J MATER RES TECHNOL. 2019;8(5):3988–3994

Available online at www.sciencedirect.com

jmr&t
Journal of Materials Research and Technology
www.jmrt.com.br

Original Article

Water absorption, thickness swelling and thermal properties of roselle/sugar palm fibre reinforced thermoplastic polyurethane hybrid composites

A.M. Radzi[a,d], S.M. Sapuan[a,b,*], M. Jawaid[a], M.R. Mansor[c]

[a] Laboratory of Biocomposite Technology, Institute of Tropical Forestry and Forest Products (INTROP), Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia
[b] Advance Engineering Materials and Composites Research Centre, Department of Mechanical and Manufacturing Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia
[c] Faculty of Mechanical Engineering, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia
[d] Faculty of Engineering and Technology, Linton University College, Persiaran Utl, Kampung Gebok Batu 12, 71700 Mantin, Negeri Sembilan, Malaysia

ARTICLE INFO

Article history:
Received 19 November 2018
Accepted 5 July 2019
Available online 31 July 2019

Keywords:
Roselle fibre
Sugar palm fibre
Hybrid composites
Thermoplastic polyurethane

ABSTRACT

The aim of this work is to investigate the effect of sugar palm fibre (SPF) loading on the water absorption, thickness of swelling and thermal properties of roselle (RF)/SPF/thermoplastic polyurethane (TPU) hybrid composites using thermogravimetric analysis (TGA). The hybridised versions of RF/SPF were prepared at different weight rations through melt mixing and hot compression at 170 °C. Water absorption and thickness of swelling properties were investigated using the water immersion time test. The thermal properties of the hybrid composites were also analysed. The water absorption and thickness of swelling results revealed that an increase in SPF content led to a decrease in water uptake and thickness of swelling of the RF/SPF hybrid composites. The lowest water absorption (7.35%) and thickness of swelling (7.15%) data were obtained from the RST-3 hybrid composite. The TGA also showed that hybrid composites with increased SPF content recorded improved thermal stability.

1. Introduction

In this decade, energy conservation and cost material reduction are key points of various manufacturing sectors such as automotive and construction. Therefore, many researchers and manufacturing industries have shifted towards eco-friendly practices and at the same time, reduce their materials' cost and preserve the environment. The use of natural cellulose fibres is very suitable and in high demand for several applications. The demand for natural fibres as new material for the production of composites has increased as several types of natural fibres have good mechanical and physical

* Corresponding author.
E-mail: sapuan@upm.edu.my (S. Sapuan).
https://doi.org/10.1016/j.jmrt.2019.07.007
2238-7854/

properties and therefore, can compete with the fibres currently used in the market [1].

Natural fibre-reinforced polymer matrix is widely used in many applications such as transport medical implants, furniture fittings, paper industry, mining, and the textile industries. It reduces production costs, is renewable, has low density and improved mechanical properties and characteristics comparable to other constituent materials. In fact, many researchers nowadays tend to choose composites [2,3].

Natural fibre can easily be found, is non-abrasive to equipment, and requires less energy for processing. The geographical distribution of natural fibres is focused mainly in South Asia such as Malaysia, Thailand, Myanmar and Indonesia. There are a variety of fibres that can easily be obtained in Malaysia to be used as reinforcement. One of these fibres is the RF. The RF is reported to have properties closely related to jute fibre. The roselle plant is classified in the hibiscus group and its scientific name is *Hibiscus Sabdariffa* L. [4]. The advantages of the roselle plant is not only in its fruit, but also its bust fibre that can be processed into rope, jute and textiles [5]. In addition, the RF has good tensile, good durability and good resistance to deterioration by sea water. It also gives a higher impact when incorporated in polymer composites.

The SPF is derived from the *Ijuk* or known as the *Arenga pinnata (Wurmb) Merr*. In addition of fibre, the tree can also produce traditional sugar blocks more commonly known as *Enau* or Gula *Kabung* which are used as food sweeteners. The SPF can be used to produce home products such as mats, brooms and ropes [6,7].

The TPU has good physical and chemical characteristics, mechanical properties, and resistance to oil, grease and abrasion; these properties enable it to be used for automotive parts, furniture and medical equipment [8]. Many previous studies have reported on the advantages of combining TPU composites with other various fibres to be used as reinforcement such as synthetic fibre, carbon fibre, kenaf fibre and curaua fibre [9].

