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ORIGINAL ARTICLE

Thermal Properties of Unsaturated Polyester Reinforced Kenaf Core Fiber with Hybrid Nanofillers of Cellulose Nanocrystal and Graphene Nanoplatelet

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ABSTRACT - The rising concern regarding plastic waste disposal issues had induced the need for versatile polymer-based materials with biodegradable properties to be optimally used in various applications. A massively abundant kenaf core fiber (KCF) is literally a great natural fiber to be explored as reinforcement filler because of its potential in mechanical, low cost and biodegradable. In this work, KCF were incorporated into unsaturated polyester (UPE) by using hand lay-up technique and hot compression moulding. The hybridization approach based on the utilization of nanofillers cellulose nanocrystal (CNC) and graphene nanoplatelet (GNP) also were added along the fabrications of UPE-KCF bio-composite to observe their impacts on thermal stability and its behaviour which were analyzed under thermogravimetric analysis (TGA) and differential scanning calorimeter (DSC) analysis. From the result, KCF shows a great potential in improving thermal stability with high residue. The compatibility between both nanofillers also provided even higher thermal stability for maximum degradation temperature (T_{max}) up to 387.8 °C and char residue with the range of 42.1-47.8% at 400-500 °C recorded by sample UKC1G2, and UKC0.5G2.5 respectively. However there were no obvious changes observed for the glass transition temperature (Tg) from DSC scan analysis upon the incorporation of single KCF or when nanofillers were added. Overall, hybrid loading of kenaf fiber with 3 wt% nanofillers shows appreciable impact on thermal properties of unsaturated polyester.

INTRODUCTION

The rising use of oil based plastics in the current times is prompting to the increased of plastic waste issues in the environment due to the longer natural degradation time. To forestall this, there is a need to produce a plastic with environment friendly properties. Unsaturated polyesters (UPE) are low cost thermoset polymers that have excellent mechanical properties, curing capability, thermal stability, no by-product during curing reaction and easily to be processed in a wide temperature range. Due to the fact of versatile properties, it has broadened the utilization of polyester in various applications such as automotive, boat frames and aircraft structures [1],[2],[3]. As to improvise into biodegradable properties, natural fiber should be a great way to be incorporated while producing other unique properties. High interest in natural fiber composites is because of their biodegradability, high performance, light weight, non-abrasive and low cost [4],[5].

Among the natural fiber, kenaf fiber which *Hibiscus cannabinus,* has great capability as reinforcement filler due to its good toughness, acceptable strength properties, low density, recyclability, low cost, good

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sound absorption and biodegradability [6],[7]. However, different kenaf fiber portray different properties indicating from bast fiber that have higher mechanical properties in contrast to core fiber (KCF) due to its higher cellulose content that in control of providing stability and strength to the bast fiber [8],[9]. Even so, the massively abundant KCF is still underexplored by the researcher should be a great opportunity to bring out its capability as reinforcement filler for a novel breakthrough and the hybridization of fiber technique also can be used as it supposedly enhances the properties of the materials. The hybridization composite is indicates to the combination of two or more reinforcement fillers. Nano-sized particle (1-100nm) can significantly enhance the thermal, mechanical and barrier properties for low content below than 10 wt.% [10].

In this work, hybridization of cellulose nanocrystal (CNC) and graphene nanoplatlet (GNP) was introduced into UPE-KCF biocomposite as to observe their impacts on thermal properties. CNC is light weight, easily (chemically) modified, reinforcing capability, abundance and biodegradability [11]. CNC also were found for their reinforcement in nano-composite that usually will exhibit properties enhancement such as biodegradability, thermal, mechanical and barrier properties when compared to neat polymer [12],[13]. While GNP are reasonable inorganic nano-size materials to be used as additional hybrid reinforcing filler which were found for their incorporation in composite can improve thermal, mechanical, physical and conductivity properties [14],[15]. Hence improving thermal properties of UPE bio-composite with an efficient nanofiller is interesting and this approach were expected to create high-performance composite with biodegradable properties

MATERIALS AND METHODOLOGY

Unsaturated polyester (UPE) was purchased from Mukmin Enviro Lab Sdn Bhd. The reinforcement material which is Kenaf core fiber (KCF) was supplied by Kenaf Agro Vet Sdn. Bhd. Other fillers referring to Cellulose nanocrystal (CNC) (BGB UltraTM) were purchased from Canada (Blue Goose Biorefineries Inc). While for Graphene nanoplatelet (GNP) (ECODEAR™), they were obtained from Sigma-Aldrich and Toray Malaysia Plastic. Both nanomaterials were in the form of powder.

