CHAPTER IV

RESULTS AND DISCUSSION

The discussions of the experimental results in this work were classified into three sections. The first section reveals the characteristic of composite waste samples that used in this work in terms of the amount of glass fiber content, functional group analysis, elemental analysis, proximate analysis, and thermal decomposition behavior of composite waste samples. The second section shows the thermal conversion of composite wastes by a fixed bed pyrolysis system. This part presents the product distributions and the characteristic of pyrolysis products including of solid, liquid, and gaseous products in relation to pyrolysis temperature, type of unsaturated polyester resin matrix, type of epoxy curing agent, and glass fiber content. The last section is the preparation and properties of composites prepared from original glass fiber and recovered glass fiber from pyrolysis process, including the morphological appearance of the fractured surface from the three point bending test samples.

4.1 The Characteristics of Composite Waste Samples

4.1.1 Glass Fiber Content in Composite Waste Samples.

The amount of glass fiber content in each composite waste sample was determined by thermogravimetric analyzer (TGA) (TGA/SDTA 851e, Mettler Toledo, Switzerland) under air condition from an ambient to 900ºC and maintained at this temperature for 10 min until no change in weight was observed. After resin matrix was burned out by oxidation reaction with oxygen under air atmosphere, the solid residues were referred to glass fiber containing in composite waste samples, which did not decompose under experimental conditions. Table 4.1 shows the amount of glass fiber or solid residue of each composite sample. It was indicated that glass fiber content in the samples was closed to 30 and 60 wt. %.

Table 4.1 The Glass fiber content (wt. %) containing in composite waste samples

Raw materials/sample designation	Glass fiber content
Ortho 30%	29.27
Iso 30%	29.40
Vinyl 30%	30.20
Epoxy/MDA 30%	29.25
Epoxy/MTHPA30%	30.50
Ortho 60%	59.74
Iso 60%	60.44
Vinyl 60%	60.28
Epoxy/ MDA 60%	59.40
Epoxy/ MTHPA 60%	60.26

4.1.2 Functional Group Analysis

The FTIR absorption spectra of unsaturated polyester and epoxy composite waste samples are presented in Figure 4.1 and 4.2 , respectively. The major FTIR spectra of three types unsaturated polyester composite wastes were very similar appearing at the same regions, consisting of the strong absorption peak of carbonyl ester at 1730 cm^{-1} (C=O) and 1285 cm^{-1} (C-O), Especially, orthophthalic and isophthalic unsaturated polyester composite waste, their FTIR spectra were almost overlapped at the same region such as the presence of C-H aromatic stretching at 3030 cm^{-1}, aromatic C=C bending and alkylenyl C=C stretching at 1700-1500 cm^{-1}, and aromatic C-H bending 700 cm^{-1} which related to their similar chemical structure as presented in Figure 4.3 (a) and (b); whereas the absorption peak of vinyl ester composite waste was different from those of them in which it presented the strong peak at 2960 and 1636 cm^{-1} which generated from the vibration of C=C in the methylene and methacrylate group, as shown in Figure 4.3 (c). Furthermore, the absorption peaks of aromatic C-H bending at 700 cm^{-1} , aromatic C-H stretching at 3030 cm^{-1}, and 1508 cm^{-1} in vinyl ester composite waste were stronger than those of orthophthalic and isophthalic

due to the higher amount of benzene ring in its repeating unit. In addition, the strong intensity at 3600- 3200 cm^{-1} of vinyl ester composite was attributed to the absorption peak of hydroxy groups (OH) on the dimethacrylate backbone.

However, it was indicated that there was no difference in absorption peak region of 30 and 60 wt.% of glass fiber composite wastes as revealed in Figure 4.1 (a) and (b), respectively, due to similar structure or functional group of the unsaturated polyester matrix.

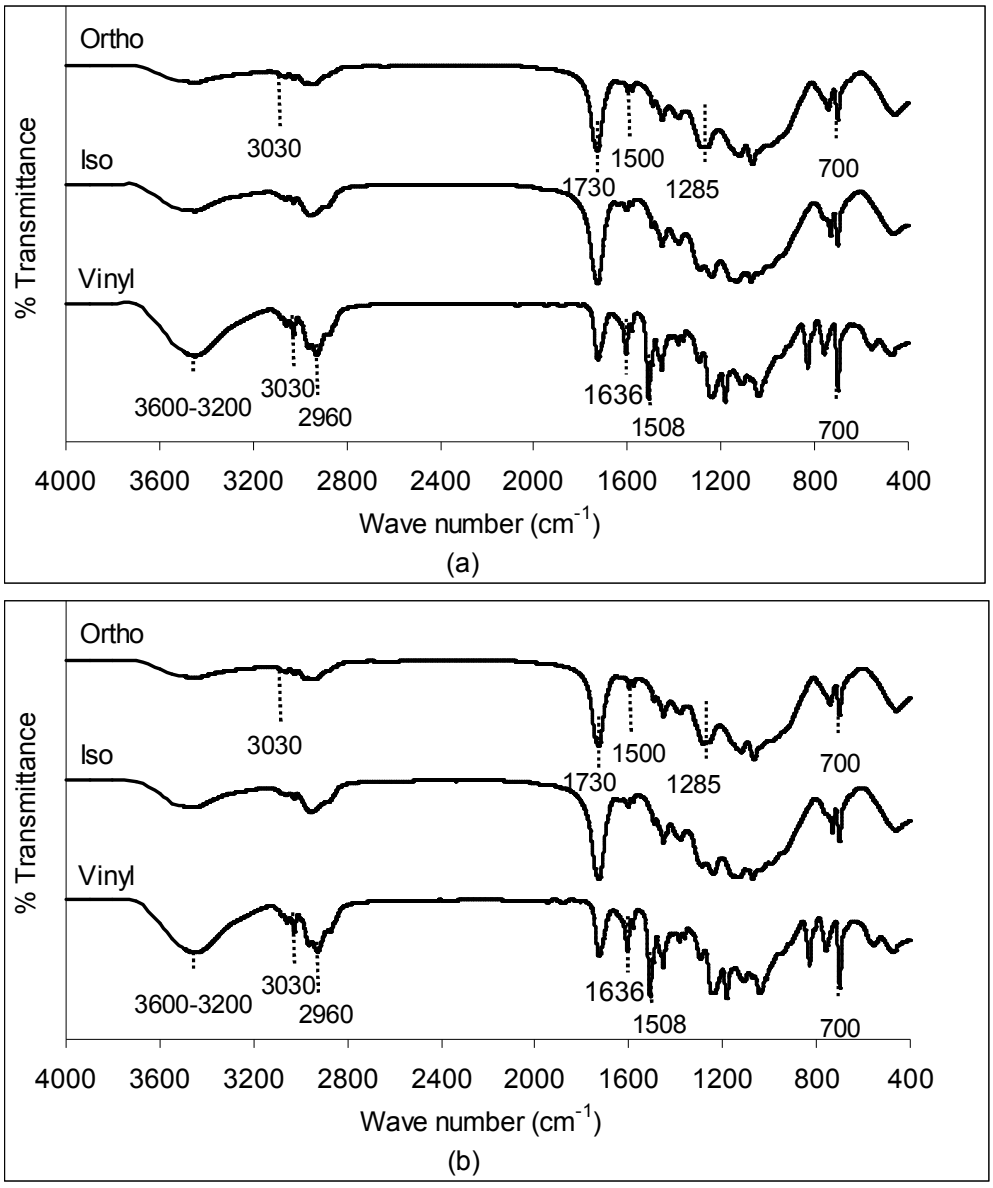

Figure 4.1 FTIR spectra of unsaturated polyester composite waste samples : (a) 30 wt% glass fiber and (b) 60 wt% glass fiber.

For the epoxy composite, the FTIR spectra of epoxy cured resin depended on the functional group of curing agent. Generally, the prominent absorption bands of epoxy resin presented at 910-890 and 1285 cm^{-1} (C-O stretching in epoxy or oxirane ring), 1630-1500 cm^{-1}(C-H bending in aromatic ring), 2930 and 2960 cm^{-1} (C-H stretching of the methylene and methyl group), and the broad peak at 3437- 3432 $cm^{-1.}$ (O-H stretching)

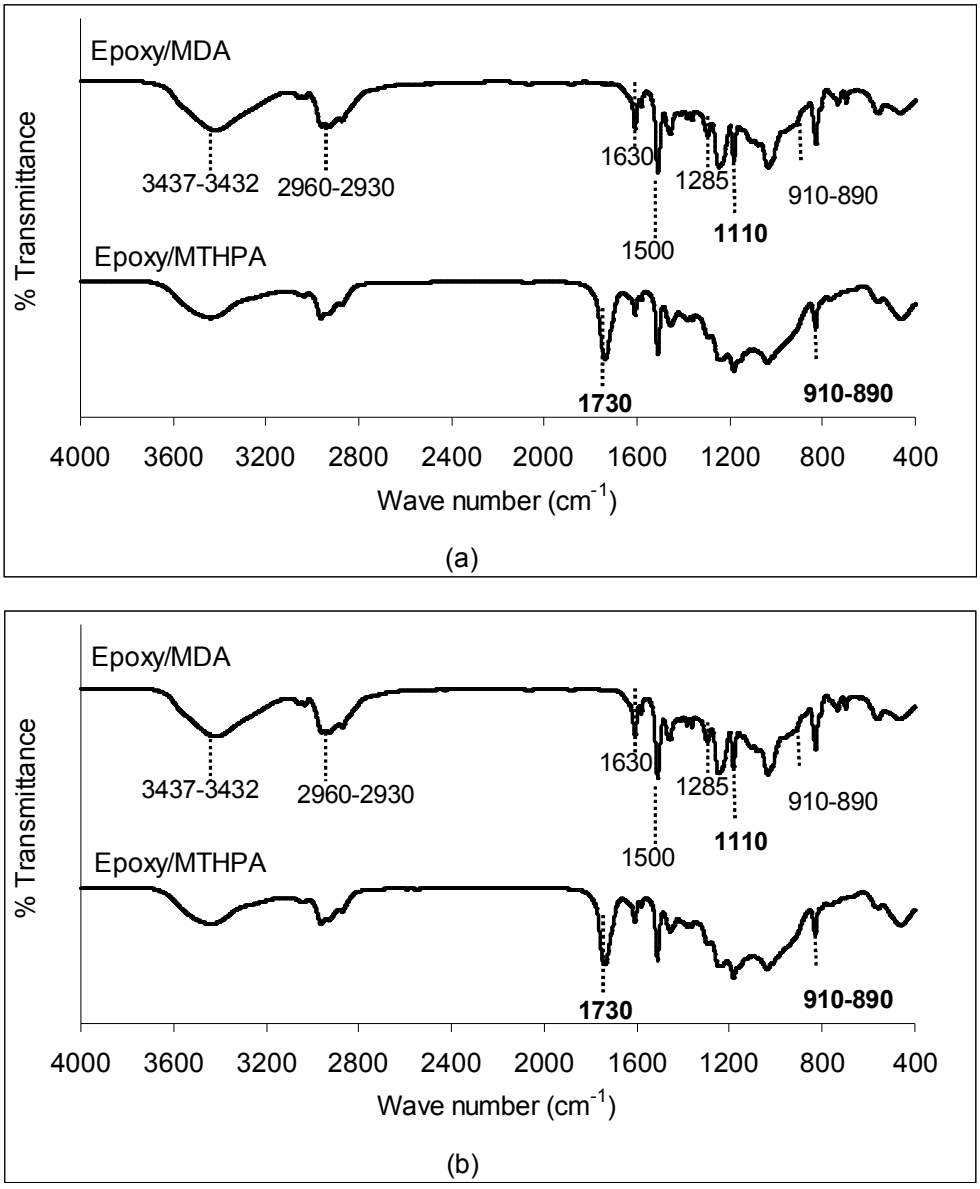

Figure 4.2 FTIR spectra of epoxy cured with amine (MDA) and anhydride (MTHPA) composite waste samples: (a) 30 wt.% glass fiber and (b) 60 wt.% glass fiber.

When epoxy was reacted with curing agent by ring opening, the intensity of absorption peak of C-O stretching t 910-890 cm^{-1} was decreased. However, the FTIR spectra of both epoxy cured with amine (MDA) and anhydride (MTHPA) matrix still presented the absorption peak which generated from the vibration in epoxy backbone such as 1630-1500 cm^{-1} (C-H bending in aromatic ring), 2960 and 2930 cm^{-1} (C-H stretching of the methyl and methylene group), and the strong broad peak at 3437-3432 $cm^{-1.}$ (O-H stretching).

In addition. Figure 4.2 (a) and (b) indicated that the appearance of C-N stretching at 1110 cm^{-1} was attributed to amine cured epoxy, while the strong carbonyl peak at 1730 cm^{-1} was used to identify the characteristic of anhydride cured epoxy. The chemical structure of epoxy cured with amine and anhydride was presented in Figure 4.4 (a) and (b), respectively. The C-O stretching t 910-890 cm^{-1} of epoxy cured with anhydride composite was stronger than that of the ones due to the higher C-O in anhydride curing agent. Similar to the spectra of unsaturated polyester composite, the spectra of epoxy reinforced with 30 and 60 wt% glass fiber presented in Figure 4.2 (a) and (b) were absolutely similar in their absorption peak region.

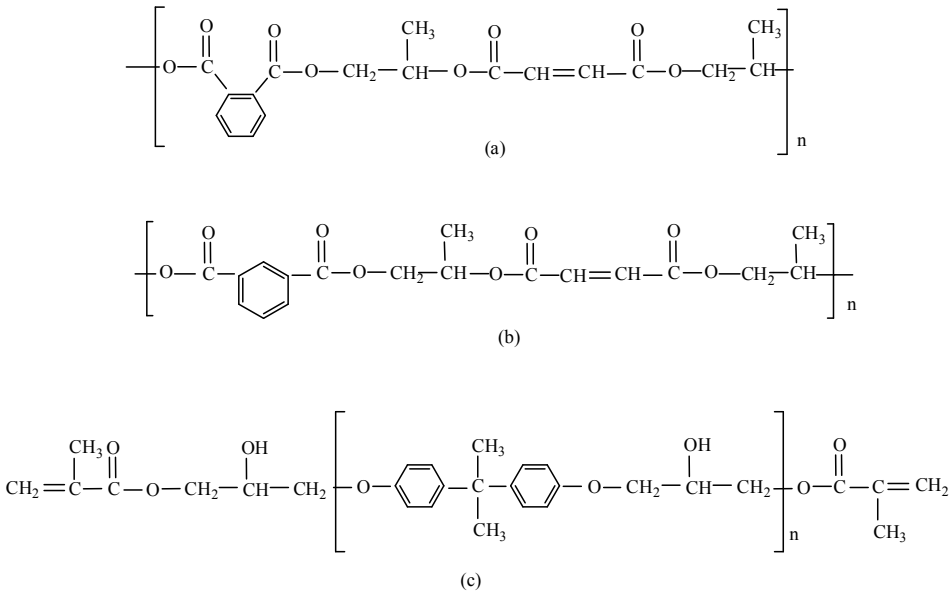

Figure 4.3 Chemical structure of unsaturated polyester resin: (a) orthophthalic polyester resin, (b) isophthalic polyester resin, and (c) vinyl ester resin.

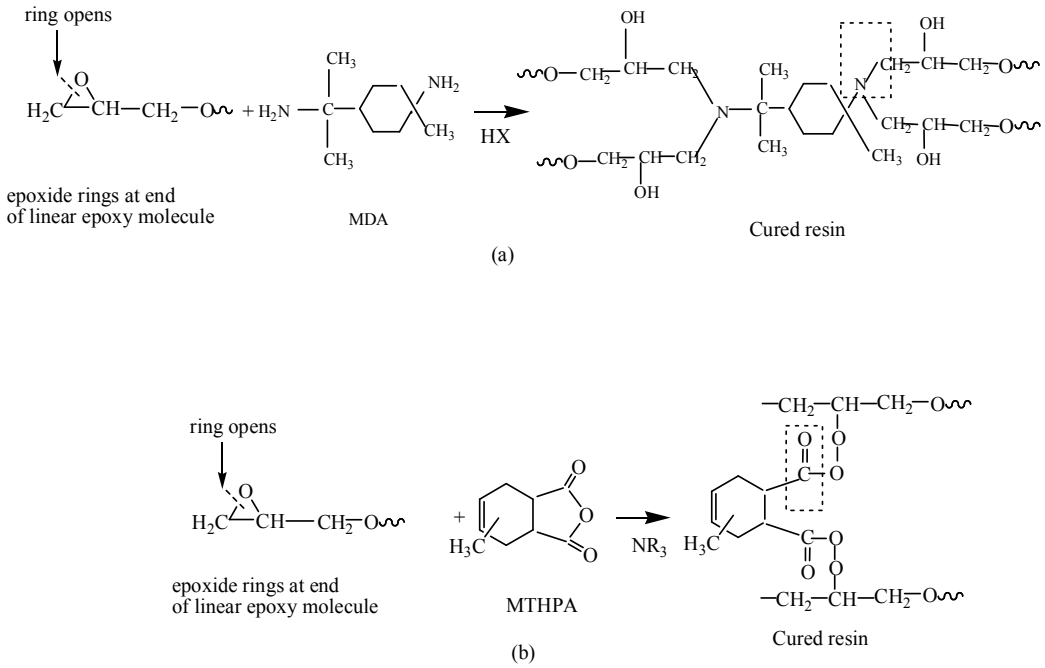

Figure 4.4 Chemical structure of epoxy cured resin: (a) Epoxy/amine (MDA) and (b) Epoxy/anhydride (MTHPA).

4.1.3 Elemental Analysis

The chemical compositions of composite samples were characterized by CHN analyzer LECO CHN-2000 as presented in Table 4.2. The percentage of CHN consisted in composite waste samples was used to evaluate the efficiency of pyrolysis process. It was indicated that how much chemical compositions in raw materials can be converted into solid, liquid, or gaseous product.

It can be seen from Table 4.2 that the chemical compositions consisted in composite samples depend on the percentage of glass fiber content. If the amount of glass fiber content in composite material was increased, the fraction of polymer matrix will be decreased resulting in the reduction of carbon, hydrogen, and nitrogen (CHN); while the amount of oxygen was increased because it was calculated from the difference in carbon, hydrogen, and nitrogen content. Both polymer matrices, unsaturated polyester and epoxy, mainly consisted of carbon, hydrogen, and oxygen. When theses elements were heated, they can be decomposed and turned into smaller

molecules in terms of light gaseous molecules such carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), or hydrogen (H_2) and some amount of these molecules may be condensed into liquid phase which can be further applied to use as fuel oil.

Table 4.2 Elemental analysis of composite waste samples.

Composite Waste Samples	Chemical Compositions (wt.%)			
	C	H	N	O*
Ortho 30%	51.79	4.76	-	43.45
Iso 30%	47.93	4.83	-	47.24
Vinyl 30%	45.07	4.51	-	50.42
Epoxy/MDA 30%	51.73	6.07	1.52	41.28
Epoxy/MTHPA 30%	45.07	4.51	-	50.42
Ortho 60%	31.35	3.25	-	65.40
Iso 60%	30.08	2.97	-	66.95
Vinyl 60%	24.41	2.27	-	73.32
Epoxy/MDA 60%	28.51	3.10	0.93	67.46
Epoxy/MTHPA60%	25.41	2.27	-	72.32

* oxygen and Silicon in glass fiber (by difference)

4.1.4 Proximate Analysis

The proximate analysis in terms of moisture content, volatile matter, fixed carbon, and ash content of composite waste samples carried out by Thermogravimetric analyzer are presented in Table 4.3. The results indicated that all of unsaturated polyester and epoxy cured with anhydride composite samples consisted of small amount of moisture content. Meanwhile, epoxy cured with amine composites showed higher moisture content. This result was attributed from amine curing agent which consisted of hydrogen polar atom that are more active towards water or moisture.

Furthermore, the composite samples were mainly consisted of volatile matters which depended on glass fiber content. When the fraction of glass fiber in composite

materials was increased, the fraction of polymer matrix was decreased resulting in the lower volatile matter. These parts were first volatilized and may be formed gaseous product for lighter molecule or condensed to liquid phase for heavier molecule under pyrolysis condition. The volatile matters generated from polymer matrix related to thermal stability of original material.

Table 4.3 Proximate analysis of composite waste samples

Composite Waste Samples	Proximate Analysis (wt. %)			
	Moisture	Volatile	FC*	Ash
Ortho 30%	0.50	70.18	0.03	29.29
Iso 30%	0.56	67.87	2.07	29.50
Vinyl 30%	0.55	66.98	2.17	30.30
Epoxy/MDA 30%	3.50	62.25	5.98	29.24
Epoxy/MTHPA 30%	0.90	61.84	6.81	30.45
Ortho 60%	0.35	39.80	0.01	59.84
Iso 60%	0.40	37.35	1.75	60.50
Vinyl 60%	0.42	37.1o	1.93	60.55
Epoxy/MDA 60%	2.40	34.74	3.36	59.50
Epoxy/MTHPA 60%	0.30	34.99	4.01	60.70

FC*: Fixed Carbon

It can be expected that under pyrolysis condition orthophthalic polyester matrix should be easily degraded and formed higher amount gas or liquid fraction than that of isophthalic and vinyl ester composite because of the presence of phthalic anhydride monomer in the orthophthalic polyester resin, which was low thermal stability. When heat was applied to orthophthalic unsaturated polyester, the phthalic anhydride was first started to degrade at lower temperature than the two other resin leading to the highest volatile matter and lowest fixed carbon.

In contrast, vinyl ester matrix yielded lower volatile matter and higher of fixed carbon upon heating because of its chemical structure was consisted of bisphenol A in the main chain. This structure was the reason for better thermal stability of vinyl ester

matrix than that the orthophthalic and isophthalic polyester matrices; as a result, result, some amount of carbon can not be degraded under the same condition and remained in the form of solid residue as fixed carbon.

Additionally, the amount of volatile matter for epoxy cured with amine and anhydride were closed and than that of unsaturated polyester matrix. This result can be explained that unsaturated polyesters were comprised of styrene diluent monomer in order to reduce their viscosity for satisfied. Obviously, the ash content, non combustible residues or inorganic matters, in all composite samples were closed to the amount of glass fiber content in the composite wastes.

4.1.5 Thermogravimetric Analysis of Unsaturated Polyester Composite

Figure 4.5 and 4.6 show the TGA and DTG thermograms of unsaturated polyester reinforced with 30 and 60 wt.% glass fiber, respectively at a heating rate of 5, 10, and 15ºC/min. Obviously, all of them started to decompose approximately at 280ºC and ended at 455ºC, and this result is in accordance with those previously reported by Evan, et al. [28]. The weight loss profiles beyond 500 ºC were mostly constant due to the step of char formation.

According to Sidney [3], thermal degradation behavior of unsaturated polyesters crosslinked with styrene composite started by the volatilization of styrene and then decarbonylation of polyester main chain by random chain scission. Unsaturated polyester cured resins were degraded at the same interval temperature ranging from 200 to 500ºC, but with minor differences in maximum weight loss temperature between 340-450ºC which related to their chemical structure regarding to the substitution position of benzene ring. From the results, it was observed that orthophthalic matrix shows slightly lower initial and final decomposition temperatures and larger DTG curves than isophthalic and vinyl ester matrices composite. This was explained by Evan, et al. [28] that orthophthalic polyester consisted of branched chain phthalic anhydride which can easily release styrene monomer at slightly lower temperature. Additionally, Sidney [3] also stated that the substitution at iso or para

position of benzene ring by using isophthalic acid or methacrylic acid in the isophthalic unsaturated polyester or vinyl ester resin resulted in the straight chain structure which showed better thermal stability than the orthophthalic polyester

The TGA data from Figure 4.5 and Figure 4.6 were summarized into Table 4.4 in order to show the effect of heating rate on thermal behavior of three types of unsaturated polyester composite wastes reinforced with 30 and 60 wt% glass fiber. It was showed that the major degradation temperature range started and ended at higher temperature heating rate increased. This characteristics were similar in all of the unsaturated polyester composite wastes reinforced with 30 and 60 wt% glass fiber.

