

Dynamic Mechanical Analysis of Treated and Untreated Sugar Palm Fibre-based Phenolic Composites

Bushra Rashid,^{a,b,*} Zulkiflle Leman,^a Mohammad Jawaid,^c Mariyam Jameelah Ghazali,^d and Mohamad Ridzwan Ishak^{c,e}

Phenolic-based sugar palm fibres (SPFs) were used as a filler for composites that were fabricated by hot pressing. The composites were prepared using various volume loadings of SPFs. Dynamic mechanical analysis (DMA) was carried out to evaluate the storage modulus (E'), loss modulus (E''), and tan delta as a function of temperature. The SPFs were treated by seawater for 30 days and a 0.5 alkaline solution for 4 days. The phenolic composites with 30% volume loading of SPFs were used to determine the effect of treatments on the DMA properties of the composites. The obtained results indicate that incorporating a SPF filler notably increased the E' and E'' properties and decreased the damping factor of the phenolic composites. Both treatments affected the DMA results. However, the alkaline-treated composites showed higher DMA properties compared with the seawater-treated and untreated fibre composites.

Keywords: Composites; Sugar palm fibre; Phenolic; Treatment; DMA

Contact information: a: Department of Mechanical and Manufacturing Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia; b: Institute of Technology, Middle Technical University, 29008 Alzafarany, Baghdad, Iraq; c: Laboratory of Biocomposite Technology, Institute of Tropical Forestry and Forest Products (INTROP), Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia; d: Department of Mechanical and Materials, Universiti Kebangsaan Malaysia, 43600, Bangi, Malaysia; e: Department of Aerospace Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia; *Corresponding author: bushrarasheed50@yahoo.com

INTRODUCTION

The main interest in using natural fibres as a reinforcement in polymer composites is because they are renewable and biodegradable. Also, natural fibres offer many advantages over synthetic fibres, such as being readily available at low cost, having low density, being non-toxic and non-abrasive, and having acceptable specific properties (Jawaid *et al.* 2013; Isma'ila *et al.* 2016; Rashid *et al.* 2017c). However, they have certain drawbacks, including poor fibre-matrix interfacial bonding, high moisture absorption, and low thermal stability. Natural fibres, such as kenaf, oil palm, jute, and sisal, are widely used in various polymer composite applications, such as in the automobile industry, building materials, and food packaging (Ridzuan *et al.* 2016; Rashid *et al.* 2017b).

Dynamic mechanical analysis is a powerful and effective technique that is used to evaluate the viscoelastic mechanical thermal properties of polymer composites (Shinoj *et al.* 2011; Saba *et al.* 2016a). The storage modulus (E') refers to the Young's modulus and corresponds to the stiffness of the material of the composites. The tendency of the materials to dissipate heat under applied energy is related to the loss modulus (E'') (otherwise, it is defined as the viscous response of the materials) (Jawaid *et al.* 2013). Tan delta or dynamic

loss typically corresponds to the internal friction and molecular motion (Saba *et al.* 2015). Polymer materials have a higher damping factor than natural fibres. The temperature at which polymer materials change from a glassy to a rubbery state is defined as the glass transition temperature. The homogeneous, heterogeneous, and changes in material structure occurring in polymer composites can be described by a Cole-Cole plot (Goriparthi *et al.* 2012; Manoharan *et al.* 2014).

Several studies have evaluated the dynamic mechanical properties of natural fibre polymer composites. For example, Ray *et al.* (2002) studied the dynamic mechanical properties of untreated and treated jute fibres reinforced vinyl ester composites. A decreasing trend in E' with an increase in temperature was observed for all composites. Also, the tan delta of the composites was lowered with the addition of the jute fibre. The reinforcement of vinyl ester with jute fibre increases E' values of the composites as a result of more interfacial stress transfer. In a different study, the DMA properties of natural flax fibre reinforced polypropylene, polylactic acid (PLA), and epoxy composites were found to be better than those of carbon and glass composites (Duc *et al.* 2014). The best compromise between stiffness and damping was reported in the case of flax fibre reinforced in semi-crystalline biodegradable PLA. The DMA stiffness analysis was conducted on celluloses microfibers (CMF) based poly (ethylene-co-vinyl acetate) (EVA) composites (Sonia *et al.* 2013). The results showed that stiffness and damping decreased with CMF loading, while the storage modulus increased up to 7.5%. However, the modulus decreases at higher CMF filler loading. Recently, the damping properties of untreated coconut and treated sheath fibre were evaluated (Kumar *et al.* 2014). The results depicted that tan δ was decreased and E' value increased for the treated composites. This is due to higher interfacial bonding between coconut sheath fiber and epoxy matrix.

Surface treatments of natural fibres greatly affect the fibre-matrix interfacial bonding (Kabir *et al.* 2012). Research studies have reported that surface treatments of natural fibres increase the interlocking bonding between the fibre and the matrix, which results in effective load transfer (Ariawan *et al.* 2015). Treatment of natural fibres with an alkaline solution causes changes such as the dissolution of hemicelluloses and part of lignin, increased fibre filamentation, and improved crystallinity, which leads to the enhancement of the behaviour of the biocomposites (Fiore *et al.* 2015).

The effect of alkaline treatment of SPF-reinforced epoxy composites has been studied (Bachtiar *et al.* 2009). The treated fibres possessed good fibre-matrix interfacial bonding, which led to improved mechanical properties of the epoxy composites. Also, the alkaline treatment reduced the hydrophilic tendency of the SPFs, which resulted in enhanced fibre-matrix bonding. However, some studies have reported that a high percentage of sodium hydroxide could result in a decrease in the properties of the composites (Kabir *et al.* 2012; Jumaidin *et al.* 2017). It has been reported that seawater can be used to treat natural fibres because it is available at low cost (Ishak *et al.* 2013; Rashid *et al.* 2017). In addition, its salinity helps to remove the rough layer from the outer surface of the fibre. Leman *et al.* (2008) concluded that treating SPFs with seawater for 30 days gave the best mechanical properties when used as reinforcement for epoxy composites.

