

Kapok Husk-Reinforced Soy Protein Isolate Biofilms: Tensile Properties and Enzymatic Hydrolysis

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The utilization of kapok husk (KH) as a reinforcing filler can enhance the properties of soy protein isolate (SPI)/kapok husk (KH) films. The properties of soy protein isolate/kapok husk films with and without the cross-linking agent glutaraldehyde (GLA) were investigated. Films with different KH contents were prepared through a solvent casting method. The addition of KH to SPI films increased the tensile strength, modulus of elasticity, and thermal stability, but reduced the elongation at break. The presence of glutaraldehyde improved the tensile and thermal properties of SPI/KH films. The tensile strength of modified SPI/KH films at 40 wt% increased by 30% compared to unmodified films. The improvement of interfacial interaction between the KH and SPI was demonstrated using a morphology study. Fourier transmission infrared spectroscopy (FTIR) analysis indicated the presence of ethylenic (C=C) groups and imine (C=N) groups. An enzymatic degradation test of SPI/KH films was performed for 14 days in a *diatase* buffer solution at 37 °C. The enzymatic degradation weight loss of unmodified films decreased with increasing KH content. In contrast, the modified SPI/KH films with glutaraldehyde retained about 50% of their original weight.

Keywords: Soy protein isolate; Kapok husk; Glutaraldehyde; Cross-linking; Films

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INTRODUCTION

In the last few decades, the development of biodegradable material based on natural polymer has notably increased to substitute petroleum-based plastics and to address concerns about environment pollution (Lodha and Netravali 2005a; Schmid *et al.* 2005). Biodegradable plastics are being developed because they are economically and environmentally friendly (Lodha and Netravali 2005b). Among the studies of all biopolymers, soy protein isolate (SPI) has been studied as a potential material to replace petroleum-based polymers because of its low cost, biodegradability, and availability (Schmid *et al.* 2005). There have been numerous studies on SPI films (Park *et al.* 2000; Lodha and Netravali 2002, 2005a,b; Chabba and Netravali 2005; Schmid *et al.* 2005; Su *et al.* 2012; Wang *et al.* 2013)

Numerous researchers have investigated the incorporation of natural filler in polymer materials such as corn cob (Chun *et al.* 2013a; Yeng *et al.* 2013a,b,c), coconut shell (Chun *et al.* 2012; Salmah *et al.* 2013), cocoa pod husk (Chun *et al.* 2013b, c), chitosan (Amri *et al.* 2013), and *Nypa fruticans* (Rasidi *et al.* 2014) to produce composites. These fillers offer several advantages, including cost effectiveness, recyclability,

biodegradability, low density, and renewability. Kapok, also known as *Ceiba pentandra*, is an agricultural waste that is derived from the fruits of silk-cotton trees with the composition of cellulose, lignin, and polysaccharides (Berry 1979). Kapok is cultivated in the northern part of Malaysia. In rural areas, the kapok seeds are consumed after removing the husk. Also, the floss has been used for pillows and cushions. Therefore, kapok husk (KH) is an abundant waste of the kapok industry. Kapok husk used as a natural filler in polymers could have an economic advantage and a low environmental impact.

Soy protein isolate (SPI) is a protein that is obtained from soybean seeds. It contains at least 90% protein, which is higher than other soy protein products such as soy protein concentrate and defatted soy flour. The proteins have a wide range of molecular weight of 8 kDa to about 600 kDa. The various components are identified as 2 s, 7 s, 11 s, and 15 s (Svedberg units) and they are usually found at 20-22, 37, 31-40, and 10-11% (of the total weight) in SPI, respectively (Hermansson 1978). Thus, SPI consist of two major components which is 7s and 11s globulins. The 7 s and 11 s is necessary to indicate the gel properties of SPI (Nagano *et al.* 1992).

There have been various attempts to modify SPI, as well as to enhance its mechanical properties, such as cross-linking, blending with polymer, and the incorporation of modifiers (Park *et al.* 2000; Lodha and Netravali 2005a,b; Schmid *et al.* 2005). In the present research, glutaraldehyde (GLA) was used as a cross-linking agent to improve the mechanical and thermal properties of SPI films. GLA is a well-known cross-linking agent that has economical and commercial availability (Migneault *et al.* 2004). GLA can react with proteins to produce cross-linked polymers, which in turn enhances the properties of SPI films. Cross-linking reactions usually occur between the aldehyde groups of GLA and the amino groups of the protein (lysine and histidine) to produce covalent intermolecular or intramolecular cross-links through a Schiff base reaction (Park *et al.* 2000). In previous studies, the addition of GLA into chitosan/corn cob biocomposite films led to improved mechanical and thermal properties (Lodha and Netravali 2002). Some researchers have proposed the incorporation of GLA as the cross-linking agent with SPI to improve its mechanical properties (Park *et al.* 2000; Chabba and Netravali 2005; Jin *et al.* 2013).



Fig. 1. SPI/KH nursery poly-bags

In this study, kapok husk was used as a reinforcing filler in SPI/KH films. The effect of filler content and a glutaraldehyde cross-linking agent on mechanical properties, thermal properties, morphology, and enzyme hydrolysis of films were investigated. Currently, the utilization of non-degradable poly-bags was one reason that caused

environment pollution in agricultural farms. The SPI/KH films were proposed as nursery poly-bags, as shown in Fig. 1, which have been initially developed at our university.

EXPERIMENTAL

Materials

Soy protein isolate with purity of 90% protein (used as the matrix) was supplied by Shandong Wonderful Industrial Group Co., Ltd., Dongying, Shandong (China). The average particle size and molecular weight of the soy protein isolate powder were 64.7 μm and 50,000 Da, respectively. Kapok husk was obtained from a kapok plantation in Perlis, Malaysia. The KH was cleaned and ground manually into a fine form. Then, the KH was dried at 80 °C for 24 h. The average particle size of KH was 16.3 μm . Table 1 shows the chemical composition of kapok husk. Glutaraldehyde was purchased from Sigma-Aldrich (Penang, Malaysia). Glycerol, which was used as a plasticizer in SPI/KH green films, was supplied by HmbG[®] Reagent Chemical (Selangor, Malaysia).

