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Improving the Physico-mechanical and Degradable Properties of Thermoplastic Polymer with Modified Starch Blend Composites for Food packaging Applications

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Abstract

Plastics are an essential item of modern life and are used in many other applications such as food packaging, building components, mulching, and much more. Plastics are a subspecies of a type of material that is not degraded in the natural environment. Plastic shopping bags are manufactured from low-density polythene (LDPE) which causes ecological problems because most plastic ingredients have been in waste storage and underground for a long time. Meanwhile, several options have been considered to increase the use of biopolymers to reduce ecological problems. In this research, sago starch (SS) was treated with sodium trimetaphosphate. Sago starch was mixed with LDPE in varying levels of starch (10%-30 wt%) and the same amount of treated SS was mixed with LDPE using additives (glycerol/urea, and epolene wax) were compounded via melt mixing technique tracked by injection molded to form sheets. The reduced tensile strength percentage of the treated composite indicates that the good distribution and uniformity of SS in the LDPE was lower but more elongated at break than in the untreated composite. Treated SS composites also show less water absorption and degradation than untreated SS plastic composites. The application of the degradable composite as an eco-friendly packaging component can be effectively judged by its percentage loss in tensile strength and elongation at break by 30.7%, 44.6%, and 20.5%, 29.9% for untreated and treated composites, respectively, after 6 months of exposure to soil burial.

Keywords: Sago starch, LDPE, blends, mechanical properties, morphology, biodegradability

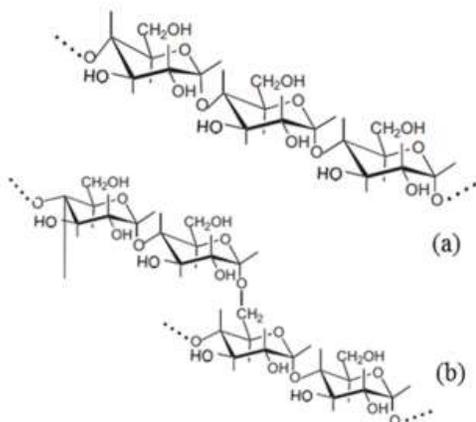
1. Introduction

The increase in plastic production across the nation has been introduced to the world as an environmental problem because most plastic materials have been in waste collection and underground for decades. Important environmental problems can contribute to this condition. Nevertheless, the use of non-easily degradable hydrocarbon plastic components has increased. Meanwhile, several alternative biopolymers have been considered to reduce environmental problems by increasing their use. Renewable natural polymers such as starch, cellulose, and chitosan alone have been incorporated into the same test with potential advances in plastic properties and biodegradation of products (1-3). Starch is a biopolymer that can be used to replace hydrocarbon plastic components.

Man-made plastic components have been used in the food, textile, housing, transport, construction, pharmaceutical, and entertainment industries. Man-made plastics such as LDPE materials are hard, light, and durable so are currently used in plastic films, distribution bottles, garbage bags,

agricultural mulch, fertilizer bags, and various molded laboratory materials for composites. Polythene is a hydrophobic polymer. It establishes a carbon-carbon connection that microorganisms cannot land easily. Plastic biodegradability is provided as a solution to the problem of misuse. Starch-based plastics have not harmed the environment and have reduced the greenhouse effect (4). Focus on ecological plastics for food packaging, medical, fishery, and agricultural applications have improved in recent years (5).

Starch is inexpensive, renewable, completely biodegradable natural ingredient (6), and abundant in agricultural resource-rich countries. It is a natural polymer that repeats 1-4- α -D glucopyranosyl units and is usually composed of a combination of linear component (amylose, secondary) and branched component (amylopectin, main) components. The structure of amylose and amylopectin is shown in Scheme 1. Like other starches, sago starch is obtained from a wide variety of palm kernels called *Metroxylon Sago*.



Scheme 1 The chemical composition of (a) amylose and (b) amylopectin.