Regarding to the effect of glass fiber content, it was observed from Table 4.4 that the presence of higher glass fiber content in unsaturated polyester composite resulted in the decreasing of the temperature at maximum weight loss (T_{max}). This result can be implied that the composite wastes reinforced with 60% glass fiber showed lower thermal stability than that of composite wastes reinforced with 30 wt% glass fiber. The presence of higher glass fiber content caused to higher local thermal conductivity resulting to start to degrade at lower temperature than those of the ones.

These the results are in agreement with those previously reported [29-32], which studied the effect of glass fiber content on the thermal behavior of glass fiber reinforced unsaturated polyester composite. They indicated that the presence of higher glass fiber content had impacted on the crosslinked density of cured network of unsaturated polyester composites by inhibiting process resulted in the decreasing in degree of cured unsaturated polyester resin [33]. As a results, their temperature at maximum weight loss were decreased comparing to those containing 30 wt% glass fiber.

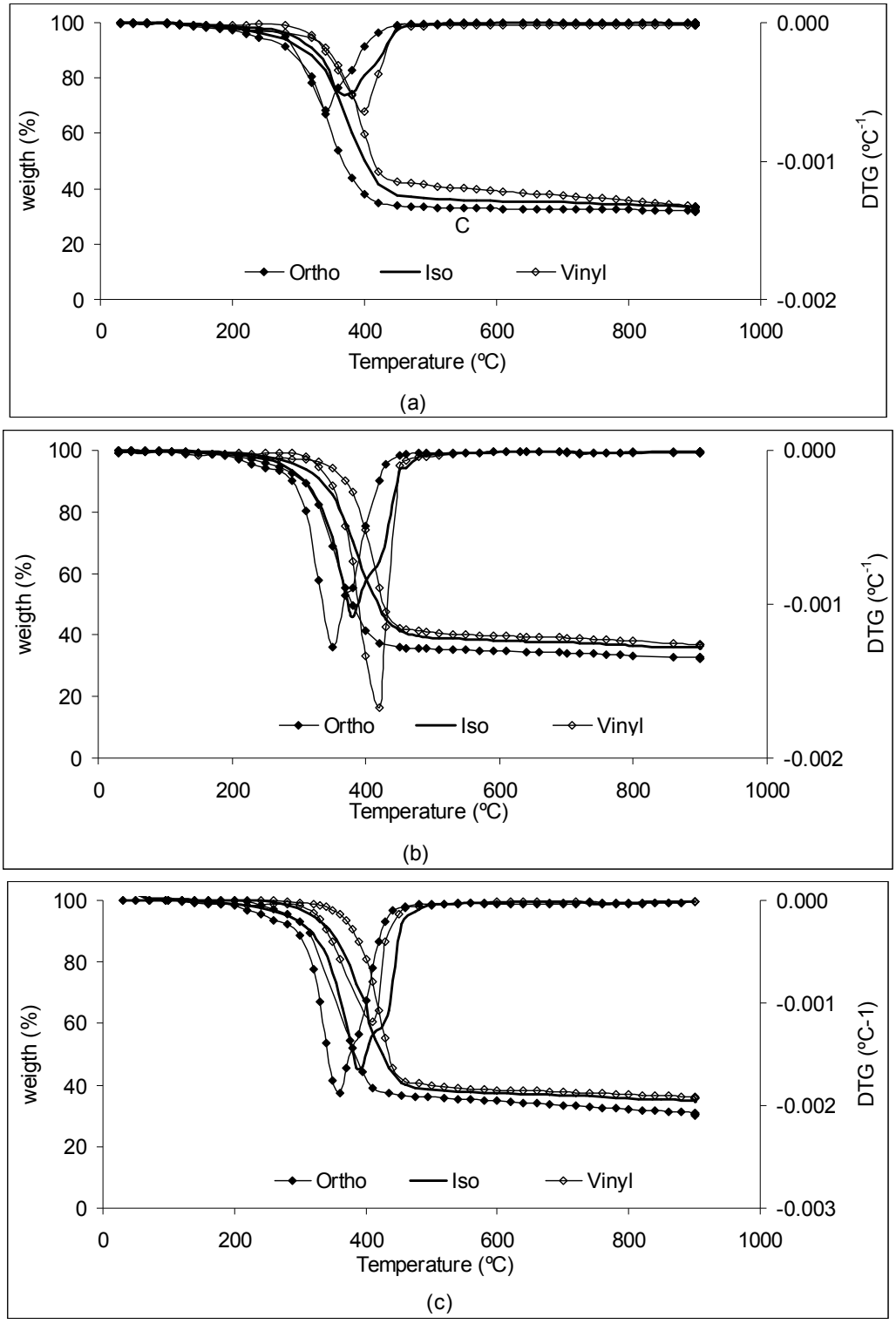

Figure 4.5 TGA and DTG thermograms of orthophthalic polyester, isophthalic polyester, and vinyl ester composite wastes reinforced with 30 wt.% glass fiber at three different heating rates : (a) 5ºC/min, (b) 10ºC/min, and (c)15ºC/min, respectively.

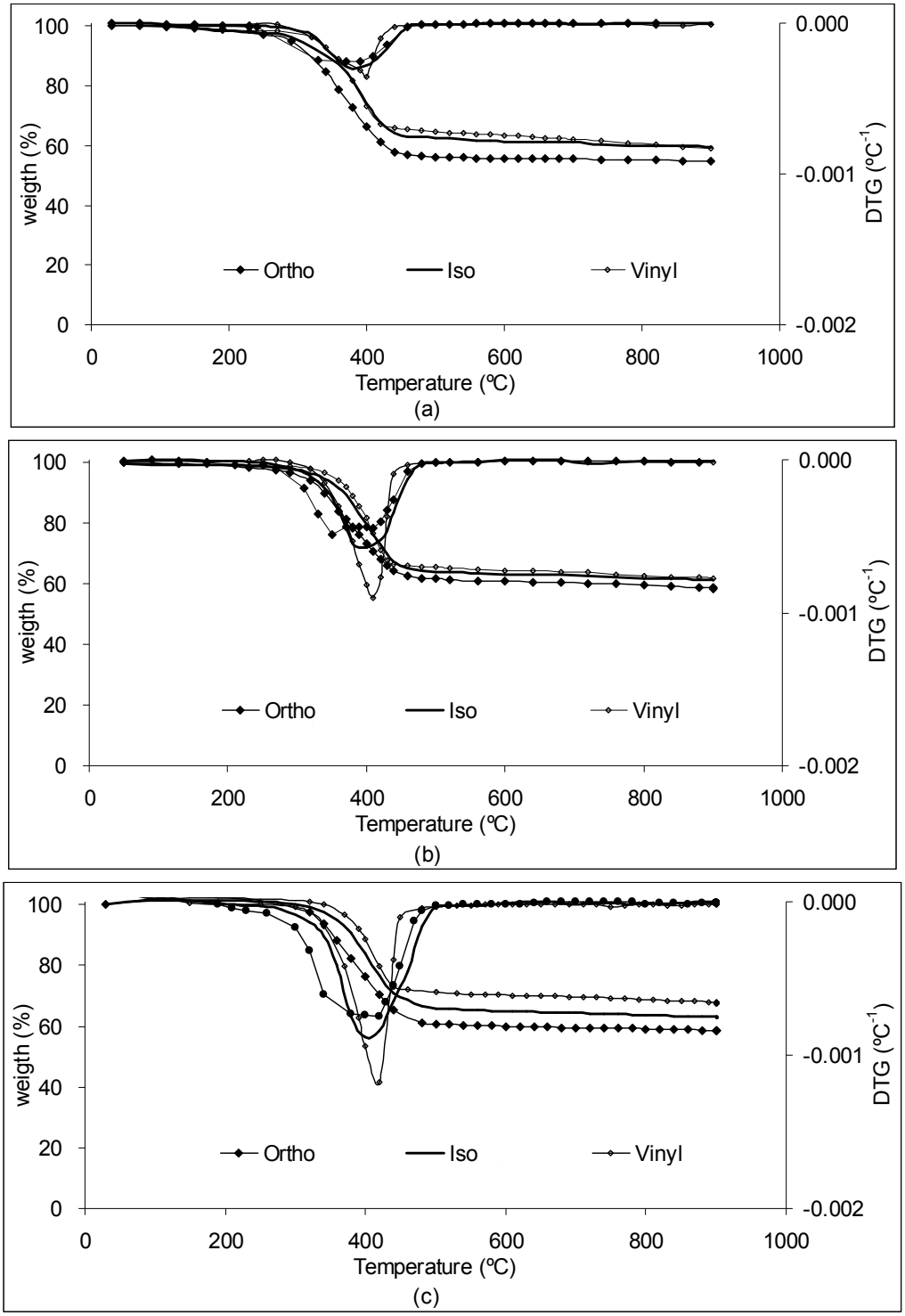

Figure 4.6 TGA and DTG thermograms of orthophthalic polyester, isophthalic polyester, and vinyl ester composite wastes reinforced with 60 wt.% glass fiber at three different heating rates : (a) 5°C/min, (b) 10°C/min, and (c)15°C/min, respectively.

Table 4.4 Summarization of TGA thermograms of unsaturated polyester composite waste samples.

Composite waste samples	Heating rate (ºC/min)	Temperature (ºC)			Weight loss at T_{max} (wt%)
		Onset[a]	Endset[b]	Maximum[c]	
Ortho 30%	5	299.80	429.62	336.50	55.50
	10	310.30	441.68	448,00	56.62
	15	314.25	457.79	355.00	57.5
Ortho 60%	5	295.80	348.50	333.40	35.32
	10	302.02	394.80	344.90	37.20
	15	312.3	404.70	251.90	39.0
Iso 30%	5	328.70	434.50	370.70	54.00
	10	341.62	450.70	382.90	55.30
	15	346.50	457.50	390.60	58.45
Iso 60%	5	323.30	415.50	365.80	37.55
	10	336.25	431.00	377.90	38.20
	15	338.80	439.70	385.60	39.02
Vinyl 30%	5	358.90	429.34	405.00	50.40
	10	377.90	436.53	418.00	53.02
	15	379.60	446.82	425.00	55.30
Vinyl 60%	5	346.63	422.33	402.06	34.55
	10	358.91	434.00	415.05	36.57
	15	362.70	442.10	422.10	37.03

a: Onset of decomposition temperature (T_{onset}) b: Endset of decomposition temperature (T_{endset})

c: The temperature at maximum weight loss (T_{max})

4.1.6 Thermogravimetric Analysis of Epoxy Composite

The TGA and DTG thermograms at different heating rates are displayed in Figure 4.7 and 4.8, respectively. Clearly, the degradation step of cured epoxy composite wastes started by the dehydration of water and the elimination of secondary alcohohol before the scission of main chain at higher temperatures which was attributed to the major decomposition step [32]. The thermal degradation behavior of epoxy composite wastes depended on types of curing agents, glass fiber content, and heating rate which were similar to those unsaturated polyester composite wastes.

The epoxy cured with amine composite waste showed initial stage of weight loss between 130 and 200ºC due to elimination of absorbed moisture and small molecule of volatile product such as ammonia from amine fragmentation [32]. The major decomposition step derived from the cleavage of the weakest C-N crosslinked bonds and -C-O- bonds of epoxy main chain [33].

Meanwhile, the onset of major degradation step of epoxy cured with anhydride showed higher thermal degradation temperature than that of amine cured matrix composite. This was resulted from the breakdown of –C-O-C- which was the crosslinked bond of epoxy main chain - anhydride curing agent, and –C-O- of epoxy main chain. These two bonds had greater thermal stability than the C-N crosslinked bonds of amine cured epoxy composites. (more thermal stability bond).

The major decomposition temperature ranges of both epoxy cured with amine and anhydride depended on heating rate and glass fiber content as summarized in Table 4.5. Considering the effect of heating rate on weight loss profile of epoxy composite wastes, similar to unsaturated polyester composite wastes, as presented in Table 4.5 the onset , end set, and the temperature at maximum weight loss were shifted to higher as increasing heating rate. This may be related to the kinetic parameter which will be discussed shortly.

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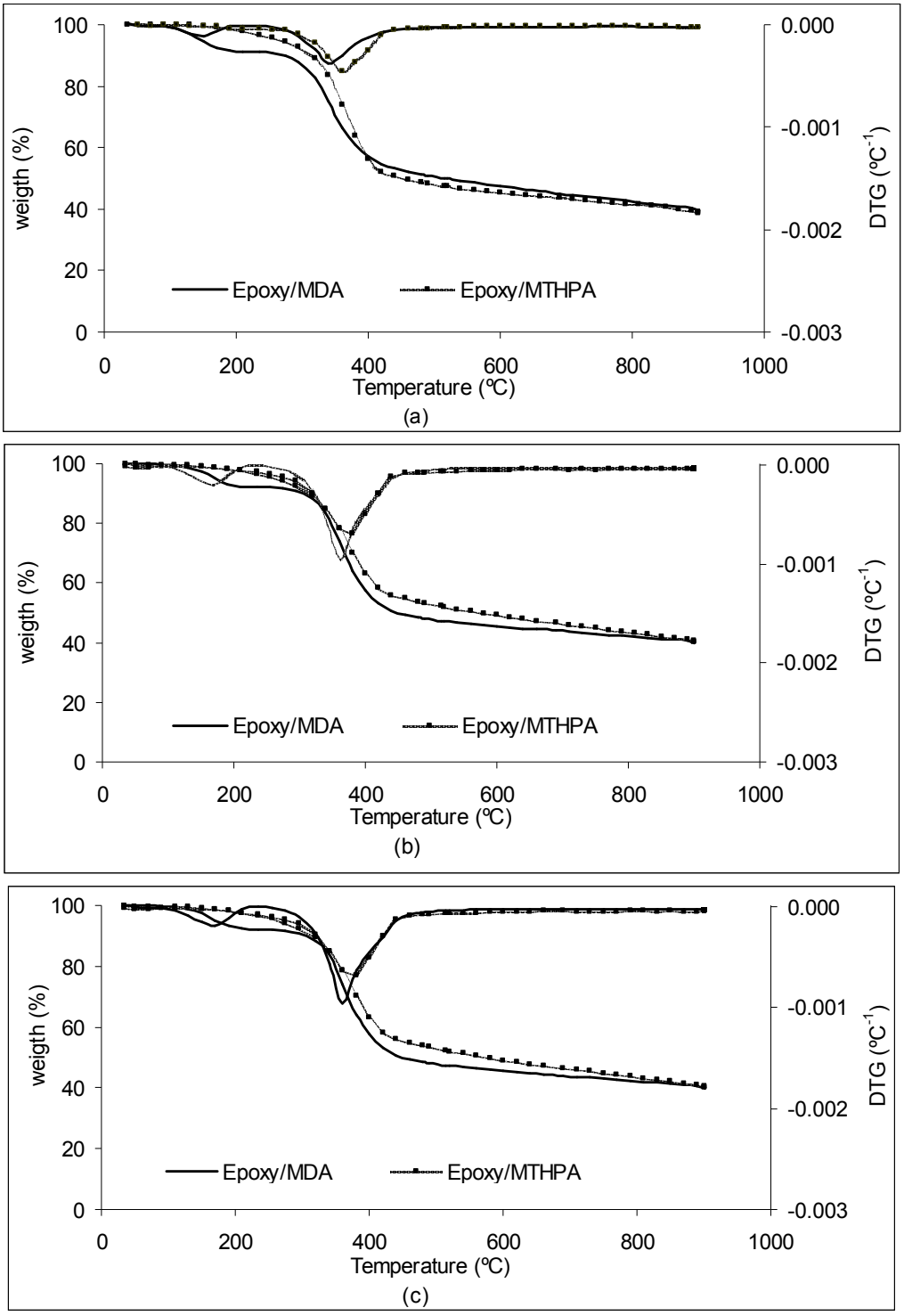


Figure 4.7 TGA and DTG thermograms of epoxy composite wastes cured with amine (MDA) and anhydride (MTHPA) reinforced with 30 wt.% glass fiber at three different heating rates : (a) 5°C/min, (b) 10°C/min, and (c)15°C/min, respectively.

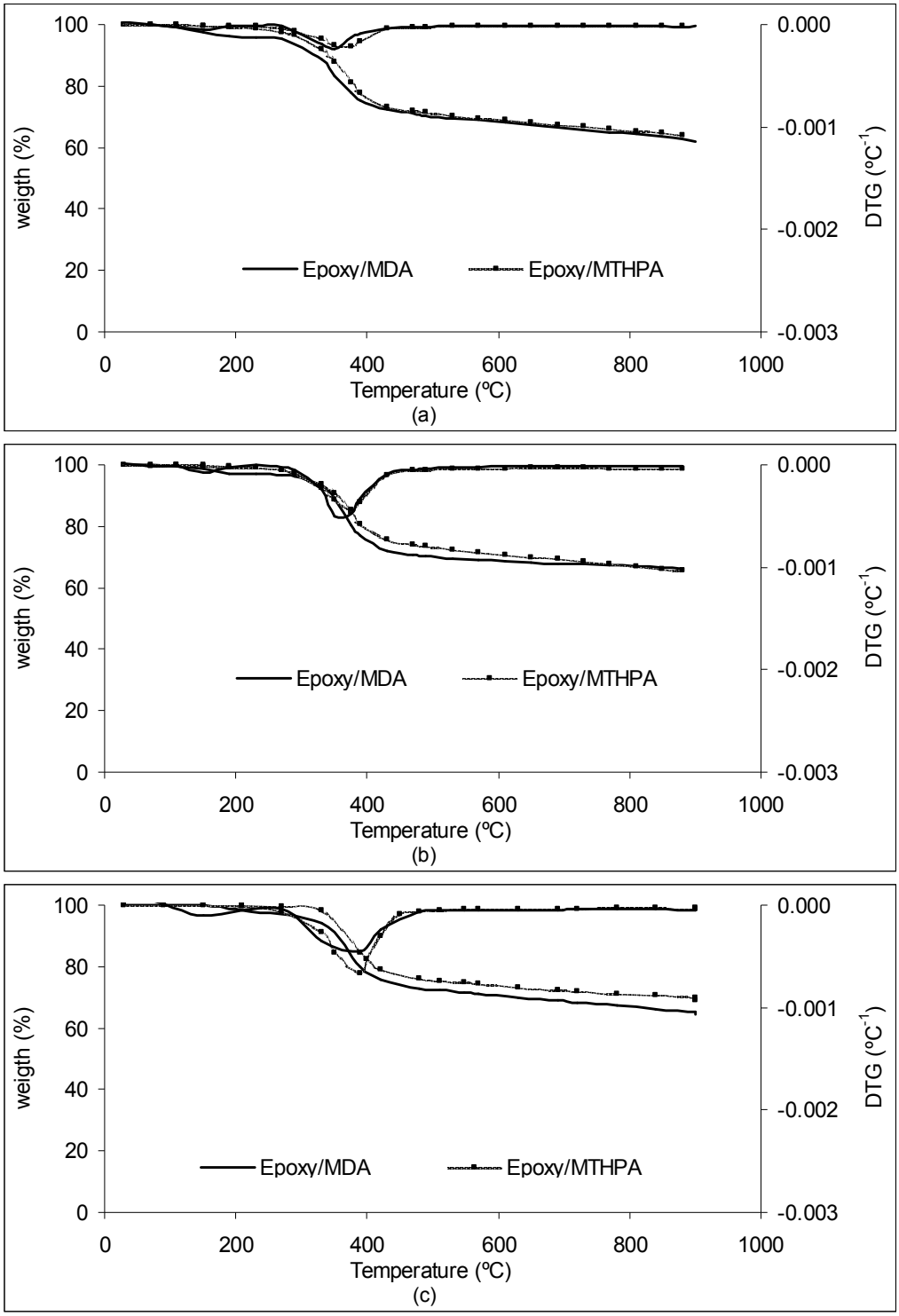


Figure 4.8 TGA and DTG thermograms of epoxy composite wastes cured with amine (MDA) and anhydride (MTHPA) reinforced with 60 wt.% glass fiber at three different heating rates : (a) 5°C/min, (b) 10°C/min, and (c)15°C/min, respectively.

Regarding to the effect of glass fiber content, as presented in Figure 4.7 and 4.8 and summarized in Table 4.5.

Table 4.5 Summarization of TGA thermograms of epoxy composite waste samples.

Composite waste samples	Heating rate (ºC/min)	Temperature (ºC)			Weight loss at T_{max} (wt%)
		Onset[a]	Endset[b]	Maximum[c]	
Epoxy/MDA 30%	5	289.71	389.73	346.20	53.50
	10	308.53	404.41	361.83	55.08
	15	314.0	414.7	369.58	58.50
Epoxy/MDA 60%	5	262.89	388.80	345.91	24.02
	10	319.39	399.53	361.70	25.10
	15	305.0	418.91	366.16	27.32
Epoxy/MTHPA 30%	5	310.03	410.52	370.00	51.50
	10	320.10	420.55	381.01	53.03
	15	330.20	425.50	389.00	57.20
Epoxy/MTHPA 60%	5	300.02	411.00	367.00	22.05
	10	307.34	421.52	378.00	24.23
	15	313.74	423.33	386.01	25.31

a: Started to decompose temperature (T_{onset}) b: Ended of decomposition temperature (T_{endset})

c: The temperature at maximum weight loss (T_{max})

The thermal degradation behavior obtained from epoxy reinforced with 60 wt% glass fiber composite waste was similar to that of 30 wt% glass fiber composite waste but the onset of major degradation temperature was lowered. This result is in agreement with Mijovic's work [34] whose indicated that the presence of glass fiber reinforcement had an effect on curing kinetic of epoxy composite by imposing

restrictions on the molecular mobility of reactive group, resulting in lower reaction rate or longer time needed to complete curing reaction than that of neat epoxy resin. As a result, cured epoxy network structure was not completed. Consequently, the results suggested that the curing process of epoxy reinforced with glass fiber content system took longer to completely cure and achieve high crosslink density network than that of neat resin

4.1.7 Kinetic Study and Activation Energy (E_a) of Decomposition Process

The studies on degradation kinetics of composite waste samples provide some usefull data for their application in terms of thermal behavior such as the prediction of maximum service temperature which relates to their properties and lifetime. Activation energy is very simplified values that contribute to overall decomposition process.

Pyrolysis kinetic parameters of unsaturated polyester and epoxy reinforced with 30 and 60 wt.% glass fiber were investigated under nitrogen atmosphere by means of thermogravimetric measurements under non isothermal conditions from an ambient to 900ºC at a heating rate of 5, 10, and 15ºC/min. The activation energies of decomposition were determined by the application of the Arrehenius equation (Eq 4.2) and Kisssinger method [35]. This method used the temperature at maximum weight loss or maximum rate of conversion (T_{max} derived from DTG curves) to calculate the activation energy of decomposition process. The rate of conversion, $d\alpha / dt$, can be expressed by

$$\frac{d\alpha}{dt} = k(T)f(\alpha) \qquad (4.1)$$

where

$$K(T) = A\exp\left(\frac{-E}{RT}\right) \qquad (4.2)$$

Here,

k(T) is the kinetic rate constant

A is a pre-exponential factor

$f(\alpha)$ is the reaction function depending on the actual reaction mechanism

E is the activation energy

R is the gas constant

T is the temperature in Kelvin, and

α is the normalized fraction conversion.