Table 1. Chemical Composition of Kapok Husk (Draman *et al.* 2014)

| Component | Weight percent (%) |
|---------------|--------------------|
| Cellulose | 53.4 |
| Hemicellulose | 29.6 |
| Lignin | 16.5 |
| Ash | 0.5 |

Preparation of Unmodified and Modified SPI/KH Films

The unmodified and modified SPI/KH films were prepared *via* a solvent casting method. First, the SPI powder (8 g) was dispersed into a glycerol with ratio 2:1 (by weight of SPI powder) solution (glycerol mixed with 100 mL distilled water) and stirred for 30 min at a temperature of 80 °C. Glycerol was added to the film-forming solutions to provide film plasticization. Then, KH was added and the solution was stirred for 15 min. For modified SPI/KH films, GLA was first dissolved in distilled water to produce a 1% v/v GLA solution. Then, the 1% GLA solution was added to the SPI/KH solution, which was stirred for 30 min. Finally, the unmodified and modified SPI/KH solutions were poured into plastic molds with size of 12 x 14 cm and dried in an oven at a temperature of 50 °C for 12 h. Table 2 shows the formulations of unmodified and modified SPI/KH films.

Table 2. Formulations of Unmodified and Modified SPI/KH Films

| Materials | Unmodified SPI/KH | Modified SPI/KH |
|------------------------------|---------------------|---------------------|
| SPI (wt%) | 100, 90, 80, 70, 60 | 100, 90, 80, 70, 60 |
| KH (wt%) | 0, 10, 20, 30, 40 | 0, 10, 20, 30, 40 |
| Glutaraldehyde (GLA) (v/v %) | - | 1 |

Tensile Testing

The tensile properties were analyzed using an Instron universal testing machine (USA), Model 5569, according to the ASTM D882 (2009) standard. The specimens were cut into rectangular shapes with sizes of 100 x 15 mm. The films were conditioned at ambient temperature and kept in a desiccator for at least 48 h prior to undergoing the tensile

test. A cross-head speed of 5 mm/s was set in the tension mode and the test was performed at 25 ± 3 °C and relative humidity (RH) of 50 ± 2 %.

Fourier Transform Infrared Spectroscopy (FTIR)

The FTIR analysis was performed using a Model L1280044 FTIR spectrometer (Perkin-Elmer; Germany). The attenuated total reflectance (ATF) method was used. Four scans in the wavelength range of 4000 to 600 cm^{-1} with a resolution of 4 cm^{-1} were used for each sample.

Thermogravimetric Analysis (TGA)

The TGA analysis was carried out using a Perkin-Elmer TGA Pyris Diamond (Germany) to determine the thermal stability of the SPI/KH films. Measurements were taken from 30 to 800 °C under a nitrogen atmosphere. The heating rate and purge gas flow rate used were 10 °C/min and 50 mL/min, respectively.

Morphology Study

The tensile fracture surface of the SPI/KH films was measured using a JEOL JSM-6460 LA scanning electron microscope (SEM) (Japan) operating at an accelerating voltage of 5 kV. The fracture surface of samples was coated with a thin layer of palladium before testing.

Enzymatic Degradation

Enzymatic hydrolysis testing was performed according to a method reported by Yoon *et al.* (1996). SPI/KH specimens were placed into a solution containing α -amylase. A buffer solution was prepared by adding 4.8 mL of 0.2 M acetic acid to 45.2 mL of 0.2 M sodium acetate solution to produce a 50-mL solution. Specimens were taken out every two days and washed thoroughly before conditioning in an oven at 50 °C for 24 h. The weight loss of enzymatic degradation of the specimens can be calculated using the equation below (Taghizadeh and Sabouri 2013),

$$D_t = \frac{w_o - w_1}{w_o} \times 100\% \quad (1)$$

where D_t is the weight loss due to enzyme degradation, w_o is the initial weight of the specimen, and w_1 is the weight of the specimen after degradation.

RESULTS AND DISCUSSION

Tensile Testing

Figure 2 shows the tensile strength of unmodified and modified SPI/KH films. The results showed that the tensile strength of SPI/KH films increased with increasing amounts of KH. The increasing trend in tensile strength for both unmodified and modified SPI/KH films indicates that the KH filler acted as a reinforcing filler in the films. The increased tensile strength of films could also be explained by the ability of the KH filler to support the stress transfer from the SPI matrix to the KH filler phase. This is attributed to the formation of intermolecular hydrogen bonds between the hydroxyl or carbonyl groups of the SPI matrix and hydroxyl groups of the KH filler. A similar result was found by Huang

and Netravali (2009). However, modified SPI/KH films with GLA exhibited higher tensile strength than unmodified. At 20 and 40 wt% KH content, the modified SPI/KH films with GLA show a significant improvement in tensile strength around 30.18 and 29.17%, respectively, as compared to unmodified films. The increment indicated that the aldehyde groups of GLA react with the amino groups of SPI to form an imine intermolecular cross-link through a Schiff base cross-linking reaction. The formation of imine linkages between GLA and SPI improved the interfacial interaction and wettability between the SPI matrix and the KH filler. Moreover, the tensile strength of modified neat SPI films (plasticized with glycerol) with GLA higher than unmodified neat SPI films due to the presence of imine linkages.

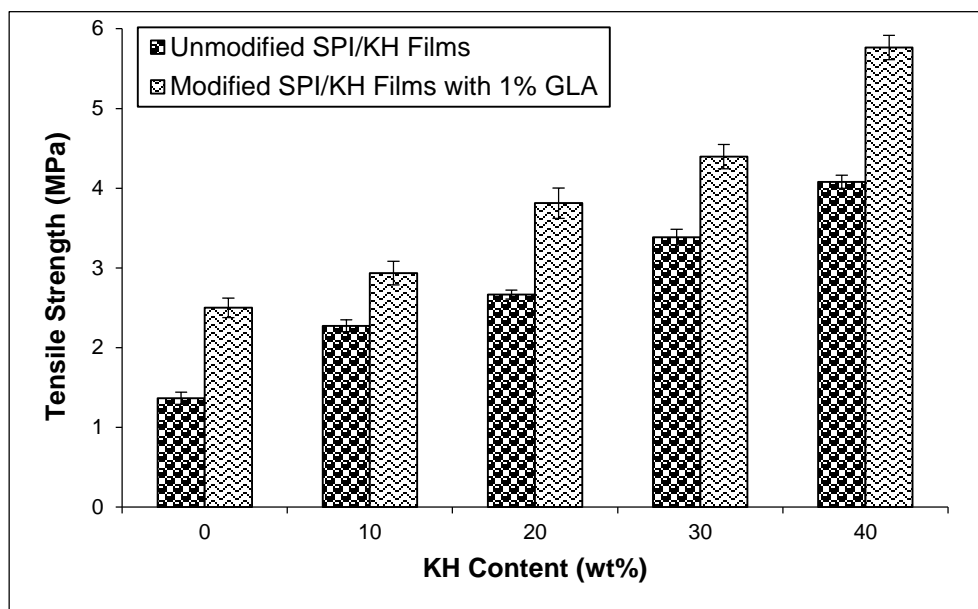


Fig. 2. Mean (\pm SD) tensile strength of unmodified and modified SPI/KH films

The proposed reaction between SPI and GLA is shown in Fig. 3. Similar observations were reported by several researchers (Park *et al.* 2000; Yeng *et al.* 2013a).