Sample preparation and fabrication method of UPE-KCF/CNC/GNP

The research were carried out from the fabrication of UPE reinforced KCF, CNC and GNP. Initially the organic fibers KCF and CNC were purposely dried in the oven for 24 hours at 105°C to diminish the moisture absorbed. The required quantity of fillers and matrix were prepared according to the formulation displayed on Table 1. Both nanomaterials CNC and GNP were dispersed into UPE and accordingly KCF were added and stirred together as to produce a homogenous mixture. During the final step of mixing, 2% of methyl ethyl ketone peroxide (MEKP) and 1 drop of cobalt napthalene (CN) were added as curing catalyst. The fabrications of bio-composites were proceeded by using a hand lay-up technique and hot compression moulding at 120°C. The fabrication process includes 10 minutes of pre-heating, and 5 minutes cooling. To characterize the thermal properties, proper amount of each samples were weighed uniformly as required.

Table 1. Formulation of UPE-KF/CNC/GNP Bio-composites

Samples	Compositions (wt. %)			
	UPE	KCF	CNC	GNP
U	100	0	0	0
UK	70	30	0	0
UKC2.5G0.5	67	30	2.5	0.5
UKC2G1	67	30	2.0	1.0
UKC1.5G1.5	67	30	1.5	1.5
UKC1G2	67	30	1.0	2.0
UC0.5G2.5	67	30	0.5	2.5

Characterization

Thermal characterizations were analyzed through thermogravimetric analysis (TGA) and differential scanning calorimeter (DSC) by utilizing TGA/DSC Mettler Toledo simultaneous thermal analysis. The analysis was done in the nitrogen atmosphere with the heating rate of 10- 600°C min^{-1} and gas flow at 20ml min^{-1}. From the test, degradation temperature at 10% mass loss ($T_{10\%}$) and maximum degradation temperature (T_{max}) were observed. Char residue (%) at 400 and 500°C also were recorded. While in DSC test, glass transition temperature (Tg) was reported based on the scan of DSC analysis.

RESULTS AND DISCUSSION

Thermogravimetric analysis (TGA)

Thermal stability of UPE and its bio-composites were studied through thermogravimetry analysis (TGA) which the result of weight loss, thermal breakdown and degradation mechanism can be observed from the integral (TGA) and derivative (DTG) thermogravimetric curves presented in Figure 1, 2. Table 2 summarizes thermal stability data based on the temperature at mass loss of 10% ($T_{10\%}$), maximum degradation temperature (T_{max}) and percentage of char residue at 400°C and 500°C (%).

**Table 2.** Thermogravimetric analysis data of UPE- based bio-composites

Sample	Degradation Temperature (°C)		Char residue (%)	
	$T_{10\%}$	T_{max}	400 (°C)	500 (°C)
U	176.7	351.6	0	0
KCF	263.7	330.4	26	20.8
UK	250.3	378.6	30	26
UKC2.5G0.5	257	386.5	20.4	12.7
UKC2G1	257	382	17.1	10
UKC1.5G1.5	257	381.7	12.7	5.3
UKC1G2	257	387.8	17.5	9.1
UKC0.5G2.5	217	387.5	47.8	42.1

According to curve TGA in Figure 1, multi-step degradation was exhibited from all of the composite samples which refer to the loss of moisture and degradation of composite. It was observed that the initial degradation of all the composites samples occur at 37–150°C due to the elimination of moisture [16]. Theoretically, the evaporation of moisture begins at the temperature of 80°C because in facts of difficulty for the moisture to completely evaporate [17],[18]. This finding has good agreement with the reported result by Ismail, et al, (2021) [19] that the initial degradation of kenaf fiber was recorded below than 200°C. Other than that, the result shows significant improvement in thermal stability which can be identified from the temperature at 10% mass loss ($T_{10\%}$) in Table 2. Based on the data, U showed the lowest $T_{10\%}$ which recorded at 176.7°C and rose up to 250.3°C for sample UK with the addition of KCF. Kenaf fiber alone presented by KCF shows the highest $T_{10\%}$ which may clarify the thermal improvement in $T_{10\%}$. Even so, KCF shows the earliest temperature of maximum degradation (T_{max}) at 330.4°C but having higher residue compared to U which might be due to the main constituent of kenaf fiber referring to cellulose, hemicellulose and lignin. According to Ayu et al, (2020) [20], hemicellulose starts to degrade attributed to its chemical structure containing random amorphous structure with lower strength. While it accordingly followed by degradation of cellulose at a higher temperature (315–390°C) compared to hemicellulose due to its high crystallinity nature and long polymer of glucose units that make it more thermally stable [21]. Hence, the lignin constituent thermally decomposed within the temperature range of 280–500°C and in control of char residue which can be related to the results obtained by KCF sample having high residue of 20-26% at 400-500°C as well as other bio-composite samples upon the incorporation of natural fiber that had contributed for the char residue while U left no residue [22]. This also may be assigned to the fiber interaction that functioned in delaying the total degradation of

composite at higher temperature [23]. Besides, the inclusion of KCF as single reinforcement not only increase char residue in sample UK shows by 26-30% at 400-500°C but also improve T_{max} exposed by the shifted curved from 357.7°C (U) to 378.6°C. Similarly to the past study that reported higher T_{max} for single reinforcement KCF into matrix polymer compared to neat polypropylene (PP). The author also mentioned that the addition of low thermal stability kenaf lowered the decomposition rate while resulting in higher residual weight [24].

