Influence of Alkaline Treatment Performance on the Mechanical, Thermal and Wettability Properties of Epoxy/Kenaf Fibre Composite

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Abstract
The development of eco-friendly composites with enhanced mechanical and thermal performance remains a challenge, particularly when using natural fibres. The studies on the mechanical and thermal properties of neat epoxy (EPX), EPX/KF and EPX/KF/NaOH at different kenaf fibre loading and at different NaOH treatment. This study investigates the effects of kenaf fibre (KF) loading and sodium hydroxide (NaOH) treatment on the properties of epoxy-based composites. Neat epoxy (EPX), EPX/KF, and EPX/KF/NaOH composites were fabricated via hand lay-up at varying fibre contents and treatment concentrations. Mechanical, thermal, and wettability analyses were conducted. Results showed that incorporating 30 wt % KF significantly improves tensile strength and Young's modulus compared to neat epoxy, while additional NaOH treatment further enhances tensile strain and impact strength. The Impact strength improved across all formulations, with the highest value observed at 40 wt % KF loading (3960.38 ± 210.5 J/m^2). Linear thermal expansion and viscoelastic parameters also improved, indicating better thermal stability and energy degeneracy. Wettability analysis showed reduced contact angle and wetting energy after fibre treatment, suggesting improved fibre–matrix adhesion. Largely, the kenaf fibre reinforcement combined with the best NaOH treatment yields composites with superior mechanical, thermal, and interfacial properties. Alkalization of kenaf fibre statistically improved interfacial interaction between epoxy and fibre, thermal stiffness and stability, alongside the effect increases with higher curing time. These findings support the use of chemically treated natural fibres in high-performance polymer composites for applications demanding improved durability and thermal stability.

Keywords: Epoxy, Kenaf-fibre, sodium-hydroxide treatment, Mechanical properties, Thermal properties and Wettability

1. INTRODUCTION

Composite materials have been used for thousands of years, with early examples including mud and straw bricks from ancient Egypt. Modern commercial composites emerged in the early 20th century when cellulose fibres were used to reinforce phenolic resins, followed by urea and melamine resins. Today, fibreglass is one of the most familiar composites, widely employed in lightweight structural applications such as automotive body panels, boat hulls, and bathroom fixtures (Sivakandhan *et al.,* 2020). Approximately 30 % of all polymers produced annually are used in civil engineering and building industries. Polymers offer significant advantages over conventional materials, including low density, corrosion resistance, and ease of processing. When combined with fibres or particles, they form composite materials with enhanced properties suitable for structural applications, ranging from construction to aerospace and satellite technologies (Musthaq *et al.,* 2023).

A composite material is defined as a combination of two or more distinct materials, each retaining its own properties, to create a new material with characteristics unattainable by any individual component. Modern composites typically consist of a fibre (e.g., glass, Kevlar, carbon, or polyethene) and a matrix (e.g., thermosetting or thermoplastic polymer) (Musthaq *et al.,* 2023). Thermosetting polymers offer several advantages over thermoplastics, including higher strength and stiffness, lower weight, and better creep and thermal resistance (Khan *et al.,* 2022). Among natural fibres, kenaf has gained attention due to its good specific mechanical properties, availability, and biodegradability. However, the hydrophilic nature of natural fibres and their poor compatibility with hydrophobic polymer matrices remain major challenges. Alkaline treatment (sodium hydroxide, NaOH) is commonly used to remove hemicellulose, lignin, and surface impurities, thereby increasing surface roughness and improving fibre-matrix adhesion (Nurazzi *et al.,* 2021; Narayana & Rao, 2023).

Several studies have investigated kenaf fibre-reinforced composites. Yunus et al. (2023) examined the fracture toughness of long kenaf/woven glass hybrid composites under water absorption conditions. Hassan et al. (2022) studied hybrid particleboards made from rubber wood (RW) and kenaf stem (KS), reporting that a 50:50 (RW:KS) ratio with 10 % resin content gave the highest strength (19.08 MPa), while a 70:30 ratio exhibited better stiffness (2.23 GPa).

Thermal properties are essential for composite Engineering applications. Khan et al. (2022) noted that thermal expansion governs processing and application behaviour, depending on polymer chemistry, cross-linking, and degree of polymerisation. Khoo et al. (2023) highlighted the importance of thermal analysis (TA) and thermal stability (TS) in industries ranging from pharmaceuticals to polymers. Saba and Jawaid (2018) compared the thermomechanical behaviour of bamboo/kenaf/epoxy hybrids, reporting improved dimensional stability with woven kenaf mats and an optimal 50:50 bamboo-kenaf ratio. Krishnasamy et al. (2019) investigated epoxy/banana/kenaf composites with different fibre orientations (vertical, horizontal, 45°) and stacking sequences, maintaining 40 – 42 % total fibre loading. The 45° orientation gave the highest coefficient of thermal expansion (CTE) at 34.61×10^{-6} /°C. Saba and Jawaid (2018) also reported that adding 3 wt% oil palm nanofiller, montmorillonite (MMT), or organo-modified montmorillonite (OMMT) reduced the CTE of kenaf/epoxy composites compared to pure kenaf/epoxy. Ilyas et al. (2021) and Gokuldass and Ramesh (2019) studied coconut fibre (C)/pineapple leaf fibre (PALF)/polylactic acid (PLA) hybrids, finding that higher coconut fibre content (C70/PALF30) gave the lowest thermal expansion.

Dynamic mechanical analysis (DMA) provides insight into polymer transition temperature, viscoelastic behaviour, and stiffness stability as a function of temperature, which is significant for applications such as automotive components (Jacob & Mamza, 2021; Gong *et al.,* 2022). Jacob and Mamza (2021) emphasised that storage modulus measures a material's ability to store elastic energy during cyclic loading.

Wettability, measured by contact angle, is another key property influencing fibre-matrix adhesion. Poor wettability leads to voids and reduced mechanical performance. Alkaline treatment can alter surface energy and reduce hydrophilicity, thereby improving adhesion (Narayana & Rao, 2023). Despite these advances, the systematic influence of alkaline treatment concentration (rather than simply treatment versus no treatment) on the combined mechanical, thermal, and wettability properties of epoxy/kenaf fibre composites has not been fully established. Most studies used one or two NaOH concentrations without exploring the concentration-dependent balance between surface cleaning and fibre degradation. Therefore, this study aims to systematically investigate the influence of alkaline treatment concentration on the mechanical, thermal, and wettability properties of epoxy/kenaf fibre composites.

2. Materials and Methods

2.1 Materials

The Epoxy (EPX) used in this research was of a commercial grade with a label name LAPOX l-12, purchased from Atul Ltd, India. The curing agent (ethylene diamine) used in this research was purchased from Atul Ltd, India, with a commercial grade name K-6. Triethylenetetramine has a molecular weight of 146.23g/mol and a density of 0.982g/mol at 25 ºC. Sodium hydroxide, NaOH (CAS number = 1310-73-2) pellets (500 g) were purchased from Nice Chemicals Limited, Kochi, India. Acetone, the solvent used as a sterilising agent, was purchased from Nice Chemicals Limited, Kochi, India. It has a molecular weight of 58.08g/mol. The kenaf fibre used in the form of non-woven mats was obtained from the Department of Polymer and Textile Engineering, ABU, Zaria.