The modulus of elasticity of unmodified and modified of SPI/KH films increased with an increasing amount of KH, as illustrated in Fig. 4. As expected, the KH natural filler would increase the modulus of elasticity of SPI/KH films cause the addition of rigid filler in soft matrix.

Abdul Khalil *et al.* (2001) claimed that the inclusion of starch or cellulosic natural filler can improve the stiffness of composite materials. Thus, the composite stiffness increases with higher filler content. Nevertheless, the modified SPI/KH films showed a higher modulus of elasticity as compared to the unmodified films. It was anticipated that the presence of GLA increased the modulus of elasticity due to the formation of imine linkages between GLA and SPI. Furthermore, from Fig. 4, it can be clearly seen that the modified neat SPI film exhibited higher modulus of elasticity than unmodified neat SPI film. This could be due to the formation of imine crosslinkages in SPI films. Yeng *et al.* (2013a) reported a similar finding, which showed that GLA enhanced the modulus of elasticity of biocomposite films.

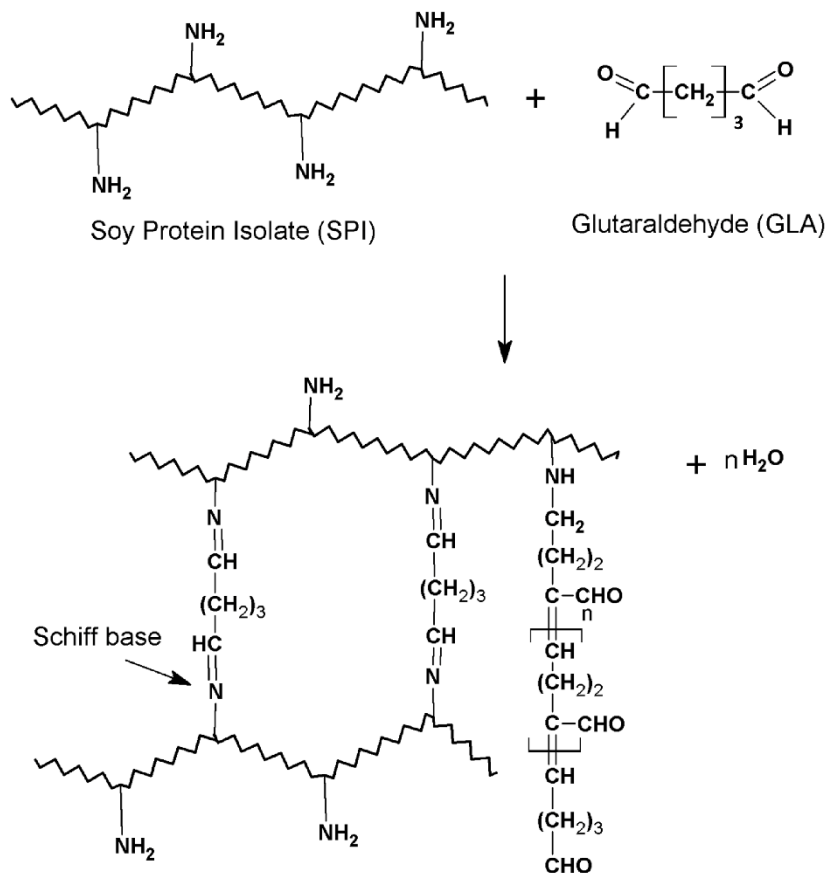


Fig. 3. Proposed cross-linking reaction between GLA and SPI

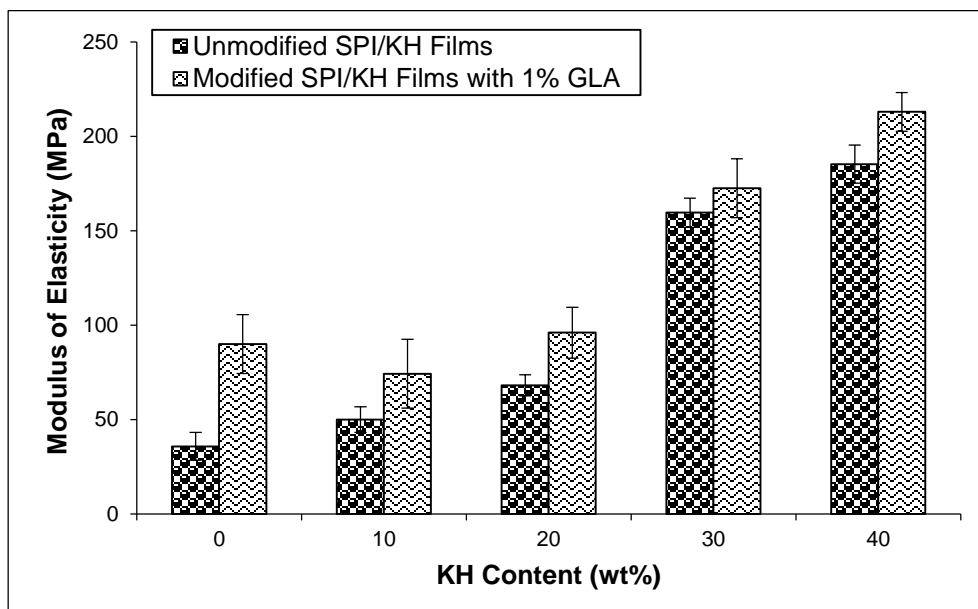


Fig. 4. Mean (\pm SD) modulus of elasticity of unmodified and modified SPI/KH films

Figure 5 displays the elongation at break of unmodified and modified SPI/KH films. The elongation at break decreased with the increasing amount of KH. This may be attributed to the incorporation of KH filler causing a decrease in SPI chain mobility and an

increase in rigidity of the SPI/KH films. In contrast, chemically modified SPI/KH with GLA showed higher elongation at break than unmodified. The increment of elongation at break value might be due to GLA improving the plasticizing effect of SPI/KH films. Similar observations were reported by Yeng *et al.* (2013c), in which films prepared from chitosan and corn cob modified with acrylic acid exhibited higher elongation at break values than unmodified chitosan/corn cob biocomposite films. Furthermore, the unmodified neat SPI film shows lower elongation at break than modified neat SPI film with GLA. These results are due to the presence of imine crosslinkages in modified neat SPI film.