Other than that, the incorporation of both nanofillers with different loading showed by sample UKC2.5G0.5, UKC2G1, UKC1.5G1.5 and UKC1G2 were not resulted for any changes with the comparable value of 257°C for $T_{10\%}$ but showed an improvement by 7°C increment when compared to the single reinforcement UK that only reaching 250°C ($T_{10\%}$). On the other hand, the result reveals differently for UKC0.5G2.5 which shows even lower $T_{10\%}$ in contrast to sample UK, but exhibits significant improvement in T_{max} by 8.9°C increments at 387.5°C. The improvement in T_{max} also can be observed from sample UKC2.5G0.5 with the loading of 2.5 wt% GNP and 0.5wt% at 386.5°C and was recorded even higher for UKC1G2 with 2wt% GNP and 1 wt% CNC loading, at 387.8°C with 9.2°C increment. It can be clearly seen from DTG curve in Figure 2 that shows the major degradation temperature shifted to a higher range of temperature compared to single reinforcement composite and neat U which might be due to the good dispersion of nanofillers and strong interfacial adhesion between the fillers and matrix affecting thermal stability. As refer to Barkoula et al, (2008) [25], the homogenous dispersion of GNP probably acts as efficient heat sink, extracting more heat over the matrix and restrict the accumulation of heat within the latter to occur, subsequently stopping oxidation at the beginning phase of degradation. This may be explained by the potential of graphene that acts as physical barrier obstructing the diffusion of available oxygen and decreasing its permeability by imparting convoluted path and hence increase the flame retardancy properties [26],[27]. The improvement observed for this sample also probably attributed to GNP shows high thermal conductivity that may cause constrain to interfacial polymer phases in the vicinity of the graphite nanoparticle surfaces and hence the energy required for decomposition could increase, modifying the ability of degraded molecules to diffuse and evaporate [28],[29]. As for the CNC, its crystallinity may contribute to thermal stability [30] which hence exposing good compatibility along the thermal conductive GNP, improved the thermal stability. Great intermolecular bonding within polymer matrix and nanocellulose had solidly impacted the thermal stability which bond dissociation energy that expected for chain cleavage of macromolecules might be enhanced by a strong intermolecular bonding [31].

