

PROPERTIES OF SAWDUST/POLY(BUTYLENE SUCCINATE) COMPOSITES: EFFECT OF FLAME RETARDANT AND COMPATIBILIZER

Rapisa Jarapanyacheep and Kasama Jarukumjorn*

Received: May 14, 2019; Revised: November 06, 2019; Accepted: November 09, 2019

Abstract

Sawdust/poly(butylene succinate) (PBS) composites were prepared at sawdust content of 30 wt%. Effect of ammonium polyphosphate (APP) contents on mechanical, flame retarding, thermal and morphological properties of the composites was investigated. Tensile strength, elongation at break, impact strength and thermal stability of PBS composites were reduced with increasing APP content while tensile modulus was enhanced. Flame retardancy of the composites was improved with the incorporation of 20 phr of APP. Increasing poly(butylene succinate grafted maleic anhydride) (PBS-g-MA) content resulted in the increment of mechanical properties but the reduction of thermal stability of the composites filled with APP. SEM micrographs confirmed the enhancement of compatibility of the composite and revealed some features of ductile fracture in the composites with the presence of PBS-g-MA.

Keywords: PBS, sawdust, APP, PBS-g-MA, mechanical properties, flame retardancy

Introduction

Nowadays, biodegradable polymers have gained more attention as a result of growing environmental awareness. Among the biodegradable polymers, poly(butylene succinate) (PBS) has attracted increasing interest in various industrial applications due to its biodegradability, melt processability and chemical resistance (Kim *et al.*, 2005).

However, utilization of this biodegradable polymer for substituting non-biodegradable polymers is restricted due to its relatively high cost. One of the possible solutions for reducing the cost of end product is to reduce the quantities of polymeric material required by incorporating low cost filler into polymers.

School of Polymer Engineering, Institute of Engineering, Suranaree University of Technology, Nakhon Ratchasima 30000, Thailand. Phone: +66-4422-4437, Fax: +66-4422-4605; E-mail: kasama@sut.ac.th

* Corresponding author

Many attempts have focused on making PBS composites from lignocellulosic materials which are low cost, renewability and biodegradability. Sawdust is one of lignocellulosic fillers obtaining from wood industries. The use of sawdust as reinforcing fillers in biodegradable polymers benefits to the environment and also adds value to the sawdust. However, the biodegradable polymers reinforced with the lignocellulosic fillers are flammable materials which could limit their applications (Dozen *et al.*, 2013). Generally, the method to improve fire resistance of flammable materials is to use flame retardants which are directly incorporated into the materials. Flame retardants are often preferred to provide low flammability to polymeric materials because the uses of flame retardants compromise between cost and properties. Ammonium polyphosphate (APP) is a very efficient halogen-free flame retardant mainly used in polyolefin composites. APP is a nontoxic and it does not generate additional quantities of smoke (Ashton, 2005). APP has also been used in PLA biocomposites (Shumao *et al.*, 2010; Chen *et al.*, 2011; Shukor *et al.*, 2014) and PBS biocomposites (Nie *et al.*, 2012; Dozen *et al.*, 2013). With adding APP, flame retarding properties of flax fiber/PBS composites were enhanced but thermal stability was reduced (Dorez *et al.*, 2013). Nie *et al.* (2012) found that microencapsulated ammonium polyphosphate improved flame retardancy of bamboo fibers/ PBS composites. However, another main drawback of the composites is incompatibility of the components which is responsible for poor properties. Compatibilizers can be used to increase the adhesion between the composite components. Thirmizir *et al.* (2011) reported that the addition of maleated PBS as a compatibilizer resulted in the increment of flexural and impact properties of kenaf bast fiber/PBS composites due to enhanced fiber-matrix interfacial adhesion. Maleic anhydride grafted BioPBS (MA-g-BioPBS) improved both mechanical and thermal properties of grape pomace (GP)/PBS composites. This was attributed to enhanced interfacial interaction between GP, BioPBS matrix and compatibilizer (Gowman *et al.*, 2018).

