

Improvements in the thermal behaviour of date palm/bamboo fibres reinforced epoxy hybrid composites

M. Jawaid^{*a}, Sameer Awad^b, H. Fouad^c, M. Asim^a, N. Saba^a, Hom N. Dhakal^d

^aLaboratory of Biocomposite Technology, Institute of Tropical Forestry and Forest Products (INTROP) Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia.

^bChemistry, School of Science and Technology, University of New England, Armidale NSW 2351, Australia

^cApplied Medical Science Department, Community College, King Saud University, P.O. Box 10219, Riyadh 11433, Saudi Arabia

^dAdvanced Polymers and Composites (APC) Research Group, University of Portsmouth, School of Mechanical and Design Engineering, Anglesea Road, Anglesea Building, PO1 3DJ, UK

^{*}Email: jawaid@upm.edu.my

ABSTRACT

Natural fibre-reinforced epoxy composites have been increasingly utilised in construction and building applications. These materials introduce cost-effective alternatives to conventional materials and their utilisation often has related to financial advantages that are immediate and can be expected over the structure service life. In this work, the results of thermal, dynamic-mechanical, and thermal-mechanical properties of date palm fibre (DPF)/bamboo fibre (BF) hybrid composite were compared to single date palm fibre-reinforced epoxy and single bamboo fibre-reinforced epoxy to demonstrate the importance of hybridization. The thermal stability was improved when DPF/BF fillers were added in epoxy resin comparatively to DPF reinforced

epoxy and BF reinforced epoxy. The glass transition temperature (T_g) was increased by incorporating the date palm fibre/bamboo hybrid composite in epoxy compared to the BF reinforced epoxy. The thermal expansion was enhanced by modifying the hybrid composites in epoxy in contrast to the single fibre composites, without hybridisation. Hence, the hybridisation technique of date palm fibre with bamboo has improved the thermal and thermal-mechanical properties suitable for several applications including non-structural applications.

Keywords: Thermal characterisations, Hybrid composite, Date palm fibre, Thermal stability.

1. Introduction

Natural fibre-reinforced polymer composites are considered as significant composite competitor materials to synthetic fibre reinforced conventional composites due to a number of attractive attributes including efficient specific stiffness, high strength and lightweight [1]. Currently, agricultural by-products or agricultural-waste materials are mainly utilised as reinforcements which include most of the cereal stalks, rice husks, bagasse, coconut fibres, corn cobs, nutshells and other wastes that might alleviate the defined shortage [2]. The renewable resources from natural fibres provide environmental advantages relating to the utilisation of ultimate disposability and raw material [1,3]. In addition, natural fibres reinforced composites present advantages for biodegradability, great accessibility, fast renewability, desirable properties with the low cost [4, 5]. On the other hand, understanding the detailed morphology, structural, and mechanical properties of natural fibres is important for exploiting their benefits as reinforcing materials in composites and for optimizing composites' service life performance [6,7].

Natural plant fibres, in particular, bamboo fibres have undoubtedly contributed to achieving excellent economic outcome and sustainability in our daily lives for various manufacturing

purposes such as textiles and construction [8]. A recent renewed interest in BF is primarily aimed at replacing or decreasing the use of non-renewable glass fibres as feedstock in the composite structures [9].

Bamboo fibres with low density (1.4 g/cm^3) exhibit an excellent mechanical properties and exhibit superior specific stiffness and bending load strength than synthetic glass fibres [10-13]. Traditionally, bamboo has been used as lightweight lattice platforms in building and construction sector due to their high modulus and strength

The fibres of date palm (*Phoenix dactylifera* L.) considered as one of the greatest accessible types of natural fibres worldwide [14, 15]. Large amounts of date palm biomass wastes are collected annually without appropriate uses. Hence, the potential interest of these quantities is to be used to support the demand of renewable feedstock in composite sector and encourage industrial sustainability by supplying alternative cheap and eco-friendly materials [16].

Natural fibre reinforced hybrid composites have great features as this approach takes the advantage of synergic effects of both hybrid reinforcements into a single matrix. In this manner, every individual material (fibre and matrix) maintains its physical and chemical characterisations, which they present that would not be conceivable if used singly. In this point, the factors of the central load-transferring have represented the fibres while the matrix includes them in orientation alongside, forming as a part of load-transferring between them, preventing the fibres from the extreme natural environments such as humidity and raised temperature [17-20]. The evolution of hybrid composite materials studies has been extensively concealed from many kinds of fibre and matrix resin, such as synthetic-synthetic fibre, natural-natural fibres, and synthetic-natural fibres properties or synergistic effects of two or more reinforcing elements [3,4, 21].

