence of such advantages as low cost, smooth spherical surface and good processability of the filled materials.⁶ In previous study,^{3,7–12} it has been reported that FA particles consist of silicon dioxide similar to silica but with the lower content of silicon dioxide than silica. Consequently, the addition of FA to rubber compounds is able to enhance rubber properties similar to that of commercial silica, but with the much smaller magnitude. It must be noted that if the reinforcement effect provided by FA is carefully considered particularly when compared with precipitated silica, FA can hardly be considered as filler with a real reinforcement potential

Correspondence to: C. Sirisinha (scsir@mahidol.ac.th).

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TABLE I
List of Materials Used in the Present Study

Materials	Trade name/grade	Suppliers	Amount (phr)
Natural rubber (NR)	STR 5L	Union rubber products, Thailand	100
Synthetic <i>cis</i> -1,4-polyisoprene (IR)	NIPOL IR 2200	Nippon zeon, Japan	100 ^a
Fly ash (FA)	—	Mae Moa power station of KNR group, Thailand	Variable

^a Only used for comparing with NR to exclude a non-rubber effect on compound properties.

because of its much lower structure and surface reactivity. However, in terms of environmental concerns, FA is an interesting filler giving reinforcement to some extent and its role on viscoelastic properties of NR compounds is of interest in the present study.

The present work aims to further the previous work³ by focusing mainly on viscoelastic properties of FA-filled NR composites measured from oscillatory and steady shear rheometers. The viscoelastic properties of NR compounds filled with various FA loadings will be discussed. It must be noted that the simple compounds containing only NR and FA with various loading are investigated in this work to exclude any interference on viscoelastic properties which might be caused by other compounding ingredients.

EXPERIMENTAL

Materials

The materials used in the present study are summarized in Table I.

Preparation of rubber compounds

Rubber compounds were prepared using a laboratory-sized two roll mill (model LRM150, Labtech, Thailand) at the set temperature of 60°C for 13 min. The loading of FA compounds was varied from 0 to 200 phr. The formulation of compound is given in Table I.

Characterization of FA

The FA originated from coals typical of those burned in electric power plant (Mae Moa Power Station). The chemical composition of FA was determined using the X-ray fluorescence spectrometer (model S4 Pioneer BrukerAXS, Germany). A scanning electron microscope (SEM) (model LEO 1455 VP, UK) was used at an accelerating voltage of 10 kV to examine the morphology of FA particles. Particle size analyzer (model Mastersizer 2000, Marvern Instruments, UK) was also used to measure the FA particle size and its distribution.

Measurement of viscoelastic properties

Viscoelastic properties of compounds under oscillatory shear flow were measured using the Rubber Process Analyzer (model RPA2000, Alpha Technologies, USA) with the time, strain and frequency sweep tests at test temperature of 70°C. For the amplitude strain sweep test, the deformation strain was swept from 0.5 to 1200% and 0.5 to 30% at 1 and 100 rad/s, respectively. In the frequency sweep test, angular frequency of 1 to 100 rad/s was used under a small strain in the linear viscoelastic region. A Goettfert rate-controlled capillary rheometer (model Rheotester 2000, Germany) was also used to measure rheological properties of compounds under steady shear flow at shear rate range of 10–500 s⁻¹ and test temperature of 70°C. A Bagley correction with three die lengths of 10, 20, and 30 mm with a given die diameter of 2 mm was performed to eliminate end effect.

RESULTS AND DISCUSSION

The chemical composition of FA as determined by the X-ray fluorescence spectrometer is shown in Table II. It is evident that the largest amount of composition in FA is of silicate (SiO₂) or silica. Figure 1 depicts SEM micrograph of FA particles used in the present study. It is clear from the micrograph that the shape of FA particles is spherical. In addition, the particle size distribution of FA used is rather broad with the sizes ranging from submicron to 200 microns as illustrated in Figure 2. The reported mean particle size of FA is of 50.311 microns. It