Hybridisation is a process of combining two fibres to strengthen its matrix in order to enhance the mechanical and physical properties of the composite. Today, many researches on hybrid composites (combination of two types of natural fibres) have been conducted with the aim of reducing the cost of materials and at the same time, increase the use of lignocelluloses fibre [10,11,12]. In fact, hybrid compositing is an approach that promises an increase in the physical properties of a material [13]. The aim of this paper is therefore to study the effect of an increase in SPF content on the density, water absorption, thickness of swelling and thermal properties of hybrid composites.

2. Materials and methodology

2.1. Materials

The RF was obtained from a roselle plantation area in Mersing, Johor, Malaysia. The RF samples were extracted from the roselle plant. It was necessary to start the procedure with a water retting process for 14 days by soaking the roselle stems in water. The retted stems were cleaned with tap water and the fibres were pulled out manually to separate the fibres from the stems. The SPF was obtained from Kuala Jempul, Negeri Sembilan, Malaysia. The SPF samples were extracted from sugar palm trees. It was necessary to first wash the samples with tap water to remove impurities. Both fibres were dried under sunlight for 24 h after all cleaning processes were completed. The RF and SPF fibres were crushed and sieved to a fibre length size range of 300–425 μm. This measurement remained fixed throughout the study [8]. The polymer matrix used was TPU with a density of 1.10 g/cm^3. TPU was supplied by Innovative Pultrusion Sdn. Bhd, Seremban, Negeri Sembilan, Malaysia.

Table 1 – Composition of the materials.

RF (%)	SPF (%)	Designation
75	25	RST-1
50	50	RST-2
25	75	RST-3

2.2. Sample preparation

The RF/SPF hybrid composites were prepared according to procedures reported in the authors' previous work [14]. The hybrid composites were prepared using a Brabender Plastograph internal mixer at 170 °C and 200 MPa. The hybrid composites were produced in the form of sheets using a hot press at 170 °C and then cooled for 5 min. The ratio of TPU was maintained at 60 wt% with a designation for single RF at 40 wt% (RF-T) and SPF at 40 wt% (SPF-T). Table 1 shows the composition of materials.

2.3. Density

The density of the hybrid composites was determined using an XS205 Mettler Toledo balance. Five samples were prepared for this test for each ratio of hybrid composite.

2.4. Water absorption

Five samples sized $10 \times 10 \times 3\,\text{mm}^3$ were prepared for the water absorption evaluation. The samples were measured as W_0 and W_1 for before and after being immersed in water for 24 h at room temperature [5]. Water absorption was determined using Eq. (1).

$$\textit{Water Absorption}, \% = \frac{W_1 - W_0}{W_0} \times 100 \quad (1)$$

where,

W_0 = The weight of sample before immersion.

W_1 = The weight of sample after 7 days of immersion in water.

2.5. Thickness swelling

Five samples with size $10 \times 10 \times 3\,\text{mm}^3$ were prepared for the thickness swelling evaluation. The samples were measured as T_0 before and T_1 after being immersed into water using a

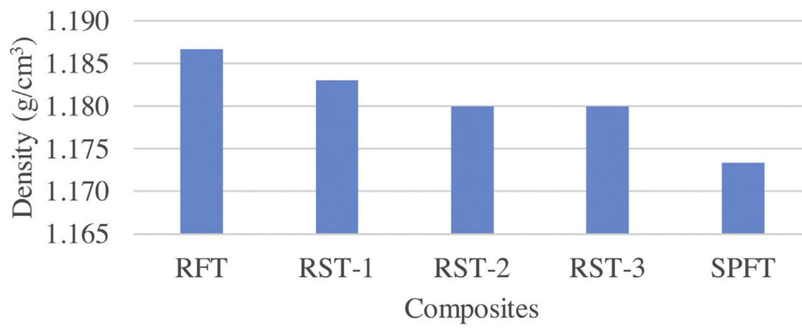

Fig. 1 – Density of RF/SPF hybrid composites.

Vernier calliper (Mitutoyo) [15]. Thickness swelling was determined using Eq. (2).

$$Thickness\,Swelling,\,\% = \frac{T_1 - T_0}{T_0} \times 100 \quad (2)$$

where,

T_0 = The thickness of sample before immersion.