The study carried out by Bahari and Krause [22] on bamboo fibre reinforced PVC thermoplastic composites has concluded that bamboo composites have a significant potential to be used as alternative fillers to develop sustainable biobased composites with greener and cleaner credentials.

The modifications in physical, mechanical, and thermal properties of thermoplastics and thermosets polymers reinforced DPF filler have been made by several attempts. However, the modification of the surface before reinforcement, usually, is required to purify and clean the fibre surface from many impurities and incomplete growths because they may cause poor adhesion between polymer and fibre. One of the studies reported better morphology and mechanical properties which was improved of polyester and epoxy matrices by incorporating the short DPF filler [23]. Largely, recycled virgin thermoplastic polymers are widely used as matrices with the filler of DPF [24]. For example, a group of researchers investigated the sorption, mechanical, sorption, and adhesion properties of wood powder from date palm /low-density polyethylene composites (LDPE) [25]. Furthermore, Asim et al. [26] reported an improvement in the mechanical (flexural, tensile and impact), morphological, and dynamic-mechanical properties of different date palm fibres incorporated as fillers with phenolic composites. They observed that the tensile modulus, and impact properties improved by incorporating 50% of DPF in the polymer matrix while the tensile strength, flexural strength and modulus were decreased.

This study has evaluated the improvements between date palm fibre and bamboo fibre which reinforced with an epoxy polymer via a hybridization technique. From the hybridization technique, the agro-waste residue from the date palm tree could minimise the environmental and health issues. With the hand-lay-up process fabrication, the date palm fibre/bamboo hybrid composite has exhibited a significant improvement of thermal stability and thermal-mechanical

characteristics when compared to single date palm fibre composite without hybridisation. Besides, date palm fibre/bamboo hybrid composite has shown a favourable attribute such as eco-friendlier and low-cost sustainable materials for non-load-bearing products.

2. Materials and Methods

2.1. Materials

The date palm/bamboo fibre reinforced epoxy hybrid composites were prepared with a mixture of short date palm fibres (DPF) sized from (0.8 – 1 mm), supplied from (King Abdul Aziz University, Jeddah, Saudi Arabia). The extraction of date palm fibre tree part was taken from date palm which is classified as following; leaf stalk (A), fruit bunch stalk (AA), leaf sheath (G) and tree trunk (L). Meanwhile, bamboo fibre was provided by Shijiangzhuang Bi Yang Technology Co. Ltd. Hebei, China (Table 1). Moreover, the matrix resin utilised in this fabrication process of date palm/bamboo hybrid was epoxy resin type D.E.R.TM 324 with epoxy hardener (Jointmine 905-3S Tazdiq Engineering Sdn supplied. Bhd., Selangor, Malaysia), the physical and mechanical properties of the epoxy and hardener which used in this work are summarised in Table 2.

Table 1. Chemical compositions of date palm fibre (DPF) and bamboo fibre [27,28,24].

Fibre Specimen	DPF - Leaf stalk (A)	DPF - Fruit bunch stalk (AA)	DPF - Leaf-sheath fibre (G)	DPF - Trunk fibre (L)	Bamboo fibre
Cellulose (%)	35.00	44.00	43.50	40.00	73.83

Hemicellulose (%)	15.40	26.00	24.00	9.75	12.49
Lignin (%)	20.10	11.00	18.00	29.50	10.15

Table 2. Epoxy resin type D.E.R.TM 324 [29] and Jointmine 905-3S properties [30].

Epoxy: D.E.R.TM 324	Density 25°C (g/ml)	Binder gel time (100 g) (min)	Tensile strength (MPa)	Flexural strength (MPa)	Compressiv e strength (MPa)
	1.16	24	13.7	31.4	87.9

Jointmine 905-3S	Amine Value (mg KOH/g)	Viscosity (25°C, cPs)	Colour (Gardn er)	Pot life(mins)	Type	Properties / Applications
	330 ± 20	200 ~ 400	<3	60 ~ 70	Cycloalipha tic Amine Adduct	Solvent-free coatings, architectural & maintenance coatings, pipe & tank lining

2.2. Fabrication of date palm/bamboo hybrid composites

Three main types of reinforced epoxy specimen were fabricated with the hand lay-up technique. In contrast, three categories were identified, such as date palm fibre composite, bamboo fibre composite and DPF/BF hybrid date palm/bamboo composite. As per categories of date palm composite (*A*, *AA*, *G*, *L*); meanwhile, bamboo mat (*B*) was categorised under single specimen and hybrid date palm/bamboo composite includes a mixture of date palm fruit leaf stalk/bamboo (*A/B*), date palm fruit bunch stalk/bamboo (*AA/B*), date palm leaf sheath/bamboo (*G/B*) and date palm tree trunk/bamboo (*L/B*). The varieties of date palm fibre/bamboo composite are due to the different chemical composition of the extracted date palm fibre (cellulose, hemicellulose and lignin) vary in yields, as displayed in Table 1 [24].

