



Physical and damping properties of kenaf fibre filled natural rubber/thermoplastic polyurethane composites

A.M. Noor Azammi ^{a, b}, S.M. Sapuan ^{a, c, *}, Mohamad R. Ishak ^d, Mohamed T.H. Sultan ^{c, d}

^a Advanced Engineering Materials and Composite Research Centre (AEMC), Department of Mechanical and Manufacturing Engineering, University Putra Malaysia, 43400, UPM Serdang, Selangor, Malaysia

^b Automotive Engineering Technology Section, Universiti Kuala Lumpur Malaysian France Institute (UniKL-MFI), 43650, Bandar Baru Bangi, Selangor, Malaysia

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

^d Department of Aerospace Engineering, Universiti Putra Malaysia, 43400, UPM Serdang, Selangor, Malaysia

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ABSTRACT

The paper presents the investigation of the effect of alkaline treatment of sodium hydroxide (NaOH) on physical and dynamic mechanical analysis (DMA) viscoelastic properties of kenaf fibre filled natural rubber (NR)/thermoplastic polyurethane (TPU) composites. The treated kenaf fiber, NR and TPU were weighed and proportioned according to the required compositions and were blended using hot mixed Brabender machine. The polymer composites were then fabricated using the hot press to form a sample board. The sample was cut and prepared and water absorption, density, thickness swelling and DMA tests were performed. As far as physical properties are concerned, composites with the highest NR amount of shows the best results, which indicates good fiber bonding adhesion. The polymer composites with the highest amount of TPU shows the highest damping properties at high temperature.

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1. Introduction

Natural fibers had been used as reinforcements for the polymer composites to produce environmentally friendly materials, which could be disposed of easily and harmless to the environment. Natural fibers can act as reinforcements or fillers in composites and are able to improve their mechanical properties [1–4]. In the past, the investigation utilizing kenaf as strengthening composites using kenaf fiber as reinforcing filler in the composites [5–8]. Kenaf tree can reach a height of 3–4 m within 4–5 months. Kenaf tree consists of 3 layers, namely core, pith and bast of which one third kenaf tree consists of bast. Kenaf bast fibre is said to have mechanical strength than other parts [9]. Recent research regarding the strengthening of the composite was made using kenaf fibre [10]. After the kenaf fibers were treated, the mechanical properties and biodegradability

of the fibers can be determined after they were combined with polymers such as rubber and polyester to form composites, which reduced the fiber hydrophilicity in the production of composites [11]. Kenaf fiber had also the potential to reinforce other thermoset and thermoplastic polymers if the fibers were compounded with maleated polypropylene/maleic acid polyethylene (MAPP/MAPE) [12–16]. Kenaf fibers had been treated with 6% of sodium hydroxide to improve the tensile properties or the bonding between the fiber and rubber [12].

Natural rubber is a biopolymer material that is largely used in producing tyres, rubber gloves and engine mountings. Natural rubber was used in engine mounting because of its excellent combined properties such as high strength, fatigue resistance, high resilience, low sensitivity to strain effect in dynamic effect and well resistant to creep [17]. The properties of natural rubber allowed the material to be blended with other materials, which created very low modulus, low damping and high strength [18,19]. A requirement of an engine rubber mount beside the rigidity of the support structure, the strength and the design geometry are also needed to be considered [20]. The thermoplastic polyurethane characteristic is resistant to wear, have a good mechanical and rubber-like elastic

* Corresponding author. Advanced Engineering Materials and Composite Research Centre (AEMC), Department of Mechanical and Manufacturing Engineering, University Putra Malaysia, 43400, UPM Serdang, Selangor, Malaysia.

E-mail address: sapuan@upm.edu.my (S.M. Sapuan).

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and tear resistant [20–22]. Hopefully with the blend of kenaf-TPU and NR will improve its durability compared to the NR composites only. In this paper, the effect of treated kenaf fiber filled natural rubber/thermoplastic polyurethane composites towards the physical and damping properties which are required in developing engine rubber mount.