Sago starch is extracted from the pith of various tropical palm stems (sago palm). This starch also points to potential biodegradable fillers in thermoplastics as a new use of sago. The blending of SS with LDPE has received considerable attention due to the potential application of this strategy in the removal of plastic waste. Starch-filled polythene composites have shown poor mechanical properties due to imperfections. When different materials are mixed with starch, the hydrophilicity of the starch results in their activities during and after the process (7). The inclusion of plasticizing agents will progress the compatibility of glycerin and starch cross-linked sodium trimetaphosphate (STMP) in LDPE/thermoplastic starch blending systems. Chemical crosslinking is a possible way to develop the physical and biological features of composite films. STMP has been nominated for this research because it has one of the most important food additives, a safe and non-toxic crosslinking agent suitable for polysaccharides to improve their functional properties, has been used in many research studies and reports (8). Although biodegradation of polythene has been extensively studied (9-11), the results have been based on starch-mixed polyethylene. The key invading agent of biodegradation is microorganisms (fungi and bacteria), which spread in soil and water. Goheen et al. (12) observed the degradation of PE/starch film in the soil using FTIR spectroscopy to evaluate the release of starch and chemical changes in the PE. Despite this work, none relies on evaluating the correlation between morphology, tensile property, and biodegradability of microbial methods. The effects of starch concentration and the addition of additives to modified SS/LDPE composites and their efficacy before and after their mechanical features and biodegradability have been investigated.

2. Experiment

2.1 Materials

LDPE pellets obtained from M/S Reliance Industries Ltd., Mumbai, India, have been used as a matrix. SS was procured from Johar, a local supplier in Malaysia. Percent moisture was 11-13% and its starch value was above 85%. Biodegradable agents are a mixture of glycerol, SS, and urea. Reagent grades glycerol (glycerin, $C_3H_8O_3$) as the plasticizing agents, STMP as a starch cross-linker in SS blends, and urea and epolene wax were purchased from Malaysia's Sigma Aldrich Chemical Company Inc.

2.2 Methods

2.2.1 Modification of sago starch

SS was modified by the Shin et al. method (13, 14) due to its poor processing and incompatibility with LDPE. SS (50 g) was modified with cross-linked STMP (5.40 g) at 45°C for 2 h. Grounded cross-linked SS was then used to dry and prepare the composite in an oven at 50°C for 2 h.

2.2.2 Sample preparation

All components were dried in a vacuum oven at 80°C for 20 h to reduce humidity before use. Granular SS was mixed with LDPE in varying levels of starch (10%-30 wt%) and the same amount of treated SS was mixed with LDPE using additives (glycerol/urea, and epolene wax) in a Mini Blender (Most Machine Builder Fairfield, New Jersey, USA) contains 3% dissolving agent (Epolene wax E-43p). Chemical compositions are presented in Table 1. The above mixer was made by melt blended with a co-rotating twin-screw extruder (model: TSE 20, GmbH & Co. KG, Germany) and then a composite sheet was formed with injection molding. The compounding process was conducted at a rotor speed of 90 rpm and the temperature die ($150^\circ\text{C}/150^\circ\text{C}$) was carried out from the feeder ($160^\circ\text{C}/160^\circ\text{C}$). The extruded components were then palletized by a pelletizer. Dumbbell-shaped specimens were made from these platelets using the injection-molded machine (Toyo, model: Si180iii-E200, Japan). The preparation of the biodegradable film is presented schematically in Figure 1.

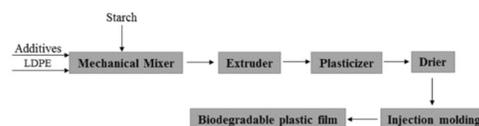


Figure 1 Schematic diagram for biodegradable film preparation.

Table 1 Compositions of the prepared samples.

Sample code	LDPE (wt%)	Starch (wt%)	*Glycerol:Urea = 2:1 (wt%)	*Epolene wax (wt%)
Virgin LDPE	100	0	–	–
L ₉₀ S ₁₀ (LUS ₁₀)	90	10	–	–
L ₈₀ S ₂₀ (LUS ₂₀)	80	20	–	–
L ₇₀ S ₃₀ (LUS ₃₀)	70	30	–	–
L ₉₀ S ₁₀ GU ₁₀ (LMS ₁₀ A)	90	10	10	3
L ₈₀ S ₂₀ GU ₁₀ (LMS ₂₀ A)	80	20	10	3
L ₇₀ S ₃₀ GU ₁₀ (LMS ₃₀ A)	70	30	10	3

L: LDPE; U: untreated SS; M: SS treated with STMP; The number after the letter S indicates the percentage of starch; A: additives; *glycerol: urea, epolene wax contents on LDPE weight basis.

