

# Mechanism of Fiber/Matrix Bond and Properties of Wood Polymer Composites Produced From Alkaline Treated *Daniella oliveri* Wood Flour

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**The effects of sodium hydroxide (NaOH) concentration and time of treatment on the mechanism of fiber/matrix bond and functional properties of *Daniella oliveri* reinforced wood polymer composites (WPCs) were investigated. The WPCs were evaluated using Fourier transform infrared (FTIR) spectroscopy, mechanical testing, scanning electron microscopy (SEM), thermogravimetric analysis (TGA), and differential scanning calorimetry (DSC). The fiber/matrix adhesion mechanism could be attributed to the disruption of hydrogen bonding in the *D. oliveri* wood fiber network structure and the removal of lignin, wax and oils covering the external surface of the fiber cell wall. This leads to an increase in desirable functional properties as alkaline concentrations reached 4 wt%, but subsequently reduced at higher concentrations, while they increased with treatment time. Analysis of the fractographs of the WPCs suggests optimization of interfacial fiber-matrix adhesion and functional properties when *D. oliveri* wood fiber was treated with a 4 wt% solution of NaOH for 150 min. POLYM. COMPOS., 00:000-000, 2015. © 2015 Society of Plastics Engineers**

## INTRODUCTION

Research into wood polymer composites (WPCs) is increasingly attracting the attention of both academic researchers and manufacturers because WPCs enable the

conversion of low-valued wood flour to high value building and automobile products, characterized by high strength, low weight, corrosion resistance, and low maintenance cost [1]. In addition, WPCs can be easily disposed of at the end of their life cycle by composting or by the recovery of their caloric value in a furnace, which is not possible for alternatives such as glass fiber [2]. However, wood fibers also exhibit some undesirable characteristics such as high moisture absorption and low thermal resistance [3]. Thermoplastic resins, such as polypropylene (PP), polyethylene (PE), polystyrene (PS), and polyvinyl chloride (PVC), soften when heated and harden when cooled. This property allows materials such as wood fiber to be mixed with plastics to form a composite product. The resulting WPCs can be easily processed into various shapes and recycled at the end of their lifecycle.

The properties of WPCs depend on the degree of interfacial bonding between the polymer matrix and the wood fiber. When wood fibers are used as reinforcement in WPCs, problems such as debinding of the wood fibers within the polymer matrix may occur at the interface due to incompatibility [2]. This is because the hydrophilic wood fibers do not adhere with the hydrophobic polymers as a consequence of the poor interfacial adhesion between the wood fibers and the matrix. This results in poor mechanical properties, leading to low engineering performance. To increase the compatibility and adhesion between the fibers and the matrix with a view to optimizing WPCs' engineering performance, the wood fibers are always subjected to various surface modification and treatments. Among fiber surface modification techniques are alkalization, benzylation,

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acetylation, peroxide treatment, and silane treatment [4]. The effectiveness of these surface treatments in improving the adhesion between the fibre and the matrix had been demonstrated with the two-fold increase in the interface strength when henequen fibre was treated with an alkali and a silane coupling agent [5, 6]. Li and co-investigators [7] carried out a comparative analytical review of some of these chemical surface modification treatments of natural fibers. Meanwhile, alkalization treatment was established to be most economical and easier way of improving the compatibility between the fiber and the matrix in comparison to other chemical surface treatment of natural fibers. Hence, the basis for the choice of alkalization treatment of wood flour in this study. The mechanism of surface modification of fibers via alkali treatment occurs via the removal of hemicelluloses and splitting of the fibers into fibrils thereby resulting in closer packing of cellulose chain as a result of the release of internal strain which consequently improve the mechanical properties of fibers [8]. According to Ray et al. [9], it is the fibrillation of the fibres which increases the effective surface area available for wetting by the matrix and improves the interfacial bonding; hence, alkalization breaks hydrogen bonds and increases the number of free hydroxyl groups on the surface of the fibres and therefore fibre reactivity.

Surface modification of the wood fibers by means of cost effective alkaline treatment is one of the major areas of current research to develop compatibility and interfacial bond strength, water absorption resistance, dimensional and thermal stability in the WPCs [2, 3, 10]. Meanwhile, there has been the challenge of varying rules in the choice of the alkalization treatment parameters to obtain the most desirable improvement in the mechanical and physical properties of WPCs. Saha et al. [11] investigated the physico-chemical properties of jute fibers treated with alkali (NaOH) solution under ambient and elevated temperatures and high pressure steaming conditions. Findings from their study established that a 30 min dipping of the fibers in 0.5wt% alkali solution followed by 30 min alkali-steam treatment lead to an increase in the tensile strength (TS) of up to 65%. Moreover, Van de Weyenberg and co-investigators [12] optimised NaOH concentration and time of treatment with a view to obtaining a better adhesion between flax fibres and epoxy matrix and developing a continuous process for the treatment and resin impregnation of unidirectional flax fibre epoxy composites. The outcome of their investigation revealed a clear improvement in the mechanical properties when flax fibres were subjected to a mild treatment in a 4% NaOH solution for 45 seconds with the transverse composite strength increasing by 30%. Roy et al. [13] treated jute fibres with 0.25–1.0% NaOH solution for 0.5–48 h to explore the variation in the hydrophilicity, surface morphology, crystallinity index, thermal and mechanical characteristics of untreated and alkali treated fibres. It was recommended that low-concentration alkali treatment of jute fibre for a prolonged time may find effective and potential industrial usage because it enhanced fibre

mechanical properties while it also reduced the hydrophilicity of jute fibre which might in turn improved fibre matrix interface bonding. Nam et al. [14] developed a poly(butylene succinate) (PBS) biodegradable composites reinforced with 25 wt% coir fibers soaked in 5 wt% NaOH concentrated solution at room temperature for 72 h. The mechanical properties of alkali-treated coir fiber/PBS composite were noted to be higher than that of the untreated fibre and pure PBS resin. Gu [15] also examined the variation in the TS of treated brown coir fibres and its composites by varying the concentration of NaOH solution between 2 wt% and 10 wt%. It was reported that lower TS was obtained for the composite when treated with 10 wt% NaOH concentration in comparison to 2–8 wt%. It is evident from the reviewed literature that the alkalization concentration and its time of treatment vary from 0.25 wt% to 8 wt% and 45 s to 72 h, respectively, depending on the type and content of the natural fibre as well as the type of the polymer matrix. Therefore, the fibre type as well as its content may be influential in the determination of the optimum alkalization treatment parameters consequent upon variation in the chemical composition of cellulose.

When developing WPCs from new reinforcing materials such as *Daniella oliveri* wood flour, which is abundantly available in Niger State in Nigeria, it is important that an insightful understanding of the surface modification treatment parameters is gained if the performance of WPCs to be optimized. This study makes use of *D. oliveri* wood flour compounded with virgin high density polyethylene (vHDPE) to produce WPCs for building and structural applications at reduced cost. It is believed that the incorporation of *D. oliveri* wood flour into vHDPE offers the possibility of increasing its economic value and developing the Nigerian industrial sector. This waste wood flour was treated with sodium hydroxide (NaOH) to improve its interfacial adhesion with the polymer matrix. vHDPE was chosen as the preferred matrix for this study due to its usability in a variety of applications such as packaging and automobile parts. Besides, vHDPE is hard, opaque and can withstand high temperatures ranging from 125°C to 130°C. Moreover, analysis of the physical, chemical, mechanical, microstructural, and thermal properties of the WPCs were carried out to explore the effects of modifying the reinforcing wood fiber in varying concentrations and time of treatment of NaOH solution. The treated wood fibers and vHDPE were compounded by using compression molding. Thereafter, characterization of WPCs samples detailing their functional properties are carried out with a view to determining the optimum concentration of NaOH and duration of treatment suitable for producing WPCs.