2. Materials and methods

2.1. Materials

Kenaf fibers, thermoplastic polyurethane (TPU) and sodium oxide (NaOH) were supplied by Innovative Pultrusion Sdn. Bhd., Seremban, Malaysia. Latex rubber was supplied by Getahindus (M) Sdn Bhd, Tangkak, Johor, Malaysia (Table 1). The type of TPU used was Estane 58311 and the specifications are shown in Table 2 [23]. Kenaf fiber was initially shieved and the kenaf fibres in the range of 300–500 μm were used. The latex rubber was initially solidified into thinned rectangle slab and cut into small cubic shapes [24].

2.2. Sample preparation

Kenaf fibres were soaked for 24 h in the 6% of sodium hydroxide (NaOH) solution for treatment. After that, the treated kenaf fibres were washed and let dry at 100 °C for 24 h. The kenaf fibers were treated by 6% of sodium hydroxide to improve the tensile properties and the bonding between the fibers and rubber [23,25,26]. All materials were weighed and mixed in the ratio shown in Table 3.

The mixed TPU/NR composites were pulverized into small cubic shape. The pulverized TPU/NR composites were pressed using a 40-tonne hot press machine and shaped into sample board in the sizes of 3 mm \times 150 mm \times 150 mm. After the sample board was ready, it was cut based on the testing specimen requirements.

2.3. Determination of water absorption

Ten specimens of 20 mm \times 20 mm \times 3 mm of size were prepared according to ASTM D570 from developed composites. Ten samples of each type of composites were prepared for the testing of thickness swelling. All samples were oven dried at 60 °C for 24 h. After oven drying, the samples were cooled in desiccators over granulated silica gel before water absorption was determined. Water absorption test was conducted by submerging the specimens in distilled water for 7 days and measured the increase in weight as compared to the original oven dry weight of the specimen. The weight measurement was performed using the weighing machine. Three specimens of each formulation were tested in an adjusted room (23 °C and 50% RH) and the results were averaged. Manual

Table 2
TPU specifications.

Physical Properties	Value	Metric
Hardness (5 s) Shore A	85 \pm 3	
Specific Gravity	1.13	
Tensile Strength	6600 psi	45.5 MPa
Ultimate Elongation	520%	
Tensile Stress at 100% Elongation	900 psi	6.2 MPa
Tensile Stress at 300% Elongation	1700 psi	11.7 MPa
Tear Strength Graves	390 lb \cdot in ⁻¹	7.0 kg \cdot mm ⁻¹
Tear Strength Trouser	120 lb \cdot in ⁻¹	2.1 kg \cdot mm ⁻¹
Tm (by DSC)	275 °F	135 °C
Tg (by DSC)	−56 °F	−49 °C
Density	1.13 g \cdot cm ⁻³	

Table 3
Composition of kenaf filled TPU-NR composites according to weight ratios.

Fiber Condition	Sample code	Kenaf	NR	TPU	Abbreviation
Treated Fiber	T1KF1NR6TPU	12.50%	12.5%	75%	T116
Treated Fiber	T1KF2NR5TPU	12.50%	25%	62.50%	T125
Treated Fiber	T1KF3NR4TPU	12.50%	37.50%	50%	T134
Treated Fiber	T1KF4NR3TPU	12.50%	50%	37.50%	T143

The materials (Fig. 1) were blended using the hot melt blending Brabender machine [25–29].

calculation for water absorption was calculated as in Eq. (1):

$$\text{Water absorption} = \frac{W_1 - W_0}{W_0} \times 100\% \quad (1)$$

where.

W_0 = The weight of specimens before immersion.

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

2.4. Determination of the density

The developed composite was cut into a square shape with the dimensions of 10 mm \times 10 mm \times 3 mm. The solid square piece was then polished with 1200 grade sandpaper in order to make a smooth and shiny surface. The density of the develop composite was determined using the densimeter XS205 Mettler Toledo balance (Fig. 2) according to ASTM D792. There were five samples from each ratio prepared for this test. The density reading was then recorded. The density was calculated using the formula in Eq. (2):

$$\text{Density}(\rho) = \frac{\text{Mass}(m)}{\text{Volume}(v)} \quad (2)$$

2.5. Determination of thickness swelling

Ten specimens of 20 mm \times 20 mm \times 3 mm samples of each type of composites were prepared according to ASTM D570 for the testing of thickness swelling. The thickness swelling (TS) was calculated using Eq. (3):

$$\text{Thickness swelling} = \frac{T_1 - T_0}{T_0} \times 100\% \quad (3)$$

where.