2.2.3 Mechanical property test

Dumbbell-shaped specimens (125 × 3 mm²) have been used to measure the tensile features of composites. Tensile features of the composite were evaluated using Shimadzu UTM (Model AG-1, Japan) by the ASTM-D 638-03 standard (14). The tensile test was achieved at a crosshead speed of 10 mm/min and a gauge length of 50 mm. All experiments were measured at 25 ± 2°C and relative humidity of 55 ± 4%. All outcomes were examined as the mean value of five specimens.

2.2.4 Water absorption (WA) calculation

WA was tested for different samples based on ASTM D570-98. The samples were dried in a vacuum oven at 80°C for 6 h to reduce humidity before use. The dumbbell-sized specimens were immersed in water for 30 days. The samples were then detached from the water at regular intervals, the surface water was removed with a dry cloth and they were weighed using a Mettler balance with an accuracy of 1 mg for excess weight of water. The water absorption capacity was determined with the following equation (2.1).

$$\text{Water absorption (\%)} = \frac{W_{\text{wet}} - W_{\text{dry}}}{W_{\text{dry}}} \times 100 \quad (2.1)$$

where W_{wet} refers to wet sample and W_{dry} refers to dry sample.

2.2.5 Morphological observation

The tensile fractured surfaces of untreated and treated SS plastic composites were analyzed by a Zeiss, Evo 50 SEM. The fracture edges of the samples were embedded in an aluminum spit and covered with a thin layer of gold to disperse the electric charge throughout the test.

2.2.6. Soil burial practice

The dumbbell-shaped specimens were dried, weighed, and then buried in perforated plastic boxes so that microorganisms and moisture could attack the specimens. The box was buried in a soil compound that mixed rockery soil, municipal waste, compost, and *P. Aeruginosa* at an additional deepness of 15–22 cm below the soil surface for six months. At various intervals, the samples were cautiously dispelled from the soil and gently washed with distilled water to mix with the soil surface, and then the samples were dried until a constant weight was obtained. The equation for weight loss percentage was as follows equation (2.2):

$$\text{Weight loss (\%)} = \frac{W_b - W_a}{W_b} \times 100 \quad (2.2)$$

Where W_b refers to mold weight before degradation; W_a refers to mold weight after degradation.

3. Outcomes and Discussions

3.1 Mechanical and morphological features

The influences of untreated and treated starch content on the percentage loss of TS and Eb for the SS plastic composite is presented in Figure 2(a) and (b). We noticed that the percentage loss for untreated composites (LUS₁₀, LUS₂₀, and LUS₃₀) of TS increased steadily compared to virgin LDPE (Figure 2a). The TS of virgin LDPE was about 9.857 MPa. Increasing the starch content has increased the loss of TS. Loss of TS may be due to the weakening of the interfacial bond between starch and LDPE. As the content of starch increases, spherical starch has a less effective cross-sectional area of LDPE. The hydroxyl group on the surface due to starch exhibits hydrophilic features and a strong intermolecular hydrogen bonding. This observation agrees with the results presented by the researchers (15). The percentage decrease in TS of treated SS plastic composites (LMS₁₀A, LMS₂₀A, and LMS₃₀A) compared to virgin LDPE has been steadily increasing but it is less than the untreated SS plastic composites (LUS₁₀, LUS₂₀, and LUS₃₀). This is probably due to better interfacial bonding that occurred after the addition of LDPE matrix and additives (glycerol: urea = 10%, ferric stearate 0.1%, and epolene wax 3%). The –OH group of cross-linked starch reacted strongly with the phosphate group in STMP, and the functional –OH group reacted less with STMP as the cross-linked starch decreased. As a result, strong hydrogen bonds between LDPE/starch and plasticizer molecules can support the desired starch/LDPE interaction between LDPE or starch molecules instead of intermolecular and intramolecular, thus reducing the loss of compatibility between LDPE and starch, which resulted in a lower loss of TS.

Figure 2(b) shows the filler load effect of Eb percent loss of untreated and treated SS plastic composites. The Eb of virgin LDPE was found to be

130.23%. We noticed that the percentage loss in Eb for composites increases with increasing filler loading. This starch can impose hydrophilic nature and may interfere with the absorption effect by absorbing moisture and reducing the effect of physical bonding between the LDPE/SS interface (16). As mentioned earlier, towards high filler contents, agglomeration can occur at higher points of stress concentration, which initiates crack propagation in mixtures. This induces a percentage loss of Eb in the mixture with increasing starch content. Wang et al. (17) observed a similar trend with natural filler-filled LDPE mixtures.