The composite specimens were fabricated using hand lay-up technique. Fabrication process comprised of three important stages, which the initial stage was the preparation of raw material such as fibre materials, where the fibres were cleaned, washed, and dried at 60°C for 24 hours to reduce moisture content until the 6–8%. The next stage was preparing steel cast mould designated with the size 150mm x 150mm x 3mm. Furthermore, in this stage, the mixture of fibres and resin with hardener was stirred at room temperature with a stoichiometric ratio of resin and hardener ratio is 2:1, respectively. Subsequently, in the final stage, the mixture was poured evenly into the steel cast mould with 50:50 of weight ratio of fibre and epoxy with total fibre loading of 50 wt.% and moved to the hot press machine and heated for 10 min at a temperature of 110°C and demould with cold press machine after 10 min cooled down. Afterwards, the samples were eliminated and cut into specific replicate size according to each standard size of experiments. Table 3 illustrates the formulations of fibres and epoxy resin composites.

Table 3. The formulation ratios of composites

Composites	Epoxy resin (wt%)	DPF (wt%)	BF (wt%)
<i>A/B</i>	50	25	25
<i>AA/B</i>	50	25	25
<i>B</i>	50	50	0
<i>G/B</i>	50	25	25
<i>L/B</i>	50	25	25

3. Characterisation of Composites

3.1. Dynamic mechanical analysis (DMA)

The viscoelastic behavior of composites was performed using a three-point bending mode DMA (TA Q-800 instrument) as a temperature function. The test was performed according to the standard of American Society for Testing and Materials (ASTM)- D4065-01, and the samples were prepared accordingly. The DMA tests were conducted in a nitrogen atmosphere at an oscillation frequency of 1 Hz and the temperature range is between 30°C to 200°C, a heating rate of 5°C/min.

3.2. Thermogravimetric analysis (TGA)

The thermal stability of all composite samples was tested by thermo-gravimetric analysis (TGA), using TGA Q500 V20.13 Build 39. The TGA analysis was performed under a nitrogen atmosphere at a 50 ml/min flow rate to avoid oxidation. The composite samples weighed (8–

8.5 mg). The heating rate was kept at 20°C/min as the samples were heated to the temperature of (30 to 700 °C).

3.3. Thermo-mechanical analysis (TMA)

TMA tests were performed with TMA Q400 V22.5 Build 31 setup controlling in the penetration mode at a heating rate of 5°C/min from 30 °C to 200°C under nitrogen atmosphere to identify the coefficient of thermal expansion (CTE) value of composites.