Sugar palm fibre or *Ijuk* is abundantly available in Southeast Asian countries, such as Indonesia and Malaysia. The main advantage of SPF is its high durability and excellent resistance to seawater. This is the reason why the local people have used it for many years to fabricate ropes. Many research studies have been undertaken to evaluate the characterisation of SPFs and their utilisation as a reinforcement material (Ishak *et al.* 2013).

Previous works have determined that using SPFs as reinforcing materials enhanced the mechanical properties of polymer composites (Rashid *et al.* 2016a). For example, the mechanical and dynamic mechanical properties of SPF-reinforced high-impact polystyrene were studied (Bachtiar *et al.* 2012). The results showed that the mechanical properties increased as the fibre loading increased, and the DMA properties showed an increase in the storage modulus.

Phenolic resin (PF) has high rigidity and possesses good adhesion properties. Furthermore, it has a dimensional stability and high thermal resistance because of its crosslinked aromatic group structure (Joseph *et al.* 2002). Thus, it is used as a binder in friction composites. The incorporation of natural fibres in phenolic resin could help to reduce the high brittleness and improve the cure shrinkage. This would contribute to overcoming the main drawbacks of phenolic resin, which limit its utilisation in a variety of applications. Phenolic resin used as a binder in friction materials plays a crucial role in determining the tribological properties, since the manufacturing condition are affected by thermal properties of the binder (Milanese *et al.* 2012). Natural fibres and phenolic resin both have a good chemical bonding created by the high polarity of phenolic resins when very sturdy hydrogen bonds are formed with -OH groups; then, good van der Waals forces are developed and chemical reactions between the fibres and the resin are promoted (Milanese *et al.* 2012).

Although SPFs have a high resistance to seawater and good strength, the full prospective utilisation of this fibre has not yet been explored. Several studies have been carried out on SPFs reinforced polymer composites such as high impact polystyrene, epoxy, unsaturated polyester, and polypropylene composites (Ishak *et al.* 2013). To the best of our knowledge, there has been no research undertaken to study the influence of various loadings of SPFs as reinforcement in phenolic composites. In previous studies, SPF/PF composites were fabricated and their physical, mechanical, and tribological properties were evaluated (Rashid *et al.* 2016a,b, 2017a,b,c). Also, it was reported that most natural fibres are stable up to 300 °C (Shalwan and Yousif 2013). Besides, phenolic resin is highly resistant to heat (Sreekala *et al.* 2005). Thus, SPFs-based phenolic composites may be used as friction composites. Hence, the objectives of the current research are to produce sugar palm fibre-based phenolic (SPF/PF) composites by compression moulding to be used as a reinforcement material for tribology applications. For friction composites, the temperature of the contact surfaces may be increased due to increases in the sliding contact forces. Thus, the temperature would highly influence the properties of these composites. Therefore, the mechanical properties as a function of temperature should be considered. This work focuses on the dynamic mechanical properties of SPF-based phenolic composites in terms of fibre loading, as well as determining the effect of different treatments on the DMA properties of the phenolic composites.

Fibre loadings with volumes of 0%, 10%, 20%, 30%, and 40% were used to prepare the composites. The SPFs were treated with seawater for 30 days (Leman *et al.* 2008) and for 4 h with a 0.5% alkaline solution to study the effect of treatments on the SPF/PF composites. The DMA analyses were carried out to evaluate the storage modulus, loss modulus, and tan delta of the SPF/PF composites.

EXPERIMENTAL

Materials

Preparation of materials

Sugar palm fibres (*Arenga pinnata*) were obtained from an area in Kampung Kuala Jempol, Negeri Sembilan, Malaysia. The fibres were black and tough. The fibres were wrapped around the whole length of the trunk of the sugar palm tree, and were cleaned and washed using tap water and then dried. Then they were ground to obtain fine particles with a size of $\leq 150 \mu\text{m}$ that were used in the current study. The sugar palm particles were oven-dried for 24 h at $80 \text{ }^\circ\text{C}$ to eliminate the moisture content in the SPFs before being used for fabrication of the composites. A typical straight novolac phenolic type resin (PH-4055) was used for the composite preparation, which was supplied by Chemovate, Bangalore, India.

Fibre treatments

Alkaline treatment: The SPFs were soaked in a 0.5% solution of sodium hydroxide for 4 h at room temperature. Then, the fibres were washed with distilled water and dried (Rashid *et al.* 2016b). **Seawater treatment:** The fibres were immersed in seawater for 30 days (Leman *et al.* 2008). Subsequently, the fibres were washed with tap water and dried.

Fabrication of composites

The composites were produced by varying the sugar palm fibre content from 0% to 40% within a range of 10% by volume in the phenolic resin. In this study, the SPFs were used in powder form with sizes $\leq 150 \mu\text{m}$, and the composites were prepared by mixing, hot pressing, and post-curing. The SPFs and phenolic powder formulations were dry-blended properly using mechanical stirring for 15 min at 250 to 500 rpm to obtain a homogeneous mixture. Then, the mixture was poured into a mould and hot-pressed at a temperature of $160 \text{ }^\circ\text{C}$ and pressure of 20 tons for 20 min. At the beginning, the pressure was released several times to release the gases involved in the cross-linking of the phenolic resin (Surojo *et al.* 2014). Then, the sample was post-cured in an oven at a temperature of $130 \text{ }^\circ\text{C}$ for 4 h to relieve the residual stress in the composites.



Fig. 1. (a) Sugar palm tree; (b) SPF and PF powders; (c) SPF/PF composite; (d) DMA samples

Finally, composites with dimensions of 150 mm x 150 mm x 3 mm were cut using a vertical band saw according to the standard ASTM D5418-15 (2015). A composite of 30% vol. SPF/PF was used to evaluate the effect of the SPF treatments in the phenolic composites on the DMA properties. In this study, composites with 30% vol. of untreated, seawater-, and alkali-treated SPF-based phenolic composites are denoted UT, ST, and AT composites, respectively. Figure 1 (parts a to d) show a sugar palm tree, sugar palm fibre and phenolic powders, SPF/PF composites, and the DMA test samples, respectively.