Many researchers focused on the effect of flame retardant or compatibilizer on the properties of lignocellulosic filler/PBS composites. However, no literature is reported on the effect of compatibilizer on the properties of lignocellulosic filler/PBS composites filled with APP. Therefore, in this study, the effect of APP content on mechanical, flame retarding, thermal and morphological properties of sawdust/PBS composites was examined. In addition, the influence of poly(butylene succinate) grafted maleic anhydride (PBS-g-MA) as a compatibilizer on the properties of PBS composites filled with APP was investigated.

Materials and Methods

Materials

PBS (FZ71PD) was purchased from PTT MCC Biochem Co., Ltd., Thailand. Indian Walnut sawdust (*Albizia lebbeck Benth*) was supplied from Huathalae Sawmill, Nakhon Ratchasima, Thailand. Ammonium polyphosphate (APP, Exolit AP 765, Clariant, average diameter of 8 μm) was supplied by Clariant Chemical (Thailand), Co., Ltd. Dicumyl peroxide (DCP, Aldrich Chemicals) and maleic anhydride monomer (MA, Sigma-Aldrich) were used.

Preparation of Sawdust

Sawdust was sieved using 40-60 mesh screen (425-600 μm). Then the sawdust was dried in an oven at 60°C for 24 h.

Preparation of Poly(butylene succinate) Grafted Maleic Anhydride (PBS-g-MA)

PBS-g-MA was prepared using an internal mixer (Hakke Rheomix, 3000P) at temperature of 140°C for 5 min and a rotor speed of 50 rpm. Dicumyl peroxide content was 1 phr and maleic anhydride monomer content was 10 phr.

Determination of MA Content

The quantity of maleic anhydride content on PBS was determined by titrating carboxylic acid groups derived from the anhydride functional groups. 1 g of PBS-g-MA was

dissolved in 100 ml of chloroform at its boiling temperature then the solution was poured into methanol and filtered. In order to remove the unreacted MA and the DCP residues, the sample was washed with methanol. Finally, the polymer was dried in an oven at 80°C for 24 h. The sample was redissolved in chloroform. Then, 5 ml of distilled water was added to the hot solution to hydrolyze the anhydride groups. Immediately, the hot solution was titrated with 0.025 N ethanolic KOH. The solution of 1% phenolphthalein in ethanol was used as an indicator. The grafting degree of MA to PBS was calculated with Equation (1).

$$\text{Grafting degree of MA} = \frac{98.06c(V-V_0)}{2 \times 1000m} \times 100\% \quad (1)$$

where V and V_0 (ml) are volumes of the KOH solution used for titration of the grafted and blank samples, respectively; c is the molar concentration of the KOH solution (mol/l); 98.06(g/mol) is the molecular weight of MA; m (g) is the weight of PBS-g-MA (Thirmizir *et al.*, 2011).

Fourier Transform Infrared (FTIR) Spectroscopy

FTIR analysis was carried out on PBS, MA, and PBS-g-MA using Fourier transform infrared spectrometer (Bruker, Tensor 27). It was performed through wavelength of 4,000-400 cm^{-1} with 32 scanning times and resolution of 4 cm^{-1} .

Composite Preparation

Composition and their designation of PBS and PBS composites are shown in Table 1. The composites were mixed in a twin screw extruder (Brabender, DSE 35/17D). The

temperatures of the four processing zones were 125, 130, 135, and 140°C. The screw speed was 20 rpm. The test specimens were molded by an injection molding machine (Tederic, TRX60C).

Composite Characterization

Tensile properties of PBS and PBS composites were examined according to ASTM D638 using a universal testing machine (Instron, 5565) with a load cell of 5 kN and a crosshead speed of 20 mm/min.

Izod impact strength of PBS and PBS composites was performed according to ASTM D256 using an impact testing machine (Atlas, BPI). Notched impact strength was tested using the impact pendulum with impact energy of 2.7 J.