The compatibility of nanofillers in the samples UKC2G1 and UKC1.5G1.5, 1.0 somehow resulted lower (T_{max}) at 382°C and 381.7°C respectively, when compared to other hybrid bio-composite, recorded at 386.5°C and above. The reason is probably due to the poor intermolecular bonding conveyed by fillers and matrix. Poor interfacial interaction will lead to higher thermal boundary resistance, which then directing to the prevention of thermal flow either from the filler to matrix or matrix to filler [32]. Furthermore, by Yin et al, (2016) [33], the difference surface energy between the hydrophobic of UPE polymer and the inclusion of hydrophilic CNC joined with their natural propensity to agglomerate will prompt the inhomogeneous dispersion and poor interfacial adhesion of nanocellulose within the polymer matrix. These could deny the potential of the compatibility between both nanofillers CNC and GNP, which the thermal stability reduction and unobvious changes had observed. By that, strong interfacial adhesion between the fillers and matrix are required to improve the thermal stability of the composite materials [34]. As for the residue, UKC0.5G2.5 shows the highest by 42.1-47.8%, while other hybrid composite shows apparent declination from 26-30% (UK) to 5.3-12.7% conveyed by UKC1.5G1.5 at 400-500 °C. Hence, it can be summarized that the inclusion of KCF into polyester had led to the slightly better thermal stability with significant enhancement in fire resistance due to the higher amount of char formation. While nanofillers does improve and also reduce both properties. Therefore, an optimum loading of nanofillers with great dispersion and strong inerfacial adhesion is required to improve the thermal stability at low degradation temperature at the same time increase the formation of char at high degradation temperature.
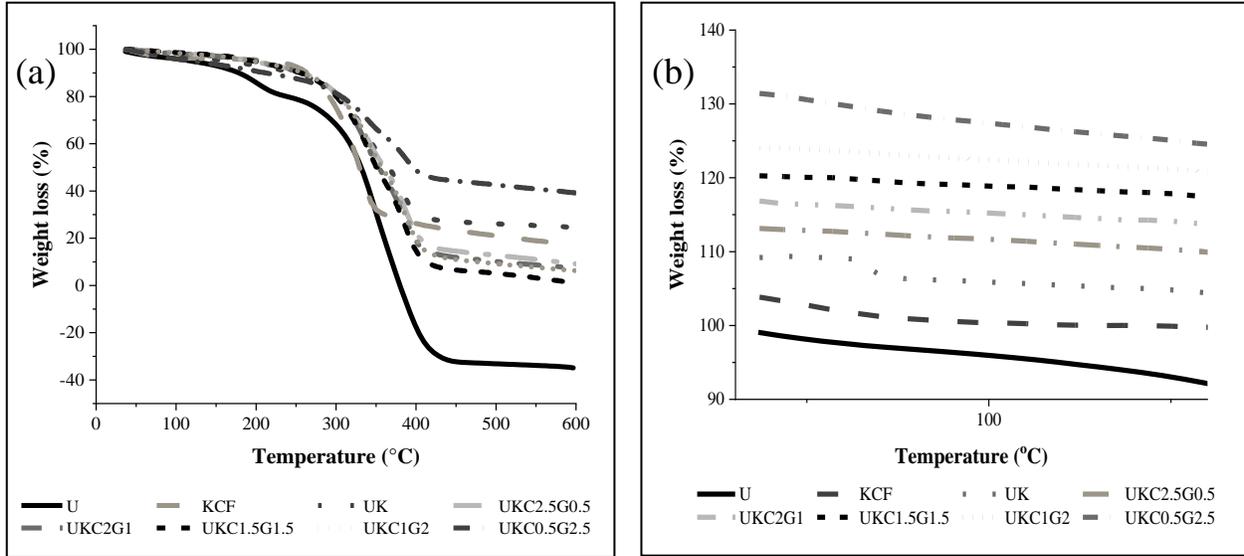
Figure 1. a) Thermogravimetric (TG) curve and b) Initial degradation for reference of sample UPE and UPE-based bio-composite.
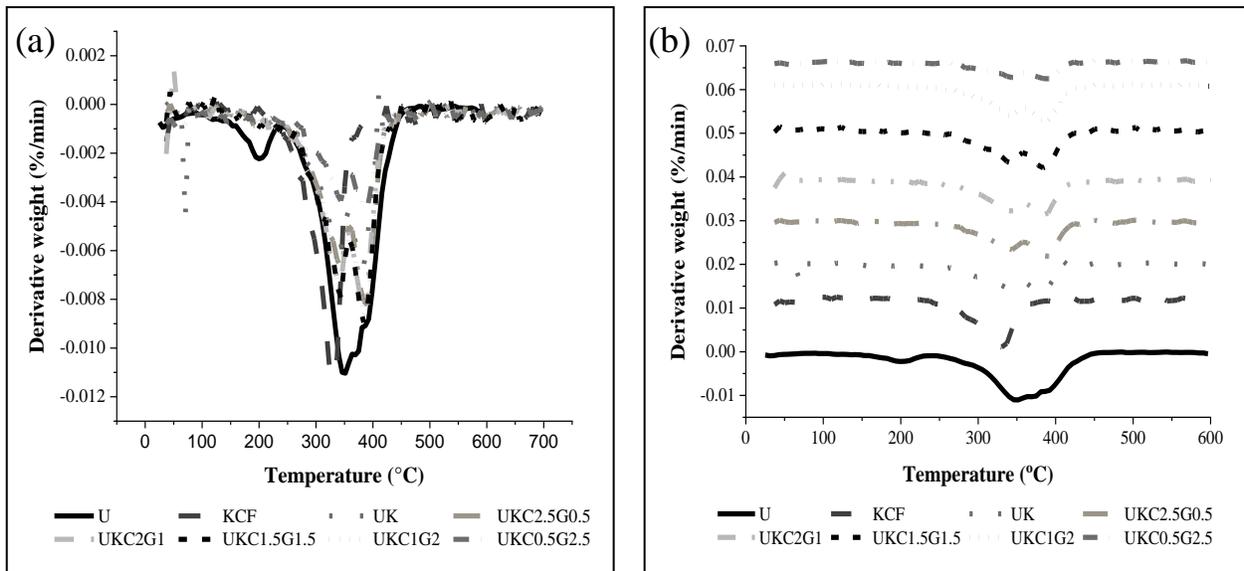
Figure 2. a) Derivative thermogravimetric (TG) curve and b) Maximum degradation curve for UPE and UPE-based bio-composite

Differential scanning calorimetry (DSC)

As refer to the Table 3, glass-transition temperature (Tg) were extracted from the DSC curve representing thermal behaviour of the tested samples in Figure 3.