Substitution of Eq. (4.2) into Eq. (4.1) gives the following equation:

$$\frac{d\alpha}{dt} = A\exp\left(\frac{-E}{RT}\right)f(\alpha) \tag{4.3}$$

According to Kissinger method, the degree of conversion at constant heating rate, $\beta = dT/dt$, and the rate of conversion at maximum weight loss temperature (T_{max}) is equal to zero , So, Eq. (4.3) can be expressed into the following equation:

$$\frac{d\alpha}{dT} = \frac{A}{\beta}\exp\left(\frac{-E}{RT}\right)f(\alpha) \tag{4.4}$$

Eq.(4.4) is the fundamental expressions of Kissinger method [35] used to calculate kinetic parameters performed by TGA. This method relates to the temperature at maximum weight loss, T_{max}, in which the rate of conversion under constant heating rate is constant, $(d\alpha/dt) = 0$, So Eq.(4.4) can be written after taking logarithms as following equation:

$$\ln\left(\frac{\beta}{T^2_{max}}\right) = \ln\frac{AR}{E} - \frac{E}{RT_{max}} \tag{4.5}$$

For each conversion, $\ln(\beta / T^2_{max})$ is plotted vs. $1/T_{max}$ giving a straight line with a slope of $-E/R$.

Arrhenius kinetic parameters of unsaturated polyester and epoxy reinforced with 30 wt.% and 60 wt.% are plotted of $\ln(\beta / T^2_{max})$ against $1/T_{max}$ giving a straight line as shown in Figure 4.9 and 4.10, respectively. The activation energy can be calculated from the slope (m = -E/R). The activation energy (E_a) of thermoset composite decomposition can be used to compare the thermal stability of polymer matrix which related to the energy that polymer matrices needed to cleavage or break the chemical bonds. It can be implied that polymer having higher activation energy value be more thermally stable than the one with lower E_a value. The calculated activation energy by Arrhenius equation of all unsaturated polyester and epoxy composite wastes were ranged from 177 to 204 KJ/mol and 179-195 KJ/mol, respectively, as summarized in Table 4.6 which were closed to those previously reported by other researches [38-39].

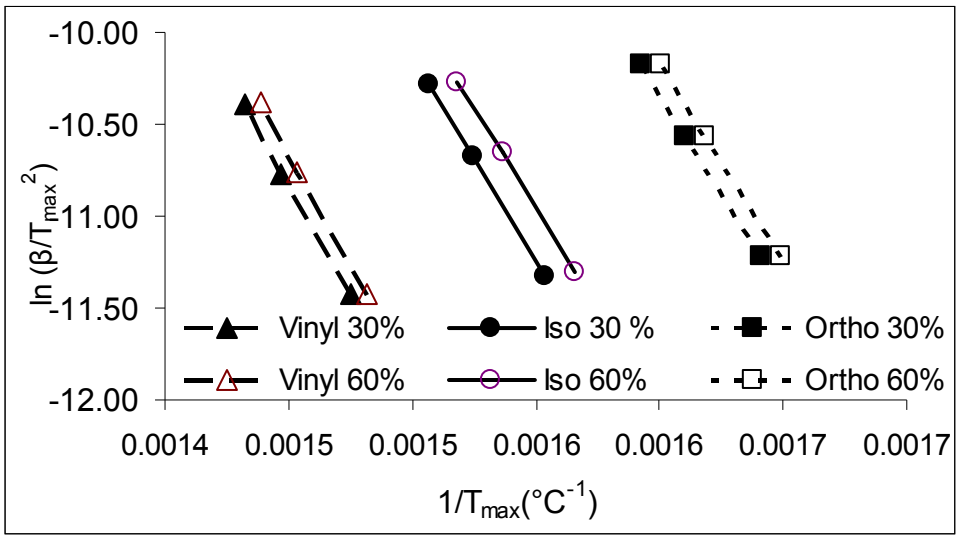


Figure 4.9 Arrhenius kinetic plot derived from TGA pyrolysis of unsaturated polyester reinforced with 30 and 60 wt.% glass fiber.

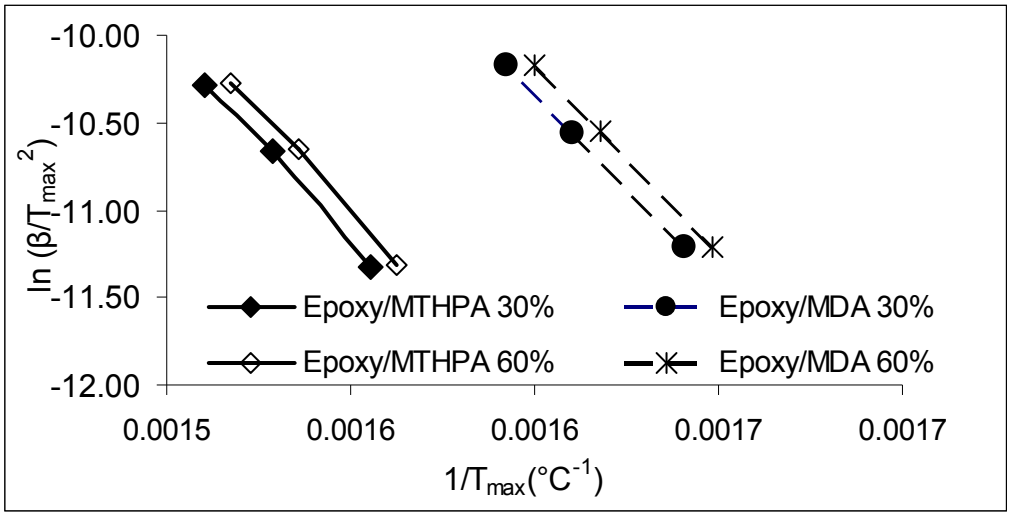


Figure 4.10 Arrhenius kinetic plot derived from TGA pyrolysis of epoxy cured with amine and anhydride reinforced with 30 and 60 wt.% glass fiber.

From the results of activation energies, it was found that the activation energy for decomposition of polymers is influenced by the chemical structure such as chain conformation, straight or branched chain or crosslinked network which decompose in a different manner when heated. Thermoplastic polymers are soften and melt before decomposing while thermoset plastic polymers are volatilized and formed char. The calculated activation energies of unsaturated polyester and epoxy composites from TGA and DTG thermograms were found to be dependent on the structure of crosslinked network and glass fiber content. Similary, Walczak [36] suggested that the activation energy of cured unsaturated polyester composite depended on their chemical structure and reinforcement content.

Comparing the effect of polyester matrix types, it was indicated the activation energy of polyester matrix associated with their decomposition steps and chemical structure. Orthophthalic polyester had lower activation energy and lager DTG peaks than isophthalic polyester and vinyl ester matrix because of the partial elimination of phthalic anhydride in branched chain. The similarity between isophthalic polyester and vinyl ester are their linear chain structure. However, the higher activation energy or thermal stability of vinyl ester matrix was due to the presence bisphenol A in the main

chain and methacrylate at the end of vinyl ester backbone, which is a steric pendant group. In addition, vinyl ester resin has reactive double bonds (ester linkage) only at the end of chains, while the orthophthalic and isophthalic unsaturated polyesters have reactive double bonds distributed throughout the chain.

Table 4.6 The activation energies (KJ/mol) of unsaturated polyester and epoxy reinforced with 30 and 60 wt% glass fiber composite waste samples.

Composite Waste Samples	Activation Energies (KJ/mol)	
	30 wt% glass fiber	60 wt.% glass fiber
Ortho	179	177
Iso	186	184
Vinyl	204	202
Epoxy/MDA	181	179
Epoxy/MTHPA	195	193

For epoxy composite wastes, the activation energy of epoxy cured with different curing agent, amine (MDA) and anhydride (MTHPA), are presented in Table 4.6 indicated that Epoxy/MTHPA system required larger energy for breaking chemical bond in its network structure than epoxy/MDA system. As previously mentioned that the activation energy of decomposition process correlated with the strength or energy of chemical bonds, the chemicals having high chemical bonding needed higher energy to breakdown their bonding before degradation.

Regarding to the effect of glass fiber content, the activation energies for thermal degradation of unsaturated polyester and epoxy composite materials are decreased with the increase of glass fiber content. For example, the activation energies of epoxy cured with amine and anhydride in Table 4.6 showed that activation energies of Epoxy reinforced with 30 wt.% glass fiber were 181 and 195 KJ/mol, respectively. while, those of epoxy reinforced with 60 wt.% glass fiber were 179 and 193 KJ/mol.

These results are in agreement with corresponded with the research of Evans, [28], Barral [37], and Montserrat [38]. Furthermore, the activation energy of three type unsaturated polyester reinforced with 60 wt% glass fiber composite waste were higher than that of previously reported by Saetiaw [39] whose work showed the activation energies of the same types of unsaturated polyester reinforced with 70 wt % glass fiber composite waste under experimented condition. As previously discussed by Britoa [40] that an addition of metallic filler or glass fiber or filler like an addition of impurities into resin matrix which resulted in lower crosslinked density and lower thermal stability.

4.2 Thermal Conversion by Pyrolysis Process

This section presents the experimental results of thermal conversion by pyrolysis process of different composite wastes using a fixed bed reactor at the temperature of 600, 700, and 800ºC. The discussion on the obtained results is classified into two parts. The fist part shows the effects of pyrolysis temperature, unsaturated polyester type or epoxy curing agent, and the amount or glass fiber content in composite samples on product distribution. The second part presents the characteristics of pyrolysis product including solid, liquid, and gaseous obtained from different pyrolysis temperature and composite waste samples.

4.2.1 Product Distributions

When polymer matrices in composite waste samples, largely consisted of C-C bond skeleton, were subjected to pyrolysis at higher temperature, they will be degraded and fragmented into smaller hydrocarbons. Generally, the products derived from thermal conversion by pyrolysis were solid residues, condensable liquid (the main target product referred to pyrolysis oil), and gaseous products providing combustible fuel gases. The lighter molecules preferred to form the gaseous products, whereas heavier molecules would condense to liquid phase. The inorganic matters or other fixed carbon still remained in forms of solid residues. The total output of product by weight fractions is

equivalent to total input raw material. The fraction of each product depended on both pyrolysis conditions (pyrolysis temperatures), and raw materials.

4.2.1.1 Effect of Pyrolysis Temperature on Product Distributions

. From the experimental results, the correlation between pyrolysis temperature and the distribution of products generated from pyrolysis of unsaturated polyester and epoxy reinforced with 30 wt% glass fiber can be plotted as shown in Figure 4.11. These results were averaged from 3 experiments having standard deviation of ± 0.5%. Obviously, decrease in solid weight resulted in the increase of liquid and gas weight. On the other hand, the reduction in liquid fraction when temperature increased from 600 to 800°C resulted from greater cracking of chemical bonds into vapors fraction.

4.2.1.2 Effect of Different Type of Unsaturated Polyester Matrix and Epoxy Curing Agent on Product Distributions

Thermal decomposition of unsaturated polyesters composite waste was probably caused by free radical depolymerization. The process results from the parts of polymer. The rate of depolymerization of polymer chain depend on their structure which resulting in different thermal stability [41]. The weight fraction of solid, liquid, and gas products involved from pyrolysis of three types unsaturated polyester reinforced with 30 wt.% fiber under the same pyrolysis temperature from 600 to 800°C are displayed in the Figure 4.12 (a-c), respectively.

It was indicated that there were negligible difference on the distribution of products obtained from different unsaturated polyester under the same condition due to the similarity of chemical composition of original monomer. The macromolecules of polyester backbone were formed by styrene crosslinked to dimensional matrix network. However there were some previous researches [28,29,40,42] reported on the relationship between chemical structure and thermal degradation behavior of unsaturated polyester. For example, Evans, et al. [28] studied the stability and the stage of degradation of unsaturated polyester of various structures such as branched chain

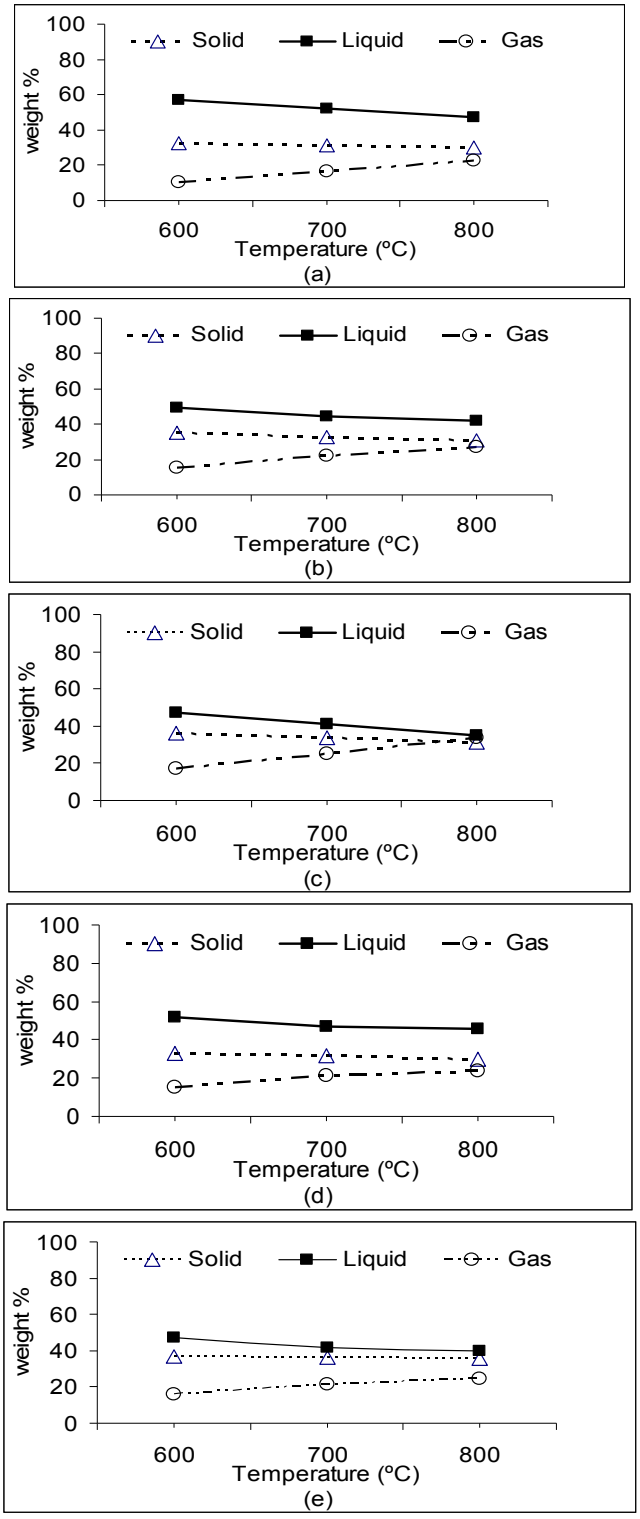


Figure 4.11 Effect of pyrolysis temperature on product yields from pyrolysis unsaturated polyester and epoxy reinforced with 30 wt% glass fiber; (a) orthophthalic, (b) isophthalic, (c) vinyl ester, (d) amine cured epoxy, and (e) anhydride cured epoxy composite waste with standard deviation of ± 0.5%

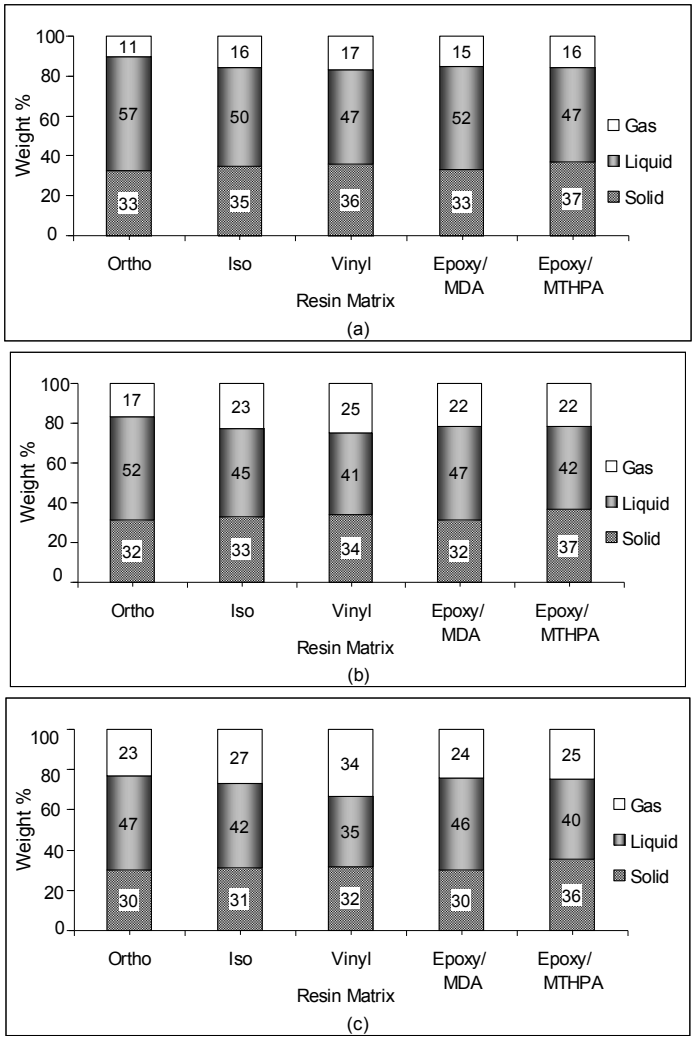


Figure 4.12 The products distribution of three types unsaturated polyester matrix and epoxy cured with amine (MDA) and anhydride (MTHPA) at pyrolysis temperature of (a) 600ºC, (b) 700ºC, and (c) 800ºC.

glycol of propylene glycol-maleic anhydride-phthalic anhydride (orthophthalic polyester), aromatic of isophthalic and terephthalate ester using thermogravimetric analyzer. They indicated that isophthalic and terephthalate ester showed higher thermal stability than that of orthophthalic polyester due to the replacement of orthophthalic with iso or terephthalic structure which increased their thermal stability. For this reason, the liquid product of pyrolyzed orthophthalic polyester at 600ºC was higher than that of iso and vinyl, while solid fraction was lower than other two resins.

On the other hand, iso and vinyl showed higher the fraction of gaseous product at 700 and 800ºC. It should be noted that ester bond (-COO-) in these matrices favored cracking into gaseous products at higher temperature. In terms of the distribution of products from pyrolysis of epoxy cured with Concerning the effect of the difference curing agent of epoxy matrix composite on the fraction of pyrolysis product also showed in Figure 4.12.

The properties of cured resins were related with chemical structure of their curing agent and also strongly depend on curing condition such as temperature. Maxwell, I.D and Richard, A.P [33] explained that crosslinked network structure of amine epoxy cured was heat sensitive due to presence of a small molecule such as ammonia, methane and nucleophilic nitrogen in the backbone leading to pyrolysis liquid fraction derived from epoxy/MDA system.

Under the same temperature slightly higher than that of epoxy/MTHPA system. pyrolysis were slightly higher epoxy/MTHPA system under the same pyrolysis condition. The fact that epoxy cured with anhydride gave lower exotherm during cure resulting in denser and brittle crosslinked network which improved its
thermal stability than that of epoxy/MDA network.

4.2.1.3 Effect of Glass Fiber Content on Product Distributions

The products distribution derived from pyrolysis unsaturated polyester and epoxy reinforced with different fiber content were considerably different on the fraction pyrolysis products as compared in Figure 4.13(a-c)

It showed that the solid, liquid and gas fraction obtained from pyrolysis composite wastes consisted of 30 and 60 wt% glass fiber at pyrolysis temperature of 600ºC showing similar trends that of 700 and 800ºC. It was clear that the increase of fiber loading from 30 to 60 wt% results in the decrease of liquid and gas fraction due to the reduction of the organic content which can be degraded and volatilized to gas before condensed to liquid phase.

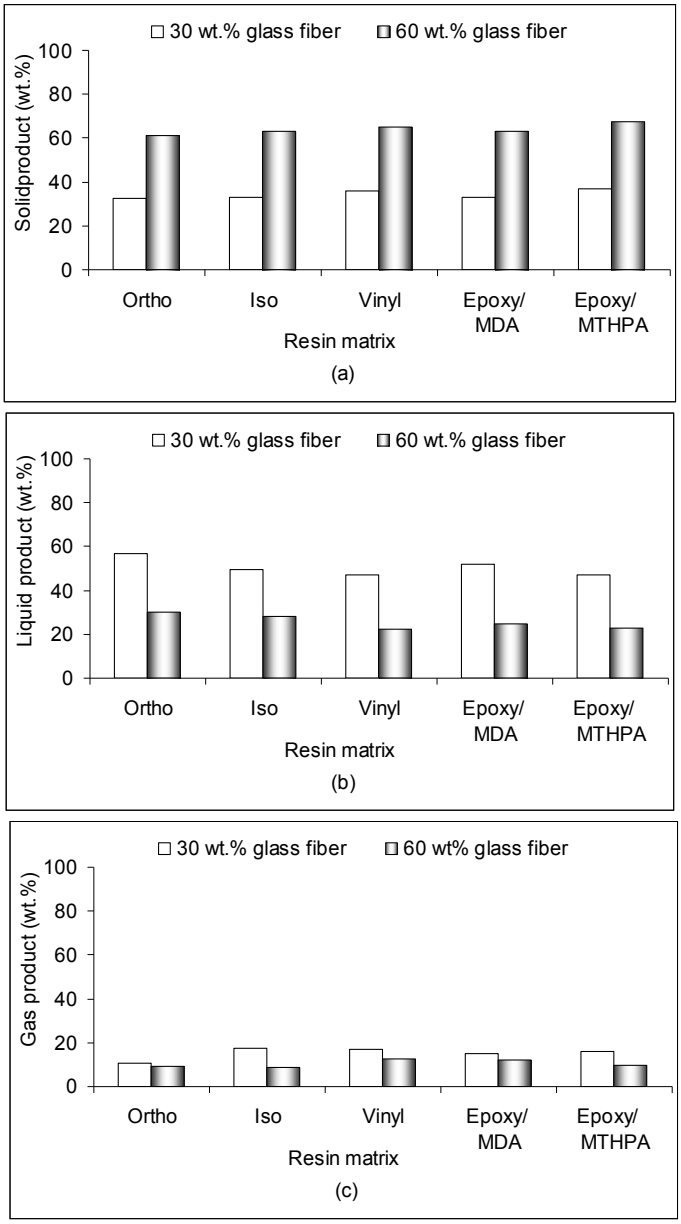


Figure 4.13 The comparison solid (a), liquid (b), and gas (c) fraction derived from pyrolysis of composite waste consisted of 30 and 60 wt% glass fiber at pyrolysis temperature of 600ºC.

4.2.2 Characterization of Pyrolysis Products

This section presents the characteristic of pyrolysis products including solid, liquid, and gas obtained from pyrolysis of different polymer matrix at pyrolysis temperature of 600, 700, 800ºC.

4.2.2.1The Characteristic of Solid residues

The solid residues retained in quartz sample boat after finished run were brittle black. They were analyzed in terms of physical appearance, functional group, elemental and proximate analyses.