Methods

The dynamic mechanical analysis of the SPF-based phenolic composites was performed using a DMA instrument (TA Instruments (New Castle, DE) model Q800 V20.24 Build 43). The analysis was carried out at a fixed frequency mode (1.0 Hz) and at a heating rate of 5 °C/min in a nitrogen atmosphere. The samples were tested in a dual cantilever mode within a temperature range of 30 to 200 °C. The dimensions of the samples were 50 mm x 10 mm x 3 mm. The storage modulus, loss modulus, and tan delta of the composites were recorded as functions of the temperature.

RESULTS AND DISCUSSION

Storage Modulus (E')

Dynamic mechanical analysis is an effective thermomechanical technique used for evaluating the viscoelastic behaviour of polymer composites in high temperature applications such as the tribology area (Sreekala *et al.* 2005). Typically, the storage modulus (E') corresponds to the Young's modulus or the stiffness of the materials (Saba *et al.* 2016a). The effect of SPF loading in the phenolic composites on the storage modulus is illustrated in Fig. 2. The results clearly show that E' increased with increasing SPF loading and decreased with an increase in temperature for all composites (Ray *et al.* 2002). In the glassy region, the storage modulus remarkably increased with the addition of filler up to 30% vol. and then declined. Moreover, the close, tightly packed components in a frozen state resulted in a high storage modulus value below glass transition temperature (T_g).

Interestingly, the 30% vol. SPF/phenolic composites displayed the highest storage modulus value in the glassy region amongst all the composites. This trend was in agreement with the flexural modulus of SPF/PF composites reported by Rashid *et al.* (2016a). It was clearly shown that the addition of the SPF filler restricted the movement of the phenolic chains. The E' curves displayed a gradually decreasing trend from approximately 65 to 180 °C, indicating a glass/rubbery state transition. It is evident that the components of composites tend to decrease the molecular mobility (Ariawan *et al.* 2015). Additionally, the gradual decrease in the E' as the temperature increased indicated the highly crosslinked density of the composites, which meant a tighter network structure and higher stiffness (Saba *et al.* 2016a; Duc *et al.* 2014). This was due to the fact that the chain motions were perturbed in the immediate vicinity of the cross-links (Sreekala *et al.* 2005). On the other hand, no notable changes were shown with respect to the rubbery state of the SPF/PF composites for all filler loadings. The results that were obtained were in agreement with the findings of other researchers (Bachtiar *et al.* 2012; Essabir *et al.* 2016; Jawaid *et al.* 2013; Manoharan *et al.* 2014).

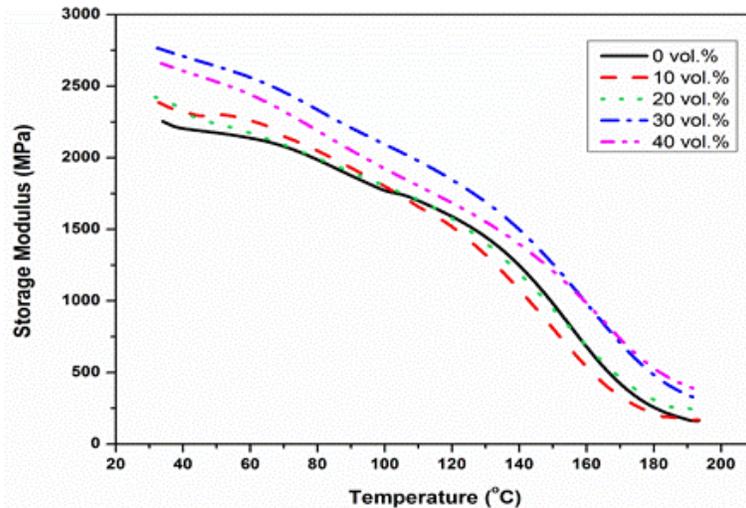


Fig. 2. Storage modulus curves of SPF/PF composites

The results also showed that the incorporation of the SPFs in the phenolic composites increased the storage modulus. Furthermore, the E' results of the composites presented values that were comparable to those of oil palm fibre/phenol formaldehyde composites (Sreekala *et al.* 2005). Also, the higher values of the storage modulus for all the SPF filler composites reflected their relatively high mechanical properties compared with those of the neat phenolic composite. However, the stiffness of the composites depended on the inherent stiffness imparted by the fibres, which enable the efficient transfer of stress (Jawaid *et al.* 2013). In a similar study conducted by Sreekala *et al.* (2005) on oil palm/glass fibre-reinforced phenolic composites, the storage modulus (E') exhibited high enhancement with fibre loading.

Loss Modulus (E'')

Loss modulus is the viscous response of a material. In other words, it is a measure of the energy that is dissipated as heat under deformation (Jawaid *et al.* 2013; Saba *et al.* 2016a). Figure 3 illustrates the variation in the loss modulus of the SPF filler in the phenolic composites with temperature. The glass transition occurred when the storage modulus decreased rapidly and the loss modulus reached a maximum value (Bachtiar *et al.* 2012; Manoharan *et al.* 2014; Saba *et al.* 2016a). All loss modulus curves reached the maximum values for the dissipation of the mechanical energy and decreased at higher temperatures as a result of the free movement of the polymer chain.

The loss modulus of the composites displayed a trend similar to that of the storage modulus. As the temperature increased, the free movement of the polymeric chains increased. It is clear from the figure that the glass transition temperatures, represented by the peak values of the curves, ranged from 150 to 157 °C for the SPF/phenolic composites. The high values of T_g could be associated with the decreased mobility of the matrix chain caused by the addition of the SPF filler and indicate a good interaction between the SPFs and the phenolic matrix. Moreover, the highest value of the E'' maximum was observed for 30% vol., suggesting that an increased fibre loading would increase the internal friction and improve the energy dissipation (Manoharan *et al.* 2014). The results that were obtained were also in line with the findings of other researchers (Sreekala *et al.* 2005; Saba *et al.* 2016b).