T_0 = The thickness of the specimen before immersion. T_1 = The

Table 1
Liquid natural rubber GIVUL MR specifications.

Properties	Specification
Total Solid Content/%	60.5 \pm 0.50
Dry Rubber Content/%	NA
Non-Rubber Content/%	NA
Alkalinity/%	0.60 min
Magnesium Content/ppm	NA
Volatiles Fatty Acid	NA
MST/s	1100 min
Coagulum Content/ppm	100 max
KoH	NA
pH	10.0–11.0
Viscosity B/F LVT/cps	Max 120
Toluene Swell/%	82–85
Density/(g \cdot cm ⁻³)	0.93



Fig. 1. Raw materials weighed and ready to be blended in Brabender machine.



Fig. 2. Densimeter XS205 Mettler Toledo balance.

thickness of specimens after 7 days of immersion in distilled water.

2.6. Dynamic mechanical analysis (DMA)

The storage modulus, loss modulus, and damping factor ($\tan\delta$) of the composite specimens were measured as a function of temperature (20°C–200 °C), using a TA 2980 DMA, equipped with a dual-cantilever bending fixture at a frequency of 1 Hz (Fig. 3).

3. Results and discussion

3.1. Water absorption

Water absorption properties for the kenaf filled NR-TPU



Fig. 3. Dual cantilever bending fixture to determine the dynamic mechanical analysis.

composites were determined for each of the composition. In Fig. 4, the water absorption value was found to increase with the increase of the TPU in the polymer composites. Meanwhile, the polymer composites with a higher amount of natural rubber T143 showed a lower value of water absorption. The T116 composition of treated kenaf fiber filled tended to absorb moisture more than other samples. This clearly indicated the increment of the moisture

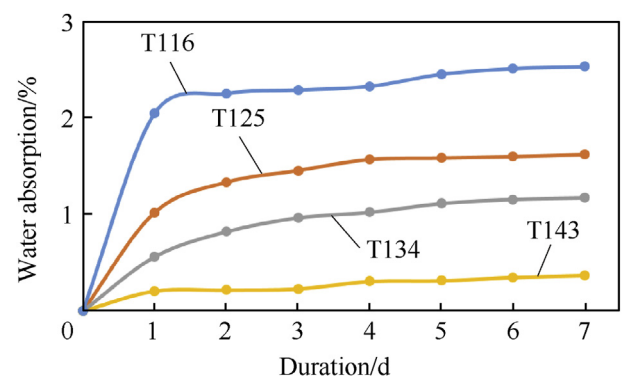


Fig. 4. Water absorption of kenaf filled TPU-NR composites.

absorption was due to poor bonding or adhesion between TPU and treated kenaf fiber. On the other hand, NR had better bonding and adhesion [30,31] for treated kenaf fiber, which had less moisture to absorb for the T143 sample. There were similar findings which showed that a treated fiber reduced the hydrophilicity of the fiber and enhanced interfacial bonding between the fiber and the matrix that prevented the fiber from absorbing the water [32,33].

3.2. Density

The density of treated kenaf filled TPU-NR polymer composites is shown in Fig. 5. From Fig. 5, it is shown that there was an increase in density as the TPU composition in the treated kenaf filled fiber composites was increased. Since the amount of TPU is dominating the composites (75%) and poor bonding interfacial between the matrixes resulting high density for the T116 composition. For the T143 sample, where the NR content was the highest among other samples which are 50%, it showed the lowest density value. This shows a good facial interaction in the matrices due to the treated kenaf fiber where the water absorption test is the lowest. It seemed that the amount of treated kenaf fiber of 12.5% in the composites had a small effect on the density of the composite samples. The density of TPU was 1.13 g/cm^3 , kenaf fiber was 1.4 g/cm^3 and NR was 0.93 g/cm^3 . Even though the density of kenaf fiber was higher than TPU and NR, 12.5% of treated kenaf fiber did not have a huge influence on the density of TPU composite or NR composite samples. The finding is similar to that of other researchers where the length of the fiber did not give any big effect on the density [34]. In this study the fiber loading was small (12.5%) and it had no influence on the density of the matrix but the ratio of NR and TPU influence the value of density.