Physical Appearance of Solid Residues

Due to the large number of different the SEM images from Scanning Electron Microscope (SEM, JEL JEM-6480 LV) of residues from pyrolysis process, therefore, only ten images from pyrolysis process of 60 wt.% glass fiber composite wastes at 600 and 800ºC were chosen for discussion as presented in Figure 4.14 and 4.15, respectively.

They are used to indicate the influence of different polymer matrix, pyrolysis temperature on appearance of solid char. Figure 4.14 and 4.15, indicated that such residues mainly composed of glass fiber and small amount of carbonaceous or coke-like materials derived from polymer matrix degradation during pyrolysis and may continue to repolymerize to gaseous phase by secondary reactions which corresponded with Lopez, et al.[43].

There was no significant difference on morphology appearance among various types polymer matrix. However, it seems that SEM micrographs in Figure 4.14 (a) and (b) of pyrolysis residue from orthophthalic polyester, and epoxy/MDA showing lower amount of carbonaceous materials covered on fiber surface than that of other resins which corresponding to higher liquid fraction.

The influence of pyrolysis temperature on physical appearance of solid residues can be compared and shown as SEM micrograph of solid residues at pyrolysis temperature of 800ºC (Figure 4.15). It was observed that the pyrolytic carbon slightly decreased with increasing temperature from 600 to 800ºC, but still well covered on fiber surface.

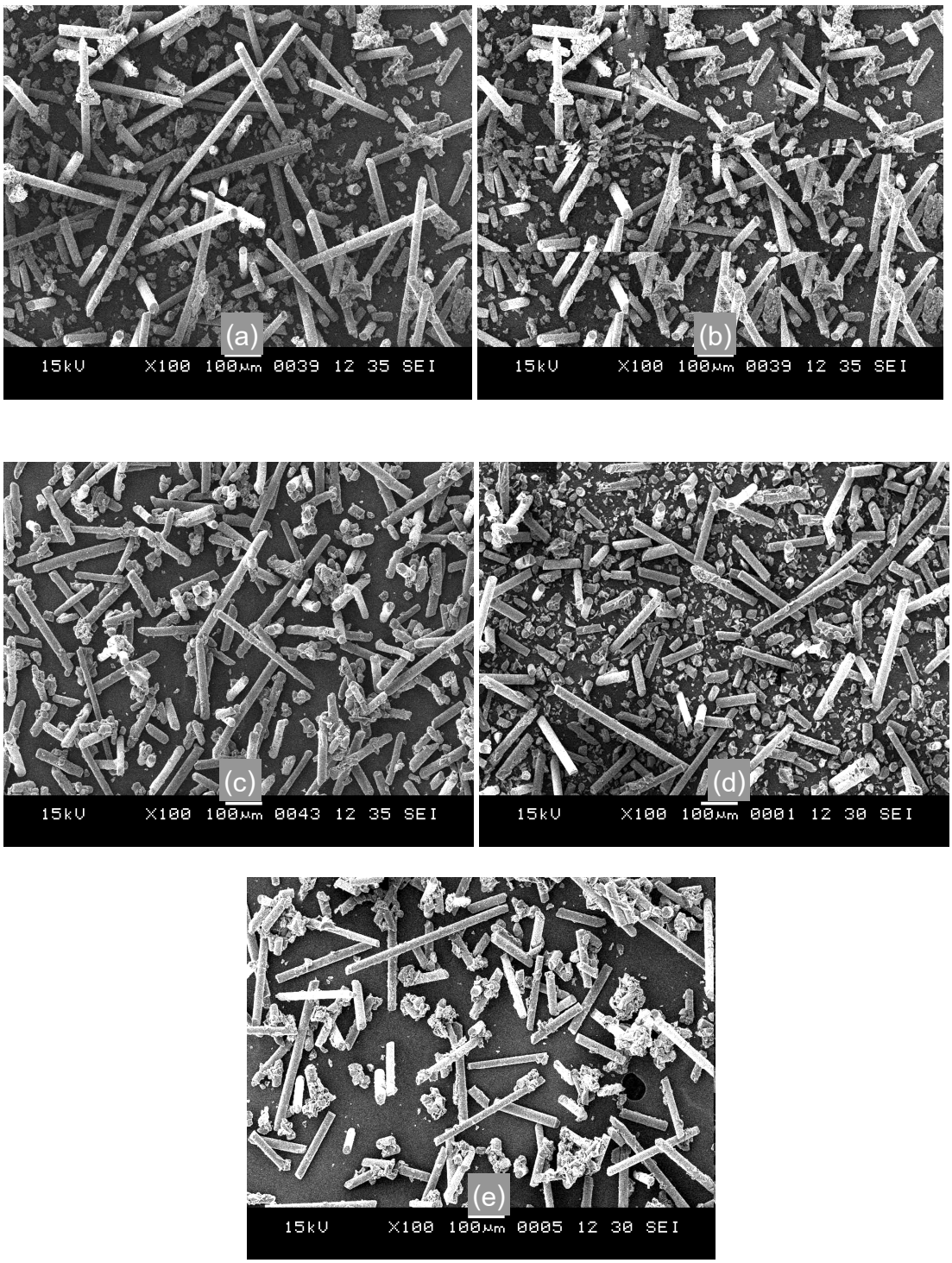


Figure 4.14 SEM micrograph of solid residues from pyrolysis of unsaturated polyester and epoxy reinforced with 60 wt.% glass fiber at temperature of 600°C: (a) orthophthalic, (b) isophthalic, (c) vinyl ester, (d) epoxy/MDA, and (e) epoxy/MTHPA.

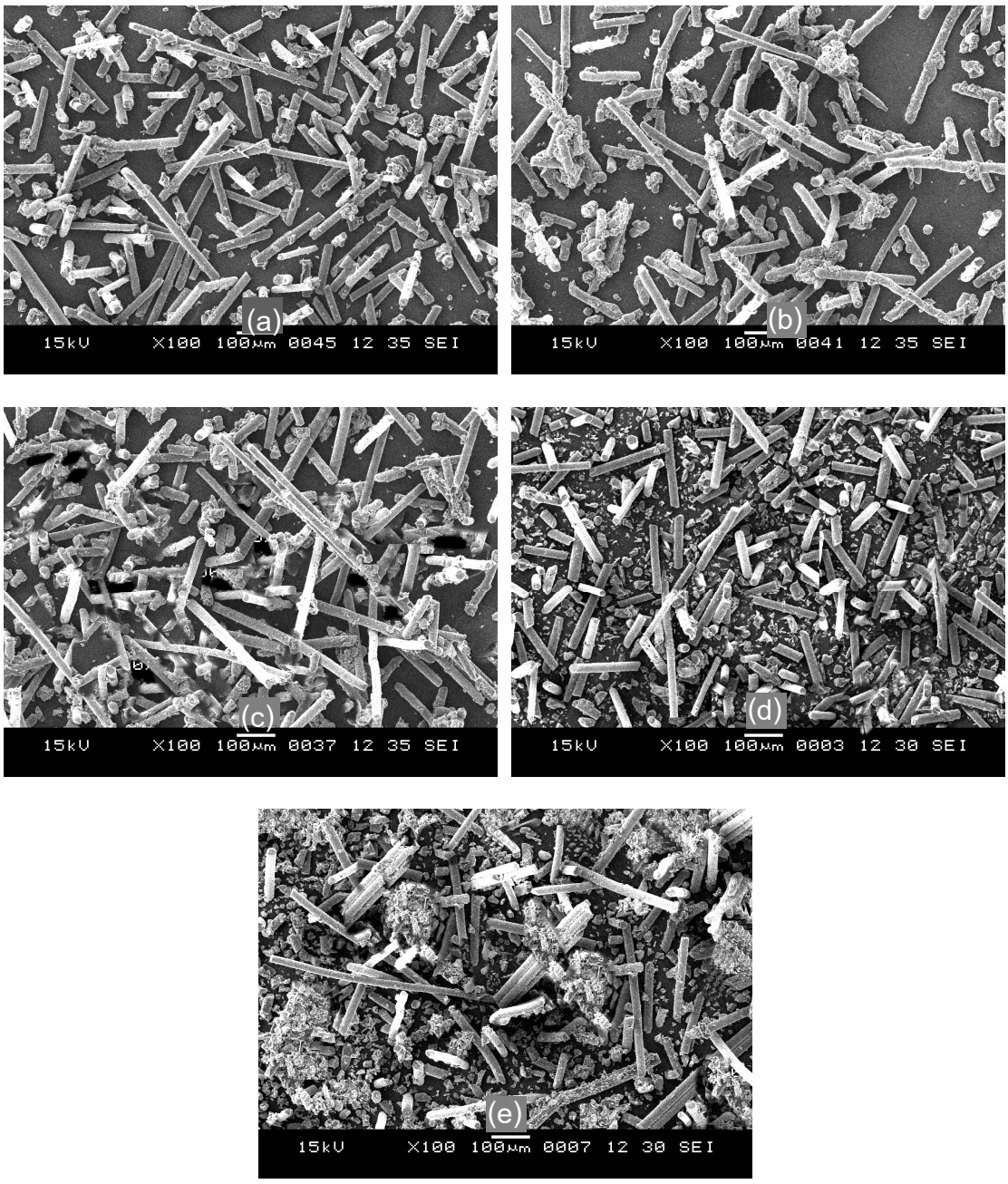


Figure 4.15 SEM micrograph of solid residues from pyrolysis of unsaturated polyester and epoxy reinforced with 60 wt.% glass fiber at temperature of 800ºC: (a) orthophthalic, (b) isophthalic, (c) vinyl ester, (d) epoxy/MDA, and (e) epoxy/MTHPA.

Functional Group Analysis of Solid Residues

The characterization in terms of functional group of resin matrix retained on glass fiber surface of solid residues which obtained from pyrolysis process at different

temperature from 600 to 800ºC were investigated by using FTIR Perkin-Elmer Spectrum and presented in Figure 4.16.

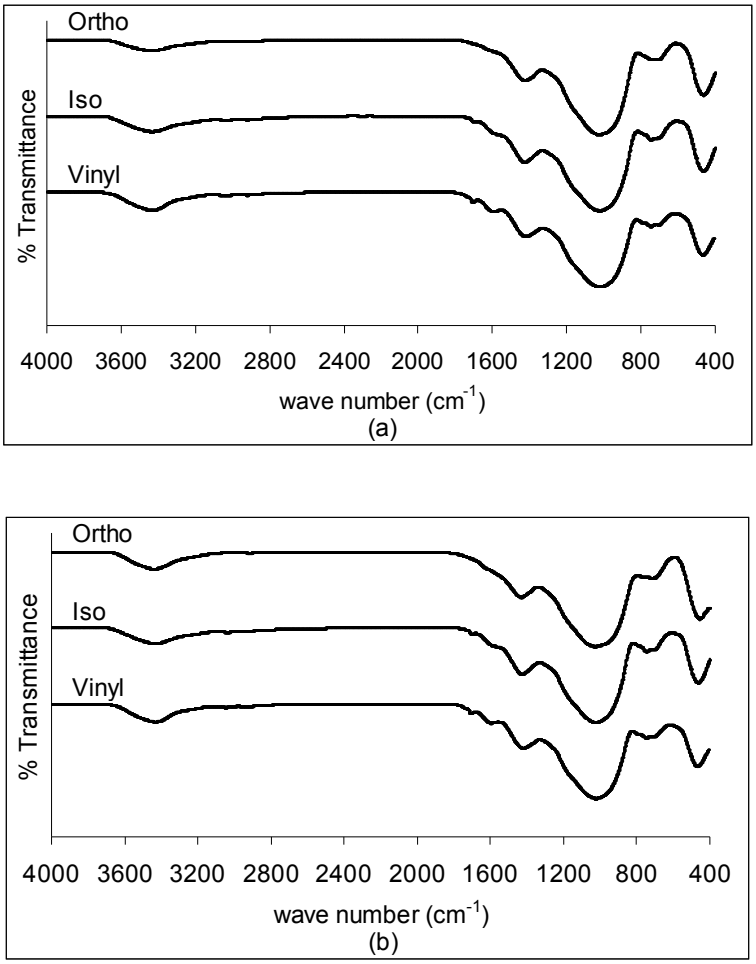


Figure 4.16 FTIR spectra of solid residues obtained from pyrolysis of three types unsaturated polyester reinforced with 30 wt.% glass fiber composite waste at temperature of (a) 600 ºC, and (b) 800ºC.

The FTIR spectra of solid residues obtained from pyrolysis of polymer matrix reinforced with 30 and 60 wt.% glass fiber composite wastes were quite similar. Hence, this section should be presented only the spectra of solid residue from pyrolysis process of unsaturated and epoxy reinforced with 30 wt% glass composite waste at the temperature of 600 and 800ºC as given in Figures 4.16 and 4.17. It was observed that FTIR spectra of solid residue of different types of unsaturated polyester and epoxy

matrix were similar which presented the broad absorption peak at 3760 cm^{-1} and 1085 cm^{-1} resulting from the vibration of Si-OH and SiO_2 in glass fiber, respectively [44,45]. Additionally, there were no evidence of absorption peak of carbonyl ester (-COO-) at 1730 and carbon nitrogen (C-N-) at 1100 cm^{-1} presented in unsaturated polyester and amine cured epoxy composite, respectively. It was implied that these chemical bonds were appreciably converted which may either be condensable liquid or gas formed.

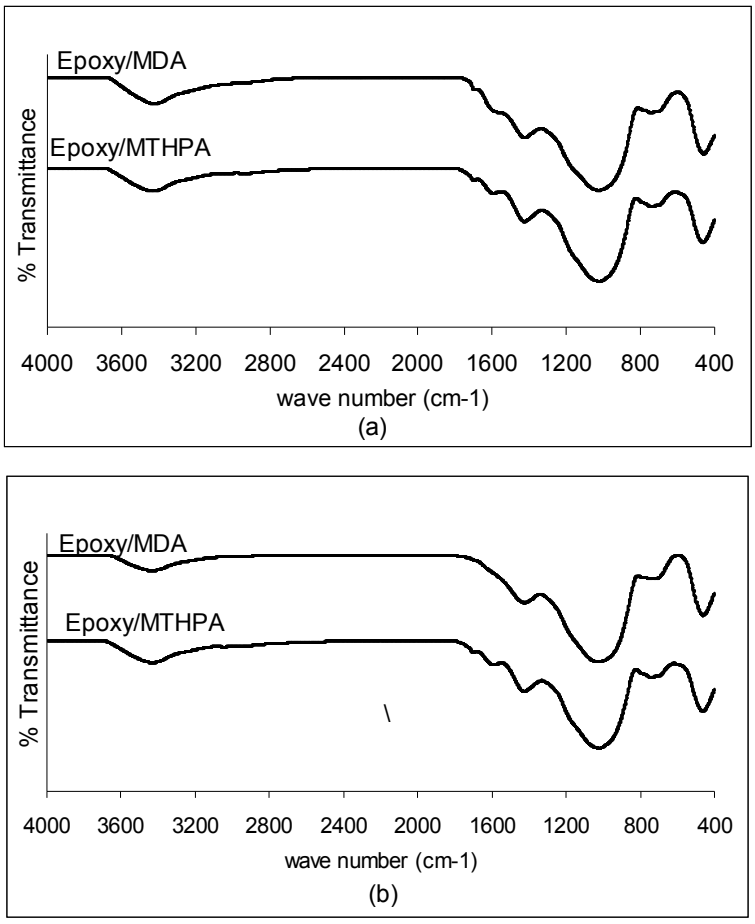


Figure 4.17 FTIR spectra of solid residues char obtained from pyrolysis of epoxy cured with amine and anhydride composite waste at temperature of (a) 600 ºC, and (b) 800ºC.

Considering the influence of pyrolysis temperature on solid residues, the spectra attributed to there are no significant on thermal degradation of polymer matrix after the temperature increased from 600ºC to 800ºC. This is due to the fact that residues solid

char were mainly glass fiber. The indicative chemical bonds in polymer matrix were first degraded at temperature of 600ºC, while some segment still remained and covered on glass fiber surface as seen from SEM micrograph before further converted by secondary decomposition at higher temperature.

Elemental Analysis of Solid Residues

The Elemental analysis results of solid residues from pyrolysis process of unsaturated polyester and epoxy reinforced with 30 and 60 wt.% glass fiber composite wastes at temperature of 600, 700, and 800ºC were determined by ECO CHN-2000 analyzer and presented in Table 4.6. The results indicate that the amount of carbon and hydrogen slightly decreased in correlation with the increase of pyrolysis temperature. The amount of oxygen, calculated by difference, increased suggesting that such residues mainly comprised of inorganic matter in formed of glass fiber.

However, the percentages of carbon and hydrogen obtained from series of unsaturated polyester and epoxy showed little different difference types of unsaturated polyester resin matrix related with thermal stability of each type of resin matrix. Orthophthalic polyester composite presented lowest the amount of carbon and hydrogen, while vinyl ester showed highest carbon and hydrogen percentages. These results corresponded with activation energy of composite waste sample. This is because chemical structure of vinyl ester resin is epoxy and unsaturated of polyester.

The elemental analysis results of solid char from pyrolysis of epoxy cured with amine were little less than that of epoxy cured with anhydride which related with the lower in dissociation energy bond of C-N and N-H network of amine cured epoxy than C-O in anhydride cured epoxy [5] resulting in epoxy cured with amine can be more degraded and less in carbon composition in solid char.

The chemical composition in solid residue from pyrolysis of 60 wt.% glass fiber showed similar trend of decreasing carbon and hydrogen with the increase of temperature related the amount of carbon and hydrogen in original feedstock composite waste.

Table 4.7 Elemental analysis (wt.%) of solid residues obtained from pyrolysis process of composite wastes with 30 and 60 wt% glass fiber.

Solid residues	Temperature (ºC)	30 wt% glass fiber				60 wt% glass fiber			
		C	H	N	O*	C	H	N	O*
Ortho	600	6.55	0.07	-	93.38	2.62	0.00	-	95.77
	700	5.46	0.05	-	94.49	2.18	0,00	-	97.82
	800	5.26	0.01	-	94.73	2.10	0.00	-	97.90
Iso	600	9.07	0.19	-	90.91	3.62	0.07	-	96.31
	700	7.09	0.13	-	92.79	2.83	0.04	-	97.09
	800	5.35	0.12	-	94.52	2.15	0.03	-	97.82
Vinyl	600	11.03	0.23	-	88.97	4.19	0.09	-	95.72
	700	10.47	0.22	-	89.31	3.97	0.08	-	96.11
	800	8.75	0.19	-	91.02	3.41	0.07	-	96.52
Epoxy/	600	10.40	0.32	0.35	88.93	3.64	0.14	0.10	96.22
MDA	700	8.36	0.23	0.15	91.26	2.92	0.09	0.06	97.17
	800	6.38	0.11	0.12	93.39	2.23	0.04	0.04	97.73
Epoxy/	600	12.40	0.63	-	86.97	4.21	0.25	-	95.54
MTHPA	700	10.36	0.26	-	89.38	3.52	0.10	-	96.38
	800	6.38	.0.17	-	93.47	2.23	0.06	-	97.71

*: oxygen and others (by difference)

Proximate Analysis of Solid Residues

Proximate analysis in terms of volatile matter and fixed carbon and ash content in solid residues from pyrolysis of composite wastes were performed by thermogravimetric analyzer and presented in Table 4.8. The results show similar trend as elemental analysis. There were some amount of volatile matter and fixed carbon remained in residues as presented in SEM micrograph in Figure 4.14, especially vinyl

Table 4.8 Proximate analysis (wt.%) of solid residues obtained from pyrolysis process of composite wastes with 30 and 60 wt% glass fiber.

Solid residues	Temperature	30 wt% glass fiber			60 wt% glass fiber		
	(ºC)	VM[a]	FC[b]	Ash	VM[a]	FC[b]	Ash
Ortho	600	5.23	0.52	94.25	2.42	0.20	97.38
	700	5.20	0.20	94.60	2.25	0.15	97.60
	800	5.13	0.13	94.74	2.20	0.07	97.73
Iso	600	8.02	1.92	90.06	2.64	0.28	97.08
	700	5.33	1.52	93.15	2.40	0.23	97.37
	800	4.93	1.34	93.73	2.30	0.13	97.57
Vinyl	600	10.35	2.00	87.65	2.72	0.45	97.08
	700	8.82	1.87	89.31	2.50	0.40	95.74
	800	7.91	1.55	90.54	2.38	0.32	96.68
Epoxy/MDA	600	9.29	1.80	88.91	2.72	0.31	96.97
	700	7.28	1.73	90.99	2.50	0.20	97.30
	800	5.83	1.32	92.85	2.38	0.32	97.47
Epoxy/MTHPA	600	11.47	2.23	86.30	4.80	0.68	96.50
	700	9.85	1.85	88.30	4.40	0.47	95.13
	800	5.15	1.50	93.35	4.03	0.35	95.62

a: Volatile matter

b: Fixed carbon

and epoxy cured with and hydride. This may be concerned with the thermal behavior of thermoset plastic which cannot melt or remold like thermoplastic, but it degraded and formed brittle char covering fiber surface. However, the lowest volatile mater and fixed carbon in residue char due to the not heat stable of phthalic anhydride in its polymer chain with the increase of condensable liquid fraction at 600ºC. It should be concerned with this resin start to degrade at lower temperature. The proximate analysis results of 60 wt.% glass fiber solid residue seems to indicate that such residue hardly contained of

volatile and fixed carbon. It should be concerned with the higher loading glass fiber resulted in lower crosslinked density network.

4.2.2.2 The Characteristic of Condensable Liquids

The condensable liquids obtained from pyrolysis of composite wastes were viscous dark brown liquid as shown in Figure 4.18. They were first centrifuged at 3000 rpm for 30 min. to separate aqueous phase or water in such liquid. After water was removed, organic phases were then analyzed the functional group, the chemical composition, simulated distillation and fuel properties.

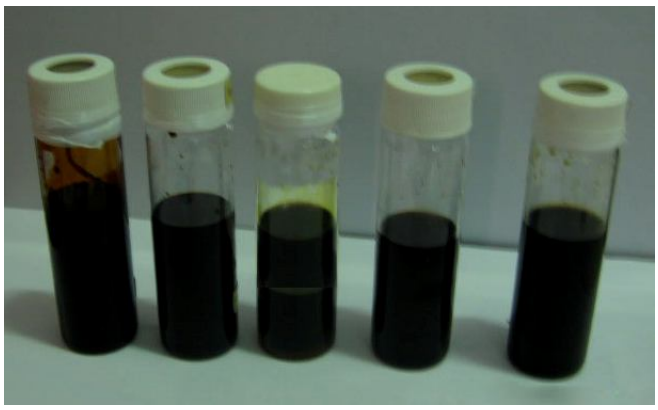


Figure 4.18 Condensable liquid obtained from pyrolysis composite wastes.

Functional Group Analysis of Condensable Liquids

The FTIR spectra produced from pyrolysis oil of unsaturated polyester composite reinforced with 30 wt.% glass fiber composites waste at pyrolysis temperature of at 600 and 800°C are presented in Figure 4.19 (a) and (b).