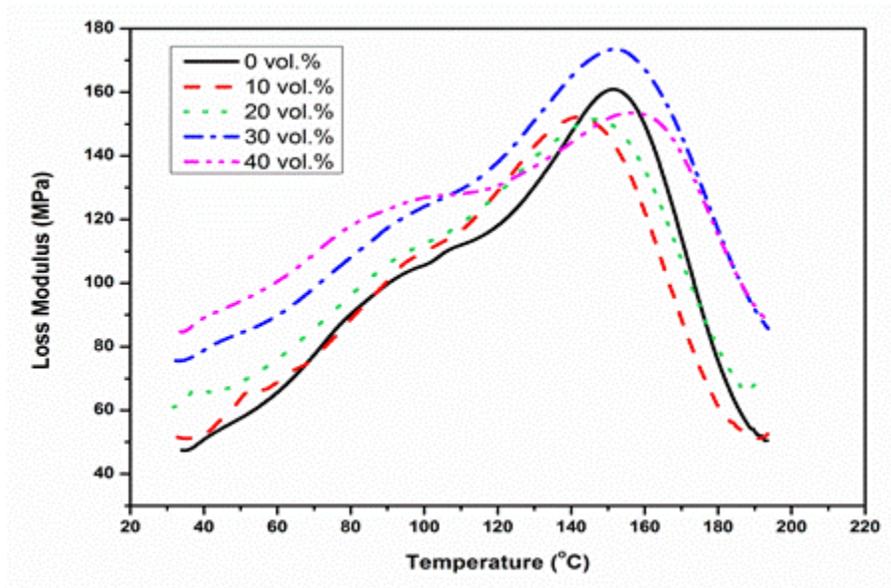


Fig. 3. Loss modulus curves of SPF/PF composites

Damping Factor ($\tan \delta$)

The damping factor ($\tan \delta$) is defined as the ratio of the loss modulus to that of the storage modulus ($\tan \delta = E''/E'$). The interlocking bonding of the fibre-matrix and composites' curing behaviour can be analysed from the damping factor (Manoharan *et al.* 2014). Figure 4 displays the effect of the SPF filler in the phenolic composites on the damping factor.

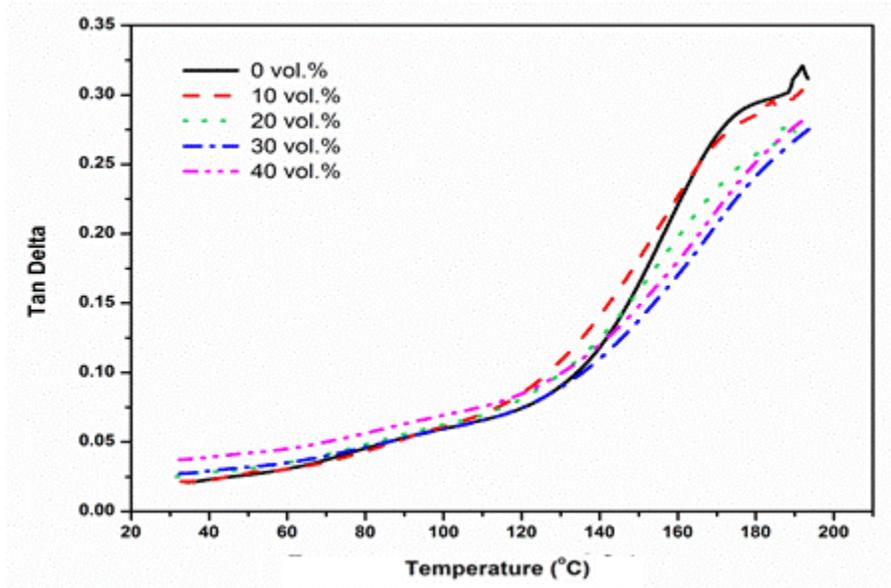


Fig. 4. Tan delta curves of SPF/PF composites

It can be clearly observed that the damping factor was relatively more affected by the introduction of the SPF filler in the phenolic composites. As the temperature increased, the chain segments became more mobile, which led to an increase in the damping factor.

Thus, the lowest degree of the molecular mobility is the lower $\tan \delta$ (Manoharan *et al.* 2014).

Observation of the figure shows that the damping factor increased as the temperature increased for all SPF-reinforced composites. Moreover, the neat phenolic composite displayed the highest damping factor amongst all the composites at high temperatures. In the rubbery region, as the amount of SPF filler increased, the damping factor decreased. The incorporation of a stiffer filler in the polymer matrix restrained the segment mobility of the matrix molecules. This was good evidence that the SPF filler embedded in the phenolic chains diminished their mobility and decreased the friction between them. Thus, the mobility of the composites decreased (Bachtiar *et al.* 2012). Hence, the addition of the SPF filler decreased damping and mechanical loss to overcome the inter-friction between the molecular chains.

A similar discussion has been reported in the literature (Jawaid *et al.* 2013; Manoharan *et al.* 2014; Saba *et al.* 2016b). However, Sreekala *et al.* (2005) reported that as the amount of oil palm fibres increased, the damping factor increased in phenolic composites. They indicated that the fibre-matrix interfacial adhesion was proportional to the fibre content. They also investigated the dynamic mechanical properties of the composites at different frequencies and found that the damping factor increased as the frequency decreased. Adding the SPF filler to the phenolic composites increased the interfacial adhesion of the fibre-matrix and served as a barrier that restricted the movement of the mobility of the chain segments. Therefore, an increase in the storage modulus and reduction of the damping factor occurred.

The composites with 30% vol. SPF/PF showed a lower damping factor, indicating that with the addition of SPFs up to 30%, better fibre-matrix interlocking bonding occurred. This promotes effective stress transfer between the fibre and the matrix and increases the load-bearing behaviour of the composites. It was clearly shown that the composites of 40% vol. SPF/PF had a damping factor less than that of the 30% vol. composites, which revealed poor fibre-matrix interfacial bonding. This may have occurred because the phenolic resin required to penetrate into the SPFs' particles to bond them is not sufficient. Thus, the fibre-matrix interfacial bonding would be weak, resulting in a lower $\tan \delta$.