2.2 Methods

2.2.1 Fabrication of EPX/KF Composites Using Hand Lay-up Technique

The composites were fabricated using a hand layup process. Primarily, a discharge gel was coated on the surface to prevent the mould from sticking to the epoxy resin to the surface. Plastic sheets were used at the top and bottom of the surface of the mould plate in order to get a good surface gloss. Non-woven kenaf fibre mats were cut according to the prescribed weight percentages of kenaf fibre (20, 30, 40, 50, 60 and 70) wt. % with respect to the dimension of the mould (Kenaf fibres were cut according to the shape of the moulds (15 x 15 cm) and placed into the mould. The epoxy resin in liquid form was mixed exhaustively in suitable proportion with the Triethylenetetramine (TETA) hardener and poured onto the surface of the mat already placed in the mould. The epoxy was spread with the aid of a roller. The prepared epoxy/kenaf fibre composites were then placed on the surface with the roller moving with an applied pressure on the epoxy/kenaf fibre composites to remove any air bubbles and voids that may develop during processing. The process was repeated for each of the epoxy/kenaf fibres at different kenaf fibre weight percentage compositions until the required composites were prepared. The dead weight was applied at the top of the mould, and the prepared samples were cured and post-cured at temperatures and times ranging from (20

– 30 ºC), 70 ºC and 24 - 48 hours. The epoxy/kenaf fibre composites post-cured at 70 ℃ were preconditioned in an oven for 3 hours and subsequently post-conditioned in a desiccator for the same time to remove any moisture absorbed from the surroundings during processing (Nurazzi *et al.*, 2021).

2.2.2 Mechanical Characterisation of EPX/KF Composites

(a) Determination of tensile strength of EPX/KF composites

Tensile tests were performed to determine the Young's modulus, tensile strength and elongation-at-break of each material. The tensile strength was determined according to the American Society for Testing and Materials (ASTM D3039) standard using a universal testing machine (UTM), Model Tinus Olsen H50KT. These tests were performed under a testing condition of 60 mm Gauge, tension mode at a strain rate of 1 mm/min at room temperature. The results presented are averages of six individual tests per sample.

(b) Determination of impact strength of EPX/KF composites

The impact test of this research was carried out in accordance with American Society for Testing Materials (ASTMD-D- 256-2018) with a Resil Release impact machine. A test piece of dimension 100 mm × 4 mm was cut and placed on the Impactor specimen holder, the impact hammer was released, and the impact values were recorded as averages. (Abioye *et al.*, 2019).

2.2.3 Determination of the contact angle of EPX/KF composites produced

Contact angle measurements were made according to ISO1409-2006 by photographing droplets of liquid on substrates using a Phoenix model instrument (Surface Electric Optics SEO), 5µL using a micro-syringe. Five microliter drops were applied to the substrate using a micro-syringe. Distilled water was used as the characterisation liquid at 25 ºC. Measurement was made on the test samples of different filler loading and 4 %, 5 % and 6 % alkaline treatment.

3. Results and Discussion

3.1 Mechanical Analysis of Pure Epoxy, EPX/KF at Different KF Loading and at Different NaOH Treatment of the Prepared Composites

3.1.1 Ultimate tensile strength of epoxy/kenaf fibre composites

The ultimate tensile strength of the EPX/KF composite produced are presented in Figure 1 (a, b and c)

Figure 1 (a) depicts the ultimate tensile strength of the prepared composite articles at different chemical at different environmental conditions. The profile depicts an increase in tensile strength with increasing of weight percentage of the kenaf fibre from 20 %wt to 40 wt. % of Kenaf fibre (KF) loadings with values of 40.30 MPa at 20 wt. %, 43.70 MPa at 30 % and 71.50 MPa at 40 wt. % of the KF, compared with 32.10 MPa, the value obtained for the unmodified Epoxy (EPX) matrix, respectively, as noticed in Figure 1a. In addition to these values obtained for the cured composite composition EPX/30 $KF_{c,}$ it displayed its maximum tensile stress of 29.10 MPa, which is lower than that of the unmodified epoxy resin matrix (control). The decreased value of tensile modulus for treated kenaf fibre reinforced composite could be due to the void matrix, deteriorated interfacial adhesion, and bond strength between matrix and fibre. However, the behaviours were enhanced by curing the composites, which decreased the number of voids in the epoxy matrix, thereby increasing the interfacial interaction between the epoxy and kenaf fibre filler. It can also be recorded that the chemical combination of chains, the Van der Waals force, and the hydrogen bonding in the molecular components of the polymer, were also improved during the kenaf fibre alkalization, which were responsible for the ability of the material to bear foreign stress (Uusi-Tarkka *et al.*, 2023). Moreover, in Figure 1b, the Young's modulus increased with the increase in weight of Kenaf fibre loading, this could be due to the decrease in the matrix mobility of EPX/KF composites, relative to the presence of the filler, which increases the stiffness of the composites at all the kenaf fibre loadings (10 – 40 wt. %) as stated earlier. Figure 1c shows the effect of fibre loading on the elongation property of the EPX/KF resin composites. It can be observed that the neat epoxy sample has the highest percent elongation value of 65.25 %, and as the Kenaf fibre weight percentage increases from 30 to 40 wt. %, the percentage elongation decreases from 50.5 % to 7.32 %, respectively. Hence, it can be deduced that the elongation property of the composites decreases with an increase in the Kenaf fibre loading. The decrease in the elongation could be attributed to the fact that, as the Kenaf fibre loading increases, the dispersion of the fibre in the matrix also increases, thereby enhancing the stiffness. Thus, the stiffer the composite materials, the lower the percentage elongation due to the difficulty of the particulate fibre to be extended (Magunga *et al.*, 2023). This behaviour is also supported by the work (Magunga *et al.*, 2023) of those who studied the effect of bagasse reinforcement on the mechanical properties of polyester composites.

Figure 2 Shows the effect of blend composition and filler loading on impact strength of unmodified EPX, EPX/KF and EPX/KF/NaOH at different filler loading and at different NaOH KF treatment respectively, from these result displayed it can be observed that, there was increase in impact strength in blend with and without filler compared to that of the pure EPX, thus, EPX/40 % KF has the highest impact strength value (3960.38 J/m^2) compared to the unmodified EPX (2328.1 J/m^2) and EPX/30 % KF/6 % NaOH (2335.82 J/m^2). The highest impact strength with respect to % NaOH KF treatment was obtained at EPX/30 % KF/5 % NaOH treatment, though it is less than that of EPX/40 % KF. This is obvious evidence that the alkalization of the prepared composite enhanced the interfacial adhesion between the epoxy and the kenaf fibre.