TABLE II
Chemical Compositions of FA Used in the Present Study

Compositions	Content (%)	Compositions	Content (%)
SiO ₂ (silica)	37.6	BaO	0.130
Al ₂ O ₃	22.4	SeO	0.123
CaO	15.1	MnO	0.101
Fe ₂ O ₃	14.5	ZnO	0.0301
K ₂ O	2.97	ZrO ₂	0.0297
SO ₃	2.71	Cr ₂ O ₃	0.0240
MgO	2.29	Rb ₂ O	0.0193
Na ₂ O	1.33	CuO	0.0187
TiO ₂	0.447	NiO	0.0121
P ₂ O ₅	0.171	Y ₂ O ₃	0.00431

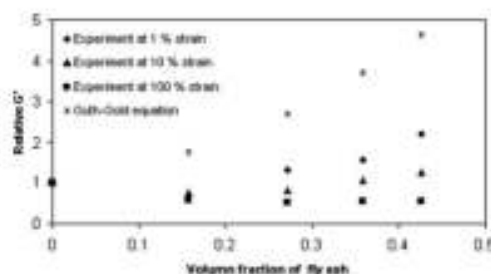


Figure 5. Relative storage modulus (relative G') at 1, 10, and 100% strain of NR compounds filled with various FA loadings as a function of FA loadings.

similar to the silicate-based filler, the interaction via H-bond between silanol groups in FA might be so small that the Payne effect is less obvious than the case of precipitated silica.

However, in the FA-filled system considered in this work, it must be noted that the magnitude of filler network effect might not be as strong as that found in silica-filled system reported previously^{14–17} because of the smaller amount of silanol group and surface area in FA particles. It is noticeable that FA particle is very similar to finely ground silicates (with limited reinforcing capabilities in rubber materials) than to precipitated silica (nearly 99.9% SiO_2) as supported by the SEM micrograph (Fig. 1). This means the changes in G' with FA loading is initially believed to be dominated by the hydrodynamic over the filler network effects. To clarify the proposed hypothesis, the Guth-Gold expression as illustrated in eq. (1) is used to generate the theoretical results of relative modulus with the exclusion of filler network effect as shown in Figure 5.^{16,19}

$$\text{Relative } G' = \frac{G'_{\text{filled}}}{G'_{\text{unfilled}}} = 1 + 2.5\phi + 14.4\phi^2 \quad (1)$$

where G'_{unfilled} and G'_{filled} are elastic moduli of unfilled and filled compounds, respectively. The ϕ is the filler volume fraction.

In theory, it is anticipated that the filled compounds would show the positive deviation relative modulus from the Guth-Gold results especially at high filler loading due to the filler network formation as found in silica filled CPE/NR blends.¹⁶ Unexpectedly, the experimental data found in this work show the negative deviation of relative modulus from the calculated Guth-Gold data, which is more pronounced at high deformation strain. It is hypothesized that the hydrodynamic effect is over-ridden by the strain amplification effect. In general, when strain is applied to the filled rubber compound, the rigid filler in the rubber matrix does not share the deformation of compound. The inclusion

of the undeformable filler in the soft rubber matrix causes the local strain in matrix to go beyond the macroscopic strain.²⁰ Thus, the rubber chains in the highly filled compounds receive greater strain, and lead to the reduction in relative storage modulus via a molecular slippage and/or a disruption of FA-NR interaction. It is noticeable that strain amplification effect would increase with increasing filler loading and strain of deformation. In addition, referred to the spherical shape of FA as shown previously in Figure 1, the molecular slippage would be further promoted via a ball-bearing effect.

Logarithmic plots of G' as a function of frequency of NR compounds filled with various FA loading are shown in Figure 6. It is evident that all compounds show an increase in G' (or elasticity) with increasing frequency which is attributed to the insufficient time for molecular relaxation at high frequency. Additionally, the magnitude of frequency-dependent responses (as determined from slopes) clearly increases with increasing FA loading. The results unexpectedly imply a decrease in elastic behavior of compounds with increasing FA loading, which is on the contrary to the previous findings on chlorinated polyethylene (CPE) system filled with precipitated silica.^{14,15} It has been reported in those work that the strong interaction between polar group of polymer matrix and silanol group of silica could form a tridimensional transient network resulting in a strong elastic response with lowered magnitude of frequency dependent behavior. Clearly, the unexpected results are in accordance with the absence of filler network as proposed previously.