Flammability of PBS and PBS composites was classified by the vertical burning test according to ASTM D3801.

Surface morphologies of tensile fractured surface of PBS composites were examined using a scanning electron microscope (SEM, JEOL JCM-6010) at 10 kV. The samples were coated with gold before examination.

Thermal behaviors of PBS and PBS composites were analyzed using a thermogravimetric analyzer (Mettler Toledo, TGA/DSC1). Thermal decomposition of each sample was examined under nitrogen atmosphere. The specimens were heated from room temperature to 800°C with a heating rate of 10°C/min.

Results and Discussion

Characterization of PBS-g-MA

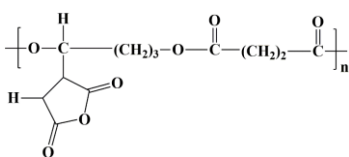
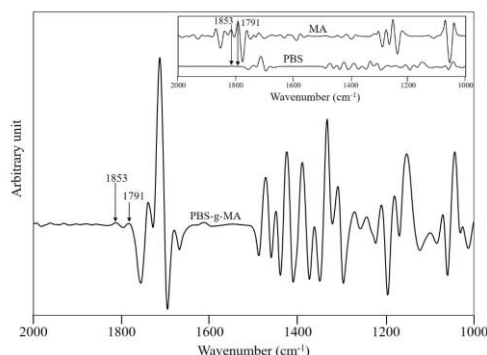
The chemical structure of PBS-g-MA is displayed in Figure 1. The second derivative of

Table 1. Composition and their designation of PBS and PBS composites

Sample	PBS (wt%)	Sawdust (wt%)	APP (phr)	PBS-g-MA (phr)
PBS	100	-	-	-
Sawdust/PBS	70	30	-	-
10APP	70	30	10	-
20APP	70	30	20	-
20APP_3C	70	30	20	3
20APP_5C	70	30	20	5
20APP_10C	70	30	20	10

Table 2. Mechanical properties of PBS and PBS composites

Sample	Tensile strength (MPa)	Tensile modulus (GPa)	Elongation at break (%)	Impact strength (kJ/m ²)
PBS	39.50±0.62	0.57±0.07	32.60±1.56	6.51±0.29
Sawdust/PBS	29.62±0.85	0.69±0.03	8.45±0.56	4.00±0.30
10APP	27.36±0.62	0.69±0.03	6.61±0.47	3.84±0.23
20APP	25.91±1.01	0.75±0.04	4.84±0.48	3.36±0.24
20APP_3C	27.72±1.23	0.80±0.02	5.58±0.22	5.62±0.18
20APP_5C	28.02±1.06	0.84±0.02	5.86±0.17	6.54±0.32
20APP_10C	32.39±1.93	0.86±0.04	6.54±0.19	7.24±0.63

**Figure 1. Chemical structure of PBS-g-MA****Figure 2. Second derivative of FTIR spectra of PBS, maleic anhydride and PBS-g-MA**

FTIR spectra were used for improving resolution and enhancing strongly overlapping peaks (Mani *et al.*, 1999). The second derivative FTIR spectra of PBS, MA and PBS-g-MA are displayed in Figure 2. The bands at 1,045 cm⁻¹ was due to -O-C-C- stretching vibrations in PBS. Peaks in the range of 1,144-1,264 cm⁻¹ resulted from the stretching of -C-O-C- group in the ester linkages of PBS. The band at the 1,713 cm⁻¹ was attributed to C=O stretching vibrations of ester groups in PBS. The peak at 1,325 cm⁻¹ was assigned to the symmetric deformational vibrations of -CH₂- groups in PBS main chains (Phua *et al.*, 2011). There

were peaks at 1,791 and 1,853 cm⁻¹ which were assigned to symmetric and asymmetric stretching of C=O bonds for the succinic anhydride groups in PBS-g-MA but these were absent in neat PBS (Mani *et al.*, 1999; Tserki *et al.*, 2006; Sclavons *et al.*, 2005). This confirmed the reaction in the grafting process. Grafting degree of PBS-g-MA from the acid titration method was 1.17%.