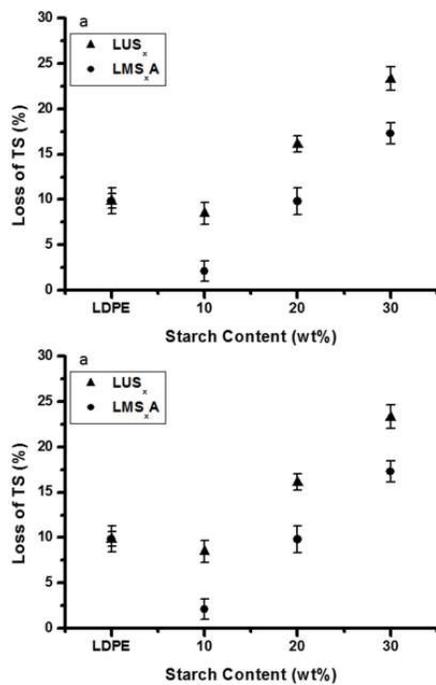


Figure 2 Effect of SS content on the percentage loss in (a) tensile strength (TS) and (b) elongation at break (Eb) of LUS_x and LMS_xA composites. LUS: LDPE/untreated SS; LMSA: LDPE/treated SS with additives. The subscript x after the letter S indicates the percentage of starch.

The morphology of SS, virgin LDPE, untreated SS plastic composite (LUS₃₀), and treated SS plastic composite (LMS₃₀A) is displayed in Figure 3. Figure 3(b) shows that the addition of SS to LDPE outcomes in a weaker spread between SS and LDPE. This micrograph proved to be the cause of the deterioration of the mechanical features of the mixture with the starch content. The interfacial bond between LDPE and starch can form in very weak stress concentration regions. Figure 3(c) displays the SEM micrograph of LMS₃₀A composite with a mixture of glycerol/urea plasticizers, and dispersing agent (Epolene wax). The morphology revealed that the

sample in LMS₃₀A of Figure 3(c) showed that phase separation between SS and matrix could not be detected. It can be noted that the interfacial morphology has improved a lot due to better interfacial adherence compared to Figure 3(b). The mixture of plasticizers, STMPs, and dispersing agents can build further hydrogen bonds with SS because glycerol/urea weakens the intermolecular and intramolecular hydrogen bonds between LDPE and starch. This clarifies the better mechanical features inspected for the LMS₃₀A composite than the LUS₃₀ composite.

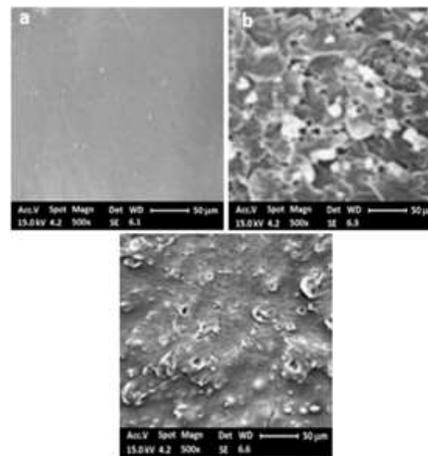


Figure 3 SEM photomicrographs of (a) virgin LDPE, (b) LUS₃₀ and (c) LMS₃₀A composites.

3.2 Influence of water absorption (WA) on mechanical and morphological properties

Figure 4 displays the influence of SS content on the WA of virgin LDPE, LUS₂₀, LUS₃₀, LMS₂₀A, and LMS₃₀A composites at five-day intervals from water immersion for thirty days. Figure 4 shows that the WA is gradually increased by increasing the amount of SS for both untreated and treated SS plastic composite. The WA of the LUS₃₀ composite is 2.21% higher than that of LDPE (0.152%) after immersion in water for thirty days. This is due to the hydrophilic nature of SS, which is found to interact with molecules, due to the presence of large -OH groups. These -OH groups have increased with starch loading with an increase related to WA. Again, as the starch load increases, the aggregate structure increases thus making it difficult to achieve a homogeneous distribution of the starch through the composite. This allows water molecules to enter the composite through voids formed in the composite, which increases the WA of the composite (18). The amount of WA has been significantly reduced by adding treated SS with additives in the mixtures. The amount of WA in the LMS₃₀A composite was found to be about 1.46%, while the WA in the LUS₃₀ composite was about 2.22% after being submerged in water for about thirty days. The

composite of treated SS plastics contributes to lower WA because the phosphate group replaces the -OH group in the starch molecule and thus the hydrophobicity of the mixture is increased.

