INTERNATIONAL JOURNAL OF AUTOMOTIVE AND MECHANICAL ENGINEERING (IJAME)
ISSN: 2229-8649 e-ISSN: 2180-1606
VOL. 17, ISSUE 4, 8351 – 8359
DOI: https://doi.org/10.15282/ijame.17.4.2020.11.0631

ORIGINAL ARTICLE

Mechanical Properties and Biodegradability of Polylactic Acid/Acrylonitrile Butadiene Styrene with Cellulose Particle Isolated from Nypa Fruticans Husk

M. S. Rasidi[1,2*], L. C. Cheah[1] and A. M. Nasib[1]

[1]Fakulti Teknologi Kejuruteraan Kimia, Universiti Malaysia Perlis, 02600 Jejawi, Perlis, Malaysia
Phone: +6049798154; Fax: +6049798178
[2]Center of Excellence Geopolymer and Green Technology (CEGeoGTech), Universiti Malaysia Perlis, Arau, Perlis 02600, Malaysia

ABSTRACT – Polylactic acid is a biodegradable polymer derived from renewable resources, showing potentials in replacing traditional petroleum-based polymers, yet its brittleness limits its applications. Thus, blending polylactic acid with acrylonitrile butadiene styrene as well as incorporation of fillers were used to enhance the mechanical and biodegradability properties of polylactic acid by extrusion compounding. The aims of this study to produce and investigate PLA/ABS blend incorporated with natural filler, NFH and IC to improve the properties pf PLA/ABS blends. Two types of fillers used were *Nypa fruticans* husk and isolated cellulose from *Nypa fruticans* husk which was obtained by using Soxhlet extraction. Transform Infrared spectroscopy analysis was used to characterize and verified the extracted substance was isolate cellulose. Tensile, impact and biodegradation test were conducted to investigate the mechanical and biodegradability properties. The optimum blend ratio for polylactic acid/acrylonitrile was 75/25 php base on previous studies, and it was found that the incorporation of both fillers, *Nypa fruticans* husk and isolated cellulose from *Nypa fruticans* husk had decreased the tensile strength, elongation at break and impact strength of the composite however increased the Young's Modulus and biodegradation weight loss. Meanwhile, at similar filler content, the tensile strength, Young's modulus and biodegradation weight loss of polylactic acid/acrylonitrile butadiene styrene blend incorporated with isolated cellulose were higher value compared to polylactic acid/acrylonitrile butadiene styrene blend incorporated *Nypa fruticans* husk. Furthermore, morphological studies showed a well-coated filler by matrix and reduction of filler pull out when isolated cellulose was incorporated in polylactic acid/acrylonitrile butadiene styrene blend. Therefore, it was found that the incorporation of isolated cellulose in polylactic acid/acrylonitrile butadiene styrene blend, shows higher mechanical and biodegradation properties than polylactic acid/acrylonitrile butadiene styrene blend incorporated with *Nypa fruticans* husk.

ARTICLE HISTORY
Received: 25th Mar 2020
Revised: 30th Sept 2020
Accepted: 14th Oct 2020

KEYWORDS
PLA;
ABS;
Isolate cellulose;
Nypa fruticans;
Blend;
Composites

INTRODUCTION

PLA is a sustainable resource and biodegradable polymer, possessing mid-range mechanical, thermal, and processing properties. It gradually becomes an economic, industrial material used in the packaging field for its transparency and glossy surface. PLA also exhibits excellent physio-chemical properties and anti-fogging properties that make it useful in food packaging [1]. PLA is capable of being shaped, moulded, and extruded easily due to its processability [2]. However, PLA possesses low impact strength and brittle, withholding its usage in high impact application [3]. To overcome it, PLA can be blend with high impact strength polymer and incorporated with filler to facilitate its weakness.

ABS is an amorphous thermoplastic terpolymer possessing an excellent balance of properties, dimensional stability, chemical resistance, and non-toxic to human. Besides that, ABS is widely used as one of blending ingredient to improve mechanical properties especially impact toughness in the brittle polymer [4, 5]. By selecting ABS as blending material for PLA, the resulting blend capable of retaining PLA strength and stiffness while remedying its weak impact properties. This can be done through melt blending with an appropriate ratio between PLA and ABS, allowing the minor polymer to be dispersed in the primary matrix and facilitate mechanical properties [6].

Recently, the emergence of employing isolated cellulose from raw fibre as reinforcing material in biocomposite has gain popularity. Many published studies strongly indicate the huge reinforcing effect observed from compounding polymer with isolated cellulose. Cellulose possesses high specific strength and modulus due to its low density and crystallinity [7]. Also, it displays a higher affinity to organic/hydrophilic polymers, allows higher interaction between them [8]. Cellulose is one of the main components presents in a lignocellulosic fiber, following hemicellulose and lignin. Elimination of hemicellulose and lignin are vital as they hinder cellulose reinforcing effect, mechanical properties, and biodegradability [9-11].