4. Results and Discussion

4.1. Thermogravimetric analysis (TGA)

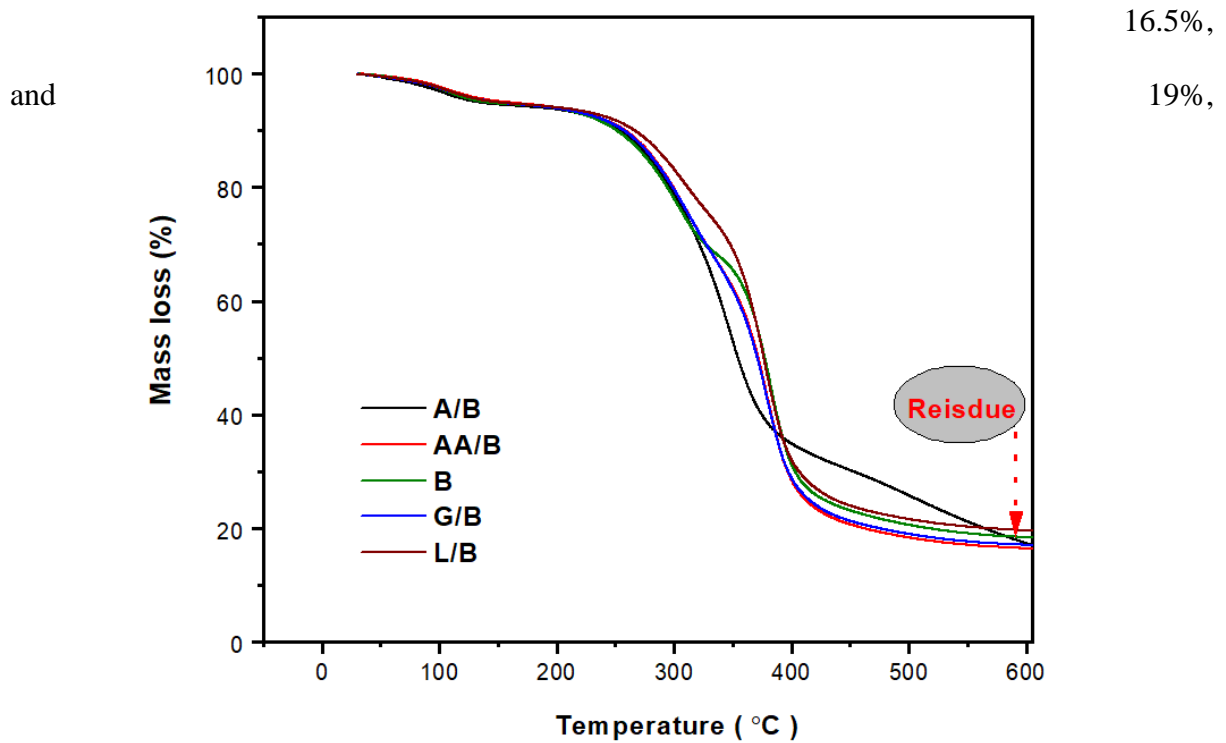
The thermal stability of the composites was identified based on temperatures corresponding to 50%, in addition to mass loss was being taken from the thermogravimetric (TG) curves. The obtained values from the mass loss (TG) and derivatives of thermogravimetric (DTG) curves are presented in Table 4 while the graphs are shown in [Figs.1 and 2](#).

Thermal degradation of sample up to 150°C is a result of the moisture content is shown in the sample [\[31\]](#). For instance, the decomposition temperatures (T50%) of samples (AA/B), (A/B), (B), (G/B) and (L/B) are about 264.0°C, 290.0°C, 255.8°C, 264.0°C, and 263.7°C, respectively, as shown in Table 4. However, the thermal decompositions of the (A/B) sample showed a significant increase in thermal decomposition in comparison to other samples. The degradation between 250 to 300°C is due to the decomposition of the cellulose and hemicelluloses present in fibre [\[32\]](#).

The maximum thermal decomposition of all composites was slightly increased and improved after incorporated the date palm fibre and palm fibre in epoxy resin. The maximum thermal decomposition (T_{max}) was 382°C, 347°C, 381°C, 379°C, and 379°C, respectively, as displayed

in Fig. 2 and Table 4. The final degradation showed the lignin of natural fibres has been decomposed [33].

The result showed that the residual amount of date palm fruit bunch stalk/bamboo (AA/B), date palm fruit leaf stalk/bamboo (A/B), hybrid date palm/bamboo composite (B), date palm leaf sheath/bamboo (G/B) and date palm tree trunk/bamboo (L/B) samples are 16%, 13.6%, 18%,



respectively (Table 4). The results are illustrated in Table 4 showed improvements in thermal stability of hybrid composites, however; the thermal decomposition at the maximum temperature showed no significant changes.