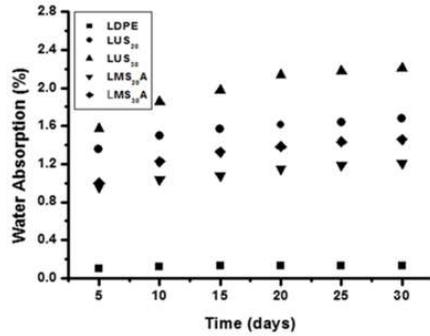


Figure 4 Water absorption (%) for virgin LDPE, LUS₂₀, LUS₃₀, LMS_{20A}, and LMS_{30A} composites with a time of immersion.

Figure 5 shows the percentage loss in tensile properties (TS and Eb) of LUS₂₀, LUS₃₀, LMS_{20A}, and LMS_{30A} composites after being submerged in water for thirty days. Figure 5 shows an increase in the percentage loss of tensile features with an increase in immersion time. In addition, during immersion, the percentage of greater tensile properties decreases. This is owing to the existence of wetness at the LDPE–starch interface, which already weakens the poor interfacial bond. After 30 days of immersion in water, LMS_{20A} and LMS_{30A} composites lost almost 20.3% and 23.7% of TS and 85.37% and 105.2% of Eb, respectively, but the LUS₂₀ and LUS₃₀ composites lost almost 28.3% and 32.4% of TS and 79.87% and 94.43% of Eb, respectively. LMS_{30A} composites retained much more of their tensile strength than does the LUS₃₀ composites during water immersion. The high biodegradation of the LUS₃₀ composite sample may increase its higher WA and lower tensile strength due to the same factors.

Figure 6 illustrates the surface morphology of the LUS₃₀ and LMS_{30A} composites before and after immersion in water for thirty days, respectively. Figure 6(a) displays that the LUS₃₀ composite had less consistency between LDPE and starch. Figure 6(b) shows that after immersion in water for thirty days, the LUS₃₀ composite surface was transformed into a rough surface by mixing large cracks, expanded cavities, and starch separation surfaces. The cavities may be owing to the elimination of starch particles due to wetness absorption, which causes the grain to swell, increase in dimension and come out of the cavity, or mechanical action due to trembling. Figures 6 (c) and (d) display that ruin samples of the composite at LMS_{30A} before and after immersion in water for 30 days. As observed, changes in the morphology of the surface of the composite at LMS_{30A} in water immersion. Figure 6(d) shows that LMS_{30A} composites had a smooth surface small of cavities.

The cavities indicate the rate of biodegradation and ensure the elimination of starch through degradation testing. More and smaller cavities were performed at the rate of biodegradation in LDPE. The smooth surface structure in the LMS_{30A} composite indicated that the LMS_{30A} composite had better consistency with LDPE than the LUS₃₀ composite.

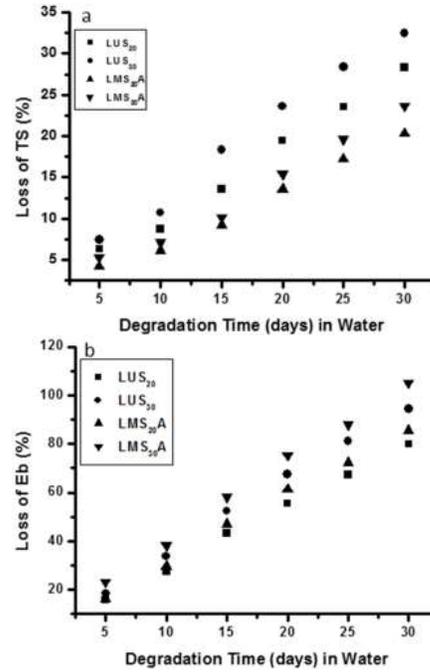


Figure 5 Percentage loss in (a) tensile strength (TS) and (b) elongation at break (Eb) of LUS₂₀, LUS₃₀, LMS_{20A}, and LMS_{30A} composites after immersion in water for 30 days.

