Effect of Natural Fiber (NF) Mix on Mechanical Strength of NF Plastic Composites (NFPC)

Dzaraini Kamarun, Engku Zaharah Engku Zawawi, Nur Hikamah Seth, Norlaily Ahmad, Siti Rafedah Abdul Karim

Abstract*: Natural fibers from plants are gaining importance and may substitute wood in the production of wood plastic composites (WPC). To ensure continuity of fiber supply and sustainability of WPC industries, fibers of various types could be mixed together to obtain Mix WPC. However, research need to be carried out to identify the contribution of different fiber type collectively to the mechanical properties of Mix natural fiber polymer composite (NFPC). In this study, preliminary work on the use of natural fibre (NF) such as kenaf, sugar palm and pineapple leaf fibers in the preparation of Mix NFPC were carried out. Four different fiber mix samples with different fiber ratio and size were formulated using polypropylene (PP) as the polymer matrix. Montmorrilonite (MMT) filler was added at constant amount for enhancement of composite mechanical properties. Samples were mixed and prepared using a twin screw extruder and mini injection moulding resepectively. Individual fibers and NFPC prepared were characterized using thermogravimetric analyzer (TGA). Tensile, flexural and impact strength of the composites were determined. Generally, it was found that addition of fiber mix at 50% fiber loading enhance the tensile and flexural strength of the various NFPC with minimal exceptions. The impact strength of the composites were comparable to that of blank PP implying that addition of fiber gives additional advantage besides being eco-friendly. It was also found that higher kenaf loading and different size of fiber mix contribute positively to the various strengths measured. In addition to that, composition of individual fibers also contribute to the mechanical properties of the NFPCs.

Keywords: Composites thermal properties, Mechanical strength, Natural fibers.

I. INTRODUCTION

In many tropical countries, agricultural waste such as palm oil empty fruit bunch (POEFB) and pineapple leaf (PAL) can be conveniently converted to fibers for consumption in various downstream industries such as wood plastic composites (WPC) and handcraft products. Natural fibers from trunk of oil palm [1, 2] and sugar palm trees [3, 4] are among other source of fibers which have potential use in WPC. Other than that, short harvest plants such as hemp, kenaf, jute, flax, sugarcane bagasse and luffa could also serve as alternatives to wood fibers [5, 6]

Due to limited supply and issues related to sustainability,WPC industries need to be non-choosy in using wood or non-wood materials as the fiber in producing composites. Wood fibers in WPC are limited to sawdust, wood powder and wood flour from timber industry waste that contains contaminants such as adhesives, resins and coatings [7]. NF from agriculture waste such POEFB consists of substantial amount of oil (~ 7.4 %) and are not suitable for consumption in WPC [8]. Alternative source of NF which are more economic and sustainable are plant fibers and fibers from tree trunks. To name a few, short harvest plants such as kenaf, jute, flax, sugarcane bagasse and luffa are some of these plant fibers available as local crops in many tropical countries. More economical would be to mix fibers of various types and sizes to ensure availability and continuity of supply. Furthermore, hybridization of one natural fibre with other natural fibre provide further advantage of reduction in the water uptake capacity compared with individual natural fibre reinforcement [9].

This paper discusses some preliminary study that were carried out in the preparation of NFPC using mix fibers of kenaf, pineapple leaf (PAL) and sugar palm (SP). All these fibers are available locally in Malaysian plantations. The purpose of using these fibers are their availability and possibility of diversifying their application in industry.

II. METHODOLOGY

A. Materials and Chemicals

Injection molding grade polypropylene (Manufacturer: Etilinas; Grade: MH03), Kenaf fibers (Lembaga Kenaf Malaysia) ; sugar palm fibers and pineapple leaf fiber were obtained as waste from a local plantation in Jempol and a pineapple plantation at MARDI Klang, Selangor, respectively. Polypropylene-graft-maleic anhydride (MAPP) were supplied by Aldrich and Cloisite 15A montmorrilonite clay (MMT) was supplied by Southern Clay Products, Gonzales Texas, USA.