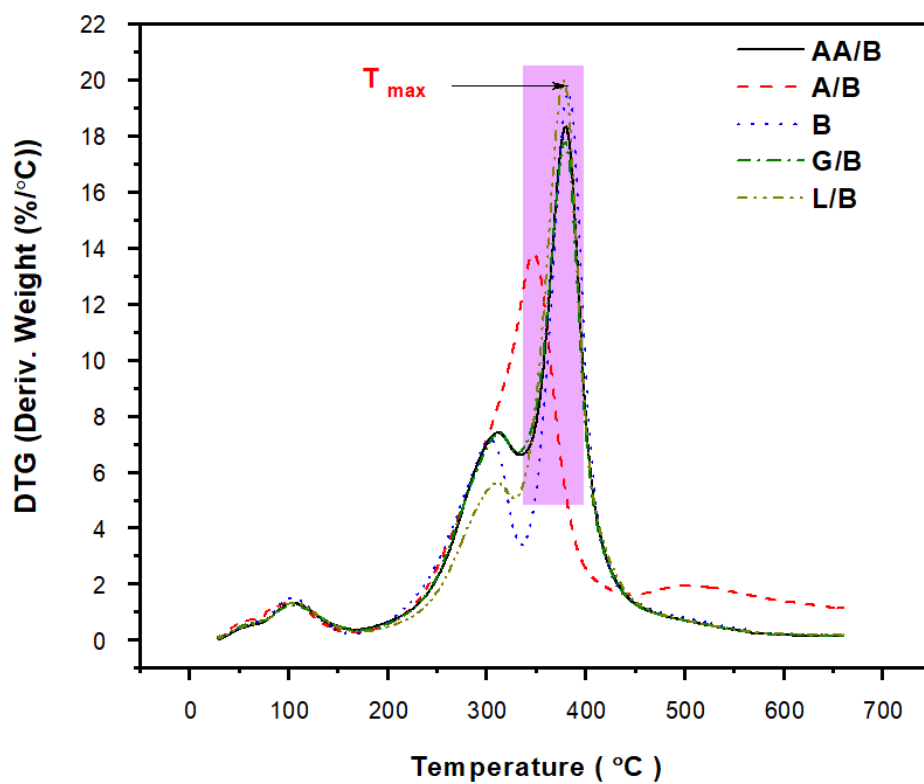


Fig. 2. Derivative thermogravimetric (DTG) curves for composites.

Composites	$T_{50\%}$ (°C)	T_{max}	Residue %
AA/B	264.0	382	16.0
A/B	290.0	347	13.6
B	255.8	381	18.0
G/B	264.0	379	16.5

Table	L/B	263.7	379	19	4.
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Therma data from TGA and DTG curves

4.2. Dynamic-mechanical Analysis (DMA) of hybrid composites

The DMA results are shown in Figs. 3, 4, and 5. The incorporation of date palm fibre and bamboo fibre in epoxy resins increased the storage modulus at room temperature. Fig. 3 shows the storage modulus (E') versus the temperature of composites. From the graph, it is seen that the samples (AA/B) and (L/B) showed the maximum values of E' (~3250 MPa and 3218 MPa), respectively. On the other hand, the storage modulus values of samples (A/B, B, G/B) were 3087.2 MPa, 3084.5 MPa, and 3109.8 MPa, respectively. Higher storage modulus attribute to the increased stiffness sample restricted molecular mobility of epoxy chains and providing greater bonding between the epoxy matrix and reinforcement [32, 34].

Fig. 4 displays the curves of modulus loss, E'' of all composites and hybrid composites versus temperature at the temperature range from 30 up to 200 °C. Loss modulus describes the viscous nature of materials along with different molecular motion types, morphology, transitions relaxation processes and structural heterogeneities [35, 36].

The E'' results in Fig.4 reveal an improvement in reducing the loss of modulus with the incorporation of DPF and BF fillers, which enhances the reinforcement impact owing to its better interaction with the polymer matrix. The incorporation of composite fibre (BF and DPF) into the epoxy matrix shifts the peak of loss modulus to a higher temperature. As shown in Fig.4 and Table 4, the loss modulus values were 349.2 MPa, 355.4 MPa, 332 MPa, 351.4 MPa, and 352.3 MPa for composites (AA/B), (A/B), (B), (G/B) (L/B) samples, respectively.

Damping is the ratio of loss modulus to the storage modulus which can be determined by getting a phase tangent variation between sinusoidal stress and strain [37]. Figure 5 illustrates

the change in the damping of all composite samples as a temperature function at the frequency (1 Hz). The minimum values of $\text{Tan } \delta$ were observed for hybrid composites in epoxy resin in comparison to the single fibre composites incorporated with epoxy, which exhibited maximum values, as shown in [Table 5](#). Comparative results also stated where the improvement of epoxy with cellulose nanofibre exhibited a minimal value than that of the epoxy resin. From the $\text{Tan } \delta$ plot, it was also evident that the damping increases with increasing the temperature and reach its maximum in the region of transition and then begins in reducing towards the rubbery region [\[37\]](#).

The glass transition temperature (T_g) of all composites was determined at a peak of $\text{tan } \delta$ (damping factor) against temperature curves as illustrated in [Fig. 5](#) while [Table 5](#) is exhibited detailed information about the values of T_g for all samples. The sample (A/B) exhibited higher T_g value (79.46°C) with higher damping factor ($\text{Tan } \delta$; 0.36) than all other composites, as shown in [Table 5](#). The incorporation of hybrid composite (DPF/BF) in epoxy exhibits an increase in glass transition temperature of hybrid fibre-epoxy composites in comparison to single fibre-epoxy composites.