## MATERIALS AND METHODOLOGY

### Materials

Waste *D. oliveri* wood flour was collected from a sawmill in Minna, Niger state, Nigeria. The vHDPE having a

density of 0.95 g/cm<sup>3</sup>, melt flow index (12 g/10 min, 190°C/2.16 kg) as measured by ASTM D1238 and molecular weight (28 g) was supplied by Echas Scientific Company, Kaduna, Nigeria. Crystalline sodium hydroxide (NaOH), obtained from Echas Scientific Company, Kaduna, Nigeria, was used in the preparation of the alkali reagents. Dilute hydrochloric (HCl) acid was obtained from Kaduna Polytechnic Soil Science Laboratory, Kaduna, Nigeria and used to rinse the treated wood flour to remove excess alkali from the treated wood fiber. The removal of excess HCl was confirmed using litmus paper.

### Methodology

**Preparation and Alkalization of Wood Fiber.** Wood flour of *D. oliveri* was dried in the sun for 48 h to remove the excess moisture content and then stored in black polythene bags at ambient temperature of 21°C. The wood flour was then soaked in hot water at 100°C for 1 h to open up the pore spaces of the hydrophilic wood fiber. At the expiration of 1 h, the wood flour was thoroughly washed and then air dried for 5 days at ambient temperature of 21°C. With the use of a hand oscillating multi-deck screen classifier, the wood flour was separated into various particle sizes and distribution of +75–150 µm, +150–210 µm, +210–300 µm, +300–425 µm, and +425–600 µm. Apart from the fact that the majority of previous research in the field of WPCs made use of wood flour of particle sizes less than 800 µm [16–18], wood flour having particle size and distribution of +425–600 µm was also selected based on the largest quantity of this portion obtained after sieving. They were then treated with varying concentration of NaOH (2 wt%, 4 wt%, 6 wt%, 8 wt%, and 10 wt%). Pre-determined amounts: 20 g, 40 g, 60 g, 80 g, and 100 g of NaOH crystals were introduced into 1000 cm<sup>3</sup> distilled water to give the required 2 wt%, 4 wt%, 6 wt%, 8 wt%, and 10 wt% NaOH concentration, respectively. The selected wood flour, 6.72 g, was measured into each of five different beakers designated A, B, C, D and E containing 2 wt%, 4 wt%, 6 wt%, 8 wt%, and 10 wt% concentrations of NaOH, respectively. The wood flour was left immersed in the alkali solution for 30 min after which it was quickly washed with dilute hydrochloric acid (HCl). Subsequently, the wood flour was thoroughly washed with distilled water. The rinsed wood flour was dried at ambient temperature of 21°C for 5 days.

**Compounding and Preparation of Wood Polymer Composite Samples.** Upon drying, the wood fiber was compounded with the vHDPE and the composite samples were produced. The mold used for the preparation of the WPC samples was prepared from a 240 × 90 × 15 mm mild steel plate fitted with both top and base plates. To enhance complete and easy removal of the composites from the mold, a polythene sheet was laid on the clean and dry mold before the compression of the compounded

wood fiber and the vHDPE into the mold at the ratio of 50:50. The ratio 50/50 was chosen for full utilization of the wood flour as well as for the optimization of the mechanical properties of the WPCs as the alkalization treatment parameters were varied [19]. The mold was clamped in a preheat stand at 70°C to prevent the molten mixture from rapid solidification that could create differential cooling zones (core and the surface) in the composites, which could deform the product after injection. A compression molding machine, Carver Hydraulic Press manufactured by Loctite Corporation (Mexico), was used in producing the WPCs. This involved pouring both the crystals of vHDPE and modified wood flour at a 50/50 ratio into the hopper of the compression molding machine. The components were thoroughly mixed to give a homogenous mixture.

The vHDPE was heated to 140°C in the barrel when the modified wood fiber was added and stirred. The homogeneous mixture was channeled through the heated barrel at different temperatures (100°C–120°C–160°C) to soften and melt the matrix. The temperature was optimized at 160°C to prevent the degradation of wood flour. Generally, wood flours start degrading at 190–210°C. At 160°C, the mold was connected to an injector through a nozzle. The molten mixture was injected into the mold through the nozzle at 160°C when the pressure was at 2.94 MPa within 5 s. After each compression, the mold was cooled by compressed air for 5 min, and then unclamped by loosening the bolted ends and scrapping the extruded part of it to give the required geometry for functional properties testing. The choice of compression molding technique is premised on the fact that it is expected to impart more desirable mechanical properties into WPCs in comparison to extrusion and injection molding [20]. Due to reduced shear stress associated with compression molding in comparison to twin screw extrusion; compression molding was adopted for this study because it allows reduced thermal degradability of HDPE molecules and reduced damage of wood flour particles. Therefore, WPCs produced through compression molding are expected to have good density with little or no voids within its structure [20]. Rectangular samples produced were machined to various geometries in accordance with the requirements of the various physical and mechanical tests as shown in Fig. 1. The samples were investigated via mechanical, physical and microstructural tests and the effect of alkaline concentration on the properties of the resulting WPCs was used to conclude on the optimum concentration for the WPC treatment. This procedure was then repeated for wood flour soaked in the optimum concentration of NaOH obtained for 30, 60, 90, 120, and 150 min. All the treated samples were then taken for compression molding.

**Characterization of Wood Polymer Samples.** *Fourier Transform Infrared Spectroscopy.* FTIR spectroscopy with a microscope was used to study the stress transfer at



FIG. 1. A typical samples produced and machined as a specimen for tensile testing. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

the molecular level. The FTIR equipment; SHIMADZU FTIR—8400s model (Japan) was used to obtain the structural analysis of the untreated/treated wood flour and WPCs samples. The FTIR was used in the transmission mode, with a resolution of  $4\text{ cm}^{-1}$  scans in the range of  $4000\text{--}400\text{ cm}^{-1}$ . FTIR was used to detect the molecular vibration to analyze the distribution of functional groups of the samples.

**Determination of Density.** The density of the WPC samples was determined by the water displacement method. The mass of each specimen sample was measured using a digital weighing balance with 0.01 g accuracy. Similarly, the volume of water displaced while the weighed specimen was completely immersed in a beaker of distilled water was also recorded. Average density was determined from three measurements with the standard deviation about the mean value also reported. The density of the specimen was calculated using Eq. 1:

$$\rho = m/v \text{ gcm}^{-3} \quad (1)$$

where  $\rho$ =density;  $m$ =mass; and  $v$ =volume.