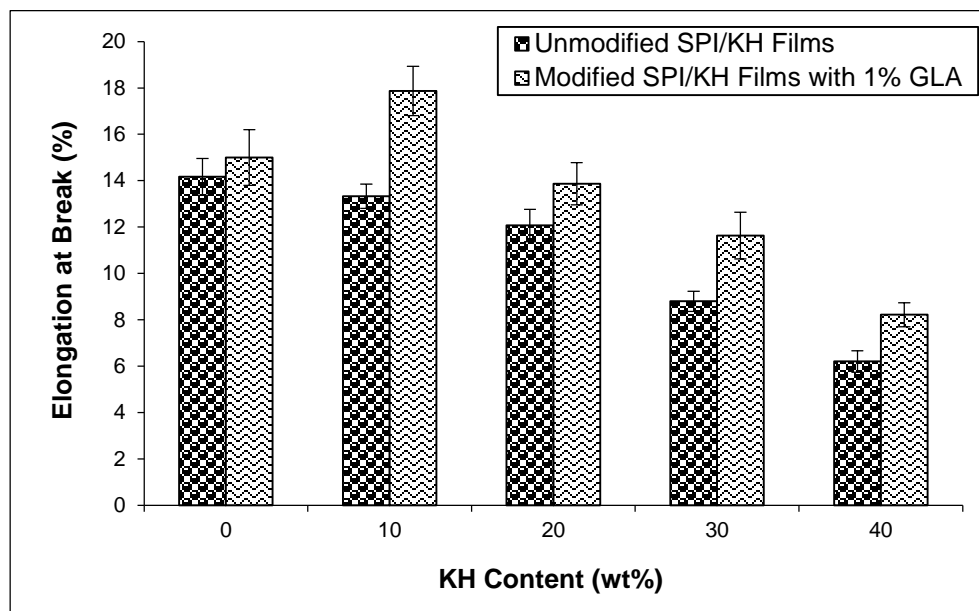


Fig. 5. Mean (\pm SD) elongation at break of unmodified and modified SPI/KH films

Fourier Transmission Infrared Spectroscopy (FTIR)

Figure 6 exhibits the FTIR spectra of unmodified and modified SPI/KH films. The broad peak at 3274 cm^{-1} was due to the $-\text{OH}$ and $-\text{NH}$ groups. SPI can form inter- and intra-molecular hydrogen bonds with other amino acid groups in the protein structure (Lodha and Netravali 2005a). The absorption band at 2936 cm^{-1} and 2876 cm^{-1} were due to the stretching of the $-\text{CH}$ bond. Peaks due to $\text{C}=\text{O}$ stretching at 1634 cm^{-1} (amide I), $\text{N}-\text{H}$ bending at 1540 cm^{-1} (amide II), and $\text{C}-\text{H}$ deformation at 1453 cm^{-1} were observed. The frequency peak at 1402 cm^{-1} was related to the $-\text{CH}_2$ deformation from cellulose or it could also be caused by the $\text{C}-\text{H}$ deformation in lignin. Moreover, $\text{C}-\text{H}$ groups were displayed by the band at 1314 cm^{-1} . The absorption peak at 1238 cm^{-1} was due to the $\text{C}-\text{N}$ bond stretching and the $\text{N}-\text{H}$ bending (amide III). Five characteristic peaks at 1107 cm^{-1} ($\text{C}-\text{O}$ stretching), 1036 cm^{-1} ($\text{C}-\text{O}$ stretching), 994 cm^{-1} , 922 cm^{-1} , and 854 cm^{-1} ($\text{C}-\text{C}$ skeletal vibrations) were observed in both SPI/KH films. A similar finding was reported by (Schmidt *et al.* 2005). From Fig. 6, it can be seen that the intensity peak at 1634 cm^{-1} increased about 2.1% after the SPI/KH films were modified with GLA. The increase in the peak intensity was mainly contributed by the formation of the imine linkages ($\text{C}=\text{N}$) from the GLA-SPI crosslinking chain overlapped with the $\text{C}=\text{O}$ stretching from the SPI. Additionally, the intensity of the band at 1540 cm^{-1} was also increased around 2.9% because the ethylenic ($\text{C}=\text{C}$) groups from the GLA-SPI crosslinking chain overlapped with the $\text{N}-\text{H}$ bend groups from

the SPI. Moreover, the intensity of hydroxyl groups was also increased about 0.2% after SPI/KH films were chemically modified with GLA. This is evidenced to the presence of inter or intra hydrogen overlapped with the –NH and –OH groups of SPI/KH films. According to previous studies (Yeng *et al.* 2013a), the presence of imine bands (C=N) and ethylenic (C=C) groups indicate a Schiff base cross-linking reaction occurred in biocomposite films.