B. Preparation of fibers.

Pineapple leaves were cleaned and dried in an oven prior to pressing in a two-roll mill to remove non-fibrous component. Kenaf and sugar palm fibers were used as received.

Revised Manuscript Received on October 30, 2019.

Dzaraini Kamarun, Orchestrated Polymer Research Group, Faculty of Applied Sciences, Universiti Teknologi MARA, Shah Alam, Selangor, Malaysia. Email: dzaraini@uitm.edu.my

Engku Zaharah Engku Zawawi*, Orchestrated Polymer Research Group, Faculty of Applied Sciences, Universiti Teknologi MARA, Shah Alam, Selangor, Malaysia. Email: engku946@uitm.edu.my

Nur Hikamah Seth, Centre of Foundation Studies, Universiti Teknologi MARA Selangor, Kampus Dengkil, 43800, Malaysia. Email: hikamah@uitm.edu.my

Norlaily Ahmad, Centre of Foundation Studies, Universiti Teknologi MARA Selangor, Kampus Dengkil, 43800, Malaysia. Email: nlaily991@uitm.edu.my

Siti Rafedah Abdul Karim, Faculty of Applied Sciences, Universiti Teknologi MARA, Shah Alam, Selangor, Malaysia. Email: srafidah@uitm.edu.my

All three fibers were crushed using a pulveriser and sieved using appropriate siever to obtain the required size i.e. 0.595 and 0.25 mm. MMT were initially dried at 70°C for 24 hours before grinded using mortar and pestle.

C. Compounding and Formulation of NFPC

NFPC compound were prepared by mixing all components in a twin screw extruder machine (PRISM TSE 16TC) at 220°C barrel temperature and roller speed of 70 rpm. Extrudates were pelletized and dried before injection molded into test pieces. Four NFPC compounds were prepared. KP01 and KP02 which include mixed fibers of kenaf and PAL at 2 different wt% ratio of kenaf: PAL but with same fibre size ratio of (0.595: 0.595) mm. KP01 and KP02 were prepared with same wt % ratio of kenaf: SP (25:25) but at two different fiber size ratio. Loading of MAPP compatibilizer and MMT filler were fixed at 4 and 2 wt % respectively. Polypropylene (PP) 100 % by wt were also prepared as blank sample.

Summary of NFPC compounds and blank PP prepared is shown in the formulation given in Table I.

Table I. Formulation of NFPC compounds prepared

Sample	Kenaf : PAL (wt % ratio)	Kenaf : SP fiber size ratio (mm)	PP (wt %)	MAPP (wt %)	MMT (wt %)
PP	-	-	100	-	-
KP01	25 : 25 (at .595:0.595 mm fiber size ratio)	-	44	4	2
KS01	-	0.595 : 0.595 (at 25:25 wt % ratio)	C44	4	2
KP02	30 : 20 (at .595:0.595 mm fiber size ratio)	-	44	4	2
KS02	-	0.595 : 0.250 (at 25:25 wt % ratio)	44	4	2

D. Preparation of Test Pieces

Compounded materials were injection molded into dumbbell (74mm × 20 mm), and bar (70 mm ×20 mm) shaped test pieces employing a micro injection molding machine, Thermo HAAKE MiniJet. The injection pressure was set at 300 bar, cylinder temperature in the range of 120 -170°C and mould temperature at 100 °C.

E. Mechanical Testing

Tensile test follows ASTM D638 using Instron Testomeric Tensile Tester machine equipped with a 5kN load cell at cross head speed of 50 mm / min. The specimens were of dumb bell shape (74 × 20) mm. Flexural test were also conducted on the same machine following ASTM D790. Specimens of (70×20) mm dimension were loaded in a three-point bending test with the recommended span-to-depth ratio of 16:1 using a load of 10 kN at 3 mm / min rate of loading. Izod impact test was carried out using Digital Izod Impact Testing Machine (model RS-8215A) following the standard ASTM D256 at 4 Joules of pendulum impact energy and 3.5 m/s hammer speed. Specimens of similar dimensions to flexural test were used.