**Determination of Water Absorption Capacity (WAC).** The samples were prepared according to the ASTM D570 standards for moisture absorption tests [21]. Before immersion, the specimens were cleaned and weighed on a digital weighing balance with 0.01 g accuracy. The initial weight of the sample  $w_0$  was then recorded. The samples were dried in an oven at  $60^\circ\text{C}$  for 12 h to constant weight. This is with a view to ensuring that any moisture present in the samples prior to WAC test did not affect the result of the absorbed water. Prior to immersion of the samples in water, they were kept in a dessicator under vacuum at room temperature. Thereafter, moisture absorption tests were conducted by immersing the specimens in a distilled water bath at temperature of  $28^\circ\text{C}$  and hot water at  $60^\circ\text{C}$ . At each of these temperatures, the absorption of the samples was observed for 24 h. During the immersion process, the specimens were allowed to rest on the edge at the bottom and completely immersed. For each observation at the end of the 24 h, the specimens were wiped with dry cloth to remove excess moisture and weighed ( $w_t$ ). In the hot water immersion process, after each immersion, the specimens were brought to room

temperature by dipping in cold water for 10 min and then weighed. Average WAC was determined from three measurements with the standard deviation about the mean value also reported. The moisture content  $M_t$ , absorbed by each specimen was calculated as follows:

$$M_t = [(w_t - w_0) \div w_0] \times 100\% \quad (2)$$

where  $w_0$ =dry weight of sample in grams;  $w_t$  = wet weight of immersed samples in grams;  $M_t$  = moisture content

**Determination of TS and Modulus of Elasticity (MOE).** The TS testing was carried out according to the ASTM D638 standard [22] by using the 5980 floor model Instron universal testing machine (United Kingdom) at a cross head speed of 5 mm/min. The initial length, width and thickness of the specimen were 90 mm, 20 mm, and 7 mm, respectively. The test was carried out at a room temperature of  $21^\circ\text{C}$  and relative humidity of  $40 \pm 2\%$ . Average TS and MOE were determined from three measurements with the standard deviation about the mean value also reported.

**Determination of Bending Strength (BS) and Modulus of Flexure (FM).** Rectangular samples with length, width and thickness of 105 mm, 20 mm, and 7 mm, respectively were prepared from WPC samples. These were used to carry out the bending test according to the ASTM D790 standard [23]. It was carried out with the 5980 floor model Instron universal testing machine (United Kingdom) at 50 rev/min. The data for bending tests were obtained from the digital screen of the Instron machine and recorded. Average BS and FM were determined from three measurements with the standard deviation about the mean value also reported.

**Microstructural Observation by Scanning Electron Microscopy (SEM).** SEM observations of unreinforced/untreated and treated wood fibers as well as tensile fractured surface of treated and untreated WPCs were carried out to study the effect of alkali treatment on the wood fiber–vHDPE interfacial adhesion. The fractured surfaces of the specimen obtained from tensile testing were analyzed using SEM equipment model JEOL 7500 SEM (Japan). All specimens were gold sputtered prior to SEM observations to avoid unnecessary charging. Each specimen was mounted on the aluminum holder of the microscope using double sided electrical conduction carbon adhesive tabs. An accelerating voltage of between 5 and 15 kV was employed. The SEM analyses of the different specimen were observed at 100 and 500 magnification.

**Thermogravimetric Analysis and Differential Scanning Calorimetry.** Thermal (TGA) gravimetric analyses were carried out on 5–10 mg of WPCs at a heating rate  $10^\circ\text{C}/\text{min}$  and constant flow rate of  $10\text{ cm}^3\text{ min}^{-1}$  in air atmosphere using a TA Instruments SDT Q600 (Germany). Thermal

decomposition of each sample occurred in a programmed temperature range of 30–800°C. Continuous records of weight loss and temperature were determined and analyzed with a view to determining the following TGA indices: onset temperature ( $T_{\text{onset}}/^\circ\text{C}$ ), peak temperature ( $T_p/^\circ\text{C}$ ), temperature at 50% mass degradation ( $T_{50\%}/^\circ\text{C}$ ), and degradation temperature range ( $^\circ\text{C}$ ).

Differential scanning calorimetry (DSC) was performed on a Mettler-Toledo DSC Q10 instrument (Germany). Samples sizes ranged from 3 to 10 mg, and were held at a heating rate of 10°C/min and temperature of 30–250°C. The experiments were performed under nitrogen atmosphere at a flow rate of 20 cm<sup>3</sup> min<sup>-1</sup>. Peak temperatures as well as melting and crystallization enthalpies were obtained from second heating and cooling cycles, respectively. These values made it possible to calculate the percentage crystallinity of the composites. Each of the data reported for TGA and DSC analyses represents an average of three runs with no significant variation between repeat tests.

## RESULTS AND DISCUSSION

### Fourier Transformation Infrared (FTIR) Spectroscopy

Figure 2a and b show the FTIR spectra of untreated and alkaline treated *D. oliveri* reinforced WPCs at different concentrations of NaOH solutions and times of treatment, respectively, whereas Fig. 2c highlights the chemical modification occurring in the unreinforced wood flour treated in 4 wt% NaOH for 150 min in comparison to the untreated and unreinforced wood flour. The absorbance at  $\nu$  3400 and 1030 cm<sup>-1</sup> assigned to the stretching vibrations of the hydrogen bonded hydroxyl (—OH) groups of cellulose and C—O groups of hemicelluloses and cellulose [24], respectively, increased with the alkalinization treatment of the wood fiber both at various concentrations of NaOH and time of treatment of the wood flour (Fig. 2a and b). The increase of the absorbance intensity upon alkali treatment suggests a reduction in inter-molecular/intra-molecular hydrogen bonding between the hydroxyl groups of cellulose and hemicellulose of *D. oliveri* wood flour [11, 25]. The absorption band at  $\nu$  1715 cm<sup>-1</sup> is attributed to the  $\nu(\text{C}=\text{O})$  of the carboxyl and ester groups in hemicelluloses of the wood fiber, which is prominent in raw wood flour as evident in Fig. 2c [26]. This absorption peak gradually decreased in the treated WPCs and the treated unreinforced wood flour as the concentration of NaOH and treatment time increased while it disappeared after 150 min of treatment in 4 wt% NaOH (Fig. 2b and c). The absorption peak at  $\nu$  1,645 cm<sup>-1</sup> in the untreated wood flour is associated with adsorbed water (Fig. 2c). The decrease in this peak intensity in the alkaline treated wood flour and its disappearance after 150 min is ascribed to the partial removal of hemicelluloses. The weak absorbance peak at  $\nu$  1463 cm<sup>-1</sup> is assigned to the in plane bending deformation of —CH<sub>2</sub> of lignin and hemicellulose. The —CH<sub>2</sub> bending