Mechanical Properties

Mechanical properties of PBS and PBS composites are listed in Table 2. With the incorporation of sawdust into PBS, tensile strength of the composite was decreased due to the lack of interfacial adhesion between sawdust and PBS which allows inefficient stress transfer between two phases of the composite (Kim *et al.*, 2005; Gowman *et al.*, 2018). Elongation at break of PBS and its composite were about 32 and 8%, respectively. The decrease in the elongation at break of the composite was attributed to poor compatibility of the components and reduced polymer chain entanglement (Nagarajan *et al.*, 2013). However, tensile modulus of sawdust/ PBS composite was higher than that of PBS indicating the ability of sawdust to impart greater stiffness to the composite (Liang *et al.*, 2010). Impact strength of PBS composite was also lower than that of PBS. This was because sawdust acted as a stress concentrator causing the impact strength to decrease.

Tensile strength, elongation at break and impact strength of sawdust/ PBS composites were reduced with increasing APP content. This was due to poor adhesion between APP and PBS matrix and agglomeration of APP in

PBS matrix (Shumao *et al.* 2010). This was confirmed by SEM micrographs in Figure 3. On the other hand, tensile modulus of the composites improved with increasing APP content. APP was a rigid small particle enhancing the stiffness of the composites (Yu *et al.*, 2014).

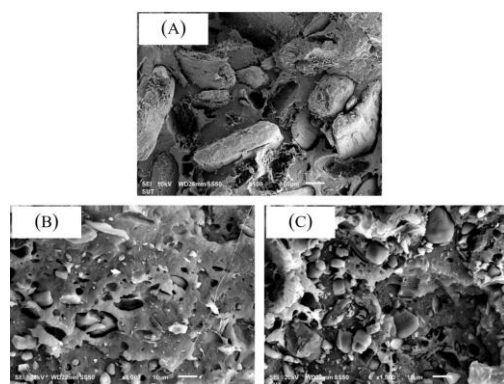


Figure 3. SEM micrographs of (A) sawdust/PBS (100X), (B) 10APP (1000X) and (C) 20APP (1000X)

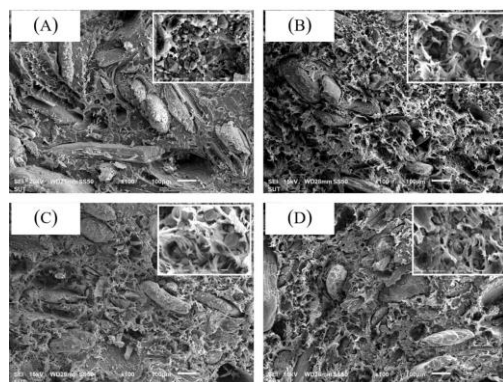


Figure 4. SEM micrographs of (A) 20APP, (B) 20APP_3C, (C) 20APP_5C and (D) 20APP_10C at magnification of 100X and 1000X

The increment of tensile strength, elongation at break, tensile modulus and impact strength of sawdust/PBS composites filled with 20 phr of APP were found with the presence of PBS-g-MA. This was due to the formation of better fillers-matrix adhesion (Demir *et al.*, 2006; Kim *et al.*, 2011) which

was confirmed by SEM images in Figure 4. Moreover, the mechanical properties of the composite continuously enhanced with increasing compatibilizer content because interfacial adhesion between fillers and matrix was enhanced (Thirmizir *et al.*, 2011). However, tensile strength and elongation at break of all the composites were lower than neat PBS.