Effect of Treatments on the Dynamic Mechanical Analyses

Natural fibre-reinforced polymer composites offer many advantages and certain drawbacks as well (Jawaid *et al.* 2013). Fibre-matrix interfacial bonding is one of the main issues affecting the strength of bio-composites. To overcome this problem, a fibre surface treatment would help to improve the fibre-matrix adhesion by increasing the roughness of the fibre (Goriparthi *et al.* 2012). Also, the treatment can increase the contact area of the cellulose exposed on the fibre surface; therefore, the number of possible reaction sites will increase, leading to the binding of the materials more tightly. The storage modulus of the UT, ST, and AT composites varied with temperature.

The effects of the treatments on the storage modulus of the SPF/PF composites are shown in Fig. 5. The storage modulus of the composites was slightly influenced by the fibre treatments. The E' values of the composites increased with increasing temperature. The AT composites exhibited the largest storage modulus, followed by the ST composites and then the UT composites. All the composites showed the same storage modulus trend in the glassy and rubbery transition phases.

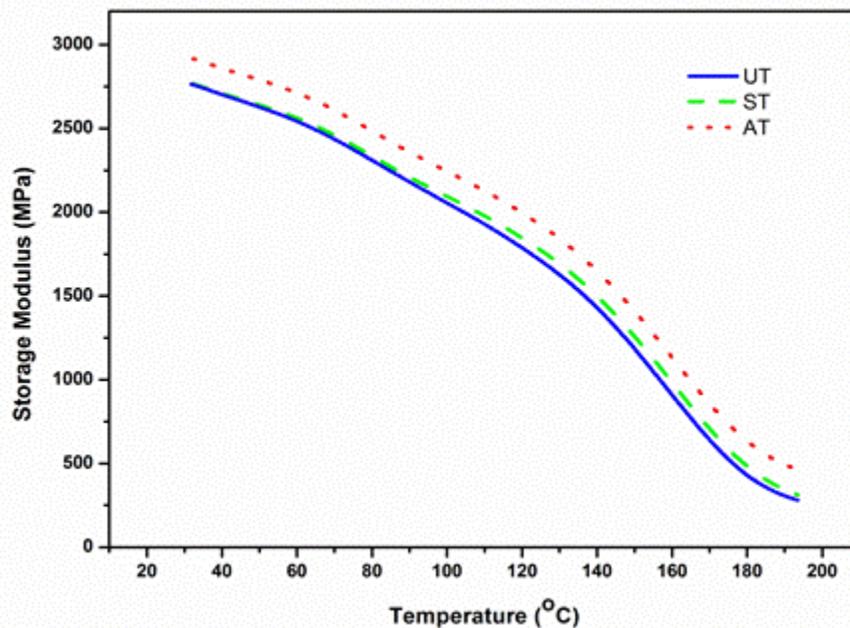


Fig. 5. Effect of treatments on the storage modulus of SPF/PF composites

It has been reported that seawater and alkaline treatments can enhance the physicochemical properties of SPFs (Rashid *et al.* 2016b). In addition, the alkaline treatment has chemical and physical effects on the SPF fibres. In terms of the chemical effects, the alkaline treatment dissolved the hemicellulose and a certain amount of lignin, which then increased the cellulose content in the fibre. Regarding its physical effect, it helped to remove the outer layer and increased the exposed area's contact with the resin. On the other hand, the seawater treatment only had a physical effect on the SPFs. Because of its salinity, seawater helped to remove the external rough surface of the SPF fibre, which is composed of pectin, wax, and impurities. This could have led to better fibre-matrix bonding and an increased storage modulus for the composites; it is a reasonable explanation for the high values of the storage modulus of the treated composites and is in agreement with other studies (Leman *et al.* 2008; Ishak *et al.* 2013). Ridzuan *et al.* (2016) found that the alkaline treatment of *Pennisetum purpureum*/glass-reinforced epoxy composites led to higher E' values compared with composites formed with untreated fibres.

The effects of fibre treatments of the SPF/PF composites on the loss modulus (E'') are shown in Fig. 6. In the glassy phase, there is a slight difference observed in the loss modulus of the untreated and treated composites. The UT composites exhibited the smallest loss modulus because of the deficiency of heat dissipation. In the rubbery transition phase, as expected, the AT composites were found to have the highest loss modulus values. The surface of the fibre became rougher and serrated after the treatment because of the removal of the wax and other impurities that covered the external surface of the fibre. Thus, the compatibility of the fibre with the matrix improved, resulting in increased interlocking bonding between the fibre and the matrix. Therefore, heat could be dissipated to the interface because of the good frictional resistance of the molecular mobility (Shinoj *et al.* 2011).

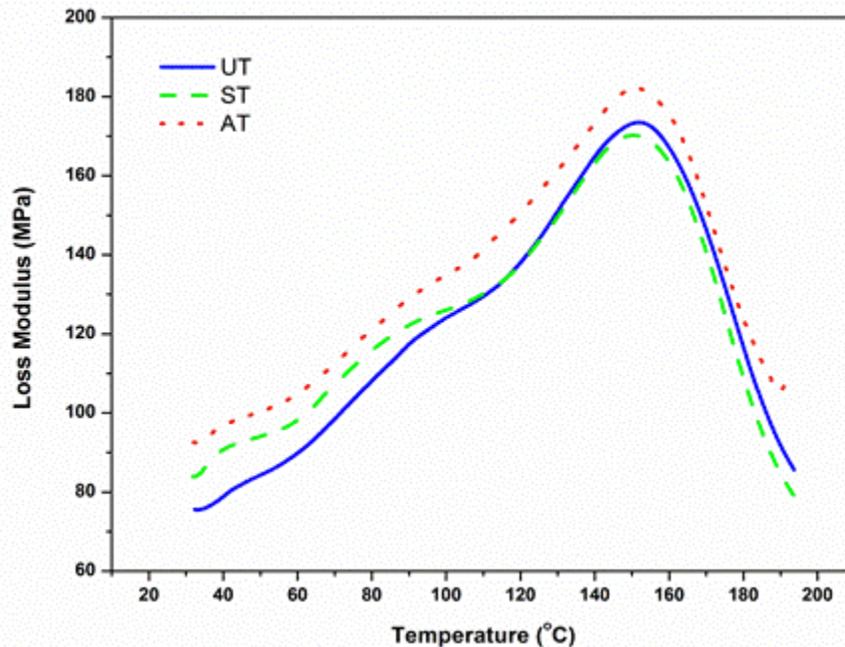


Fig. 6. Effect of treatments on the loss modulus of SPF/PF composites

The seawater treatment helped to remove the outer layer of the fibre and improved the fibre-matrix bonding, which resulted in a high amount of energy being dissipated as heat. On the other hand, the alkaline treatment removed the outer surface and dissolved the hemicellulose content in the fibre as well. This led to a high fibre-matrix adhesion and improved the mechanical strength of the composites. Thus, the AT composites exhibited the highest loss modulus in the glassy phase. The UT, ST, and AT composites showed relatively similar values of loss modulus. This could correspond to the good bonding between the fibre and the matrix, which reduced the molecular disorder at the bonding surface (Goriparthi *et al.* 2012).