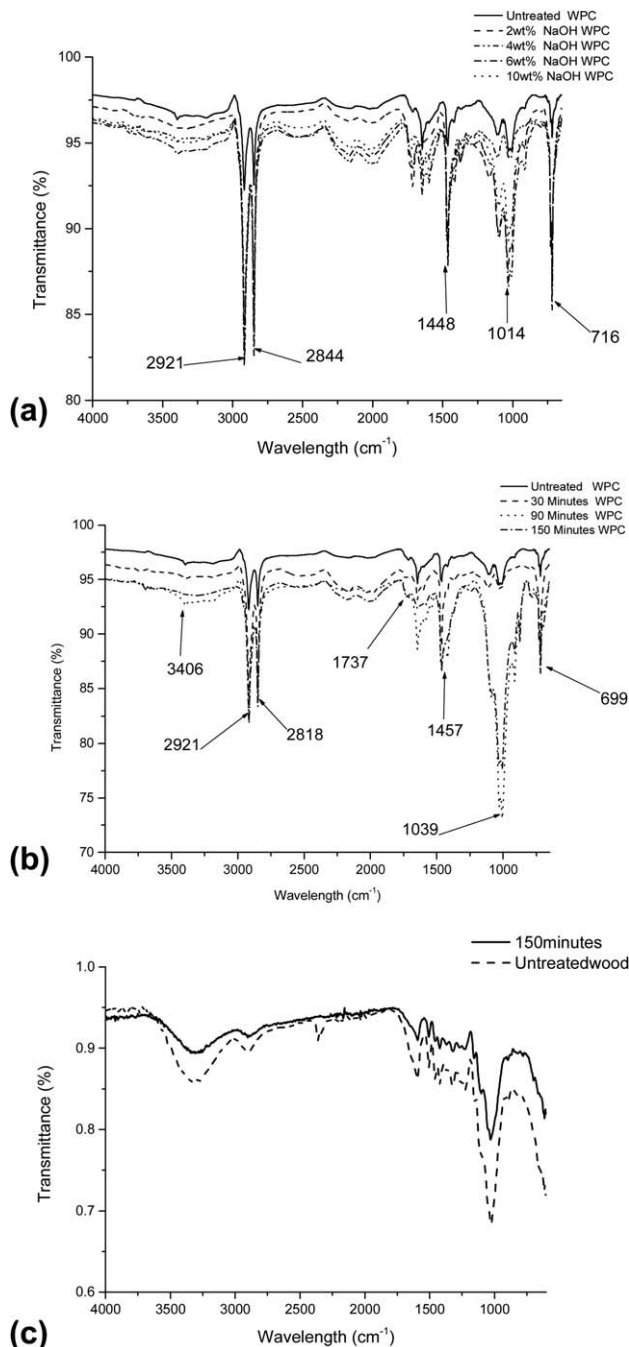


FIG. 2. FTIR spectra of untreated and alkaline treated *D. oliveri* wood fiber reinforced polymeric composites at different (a) concentrations of NaOH solutions, (b) times of treatment; and (c) untreated wood fiber and treated wood fiber modified in 4 wt% NaOH concentration for 150 min.

of cellulose component of the wood flour which remained unaffected after alkali treatment as seen in Fig. 2c [27] may increase the wood flour TS and consequently the mechanical properties of WPCs [28]. The sharp peaks observed between  $\nu$  2848 and 2915 cm<sup>-1</sup> indicate the main polymer chain of vHDPE (Fig. 2a and b). In summary, the alkaline treatment of wood fiber at varying concentrations and time of treatment changes the super molecular structure of the wood fiber (Fig. 2c) while the chemical structure is not significantly affected. Due to the

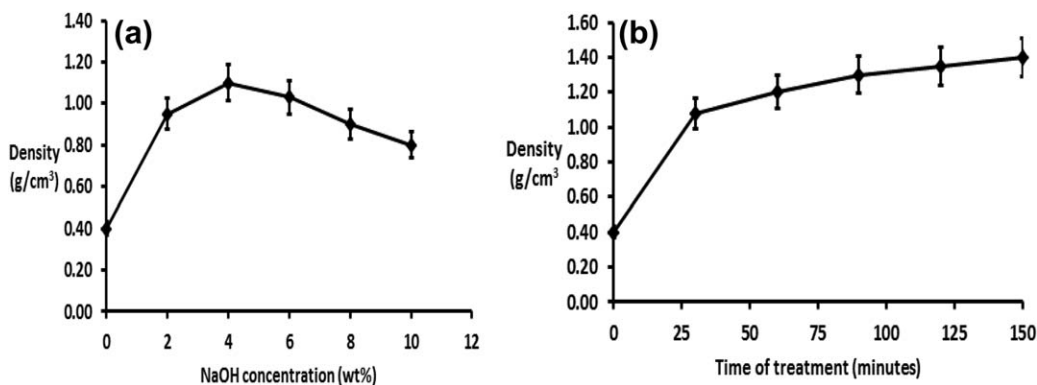


FIG. 3. Relationship between the density of the wood polymer composites and varying (a) concentration; (b) time of treatment in 4 wt% concentrated NaOH solution.

intra- and inter-fibrillar swelling, the accessibility of wood fiber changes with the removal of hemicelluloses and other alkali soluble materials from the surface of the wood fiber, thus increasing the softness of the fiber to allow better adhesion with the polymer matrix.

#### Density

From Fig. 3a, it can be seen that the density of WPCs increases to the optimum ( $1.095 \text{ g/cm}^3$ ) as NaOH concentration increases to 4 wt% while a subsequent increase in the concentration of NaOH resulted in reduction of the density at the same time of treatment (30 min). Figure 3b shows that the density of WPCs increases to  $1.40 \text{ g/cm}^3$  as the time of treatment of the wood flour in the 4 wt% concentrated NaOH solution increases to 150 min. Meanwhile, it is evident from both Fig. 3a and b that the density values ( $0.95\text{--}1.40 \text{ g/cm}^3$ ) of treated WPCs are higher than that of the untreated WPC ( $0.40 \text{ g/cm}^3$ ). Besides the untreated wood fibers acting as diluents within the vHDPE matrix such that it could not induce effective bonding with the vHDPE which consequently resulted in the production of highly porous WPCs ( $0.40 \text{ g/cm}^3$ ); the reduced density of the untreated WPC could possibly be attributed to incorrect processing or compounding of the untreated wood fibers and the virgin polymer. In addition, improvement in the density of WPCs noted upon reinforcing the vHDPE with alkaline treated wood fibers suggests that surface modification of wood fibers with NaOH resulted in the removal of non-cellulosic materials thereby promoting wood flour-matrix adhesion leading to improved densification of the WPCs as seen in Fig. 3a and b. Finally, treating *D. oliveri* wood flour with 4 wt% concentration of alkali solution for extended times (150 min) optimized the density of WPCs. This is expected to result in optimization of other physical, mechanical, and thermal properties of WPCs.