Tg is the range of temperature when the amorphous rigid structure of thermoset polymer shifts from a stiff to a flexible structure or rubbery state when a certain heat energy level is achieved and the polymer molecules are permitted to freely move around. It is mainly impacted by chain mobility, crosslinking, intermolecular reaction, and branching [35]. Moreover, if the temperature that applied on the polymer is higher than Tg then, it will negatively affect the mechanical properties (strength and stiffness), and continue to keep up parts of mechanical properties until it reach the melting temperature Tm [36]. By this, Tg is a significant material property to be considered for the composites to be used in specific end-use application. From the result listed in Table 3, it shows slightly improvement from 48.4°C to 50.3°C with the incorporation of KCF loading indicated by the single reinforcement sample, UK when compared

to neat U. According to Nielsen et al, (1993) [37], the mobility of polymer chain may be restricted upon the addition of fibers or fillers and hence leading for the increment in glass transition temperature. Similarly to other bio- composite shows higher Tg compared to U with the incorporation of KCF.

Table 3. Glass transition temperature of DSC scan analysis for UPE and its bio-composite

Sample	T_g (°C)
U	48.4
UK	50.3
UKC2.5G0.5	50.5
UKC2G1	50.4
UKC1.5G1.5	50
UKC1G2	50.5
UKC0.5G2.5	50.8

Normally, the utilization of nanofillers will induce the thermal behaviour improvement. According to Qin et al, (2015) [38], the size of nanoparticle can affect the Tg enhancement which performed by the nano-sized particle in providing maximium interphase formation and allow for the greater nanoconfinement influence on bio-composite Tg value. The inclusion of nanofillers into polymer matrix also would influence the thermal behaviour by causing a restriction of chain mobility in polymer matrix [39, 40]. However, hybrid samples revealed differently which the inclusion of nanofillers does not contribute for thermal behaviour improvement with the comparable value of Tg showed at any different loading level of CNC and GNP. The temperature values shows at 50.5°C, 50.4°C, 50°C, 50.5°C, 50.8 °C representing samples UKC2.5G0.5, UKC2G1, UKC1.5G1.5, UKC1G2 and UKC0.52.5 respectively. The unobvious shifting observed from the results would be attributed to the agglomerations of GNP occur at the interface of the polymer and reinforcement leading for the obstruction of polymer cross linking [41, 42].
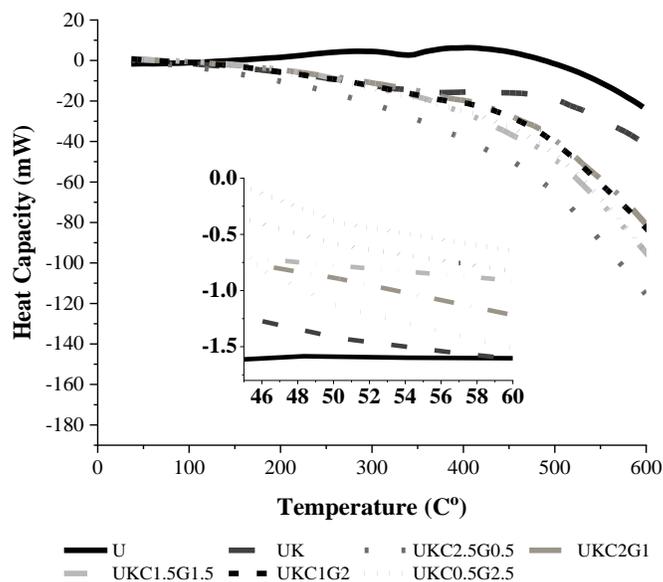
Figure 2. DSC curve for UPE and its bio-composite sample

The different loading of CNC also did not clearly shows the improvement instead giving the comparable value for all loadings. As refer to Qin et al, (2015) [38], weak interfacial attraction portrayed by the matrix and filler may lead the unwanted agglomeration to occur contributing for the weak interface and phase separation, accordingly lowering the glass transition temperature. In this study, the

incorporation of hybrid nanofillers CNC and GNP did not affect Tg value which can be related to the previous study that experienced unaffected Tg upon the inclusion of nanocellulose into thermoset polymer [43]. Also, Chiou et al, (2019) [44] noticed the trend of unchanged Tg upon the inclusion of GNP and carbon fiber in the matrix polymer.

CONCLUSION

The effect of KCF and the compatibility of CNC and GNP nanofillers were studied through TGA and DSC analysis. From the result, it was found that KCF is a potential single fiber as it shows remarkable improvement in thermal stability with high residue. The inclusion of both CNC and GNP also contribute even higher thermal stability for maximum degradation temperature (T_{max}) up to 387.8 °C by sample UKC1G2 and the highest char residue with the range of 42.1-47.8% at 400-500 °C was recorded by sample UKC0.5G2.5. Nevertheless, unaffected changes in T_g were noticed for all of hybrid bio-composite through DSC analysis. This work shall to be further continued with the proper treatment applied on KCF and critically for nanofillers CNC and GNP as to enhance the fiber-matrix interaction within the materials and produce an efficiency dispersion of nanofillers which the agglomeration issues can be reduced creating opportunity for the better properties improvement.