Nypa fruticans (NF) is a palm plant species that thrives mostly in Southeast Asia such as Malaysia [12]. NF is also four seasons accessible, and recurrence of harvesting does not restrict its growth rate, ensuring long term supply of its

*CORRESPONDING AUTHOR | M. S. Rasidi | ✉ syahmie@unimap.edu.my

plant parts [13]. However, NF currently has limited usage in the field of agriculture. Several reviews on NF plant parts concludes that they are suitable to serve as natural fillers through further refine and treatment [14- 17]. A chemical characterisation of Nypa fruiticans husk (NFH) reviewed by Tamunaidu [18] shows that it has high cellulose content about 36.5%, making it a feasible source of cellulose. Till date, application of NFH cellulose in biocomposite has yet to be covered. In laboratory scale, isolation of NFH cellulose can be done through Soxhlet extraction method along with several chemical treatments as demonstrated by Norfatihah [19].

The objective of this study is to investigate the effect of natural filler, isolated cellulose (IC) from NFH and NFH toward the properties of PLA/ABS blend. The effect of IC and NFH loading in PLA/ABS biocomposites were evaluated in term of mechanical and morphological properties. Biodegradability of IC and NFH filled PLA/ABS biocomposites were assessed through enzymatic degradation. Further comparison between IC and NF filler were also evaluated.

METHODOLOGY

Materials

Ingeo^{TM} biopolymer 2003D PLA pellet was supplied from Innovative Pultrusion Sdn. Bhd., Penang. Whereas, ABS pellet was obtained from TT Biotechnologies Sdn. Bhd., Penang. Toluene, ethanol, sodium chlorite ($NaClO_2$), glacial acetic acid, sodium hydroxide solids, sodium acetate anhydrous, and diastase from malt were purchased from HmbG Chemical, Syarikat Saintifik Jaya, Malaysia.

NFH was obtained from the plantation at Simpang Empat, Perlis. The NFH was roughly cleaned and soaked for 24 hours before proceeding oven-dried at 80°C for 24 hours. The dried husks were then grounded into a fine powder and sieved with mesh size 75µm.

Method

The extraction of wax and impurities follow the method by Norfatihah [19]. The extraction was proceeded through Soxhlet extraction with a mixture of toluene and ethanol 2:1 v/v, with NFH powder filled into cellulose extraction thimble. The husks were refluxed for 48 hours for complete removal. Addition of boiling chips was to facilitate boiling distribution. The refluxed sample was retrieved and proceeded for water-soluble extraction.

Further extraction of water-soluble materials was done by refluxed the sample with its thimble was submerged in sufficient deionised water and boiled for 3 hours in a large beaker with constant stirring. Upon completion, the refluxed sample was rinsed repeatedly four times per 30 minutes with deionised water without heating and air-dried at room temperature. The boiled sample was retrieved and proceeded for the bleaching process.

Bleaching solution was prepared by dissolving 8g of sodium chlorite ($NaClO_2$) in 800 ml deionised water while stirred, followed by addition of 4 ml glacial acetic acid (CH_3COOH) and heated to 70°C. Five boiled samples in its thimble were added into bleaching solution and bleached until the reaction was completed to remove lignin. The endpoint of the bleaching process was determined by observing the solution maintained its bright yellow colour for more than 12 hours. During the reaction, the solution temperature, pH, and solution colour were maintained at 70°C, pH 4, and bright yellow colour by controlling heating temperature and balance the addition of sodium chlorite and glacial acetic acid. Once the reaction was completed, the bleached samples were dried and rinsed five times with deionised water about 30 minutes with stirring until conductivity less than <10 µS/cm. The bleached samples were then retrieved and proceeded for alkaline fibre treatment.

A 500ml of 17 wt.% sodium hydroxide (NaOH) solution was prepared by dissolving 85g of sodium hydroxide solid in 500ml of deionised water without extra heating due to exergonic reaction. Five bleached samples were immediately rinsed with the sodium hydroxide solution while stirring for 60 minutes to remove hemicellulose. The rinsing process was repeated five times. When completed rinsing, the samples were neutralised with 500ml of 10% v/v acetic acid solution while stirring for 60 minutes. The treated samples were then again rinsed with deionised water until conductivity less than <10 µS/cm. The treated samples were then dry in an oven at 70°C overnight, retrieved and then been ground before stored the IC in airtight condition.

Preparation of PLA / ABS / NFH and PLA / ABS / IC biocomposites

The PLA/ABS/NFH blends were compounded using LabTech Scientific 20 mm co-rotating twin-screw extruder. All specimens were compounded at 100 rpm screw speed, 180-200°C temperature profile, and residence time of 2 minutes. The extrudates were then pelletised and store for testing and characterisation. Similar compounding method for PLA/ABS/IC was used, and the formulation is as in Table 1.

Table 1. Formulation of PLA/ABS/NFH and PLA/ABS/IC biocomposites.