F. Thermogravimmetric Analysis

Thermal stability of fibers and composites were determined using Netzsch TG209 F3Tarsus TGA machine. Samples were crimped and placed in the sample holder and heated at a heating rate of 30^{0}C/min from (30 – 600)^{0}C under N_2 environment.

III. RESULTS AND DISCUSSION

A. Thermogravimmetric Analysis (TGA)

The degradation temperatures reported were measured from the peaks observed in the DTG curves. The peaks represent the highest rate of degradation. Fig.1 shows DTG curves of the 3 fibers used in this study. Table II summarized the data obtained from the each curves.

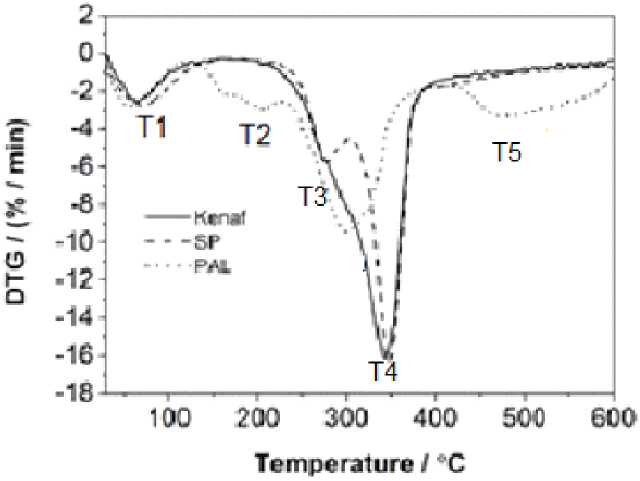


Fig. 1. DTG curves of kenaf, SP and PAL fibers

Table II. Degradation temperatures of fibers used in the preparation of NFPC

Fiber	Degradation temperatures/T_{deg} (°C)				
	T1	T2	T3	T4	T5
Kenaf	60	-	280	350	-
PAL	60	150-200	260	300	450-600
SP	60	-	280	350	-

All fibers show at least 3 degradation temperatures in the regions: a) below 100°C (T1), b) (150- 200)°C (T2), c) (260 - 280)°C (T3), (300 - 350)°C (T4) and d) (450 – 600)°C (T5). These degradation peaks correspond to degradation of the components present in the fibers. PAL consists of 2 extra degradation temperatures as shown by peaks at T2 and T5.

It is known that NF are hydrophilic in nature consisting of components such as hemicellulose, cellulose and lignin [10]. In terms of heat stability the following order was followed: lignin > cellulose > hemicellulose [11]. Cellulose which has lower heat stability than lignin degraded in the temperature range (300 – 350)°C(T4) as shown by all 3 fibers. Both SP and kenaf consist of cellulose with almost similar degradation temperature at 350 °C. Cellulose of PAL degraded at lower temperature which is 300 °C. PAL fiber showed a broad degradation peak at (450-600)°C (T5) which corresponds to the degradation of lignin higher structures. Hemicellulose component of the three fibers degrade at temperatures 260^{0}C (PAL) and 280°C (kenaf and SP) and are denoted as T3 in Fig.1. Lower structures of lignin in PAL decomposed at lower temperatures in the region (150-200)°C (T2).

Lignin is a complex three-dimensional aromatic molecule composed of phenyl groups [12] decomposes slower, over a broader temperature range (200-500 °C) than cellulose and the hemicellulose components of biomass [13]. Both SP and kenaf do not contain a measurable amount of hemicellulose and lignin as no peaks of this components are detected in the DTG curves of the fibers.

Due to the polarity of NF, bonded water was retained in its structure which dissociates below 100°C when heated. TG curves (Fig.2) of these fibers show that all fibers PAL, SP and kenaf retained about 10% of bonded water within the cell wall of their structure.