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REFERENCES

[1]   Ansari, F., Skrifvars, M., & Berglund, L. (2015). Nanostructured biocomposites based on unsaturated polyester resin and a cellulose nanofiber network. *Composites Science and Technology*, *117*, 298–306. https://doi.org/10.1016/j.compscitech.2015.07.004

[2]   Arjmandi, R., Yıldırım, I., Hatton, F., Hassan, A., Jefferies, C., Mohamad, Z., & Othman, N. (2021). Kenaf fibers reinforced unsaturated polyester composites: A review. Journal of Engineered Fibers and Fabrics, 16. https://doi.org/10.1177/15589250211040184

[3]   Ren, X., & Li, K. (2013). Investigation of vegetable-oil-based coupling agents for kenaf-fiber-reinforced unsaturated polyester composites. *Journal of Applied Polymer Science*, *128*(2), 1101–1109. https://doi.org/10.1002/app.38303

[4]   Balla, V. K., Kate, K. H., Satyavolu, J., Singh, P., & Tadimeti, J. G. D. (2019). Additive manufacturing of natural fiber reinforced polymer composites: Processing and prospects. *Composites Part B: Engineering*, *174*(May), 106956. https://doi.org/10.1016/j.compositesb.2019.106956

[5]   Li, S., Zheng, T., Li, Q., Hu, Y., & Wang, B. (2019). Flexural and energy absorption properties of natural-fiber reinforced composites with a novel fabrication technique. *Composites Communications*, *16*(August), 124–131. https://doi.org/10.1016/j.coco.2019.09.005

[6]   Asdrubali, F. (2006). Survey on the acoustical properties of new sustainable materials for noise control. *EURONOISE 2006 - The 6th European Conference on Noise Control: Advanced Solutions for Noise Control*, 1–10.

[7]   Mohamad Jani, S., & Izran, K. (2012). Kenaf core particleboard and its sound absorbing properties. *Journal of Science and Technology*, *4*(2), 23–34.

[8]   Ishak, M. R., Leman, Z., Sapuan, S. M., Edeerozey, A. M. M., & Othman, I. S. (2010a). Mechanical properties of kenaf bast and core fibre reinforced unsaturated polyester composites. *IOP Conference Series: Materials Science and Engineering*, *11*(December 2016), 012006. https://doi.org/10.1088/1757-899x/11/1/012006

[9]   Rouison, D., Sain, M., & Couturier, M. (2004). Resin transfer molding of natural fiber reinforced composites:

Cure simulation. *Composites Science and Technology*, *64*(5), 629–644. https://doi.org/10.1016/j.compscitech.2003.06.001

[10] Chee, W. K., Lim, H. N., Huang, N. M., & Harrison, I. (2015). Nanocomposites of graphene/polymers: a review. *RSC Advances*, *5*(83), 68014–68051. https://doi.org/10.1039/c5ra07989f

[11] Uddin, A. J., Araki, J., & Gotoh, Y. (2011). Toward "Strong" sreen nanocomposites: Polyvinyl alcohol reinforced with extremely oriented cellulose whiskers. *Biomacromolecules*, *12*(3), 617–624. https://doi.org/10.1021/bm10128of

[12] Azizi Samir, M. A. S., Alloin, F., Sanchez, J. Y., El Kissi, N., & Dufresne, A. (2004). Preparation of Cellulose Whiskers Reinforced Nanocomposites from an Organic Medium Suspension. *Macromolecules*, *37*(4), 1386–1393. https://doi.org/10.1021/ma030532a

[13] Ferreira, F. V., Dufresne, A., Pinheiro, I. F., Souza, D. H. S., Gouveia, R. F., Mei, L. H. I., & Lona, L. M. F. (2018). How do cellulose nanocrystals affect the overall properties of biodegradable polymer nanocomposites: A comprehensive review. *European Polymer Journal*, *108*, 274–285. https://doi.org/10.1016/j.eurpolymj.2018.08.045

[14] Jun, Y. S., Um, J. G., Jiang, G., & Yu, A. (2018). A study on the effects of graphene nano-platelets (GnPs) sheet sizes from a few to hundred microns on the thermal, mechanical, and electrical properties of polypropylene (PP)/GnPs composites. *Express Polymer Letters*, *12*(10), 885–897. https://doi.org/10.3144/expresspolymlett.2018.76

[15] Kong, N., Khalil, N. Z., & Fricke, H. (2021). Moisture absorption behavior and adhesion properties of gnp/epoxy nanocomposite adhesives. *Polymers*, *13*(11), 1–17. https://doi.org/10.3390/polym13111850