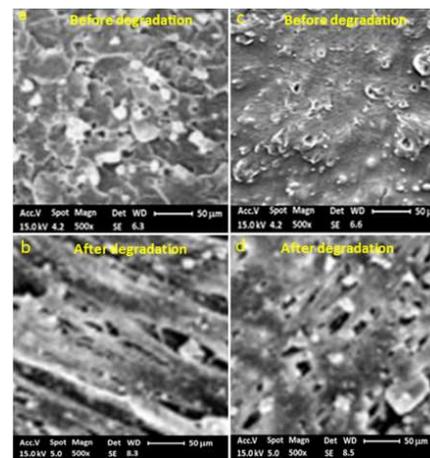


Figure 6 SEM images (magnification 100×) of LDPE/SS blends: (a) LUS₃₀ and (c) LMS_{30A} composites before immersion in water; (b) LUS₃₀ and (d) LMS_{30A} composites after water immersion for 30 days.

3.3 Soil burial test

The biodegradation studies on composite behavior are of vital importance on the environmental application of biocomposites. Soil burial is the most appropriate test for measuring the biodegradability of SS blended with LDPE. All samples were placed in the ground and weight loss was recorded 1, 3, and 6 months later. Figure 7 shows the weight loss (%) of virgin LDPE, LUS₃₀, and LMS₃₀A composites during soil burial time. weight loss was observed at each time point of the study was greater with increasing degradation time. This means that the weight loss of both the LUS₃₀ and LMS₃₀A composites increases continuously with increasing degradation time, which indicates that the samples deteriorate with increasing time. After 1 month of exposure, LDPE showed no significant weight loss (approximately 0.07%) since its mixtures with starch showed even greater weight loss. It was about 4.72% weight loss in the LUS₃₀ composite after six months of exposure to a land situation. LUS₃₀ composites showed maximum weight loss while LDPE showed minimum weight loss over time. This search was accredited to the SS content in the composite because SS is more disinfectant than virgin LDPE. Soil background comprises a dissimilar kind of microbes and macroorganisms. Weight loss of soil polymer fragments can be taken as an indicator of bio-growth in laneways. Soil microbes invaded the polymer fragments. First, the microbes were invaded in the SS of the mixtures. 30 wt% starch composites were ruined by the presence of cavities on the surface of the sample. The small cavities displayed on the surface of the samples ensure the elimination of starch, which can be invaded by microbes. Thus, O₂ can invade the freshly produced surface with the formation of peroxides and hydroperoxides. These radicals raise the LDPE key chain into tiny strips that are more susceptible to invade by microbes such as fungi and bacteria. These outcomes have ensured weight loss measures. However, a soil environment of 3.85% weight loss was achieved for the LMS₃₀A composite after six months of exposure. As clearly depicted in Figure 7, the decay rate of the LMS₃₀A composite was slightly lower than that of LUS₃₀. This may be due to the interfacial bond between the phosphate group of STMP and the -OH groups of crosslinked starch, which represent or inhibit the use of SS from microbes in the soil.

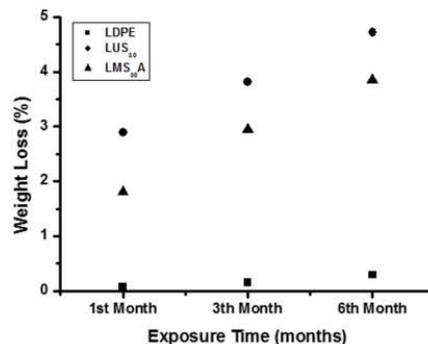


Figure 7 Weight loss of virgin LDPE, LUS₃₀, and LMS₃₀A blend composites after six months of exposure to soil burial.

In most applications envisaged for films or fibers in contact with soil, the most relevant practical criterion for determining its degradation is the reduction of the tensile properties (19). The percentage loss in TS and Eb of the composite of LUS₃₀ and LMS₃₀A along with virgin LDPE after biodegradation tests in natural soils for 6 months is illustrated in Table 2. As the exposure time increased, the percentage loss in TS and Eb of the virgin LDPE, LUS₃₀, and LMS₃₀A composites increased. After 6 months, the percentage loss in TS and Eb of virgin LDPE film increased by 5.78% and 11.7%, respectively, where to the inclusion of 30 wt% starch in LDPE resulted in a loss of 30.7% in TS and 44.6% in Eb. The cause for this loss is the depletion of starch by microbes. The bond between the two phases is broken, resulting in starch degradation and the formation of large cavities in LDPE. This loss may be related to the shortage of consistency between LDPE and starch. These variations reflect the tensile features of the composites. However, the LMS₃₀A composite showed less loss in tensile properties than the LUS₃₀. After 6 months, the percentage loss in Eb increased about 44.6% whereas that of the LMS₃₀A composite increased 29.9% compared to the untreated ones. The LUS₃₀ composite has more surface area for invasion by microbes, in which more microscopic cavities are arbitrarily distributed in the composite than in the LMS₃₀A composite. Subsequently, the percentage loss in tensile features of the LUS₃₀ composite was greater than those of the LMS₃₀A composite. These outcomes have a good contract with the SEM micrograph shown in Figure 8.