Filler	Specimen	PLA/ABS (wt%)	Filler Loading
	Control	75/25	0
NFH	PLA/ABS/NFH	75/25	10, 20, 30, 40
IC	PLA/ABS/IC	75/25	10, 20, 30, 40

Testing and Characterisation

Tensile and impact test

The tensile test was conducted by using 5960 Series universal testing systems Instron machine with accordance to ASTM D638 to determine the tensile properties of polymer blend and biocomposites. Izod impact test was conducted with reference to ASTM D256 using Izod impact tester. Tensile test specimens are dumbbell bar type IV with a thickness of 2 mm [20], while the impact test specimens are rectangular of 4 mm thickness, without notch [21]. Specimens were prepared from polymer blends and biocomposites using GT-7014-P GOTECH Plastic Hydraulic Molding Press at compression temperature of 220 °C with 1 minute preheat, 4 minutes of compression, and 4 minutes of cooling.

Enzymatic degradation test

Enzymatic degradation test was performed by referring to Yoon et al. method [22]. The neat PLA, neat ABS, PLA/ABS blend, PLA/ABS/NFH biocomposite, and PLA/ABS/IC biocomposite were immersed in a buffer solution containing α-amylase at 50 ºC. Buffer solution with pH 7.3 was prepared by adding 4.8 ml of 0.2M acetic acid to 45.2 ml of 0.2M sodium acetate solution, produced a 50 ml solution containing 10 mg of α-amylase. Specimens with a dimension of 2×2×0.2 cm were immersed in the solution and taken out every two days. The specimens were then rinsed with deionised water to remove excess α-amylase enzyme and oven-dried at 50 ºC for 24 hours. The total percentage of weight loss of specimens after 14 days was calculated using Eq. (1).

$$D_t = \frac{W_0 - W_1}{W_0} \times 100\% \tag{1}$$

where D_t is degradation test, W_0 is the initial weight of sample, and W_t is the weight of sample after immersed in buffer solution.

Morphological analysis

Morphological study of impact fractured surface of the polymer blends and biocomposites were carried out using a scanning electron microscope (SEM), Model JSM 6260 LE JOEL. The physical appearance of NFH, IC, and enzymatically degraded specimens were also observed using a similar machine. The impact fractured end of the specimens, NFH/IC powders, and the surface of enzymatically degraded specimens was mounted on aluminium stubs and sputter-coated with a thin layer of platinum to avoid electrostatic charging during the examination. The SEM micrographs were obtained at 10 Kv.

Fourier Transmission Infrared Spectroscopy Analysis (FTIR)

FTIR was used to characterise the presence of functional groups in NFH and IC. The FTIR analysis was carried out by using an FTIR spectroscopy, Perkin Elmer, Model L1280044. The attenuated total reflectance (ATR) method was used. Four scans were recorded per sample in a frequency range of 700 – 4000 cm^{-1} with a resolution of 0.5 cm^{-1}.

RESULTS AND DISCUSSION

Tensile Properties

The tensile strength of PLA/ABS/NFH and PLA/ABS/IC biocomposites is shown in Figure 1 as a function of filler loading. Apparently, all filler filled PLA/ABS biocomposites showed lower tensile strength compared to the unfilled PLA/ABS blend. These were due to poor interfacial adhesion between polymer matrix and filler due to incompatibility between fibre and matrix [23]. As the filler loading (for both NFH and IC) increased to 30 php, the resultant biocomposites exhibit gradual improvement of tensile strength. This behaviour was mainly due to the increased ability to transfer applied stress from the polymer matrix to the fibre. However, when exceeding 30 phr filler loading, the tensile strength decreased because of reduced in interfacial adhesion between matrix and fibre. Excess loading of filler also resulting in agglomeration of filler, causing stress unable to transfer efficiently across the interface. This incites formation of microcrack and discourages strong interfacial bonding between matrix and fibre. [24] Meanwhile, at similar filler loading, PLA/ABS/IC biocomposite showed higher tensile strength than PLA/ABS/NFH biocomposite. The absence of hemicellulose and lignin in IC contribute the higher tensile strength. Removal of hemicellulose and lignin had attributed to the improvement of molecular orientation and packing of cellulose chains. Orientated cellulose fibre possess greater tensile properties than its counterparts. Since the removal of hemicellulose and lignin, which act as cementing materials in the plant, this enables cellulose fibre to rearrange and conform themselves to the stress direction [25].

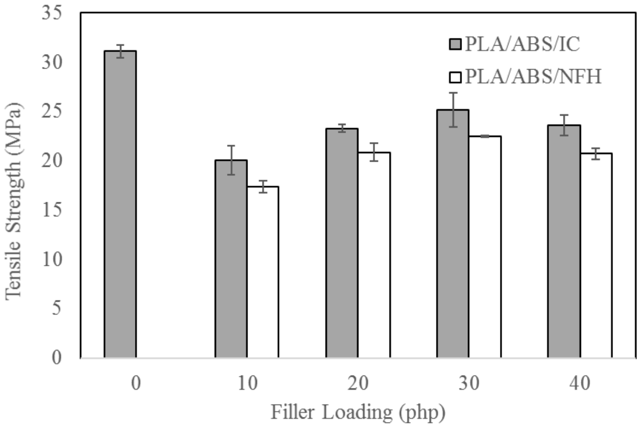


Figure 1. Effect of filler loading on the tensile strength of PLA/ABS blends.