T_1 = The thickness of sample after 7 days of immersion in water.

2.6. Thermogravimetric analysis (TGA)

The thermal properties of hybrid composites are practically significant in studies related to temperature as it is important to determine fibres' level of degradation after alkaline treatment. A thermogravimetric analysis (TGA) was performed using a Q-series thermal analysis machine model TA Instrument (TGA Q500). The analysis was done at a temperature range between 30 to 600°c with a heating rate of 10 °C/min.

3. Result and discussions

3.1. Density

Fig. 1 shows the density of the RF/SPF hybrid composites. Physical characteristics such as density and porosity are important elements in determining the tensile and flexural strength and mechanical properties of composites [15]. The results showed that density slightly decreased with an increase in the SPF content of hybrid composites. Hybrid composites RST-3 shows the lowest density value compared to other hybrid composites. For single SPFT composites it also shows lowest density value compared to single RFT composites. The highest density value for hybrid composites was recorded by RST-1 (1.183 g/cm^3) followed by RST-2 (1.18 g/cm^3). The reductions of this density value maybe due to the low density of SPF compared to RF. In addition, influence from the manufacturing processes such as heat and temperature are also important because it can affect and could be a reason behind the decrease of mechanical and physical properties due to the production of high voids due to air trap on the composites and incorporating two types of fibres into the matrix in the composites [17–19]. The impact of these composite density reductions has also been reported in previous work [20]. Jumaidin et al. [15] conducted a study on the effects of seaweed/SPF hybrid composites on the physical properties where increased the SPF content was shown to decrease the density of composites and Ramanaiah et al. [20] study on the effect of Borassus seed shoot fibre reinforced polyester composites found that an increase in the fibre content of polyester composites resulted in a decrease in the density value of the composites.

3.2. Water absorption

Hybrid composites based on natural fibre sources are sensitive to water (hydrophilic). Therefore, water absorption characteristics are important and thus prepared for in this study. This test was conducted on the RF/SPF hybrid composites as it is one of the important physical properties to be assessed in order to determine the amount of the water absorbed at certain conditions and times. Physical properties in terms of moisture diffusion depend on some factors such as void/pore, humidity, temperature, volume fraction of fibre and viscosity of matrix. Fig. 2 shows the water absorption levels for the RF/SPF hybrid composites. The highest water uptake value of hybrid composites after 168 h was 8.53% (RST-1), followed by 7.48% (RST-2) and 7.35% (RST-3). From the observation, water uptake was faster in the early stages and became slower when the immersion time increased. The capacity of water absorption and swelling thickness is directly related to the density, the presence of voids and the bond between the fibre and the matrix [13]. This factor causes water to be trapped in the void and increase composite weight. All composite shows decrease with increasing the SPF content after 7 days immersed in distilled water. All composites showed a decrease with increasing SPF content after 7 days immersed in distilled water. In addition, the combination of SPF with RF indicated a decreased in water absorption of all composition of hybrid composites. The lowest water absorption level was recorded by a single SPF composite, while the highest was for single RF composites. Therefore, the combination of SPF and natural fibre composites becomes one of the factors behind the decline as it can decrease the water uptake of composite materials. SPF is used in composites to reduce water uptake as it has good water resistance. Jumaidin el al. [15], Ishak et al. [21] and Sahari et al. [22] with the use of SPF it can reduce the water uptake on composites. In addition, according the Jumaidin et al. [15] and Remirez et al. [23], lower water absorption factors of natural fibres materials can be attributed such as interfaces adhesion

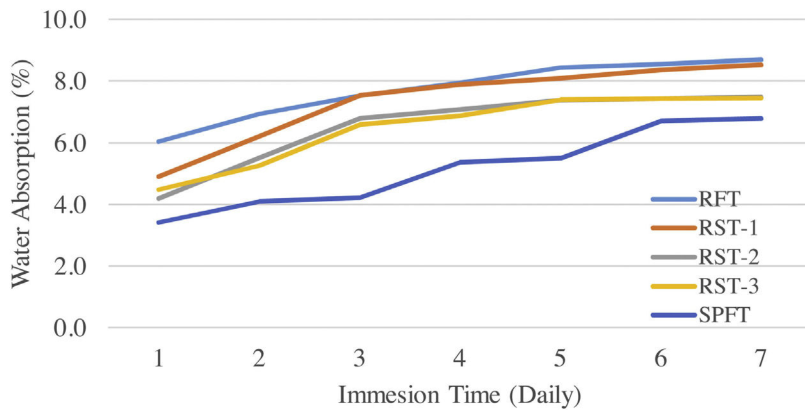

Fig. 2 – Water absorption of RF/SPF hybrid composites.