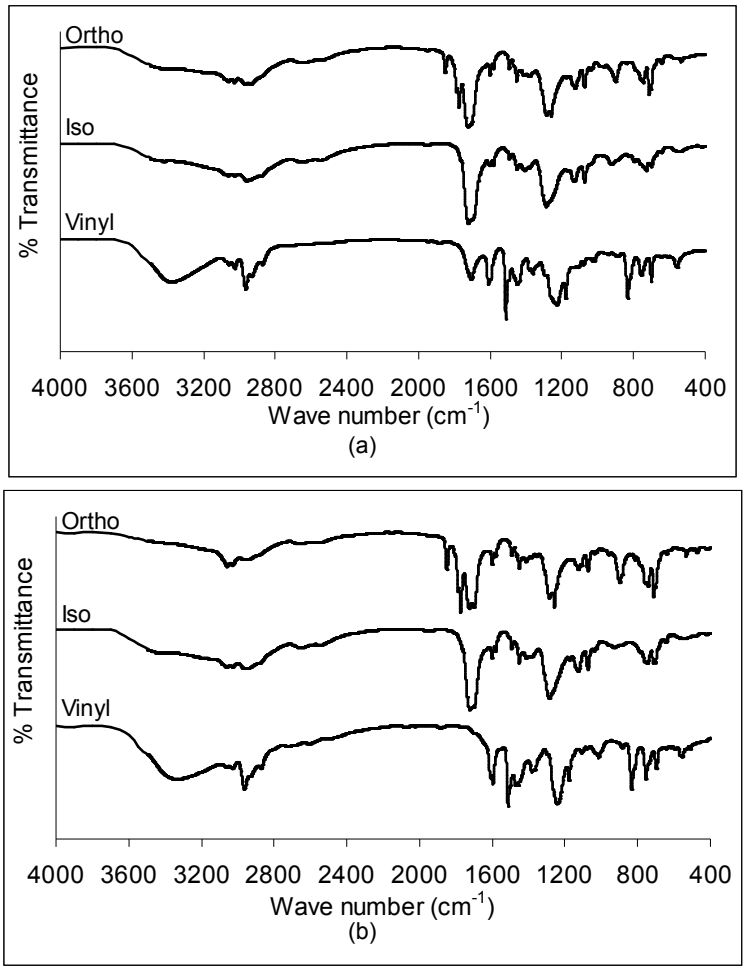


Figure 4.19 FTIR spectra of pyrolysis liquid oil obtained from pyrolysis of unsaturated polyester reinforced with 30 wt.% glass fiber composite waste at temperature of (a) 600 ºC, and (b) 800ºC.

The distinguished strong peaks were the broad peak at 3432-3437 cm^{-1} derived from O-H stretching of alcoholic compounds consisting of the breadth peaks which indicated to carboxylic groups. The prominent absorption peaks at 1730 and 1285 cm^{-1} were assigned to C=O and C-O stretching of carboxylic or ester compounds and may be used to identify oxygenated compounds in the decomposition products of polyester. The presence of sharp peaks at 1500-1600 and 700-910 cm^{-1} corresponded to skeletal vibrations and C-H bending of aromatic rings. The ratio of methyl groups (CH_3) at 2930 cm^{-1} to methylene groups (CH_2) at 2960 cm^{-1} was rather low which indicated a high proportion of methylene groups in the oil [46]. These spectra suggested that the oil

composed mainly of aromatic structure and oxygenated compounds derived from the chain scission of crosslinked bond and the breakdown of ester group (COO).

The effect of glass fiber content on FTIR spectra of pyrolysis liquid oil obtained from pyrolysis of unsaturated polyester reinforced with 30 and 60 wt % glass fiber under the same pyrolysis temperature of 700 ºC are presented in Figure 4.20 (a) and (b), respectively.

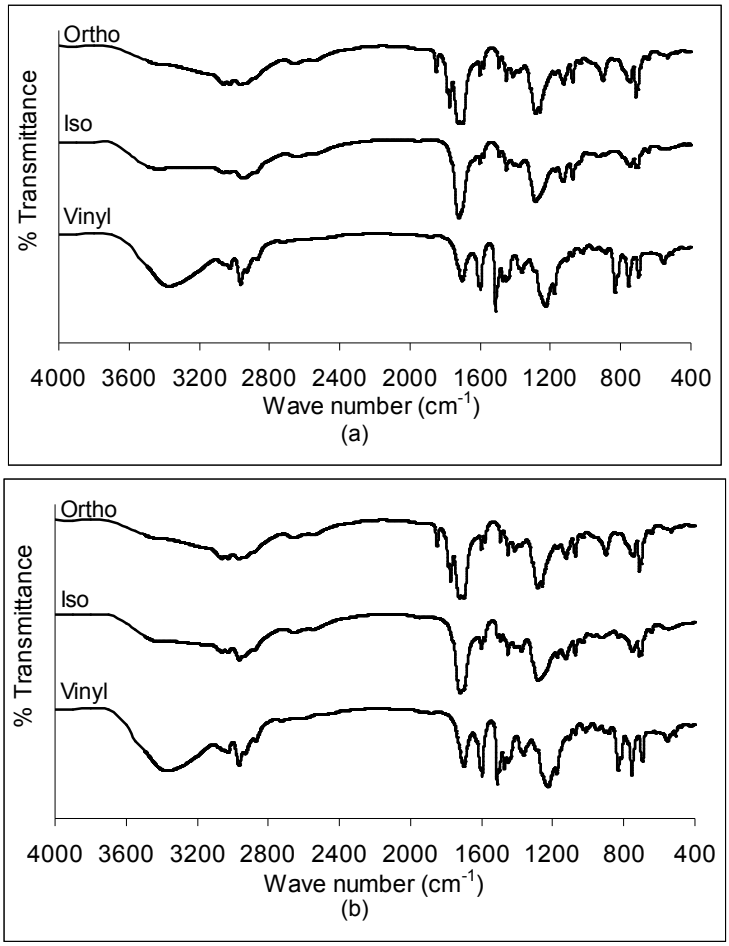


Figure 4.20 FTIR spectra of pyrolysis liquid oil obtained from pyrolysis of unsaturated polyester composite waste at temperature of 700ºC: (a) 30 wt.% glass fiber, and (b) 60 wt.% glass fiber.

From Figure 4.19 and 4.20, it can be suggested that pyrolysis temperature did not have significant correlation with aromatic content. The chemical structure of unsaturated polyester matrix on the their composition of liquid oil similarly composed of

aromatic and oxygenated compound but it may be different in its weight fraction which corresponded with Torres, et al. [13,15].

Functional group analysis of liquid oil from pyrolysis of epoxy composite at 600 and 800ºC are presented in Figure 4.21 (a) and (b), respectively.

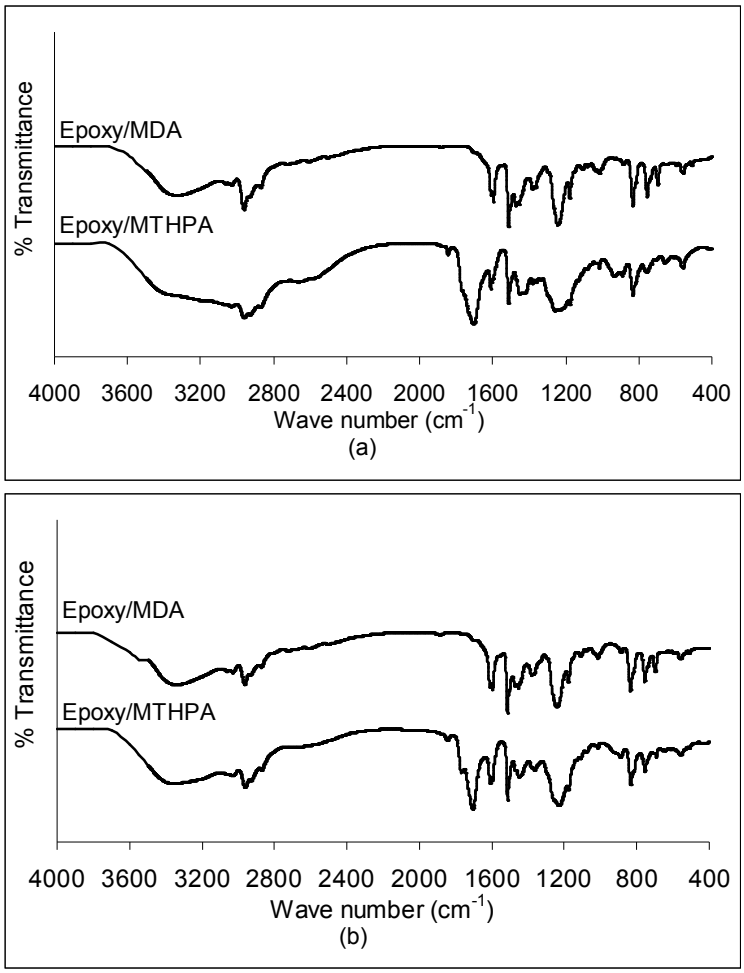


Figure 4.21 FTIR spectra of pyrolysis liquid oil obtained from pyrolysis of amine and anhydride cured epoxy reinforced with 30 wt.% glass fiber composite waste at temperature of (a) 600 ºC, and (b) 800ºC.

It was indicated that their compositions were similar to those seen from polyester due to the presence of the broader peak from O-H stretching of alcoholic compounds at 3432-3437 $cm^{-1}$and aromatic absorption peak at 1500-1630 cm^{-1}. However, it was seen that the ratio of groups (at 2930 cm^{-1}) to methylene groups (at 2960 cm^{-1}) and methyl groups was rather higher than unsaturated polyester, suggesting greater aliphatic chain.

The comparison of FTIR spectra of pyrolysis liquid oil of amine and anhydride cured epoxy is displayed in Figure 4.22. Both materials has similar alcoholic and oxygenated compound. Though, each one presented its characteristic absorption peak derived from the breakdown of chemical bonds in curing agent such as the presence of absorption peak C-N bond at 1100 cm^{-1} for amine curing and carbonyl ester C=O at 1730 and 1285 of C-O four anhydride structure.

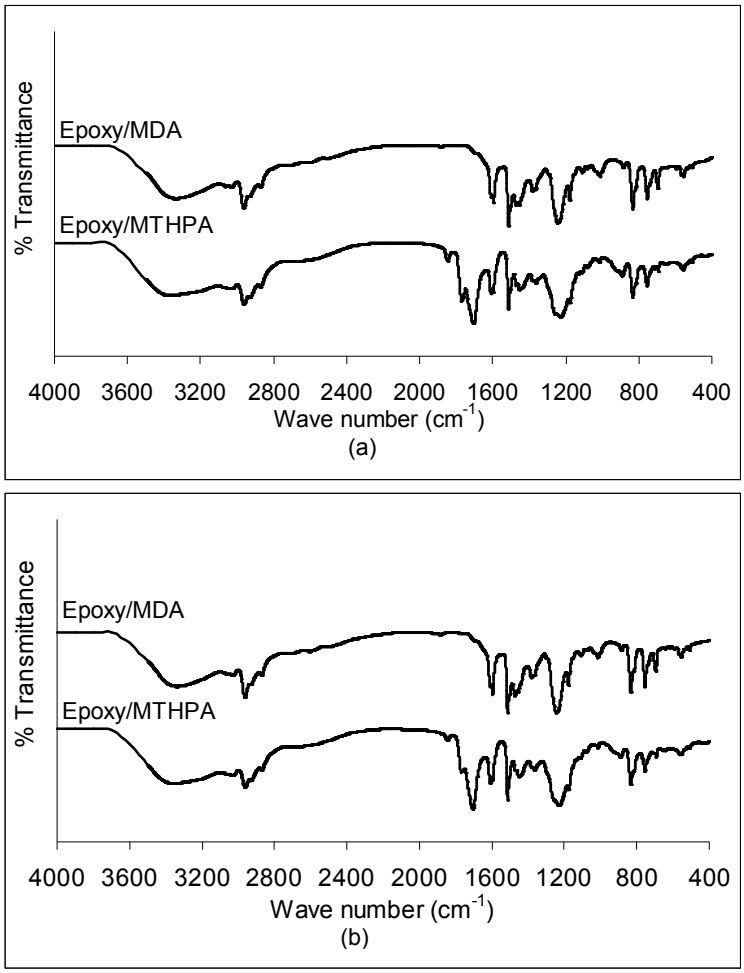


Figure 4.22 FTIR spectra of pyrolysis liquid oil obtained from pyrolysis of amine and anhydride cured epoxy composite at temperature of 700 ºC: (a) 30 wt.% glass fiber composite waste, and (b) 60 wt.% glass fiber.

Elemental Analysis of Condensable Liquids

After the water in condensable liquid was removed by centrifugation, the organic phase, referred to liquid oil, was analyzed for the amount of chemical

composition using LECO CHN-2000 analyzer and calculated for atomic ratio of hydrogen to carbon (H/C), and oxygen to carbon (H/C). Additionally, the liquid oil were measured for gross calorific value (GCV) by bomb calorimeter. These results are summarized in Table 4.9 and 4.10, respectively.

Table 4.9 Elemental analysis, water content, and gross calorific value (GCV) of condensable liquids obtained from pyrolysis process of composite wastes with 30 wt.% fiber.

Resin	Temperature	Elemental Analysis (wt %)				Atomic ratio		Water	GCV
Matrix	(ºC)	C	H	N	O*	H/C	O/C	(wt%)	(MJ/kg)
Ortho	600	72.00	6.42	-	21.58	1.07	0.22	5.0	28.20
	700	74.00	6.27	-	19.73	1.02	0.20	4.8	29.60
	800	75.30	6.20	-	14.43	0.99	0.18	4.5	30.60
Iso	600	67.53	7.40	-	25.07	1.31	0.28	3.80	25.90
	700	68.41	6.27	-	24.26	1.29	0.27	3.65	26.30
	800	70.62	6.57	-	23.04	1.08	0.24	3.10	27.70
Vinyl	600	79.34	7.40	-	12.88	1.18	0.12	4.50	29.20
	700	80.3	7.33	-	11.49	1.22	0.11	4.30	29.80
	800	81.72	6.34	-	10.31	1.17	0.09	3.90	30.70
Epoxy/	600	77.08	8.41	2.45	12.06	1.31	0.12	5.0	33.60
MDA	700	77.18	8.09	2.42	12.31	1.26	0.12	4.8	33.90
	800	78.53	7.66	2.63	11.18	1.17	0.11	4.5	34.60
Epoxy/	600	71.74	7.53	0	20.73	1.26	0.22	3.6	29.20
MTHPA	700	73.98	7.35	0	18.67	1.19	0.19	3.4	29.80
	800	80.00	7.25	0	12.75	1.09	0.12	3.0	30.70

*: oxygen and others (by difference)

Table 4.10 Elemental, water content, and gross calorific value (GCV) of condensable liquids obtained from pyrolysis process of composite wastes with 60 wt.% fiber.

Resin Matrix	Temperature	Elemental Analysis (wt %)				Atomic ratio		Water	GCV
Liquid	(ºC)	C	H	N	O*	H/C	O/C	(wt%)	(MJ/kg)
Ortho	600	72.19	7.74	-	21.00	1.11	0.21	4.20	25.90
	700	73.65	6.24	-	20.11	1.02	0.20	4.00	27.70
	800	75.09	5.93	-	18.98	0.95	0.19	3.5	29.20
Iso	600	73.51	6.98	-	19.51	1.14	0.20	3.40	29.50
	700	74.53	6.66	-	18.81	1.09	0.19	3.12	29.60
	800	76.31	6.56	-	17.13	1.03	0.17	2.80	29.80
Vinyl	600	78.00	7.34	-	14.70	1.13	0.14	4.0	30.60
	700	78.12	7.18	-	14.60	1.10	0.14	3.8	30.70
	800	80.16	6.99	-	12.85	1.05	0.12	2.5	31.70
Epoxy/MDA	600	76.33	9.55	2.11	12.01	1.50	o.12	4.80	32.40
	700	77.00	9.28	2.11	11.61	1.42	0.11	4.50	33.90
	800	78.39	8.19	2.38	11.04	1.25	0.10	4.10	34.40
Epoxy/MTHPA	600	73.50	7.09	-	19.41	1.16	0.20	3.2	33.60
	700	75.00	7.02	-	17.98	1.12	0.20	2.8	34.30
	800	76.00	6.24	-	17.76	0.99	0.20	2.5	34.30

*: oxygen and others (by difference)

It was observed from Table 4.8 and 4.9 that the difference among resin matrices and glass fiber content did not strongly effect the chemical composition of condensable liquids and their GCVs. The amount of carbon increased as function temperature corresponded with the decrease of carbon in solid residue. Hydrogen and hydrogen composition decreased as the increase of temperature due to further vapor to gas phase resulting in the higher in GCV. H/C atomic ratio obtained from pyrolysis oils of orthophthalic polyester composite were rather low (~1.0-1.1) which corresponded with Torres, et al. [14] who suggested that such oil is mainly of aromatic compound. Whereas other matrices showed more H/C atomic ratio (~1.2-1.3), suggesting that the oils were

less aromatics and more aliphatic hydrocarbon chain. In addition, it was suggested that water content in pyrolysis liquids were ranged of Torres, et al. [14] derived from the breakdown of COOH and may be from condenser system.

Simulated Distillation of Condensable Liquids

In order to evaluate the utility of fuel oil, the pyrolysis liquid oils derived from pyrolysis temperature of 600ºC of 30 wt.% glass fiber composite waste, the highest liquid yields, were selected to distill for analysis of the boiling point range of the products as crude petroleum oil by using GC Simulated Simulation CP3800. The distillation results are presented in Figure 4.23.

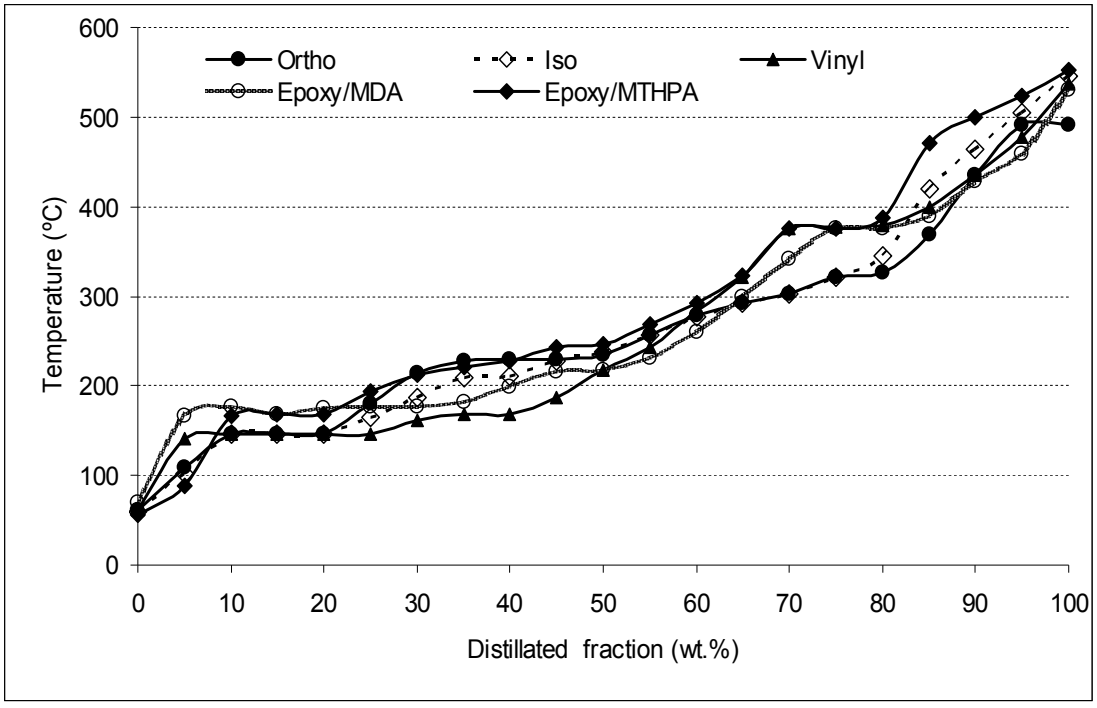


Figure 4.23 Distillated fractions of pyrolysis liquid oils obtained from pyrolysis of unsaturated and epoxy reinforced with 30 wt.% glass fiber composite waste at temperature of 600 ºC.

From the distillation curve in Figure 4.23, boiling point ranges of fuel product fractions are summmarized in Table 4.11. It shows that such liquid oil mainly consisted of hydrocarbon distillated of the boiling point range between 30 and 200 which referred

to the fraction of gases consisting of 1-4 carbon atoms and gasoline or naphtha which can be used for commercial petrol. This fraction is rich in aromatic. It was suggested that the liquid remained largely comprised of stable aromatic structure corresponded with the results of elemental analysis which showed H/C atomic ratio ranging between 1.0-1.1 (range of aromatic structure). However, the initial boiling of pyrolysis oil derived from pyrolysis of orthophthalic unsaturated polyester composite was 60ºC which is better than that reported by Torres, et al [13,14]. The oils having high initial boiling point temperature (70ºC) may cause problems during engine starting and warming up. Therefore, it should be mixed with light hydrocarbon fraction before used as petrol oil.

Table 4.11 The Distillated products of pyrolysis liquid oil obtained from pyrolysis of unsaturated and epoxy reinforced with 30 wt.% glass fiber composite waste at temperature of 600 ºC.

Distillated products	Boiling range (ºC)	Distilled fraction (wt.%)					Utility
		Ortho	Iso	Vinyl	Epoxy/MDA	Epoxy/MTHPA	
Gases/Naphtha	30-200	28	32	47	40	26	Gasoline oil
Kerosene	200-250	26	21	9	18	25	Kerosene oil
Light gas oil	250-300	15	15	7	7	11	Diesel oil
Heavy gas oil	300-370	17	7	6	8	8	Lubricant oil
Residue	>370	14	25	31	31	27	Asphalt

Furthermore, Table 4.11 also suggested that the distillated fraction is related to the type of resin. For polyester matrices, their thermal degradation were initiated by random scission of ester linkage and then further cracked into smaller molecule which may be in various forms such as phthalic anhydride ($C_6 H_4 (CO)_2O$), terephthalic acid (HOOC-ph-COOH), carbon dioxide(CO_2), propylene glycol ($HOCH_2CHOHCH_3$), propylene (CH_2=CH-CH_3), and methane (CH_4) according to Sarah [47]. According to Maxwell and Richard [33] the degradation products of amine cured resin occurred from the initial cleavage of the weakest C-N bonds and O-CH_2- were hydrogen, methane

propene, ammonia, methylamine, trimethylamine, water, phenol. Whereas the products generated from thermal degradation of epoxy cured with anhydride were suggested by Vlastaw, A. S. [32] which started by the dyhydration of water and further decomposed into carbon dioxide, toluene, pyridiene, acetone, methyl formate, hexatriene, methyl acetone, and butadiene.

In addition, the pyrolysis liquid oils, derived from pyrolysis temperature of 600ºC of 30 wt.% glass fiber composite waste, were also analyzed for the general properties of liquid fuel such as flash point, pour point, viscosity gross calorific value (GCV) specific gravity, and acidity and compared to other fuels as presented in Table 4.12. Pour point and viscosity of liquid oil derived from pyrolysis process of ortho and iso phthalic unsaturated polyester composite waste were clearly lower than that of vinyl or epoxy composite. This is because the oil of ortho and iso phthalic polyester mainly consisted of waxy crystal compound than that of vinyl or epoxy composite. It can be observed that the fuel properties of pyrolysis vinyl oil were ranged between ortho or iso phthalic unsaturated polyester and epoxy resin which corresponded with Cunliffee, et al. [48]. As mentioned in chapter II, vinyl ester resin was the modified resin of unsaturated polyester and epoxy resin which essentially comprise an epoxy-based and crylic ester end groups.