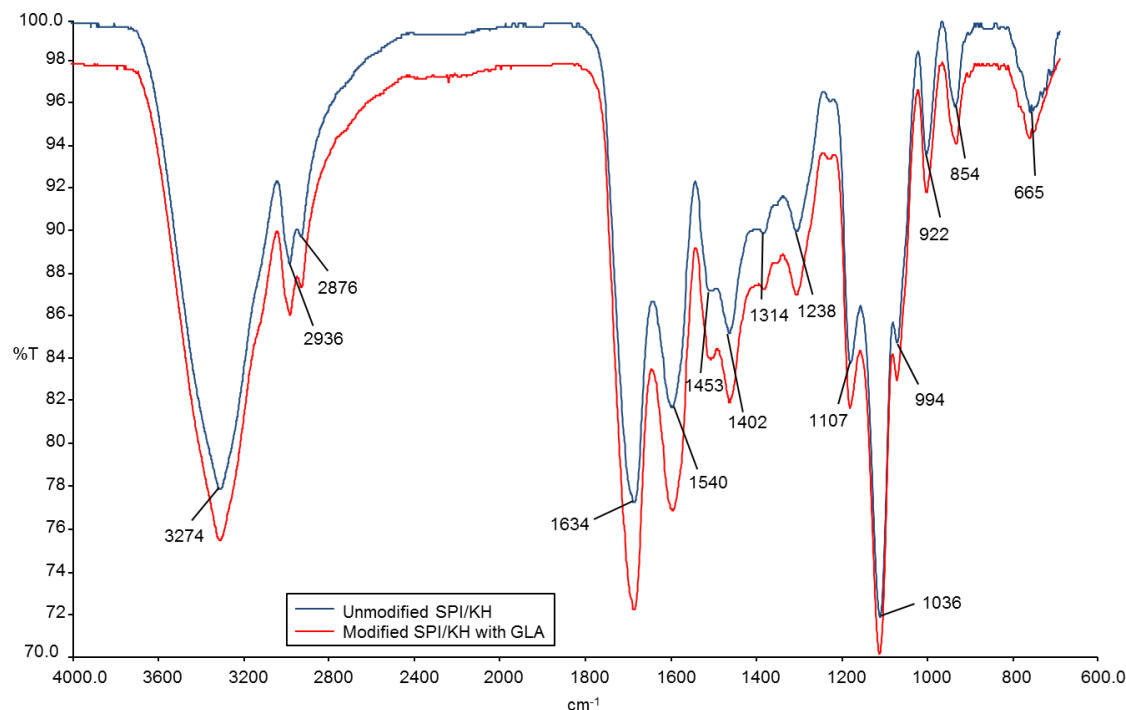


Fig. 6. FTIR analysis of unmodified and modified SPI/KH films

Thermogravimetric Analysis

Figure 7 illustrates the TGA curves of the unmodified and modified SPI/KH films. The TGA curve for neat SPI film (plasticized with glycerol) exhibits two stages of degradation of weight loss. KH also was decomposed in two stages. The first stage of weight loss was attributed to evaporation of water, and the second stage showed a decomposition peak within the range temperature of 200 to 500 °C, which was related to the decomposition of hemicellulose, cellulose, and lignin. However, unmodified and modified SPI/KH films showed three stages of degradation. The initial weight loss of neat SPI, unmodified and modified films began below 100 °C due to the evaporation of the absorbed moisture. The second stage of weight loss occurred approximately between 200 and 400 °C due to the KH degradation and decomposition of hemicellulose, cellulose, and lignin. Fisher *et al.* (2002) reported that thermal decomposition of cellulosic materials will take place at a temperature in the range of 200 to 400 °C. The weight loss at temperature of 200, 300, and 400 °C of unmodified SPI/KH films decreased as KH content increased, as revealed in Table 3. Besides that, the KH filler showed lower weight loss at temperatures of 200, 300, and 400 °C compared to neat SPI film. This finding is evidence of the chemical composition of KH filler as shown in Table 1. Accordingly, lignin decomposed at temperatures from 250 to 500 °C due to its thermally stable nature, therefore contributing to the formation of char residue (Abdullah *et al.* 2010). Besides, the weight loss of modified

SPI/KH with GLA increased with increasing KH content. Nevertheless, at a similar KH content, the weight loss at 200, 300, and 400 °C of modified SPI/KH films was lower compared to the unmodified films. These results indicate that the modified films with GLA had a higher degradation temperature compared to the unmodified films. From Table 2, it can be seen that the residue remaining after 600 °C of neat SPI film was lower than with KH filler. The unmodified SPI/KH films exhibited an increase trend in percentage of residue remaining at 600 °C. These results indicated that the incorporation of KH filler enhanced the thermal stability of SPI/KH films. Moreover, the residue remaining at 600 °C of modified SPI/KH films was increased with increasing KH filler. However, the percent residue at 600 °C of modified SPI/KH films was higher than unmodified films. This is because the GLA cross-linking agent enhanced the thermal stability of SPI/KH films with formation of imine linkages. The formation of char residue provided a barrier between the heat source and the polymeric materials.