The elongation at break of PLA/ABS/NFH and PLA/ABS/IC biocomposites was illustrated in Figure 2. From the figure, the general trend of elongation at break for biocomposites showed gradual reduction due to the stiffening effect by filler which restricted the polymer chain movement. The elongation at break of PLA/ABS/IC biocomposites were lower than PLA/ABS/NFH biocomposites when comparing at similar loading of filler. This was mainly due to higher interfacial adhesion exists between IC and matrix [26]. IC possesses higher surface chemical interaction such as hydrogen bond comparing to NFH. This chemical interaction form linkage in between hydroxyl group on cellulose with a terminal hydroxyl group and carbonyl group in the ester group of PLA. This, in turn, achieving strong filler-matrix interaction and further reducing the elongation of biocomposites [27].

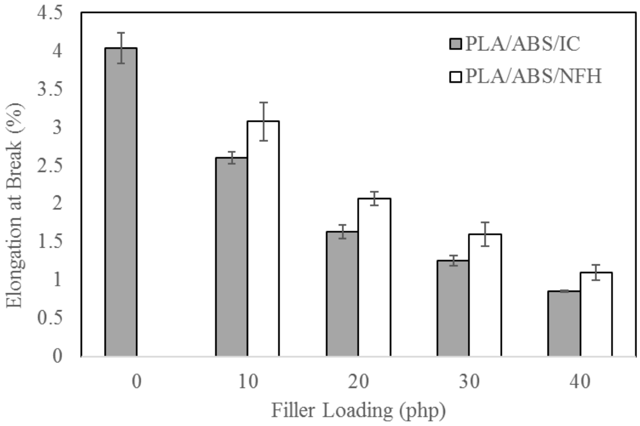


Figure 2. Effect of filler loading on the elongation at break of PLA/ABS blends.

The Young's modulus of PLA/ABS/NFH and PLA/ABS/IC biocomposites is shown in Figure 3. The increment of Young's modulus was a norm in filled polymer system, where filler restricted the mobility of polymer chains and hindered their ability to deform, which stiffen the biocomposites. The Young's modulus of biocomposites influenced by the properties of filler and matrix. In general, cellulosic fibre possessed higher Young's modulus than polymer matrix, which raised Young's modulus of biocomposites. Meanwhile, PLA/ABS/IC biocomposite showed a greater effect on Young's modulus compared to PLA/ABS/NFH biocomposites because of high chemical interaction [25]. As previously discussed, IC capable of forming high hydrogen bonding to PLA matrix and improved interfacial adhesion between the filler and matrix. Furthermore, the absence of hemicellulose and lignin grants molecular orientation and close packing of cellulose chains resulting in high crystallinity structure and high Young's modulus.

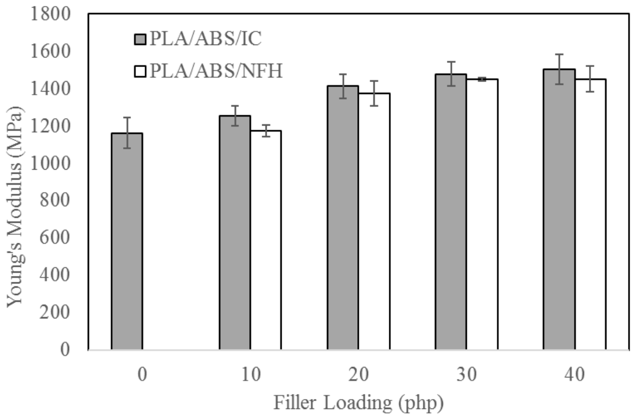


Figure 3. Effect of filler loading on the Young's Modulus of PLA/ABS blends.

Impact Properties

Figure 4 illustrates the impact strength of PLA/ABS/NFH and PLA/ABS/IC biocomposites as a function of filler loading. In general, incorporation of both types of filler in biocomposites decreased the impact strength. Incorporation of high Young's modulus filler like NFH and IC changed the mode of failure in biocomposites from ductile to brittle. Filler restricted polymer chain movement during the fast pace impact test. This resulted in a polymer chain unable to move and accommodate to the deformation, which required less energy to break the biocomposites in a brittle manner. Flaws also existed when inappropriate incorporation and dispersion of filler such as filler agglomeration and void formation. These flaws further reduced the required energy to break the biocomposites as they serve as stress concentration and crack propagation. The reduction in impact strength when loading with fibre was due to incompatibility between filler and polymer matrix. These caused poor interfacial adhesion between them and inefficient to transfer stress in between them.