Table 4.12 also showed acid value of pyrolysis oil from unsaturated polyester. Flash point of pyrolysis oil of ortho or iso unsaturated polyester and vinyl ester were lower than that of epoxy and convention fuel oil. Scheirs and Kaminsky [46] explained that the original feed stock of unsaturated polyester contained styrene monomer which was sensitively ignitable material. However, Cunliffe and Williams [16] suggested that these pyrolysis oils can be recycled as chemical feedstock for polyester resin production process.

From comparison on acidity of condensable liquid oil derived from pyrolysis process of unsaturated, epoxy composite wastes and conventional oil, it can be seen that the liquid oil from ortho and iso phthalic polyester are significantly greater than that of epoxy and conventional oil. Recalling to the chemical compositions of pyrolysis liquid

presented in Table 4.9, two of these oils largely consisted of oxygen content with may be in formed of acid (COOH). The presence of high acidity in liquid oil resulted in low gross calorific value and may cause corrosion of engine tank and formation of gum.[49]

Table 4.12 Fuel properties of pyrolysis liquid oil obtained from pyrolysis of unsaturated and epoxy reinforced with 30 wt.% glass fiber composite waste at temperature of 600ºC compared to other conventional fuels.

Properties:	Flash point	Pour point	Viscosity[a]	GCV[b]	SG[c]	Acidity
Unit:	(ºC)	(ºC)	(CPs.)	(MJ/kg)	(g/cm^3)	(mgKOH/oil)
ASTM	D6450	D97	D 445	D240	D4052	D644
This research						
Ortho	44.4	<-20	21.5	29.4	1.3	141.2
Iso	45.3	<-20	19.5	25.9	1.1	155.1
Vinyl	36.4	3	15.3	33.5	1.1	34.8
Epoxy/MDA	84.0	3.1	74.7	37.2	1.1	8.81
Epoxy/MTHPA	84.3	7.9	70	34	1.2	88.2
[9][d]	26	NR[e]	3.9	33.6	0.83	NR[e]
[28][f]	>52	<10	1.8-4.1	16-26	0.81-0.87	NR[e]
[29][g]	40-70	NR[e]	3.5-5	14-16	NR[e]	<0.5

a: @40ºC

b : Gross calorific value

c: Specific density at15ºC

d: Liquid oil obtained from pyrolysis of 30 wt.% glass fiber orthophthalic polyester at 450ºC

e: Not recommend

f: Automotive diesel oil

g: Bio diesel

4.2.2.3 The Characteristic of Gaseous Product

The gaseous products from pyrolysis of composite wastes collected in gas sampling bags were analyzed and quantified by Gas chromatography (GC) SRI 8610C. The analysis results of evolution gases from pyrolysis process of unsaturated polyester

reinforced with 30 and 60 wt.% glass fiber composite wastes in relation to final pyrolysis temperature are presented in Figure 4..24 and 4.25, respectively. It was demonstrated that product of gases were rich in carbon dioxide (CO_2); 60-70 vol.% which directly derived from the breakdown of ester bonds (-COO-) with resin, methane(CH_4); 30-40 vol.%, and some amount of carbon monoxide (CO), and hydrogen (H_2).

The influence of final temperature on gas composition, Figures 4.24 and 4.25 show that carbon dioxide decreased with the increase of pyrolysis temperature from 600 to 800ºC due to secondary decomposition into carbon monoxide. Significant increased of carbon monoxide resulted from both secondary reaction decomposition of carbon dioxide and carbonization of atomic structure in solid residue at elevated temperature. Further cracking of evolved volatile matter [15] also resulted in greater yield of methane and hydrogen.

Gaseous products generated from pyrolysis of epoxy composite waste are shown in Figure 4.26 which largely consisted of methane gas (50-60 vol.%) derived from the breakdown of alkyl groups in the epoxy chain [6,15,16], carbon dioxide (40-50 vol.%), carbon monoxide (3-5 vol.%) and some amount of hydrogen gas (0.1-0.2 vol%)

The comparison of gross calorific value (GCV) of generated gas from various resin matrix reinforced with 30 and 60 wt.% glass at pyrolysis temperature from 600 to 800ºC are displayed in Figure 4.27 (a-c).

It can be seen that the GCVs of evolution gases from composite consisted of 30 wt.% glass fiber were slight higher than that of 60 wt.% glass fiber, resulting from larger in volumetric gas evolution. The GCVs of nitrogen free gaseous product involved from pyrolysis 30 wt.% glass fiber reinforced unsaturated polyester composite wastes were ranging from 12-14 MJ/m^3 and comparable to previous literature [13,15].

The GCVs of gas obtained from pyrolysis of epoxy cured amine and anhydride composite waste shown in Figure 4.27 were no significantly different (17-21 MJ/m^3) which clearly higher than that of unsaturated polyester composite due to greater methane gas..

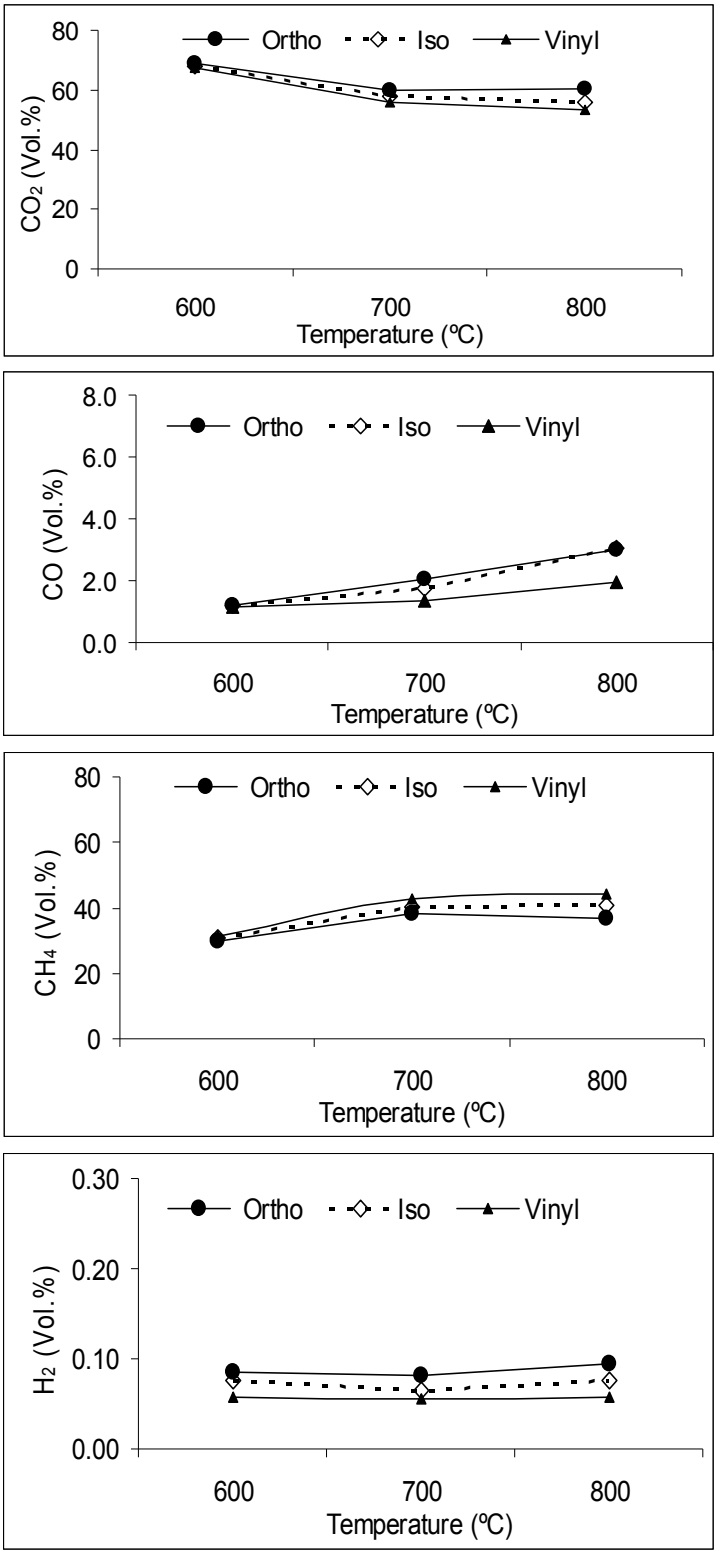


Figure 4.24 Gaseous product (nitrogen free) involved from pyrolysis process of three types unsaturated polyester reinforced with 30 wt% glass fiber composite wastes.

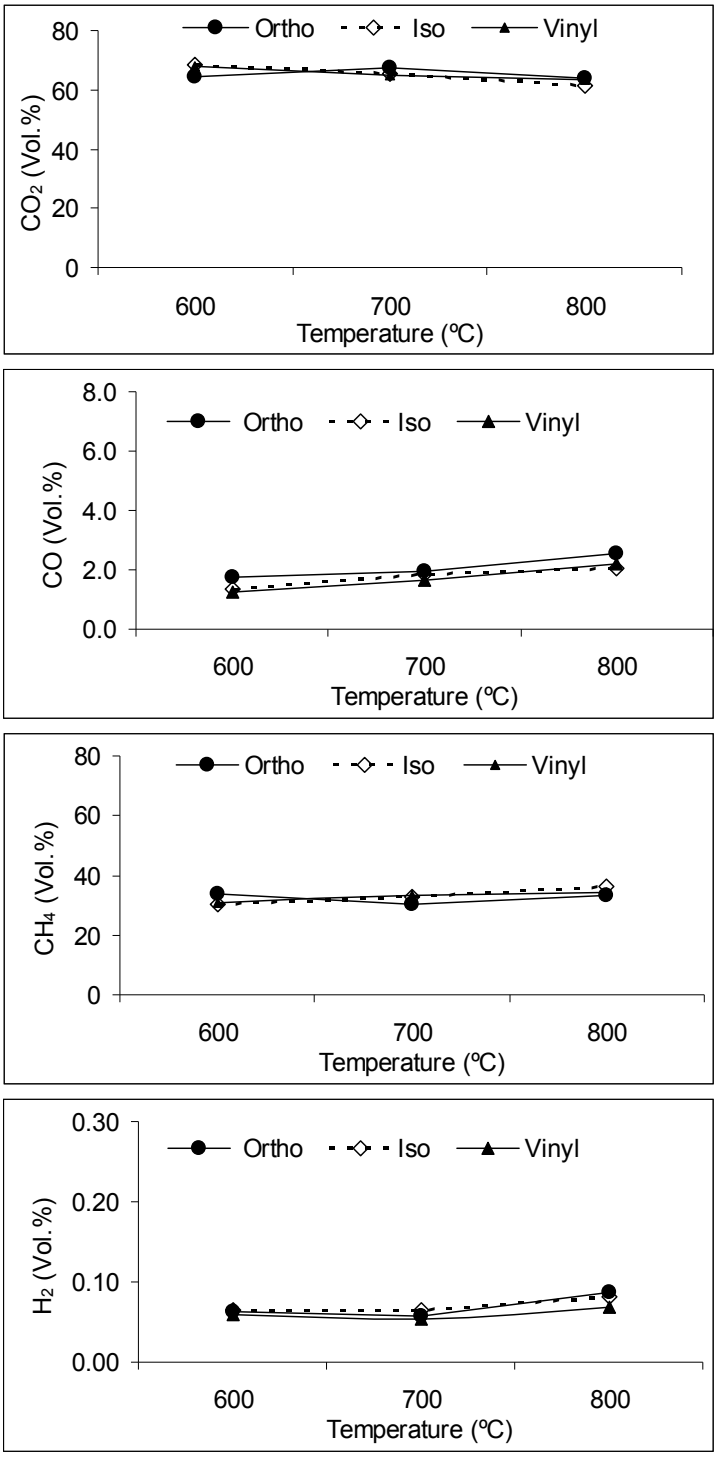


Figure 4.25 Gaseous product (nitrogen free) involved from pyrolysis process of three types unsaturated polyester reinforced with 60 wt% glass fiber composite wastes.

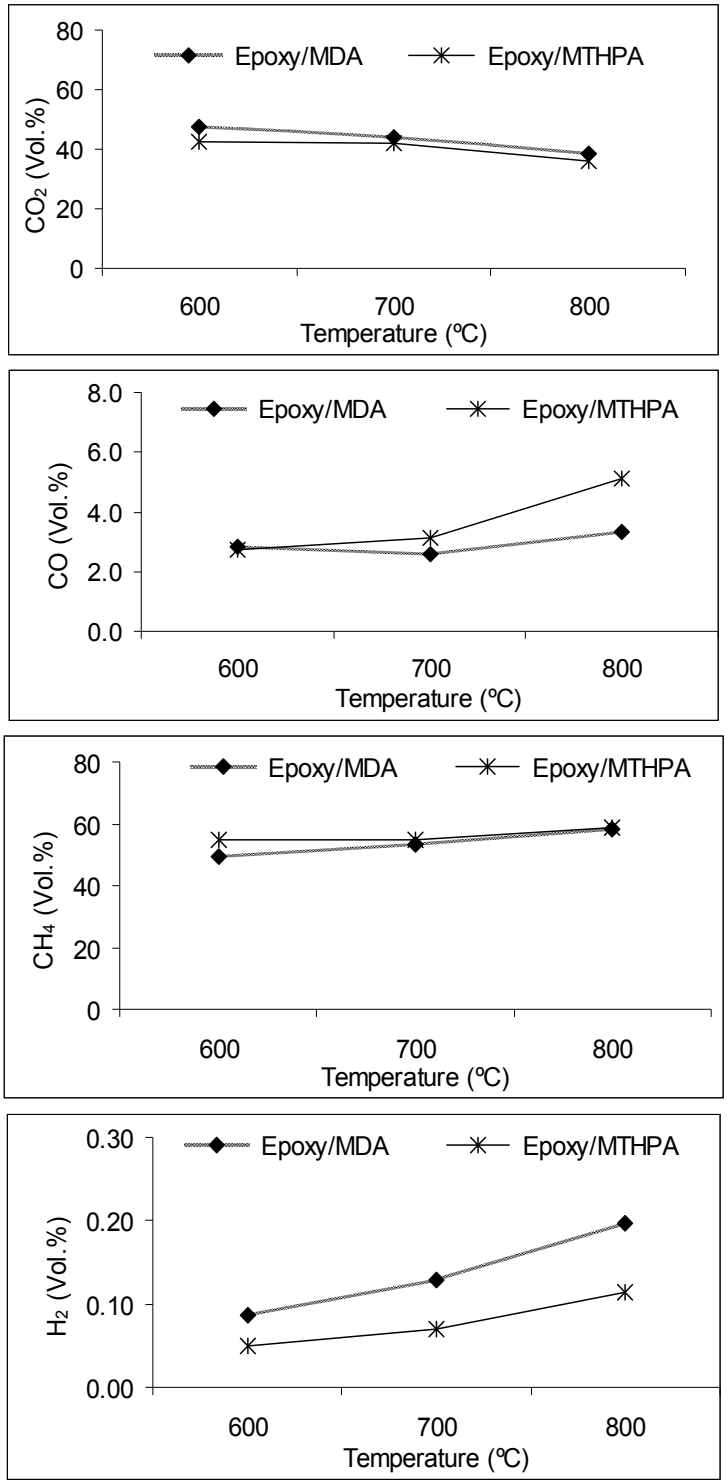


Figure 4.26 Gaseous product (nitrogen free) involved from pyrolysis process of amine and anhydride cured epoxy reinforced with 30 wt% glass fiber cured composite wastes.

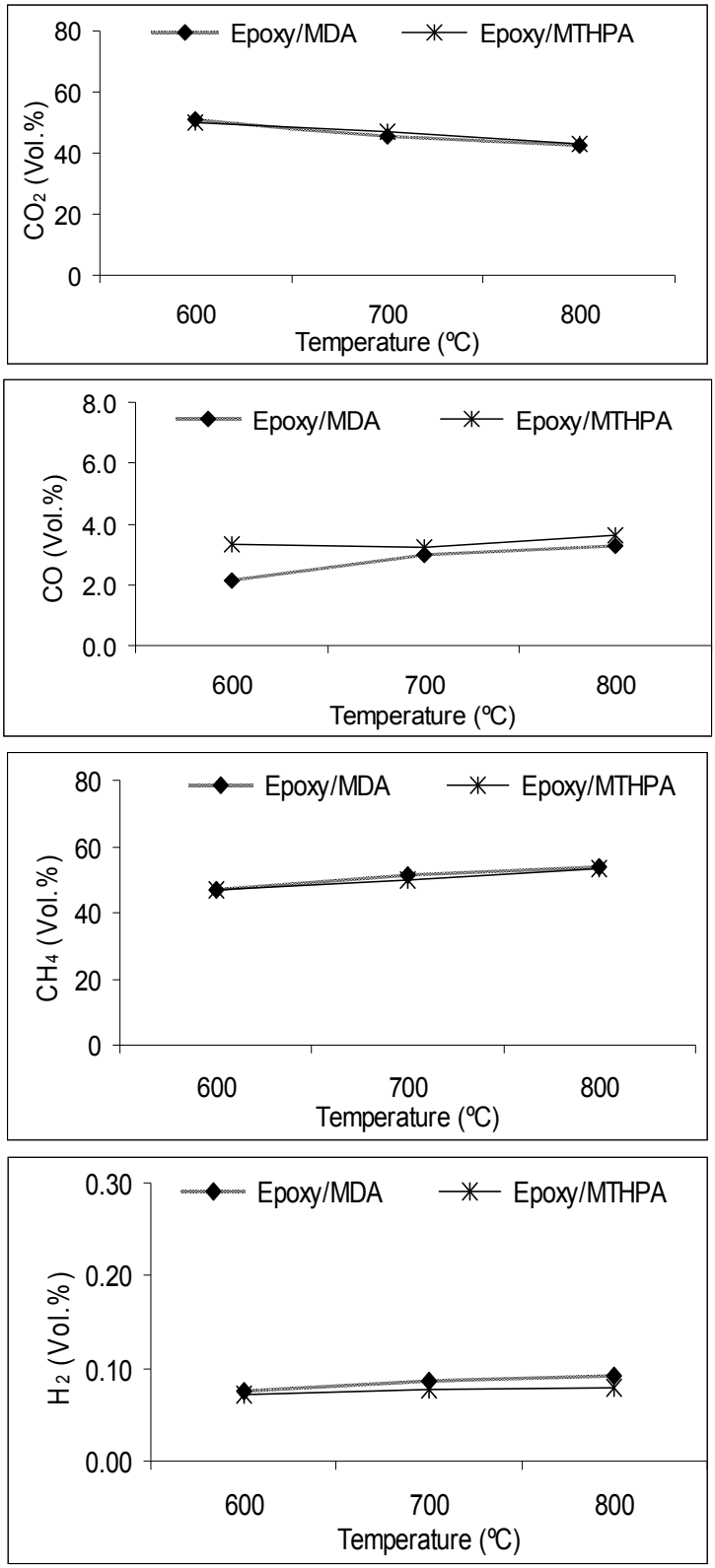


Figure 4.27 Gaseous product (nitrogen free) involved from pyrolysis process of amine and anhydride cured epoxy reinforced with 60 wt% glass fiber cured composite wastes.

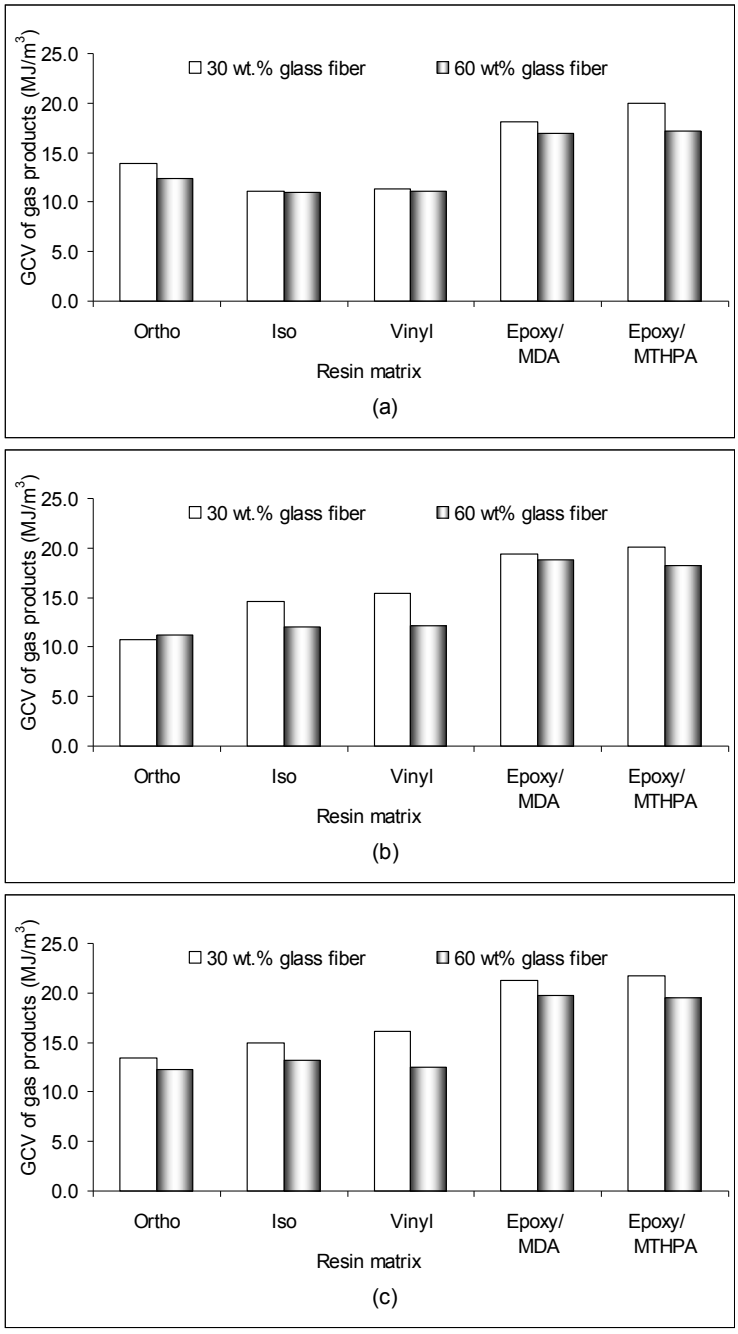


Figure 4.28 The comparison of nitrogen free gross calorific value (GCV) of gaseous products obtained from pyrolysis of unsaturated polyester and epoxy reinforced with 30 and 60 wt% glass fiber at pyrolysis temperature of (a) 600ºC, (b) 700ºC, and (c) 800ºC.

4.3 Characterization of Composites Prepared from Original Glass Fiber and Recycled Glass Fiber from Pyrolysis Process

This section presents the mechanical properties in terms of flexural and impact properties of composite prepared from original glass fiber (125 μm) and recycled glass fiber (mean length of 130-200 μm and particle size of 420 μm) obtained from pyrolysis of 60 wt.% glass fiber composite waste samples at temperature of 600 and 800 ºC in order to investigate the effect of pyrolysis temperature and the proportion of recycled fiber that can be used for replacing original fiber with no significant effect on mechanical properties . The proportion of fibers was varied from 5 to 20 wt. % of the total composite weight. Furthermore, the fractured surface of three-point bending test specimens was observed by Scanning Electron Microscope (SEM) to investigate the relationship between interfacial bonding of fiber- matrix and strength.