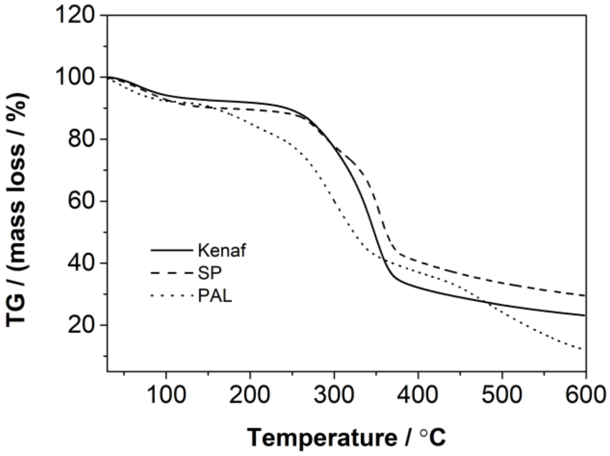


Fig.2. TG curves of kenaf, PAL and SP fibers

Fig. 3 shows the TG curves of KS01 and KP01. It can be seen that the water content of the composites were less than 5% which is important in producing composites of good properties.

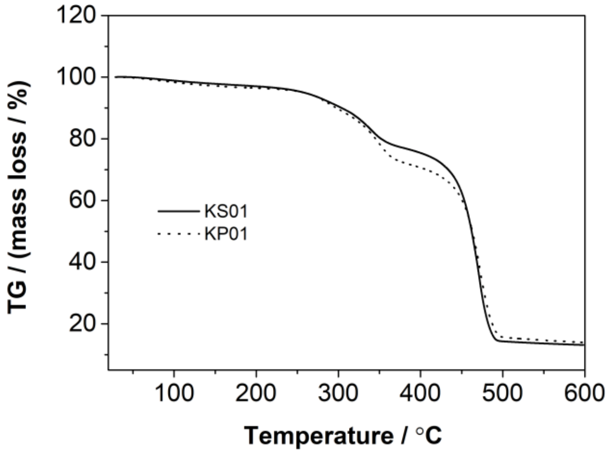


Fig.3. TG curves of KS01 and KP01 composites

Cellulose component of the fibers were still present in both composites as indicated by the presence of peaks at 339°C and 350°C of KS01 and KP01 respectively in Fig. 4. However peaks of lignin in PAL was not visible in the KP01 curves. This could be due to the fact that lignin composition in PAL is not significantly high to be detected in the composite. Peaks at ~470°C represented the degradation temperature of PP.

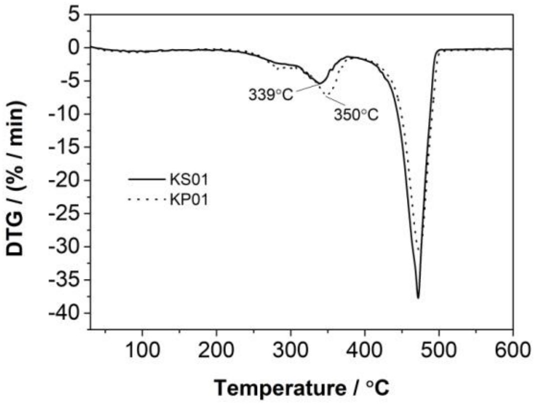


Fig. 4. DTG curves of KS01 and KP01 composites

B. Mechanical Strength

Table III summarized the tensile, flexural and impact strengths of PP and NFPC composites.

Table III: Mechanical strength of PP and NFPC composites

Samples	Tensile Strength / MPa	Flexural Strength / MPa	Impact Strength / kJm^{-1}
PP	24	30	19
KP01	40	19	26
KS01	41	17	28
KP02	44	29	22
KS02	44	30	29

A clearer relationship are shown by the bar chart presentation of tensile, flexural and impact strength respectively of PP and the various composites as in Fig.5a, 5b and 5c.

It can be seen that tensile strength of all the composites are higher by at least 80% compared to blank PP (Fig.5a). MAPP as compatibilizer and MMT as fillers cumulatively contribute to the increased in tensile strength of the composites. The flexural strength however do not follow the same trend.