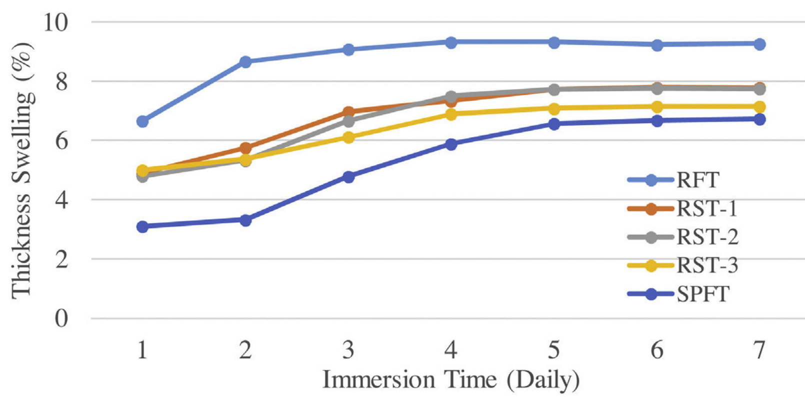

Fig. 3 – Thickness swelling of RF/SPF hybrid composites.

between fibres /matrix, greater water affinity of the polymer matrix compared natural fibres, prevents water absorption by matrices produced by fibre and reduce void content in the composites.

3.3. *Thickness swelling*

A thickness of swelling test was conducted on the RF/SPF hybrid composites to analyse the changes of dimensional stability of the hybrid composites. Fig. 3 shows the thickness of swelling of the hybrid composites. The results showed a similar trend with water absorption where the thickness of swelling of RF/SPF hybrid composites had increased following the 7 days of immersion in distilled water. The thickness of swelling was more stable and can be seen after 4 days of immersion. The highest value for thickness of swelling for the hybrid composites after 168 h was 7.78% (RST-1) followed by 7.75% (RST-2) and 7.15% (RST-3). From the result, the thickness swelling is faster in the early stages and become slower when the immersion time increased and all composite shows decrease with increasing the SPF content after 7 days immersed in distilled water. Overall, the increase of SPF in RF composites showed a decline for thickness of swelling in all hybrid composites compared to values recorded for single RF composites. The effect of thickness of swelling was noticeable after two days of immersion and became stable after 4 days of soaking in water. This phenomenon can be said to be similar to water uptake behaviour where the presence of SPF with less hydrophilic characteristics compared the RF reduced the water absorption in composites, thereby providing better water resistance and a reduction in the thickness of swelling when immersed in water. This findings agreed with the reported by Edihirej et al. [24] and Sahari et al. [25], the contribution of SPF with relation the water absorption and thickness swelling behaviour. In addition, the poor interaction between fibre/matrix and distribution of fibres affects thickness swelling and dimensional stability composite [26,27]. With good interface and distribution between RF and SPF it can limit the swelling and percentage of dimensional stability of composites.

3.4. *Thermal properties of hybrid composites*

3.4.1. *Thermogravimetric analysis (TGA)*

RF and SPF are natural fibres that are hybridised and reinforced with TPU composites. These fibres should be examined for durability from heat resistance and thermal stability to ensure that both fibres can withstand certain levels of temperature during service. TGA analysis was performed to investigate the decomposition and thermal stability of the RF/SPF hybrid composites. The results are presented in thermogravimetric and derivative thermogravimetric forms. Figs. 4 and 5 demonstrate the TGA and Differential Thermogravimetric (DTG) results for the single fibre and hybrid composites. The overall trend