3.3. Thickness swelling

Fig. 6 shows the thickness swelling of treated kenaf filled NR-TPU composites. The T116 composites contained a large amount of TPU that had gained the highest thickness swelling, which was 8%. This indicates the treated fiber has weak interfacial bonding within the TPU composites. Thus it will create micro-crack between the treated fiber and the TPU which allow a large amount of water to penetrate the T116 composites. This resulted in the T116 obtain high water absorption and thickness swelling result. Meanwhile, the T143 with a large amount of NR had the lowest thickness swelling, which was 3%. This clearly shows that the treated kenaf fibers had better bonding and adhesion with NR. This indicated that the hydrogen bonding existed between the kenaf fiber, sodium oxide (NaOH) and NR. This indicates that there were less micro-crack between the treated fiber and the NR. Due to less water

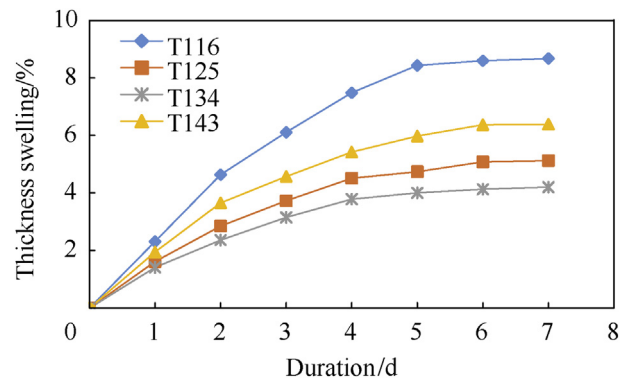


Fig. 6. Thickness swelling of kenaf filled TPU-NR composites.

absorption, the T143 composites have become a water-barrier composite. Similar findings by other researchers were reported regarding the hydrogen bonding between NR with treated fibers. It was found that hydrogen bonding improved the interfacial bonding between fibers and matrices [12,23]. With the improved bonding between the fibres and matrices, this helped to prevent water from entering the composites and the swelling effect was minimized.

3.4. Dynamic mechanical properties

3.4.1. Storage modulus

Fig. 7 shows the storage modulus against temperature for all 4 types of composite compositions conducted in this experiment. In general, the available storage modulus was inversely proportional to the temperature. The higher the temperature the lesser the storage modulus recorded. For samples T116 and T125, both graphs intersected at temperatures of 60°C and 145°C . This shows that both compositions shared the same high storage modulus at 60°C , which was 40 MPa and the lowest storage modulus at 145°C , which was 13 MPa. The T143 sample recorded the lowest storage modulus among other composites and dropped to 0 MPa at 150°C . The storage modulus decreased with the increase in the amount of NR and with the decrease in the amount of TPU in the composite samples. A similar finding was observed in other work, where the treated kenaf composites had higher storage modulus compared to untreated kenaf fiber composites [35].

3.4.2. Loss modulus

Loss modulus is generally defined as a viscose response of a material. It is an indication of the energy dissipation when a material is deformed after heat is applied [36,37]. Fig. 8 shows the

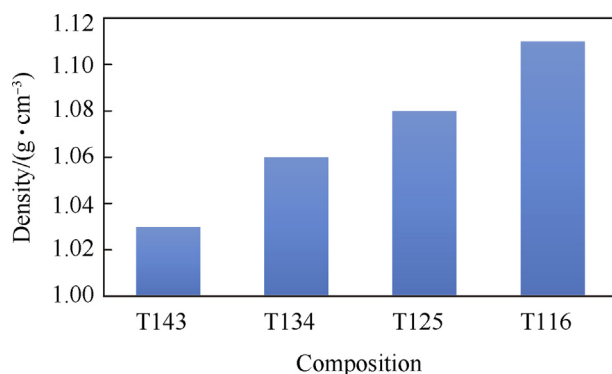


Fig. 5. Density of kenaf filled TPU-NR composites.