The influence of the fibre surface treatments on the damping factor ($\tan \delta$) values is presented in Fig.7. There are many factors that influence the damping factor of polymer composites, such as fibre breakage, fibre-matrix interlocking, matrix cracking, interphase zone, and frictional resistance (Ridzuan *et al.* 2016). In the current study, there was no notable change in the damping factor for any of the composites in the glassy region. It has been reported that lower damping factors are associated with better load bearing properties of composites (Jawaid *et al.* 2013). As the interfacial bonding between the fibre and the matrix increased, the molecular chain mobility decreased at the fibre-matrix interface, which resulted in the decrease in the damping factor. The figure shows that the $\tan \delta$ increased with increasing temperature for all composites. At the rubbery transition phase, the AT composites revealed lower damping factor values than those for the ST and UT composites. This indicated a strong interlocking of the fibre-matrix bond and revealed a high restriction of the molecular mobility (Goriparthi *et al.* 2012). Amongst the three composites, the AT composites exhibited the highest damping factor values, followed by the ST and UT composites. This gives credence to the effect of the treatment on the fibre-matrix interfacial bonding. The damping properties of heterogeneous materials depend on certain factors, for example, breakage or bonding of fibres, matrix cracking, and holes caused by fibre pull-out (Ridzuan *et al.* 2016).

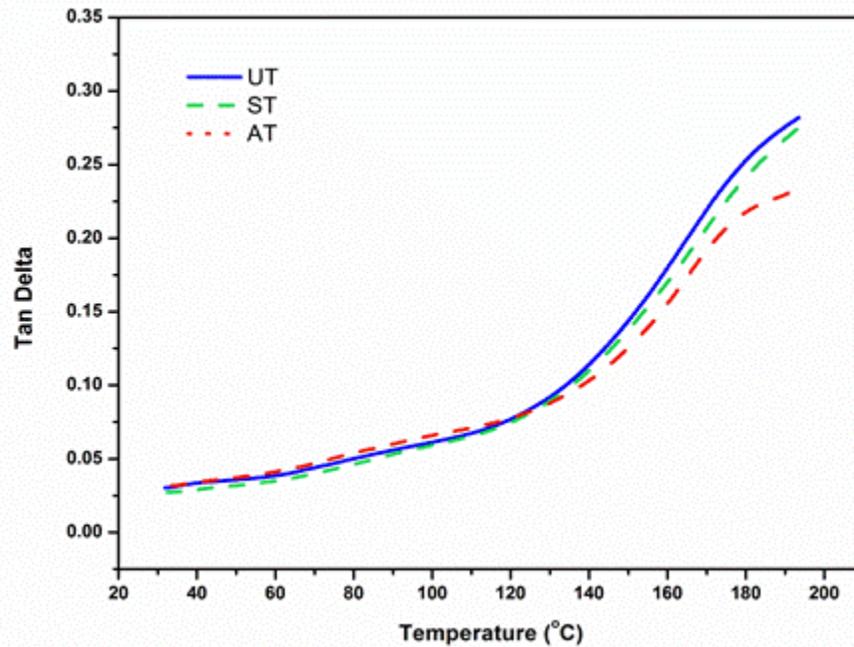


Fig. 7. Effect of treatments on the $\tan \delta$ of SPF/PF composites

A Cole-Cole plot is an effective and powerful tool to evaluate the relationship between the dynamic modulus (E') and dynamic loss modulus. Homogeneous polymer composites are associated with smooth, semi-circular arc curves in Cole-Cole plots, whereas irregularly shaped curves indicate nonhomogeneous dispersion and heterogeneity of polymer phase composites.