4.3.1 Flexural Properties

The flexural properties in terms of flexural strength and flexural modulus of unsaturated polyester and epoxy reinforced with original glass fiber and recovered glass fiber obtained from pyrolysis process were tested by using a Universal Testing Machine (LLOYD LR 100 N) in form of flat wise specimens according to ASTM D790-03 standard method for three point bending test.

4.3.1.1 Flexural Strength

The flexural strength of bulk molding compound (BMC) unsaturated polyester reinforced with original and recycled glass fiber at various content varied from 5 to 20 wt.% are presented in Figure 4.29. The results showed that vinyl ester resin had higher flexural strength than isophthalic and orthophthalic polyester resin about 9 and 27%, respectively, which can be related to their its fracture surface morphology by SEM images in Figure 4.30 (a-c). It was revealed that fracture surface of vinyl ester and isophathalic polyester cured resin matrix showed rib-like of fracture path propagation, especially vinyl ester resin, while orthophthalic polyester resin showed smooth and

presence of spot which was defection area leading to lower flexural strength than the other two resins.

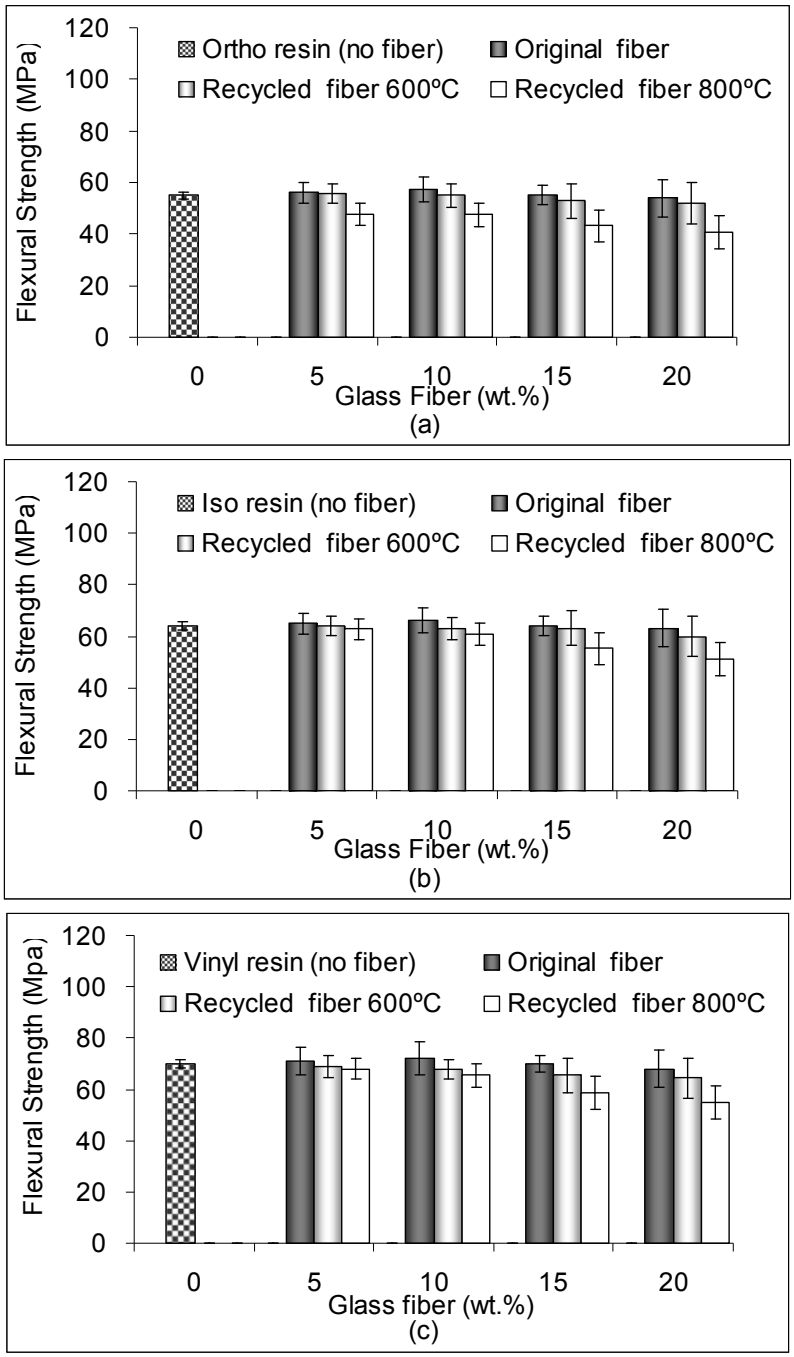


Figure 4.29 Effect of glass fiber content on flexural strength of unsaturated polyester resin and BMC composite: (a) orthophthalic polyester, (b) isophthalic polyester, and (c) vinyl ester composites.

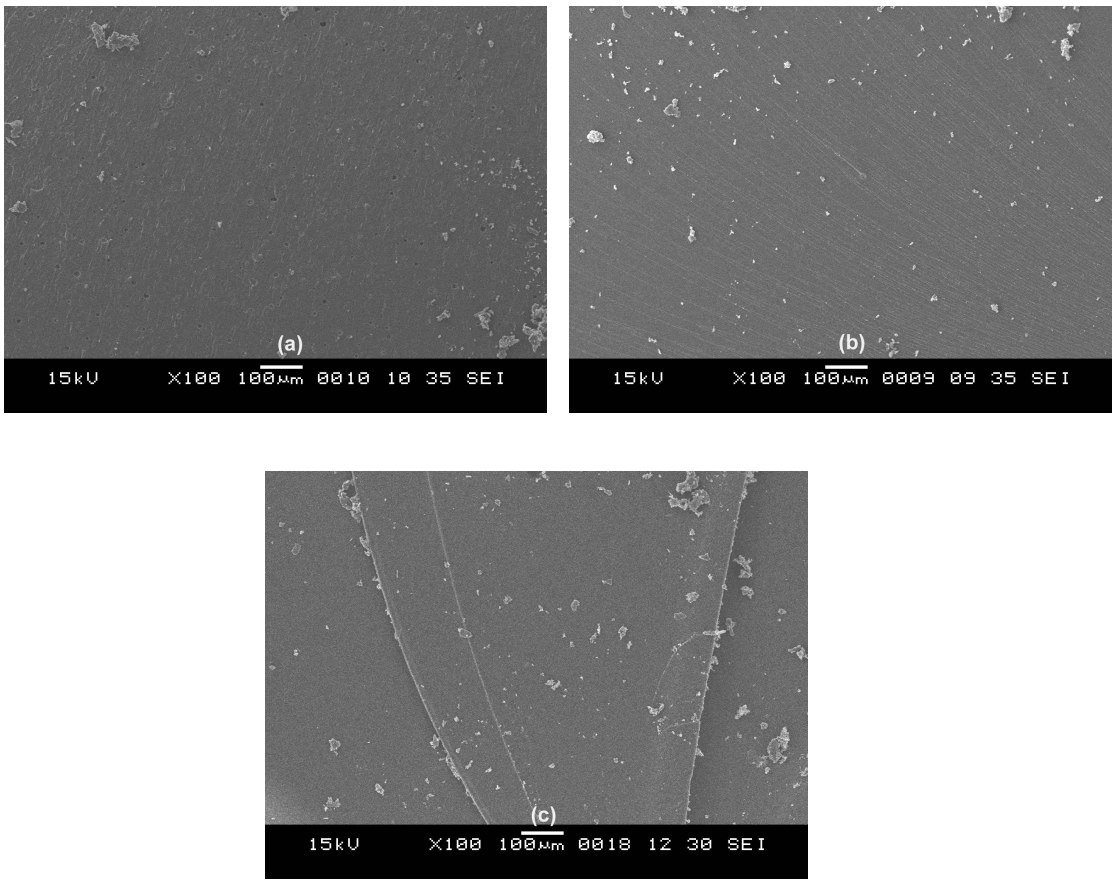
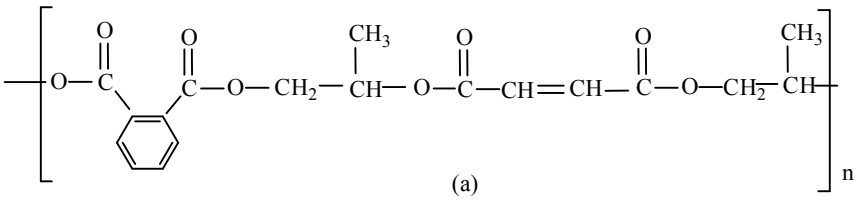
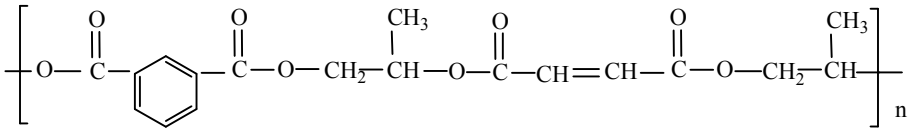


Figure 4.30 SEM micrographs of fracture surface from three point bending test specimens of unsaturated polyester resin: (a) orthophthalic polyester, (b) isophthalic polyester, and (c) vinyl ester resin.

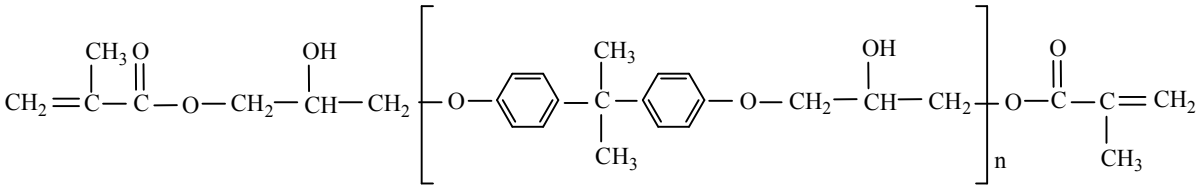
This is resulted from the difference in their chemical structure which are presented in Figure 4.31 It can be seen that the chemical structure of vinyl ester resin consisted of bisphenol A in its straight main chain which is very stiff and strong and high rigidity structure resulting in higher flexural strength than the other two resins. Visco, et. al. [50] indicated that the steric hindrance structure of orthophthalic polyester resulted in lower density structure which attributed to its lower flexural strength comparing to the isophthalic polyester resin. In addition, The chemical structure of isophthalic polyester showed higher symmetry than that of orthophthalic polyester, leading to higher strength when applied stress.



(a)



(b)



(c)

Figure 4.31 Chemical structure of three types unsaturated polyester resin: (a) orthophthalic polyester, (b) isophthalic polyester resin, and (c) vinyl ester resin.

For the effect of different curing agent on flexural strength of epoxy resin, as presented in Figure 4.32, it was found that the amine (MDA) cured epoxy resin had lower flexural strength than of and anhydride (MTHPA) cured epoxy resin of about 20% which was resulted from a few reasons. Firstly, because amine curing agent had higher viscosity (400 centipoise, cps.) than anhydride curing agent (260 cps) which affected to the homogeneity of resin mixed at room temperature. Secondary, the pot life of resin mixed with amine curing was short which resulted in the fast and into high exothermic reaction, In additin, unreacted molecules may be presented which were trapped into network structure during curing reaction as white haze sports, contributing to weak point when applying load as shown in SEM image in Figure 4.33 (a) Finally, is due to its chemical structure was cycloaphthic amine which preeminently showed in terms of toughness .While anhydride curing which low viscosity and can be reacted with epoxy

resin at high temperature (150ºC) leading to more homogeneity of resin mixed as the results from SEM image in Figure 4.33 (b) Furthermore, the presence of benzene ring in its structure as shown in Figure 4.34 providing good flexural strength.

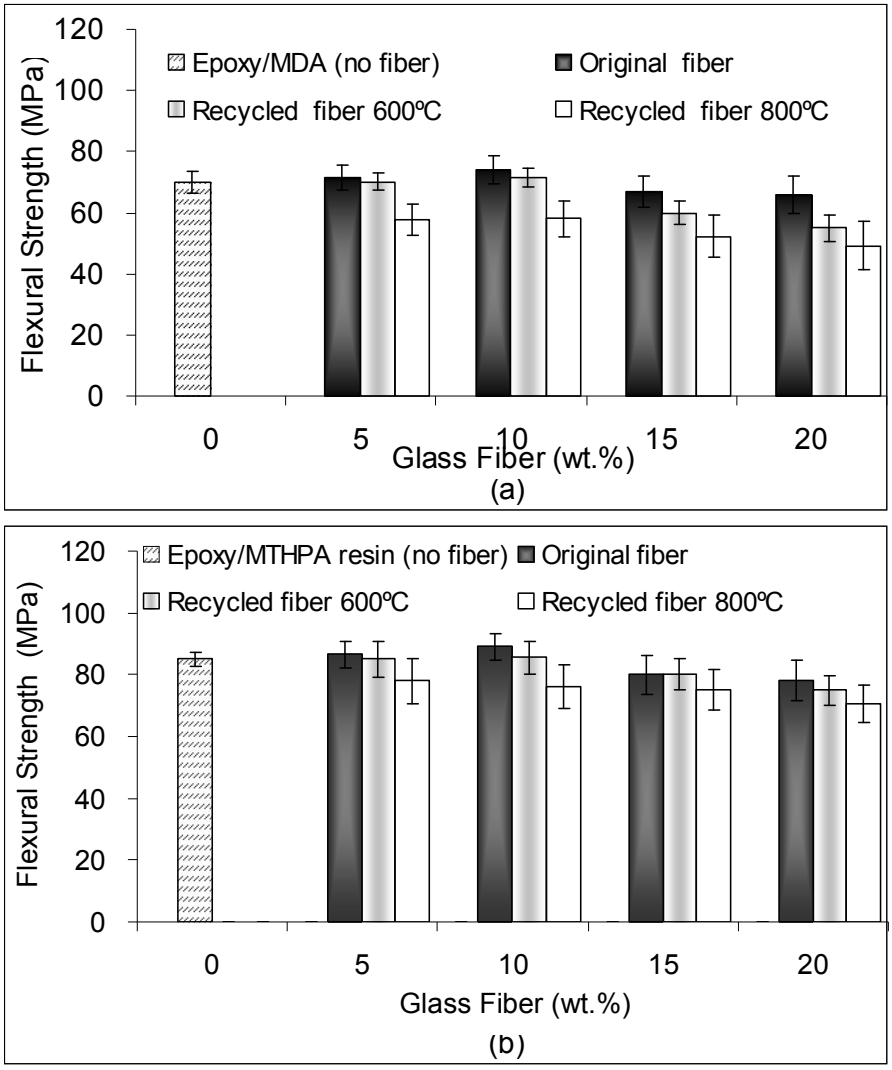


Figure 4.32 Effect of glass fiber content on flexural strength of epoxy resin and BMC composite: (a)epoxy/MDA, and (b) epoxy/MTHPA composites.

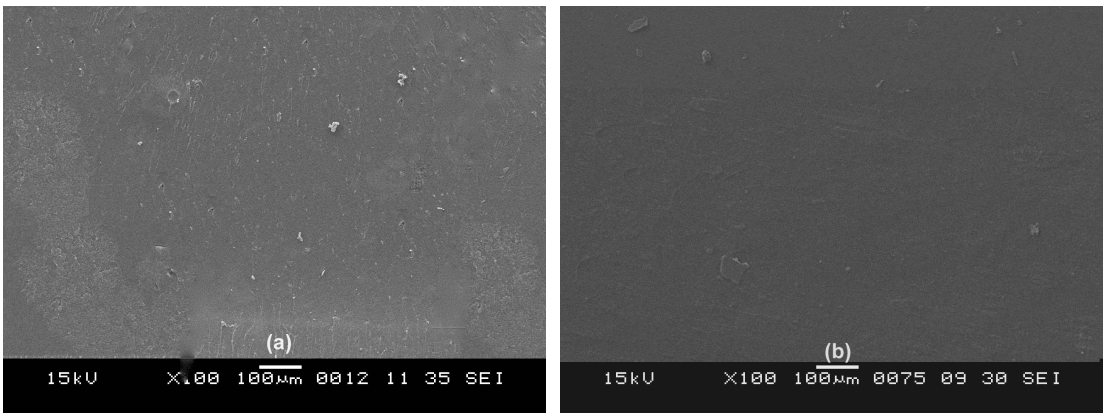
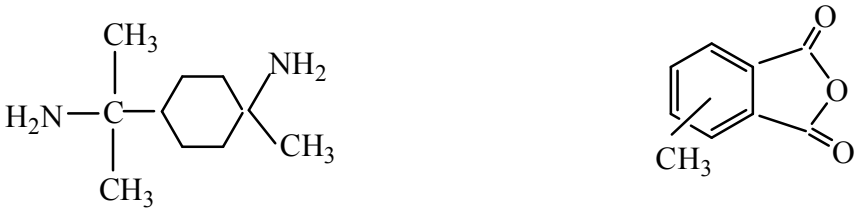


Figure 4.33 SEM micrographs of fracture surface from three point bending test specimens of of epoxy resin: (a) epoxy/MDA and (b)epoxy/MTHPA.



MDA MTHPA

Figure 4.34 Chemical structure of Methanediamine (MDA) and Methyltetra hydrophthalic anhydride (MTHPA).

According to rule of mixture [10], it was observed from Figure 4.29 and 4.32 that the flexural strength of both BMC composites containing original glass fiber were increased 2-5 % when glass fiber content was significantly increased from 5 to 10 wt% but it was slightly decreased (1-2%) when the original glass fiber content was increased to 20 wt% This result may be affected from the agglomeration of original glass fiber as presented in SEM images in Figure 4.35 and 4.36, resulting in the reduction of strength at fiber fraction from 15 -20 wt%. However, their flexural strength was shill higher than that of neat resin and no significant effect from the difference types of unsaturated polyester and epoxy curing agent was observed.

Considering the flexural strength of both BMC composites reinforcing with recycled glass fiber from pyrolysis process at temperature of 600 and 800ºC, unlike the

BMC composites having original glass fiber, upon increasing recycled glass fiber content, their flexural strength was decreased. In addition, it was found that the flexural strength of both BMC composites containing 20wt% recycled glass fiber at 800ºC were clearly lower than that of the BMC composites containing original glass fiber (10 - 15%) and recycled fiber 600 ºC(3-7%). It can be seen from SEM images in Figure 4.37 and 4.38 respectively, that the fracture surface of the unsaturated polyester and epoxy composites prepared from recycled glass fiber at 600 and 800ºC showed poor interfacial bonding between matrix and glass fiber as evidenced by the presence of many holes. Particularly, the composites containing recycled glass fiber at 800ºC showed several tiny holes greater than the ones with recycled glass fiber at 600ºC.

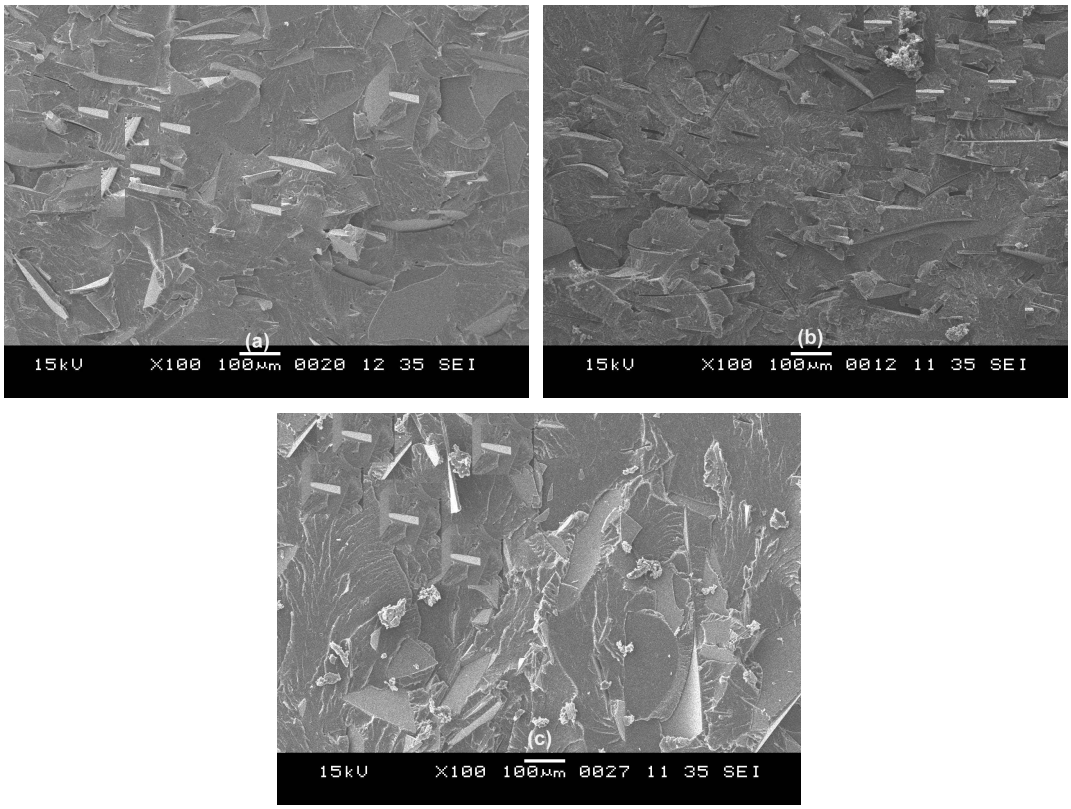


Figure 4.35 SEM micrographs of fracture surface from three point bending test specimens of unsaturated polyester composite: (a)orthophthalic polyester with 15 wt% original glass fiber, (b) isophthalic polyester 15 wt% original glass fiber, and (c) vinyl ester 15 wt% original glass fiber composites.

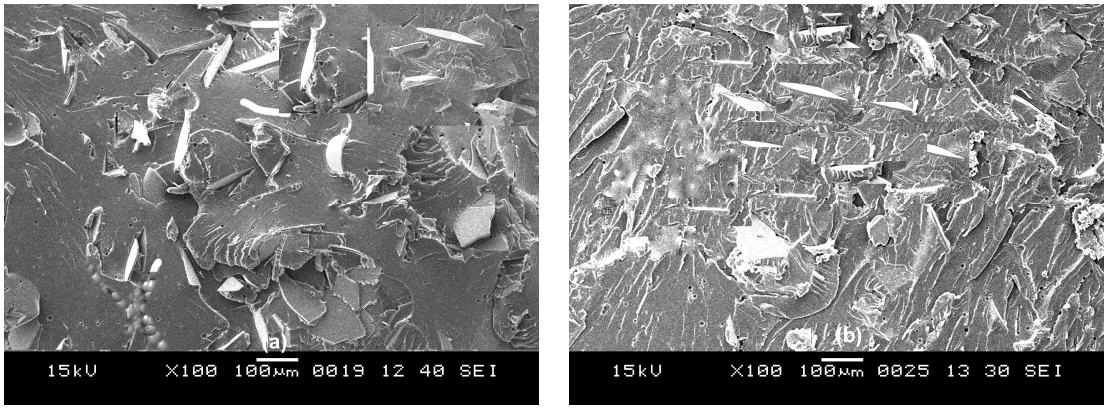


Figure 4.36 SEM micrographs of fracture surface from three point bending test specimens of epoxy composite: (a) Epoxy/MDA with15 wt% original glass fiber, (b) Epoxy/MTHPA with15 wt% original glass fiber composites.