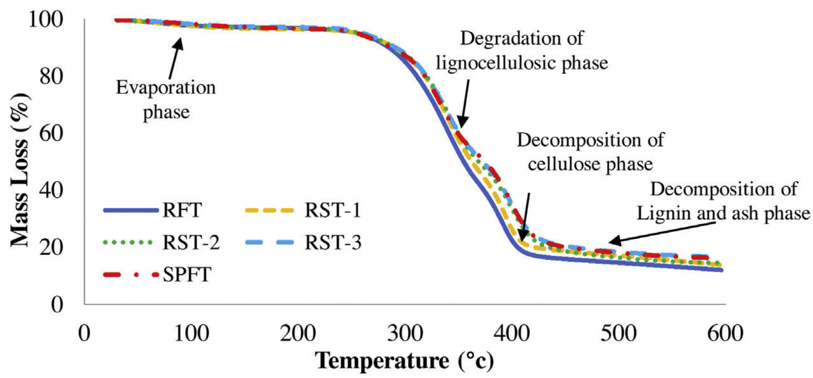

Fig. 4 – TGA of RF/SPF hybrid composites.

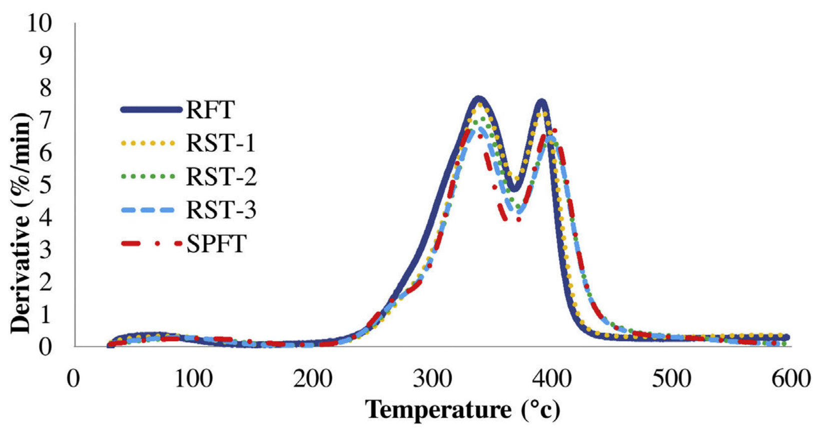

Fig. 5 – DTG of RF/SPF hybrid composites.

Table 2 – Thermal degradation analysis of RF/SPF hybrid composites.

Sample	First degradation		Second degradation		Char at 600 °C (%)	Ref
	T_{on}	T_{max}	T_{on}	T_{max}		
RFT	300.24	340.06	384.7	398.56	9.51	[8]
RFT-1	299.82	339.71	381.13	391.13	13.75	Current study
RFT-2	299.09	340.19	385.95	398.36	14.54	
RFT-3	296.31	337.72	385.32	398.17	16.67	
SPFT	296.5	333.85	383.73	398.01	16.23	

showed similar trends of degradation. Observation of thermal degradation for the single fibre and hybrid composite samples occurred within a set temperature range of 30 °C to 600 °C. Generally, the thermal decomposition of these composites is divided into four core phases. The initial phase is evaporation of moisture at 30–100 °C, followed by decomposition of lignocellulosic components at 220–315 °C, cellulose components at 315–400 °C, lignin at 165–900 °C, and ash at 1723 °C [28–31]. Edhrej et al. [24] and Yang et al. [31] have reported that hemicellulose decomposes when the temperature reaches 220 °C and 315 °C the decomposition has been completed.

As shown in Fig. 4, in the initial stage (evaporation of moisture) all composites showed similar moisture trends and peak degradation temperatures. In the second phase, the degradation of lignocellulosic occurred at a temperature range between 332–340 °C. The third phase which is the decomposition of cellulose occurred at a temperature range between 391–399 °C. After all the hemicellulose had decomposed, the decomposition of cellulose then occurred as the third phase. The decomposition of lignin, cellulose and ash occurred in this phase [32–34]. The temperature for decomposition rate is determined by the maximum decomposition rate represented by the peak of the curve. Table 2 shows the result of thermal degradation analysis of RF/SPF hybrid composites. From the result, the percentage of residue char recorded by RFT-3 is the highest was 16.67% and the lowest residue char percentage is RFT was 9.51%.

4. Conclusions

In the present study, hybrid composites from natural fibres (RF and SPF) reinforced with TPU composites were successfully produced using pre-mix and compression moulding processes. The addition of SPF affects the density, water absorption, thickness swelling and thermal properties of RF/SPF hybrid composites. The water absorption, thickness of swelling and thermal properties showed increases as the SPF content reduced the water uptake and thickness of swelling. The TGA conducted also showed better thermal stability. Density was proven to be reduced with the increase in SPF content.