Meanwhile, the impact strength of PLA/ABS/IC biocomposites showed a lower impact strength as compared to the PLA/ABS/NFH biocomposites. IC possessed a higher Young's modulus than NFH because of the absence of hemicellulose and lignin after the extraction process. The higher Young's modulus fibre further contributed to the brittle failure behaviour and thus required lesser fracture energy.

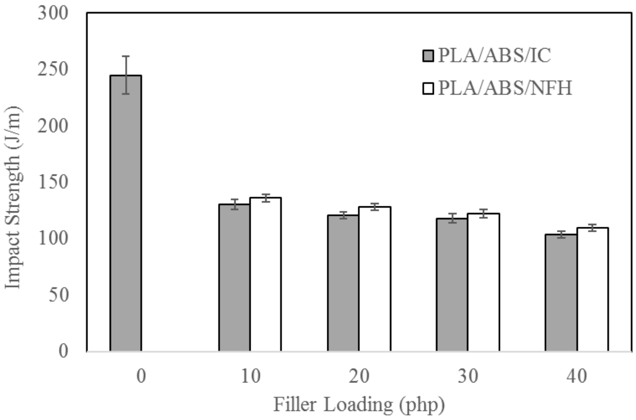


Figure 4. Effect of filler loading on the impact strength of PLA/ABS blends.

Fourier Transform Infrared Spectroscopy Analysis

Figure 5 illustrates the infrared spectrum of NFH and IC where NFH undergoes series of chemical treatment to remove hemicellulose and lignin content to yield IC. In the NFH spectrum, the absorption bands around 3401 cm^{-1} represented as the stretching vibration regarding to –O–H groups. This peak is mainly composed of cellulose, hemicellulose, lignin, and moisture content present in NFH. The peak of 2919 cm^{-1} depicts the asymmetric stretching vibration of CH_2. The following peak of 1738 cm^{-1} represents the carbonyl C=O stretching vibration for acetyl and uranic ester group in hemicellulose or ester linkage of the carboxylic group of ferulic and p-coumaric acids in lignin or hemicellulose. The transmittance peak around 1615-1651 cm^{-1} was verified as OH bending vibration by absorbed water. The detected peak of 1517 cm^{-1} represents the characteristic peak of lignin corresponding to its C=C aromatic skeletal vibration. The 1377 cm^{-1} peak was attributed to bending vibration of C–H and C–O bonds in polysaccharide aromatic rings. The peak at 1254 cm^{-1} is identified as C–O bond, out of plane stretching for aryl group in lignin. The following peak of 1055 cm^{-1} is characterised as the C–O stretch in anhydro glucose chains.

After the isolation of cellulose from NFH, it was found that the peaks at 1738 cm^{-1}, 1517 cm^{-1}, and 1254 cm^{-1} disappeared from IC spectrum, where each peak represents the characteristics of hemicellulose and lignin. The disappearance of these peaks means that hemicellulose and lignin were successfully removed throughout the chemical processes. Besides that, a new peak was observed at 897 cm^{-1} in IC represents as C–H glycosidic deformation, with ring vibration contribution and OH bending. These characteristics identified as the beta-glycosidic bond between anhydro glucose units of cellulose in all of the fibers. This band of the peak is also known as the amorphous band.

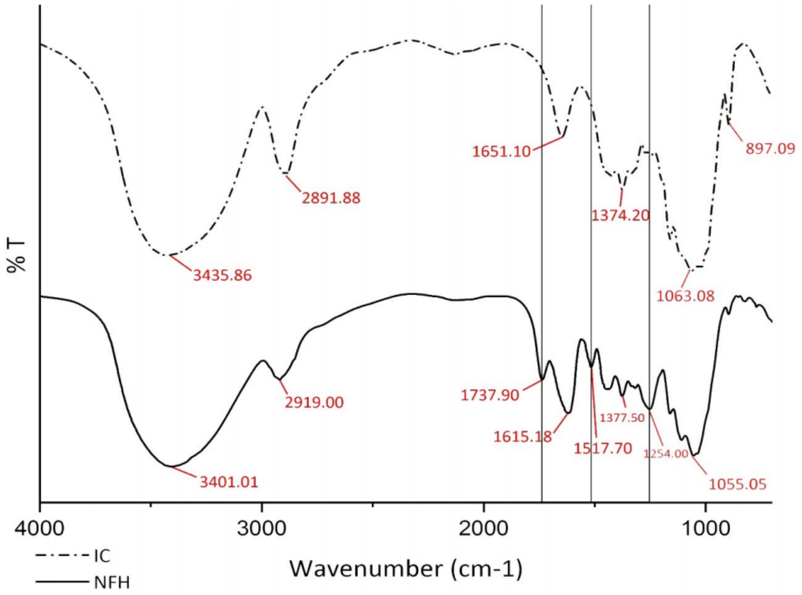


Figure 5. FTIR Spectrum of NFH and IC.