#### Water Absorption Capacity (WAC)

The water absorption behavior (WAC) of WPCs is a complex phenomenon which is a function of the

strength of the interfacial adhesion between the reinforcing wood fillers and the matrix; the degree of hydrophilicity of the fillers upon alkaline treatment and the fillers' morphological characteristics. Figure 4a and b reveal the outcomes of WAC tests as the concentration and time of treatment are varied, respectively. WAC decreases as concentration reaches 4 wt% (0.70% absorption) at a temperature of  $28^\circ\text{C}$  (Fig. 4a). Thereafter, WAC increases as concentration increases above 4 wt%. Figure 4b shows that water absorption decreased from 0.70% at 30 min to 0.35% at 150 min. Considering that the degree of densification of WPCs is indicative of the strength of the interfacial adhesion between the reinforcing wood fillers and the vHDPE matrix, hence, higher densification of WPCs results in stronger interfacial adhesion which lowers WAC in WPCs. The reported lowest value of WAC at  $28^\circ\text{C}$  (Fig. 4b) when the WPC was treated with 4 wt% NaOH concentration for 150 min confirms that both the density and interfacial adhesion of WPC were optimized at the same treatment parameter (Fig. 3b). This is because the WAC of WPCs is dependent upon the degree of reduction of the hydrophilicity of the *D. oliveri* wood flour upon treatment with NaOH solution. Therefore, the minimization of WAC at alkaline treatment parameters of 4 wt% and 150 min indicate the occurrence of least hydrophilicity in the wood fibers at which non-cellulosic materials were mostly effectively removed. This then imparted the strongest interfacial adhesion strength which is seen to have most effectively resisted water absorption in the WPCs produced with the optimal treatment parameters. Figure 4a and b establish that WPCs possess greater WAC at higher temperature ( $60^\circ\text{C}$ ) than at lower temperature ( $28^\circ\text{C}$ ) irrespective of the treatment parameters. These findings are in agreement with Mylsamy and Rajendran [3] who reported that WAC of WPCs significantly increases as the temperature of immersion is increased due to the destruction of the rigid cellulose structure by the percolating water molecules. Therefore, as WAC of WPCs increases at higher temperature, the interlocked water molecules attains greater propensity to

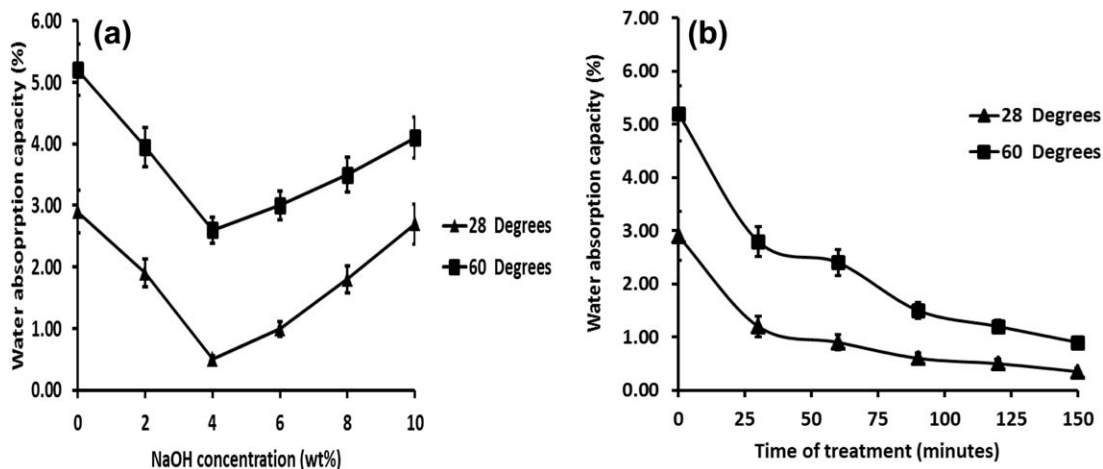


FIG. 4. Relationship between the water absorption capacity of the wood polymer composites and varying (a) concentration; (b) time of treatment in 4 wt% concentrated NaOH solution.

actively attack the interface, resulting in debonding of the wood fiber and the matrix internally in the composite [3].

### Mechanical Properties

**Tensile Strength and Moduli of Elasticity.** Figure 5a and c show that TS and MOE increase with NaOH concentration up to 4 wt% while a further increase in NaOH concentration leads to their reduction. Meanwhile, both TS and MOE increase with treatment time (Fig. 5b and d). Analysis of Figure 5 establish that TS and MOE of WPCs increased to 16 MPa and 96.10 MPa, respectively, when treated with 4 wt% NaOH concentration for 150 min (Fig. 5b and d) in comparison to the optimal values of 11 MPa and 34 MPa obtained when treated in 4 wt%

NaOH concentration for 30 min (Fig. 5a and c). It is clear from Fig. 5 that TS and MOE treated WPCs are higher than that of untreated WPCs.

**Bending Strength and Flexural Modulus.** Analysis of the effects of NaOH concentration and time of treatment on BS (Fig. 6a and b) and flexural modulus (FM) (Fig. 6c and d) of WPCs follows the same trend already reported for TS and MOE. Optimum values of BS and FM of WPCs were 103 MPa and 165.10 MPa, respectively, when wood flour was treated with 4 wt% NaOH for 150 min (Fig. 6b and d). This outcome again confirms that best interfacial adhesion of the vHDPE matrix with the reinforcing wood flour can be obtained when WPCs are treated with 4 wt% NaOH concentration for 150 min.

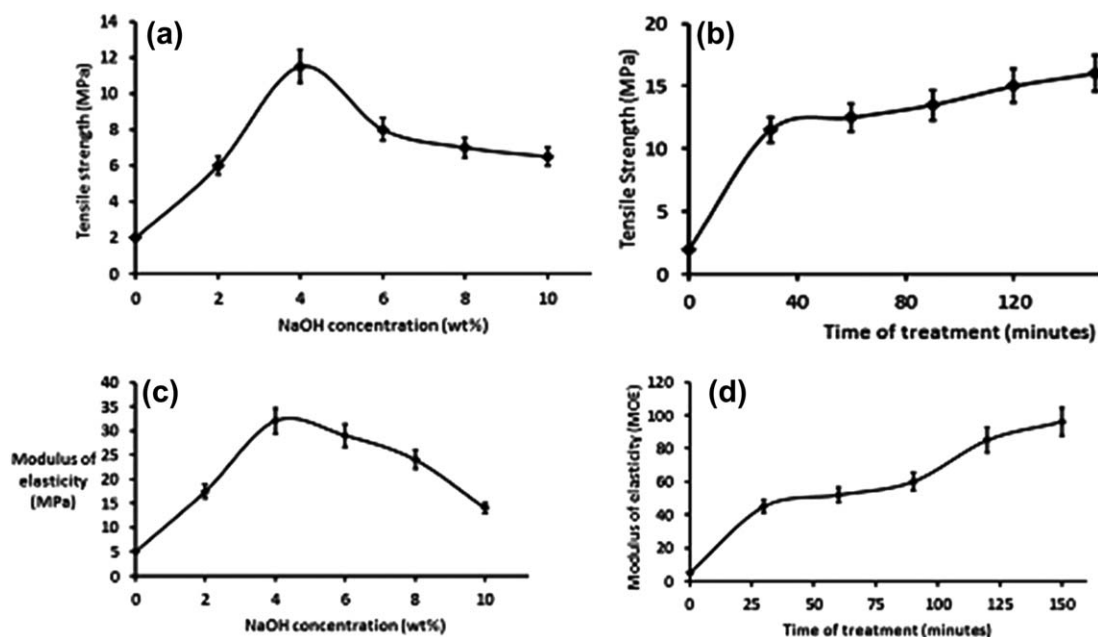


FIG. 5. Relationship between tensile properties of wood polymer composites and varying concentration (a, c) and time of treatment (b, d) in 4 wt% concentrated NaOH solution.