Damping properties of NR compounds with various FA loading are exhibited in Figure 7. Obviously, the reduction in damping factor with increasing frequency could be observed in all compounds indicating a rise in elastic behavior caused by the insufficient time for molecular relaxation as discussed in G' results. Similar to the case of G' , there is the unexpected increase in damping factor with increasing ash loading. As mentioned previously, it

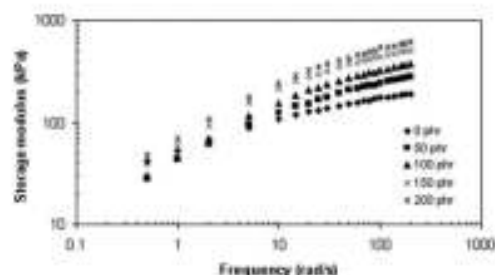


Figure 6. Storage modulus (G') as a function of frequency of NR compounds filled with various FA loadings.

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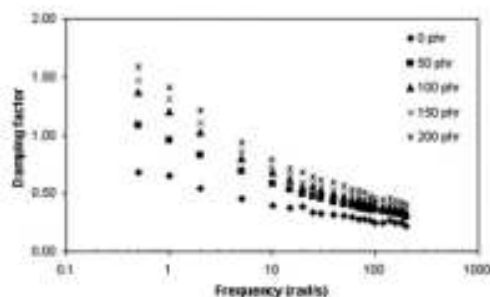


Figure 7 Damping factor ($\tan \delta$) as a function of frequency of NR compounds filled with various FA loadings.

is initially anticipated that there would be the increase in elastic behavior of compounds with increasing FA loading via the filler network formation similar to the previous work on silica-filled CPE system.^{14,15} The unexpected difference in result trends are believed to be the results of a viscous response promotion given by molecular slippage via the FA ball-bearing effect (i) and molecular degradation of NR matrix which might be accelerated by the presence of nonrubber component and/or of metal such as iron and copper in FA as illustrated in Table II (ii).^{21,22}

To validate the proposed explanation of nonrubber component effect, further experiment was performed on a synthetic isoprene rubber (IR) which is free from nonrubber substances. It can be seen from Figure 8 that, with increasing FA loading, the increase in damping factor is still noticeable, but the magnitude of discrepancy in damping factor of compounds with different FA loading is much less apparent than that of NR compounds (Fig. 7). This implies the lower viscous response in IR compounds than NR compounds. Thus, it might be summarized

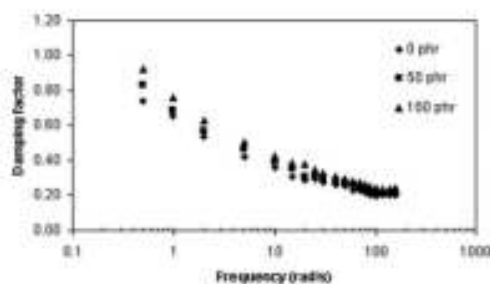


Figure 8 Damping factor ($\tan \delta$) as a function of frequency of synthetic isoprene rubber (IR) compounds filled with various FA loadings.

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that the nonrubber substances presented in NR play role in such increase in magnitude of viscous behavior to some extent. According to previous work, the inorganic constituents found in nonrubber component of NR including manganese, iron and copper are able to catalyze the oxidation breakdown of NR.^{21,22}

An existence of small magnitude of change in viscous response observed in IR compounds filled with FA might be due to the molecular slippage via the ball-bearing effect of FA and/or the occurrence of molecular degradation of rubber matrix by a thermo-oxidative process, as mentioned previously. It is likely that a small amount of heavy metals in FA including iron or copper as shown in Table II could act as good oxidizing agents, which are believed to accelerate the degradation of rubber molecules.²¹

Figure 9 reveals the processability of the NR compounds with various FA loadings via complex viscosity as a function of frequency. As expected, the complex viscosity evidently decreases with increasing frequency indicating the pseudoplastic behavior. Additionally, the increase in complex viscosity with increasing FA loading could be observed. Referred to the results of damping properties shown in Figure 7, the highly filled compounds reveal high damping factor particularly at low frequency that means the increase in complex viscosity is caused by the increase in magnitude of viscous response via molecular slippage as discussed earlier.