Conflict of interest

The authors declare no conflicts of interest.

REFERENCES

[1] Sathishkumar TP, Navaneethakrishnan P, Shankar S, Rajasekar R. Mechanical properties and water absorption of short snake grass fiber reinforced isophthallic polyester composites. Fibers Polym 2014;15(9):1927–34.

[2] Venkateshwaran N, ElayaPerumal A, Alavudeen A, Thiruchitrambalam M. Mechanical and water absorption behaviour of banana/sisal reinforced hybrid composites. Mater Des 2011;32((7)August):4017–21.

[3] Sapuan SM, Pua F, El-Shekeil YA, AL-Oqla FM. Mechanical properties of soil buried kenaf fibre reinforced thermoplastic polyurethane composites. Mater Des 2013;50(September):467–70.

[4] Nadlene R, Sapuan SM, Jawaid M, Ishak MR, Yusriah L. A review on roselle fiber and its composites. J Nat Fibers 2016;13(1):10–41.

[5] Razali N, Salit MS, Jawaid M, Ishak MR, Lazim Y. A study on chemical composition, physical, tensile, morphological, and thermal properties of roselle fibre: effect of fibre maturity. BioResources 2015;10(1):1803–23.

[6] Ilyas RA, Sapuan SM, Ibrahim R, Abral H, Ishak MR, Zainudin ES, et al. Sugar palm (Arenga pinnata (Wurmb.) Merr) cellulosic fibre hierarchy: A comprehensive approach from macro to nano scale. J Mater Res Technol 2019;8:2753–66, http://dx.doi.org/10.1016/j.jmrt.2019.04.011.

[7] Huzaifah MRM, Sapuan SM, Leman Z, Ishak MR, Maleque MA. A review of sugar palm (*Arenga pinnata*): application, fibre characterisation and composites. Multidiscip Model Mater Struct 2017;13(November (4)):678–98.

[8] Radzi AM, Sapuan SM, Jawaid M, Mansor MR. Mechanical and thermal performances of roselle fiber-reinforced thermoplastic polyurethane composites. Polym Plast Technol Eng 2018;57(May (7)):601–8.

[9] Radzi AM, Sapuan SM, Jawaid M, Mansor MR. Influence of fibre contents on mechanical and thermal properties of roselle fibre reinforced polyurethane composites. Fibers Polym 2017;18(July (7)):1353–8.

[10] Wan Busu WN, Anuar H, Ahmad SH, Rasid R, Jamal NA. The mechanical and physical properties of thermoplastic natural rubber hybrid composites reinforced with Hibiscus cannabinus, L and short glass Fiber. Polym Plast Technol Eng 2010;49(September (13)):1315–22.

[11] Thiruchitrambalam M, Alavudeen A, Venkateshwaran N. Review on kenaf fiber composites. Rev Adv Mater Sci 2012;32(2):106–12.

[12] Idicula M, Joseph K, Thomas S. Mechanical performance of short Banana/Sisal hybrid Fiber reinforced polyester composites. J Reinf Plast Compos 2009;29(1):12–29.

[13] Nunna S, Chandra PR, Shrivastava S, Jalan A. A review on mechanical behavior of natural fiber based hybrid composites. J Reinf Plast Compos 2012;31(11):759–69.

[14] Radzi AM, Sapuan SM, Jawaid M, Mansor MR. Mechanical performance of Roselle/Sugar palm fiber hybrid reinforced polyurethane composites. BioResources 2018;13(3):6238–49.

[15] Jumaidin R, Sapuan SM, Jawaid M, Ishak MR, Sahari J. Thermal, mechanical, and physical properties of seaweed/sugar palm fibre reinforced thermoplastic sugar palm Starch/Agar hybrid composites. Int J Biol Macromol 2017;97(April):606–15.

[16] Akhtar MN, Sulong AB, Radzi MKF, Ismail NF, Raza MR, Muhamad N, et al. Influence of alkaline treatment and fiber loading on the physical and mechanical properties of kenaf/polypropylene composites for variety of applications. Prog Nat Sci Mater Int 2016;26:657–64.