[16] Izwan, S. M., Sapuan, S. M., Zuhri, M. Y. M., & Mohamed, A. R. (2021). Thermal stability and dynamic mechanical analysis of benzoylation treated sugar palm/kenaf fiber reinforced polypropylene hybrid composites. *Polymers*, *13*(17). https://doi.org/10.3390/polym13172961

[17] Monteiro, S. N., Calado, V., Rodriguez, R. J. S., & Margem, F. M. (2012). Thermogravimetric behavior of natural fibers reinforced polymer composites-An overview. *Materials Science and Engineering A*, *557*, 17–28. https://doi.org/10.1016/j.msea.2012.05.109

[18] Sulong, A. B., Afiqah, N., Radzuan, M., Rizal, M., Tharazi, I., Tholibon, D., … Saleh, M. (2018). Kenaf Reinforced PLA Composite Thermoforming : A Numerical Simulation. *International Journal of Integrated Engineering*, *10*, 15–20.

[19] Ismail, N. F., Mohd Radzuan, N. A., Sulong, A. B., Muhamad, N., & Che Haron, C. H. (2021). The effect of alkali treatment on physical, mechanical and thermal properties of kenaf fiber and polymer epoxy composites. *Polymers*, *13*(12). https://doi.org/10.3390/polym13122005

[20] Ayu, R. S., Khalina, A., Harmaen, A. S., Zaman, K., Mohd Nurrazi, N., Isma, T., & Lee, C. H. (2020). Effect of Empty Fruit Brunch reinforcement in PolyButylene-Succinate/Modified Tapioca Starch blend for Agricultural Mulch Films. *Scientific Reports*, *10*(1), 1–7. https://doi.org/10.1038/s41598-020-58278-y

[21] Qu, T., Guo, W., Shen, L., Xiao, J., & Zhao, K. (2011). Experimental study of biomass pyrolysis based on three major components: Hemicellulose, cellulose, and lignin. *Industrial and Engineering Chemistry Research*, *50*(18), 10424–10433. https://doi.org/10.1021/ie1025453

[22] Tomczak, F., Satyanarayana, K. G., & Sydenstricker, T. H. D. (2007). Studies on lignocellulosic fibers of Brazil: Part III - Morphology and properties of Brazilian curauá fibers. *Composites Part A: Applied Science and Manufacturing*, *38*(10), 2227–2236. https://doi.org/10.1016/j.compositesa.2007.06.005

[23] Da Silva, T. T., Mendonça Da Silveira, P. H. P., Ribeiro, M. P., Lemos, M. F., Da Silva, A. P., Monteiro, S. N., & Cassiano Nascimento, L. F. (2021). Thermal and chemical characterization of kenaf fiber (Hibiscus cannabinus) reinforced epoxy matrix composites. *Polymers*, *13*(12), 1–15. https://doi.org/10.3390/polym13122016

[24] Ahmad Saffian, H., Talib, M. A., Lee, S. H., Md Tahir, P., Lee, C. H., Ariffin, H., & Asa'ari, A. (2020). Mechanical Strength, Thermal Conductivity and Electrical Breakdown of Kenaf Core Fiber/Lignin/Polypropylene Biocomposite. *Polymers*, *12*(8), 1833. https://doi.org/10.3390/polym12081833

[25] Barkoula, N. M., Alcock, B., Cabrera, N. O., & Peijs, T. (2008). Flame-Retardancy Properties of Intumescent Ammonium Poly(Phosphate) and Mineral Filler Magnesium Hydroxide in Combination with Graphene. *Polymers and Polymer Composites*, *16*(2), 101–113. https://doi.org/10.1002/pc

[26] Lee, J., Yun, Y. S., Kim, B., Cho, S. Y., & Jin, H. J. (2014). Nylon 610/graphene oxide composites prepared by in-situ interfacial polymerization. *Journal of Nanoscience and Nanotechnology*, *14*(8), 5703–5707.

https://doi.org/10.1166/jnn.2014.8793

[27] Xu, J. Y., Liu, J., Li, K. D., Miao, L., & Tanemura, S. (2015). Novel PEPA-functionalized graphene oxide for fire safety enhancement of polypropylene. *Science and Technology of Advanced Materials*, *16*(2). https://doi.org/10.1088/1468-6996/16/2/025006

[28] Lin, W., Xi, X., & Yu, C. (2009). Research of silver plating nano-graphite filled conductive adhesive. *Synthetic Metals*, *159*(7–8), 619–624. https://doi.org/10.1016/j.synthmet.2008.12.003