It can be seen from fig. 5b that NFPC composites KP01 and KS01 show lower flexural strength than KP02 and KS02 and the flexural strength of KP02 and KS02 are comparable to PP blank. KP02 incorporated higher amount of kenaf compared to KP01 and show higher flexural strength. KS02 which have smaller sized SP fibers in the mix also show higher flexural strength than KS01 which utilize similar sized kenaf and SP fibers. It can be concluded that higher kenaf loading in KP02 composites and smaller sized SP fibers as compared to kenaf fibers in KS02 contribute to the increase in flexural strength of the NFPCs. Flexural strength is a measure of the ability of materials to withstand bending force that is applied perpendicular to its longitudinal axis [14]. Kenaf fiber with higher loading of kenaf in KP02 has higher amount of crystalline cellulose which contribute to higher tensile strength. However when the concentration of kenaf is lower as in KP01, the amount of crystalline cellulose is lower thus causing it to be more brittle. As a result the flexural strength is reduced.

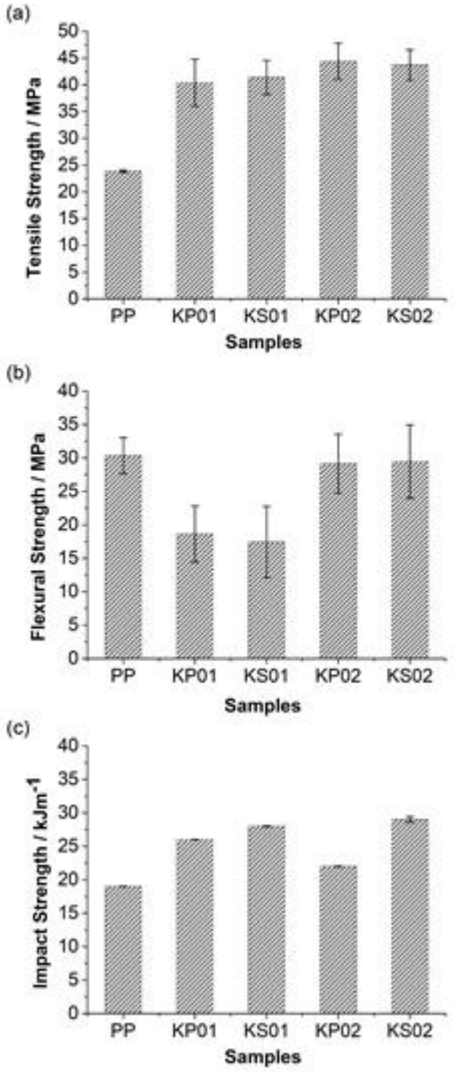


Fig. 5. Bar chart of (a) tensile strength, (b) flexural strength and (c) impact strength of PP and the various composites.

Incorporation of fiber increased the impact strength of NFPC composites compared to blank PP by ~38%. However, the impact strength of KP02 increased minimally compared to PP blank which could be due to the reduction in the amount of PAL fiber. Impact strength of KP02 is also less than KP01; KP02 which has different ratio of K: PAL fibers has lower loadings of PAL compared to KP01. Lignin which is a complex three-dimensional aromatic molecule is composed of phenyl groups [12]. Previous research shows the importance of lignin in improving impact strength of NF composites [15]. Lower loadings of PAL in KP02 compared to KP01 caused a reduction of lignin composition in KP02 which result in lower impact strength of KP02 compared to KP01. Furthermore, the difference in impact strength of these two composites could also be due to the difference in stiffness and strength of the two material which contribute to different energy absorption behavior [16]. Similarly KS02 shows higher impact strength compared to KS01. Shorter fibers of sugar palm in KS02 have the capability to fill up voids formed in between fibers to increase the fiber volume fraction and consequently its rigidity and stability.