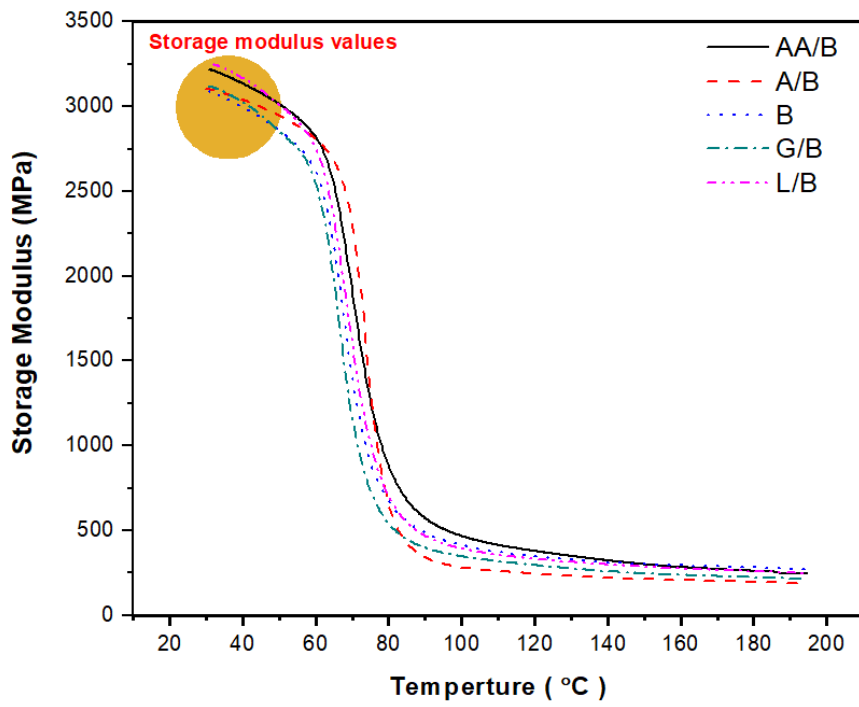


Fig. 3. Storage Modulus vs temperature of date palm, date palm/bamboo and bamboo composites

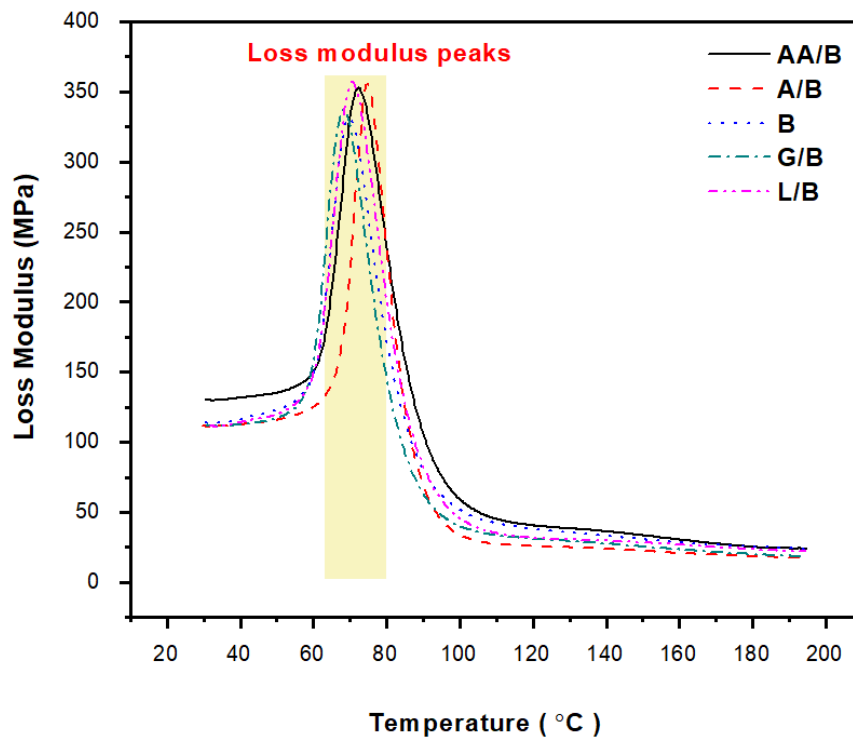


Fig. 4. Loss modulus vs temperature of date palm, date palm/bamboo and bamboo composites