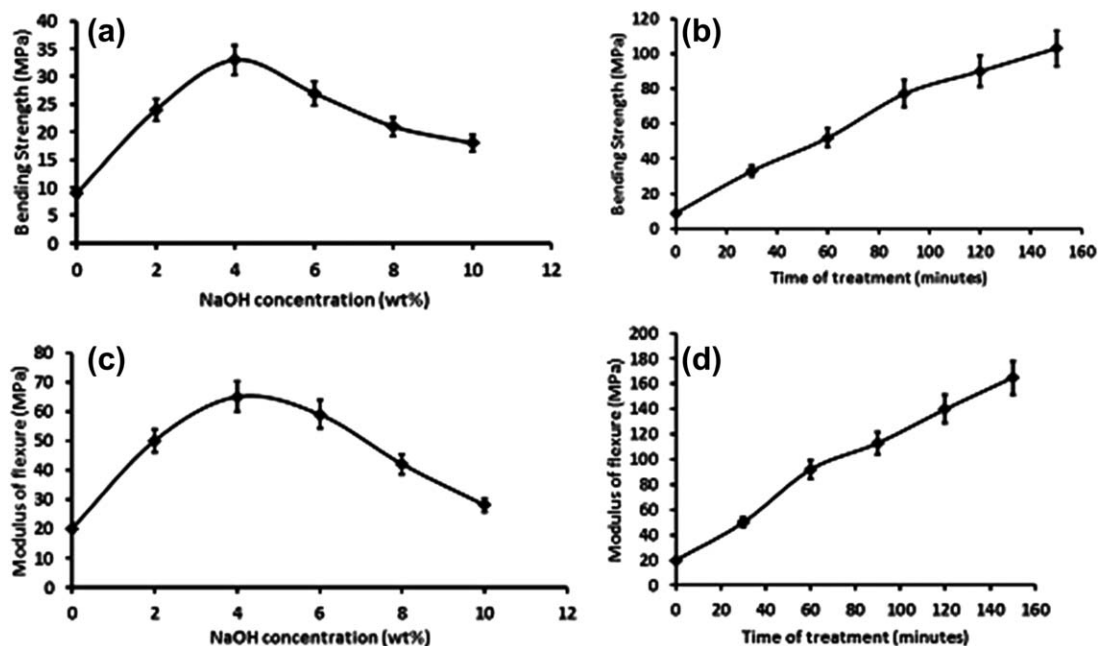


FIG. 6. Relationship between bending/flexural properties of wood polymer composites and varying concentration (a, c) and time of treatment (b, d) in 4 wt% concentrated NaOH solution.

Alkaline treated WPC samples have higher BS and FM in comparison to the untreated WPC samples [29].

#### Fiber/Matrix Bond Mechanism

The SEM images shown in Fig. 7 elucidate the variations in the surface morphologies of *D. oliveri* wood fibers upon treatment with an alkaline solution (Fig. 7b–d) in comparison with the untreated fibers (Fig. 7a). This approach had been undertaken to highlight how the surface morphologies of alkaline treated fibers influenced the fiber/matrix bond mechanism in the resultant WPCs. Figure 7b–d present evidence of alteration in the wood fibers' microstructure as a result of its alkaline modification at varying time of treatment in comparison to the porous structure characterizing the untreated fiber (Fig. 7a). It is evident that microstructural pores of *D. oliveri* wood fibers became clearer consequent upon the leaching of the waxy cuticle layer as the alkaline time of treatment increases up to 150 min when soaked in 4 wt% NaOH concentration (Fig. 7b–d). The surface of the wood fiber appeared curly and soft upon alkaline treatment. This consequently resulted in the enhancement of mechanical interlocking at the interface.

SEM fractographs of WPCs produced from wood flour treated in NaOH solution of varying concentration at different time durations are presented in Figs. 8 and 9, respectively. Observation of the fractographs of tensile test samples of WPCs was undertaken to clarify the importance of fiber adhesion on the physical and the mechanical test results while correlating the findings to the nature of alteration of surface morphologies of treated

wood fibers (Fig. 7). Figures 8a and 9a reveal that most regions on the fractographs of untreated WPCs appear to be free of vHDPE adhering to the wood flour (region AA), thus indicating poor flour–matrix adhesion. There are also few regions where the untreated wood flour partly adhered to the vHDPE (region BB in Figs. 8a and 9a), thereby, suggesting the occurrence of weak fiber matrix adhesion. Moreover, the untreated WPC is seen to have failed via fiber pull-out from the vHDPE matrix and display what appears to be a considerable amount of impurities/debris on the wood fiber surface (portion arrowed DD in Fig. 8a).

According to Mysamy and Rajendran [3] and Venkateshwaran et al. [30], the presence of impurities/debris on the wood flour surface (Fig. 7a) is responsible for poor wood flour–matrix interface adhesion. The compatibility between the untreated wood flour and the matrix is reduced and this lowered the mechanical properties when compared with the treated wood fiber composites. Furthermore, the occurrence of poor stress transfer between the vHDPE matrix and the untreated wood flour could be attributed to the formation of voids within the microstructure of the WPC which resulted from the lack of interaction at the interface [31]. The occurrence of voids between the matrix and the untreated wood flour could also be explained by weak interactions between the vHDPE matrix and the untreated wood fiber which promotes large shrinkage experienced by the vHDPE matrix as its specific volume reduces upon cooling after compounding with the wood fiber [31].

Observation of the fractographs of alkaline-treated WPCs suggests a reduction in the degree of occurrence of