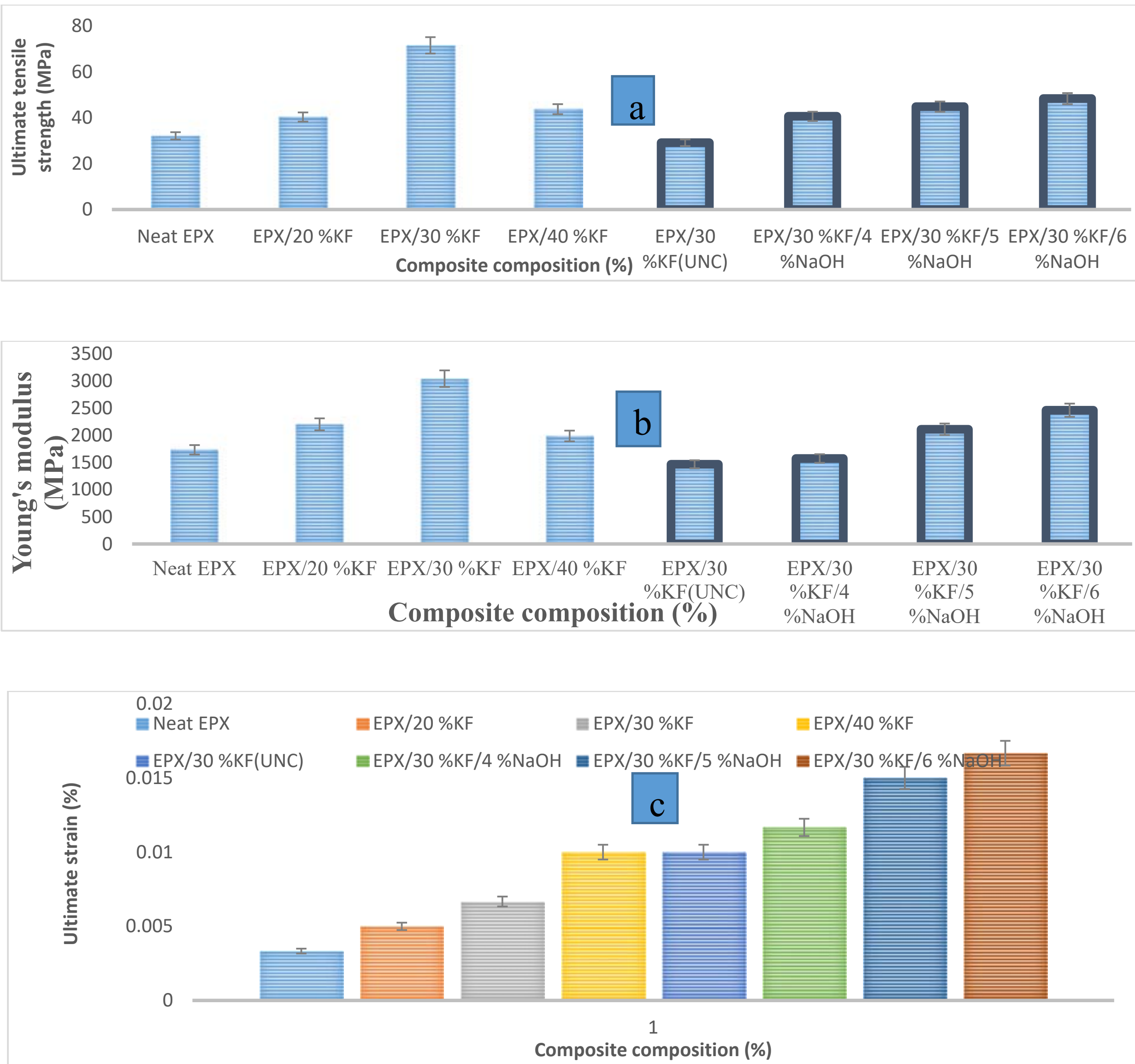

Figure 1 (a, b and c): Effect Kenaf fibre loading, concentration of NaOH treatment and post curing on tensile properties of epoxy/kenaf fibre composites

3.2 Impact strength of the epoxy/kenaf composites

The impact strength of pure EPX, EPX/KF and EPX/KF/NaOH at different filler loading and at different NaOH KF treatments is presented in Figure 2.

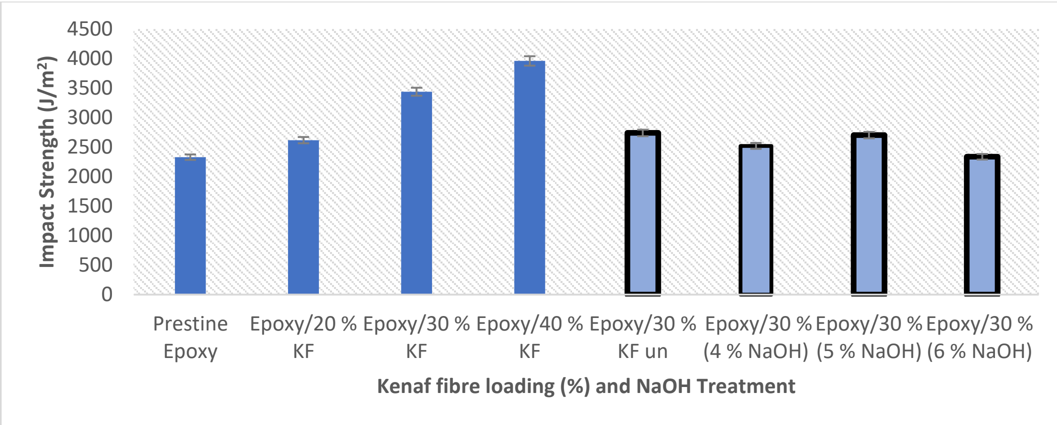

Figure 2: The effect of fibre loading, % concentration of NaOH treatment and post-curing on impact strength of the composites

3.3 Thermal properties of EPX/KF Composites: Thermomechanical Analysis (TMA) and Dynamic Thermal Mechanical Analysis (DTMA) and the results are presented in Figure 3 (A – E)

3.3.1 Thermomechanical analysis (TMA) of EPX/KF composites was presented in Figure 3 (A - E)

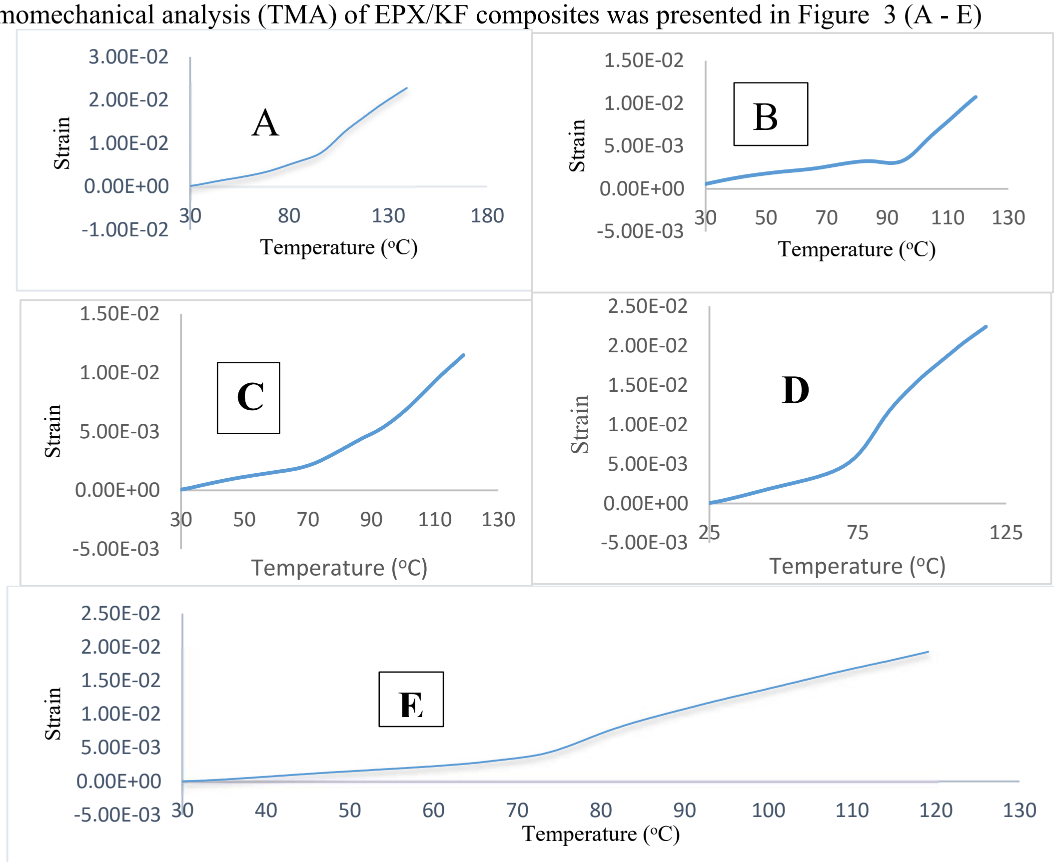

Figure 3: Effect of temperature on the thermomechanical properties of EPX/30 %KF_u (A), EPX/30 %KF_c, (B), EPX/30 %KF/4 % NaOH (C), EPX/3%KF/5 %NaOH (D) and EPX/30 %KF/6 %NaOH (E)

The LTEC of the pure EPX, uncured EPX/30 % KF, cured EPX/30 %KF and treated EPX/30 % KF with 4, 5 and 6 % NaOH were displayed in Figure 3 (A, B, C, D and E) as a function of temperature change. From the result recorded composite treated with 4, 5 and 6 % NaOH have higher a(LTEC), though the linear thermal expansion was not linearly observed, similar result was observed and reported by (Saba & Jawaid, 2018).