Enzymatic Biodegradation

Figure 6 illustrates the weight loss percentage of PLA/ABS blend, PLA/ABS/NFH biocomposites and PLA/ABS/IC biocomposites at 30 php of filler. The weight loss percentage of all specimens increased with increasing time. This was due to the incorporation of PLA, which biodegraded when exposed to a hydrolytic enzyme such as amylase. As for biocomposites, both PLA/ABS/NFH and PLA/ABS/IC biocomposites exhibited a higher weight loss percentage compared to PLA/ABS blend, respectively. The NFH is composed of cellulose, hemicellulose, and lignin. The presence of the hydrophilic hydroxyl group of NFH that responsible for absorbing more of the amylase solution. The higher absorbance of α-amylase solution facilitated the penetration of α-amylase into PLA/rLDPE/NF biocomposites and subsequently enhanced the amylase attack on the NFH. Thus, incorporation of NFH accelerated biodegradation and disintegration rate of filler filled PLA/ABS biocomposites when subjected to enzymatic hydrolysis. Meanwhile, IC composed of only pure cellulose, that was abundant in the hydroxyl group. It contributes to the higher polarity character in the filler filled PLA/ABS biocomposites. Therefore, PLA/ABS/IC biocomposites possessed the highest weight loss percentage since IC contained a higher number of hydroxyl group compared to NFH [28]. Besides that, alkaline fibre treatment employed in the isolation of cellulose had removed waxy substances in NFH and increased IC filler surface accessibility. Incorporation of IC in PLA/ABS/IC biocomposite contributed greater water uptake from the environment due to the presence of the hydroxyl group. As the IC swelled after water absorption, microcrack was formed around the swollen IC. This microcrack served as a precursor area to leach surrounding water into the matrix. These phenomena accelerated the biodegradation process as compared to PLA/ABS/NFH biocomposite due to the difference in water uptake rate [29].

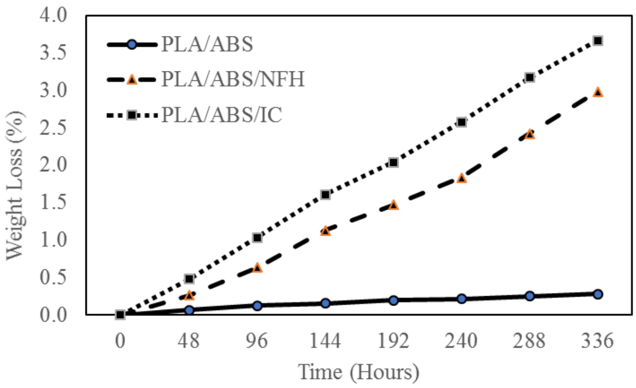
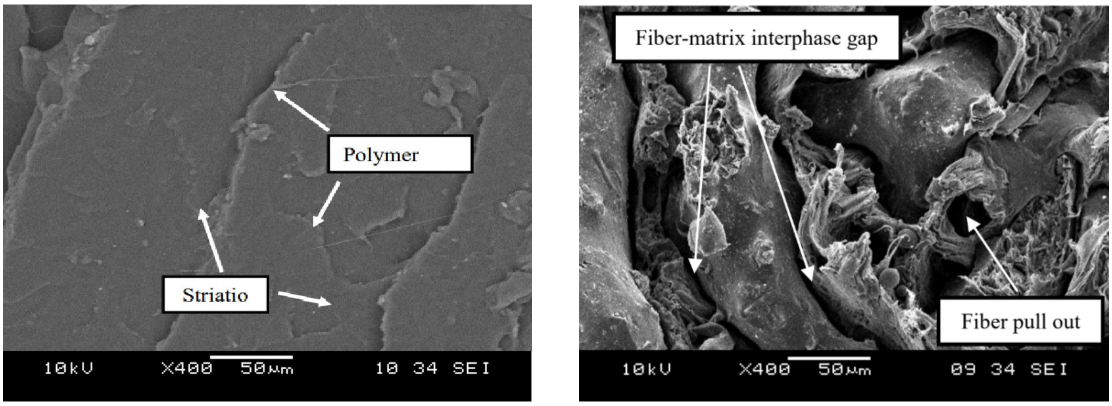


Figure 6. Weight loss percentage of PLA/ABS, PLA/ABS/NFH and PLA/ABS/IC.

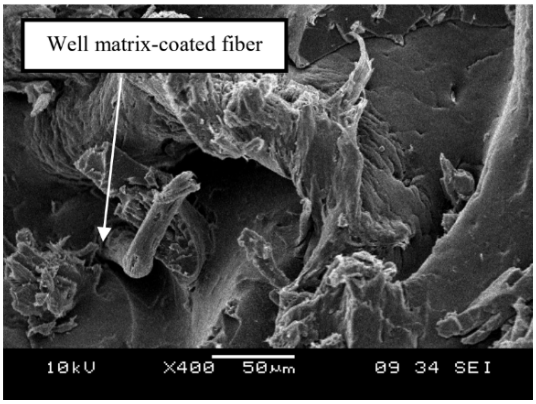
Morphological Study

Figure 7(a) shows the fractured surface of PLA/ABS blend. The fractured surface is presented with striation to the direction of fracture and drawing of polymer fibril. This indicated that the blend was fractured in ductile deformation manner as opposed to pure PLA. The incorporation of ABS into PLA has improved the blend ductility and impact strength as previously proven in tensile and impact test.