Table 2 Percentage loss in tensile properties (TS and Eb) of virgin LDPE, LUS₃₀, and LMS₃₀A composites during the soil burial.

Loss in properties (%)	Time (months)	Samples		
		LDPE	LUS ₃₀	LMS ₃₀ A
Loss of TS (%)	1	1.83±0.5	13.9±0.6	6.9±0.7
	3	3.98±0.8	21.6±0.7	14.8±0.6
	6	5.78±0.6	30.7±0.9	20.5±0.8
Loss of Eb (%)	1	4.9±0.6	19.7±0.6	14.2±0.6
	3	6.8±0.7	32.5±0.7	23.7±0.8
	6	11.7±0.8	44.6±0.8	29.9±0.7

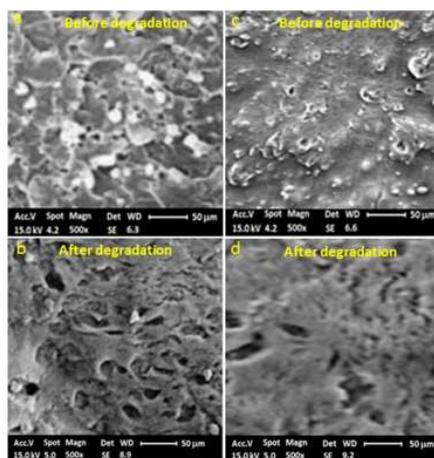


Figure 8 SEM images (magnification 100×) of LDPE/SS blends: (a) LUS₃₀ and (c) LMS₃₀A composites before exposed to soil burial; (b) LUS₃₀ and (d) LMS₃₀A composites after exposure to soil burial.

Figure 8 shows the SEM micrographs of the LUS₃₀ and LMS₃₀A composites, respectively, before and after burying the soil. SEM micrographs confirmed the destruction of starch particles in the blend. Starch particles are reduced in all mixtures regardless of the corrosive environs. The degree of degradation increases with the period of degradation. This event observed the erosion of all specimens. They are either isolated or performed on the surface of their mixture. The separation of the starch particles brings about a change in the structure of the mixture surface. The sample buried under moist soil for a six-month test proves that the starch particles are completely broken down when viewed by SEM. Figures 8 (b) and (d) show that after the soil burial test the LUS₃₀ and LMS₃₀A composite surface became larger cavities than the control composite before the erosion process shown in 8 (a) and (c). Besides that, during 30 wt% SS loading, uninterrupted cavities are observed due to the starch leaking. As a result of tiny cracks it breaks down and starch granules appear embedded on the surface of the matrix. Some changes are present in the mixtures in contact with the total

collapse, the embrittlement of the matrix, and the embedded SS granules. The outcomes agree with the mechanical assessment.

4. Conclusions

SS was treated by cross-linking with STMP, had better consistency and interaction with LDPE than untreated SS. Therefore, the reduction in the tensile strength percentage of the treated SS plastic composite is less but more in the elongation at break. This is due to the good interfacial bond between LDPE and thermoplastic SS, although both mixtures have shown a decrease in tensile properties with increasing soil burial period. The rate of biodegradation of composites has increased over time. Also, the phosphate group treatment increases the hydrophobicity of SS plastic composites resulting in increased water tolerance and consequently reduces the plastic composite corrosion by replacing the -OH group in the starch molecule. The data show that this new polymer matching is related to an industry where rapid decay can be applied to the desired image in agricultural plastics, flower cases and as bags, horticulture, packaging, and so on.

Declaration of conflicting interests

The authors declared that they have no conflicts of interest in the research, authorship, and this article's publication.

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