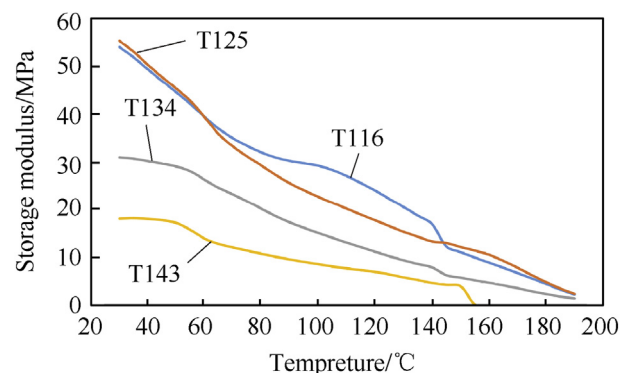


Fig. 7. Storage modulus of kenaf filled TPU-NR composites.

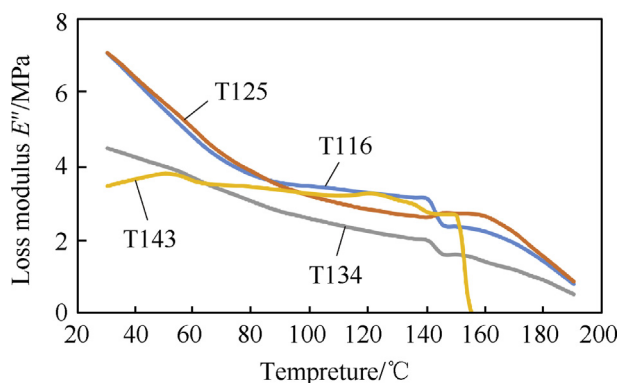


Fig. 8. Loss modulus of kenaf filled TPU-NR composites.

results of loss modulus against temperature for treated kenaf filled TPU-NR composites. From Fig. 8, it is clearly shown that the loss modulus decreased as the temperature was increased, which occurred for all samples. For the samples of T116, T125 and T143 the curves showed an intersection at 90 °C and 3.3 MPa, which meant the 3 samples shared the same loss moduli. This indicated that the T116, T125 and T143 composite compositions shared the same capability to dissipate energy at 90 °C and 3.3 MPa.

3.4.3. Damping factor ($\tan\delta$)

Fig. 9 shows the damping factor of treated kenaf filled TPU-NR composites. A good damping factor was $\tan\delta > 0.3$ [30–33]. The T143 sample showed an increase in $\tan\delta$ up to 0.6 at the temperature of 135 °C. Then the T143 sample started to degrade until 140 °C. For other samples, T124, T134 and T116, the $\tan\delta$ value did not reach above 0.3, which meant these three samples had low damping properties. After the temperature reached 140 °C, these three samples started to degrade until 145 °C, which resulted in $\tan\delta$ value to be below 0.25. This clearly indicated that the sample T116 had good damping properties [27] at the high-temperature condition.

4. Conclusions

From this experiment, the effect of NaOH on kenaf fiber filled TPU-NR composites towards the physical and dynamic mechanical analysis (viscoelastic parameters) had been determined. From the result above it is clearly shows that the treated kenaf fiber in the T143 composite, has the better result in water absorption and thickness swelling due to good interfacial bonding between the fiber and the NR matrices. This clearly indicated that the fiber

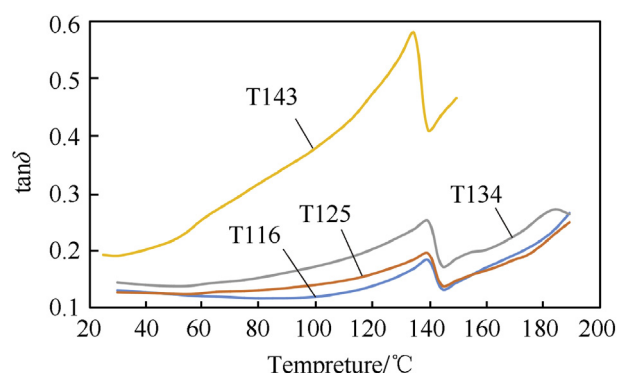


Fig. 9. $\tan\delta$ of kenaf filled TPU-NR composites.

adhesion due to the hydrogen bonding effect has made the composites into a water barrier material. From the DMA results, it was shown that the T143 sample had good damping properties in the high-temperature condition up to 135 °C. In this condition, the T143 composite had the potential to become a damping material in the high-temperature application and water resistant engine rubber mounting.

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