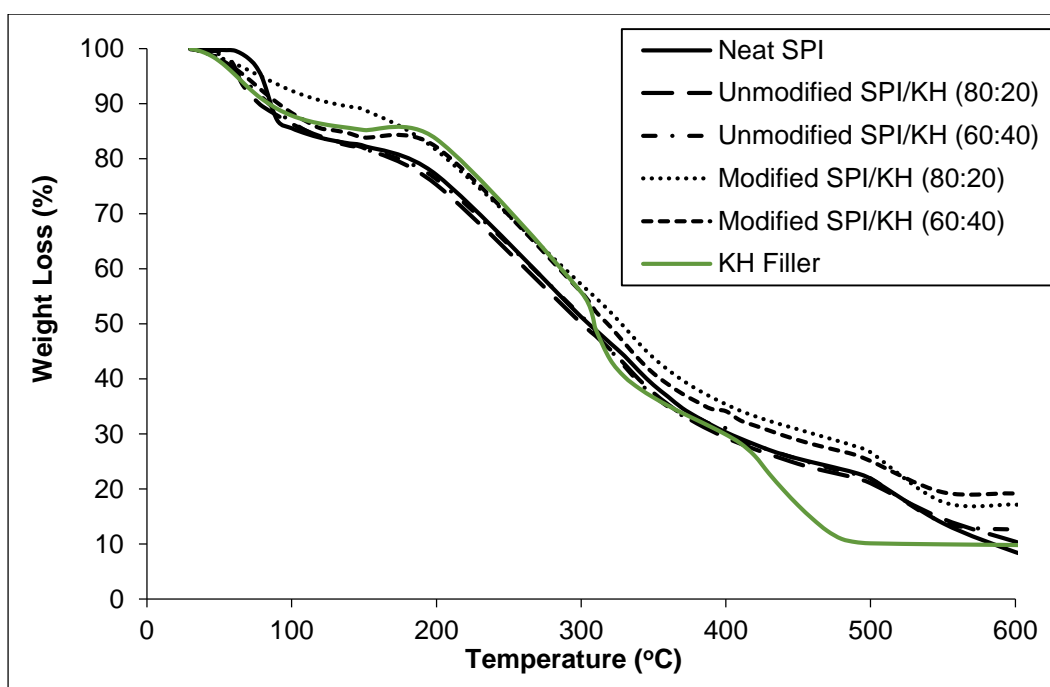


Fig. 7. TGA curves of unmodified and modified SPI/KH films

Table 3. TGA data of Unmodified and Modified SPI/KH Films

| Sample | Weight Loss (%) | | | Residue remaining at 600 °C (%) |
|----------------------------------|---------------------|---------------------|---------------------|---------------------------------|
| | $T_{200\text{ °C}}$ | $T_{300\text{ °C}}$ | $T_{400\text{ °C}}$ | |
| KH filler | 16.39 | 44.20 | 67.98 | 9.83 |
| Neat SPI film | 22.88 | 48.68 | 69.65 | 8.59 |
| Unmodified SPI/KH (80:20) | 24.68 | 49.85 | 70.64 | 10.47 |
| Unmodified SPI/KH (60:40) | 23.68 | 48.47 | 68.92 | 12.62 |
| Modified SPI/KH (80:20) with GLA | 18.41 | 42.81 | 64.59 | 17.17 |
| Modified SPI/KH (60:40) with GLA | 17.85 | 44.19 | 65.49 | 19.24 |

Morphology Study

Figure 8 displays the SEM micrograph of KH filler. This figure indicates that KH incorporates various shapes of filler particulates, fibers, *etc.* The SEM micrograph of neat SPI is shown in Fig. 9. The tensile fracture surface of neat SPI was homogeneous and smooth. The SEM micrograph of the unmodified film at 20 wt% KH gave evidence of poor interfacial adhesion between SPI matrix and KH filler, as illustrated in Fig. 10. Figure 10 also indicates that some KH was pulled out and detached from the matrix.

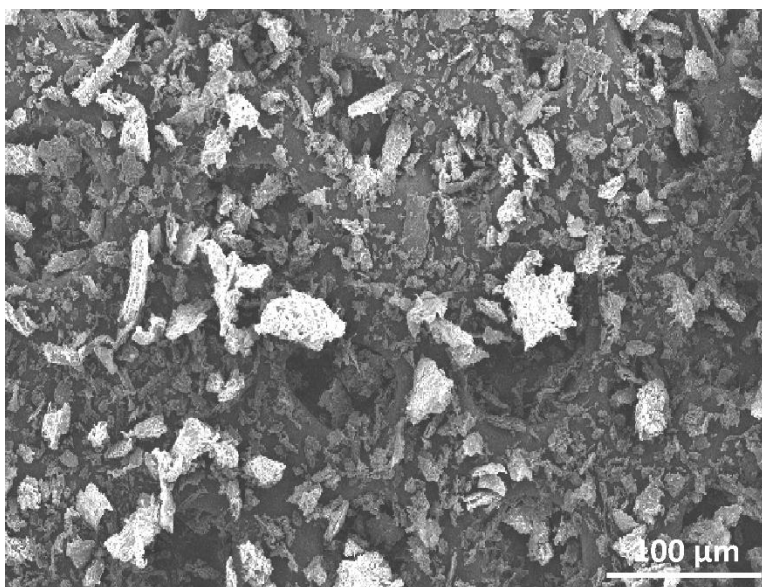


Fig. 8. SEM micrograph of KH filler

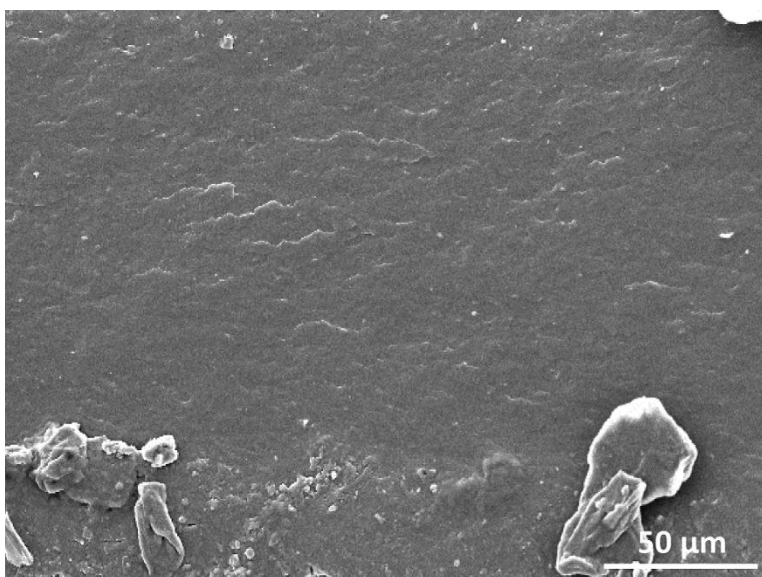


Fig. 9. SEM micrograph of neat SPI

The SEM micrograph of film at 40 wt% KH, as shown in Fig. 11, exhibited better wettability between the filler and the matrix. This result was supported by the tensile strength of the unmodified film, which increased with an increasing amount of KH. Figure 12 shows the SEM micrograph of modified neat SPI film with GLA.

The tensile fracture surface of modified neat SPI film shows a rough surface and a discontinuous fracture path, which are characteristic of a SPI/KH film with the presence of GLA. Yeng *et al.* (2014) reported that the discontinuous fracture path on tensile fracture surface of neat polymer film might be due to the crosslinked state of a polymer film. The presence of GLA improved the adhesion of the modified films as shown in Figs. 13 and 14, respectively. Both SEM micrographs of the modified films showed that the tensile fracture surface had better adhesion and wettability in the SPI matrix compared to the SEM micrograph of the unmodified films.