IV. SUPERIORITY OF KENAF FIBER

All four composites incorporate kenaf as one of the mix fiber. Kenaf fiber is already known to improve tensile strength of PP composites due to its high cellulose content [17]. All four composites show higher tensile and flexural strength compared to PP blank except for the flexural strength of KP01 and KS01. Cellulose in kenaf and SP fibers degrade at higher temperatures 350 ^{o}C compared to PAL fibers (300 ^{o}C). This could be due to different crystalline structures that exist in these fibers [12]. Cellulose content of natural fibers differ in crystallinity, allomorph type and degree of crystallinity [18]. Because of the difference in crystalline structure and thermal stability of kenaf cellulose compared to PAL fiber, an increase in the loading of kenaf in KP02 contribute to the increase in the tensile and flexural strength of KP02 compared to KP01. SP which probably has similar crystalline structures to kenaf fibers as shown by the degradation temperatures of the cellulose component of these fibers show comparable tensile strength to kenaf as shown by KS01 and KS02. However, both higher loading of kenaf and mixed fiber size of kenaf and SP fibers contribute to the increase in flexural strength of the composites.

V. CONCLUSION

Kenaf and sugar palm fibers have comparable properties in terms of their composition and thermal stability. Pineapple leaf fibers have promising properties from the presence of lignin component. Mixing these fibers in eco-friendly NFPC provide opportunities for sustaining plastics composite industries as well as paving new ideas for development of biodegradable products.

ACKNOWLEDGMENT

The authors would like to thank Institute of Research Management & Innovation (IRMI), Universiti Teknologi MARA for their support and assistance in securing the research grant 600IRMI/PERDANA 5/3 MITRA (003/2018) awarded for this research. Also highly appreciated are the effort contributed by Nur Syahmi Syafiqah binti Rahman and Siti Norhanisah binti A Rahim, undergraduate students of Polymer Technology, Faculty of Applied Sciences, UiTM Shah Alam.

REFERENCES

1. M. J. Mochane, T. C. Mokhena, T. H. Mokhothu, A. Mtibe, E. R. Sadiku1, S. S. Ray," Recent progress on natural fiber hybrid composites for advanced applications: A review", *eXPRESS Polymer Letters*, Vol.13, No.2 ,2019, pp. 159–198
2. I. D. Ibrahim, O. O. Daramola, N. Phreecha and W. Chinpa, "Environmentally Friendly Composites from Seabass Scale and Oil Palm Empty Fruit Bunch Waste*", Journal of Polymers and the Environment*, Volume 27, 2009, Issue 5, pp 1043–1053
3. M.H. Alaaeddin , S.M. Sapuan, M.Y.M. Zuhri, E.S. Zainudin and, Faris M. AL- Oqla, "Physical and mechanical properties of polyvinylidene fluoride sugar palm fiber nanocomposites", *Journal of Cleaner Production*, 235, 2019, pp. 473-482
4. M. R. M. Huzaifah, S. M. Sapuan, Z. Leman and M. R. Ishak. "Effect of Fibre Loading on the Physical, Mechanical and Thermal Properties of Sugar Palm Fibre Reinforced Vinyl Ester Composites", *Fibers and Polymers*, 2019, Volume 20, Issue 5, pp. 1077–1084