Figure 7(b) and 7(c) illustrate the SEM micrographs of PLA/ABS/NFH and PLA/ABS/IC biocomposites at 30 php filler loading, respectively. In Figure 7(b), the surface shows coarsen morphology of the matrix phase and presence of an interphase gap in between NFH and polymer matrix. Numerous of filler pull-outs were also observed on the fractured surface. This showed clear evidence on the poor interfacial interaction in between filler and matrix. However, a well coated of IC by polymer matrix is observed in Figure 7(c) where a smoother surface between IC and polymer matrix improved. Besides that, there was a notable enhancement in polymer matrix morphology where a smoothen topology is displayed on the fractured surface when filled with IC. Furthermore, there were no significant counts of fibre pull-outs presented on the fractured surface in PLA/ABS/IC biocomposite. In comparison between NFH and IC fibre, the removal of hemicellulose and lignin resulted in an improvement of interfacial interaction, which strengthens the mechanical properties of biocomposites, as proven by the micrographs.



(a) PLA/ABS blend (b) PLA/ABS/NFH



(c) PLA/ABS/IC

Figure 7. SEM micrographs of impact fractured surface.

CONCLUSION

Biocomposite from PLA/ABS blend incorporated with two types of fillers, NFH and IC from NFH were formulated by using extrusion compounding methods. IC from NFH was successfully extracted by using Soxhlet extraction method. It was found that the mechanical properties of PLA/ABS incorporated with both fillers to be deteriorated as the filler loading increasing due to the non-reinforcing effect of the fillers which lead to poor interfacial adhesion between the filler and matrix. However, the biodegradation rate of the biocomposites was faster, indicating that the filler facilitated the penetration of the amylase into the PLA/ABS matrix, which enhanced the amylase attack. At similar filler loading, the mechanical properties of IC were found to be higher than NFH because of the removal of hemicellulose and lignin, which contributes to the improvement of molecular orientation and packing of the cellulose chain. Moreover, the biodegradation rate of PLA/ABS/IC was the highest compared to PLA/ABS blend and PLA/ABS/NFH since IC contained a higher number of hydroxyl group compared to NFH.

ACKNOWLEDGEMENT

The author would like to acknowledge the support from the University Malaysia Perlis (UniMAP) for the publication incentive.

REFERENCES

[1] Friné VC, Hector AP, Manuel NDS, Estrella ND, Antonio GJ. Development and characterisation of a biodegradable PLA food packaging hold monoterpene–cyclodextrin complexes against alternaria alternata. Polymers 2019; 11(10): 1720-1735.

[2] Awal A, Rana M, Sain M. Thermorheological and mechanical properties of cellulose reinforced PLA bio-composites. Mechanics of Materials 2015; 80: 87-95.

[3] Ahsan Q, Carron TSS, Mustafa Z. On the use of nano fibrillated kenaf cellulose fiber as reinforcement in polylactic acid biocomposites. Journal of Mechanical Engineering and Sciences 2019; 13(2): 4970-4988.

[4] Kühnel T, Wagner F, Berthold A, Langer B, De Keyzer J. Influence of label impurities in recycling on the mechanical properties of PC/ABS materials. In: Macromolecular Symposia, pp. 1900037-1900044; 2019.

[5] Chaikeaw C, Srikulkit, K. Preparation and properties of poly (lactic acid)/PLA-g-ABS blends. Fibers and Polymers 2018; 19(10), 2016-2022.

[6] Adeniyi A, Agboola O, Sadiku ER, Durowoju MO, Olubambi, PA, Reddy AB, Ibrahim ID, Kupolati WK. Thermoplastic-thermoset nanostructured polymer blends. In: Sabu T, Robert S, Sarathchandran C, editors. Design and applications of nanostructured polymer blends and nanocomposite systems, Oxford: William Andrew Publishing, 2016, p 15-38.

[7] Ahvenainen P, Kontro I, Svedström K. Comparison of sample crystallinity determination methods by X-ray diffraction for challenging cellulose I materials. Cellulose 2016; 23(2): 1073-1086.

[8] Yu L, Dean K, Li L. Polymer blends and composites from renewable resources. Progress in Polymer Science 2006; 31(6): 576-602.

[9] Essabir H, Raji M, Bouhfid R, Qaiss AEK. Nanoclay and natural fibers based hybrid composites: Mechanical, morphological, thermal and rheological properties. In: Jawaid M, Qaiss A, Bouhfid R, editors. Nanoclay reinforced polymer composites, Singapore: Spinger, 2016, p 29-49.

[10] Luo Z, Li P, Cai D, Chen Q, Qin P, Tan T, Cao H. Comparison of performances of corn fiber plastic composites made from different parts of corn stalk. Industrial crops and products. 2017; 95: 521-527.