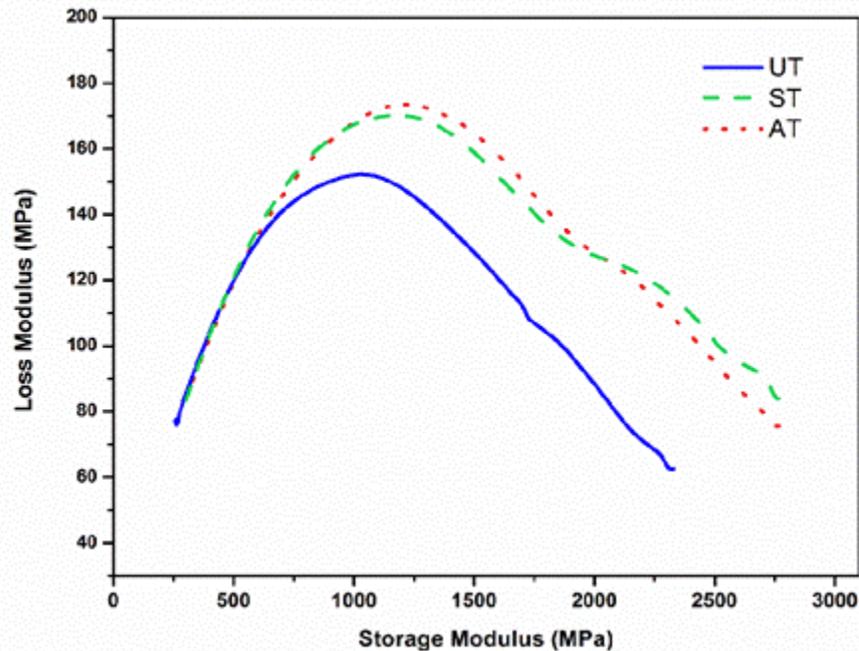


Fig. 8. Cole-Cole curves of UT, AT, and ST composites

Figure 8 shows the Cole-Cole curves of the UT, ST, and AT composites. The dynamic loss values were plotted as a function of the dynamic modulus. From the curves, it is clearly apparent that all the composites displayed nearly semi-circular Cole-Cole curves, indicating that all the composites had good fibre-matrix interfacial bonding. This confirms the idea that there is a positive compatibility and high affinity between natural fibres and phenolic resin because of the similar aromatic ring in their molecular structure (Saba *et al.* 2016a). However, the AT and ST composites' Cole-Cole curves are shown to be relatively higher than the UT composites. This was good evidence that the external surface treatments improved the fibre-matrix bonding (Rashid *et al.* 2016b).

CONCLUSIONS

1. This study evaluated the dynamic mechanical analyses of sugar palm fibres embedded in phenolic composites. Moreover, the influence of seawater and alkaline treatments of SPFs on the DMA analyses was also studied.
2. The results clearly indicated that the composites of 30% vol. SPF/PF had the highest storage modulus and loss modulus values. The SPFs introduced to the phenolic composites acted as barriers to restrict the molecular chain mobility.
3. The value of the storage modulus of the neat phenolic composite was the lowest amongst all the SPF/phenolic composites. As the fibre loading increased, the effectiveness of the stress transfer increased.
4. There was an increase in the loss modulus in the glassy and rubbery regions for all the composites. Also, the addition of the SPF to the phenolic composites reduced the damping factor, reflecting a good fibre-matrix interface.
5. The seawater- and alkaline-treated sugar palm fibres slightly affected the DMA properties of the phenolic composites. The Cole-Cole plots of the treated fibre composites showed a regular shape, revealing homogeneous polymer composites.
6. Adding sugar palm fibres may considerably enhance the DMA properties of phenolic composites. In short, the composites can be effectively used at elevated temperatures such as friction materials.

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REFERENCES CITED

- Ariawan, D., Mohd Ishak, Z., Salim, M., Mat Taib, R., Ahmad Thirmizir, M., and Pauzi, H. (2015). "The effect of alkalization on the mechanical and water absorption properties of nonwoven kenaf fiber/unsaturated-polyester composites produced by resin-transfer molding," *Polym. Compos.* 37(12), 3516-3526. DOI: 10.1002/pc.23551
- ASTM D5418-15 (2015). "Standard test method for plastics: Dynamic mechanical properties: In flexure (dual cantilever beam)," ASTM International, West Conshohocken, PA.
- Bachtiar, D., Sapuan, S., and Hamdan, M. (2009). "The influence of alkaline surface fibre treatment on the impact properties of sugar palm fibre-reinforced epoxy composites," *Polym. Plast. Technol. Eng.* 48(4), 379-383. DOI: 10.1080/03602550902725373
- Bachtiar, D., Sapuan, S., Khalina, A., Zainudin, E., and Dahlan, K. (2012). "The flexural, impact and thermal properties of untreated short sugar palm fibre reinforced high impact polystyrene (HIPS) composites," *Polym. Polym. Compos.* 20(5), 493. DOI: 10.1007/s12221-012-0894-1
- Duc, F., Bourban, P. E., Plummer, C. J. G., and Månson, J. A. E. (2014). "Damping of thermoset and thermoplastic flax fibre composites," *Compos. Part A: Appl. Sci. Manuf.* 64, 115-123. DOI:10.1016/j.compositesa.2014.04.016.
- Essabir, H., Bensalah, M. O., Rodrigue, D., Bouhfid, R., and Qaiss, A. (2016). "Structural, mechanical and thermal properties of bio-based hybrid composites from waste coir residues: fibers and shell particles," *Mech. Mater.* 93, 134-144. DOI: 10.1016/j.mechmat.2015.10.018
- Fiore, V., Di Bella, G., and Valenza, A. (2015). "The effect of alkaline treatment on mechanical properties of kenaf fibers and their epoxy composites," *Compos. Part B-Eng.* 68, 14-21. DOI: 10.1016/j.compositesb.2014.08.025
- Goriparthi, B. K., Suman, K., and Rao, N. M. (2012). "Effect of fiber surface treatments on mechanical and abrasive wear performance of polylactide/jute composites," *Compos. Part A-Appl. S.* 43(10), 1800-1808. DOI: 10.1016/j.compositesa.2012.05.007
- Ishak, M. R., Sapuan, S. M., Leman, Z., Rahman, M., Anwar, U., and Siregar, J. (2013). "Sugar palm (*Arenga pinnata*): Its fibres, polymers and composites," *Carbohydr. Polym.* 91(2), 699-710. DOI: 10.1016/j.carbpol.2012.07.073
- Isma'ila, M., Leman, Z., Ishak, M. R., and Zainudin, E. S. (2016). "Sugar palm fibre and its composites: a review of recent developments," *BioResources* 11(4), 10756-10782. DOI: 10.15376/biores.11.4.10756-10782
- Jawaid, M., Khalil, H. A., Hassan, A., Dungani, R., and Hadiyane, A. (2013). "Effect of jute fibre loading on tensile and dynamic mechanical properties of oil palm epoxy composites," *Compos. Part B-Eng.* 45(1), 619-624. DOI: 10.1016/j.compositesb.2012.04.068
- Joseph, S., Sreekala, M., Oommen, Z., Koshy, P., and Thomas, S. (2002). "A comparison of the mechanical properties of phenol formaldehyde composites reinforced with banana fibres and glass fibres," *Compos. Sci. Technol.* 62(14), 1857-1868. DOI: 10.1016/S0266-3538(02)00098-2
- Jumaidin, R., Sapuan, S. M., Jawaid, M., Ishak, M. R., and Sahari, J. (2017). "Effect of seaweed on mechanical, thermal, and biodegradation properties of thermoplastic