[17] Asim M, Paridah MT, Saba N, Jawaid M, Alothman OY, Nasir M, et al. Thermal, physical properties and flammability of silane treated kenaf/pineapple leaf fibres phenolic hybrid composites. Compos Struct 2018;202:1330–8.

[18] Jawaid M, Abdul Khalil HPS, Noorunnisa Khanam P, Abu Bakar A. Hybrid composites made from oil palm empty fruit Bunches/Jute fibres: water absorption, thickness swelling and density behaviours. J Polym Environ 2011;19(March (1)):106–9.

[19] Alamri H, Low IM. Effect of water absorption on the mechanical properties of nanoclay filled recycled cellulose fibre reinforced epoxy hybrid nanocomposites. Compos Part A Appl Sci Manuf 2013;44(January (1)):23–31.

[20] Ramanaiah K, Ratna Prasad AV, Hema Chandra Reddy K. Effect of fiber loading on mechanical properties of borassus seed shoot fiber reinforced polyester composites. J Mater Environ Sci 2012;3(2):374–8.

[21] Ishak MR, Sapuan SM, Leman Z, Rahman MZA, Anwar UMK, Siregar JP. Sugar palm (*Arenga pinnata*): its fibres, polymers and composites. Carbohydr Polym 2013;91(2):699–710.

[22] Sahari J, Sapuan SM, Zainudin ES, Maleque MA. Mechanical and thermal properties of environmentally friendly composites derived from sugar palm tree. Mater Des 2013;49:285–9.

[23] Lomelí Ramírez MG, Satyanarayana KG, Iwakiri S, De Muniz GB, Tanobe V, Flores-Sahagun TS. Study of the properties of biocomposites. Part I. Cassava starch-green coir fibers from Brazil. Carbohydr Polym 2011;86(4):1712–22.

[24] Edhirej A, Sapuan SM, Jawaid M, Zahari NI. Cassava/sugar palm fiber reinforced cassava starch hybrid composites: physical, thermal and structural properties. Int J Biol Macromol 2017;101:75–83.

[25] Sahari J, Sapuan SM, Ismarrubie ZN, Rahman MZA. Comparative study of physical properties based on different parts of sugar palm fibre reinforced unsaturated polyester composites. Key Eng Mater 2011;471–472(February):455–60.

[26] Kaymakci A, Gulec T, Hosseinihashemi SK, Ayrilmis N. Physical, mechanical and thermal properties of wood/zeolite/plastic hybrid composites. Maderas Cienc y Tecnol 2017;19(ahead):339–48.

[27] Ashori A, Sheshmani S. Hybrid composites made from recycled materials: moisture absorption and thickness swelling behavior. Bioresour Technol 2010;101(12):4717–20.

[28] Ilyas RA, Sapuan SM, Ishak MR. Isolation and characterization of nanocrystalline cellulose from sugar palm fibres (*Arenga Pinnata*). Carbohydr Polym 2018;181(February):1038–51.

[29] El-Shekeil YA, Sapuan SM, Jawaid M, Al-Shuja'a OM. Influence of fiber content on mechanical, morphological and thermal properties of kenaf fibers reinforced poly(vinyl chloride)/thermoplastic polyurethane poly-blend composites. Mater Des 2014;58(June):130–5.

[30] Boopalan M, Niranjanaa M, Umapathy MJ. Study on the mechanical properties and thermal properties of jute and banana fiber reinforced epoxy hybrid composites. Compos Part B Eng 2013;51:54–7.

[31] Yang H, Yan R, Chen H, Lee DH, Zheng C. Characteristics of hemicellulose, cellulose and lignin pyrolysis. Fuel 2007;86(August (12–13)):1781–8.

[32] Elkhaoulani A, Arrakhiz FZ, Benmoussa K, Bouhfid R, Qaiss A. Mechanical and thermal properties of polymer composite based on natural fibers: moroccan hemp fibers/polypropylene. Mater Des 2013;49(August):203–8.
[33] Bakare IO, Okieimen FE, Pavithran C, Abdul Khalil HPS, Brahmakumar M. Mechanical and thermal properties of sisal fiber-reinforced rubber seed oil-based polyurethane composites. Mater Des 2010;31(9):4274–80.
[34] Arrakhiz FZ, Benmoussa K, Bouhfid R, Qaiss A. Pine cone fiber/clay hybrid composite: mechanical and thermal properties. Mater Des 2013;50(September):376–81.