This increase in the LTEC observed in the composites is attributed to agglomeration of the kenaf fibre reinforcement. Similar trend was reported by (Hassan *et al.*, 2022) explained that the LTEC of epoxy/ kenef fibre in the glassy region is associated to the free expansion process consequently yielding a variability of the values. Thus, recording values of the LTEC of both untreated and treated composite having higher values compared to the values recorded for the unmodified EPX.

The highest value of LTEC was observed for EPX/30 % KF this attributed to the inrease in the epoxy, chain expansion, which could be the chance for the kenaf fibre to adhere to the epoxy that could lead to better KF reinforcement. Similar result was reported by (Saha & Kumari, 2023). Also this decreased in CTE was observed as the weight percentage of KF was incorporated into the epoxy which restricts the epoxy chains mobility resulting in greater interaction between the epoxy and the kenaf fibre which was also improved by the treatment of composites with different percentage of NaOH (4, 5 and 6 %), similar observation was made by (Radzi *et al.*, 2023).

3.3.2 Dynamic thermal mechanical analysis of the EPX/KF composites

The effect of temperature on the dynamic mechanical parameters of storage modulus (E'), loss modulus (E''), and tan δ of unmodified Epoxy Resin, EPX/30 % KF and EPX/30 %KF/ at 4, 5 and 6 NaOH % were presented in Figure 4a – 4f (A - M).

Figure 5 (I – M) depicts visco-elastic parameters such as storage modulus (E'), loss modulus (E''), and tan δ of unmodified epoxy resin (control), EPX/30 % KF and EPX/30 %KF/ at 4, 5 and 6 NaOH %. With increasing temperature at oscillation frequencies of 2, 5 and 10 Hz, respectively.

The storage moduli $E'(MPa)$ curves show the thermal stability of the unmodified matrix under dynamic loading, in which the onset temperature is 85.3 °C with a storage modulus E' (590 MPa) and its point of inflexion (mid-point), usually taken as the glass transition temperature, is 91.7 °C at 10 Hz. The E''(429 MPa) curve depicts peak maximum temperature at 110.5 °C while the tan δ curve describes the viscoelasticity of the unmodified epoxy resin matrix with corresponding damping factor (0.797) at 10 Hz. On the other hand, Figure 5 (J), displays the combined curves of dynamic mechanical analysis of EPX/30 %KF_c, illustrating the storage modulus E', the loss modulus E'', the damping factor tan δ, the creep deformation dL, the transition temperature T_g and the activation energy (E_a) respectively (I - M). Here, a decrease in onset temperature was observed as compared to that of the unmodified epoxy (77.6 °C). The value of the storage modulus of the epoxy composite composition (798 MPa) increased as compared to that of the unmodified epoxy with a value (590 MPa), and the loss modulus E'' (800 MPa) largely increased as compared to the value recorded for the unmodified epoxy (429 MPa) similar observation was reported by (Kumar *et al.*, 2020). Figure 5 (A) shows thermal stability of the EPX/30 % KF_{pc} composite under dynamic loading, where its inflexion temperature was at 93.7 °C, which was greater than the value recorded for both the neat and the cured EPX/30 %KF composite. This value was obtained at a 10 Hz frequency. However, the storage modulus E^1 (600 MPa) increased as compared to the value obtained for the neat epoxy, that is for both uncured and cured composite for 30 % Kenaf fibre loading improvement in the thermal stability of the composite of the filler composition increased which was an indication of reinforcing ability of the Kenaf fibre to interact with the epoxy and condensed with the polymer resin resulting in the formation of rigid, thermally resistant and stiffer product as reported by (Shuaibu *et al.*, 2020). Also, the Tg of this composite increases as the curing time increases, where the value recorded was 108.3 °C.

The $E''(MPa)$ curve depicts EPX/30 %KF_c composites, which shows a decrease in energy dissipation at maximum peak temperature of 110 °C (667 MPa). While the tan δ curve revealed an increase in its value compared to both the neat and uncured composite of the same Kenaf fibre composition (0.313), which implies an increase in both the glass transition temperature and stiffness stability of the composite of the cured Kenaf fibre loading. This also means that the epoxy chains' mobility is restricted by the Kenaf fibre distribution, thereby transferring its stressing capability into the epoxy, resulting in the epoxy rigidity increasing its ability to absorb more thermal energy in order to attain its Tg (Yaro, 2023). This result agrees with the work of Yaro (2023), who worked on the effect of variation in frequencies on the dynamic mechanical properties of jute-reinforced epoxy composites. Similar positive shift of T_g from the tan δ curve at 10 Hz was also observed by (Akter *et al.*, 2023). There was a gradual drop in storage modulus with increasing Kenaf fibre and temperature because of the loss in stiffness at elevated temperatures. This could be attributed to the relaxations in the polymer matrix associated with the change in glass transition temperature (T_g) from crystalline to amorphous state, as suggested by Zhang *et al.*, (2023). It is clearly observed that EPX/30 %KF_c composites have the highest stiffness stability because the storage modulus of 790 MPa in the glassy region, when compared to other composites of different filler wt. %. This occurs as a result of the strong interfacial adhesion, which indicates superior dynamic mechanical properties (Musthaq *et al.*, 2023). Slight decrease in the storage modulus for EPX/30 %KF_c, however, as this value (600 MPa) obtained was still greater than the storage modulus of (Musthaq *et al.*, 2023) the unmodified matrix (590 MPa), this could be associated with the low stiffness, which tends to reduce the viscoelasticity of the epoxy matrix. Thus, the current study revealed that the EPX/30 %KF_c composite had the highest value of storage modulus in the rubbery region this account for its better interface bonding than the neat EPX and EPX/30 %KFc. This result was supported by the results established by (Ribeiro *et al.,* 2021) and (Huang *et al.*, 2023).