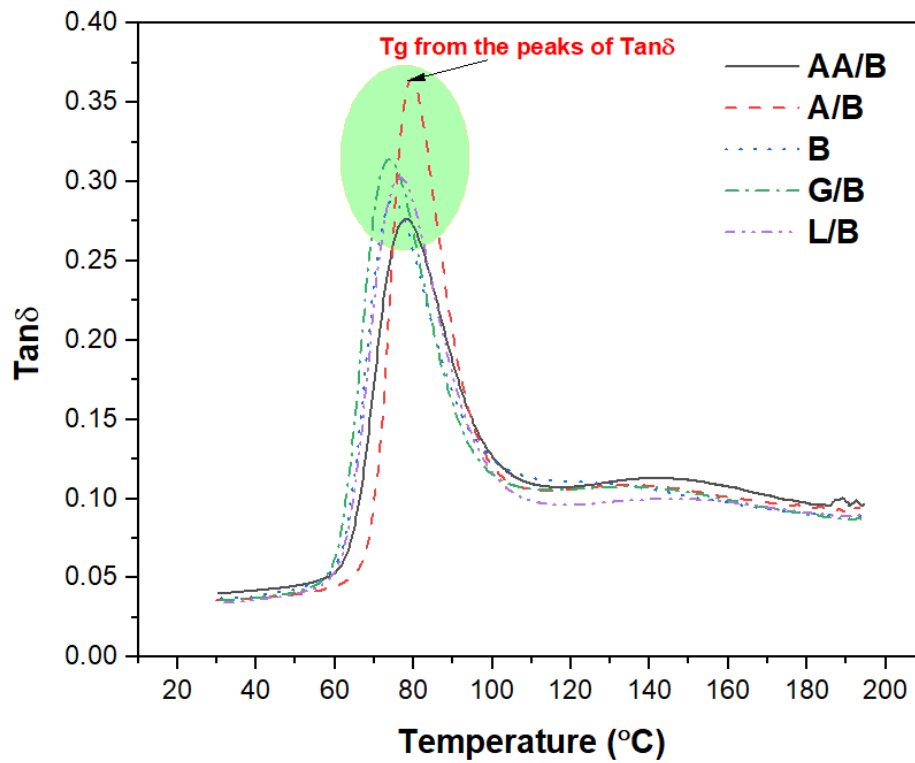


Fig. 5. Tan δ vs temperature of date palm, date palm/bamboo and bamboo composites

Table 5. DMA results of date palm, date palm/bamboo and bamboo composite fibre

Composites	Storage Modulus (MPa)	Loss Modulus (MPa)	Tan δ	T _g from Tan δ (°C)
AA/B	3218.4	349.2	0.27	78.02
A/B	3087.2	355.4	0.36	79.47
B	3084.5	331.6	0.29	74.83
G/B	3109.8	351.4	0.31	73.87
L/B	3249.8	352.3	0.30	76.56

4.3. Thermo-Mechanical Properties (Thermal Expansion)

Thermal expansion of samples was evaluated by utilising a thermo-mechanical analyzer (TMA). Fig.6 shows the dimensional change as a function of temperature of fibres composites. Thermomechanical analyses of the date palm fibre/bamboo hybrid composite and to single date palm fibre-reinforced epoxy and single bamboo fibre-reinforced epoxy were performed by identifying in both areas the coefficient of thermal expansion and dimensional changes [38]. Through composite fabrication, quenching and stretching formed interior stress in the composites. Within the analysis, the external load is operated on a specimen in an axial direction with temperature, the porosity of the specimen begins to collapse, and the specimen indicated three stages of deformities. From the results are shown in Table 6, the composite samples (AA/B) exhibited a higher coefficient of thermal expansion ($199.5 \mu\text{m} / \text{m}.\text{°C}$) in comparison to other samples, which were 135.7, 92.3, 194.9, and $196.6 \mu\text{m} / \text{m}.\text{°C}$ for hybrid composite (A/B), the bamboo fibre composite (B), hybrid composite (G/B), and hybrid composite(L/B), respectively.