- sugar palm starch/agar composites," *Int. J. Biol. Macromolec.* DOI: 10.1016/j.ijbiomac.2017.02.092
- Kabir, M., Wang, H., Lau, K., and Cardona, F. (2012). "Chemical treatments on plant-based natural fibre reinforced polymer composites: An overview," *Compos. Part B-Eng.* 43(7), 2883-2892. DOI: 10.1016/j.compositesb.2012.04.053
- Kumar, S. M. Suresh, Duraibabu, D., and Subramanian, K. (2014). "Studies on mechanical, thermal and dynamic mechanical properties of untreated (raw) and treated coconut sheath fiber reinforced epoxy composites," *Mater. Des.* (59), 63-69. DOI: 10.1016/j.matdes.2014.02.013.
- Leman, Z., Sapuan, S., Azwan, M., Ahmad, M., and Maleque, M. (2008). "The effect of environmental treatments on fiber surface properties and tensile strength of sugar palm fiber-reinforced epoxy composites," *Polym. Plast. Technol. Eng.* 47(6), 606-612. DOI: 10.1080/03602550802059451
- Manoharan, S., Suresha, B., Ramadoss, G., and Bharath, B. (2014). "Effect of short fiber reinforcement on mechanical properties of hybrid phenolic composites," *J. Mater.* 2014, 478549. DOI: 10.1155/2014/478549
- Milanese, A. C., Cioffi, M. O. H., and Voorwald, H. J. C. (2012). "Thermal and mechanical behaviour of sisal/phenolic composites," *Compos. Part B-Eng.* 43(7), 2843-2850. DOI: 10.1016/j.compositesb.2012.04.048
- Rashid, B., Leman, Z., Jawaid, M., Ghazali, M. J., and Ishak, M. R. (2016a). "The mechanical performance of sugar palm fibres (ijuk) reinforced phenolic composites," *Int. J. Precis. Eng. Manuf.* 17(8), 1001-1008. DOI: 10.1007/s12541-016-0122-9
- Rashid, B., Leman, Z., Jawaid, M., Ghazali, M. J., and Ishak, M. R. (2016b). "Physicochemical and thermal properties of lignocellulosic fiber from sugar palm fibers: Effect of treatment," *Cellulose* 23(5), 2905-2916. DOI: 10.1007/s10570-016-1005-z
- Rashid, B., Leman, Z., Jawaid, M., Ghazali, M., and Ishak, M. (2017a). "Effect of treatments on the physical and morphological properties of SPF/phenolic composites," *J. Nat. Fibers* 1-13. DOI: 10.1080/15440478.2016.1266291
- Rashid, B., Leman, Z., Jawaid, M., Ghazali, M. J., and Ishak, M. R. (2017b). "Influence of treatments on the mechanical and thermal properties of sugar palm fibre reinforced phenolic composites," *BioResources*, 12(1), 1447-1462. DOI: 10.15376/biores.12.1.1447-1462
- Rashid, B., Leman, Z., Jawaid, M., Ghazali, M. J., Ishak, M. R., and Abdelgnei, M. A. (2017c). "Dry sliding wear behavior of untreated and treated sugar palm fiber filled phenolic composites using factorial technique," *Wear*. DOI: 10.1016/j.wear.2017.03.011
- Ray, D., Sarkar, B. K., Das, S., and Rana, A. K. (2002). "Dynamic mechanical and thermal analysis of vinylester-resin-matrix composites reinforced with untreated and alkali-treated jute fibres," *Compos. Sci. Technol.* 62, 911-917. DOI: 10.1016/S0266-3538(02)00005-2.
- Ridzuan, M., Majid, M. A., Afendi, M., Mazlee, M., and Gibson, A. (2016). "Thermal behaviour and dynamic mechanical analysis of Pennisetum purpureum/glass-reinforced epoxy hybrid composites," *Compos. Struct.* 152, 850-859. DOI: 10.1016/j.compstruct.2016.06.026

- Saba, N., Paridah, M., and Jawaid, M. (2015). "Mechanical properties of kenaf fibre reinforced polymer composite: A review," *Constr. Build. Mater.* 76, 87-96. DOI: 10.1016/j.conbuildmat.2014.11.043
- Saba, N., Jawaid, M., Alothman, O. Y., and Paridah, M. (2016a). "A review on dynamic mechanical properties of natural fibre reinforced polymer composites," *Constr. Build. Mater.* 106, 149-159. DOI: 10.1016/j.conbuildmat.2015.12.075
- Saba, N., Paridah, M., Abdan, K., and Ibrahim, N. (2016b). "Dynamic mechanical properties of oil palm nano filler/kenaf/epoxy hybrid nanocomposites," *Constr. Build. Mater.* 124, 133-138. DOI: 10.1016/j.conbuildmat.2016.07.059
- Shalwan, A., and Yousif, B. (2013). "In state of art: Mechanical and tribological behaviour of polymeric composites based on natural fibres," *Mater. Des.* 48, 14-24.
- Shinoj, S., Visvanathan, R., Panigrahi, S., and Varadharaju, N. (2011). "Dynamic mechanical properties of oil palm fibre (OPF)-linear low density polyethylene (LLDPE) biocomposites and study of fibre-matrix interactions," *Biosyst. Eng.* 109(2), 99-107. DOI: 10.1016/j.biosystemseng.2011.02.006
- Sonia, A., Dasan, K. P., and Alex, R. (2013). "Celluloses microfibrils (CMF) reinforced poly (ethylene-co-vinyl acetate) (EVA) composites: Dynamic mechanical, gamma and thermal ageing studies," *Chem. Eng. J.* (228), 1214-1222. DOI: 10.1016/j.cej.2013.04.091.
- Sreekala, M., Thomas, S., and Groeninckx, G. (2005). "Dynamic mechanical properties of oil palm fiber/phenol formaldehyde and oil palm fiber/glass hybrid phenol formaldehyde composites," *Polym. Compos.* 26(3), 388-400. DOI: 10.1002/pc.20095
- Surojo, E., Malau, V., and Ilman, M. (2014). "Effects of phenolic resin and fly ash on coefficient of friction of brake shoe composite," *J. Eng. Appl. Sci.* 9(11), 2234-2240.

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