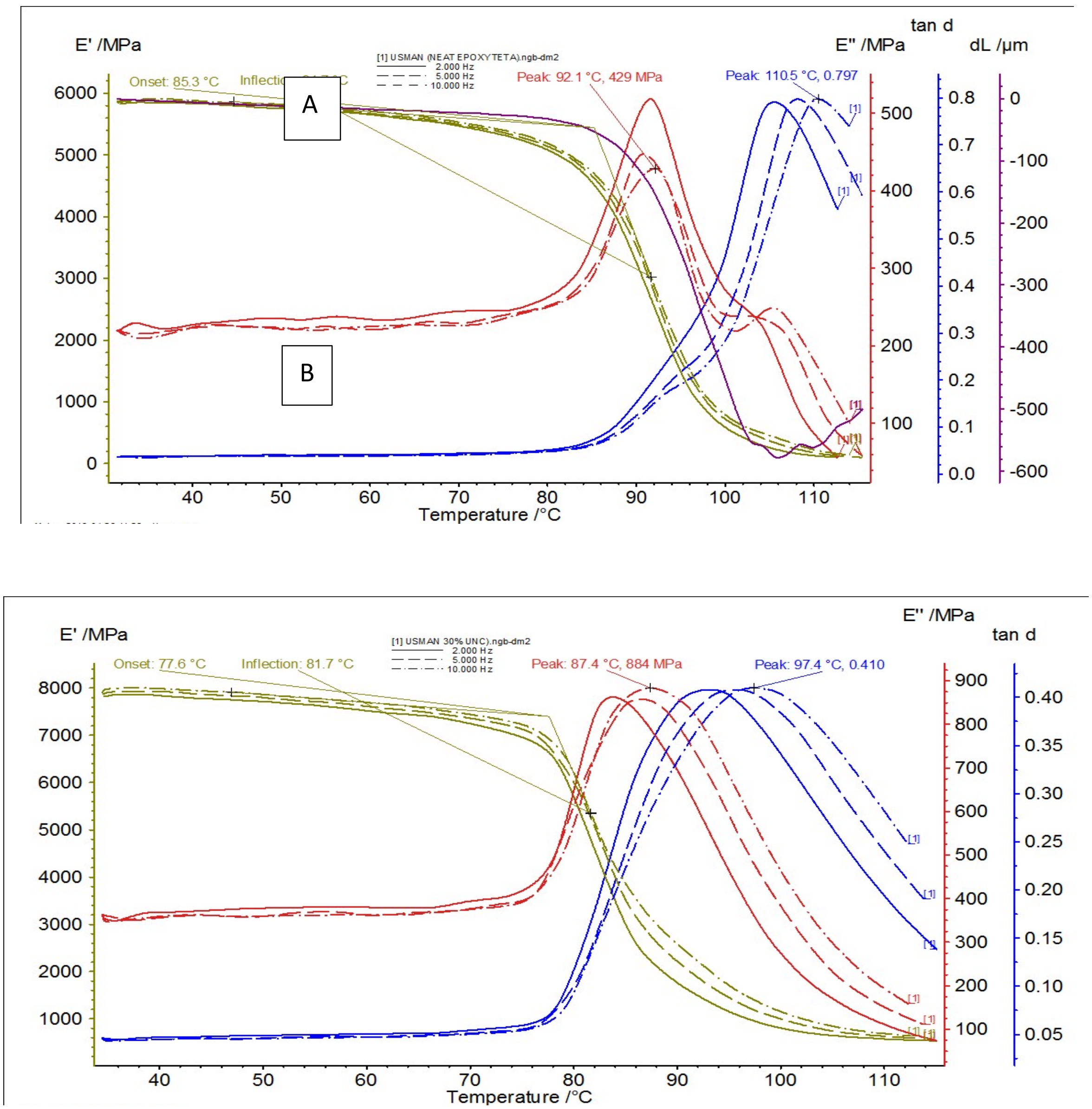

Figure 4a: Effect of Temperature on storage modulus, loss modulus, Tan δ and creep deformation of unmodified Epoxy (A) EPX/30 %KFc (B) at 2.0, 5.0 and 10.0 Hz

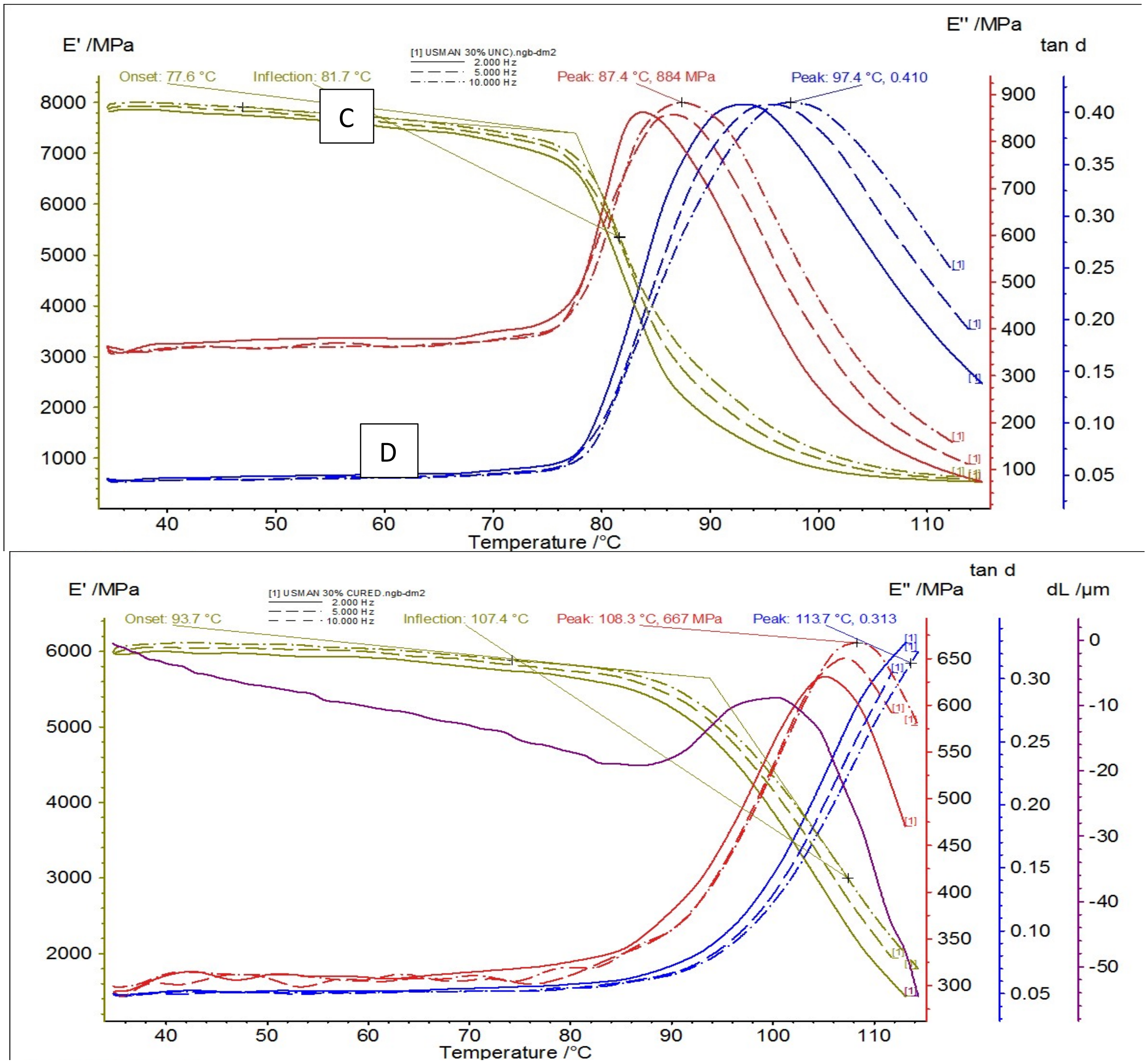

Figure 4b: Effect of Temperature on the Dynamic Thermal Mechanical Properties of EPX/30 %KF_c (C) and EPX/30 %KF_{pc} (D)

Figure 5 (A - G) shows the red colored graphs showing the variation of loss modulus with increasing temperature at 10 Hz oscillation frequency. From the curve, it was observed that the loss modulus increases with an increase in temperature, with the neat epoxy having the least loss modulus of 429 MPa. The highest value of the loss modulus was obtained for EPX/30 %KF_c (800 MPa). In addition, the incorporation of kenaf fibre content caused broadening of curves, which depicts an increase in thermal stability of composite materials in comparison with the neat epoxy loss module. This may be attributed to the decrease in the molecular mobility in the matrix. Therefore, it can be established here that the loss modulus increases with an increase in Kenaf fibre loading. A similar observation was reported by (Tarih *et al.,* 2023), who studied the effect of groundnut shell powder on the viscoelastic properties of recycled high-density polyethene composites. This implies that a strong matrix – fibre interaction results in good adhesion, which could lead to a decrease in the mobility of the polymer chain, thereby decreasing damping (Narayana & Rao, 2023). For instance, the unmodified epoxy had the highest Tan δ of 0.797, while EPX/30 %KFc had the lowest Tan δ value composite had the lowest of 0.313; hence, there is a gradual increase in damping coefficients (Tan δ) as the Kenaf fibre content, NaOH treatment of the KF, curing time and temperature increase (Gupta, 2022). The enhancement in storage modulus observed for chemically treated fibre composites is due to the better interfacial adhesion between the EPX and the KF fibre treated by the 4 %NaOH to the prepared composite. That is, the chemical treatment increases the matrix/Kenaf fibre adhesion causing reduced molecular mobility in the interfacial region. The highest improvement in the storage modulus was observed with respect of 6 % NaOH treated EPX/30 %KF composite.