Morphological Properties

SEM micrographs of tensile fractured surface of sawdust/PBS composites with and without APP are shown in Figure 3. It can be seen that not only holes left after sawdust pull out from PBS matrix but also gap between sawdust and matrix were observed as displayed in Figure 3(A) because of incompatibility between sawdust and PBS. Moreover, poor adhesion and agglomeration of APP were found in matrix as shown in Figure 3(B-C). This indicated the poor interfacial adhesion of fillers and PBS matrix leading to the reduction of the tensile strength, elongation at break and impact strength of PBS composites.

Adding PBS-g-MA into the composites resulted in improved the compatibility of fillers and matrix. In addition, the enhancement of ductile deformation of the matrix was confirmed as shown in Figure 4(B-D). This was evidence from long fibrils pulled out during testing whereas this did not occur in the composite without PBS-g-MA as displayed in Figure 3(A). This led to an increase in elongation at break and impact strength of the composites.

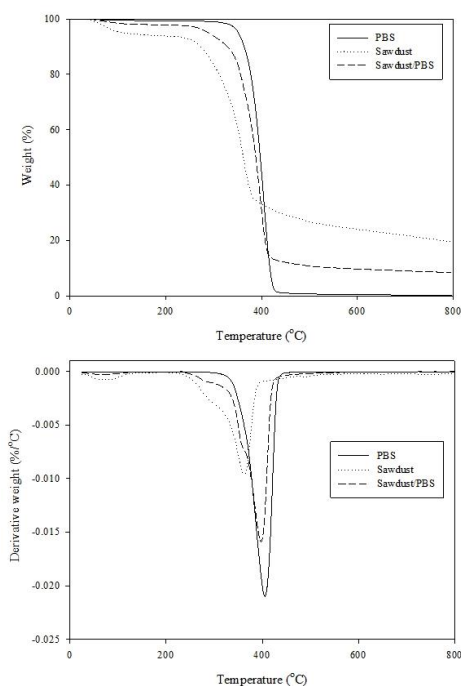
Flammability

UL-94 test results of PBS, sawdust/PBS composite and PBS composites with APP are shown in Table 3. It can be seen that PBS was classified as V-2 in UL-94 classification. After the addition of sawdust in PBS, the composite was not classified in UL-94 classification indicating that sawdust/PBS composite was a flammable material. The composite with 10 phr of APP was also not classified in UL-94 classification. This was because APP content was not enough to form sufficient char protective layer during the combustion. This suggested that higher loading of APP was

Table 3. Weight loss temperature and UL-94 classification of PBS, sawdust and PBS composites

Sample	Weight loss temperature (°C)			Residue (%)	UL 94 classification
	1 st	2 nd	3 rd		
PBS	-	-	405.67	0	V-2
Sawdust/PBS	278.62	355.63	397.50	8.39	NC
10APP	280.93	356.33	376.33	10.98	NC
20APP	282.01	-	344.17	15.69	V-2
20APP_3C	280.54	-	343.83	16.58	V-2
20APP_5C	280.52	-	342.50	14.80	V-2
20APP_10C	275.93	-	341.33	14.72	V-2

NC: no classification, complete combustion of the specimens

**Figure 5. TGA and DTG curves of PBS, sawdust and PBS composites**

required to increase the flame retardancy of the composite. With the incorporation of APP at 20 phr, PBS composite was classified as V-2. This was because APP produced char protective layer separating the composite from heat and oxygen (Shumao *et al.*, 2010). This can be confirmed by TGA results (Table 3) that char residue of the composites increased with increasing APP content. On the other hand, PBS-g-MA content insignificantly affected UL-94 test results of the composites. This was

corresponded with char residue of the composites that did not substantially change with PBS-g-MA content.