In the case of rheological properties under steady shear flow as measured from a capillary rheometer, Figure 10 reveals a similar result trend to that of complex viscosity under oscillatory flow, i.e., there is a slight increase in true shear viscosity with increasing FA loading at any given shear rate. Such small magnitude of increase in shear viscosity of NR compounds filled with FA is in good agreement with previous work.²³ The promotion of viscous response by molecular slippage via the ball-bearing effect

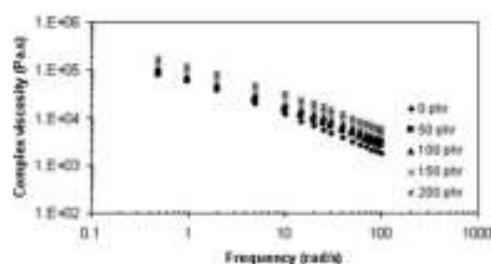


Figure 9 Complex viscosity as a function of frequency of NR compounds filled with various FA loadings.

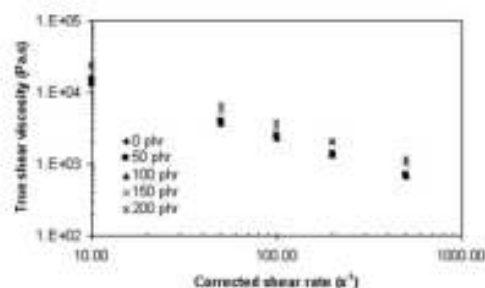


Figure 10 True shear viscosity as a function of corrected shear rate of NR compounds filled with various FA loadings.

and/or molecular chain-scission is believed to be responsible for the slight viscosity rise observed despite with high loading of FA.

To correlate rheological data under oscillatory and steady shear flow, the Cox-Merz concept²⁴ was applied, and the results obtained are illustrated in Figure 11. It is obvious that, in the case of unfilled NR compound, complex viscosity measured under oscillatory shear flow is slightly higher than apparent shear viscosity measured under steady shear flow, which is due probably to the end effect taking place via elasticity of compounds. This end effect leads to an additional pressure measured. On the contrary, in FA-filled compounds, both plots of