[29] Phua, J. L., Teh, P. L., Ghani, S. A., & Yeoh, C. K. (2016). Effect of Heat Assisted Bath Sonication on the Mechanical and Thermal Deformation Behaviours of Graphene Nanoplatelets Filled Epoxy Polymer Composites. *International Journal of Polymer Science*, *2016*. https://doi.org/10.1155/2016/9767183

[30] Santmartí, A., & Lee, K.-Y. (2018). Crystallinity and Thermal Stability of Nanocellulose. *Nanocellulose and Sustainability*, (January), 67–86. https://doi.org/10.1201/9781351262927-5

[31] Maiti, S., Jayaramudu, J., Das, K., Reddy, S. M., Sadiku, R., Ray, S. S., & Liu, D. (2013). Preparation and characterization of nano-cellulose with new shape from different precursor. *Carbohydrate Polymers*, *98*(1), 562–567. https://doi.org/10.1016/j.carbpol.2013.06.029

[32] Phua, J. L., Teh, P. L., Ghani, S. A., & Yeoh, C. K. (2016). Effect of Heat Assisted Bath Sonication on the Mechanical and Thermal Deformation Behaviours of Graphene Nanoplatelets Filled Epoxy Polymer Composites. *International Journal of Polymer Science*, *2016*. https://doi.org/10.1155/2016/9767183

[33] Yin, Y., Tian, X., Jiang, X., Wang, H., & Gao, W. (2016). Modification of cellulose nanocrystal via SI-ATRP of styrene and the mechanism of its reinforcement of polymethylmethacrylate. *Carbohydrate Polymers*, *142*, 206–212. https://doi.org/10.1016/j.carbpol.2016.01.014

[34] Gan, P. G., Sam, S. T., Abdullah, M. F. bin, & Omar, M. F. (2020). Thermal properties of nanocellulose-reinforced composites: A review. *Journal of Applied Polymer Science*, *137*(11). https://doi.org/10.1002/app.48544

[35] Frone, A. N., Berlioz, S., & Panaitescu, D. M. (2013). *Morphology and thermal properties of PLA − cellulose nanofibers composites*. *91*, 377–384. https://doi.org/10.1016/j.carbpol.2012.08.054

[36] Neto, J. S. S., de Queiroz, H. F. M., Aguiar, R. A. A., & Banea, M. D. (2021). A review on the thermal characterisation of natural and hybrid fiber composites. *Polymers*, *13*(24). https://doi.org/10.3390/polym13244425

[37] Nielsen, L. E., & Landel, R. F. (1993). Mechanical properties of polymers and composites, second edition, revised and expanded. In *Mechanical Properties of Polymers and Composites, Second Edition, Revised and Expanded*.

[38] Qin, X., Xia, W., Sinko, R., & Keten, S. (2015). Tuning Glass Transition in Polymer Nanocomposites with Functionalized Cellulose Nanocrystals through Nanoconfinement. *Nano Letters*, *15*(10), 6738–6744. https://doi.org/10.1021/acs.nanolett.5b02588

[39] Bastiurea, M., Rodeanu, M. S., Dima, D., Murarescu, M., & Andrei, G. (2015). Thermal and mechanical properties of polyester composites with graphene oxide and graphite. *Digest Journal of Nanomaterials and Biostructures*, *10*(2), 521–533.

[40] Montes, S., Etxeberria, A., Mocholi, V., Rekondo, A., Grande, H., & Labidi, J. (2018). Effect of combining cellulose nanocrystals and graphene nanoplatelets on the properties of poly(lactic acid) based films. *Express Polymer Letters*, *12*(6), 543–555. https://doi.org/10.3144/expresspolymlett.2018.45

[41] Gu, J., Yang, X., Lv, Z., Li, N., Liang, C., & Zhang, Q. (2016). Functionalized graphite nanoplatelets/epoxy resin nanocomposites with high thermal conductivity. *International Journal of Heat and Mass Transfer*, *92*, 15–22.

[42] Vallés, C., Beckert, F., Burk, L., Mülhaupt, R., Young, R. J., & Kinloch, I. A. (2016). Effect of the C/O ratio in graphene oxide materials on the reinforcement of epoxy-based nanocomposites. *Journal of Polymer Science, Part B: Polymer Physics*, *54*(2), 281–291. https://doi.org/10.1002/polb.23925

[43] Aydemir, D. (2015). Morphological and Thermal Properties of Cellulose Nanofibrils Reinforced Epoxy Nanocomposites Morfološka i toplinska svojstva epoksidnih nanokompozita oja č anih celuloznim nanofibrilima. 66(1), 35–40. https://doi.org/10.5552/drind.2015.1403

[44] Chiou, Y. C., Chou, H. Y., & Shen, M. Y. (2019). Effects of adding graphene nanoplatelets and nanocarbon aerogels to epoxy resins and their carbon fiber composites. Materials and Design, 178, 107869. https://doi.org/10.1016/j.matdes.2019.107869