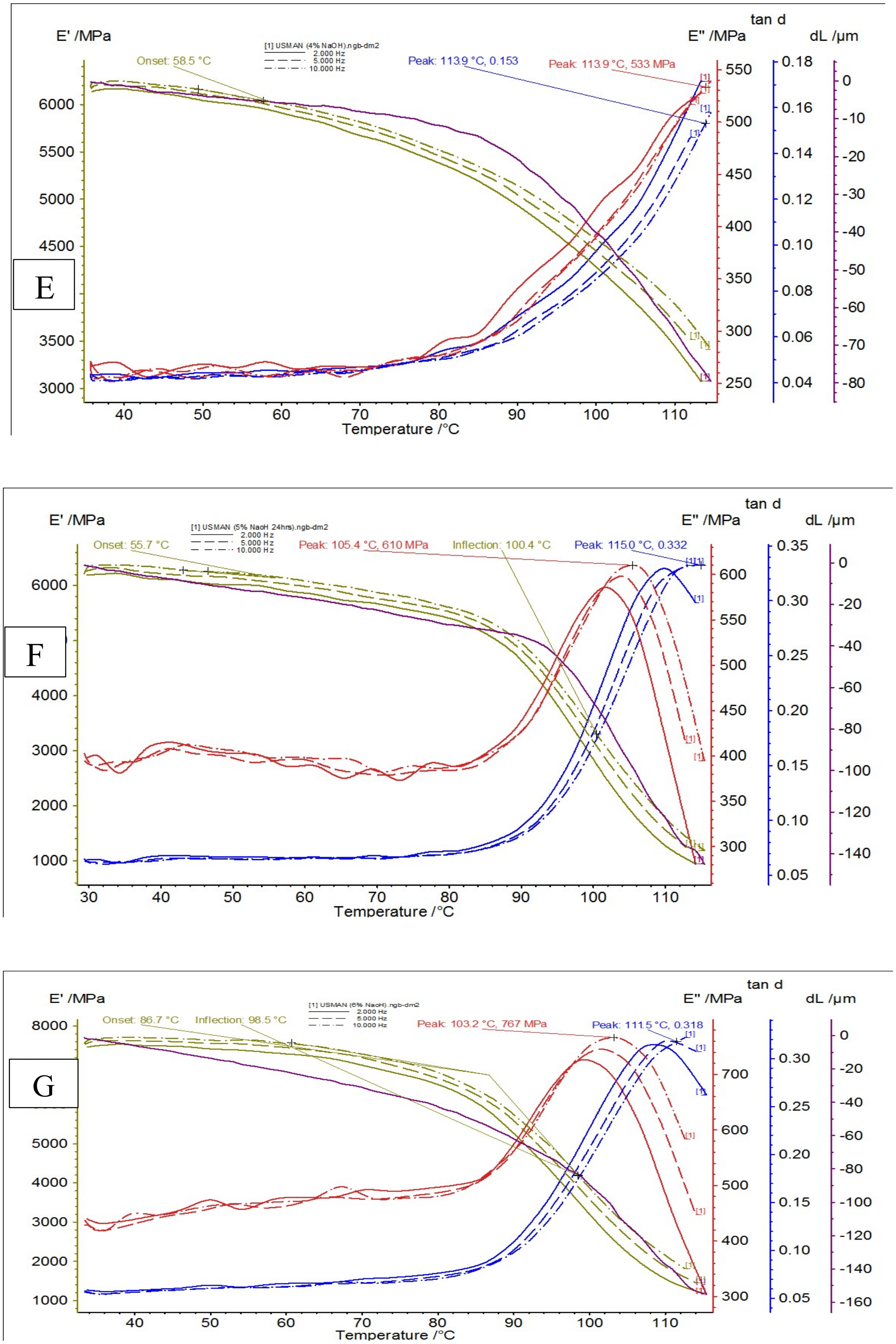

Figure 4c: Effect of Temperature on the Dynamic Thermal Mechanical Properties of EPX/30 %KFp/4 %NaOH (E), EPX/30 %KFp/5 %NaOH (F) and EPX/30 %KFp/6 %NaOH (G) at 2, 5 and 10 Hz

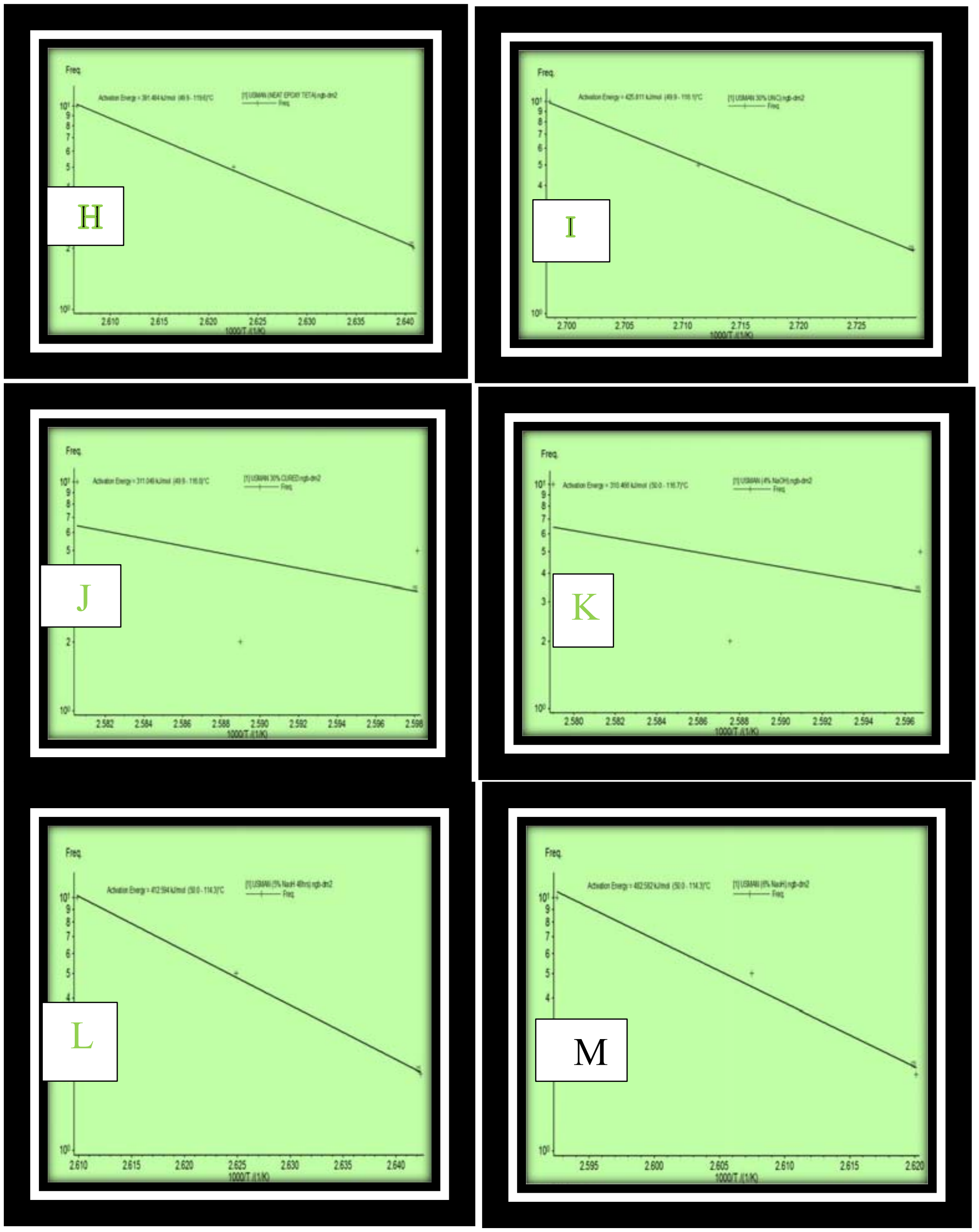

Figure 5 (H – M): Effect of Temperature on the Activation Energy of EPX/30 %KF_{pc} and EPX/30 %KF/4 %NaOH

The increase of storage modulus of the untreated and treated composites was found in the order of untreated <4 %NaOH of EPX/30 %KF treated < 5 %NaOH < 6 %NaOH EPX/30 % KF. The corresponding observation was given by (Huang *et al.*, 2023; Yang *et al.*, 2023).