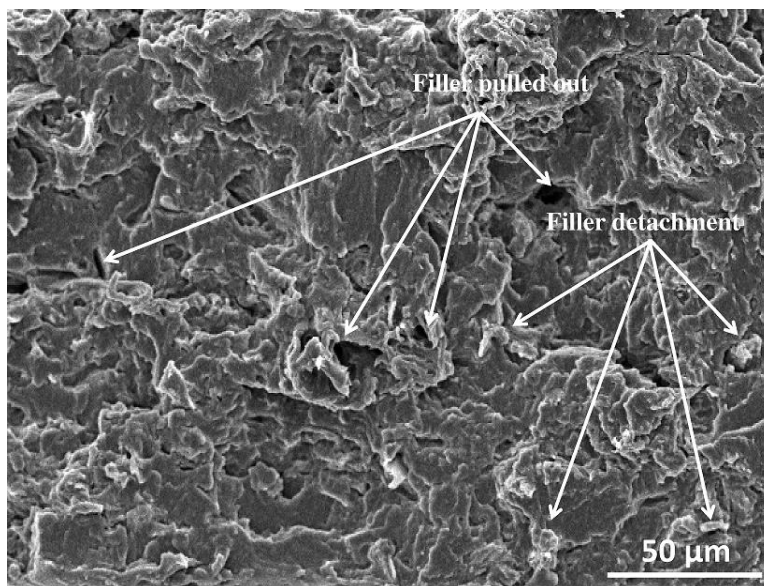


Fig. 10. SEM micrograph of unmodified SPI/KH film at 20 wt% KH

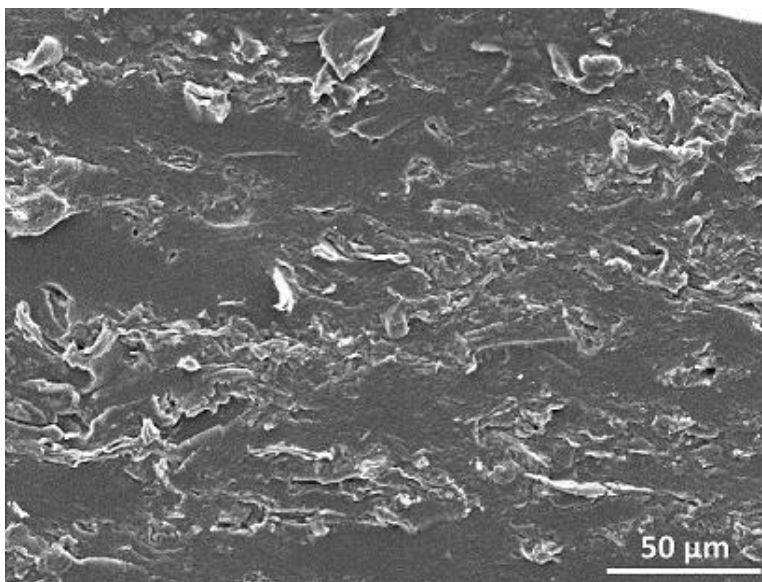


Fig. 11. SEM micrograph of unmodified SPI/KH film at 40 wt% KH

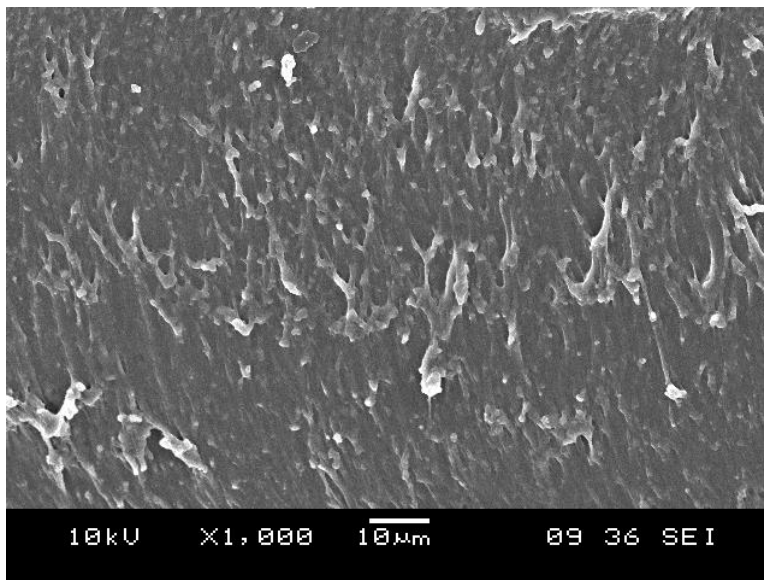


Fig. 12. SEM micrograph of modified neat CS film

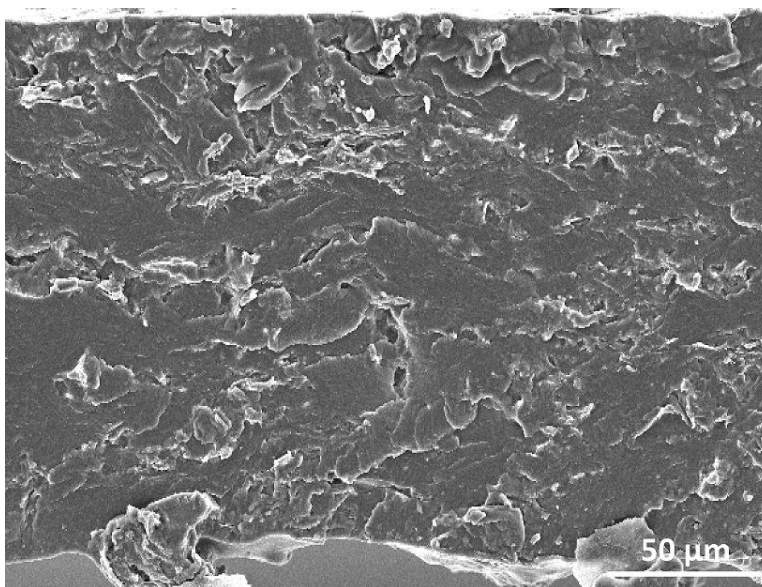


Fig. 13. SEM micrograph of modified SPI/KH at 20 wt% KH

Enzymatic Degradation

The effects of KH content on enzyme degradation of both unmodified and modified neat SPI and SPI/KH films are illustrated in Fig. 15. Table 4 summarizes the degree of enzyme degradation after 14 days. It was found that the neat SPI was hydrolyzed completely in 14 days. The weight loss after 14 days was reduced with increasing KH content. This is due to the fact that the KH has a higher lignin content (Berry 1979), which is resistant to degradation by α -amylase. Additionally, the lignin in the KH filler formed a barrier that inhibited cellulose in KH from degradation.

In contrast, the modified neat SPI film with GLA exhibited lower weight loss of degradation α -amylase buffer solution than unmodified neat SPI film. Similarly, modified SPI/KH films with GLA showed more resistance to hydrolysis in the α -amylase buffer solution than the unmodified form. This is due to the modification of neat SPI and SPI/KH

films with GLA, which exhibited a lower moisture absorption compared to the unmodified films.