These holes and voids were resulted from the lack of adhesion between the two phases; as a result, the pulled out of fiber under load was occurred which was more clearly observed in recycled glass fiber 800ºC as presented in Figure 4.37 and 4.38 (b).This is an important factor having impact on mechanical strength of composite material. The reduction in strength of BMC composites with recycled glass fiber may be. resulted from the breakage of fiber during applied load due to thermal degradation by pyrosis process Moreover, the coupling agent of glass fiber, which was widely used for unsaturated polyester and epoxy resin as presented in Figure 4.39, was probably degraded at these pyrolyzed tem perature, leaving the recycled glass fiber without any surface treatment, which was usually helped promoting the interfacial adhesion between matrix and fiber.

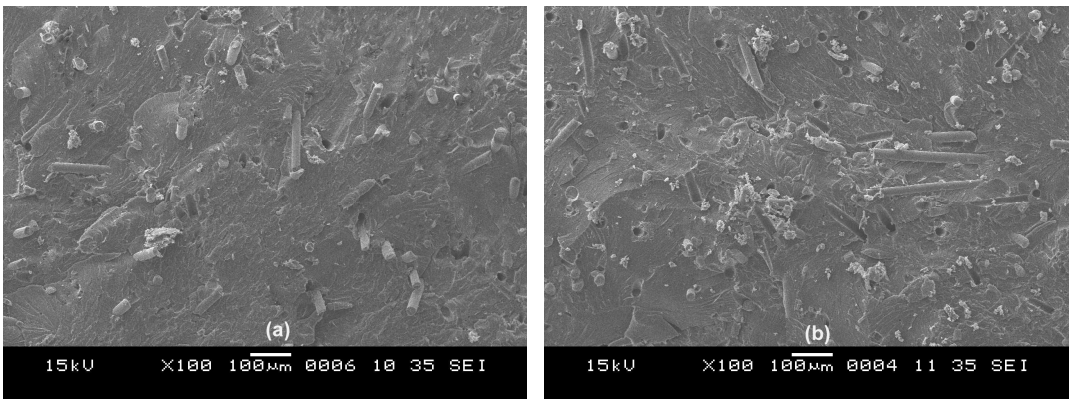


Figure 4.37 SEM micrographs of fracture surface from three point bending test specimens of (a) orthophthalic polyester with 15 wt% recycled glass fiber (600ºC) composite and (b) orthophthalic polyester with 15 wt% recycled glass fiber (800ºC) composite.

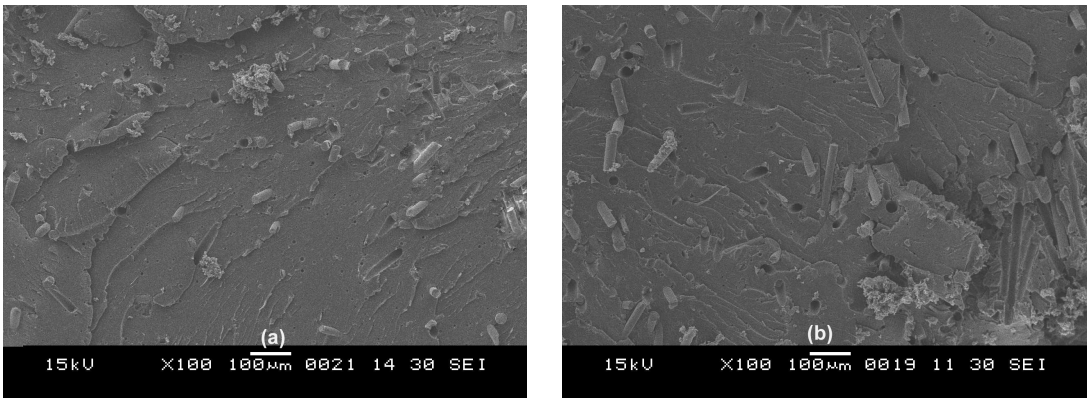
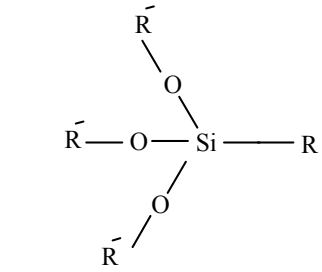


Figure 4.38 SEM micrographs of fracture surface from three point bending test specimens of (a) epoxy/MDA with 15 wt% recycled glass fiber (600ºC) composite and (b) epoxy/MDA with 15 wt% recycled glass fiber (800ºC) composite.



Where R is CH_2 or CH_3

$\bar{R}$ is H_2C(O)$CH\,CH_2OCH_2CH_2CH_2$ for epoxy marix, and

CH_2=CH for unsaturated polyester matrix

Figure 4.39 Typical coupling agent for unsaturated polyester and epoxy resin base [53].

4.3.1.2 Flexural Modulus

The flexural modulus of three types unsaturated polyester resin matrix is presented in Figure 4.40.similar to flexural strength, their modulus values were different depending on their chemical structure. Vinyl ester resin had higher flexural modulus value than isophthalic and orthophthalic polyester resin about 25 and 65 %, respectively. This result was clearly attributed to the effect of chemical structure as previously mentioned on flexural strength.

Similar to flexural strength, the flexural modulus of epoxy resin matrix was significantly affected from the different curing agent as previously discussed. It was found from Figure 4.41 that epoxy cured with anhydride curing agent showed ap proximatly 15% higher in flexural modulus than that of epoxy cured with amine about 15 %.

Considering the flexural modulus of all BMC composites containing original glass fiber, it was found that their modulus was increased with increasing original glass fiber content, suggesting that BMC composites became stiffer, similar to other previous works [52-53].

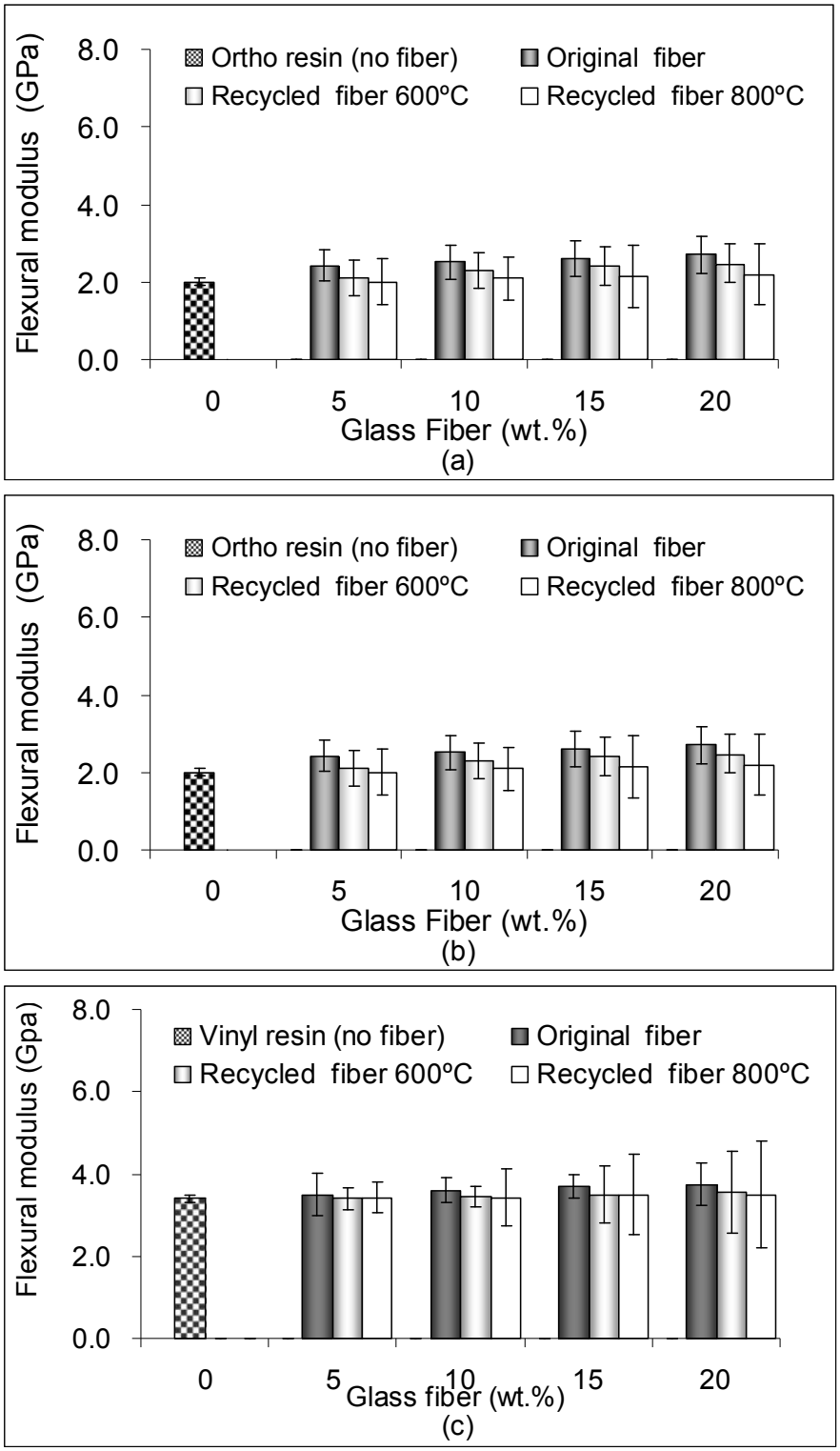


Figure 4.40 Effect of glass fiber content on flexural modulus of unsaturated polyester resin and BMC composite:(a) orthophthalic polyester, (b) isophthalic polyester, and (c) vinyl ester composites.

This is also based on rule of mixture which was proved that the elastic modulus of composite related to elastic moduli of fiber and matrix and their volume fraction of reinforcement material. The increase in flexural modulus of each unsaturated polyester containing original glass fiber was different depending on type of unsaturated polyester matrix.

Comparing with the resin matrix (o wt% fiber), it was found that the flexural modulus of orthophthalic polyester composite was significantly increased from 20 to 35 % when the original glass fiber was increased from 5 to 20 wt% While it was increased from 10 to 20 % for isophthalic unsaturated polyester matrix and from 6 to 10% for vinyl ester resin matrix. Similarly, the increase in flexural modulus of epoxy composite depended on type of curing agent the flexural modulus of Epoxy/MDA composites was increased from 7 to 25% when the original glass fiber was increased from 5 to 20 wt% compared to neat resin. While those of epoxy/MTHPA were increased from 2-20 %. The significantly increased in flexural modulus may by resulted from the stiffness of original glass fiber.

Regarding to the effect of recycled glass fiber, it was found that the flexural modulus of both BMC composites showed similar trend as the ones with original glass fiber that the flexural modulus was increased as increasing of recycled glass fiber content. However, the percentage of increasing in the flexural modulus was significantly depended on pyrolyzed glass fiber temperature. It was found that the recycled glass fiber at 800ºC. had less effct on in creasing in flexural less than that of recycled glass fiber 600ºC, which was affected from the loss of sizing during pyrolysis at higher temperature.

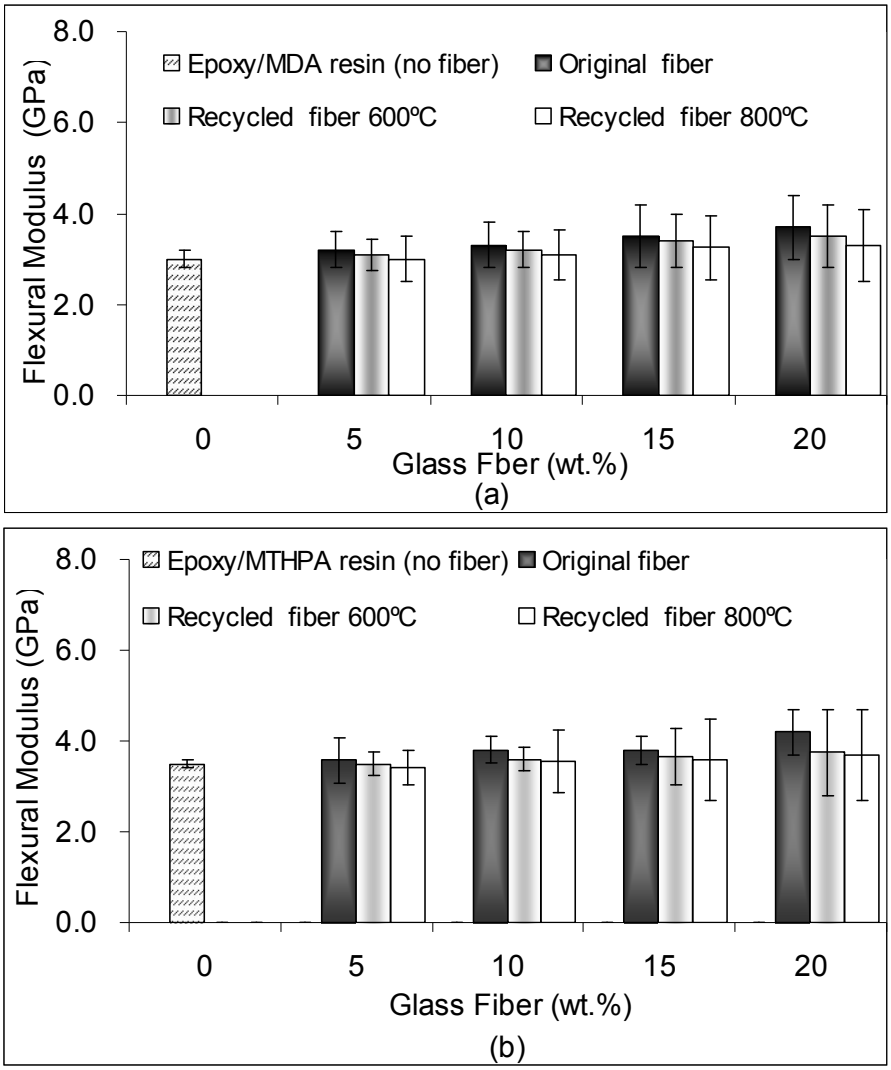


Figure 4.41 Effect of glass fiber content on flexural modulus of epoxy resin and BMC composite: (a) epoxy/MDA and (b) epoxy/MTHPA composites.

4.3.2 Impact Properties

The impact properties of unsaturated polyester and epoxy resins and their BMC composites carried out by impact tester (Zwick) according to ASTM D256-04 were presented in Figures 4.42 and 4.43, respectively. Similar to flexural properties, the impact strength of vinyl ester matrix was greater than that of isophthalic and orthophthalic approximately 7 and 25 % respectively. This is due to the effect of chemical structure of vinyl ester which was based on a bisphenol A backbone as

previously mentioned. On the other hand the impact strength of epoxy cured with MDA was higher than that of epoxy cured with MTHPA because of the cycloaliphatic amine structure of MDA which contributing to better toughness of the epoxy composite than the one wilh MTHPA which had rigid structure of benzene ring.

The impact strength of both BMC composites containing original fiber and recycled fiber tended to decrease with higher glass fiber loading, suggesting that the composites became more brittle upon increasing glass fiber content. This is due to the stiffness of the fiber comparing to the matrix.

Considering the impact strength of BMC composites containing original glass fiber, the results showed that the addition of 5 wt % glass fiber had no influence on their impact strength values. This result was found in both systems. Further loading the glass fiber resulted in the decrease in impact strength, owing to the agglomeration of glass fiber leading to the increase of localized stress and reduction of the elasticity of matrix material. As a result, the impact strength was decreased. Another possible reason also was discussed by Surai, et. al. [54] that the particle of reinforcement in the scale of micrometer led to the generation of crack around reinforcement and short energy distance for energy dissipation, whereas the high volume fraction of nanoparticle size can be used to improve toughness of brittle plastic because of the dense of interfacial region. Therefore, the impact strength of BMC composites containing original glass fiber (125 μm) may be affected from the length of glass fiber, resulting in the decrease of strength values.

Regarding to the effect of recycled glass fiber obtained from pyrolysis process on the impact strength of BMC composites, it can be seen from Figure 4.40 and 4.41 that the impact strength of both BMC composites containing recycled glass fiber at 600 and 800°C showed similar trend that the impact strength was decreased with the addition of recycled glass fiber and was lowered than that of neat resin and BMC composite with original glass fiber. This result was agreed with previous research reported by Marco, et al. [26].

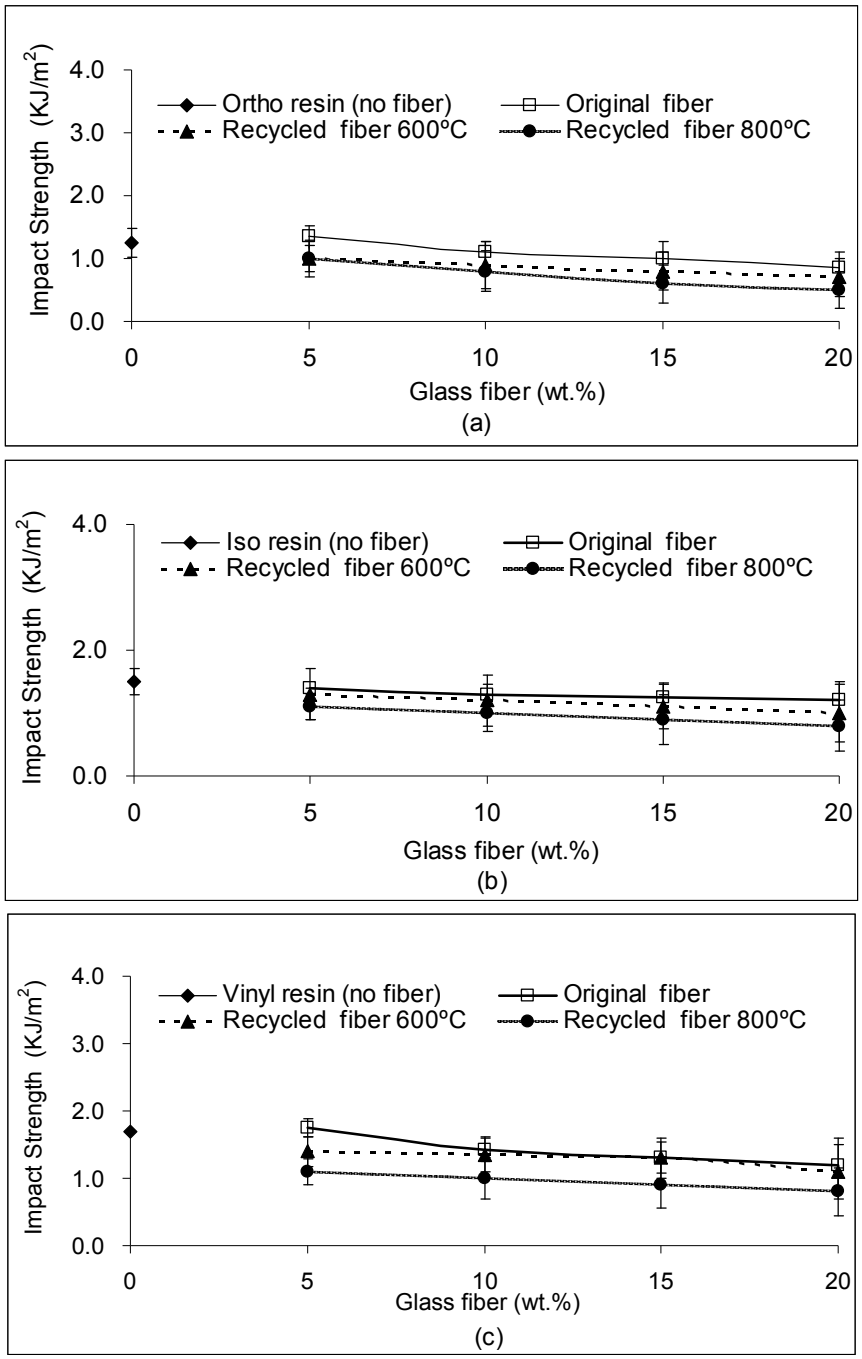


Figure 4.42 Impact strength of unsaturated palyester resin and BMC composites in relation to weight fraction of glass fiber (a) orthophthalic polyester, (b) isophthalic polyester, and (c) vinyl ester composite.

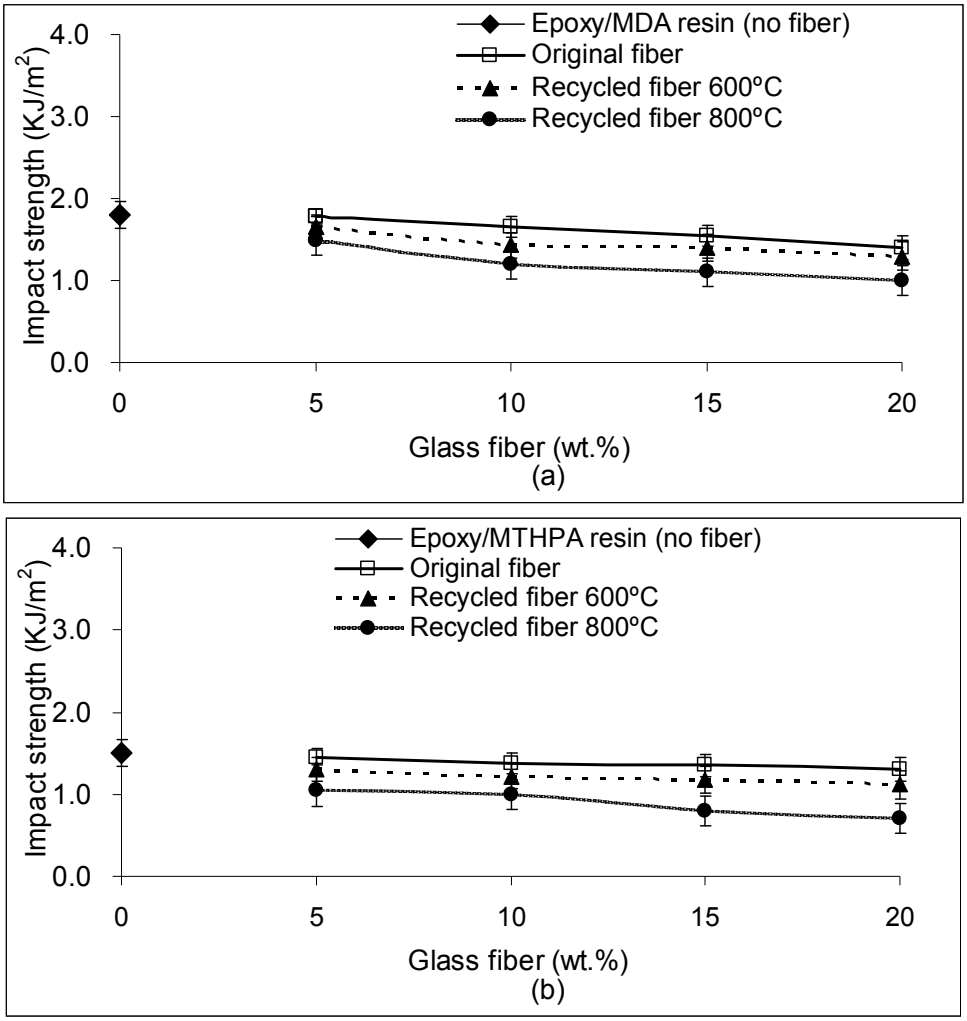


Figure 4.43 Impact strength of epoxy resin and BMC composites in relation to weight fraction of glass fiber (a) epoxy cured with amine (MDA), (b) epoxy cured with anhydride (MTHPA).

Comparing between two types of recycled glass fiber, both BMC composite systems containing recycled glass fiber at 600ºC exhibited better impact strength than their counterparts. This is because recycled glass fiber at 800 ºC showed poorer interfacial bonding between matrix and glass fiber as evidenced by the presence of many holes as previously presented by SEM micrographs in Figures 4.37 and 4.38.