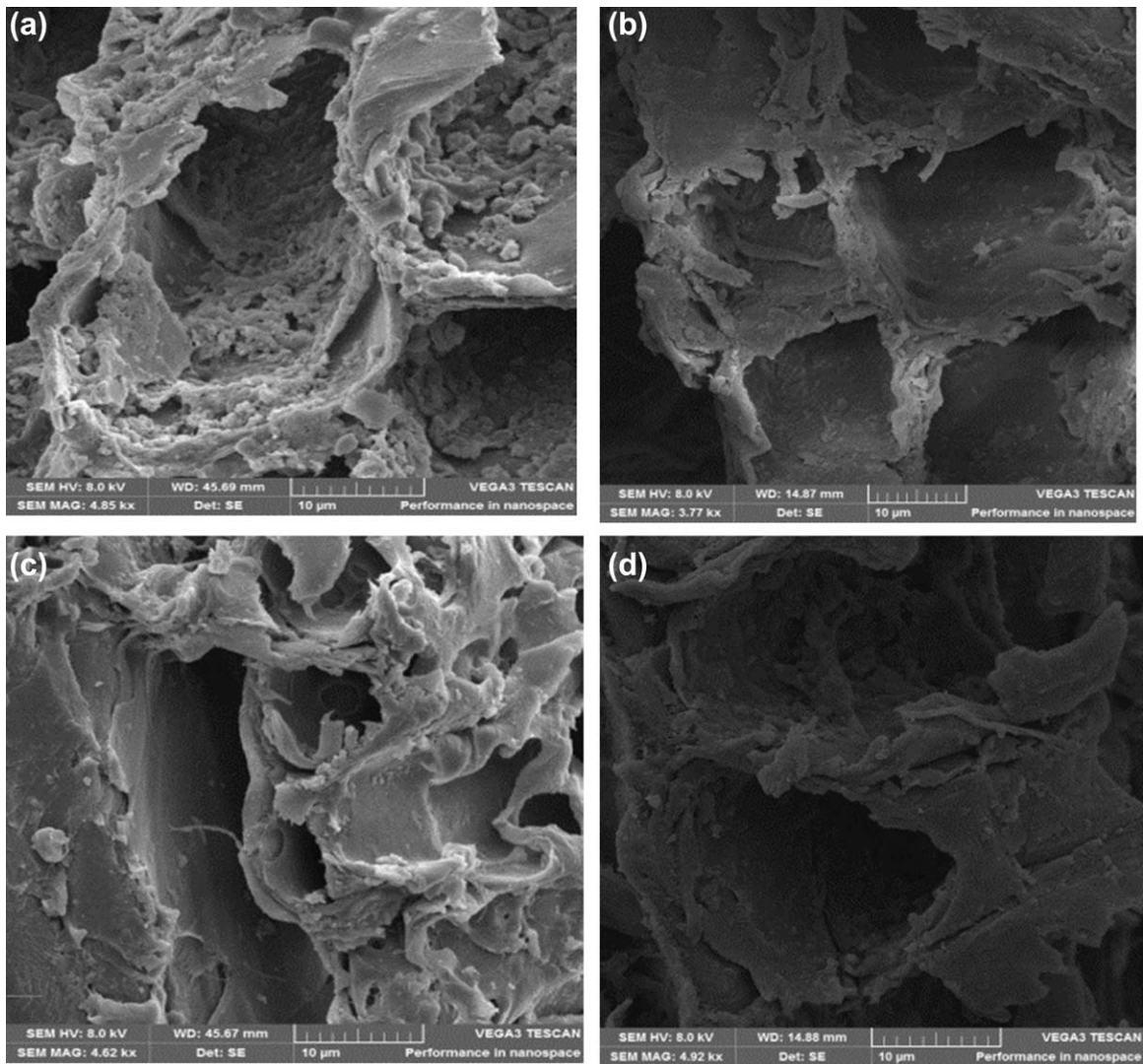


FIG. 7. SEM images showing the surface morphologies of (a) untreated wood fibers; and wood fibers treated for (b) 30 min; (c) 90 min; (d) 150 min in 4 wt% NaOH solution.

uneven matrix cracking and void; an indication of the good laminar bonds between the treated wood flour and the vHDPE (Figs. 8b–f and 9b–f). The regions labelled CC and XX on the fractographs indicate occurrence of uneven matrix cracking and void in the WPCs microstructure and clean surfaces of wood fiber particles (pull-out), respectively. The preponderance of regions CC and XX in the composite's microstructure suggests occurrence of poor interfacial adhesion between the wood flour and the vHDPE matrix. Regions labelled YY on the fractographs indicate rough portions of the matrix found on the surface of the wood fiber particles, thereby indicating better adhesion. A close observation of the regions labelled YY reveals that more tearing of the flour occurred on the fractographs coupled with the absence of voids resulting from the pulled out wood flour particles. The existence of region YY suggests the occurrence of effective chemical bonding of the vHDPE matrix to the wood flour's surface via alkaline treatment. In this region, wood flour particles

are evenly distributed on the composite surfaces with the fractured surfaces covered up by the matrix materials.

In the non-optimized WPCs, flour fracture was noted to be very small while the predominant failure mechanism was flour pull-out. Hence, the voids created on the failed surfaces of these WPC samples could be ascribed to flour pull-out. When the optimized WPC sample produced with wood flour treated with 4 wt% NaOH for 150 min was subjected to tensile load, the initiation of cracks at the fiber-matrix interface could be attributed to the heterogeneous connection. Consequently, cracks were propagated along the matrix because the TS of the matrix is lower than that of WPCs [32]. Thereafter, the adjacent wood flour particles in the failure plane of the optimized WPC sample were interconnected by the cracks. As a consequence of the fracture of the matrix in the failure plane of the optimized WPC sample, the applied tensile load was totally carried by the wood flour. With time, the wood flour were fractured in the failure plane of the

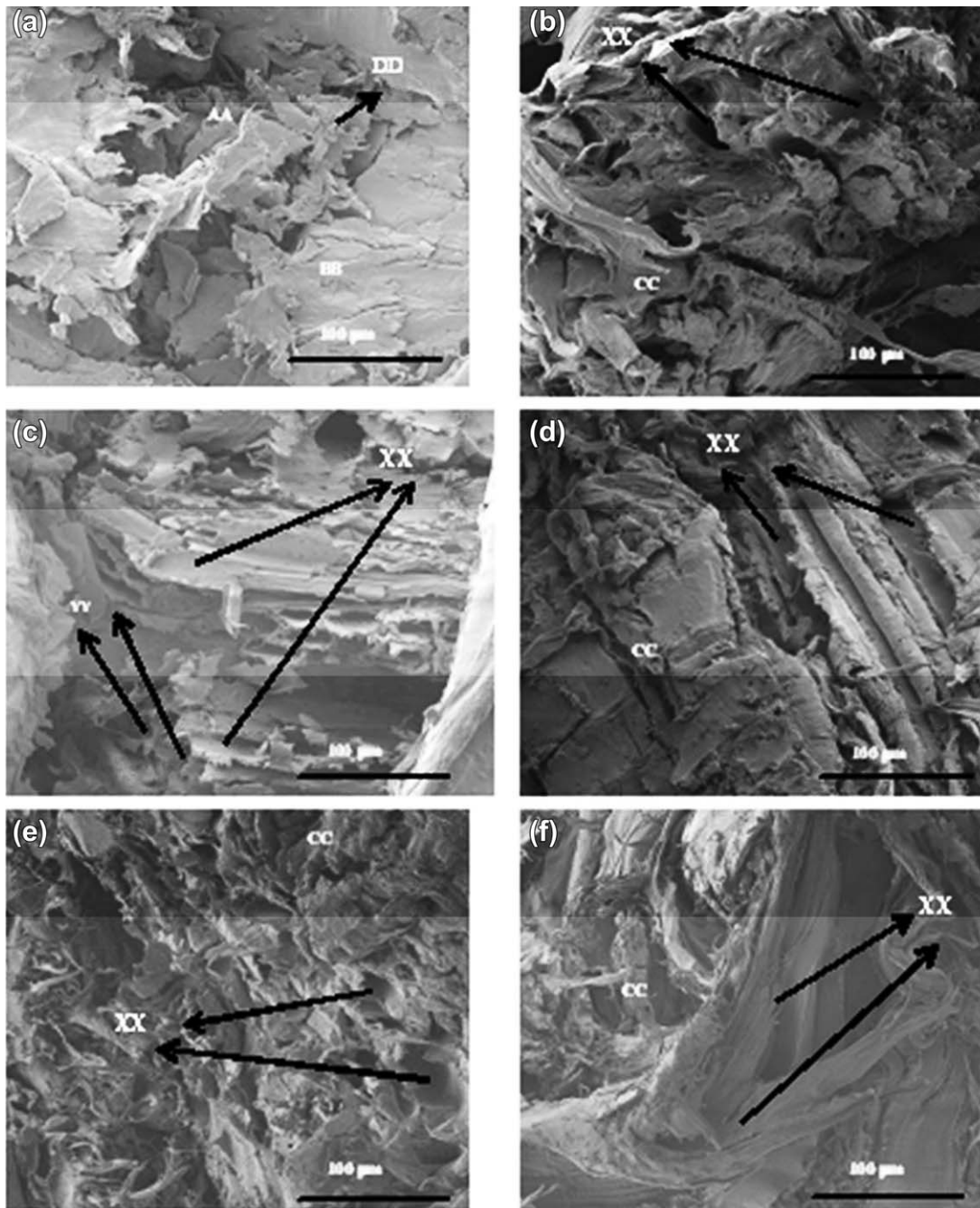


FIG. 8. SEM fractographs of wood polymer composites reinforced with wood fiber treated with varying concentration of NaOH solution for 30 min (a) Untreated (b) 2 wt% (c) 4 wt% (d) 6 wt% (e) 8 wt% (f) 10 wt%.