Again, as explained earlier, the presence of GLA resulted in the improvement of the interfacial interaction between the matrix and the filler, as well as the formation of imine linkages, which led to a reduction in the enzyme degradation.

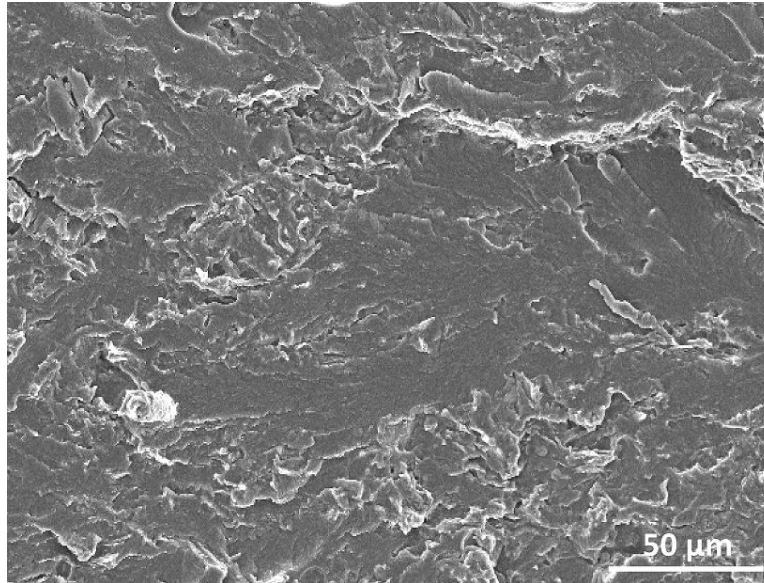


Fig. 14. SEM micrograph of modified SPI/KH film at 40 wt% KH

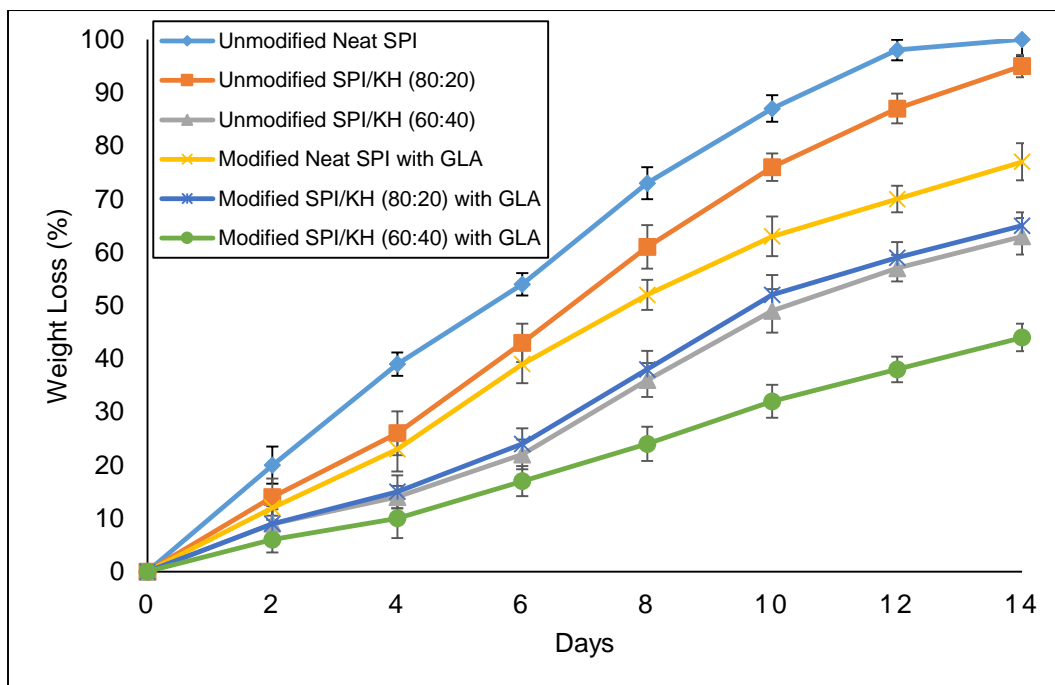


Fig. 15. Weight loss of both unmodified and modified SPI/KH in enzyme degradation

Table 4. Weight Loss of Enzyme Degradation of both Unmodified and Modified SPI/KH Films after 14 Days

| Materials | Weight loss of enzyme degradation after 14 Days (%) |
|----------------------------------|---|
| Unmodified Neat SPI | 100 ± 3.2 |
| Unmodified SPI/KH (80:20) | 95 ± 2.1 |
| Unmodified SPI/KH (60:40) | 63 ± 3.2 |
| Modified Neat SPI with GLA | 77 ± 3.1 |
| Modified SPI/KH (80:20) with GLA | 65 ± 2.5 |
| Modified SPI/KH (60:40) with GLA | 44 ± 2.6 |

CONCLUSIONS

1. The tensile strength and modulus of elasticity of both unmodified and modified neat SPI and SPI/KH films increased with increasing KH content. In contrast, the elongation at break was reduced with increasing KH content. GLA, which was used as the cross-linking agent in the SPI/KH films, improved the tensile and thermal properties of the films due to formation of imine linkages between aldehyde groups of GLA and amino groups of SPI through Schiff base reaction.
2. The weight loss enzyme degradation of SPI/KH films decreased with increasing KH content due to the fact that the KH filler contained high lignin content, which contributes resistance to being hydrolyzed by α -amylases enzyme. Nevertheless, GLA modified neat SPI and SPI/KH films showed lower weight loss of enzymatic degradation than unmodified films.
3. The FTIR results of SPI/KH films indicated the presence of imine C=N and ethylenic C=C bond after modification.
4. GLA enhanced the adhesion between the SPI matrix and the KH filler, as demonstrated by the SEM morphology study.

ACKNOWLEDGMENTS

This research was supported by the Ministry of Education (MOE). The authors would like to thank the Research Acculturation Collaboration Effort (RACE) 2014 for financial support.

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Article submitted: May 29, 2014; Peer review completed: July 11, 2014; Revised version received and accepted: July 22, 2014; Published: July 29, 2014.