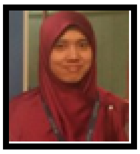
5. W.Westerhuis, S.H.van Delden, J.E.G.van Dam, J.P.Pereira Marinho, P.C.Struik, and T.J.Stomph, "Plant weight determines secondary fibre development in fibre hemp (Cannabis sativa L.)", *Industrial Crops and Products*, Volume 139, 2019.
6. M.Ramesh, "Flax (Linum usitatissimum L.) fibre reinforced polymer composite materials: A review on preparation, properties and prospects", *Progress in Materials Science*, Volume 102, May 2019, pp. 109-166.
7. L.Teuber, V.S. Osburg, W. Toporowski, H. Militz and A. Krause , "Wood polymer composites and their contribution to cascading utilization", *Journal of Cleaner Production*. 110, 2016, pp. 9–15.
8. A. N.Ahmad Kurnin, I. Mohd Halim Shah, H.Yoshida and I.Shamsul. "Recovery of palm oil and valuable material from oil palm empty fruit bunch by sub-critical water", *Journal of Oleo Science*, 65, 4, 2016, pp. 283-289, 2016.
9. S.S.Todkar and S.A.Patil."Review on Mechanical Properties Evaluation of Pineapple leaf fibre (PALF) Reinforced Polymer Composites". *Composites Part B.* 174, 2019.
10. B. Madsen and E.K. Gamstedt , "Wood versus plant fibers: Similarities and differences in composite applications", *Advances in Materials Science and Engineering*. 2013 doi:10.1155/2013/564346.
11. Y. Haiping, Y.Rong, C.Hanping, H.L.Dong and .Chuguang. "Characteristics of hemicellulose, cellulose and lignin pyrolysis". *Fuel* 86, 2007, pp. 1781–1788.
12. M. Sorieul, A. Dickson, S. J. Hill and H. Pearso "Plant Fibre: Molecular Structure and Biomechanical Properties, of a Complex Living Material, Influencing Its Deconstruction towards a Biobased Composite". Materials 2016, 9, pp. 618
13. M. Brebu and C. Vasile, "Thermal Degradation of Lignin – A Review", *Cellulose Chem. Technol*., 44, 9, 2010, pp. 353-363.
14. K.L.Pickering, M.G.A.Efend and T.M.Le. "A review of recent developments in natural fibre composites and their mechanical performance". *Compose Part A.* , 2016; 83, pp.98–112.
15. M.F. Daud, E.Z. Engku Zawawi and D. Kamarun. "Mechanical Properties of Luffa Acutangula-Filled Polypropylene". Advanced Materials Research", 2013, 812, pp. 87-92.
16. E. López-Alba, S. Schmeer and F. Díaz, "Energy Absorption Capacity in Natural Fiber Reinforcement Composites Structures" *Materials* 2018, 11, pp. 418.
17. L.Y. Mwaikambo, "Review of History, Properties and Application of Plant Fiber", *African Journal of Science and Technology*, 2006, 7, pp. 120-133
18. J. He, Y. Tang, and S.Y. Wang, "Differences in Morphological Characteristics of Bamboo Fibres and other Natural Cellulose Fibres: Studies on X-ray Diffraction, Solid State 13C-CP/MAS NMR, and Second Derivative FTIR Spectroscopy Data", *Iranian Polymer Journal*, 16, 12, 2007, pp. 807-818

AUTHORS PROFILE

Dzaraini Kamarun, PhD, is an Associate Professor cum Lecturer at the Universiti Teknologi MARA, Malaysia. She has experience in writing and editing academic literature for tertiary education and was the chief editor for her first published book, Progress in Polymer and Rubber Technology. She has published many research articles in journals and proceedings as well as in several local magazines. Her most recent published work was co-author the book entitled 'Tensile and Tear Strengths of Unfilled and Carbon-Black Filled NR' published by LAP Lambert Academic Publishing in 2018. She has over 30 years of teaching and writing experience. She received her PhD in Biosensors from the Queen Mary University of London, UK.

Engku Zaharah Engku Zawawi is a senior lecturer at the Universiti Teknologi MARA, Malaysia. She received her PhD and MSc. from Universiti Kebangsaan Malaysia. She obtained her BEng in Polymer Engineering from University of North London, UK. Her research interests are polymer composites, thermoplastic elastomer nanocomposites, polymer blending and polymer processing.

Nur Hikamah Seth, MSc. is a senior chemistry lecturer at Centre of Foundation Studies, Dengkil Campus, Universiti Teknologi MARA, Malaysia. She has 9 years experiences in teaching with UiTM and published several articles, laboratory manuals, chapters in book, STEM modul and developed innovative products as teaching kit.



Norlaily Ahmad, PhD, is a chemistry lecturer at the Centre of Foundation Studies, Dengkil Campus, Universiti Teknologi MARA, Malaysia. She has 9 years experiences in teaching with UiTM and published several articles in journals and proceedings.

Siti Rafedah Abdul Karim is a senior lecturer of the Bio Composite Technology Department, Universiti Teknologi MARA, Malaysia. Hold PhD in Wood Sciences from University of Wales, Bangor, UK. Her specialization is in wood and non-wood forest product modification, protection and treatment.