Comparing the tan δ maximum values of the different composites, the unmodified EPX has the highest value and EPX/30 %KF/6 %NaOH treated composites show the minimum value Neves *et al.* (2023). This result is in agreement with the observation of the higher modulus values of the chemically modified fibre composites reported by (Huang *et al.*, 2023; Yang *et al.*, 2023).

The activation energy E_a, for the glass transition of the unmodified EPX, the EPX/30 %KFu, EPX/30 %KF_c and the EPX/30 %KF composites treated with different percentages of NaOH solution.

The activation energy was found to decrease with the incorporation of KF and was increased by the alkaline treatment and the maximum value was attained at EPX/30 % KF/6 % NaOH composite (Figure 5 [I- M]). Chemical treatment of Kenaf fibre increases the activation energy of the composites and the value for the activation energy is found to be maximum for the alkali-treated fibre composites. The result is in agreement with the storage modulus values, which is high for NaOH-treated composites as reported by (Huang *et al.*, 2023; Yang *et al.*, 2023). The highest result of the activation energy obtained for EPX/30 % KF/6 % NaOH composites can be attributed to a better interfacial adhesion existing between the EPX, the KF and the 6 % NaOH treatment. (Saha & Kumari, 2023).

3.4 Contact angles

The mean contact angles and wetting surface energies of water on the composites produced are listed in Table 1

Table 1: Contact angles and wetting surface energies of the EPX/KF composites

S/N	Composite composition (%)	Average contact angle (°)	Wetting energy
1	Neat EPX	57.4431	28.6957
2	EPX/20 %KF	68.8094	26.2595
3	EPX/30%KF_C	72.64909	21.70992
4	EPX/30 %KF_{Pc}	81.9497	62.6401
5	EPX/40 %KF	66.9091	27.6554
6	EPX/30 %KF/4 %NaOH	74.65417	19.2653
7	EPX/30 %KF/6 %NaOH	84.41648	16.5910

The wettability results of unmodified EPX and EPX/KF composites at different filler wt. % and at different alkaline treatments were summarised in Table 1. Figure 6 displayed the results of the wettability studies. The unmodified epoxy has shown a low contact angle of 57.4 °. Purely based on the measurement of water-air-solid contact angles. Significant uncertainty has arisen in the literature as to whether the commonly used epoxies are hydrophobic or hydrophilic (Jakes *et al.*, 2019). Similar observations on the flat smooth piece of cured epoxy, water had a contact angle of 20 ° in air, implying that the epoxy is hydrophilic. (Erbil *et al.*, 2021) reported contact angles between 61.27° and 64.72° for polished epoxy surfaces, 53.00° and 61.17° for cast epoxy surfaces and 58.91° and 62.96° for roughened epoxy surfaces(Erbil, 2021). These measurements could represent averages, on the basis of these findings, compared with the results determined and reported by (KOÇYİĞİT, 2023), that the epoxy/KF composite falls into the range of intermediate wettability. The researchers then compared these results to water-air contact angles on Smooth granite (53.00 ° – 59.35 °) and on glass (7.47 ° – 34.30 °) and determined that the epoxy was a better match for the granite than glass, which is frequently taken to be a good representative of silicate mineral surfaces. The result indicates that with an increase in fibre loading up to 30 %, the composites have improved the contact angle, indicating a reduction in hydrophilicity compared to the neat epoxy, but declined on fibre loading of 40 %, which obviously may be due to the rough surface of the sample which was caused by incomplete immersion of the kenaf fibre in the epoxy resin. On the other hand, the post-cured sample EPX/30 %KF_P (Figure 6 [D]) has shown a better result than the one cured at room temperature EPX/30 %KF_C which may be attributed to the excellent and strong adhesion between the fibre and the epoxy as a result of total crosslinking of Epoxy and amine, as well as completion of reaction of the matrix. The fibre loading at 40 % have increased the wettability of the composite. For instance, at a kenaf fibre loading of 30 %, the maximum wettability (water absorption uptake) decreased from contact angle 81.9497° to 66.9091° with corresponding wetting energy 62.6401 J to 27.6554 J for the post-cured EPX/30 % KF_p and the EPX/40 % KF_p composites, respectively. A similar observation was reported by (Taj *et al.*, 2023). The sodium hydroxide treatment of the filler significantly reduced the wettability of the composite. The contact angle has improved from 66.9091° for the untreated EPX/40%KF to 74.65417° which further increased to 84.41648° for both treated EPX/30 % KF/4 % NaOH and EPX/30 % KF/6 % NaOH, respectively. It can be established here that this reduction in the contact angles as the alkaline fibre treatment proceeds may be attributed by the chemical modification of the composite which resulted from the better wetting of the KF particles by the polymer and the enhancement in the interfacial adhesion between the fibre and the matrix. The decreased hydrophilic character of the Kenaf fibre after treatment with $NaOH_{(aq)}$ is also another reason contributing to the decrease in the water sensitivity of the composite which is accompanied by the removal of fats, oil and lignocellulose components of the Kenaf fibre, as corresponding results were reported by numerous research groups (Parashar & Chawla, 2023). In addition to this observation, it can be deduced and established a defined synergy between the wettability results and those reported for mechanical properties like tensile and impact strength properties because of the similar trend that was observed in the case of their results when compared to the results of the contact angle, that is, a strong relationship can be established between the structural property and the mechanical performance. It was also noted from the results of contact angle measurement that the interfacial energies of the EPX/KF/4 % NaOH, EPX/30 % KF/6 % NaOH are much lower than that of EPX/30 % KF which indicates a good interfacial interaction between epoxy matrix and Kenaf fibre components of the prepared composite, due to the similar polarity associated to the epoxy matrix and Kenaf fibre which was improved by the alkaline treatment. A similar result was reported by

(Al-Meer & Al-Kuwari, 2023; Javidrad & Javidrad, 2023). This decreased the accessibility of hydroxyl groups and enhanced the surface roughness of the epoxy/kenaf fibre after alkali treatment, resulting in the improved contact angle of the alkali-treated composite when compared to the untreated one. From the contact angle studies of the treated and untreated EPX/KF composite, it could be concluded that the 30 % fibre loaded and post-cured composite and that of the alkali-treated EPX/KF composite have shown improved wettability and tend to become hydrophobic in nature (Sooraj, 2021).