Thermal Properties

TGA and DTG curves of PBS, sawdust and sawdust/ PBS composite are shown in Figure 5 and their weight loss temperature and char residue are listed in Table 3. Decomposition temperature of PBS was 405.67°C. Sawdust showed three stages of decomposition process. The 1st weight loss step was due to evaporation of moisture at temperature of 63.72°C. The 2nd weight loss step and the 3rd weight loss step were caused by hemicellulose degradation and cellulose degradation, respectively (Ouajai *et al.*, 2005). Char residue of sawdust was 19.52%. Sawdust/ PBS composite showed three weight loss temperature at 278.62, 355.63, and 397.50°C due to decomposition of hemicellulose, cellulose and PBS, respectively. Thermal stability of PBS was reduced with the incorporation of the sawdust due to low thermal decomposition temperature of sawdust. Lee and Wang (2006) also observed similar results in bamboo fiber/PBS composites.

From TGA and DTG curves of PBS composites filled with APP as displayed in Figure 6, with adding 10 phr of APP into PBS composite, the weight loss temperatures were observed at 280.93, 356.63, and 376.33°C due to hemicellulose, cellulose and APP, and PBS degradation, respectively. APP had decomposition temperature about 300-500°C (Samyn *et al.*, 2007). However, there were two weight loss steps in the composite filled with 20 phr of APP at temperature of 282.01 and 344.17°C. This indicated that weight loss temperature of

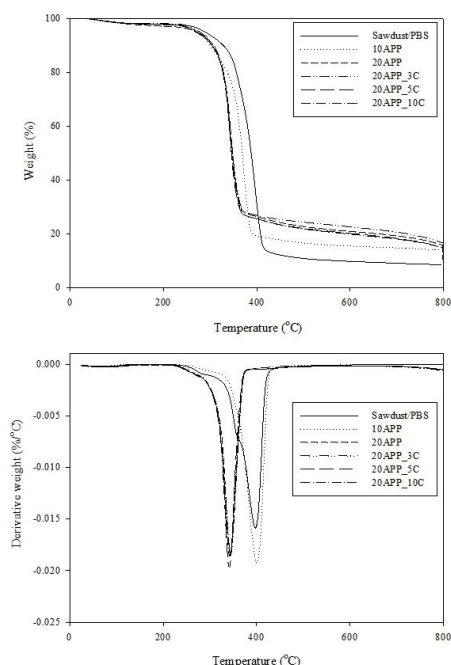


Figure 6. TGA and DTG curves of PBS composites with different APP and PBS-g-MA contents

PBS was reduced and overlapped with the weight loss temperature of APP since thermal decomposition of APP might accelerate thermal decomposition of polymer matrix x (Siat *et al.*, 1997; Duquesne *et al.*, 2001). The weight loss temperature of PBS in the composites continuously decreased with an increase in APP content. Char residue of the composites increased with APP content as listed in Table 3, especially with filling 20 phr of APP, thus this resulted in enhanced flame retardancy. Dorez *et al.* (2003) also found that adding APP led to the reduction of thermal stability but the increment of residue of flax fiber/PBS composites.

The incorporation of PBS-g-MA slightly reduced thermal stability of the composites as displayed in Table 3. Lee *et al.* (2013) found that decomposition of PBS-g-MA compatibilized kenaf fiber/PBS composites was lower than that of uncompatibilized composite. It could probably be due to the presence of

carboxylic acid groups of MA which were formed during its decomposition catalyzed the decomposition of polymer (Roumeli *et al.*, 2015). In addition, the presence of peroxide residue used for grafting process can cause the initiation of polymer decomposition at lower temperatures (Pracella *et al.*, 2006; Araujo *et al.*, 2008; Lee *et al.*, 2013). Compatibilizer content insignificantly influenced char residue of the composites.

Conclusions

With the addition of sawdust, tensile strength, elongation at break, impact strength, thermal stability and flame retardancy of PBS were deteriorated whereas tensile modulus was found to improve. Increasing APP content resulted in the reduction of tensile strength, elongation at break, impact strength and thermal stability but the enhancement of tensile modulus of sawdust/PBS composites. Flame retardancy of the composite was enhanced when 20 phr of APP was added. With adding PBS-g-MA, the composites showed the improvement in tensile properties and impact strength. However, tensile strength and elongation at break of the all composites were lower than neat PBS. The compatibilized composites were less thermally stable than the uncompatibilized composite. Compatibilizer content had no influence on flame retarding properties of the composites. The compatibilized composites showed good interface adhesion between constituents of the composites and also revealed ductile deformation which indicated by extensive elongated matrix in the fractured surface.