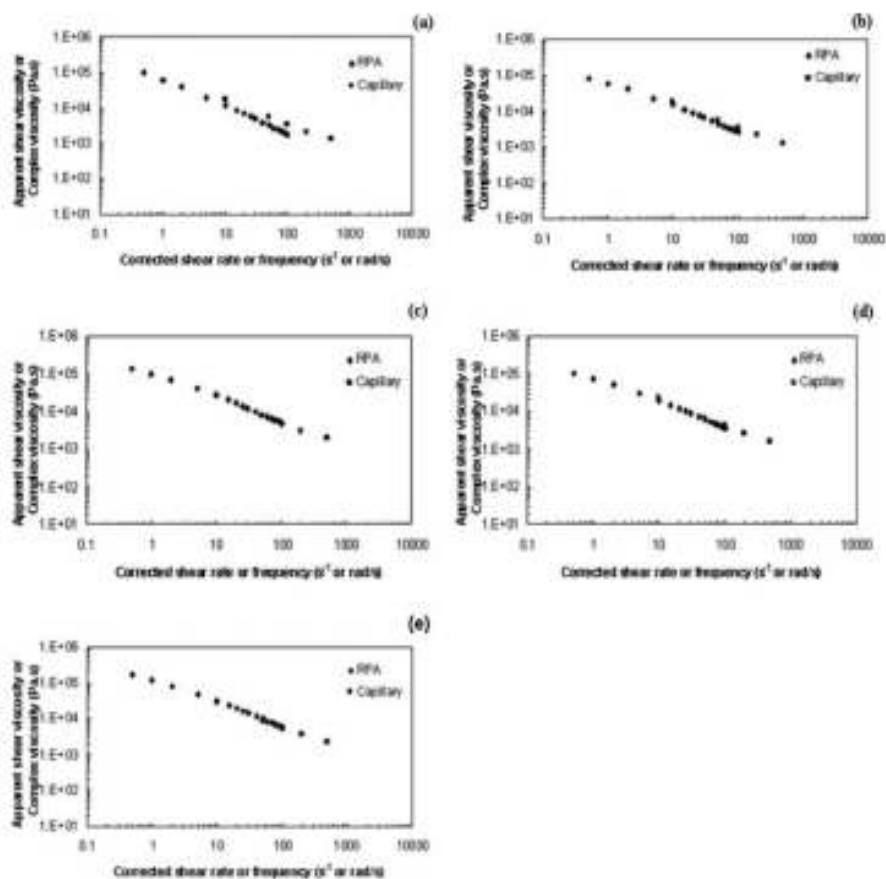


Figure 11 Plots of complex viscosity determined from RPA2000 as a function of frequency (◆) and apparent shear viscosity measured from capillary rheometer as a function of shear rate (■) of NR compounds filled with various FA loadings : (a) 0, (b) 50, (c) 100, (d) 150, and (e) 200 phr.

complex and apparent shear viscosity become superimposed. This is because of the flow promotion via viscous response provided by FA and/or nonrubber component, as discussed earlier.

In summary, it is proposed that, in NR compounds filled with FA, there is an occurrence of molecular degradation of NR which would therefore cause an increase in magnitude of molecular slippage. Furthermore, the spherical shape of FA would promote such molecular slippage via the ball-bearing effect. These two phenomena would end up with an increase in magnitude of viscous response of NR compounds as FA loading increases. On the contrary, in IR compounds without nonrubber substances, the smaller magnitude of molecular degradation yields the greater degree of molecular entanglement and thus the lower viscous response than NR. The molecular degradation is believed to be attributed to the presence of nonrubber component in NR and heavy metals in FA.

CONCLUSIONS

The FA-filled NR compounds are investigated in this article, and the following conclusions can be drawn. The addition of FA in NR increases G' and viscosity under both oscillatory and steady shear flow. Moreover, the higher FA loading, the larger magnitude of damping properties (viscous response) in the FA-filled NR compounds is observed. The explanation is proposed in terms of ball-bearing effect of FA associated with molecular degradation induced by inorganic constituents particularly manganese, iron and copper in nonrubber component of NR as well as the small amount of heavy metals including iron and copper in FA. The isoprene rubber (IR) containing no nonrubber component was used to validate the explanation. With the use of Cox-Merz concept, the results of both oscillatory and steady shear flows can effectively be superimposed in the case of FA-filled compounds, supporting the

promotion of viscous response provided by the addition of FA.

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Storage Instability of Fly Ash Filled Natural Rubber Compounds

Thanunya Saowapark, Pongdhorn Sae-oui, Narongrit Sombatsompop, Chakrit Sirisinha

Generally, fly ashes (FA) could function as either semi-reinforcing or non-reinforcing fillers in polymeric systems, depending on particle size, specific surface areas and surface chemistry of FA particles. Typically, FA particles are spherical with smooth surfaces having significant influences on viscoelastic and mechanical properties. Additionally, the presence of heavy metals in FA particles could play role on degradation process of rubber molecules to some extent. In this article, the storage instability and thermal aging properties of FA filled natural rubber (NR) compounds were focused via changes in viscoelastic responses. Results obtained reveal that the storage duration of FA filled NR compounds leads to decreases in elastic modulus and molecular weight, particularly in the compounds with high FA loading. By replacing NR with polyisoprene (IR) containing no non-rubber substances, the storage stability is significantly enhanced. It is believed that the presence of metal ions in both FA and non-rubber substances in NR could catalyze the degradation process of rubber molecules. Such degradation process could effectively be suppressed by the addition of amine-based antioxidant. © 2012 Applied Rheology.

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BIOGRAPHY

NAME	Miss Thanunya Saowapark
DATE OF BIRTH	December 1, 1980
PLACE OF BIRTH	Bangkok, Thailand
INSTITUTIONS ATTENDED	Silpakorn University, 1998-2002: Bachelor of Science (Chemistry) Mahidol University, 2002-2005: Master of Science (Polymer Science and Technology) Mahidol University, 2006-2012: Doctor of Philosophy (Polymer Science and Technology)
RESEARCH GRANTS	The Development and Promotion of Science and Technology Talent Project (DPST), 2001-2010
HOME-ADDRESS	1/22 Leelavadee Village, Wangtagoo, Mueang NakhonPathom, NakhonPathom, 73000 Tel. 089-818-8131 E-mail: bbiirddtt@hotmail.com

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