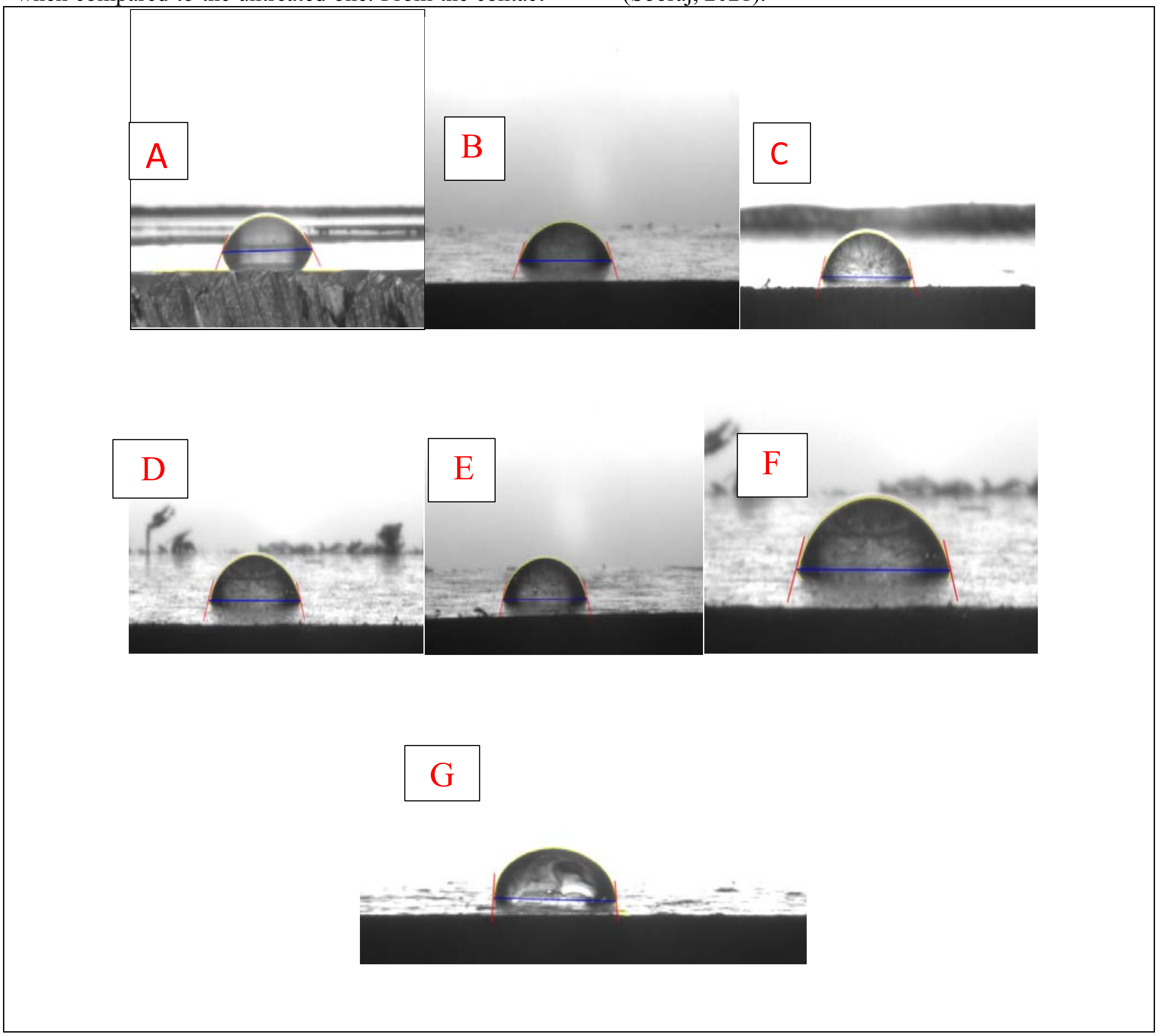

Figure 6: Contact Angle of unmodified Epoxy (A), EPX 20 %KF (B), EPX/30 % KFc (C), EPX 30 %KFps (D), EPX/40 % KF (E), EPX/30 %KF/4 %NaOH (F) and EPX/30%KF/6 %NaOH

The summary of ANOVA results was tabulated in Table 2. Values were computed based on a low p-value (≤0.05). P-value serves as an alternative to rejection to provide the smallest level of significance at which the null hypothesis would be rejected. A smaller p-value means that there is strong evidence in favour of the alternative hypothesis. For the impact strength and crystallinity of the prepared epoxy-kenaf fibre composite, since the p-value is less than the f-critical value at a 0.05 significance level, this implies that there is a significant difference between the impact strength results and that of the crystallinity. This statistically shows that there is a strong relationship between the impact strength analysis of the prepared EPX/KF composite and that of the structural crystallinity of the same composites for both uncured and cured composites at different kenaf fibre loading and at all the alkaline treatments. This strong relationship could be due to good interaction between the epoxy and kenaf fibre matrix, which was improved by the sodium hydroxide treatment of the fibre, as observed by the experimental analysis. Dan-as be et al. (2021) reported similar observations. For the storage modulus of the uncured and cured EPX/KF composites, the p-value is also less than the f-critical, which statistically means that there is a significant difference between the uncured and the cured EPX/KF composites. This result showed that after curing the EPX/KF composites, there was better interfacial interaction between the epoxy and kenaf fibre filler. Although the p-value is still less than the f-critical, thus implying a decrease in the relationship between the filler, the alkaline treatment and the curing time. This could be due to the removal of cellulosic lignin in the fibre by the sodium hydroxide solution during curing; a similar observation was made by Esonye *et al.* (2021). For the theoretical and the experimental density, values computed were statistically analysed, the results showed that there is a significant difference between the theoretical and the experimental densities as the p-value (0.00001187) of the analysis is smaller than the f-critical (19). This could be as a results of the incorporation of the kenaf fiber in to the epoxy or the alkalization of the fiber, the since at the different percentage composition of the sodium hydroxide fiber treat and different curing time both lignin and cellulose were removed from the kenaf fiber this could be the reason for enhancement of the interfacial adhesion between the matrix and the kenaf filler which may probably reduce the experimental density of the composite as compared to that of theoretical determination. This means that the alkalization of the EPX/KF composite could also improve the mechanical properties, such as toughness and stiffness of the prepared composite; a similar result was reported by (Dan-asabe *et al.,* 2019). For the contact angle measurement of uncured and cured composites, a similar trend was observed for the density results discussed above, as the p-value recorded is less than the f-critical. Thus, this illustrates that the presence of the kenaf in the epoxy matrix reduced the porosity among the epoxy particles that could be produce during formulation and processing the composite which were further minimized by the sodium hydroxide fiber treatment and curing, this could be the reason why the p-value obtained is less than the f-critical indicating the significance of the filler incorporation into the epoxy and fiber sodium hydroxide treatment, similar result was reported by (Dan-asabe *et al.,* 2019). Generally, since the p-value is less than the f-critical value at 0.05 significance level, there is a significant difference. Hence, this result confirmed that the curing of EPX/30% KFcu statistically improved the interfacial interaction between the epoxy and the Kenaf fibre; thus, the higher the curing time, the greater the interfacial adhesion between the epoxy and the Kenaf fibre. Similarly, the alkalization of the Kenaf fibre improved the thermal stiffness and stability. A similar result was portrayed by (Dan-asabe *et al.,* 2021).

Table 2: Statistical Analysis results and discussion

S/N	Source	df	Sum of squares (SS)	Mean of squares (MS)	f	p-value	f-critical
1	Impact strength and (MPa) crystallinity (%)	6	21490915	21490915	95.51829	0.0000101	5.31765507
2	Storage modulus of uncured and cured EPX/KF	7	2021879	336979.9	2.113301	0.172372	3.787044
3	Density (g/cm^3) of theoretical and experimental	8	84244.62623	1316.32228	84244.63	0.00001187	19
4	Contact angle of (°) treated uncured and treated cured composites	1	4877.962	4877.962	28.04858	0.000349	4.964603

4. CONCLUSION

The studies on the mechanical properties of the unmodified epoxy (EPX), EPX/KF and EPX/KF/NaOH at different kenaf fibre weight percentages and different concentrations of the NaOH treatment were successfully and effectively analysed. The results obtained revealed that the integration of kenaf fibres in epoxy resin greatly improves the mechanical, thermal, and viscoelastic properties of the fabricated biocomposite. These properties include tensile strength, modulus of elasticity, impact resistance, and strain at failure, with more improvement recorded at particular fibre content. Importantly, it was shown that sodium hydroxide pretreatment of the kenaf fibre is essential in optimising the mechanical, thermal, and viscoelastic properties, including the modulus of elasticity, thermal expansion coefficient, storage, and loss moduli, as well as the damping ratio. It is clear that the increase in the contact angle after pretreatment shows an improvement in hydrophobicity, which is beneficial in terms of moisture protection and interfacial bonding. The results finally confirmed that the curing of EPX/30 % KFcu statistically improved the interfacial interaction between the epoxy and the Kenaf fibre; thus, the higher the curing time, the greater the interfacial adhesion between the epoxy and the Kenaf fibre. Similarly, the alkalization of the Kenaf fibre improved the thermal stiffness and stability. For all properties analysed, the p-value was less than the f-critical value at the 0.05 significance level. This consistently indicates statistically significant differences between the compared groups, confirming that the null hypothesis is rejected.

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