The values of T_g for the pure epoxy and composites appear between 60°C and 62°C , as shown in Table 6. Generally, T_g was decreased slightly because of the incorporation of DPF and BF into the epoxy matrix. The values of hybrid composite (AA/B) exhibited a higher thermal expansion coefficient ($199.5 \mu\text{m}/\text{m}.\text{°C}$), in comparison to other samples, which were 135.7, 92.3, 194.9, and $196.6 \mu\text{m}/\text{m}.\text{°C}$ for hybrid composite (A/B), the bamboo fibre composite (B), hybrid composite (G/B), and hybrid composite(L/B), respectively. In the polymers, the CTE above the T_g is relying on the expansion of free volume, which occurs due to volumetric changes in the polymer under the thermo-mechanical convection. the free volume expansion above the T_g decreases the density, increased the mobility of chain and the rise in the number of conformational changes within the epoxy matrix chains [39].

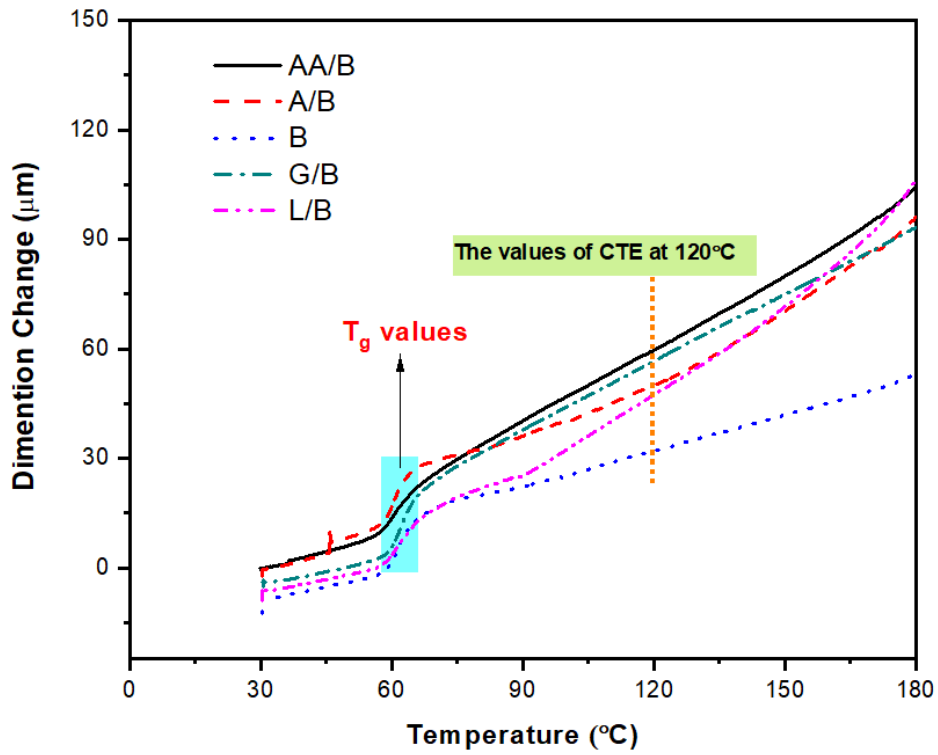


Fig.6. TMA results showing dimensional changes as a function of temperature

Table 6. CTE and T_g values from DMA data of all hybrid composites

Composite	CTE ($\mu\text{m} / \text{m} \cdot ^\circ\text{C}$)	T_g ($^\circ\text{C}$)
AA/B	199.5	60.30
A/B	135.7	60.26
B	92.3	61.23
G/B	194.9	61.95
L/B	196.6	61.20

5. Conclusions

In this research work, an investigation has been made to reinforce stiffer bamboo fibre as a hybrid composite filler to modify the thermal and mechanical-thermal characteristics of date palm fibre with epoxy resin. To investigate the advantages of bamboo fibre into date palm fibre waste residue, the investigation of thermal stability and mechanical-thermal properties was assessed with the effect of a variety of date palm fibre type of leaf stalk, fruit bunch stalk, leaf sheath fibre and trunk fibre. The results of the hybrid composite were comparable with a single bamboo fibre composite, furthermore, the contrast characterization of natural fibre found from various categories of the date palm tree with bamboo fibre have significantly improved the thermal stability and mechanical-thermal characteristics. The hybrid fibre composites incorporated epoxy matrix exhibited better thermal stability than single fibre composites. The dynamic mechanical properties improved with hybrid fibre composites reinforced epoxy in comparison to single fibre composites without hybridisation. Thus, the conclusion from the tests of this study stimulates the potential of DPF when hybridising with other excellent fibre filler to make lightweight material suitable for various applications. In addition, these materials provide significant advantages in building and construction industry.

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Data Availability

The raw/processed data required to reproduce these findings cannot be shared at this time as the data also forms part of an ongoing study.

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