Acknowledgements

The authors wish to acknowledge Suranaree University of Technology and National Research Council of Thailand for the financial support, Clariant Chemical (Thailand), Co., Ltd. for providing ammonium polyphosphate, and Huathalae Sawmill for providing sawdust.

References

- Araujo, J.R., Waldman, W.R., and De Paoli, M.A. (2008). Thermal properties of high density polyethylene composites with natural fibres: Coupling agent effect. *Polym. Degrad. Stabil.*, 93(10):1,770-1,775.
- Ashton, H.C. (2005). Fire retardants. In: *Functional Fillers for Plastics*. Xanthos, M (ed). Wiley-VCH, Weinheim. p. 303-306.
- Chen, D., Li, J., and Ren, J. (2011). Combustion properties and transference behavior of ultrafine microencapsulated ammonium polyphosphate in ramie fabric-reinforced poly(L-lactic acid) biocomposites. *Polym. Int.*, 60(4): 599-606.
- Demir, H., Balkose, D., and Ulku, S. (2006). Influence of surface modification of fillers and polymer on flammability and tensile behavior of polypropylene-composites. *Polym. Degrad. Stabil.*, 91(5):1,097-1,085.
- Dorez, G., Taguet, A., Ferry, L., and Lopez-Cuesta, J.M. (2013). Thermal and fire behavior of natural fibers/PBS biocomposites. *Polym. Degrad. Stabil.*, 98(1): 87-95.
- Duquesne, S., Le Bras, M., Bourbigot, S., Delobel, R., Camino, G., Eling, B., Lindsay, C., Roels, T., and Vezin, H. (2001). Mechanism of fire retardancy of polyurethane using ammonium polyphosphate. *J. Appl. Polym. Sci.*, 82(13):3,262-3,274.
- Gowman, A., Wang, T., Rodriguez-Urbe, A., Amar, K., Mohanty, A.K., and Misra, M. (2018). Bio-poly (butylene succinate) and its composites with grape pomace: Mechanical performance and thermal properties, *ACS Omega*, 3:15,205-15,216.
- Kim, H.S., Yang, H.S., and Kim, H.J. (2005). Biodegradability and mechanical properties of agro-flour-filled polybutylene succinate biocomposites. *J. Appl. Polym. Sci.*, 97(4):1,513-1,521.
- Kim, S., Lee, B.H., Lee, S., Kim, H.J., and Dorgan, J.R. (2011). Enhanced interfacial adhesion, mechanical and thermal properties of natural flour-filled biodegradable polymer bio-composites. *J. Therm. Anal. Calorim.*, 104:331-338.
- Lee, J.M., Mohd Ishak, Z.A., Mat Tai, R., Law, T.T, and Thirmizir, M.Z.A. (2013). Mechanical, thermal and water absorption properties of kenaf fiber-based polypropylene and poly(butylene succinate). *J. Polym. Environ.*, 21(1):293-302.
- Lee, S.H., and Wang, S. (2006). Biodegradable polymers/ bamboo fiber biocomposite with bio-based coupling agent. *Compos. Part A-Appl. S.*, 37:80-91.
- Liang, Z., Pan, P., Zhu, B., Dong, T., and Inoue, Y. (2010). Mechanical and thermal properties of poly(butylene succinate)/plant fiber biodegradable composite. *J. Appl. Polym. Sci.*, 115(6):3,559-3,567.
- Mani, R., Bhattacharya, M., and Tang, J. (1999). Functionalization of polyesters with maleic anhydride by reactive extrusion. *J. Polym. Sci. Part A*, 37(11):1,693-1,702.
- Nagarajan, V., Mohanty, A., and Misra, M. (2013). Sustainable green composites: Value addition to agricultural residues and perennial grasses. *ACS Sustain. Chem. Eng.*, 1:325-333.