[11] Mtibe A, Mandlevu Y, Linganiso LZ, Anandjiwala RD. Extraction of cellulose nanowhiskers from flax fibres and their reinforcing effect on poly (furfuryl) alcohol. Journal of Biobased Materials and Bioenergy. 2015; 9(3): 309-317.

[12] Suga, K, Watanab, S, Kuishi T, Imura S, Ishigaki K, Yokota M, Yanagawa S, Suyama Y. Extremely low genetic diversity of the northern limit populations of Nypa fruticans (Arecaceae) on Iriomote Island, Japan. Conservation genetics 2016; 17(1): 221-228.

[13] Robertson AI, Dixon P, Daniel PA, Zagorskis I. Primary production in forests of the mangrove palm Nypa fruticans. Aquatic Botany 2020; 167: 103288-103298.

[14] Rasidi MS, Husseinsyah S, Leng TP. The effect of filler content and ultraplast TP01 of Nypa fruticans fiber-filled polylactic acid/recycled low density polyethylene biocomposites on tensile properties and morphology. Polymer-Plastics Technology and Engineering 2015; 54 (3): 253-258.

[15] Rasidi MSM, Husseinsyah S, Leng TP. Chemical modification of Nypa fruticans filled polylactic acid/recycled low-density polyethylene biocomposites. BioResources. 2014; 9(2): 2033-2050.

[16] Syahmie Rasidi M, Salmah H, Leng TP, Ismail H. Mechanical and morphological properties of polylactic acid/recycled low density polyethylene/nypa fruticans biocomposites compatibilised with polyetylene-co-acrylic acid. Applied Mechanics and Materials 2015; 754: 54-58.

[17] Rasidi MS, Salmah H, Teh PL. Properties of silanised nypa fruticans filled polylactic acid/recycled low density polyethylene biocomposites. Polymer Engineering & Science. 2015; 55(8): 1733-1740.

[18] Tamunaidu P, Saka S. Chemical characterisation of various parts of nipa palm (Nypa fruticans). Industrial Crops and Products. 2011; 34(3): 1423-1428.

[19] Fauzee SN, Othaman R. Extraction and dissolution of cellulose from Nypa fruit husk for nanofibers fabrication. In AIP Conference Proceedings, American Institute of Physics. 1571 (1), pp. 904-910; 2013.

[20] ASTM D638-14, Standard test method for tensile properties of plastics, ASTM International, West Conshohocken, PA, 2014, www.astm.org

[21] ASTM D256-10e1, Standard test methods for determining the izod pendulum impact resistance of plastics, ASTM International, West Conshohocken, PA, 2010, www.astm.org

[22] Yoon BS, Suh MH, Cheong SH, Yie JE, Yoon SH, Lee, SH. Studies on the degradable polyethylene: Use of coated photo degradants with biopolymers. Journal of Applied Polymer Science 1996; 60(10): 1677-1685.

[23] Syahmie, MM, Leng TP, Najwa ZN. Effect of filler content and chemical modification on mechanical properties of polylactic acid/polymethyl methacrylate/nypa fruticans husk biocomposites. Management Science and Engineering 2018; 318(1): 012011-012017.

[24] Dinesh S, Kumaran P, Mohanamurugan, S, Vijay R, Singaravelu DL, Vinod A, Bhat KS. Influence of wood dust fillers on the mechanical, thermal, water absorption and biodegradation characteristics of jute fiber epoxy composites. Journal of Polymer Research 2020;27(1): 9.

[25] Majhi SK, Nayak SK, Mohanty S, Unnikrishnan L. Mechanical and fracture behavior of banana fiber reinforced Polylactic acid biocomposites. International Journal of Plastics Technology 2010; 14(1): 57-75.

[26] Tawakkal ISM, Talib RA, Abdan K, Ling CN. Mechanical and physical properties of kenaf-derived cellulose (KDC)-filled polylactic acid (PLA) composites. BioResources 2012; 7(2): 1643-1655.

[27] Haroosh HJ, Dong Y. Electrospun poly (lactic acid) (PLA): poly (ε-caprolactone) (PCL)/halloysite nanotube (HNT) composite fibers: synthesis and characterisation. In Yu D, Rehan U, Alan Kin-Tak L, editors. Fillers and reinforcements for advanced nanocomposites, Cambridge: Woodhead Publishing, p 59-80.

[28] Tanjung FA, Arifin Y, Husseinsyah S. Enzymatic degradation of coconut shell powder–reinforced polylactic acid biocomposites. Journal of Thermoplastic Composite Materials 2020; 33(6): 800-816.

[29] Agung EH, Hamdan MHM, Siregar JP, Bachtiar D, Tezara C, Jamiluddin, J. Water absorption behaviour and mechanical performance of pineapple leaf fibre reinforced polylactic acid composites. International Journal of Automotive and Mechanical Engineering 2018; 15(4): 5760-5774.