- Nie, S., Liu, X., Dai, G., Yuan, S., Cai, F., Li, B., and Hu, Y. (2012). Investigation on flame retardancy and thermal degradation of flame retardant poly (butylene succinate)/bamboo fiber biocomposites. *J. Appl. Polym. Sci.*, 125(S2):E485-E489.
- Ouajai, S. and Shanks, R.A. (2005) Composition, structure and thermal degradation of hemp cellulose after chemical treatments. *Polym. Degrad. Stabil.*, 89(2): 327-335.
- Phua, Y.J., Chow, W.S., Mohd Ishak, Z.A. (2011). The hydrolytic effect of moisture and hygrothermal aging on poly(butylene succinate)/ organo-montmorillonite nanocomposites. *Polym. Degrad. Stabil.*, 96(6):1,194-1,203.
- Pracella, M., Chionna, D., Anguillesi, I., Kulinski, Z., Piorkowska, E. (2005). Functionalization, compatibilization and properties of polypropylene composites with hemp fibres. *Compos. Sci. Technol.*, 66(13): 2,218-2,230.
- Roumeli, E., Terzopoulou, Z., Pavlidou, E., Chrissafis, K., Papadopoulou, E., Athanasiadou, E., Triantafyllidis, K., Bikiaris, D.N. (2015). Effect of maleic anhydride on the mechanical and thermal properties of hemp/high-density polyethylene green composites. *J. Therm. Anal. Calorim.*, 121(1):93-105.
- Samyn, F., Bourbigot, S., Duquesne, S., and Delobel, R. (2007). Effect of zinc borate on the thermal degradation of ammonium polyphosphate. *Thermochim. Acta.*, 456(2):134-144.
- Sclavons, M., Laurent, M., Devaux, J., and Carlier, V. (2005). Maleic anhydride-grafted polypropylene: FTIR study of a model polymer grafted by ene-reaction. *Polymer*, 46(19):8062-8067.
- Shukor, F., Hassan, A., Islam, M.S., Mokhtar, M., and Hasan, M. (2014). Effect of ammonium polyphosphate on flame retardancy, thermal stability and mechanical properties of alkali treated kenaf fiber filled PLA biocomposites. *Mater. Design.*, 54:425-429.
- Shumao, L., Jie, R., Hua, Y., Tao, Y., and Weizhong, Y. (2010). Influence of ammonium polyphosphate on the flame retardancy and mechanical properties of ramie fiber-reinforced poly(lactic acid) biocomposites. *Polym. Int.*, 59(2):242-248.
- Siat, C., Bourbigot, S., and Le Bras, M. (1997). Thermal behaviour of polyamide-6-based intumescent formulations a kinetic study. *Polym. Degrad. Stabil.*, 58(3):303-313.
- Thirmizir, M.Z.A., Ishak, Z.A.M., Taib, R.M, Rahim, S., and Jani, S.M. (2011). Kenaf bast fiber filled biodegradable poly(butylene succinate) composites: effects of fiber loading, fiber length, and maleated poly(butylene succinate) on the flexural and impact properties. *J. Appl. Polym. Sci.*, 122(5):3,055-3,063.
- Tserki, V., Matzinos, P., and Panayiotou, C. (2006). Novel biodegradable composites based on treated lignocellulosic waste flour as filler. Part II. Development of biodegradable composites using treated and compatibilized waste flour. *Compos. Part A-Appl. S.*, 37(9):1,231-1,238.
- Yu, T., Li, Y., and Wang, Y. (2014). Flammability and mechanical properties of ramie reinforced poly(lactic acid) composites by using DOPO. *J. Eng. Sci.*, 10:9-18.