optimized composites, when the applied tensile load was greater than the ultimate TS of the wood flour particles. Similar to findings from Sreenivasan et al. [32], it can be seen from Fig. 9f that wood flour–matrix debonding and wood flour pull-out were hard to occur in the optimized WPC samples as a result of the existence of good interfacial bond between the wood flour particles and the vHDPE matrix. In the optimized WPC samples, matrix and wood flour fractures were the predominant failure mechanisms.

A study of the fractographs of WPCs (Figs. 8b, d–f; 9b–d and e) treated with NaOH concentration and time other than 4 wt% and 150 min, respectively, reveals occurrence of regions CC and XX in greater degree in comparison to WPCs samples treated for 150 min in 4 wt% NaOH concentration (Fig. 9f). Moreover, findings on the fractographs compares favorably with density, WAC and mechanical data of WPCs such that the sample treated with 4 wt% NaOH concentration for 150 min have the most desirable physical and mechanical

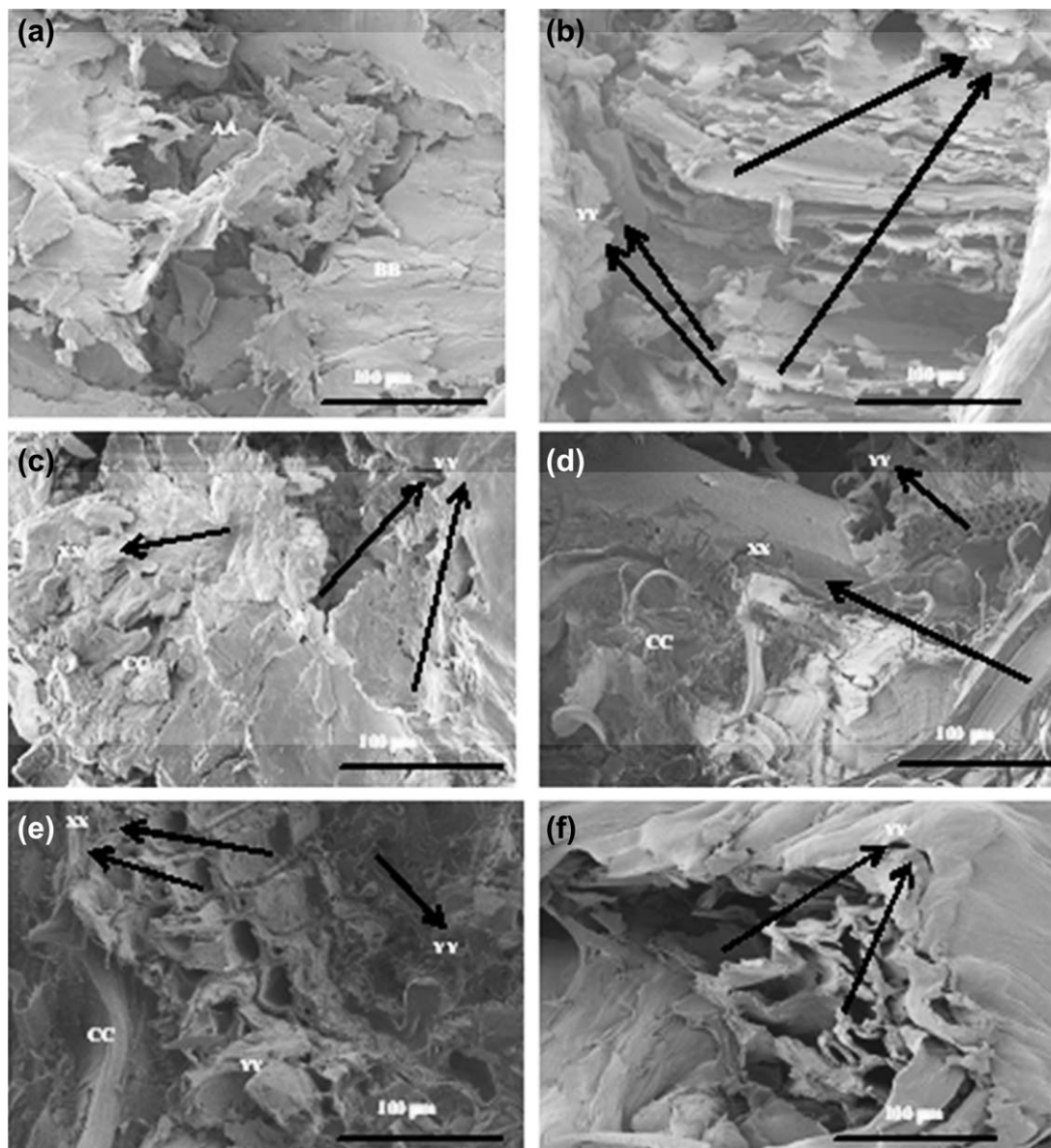


FIG. 9. SEM fractographs of wood polymer composites reinforced with wood fiber treated with varying time of treatment at 4 wt% NaOH concentration (a) Untreated (b) 30 (c) 60 min (d) 90 min (e) 120 min (f) 150 min.

properties (Figs. 2, 5, and 6). The presence of voids in the fractographs of WPCs treated with parameters other than 4 wt% NaOH concentration and 150 min indicates a lower degree of densification and the development of less desirable physical and mechanical properties during processing due to inappropriate choice of concentration of NaOH solution or time of treatment of the wood fiber.

Meanwhile, optimization of the physical and mechanical properties of WPC samples when the polymer matrix was reinforced with wood fiber treated with 4 wt% NaOH concentration for 150 min could be attributed to the removal of non-cellulosic materials and impurities (as seen in Fig. 7d) which remains dispersed in the interfibrillar region of the wood [15, 33]. In this instance, treatment of wood flour enhanced their capability for re-

arrangement along the direction of tensile deformation since the inter-fibrillar region becomes less dense and less rigid due to the removal of hemicellulose. Therefore, when the fibers are stretched, re-arrangement amongst the fibrils results in higher stress development due to better load sharing [8]. Consequently, as hemicellulose leaches out due to the alkali treatment, close packing of the cellulose chains in fibers takes place owing to the release of internal strain leading to improved physical and mechanical properties [34]. In addition, the increasingly rough surface or grooves on the surface of wood flour (Fig. 7d) enhanced wood flour–matrix adhesion by providing additional sites for mechanical interlocking and eventually leading to better physical and mechanical properties of the WPCs [35]. Meanwhile, when wood fibers were

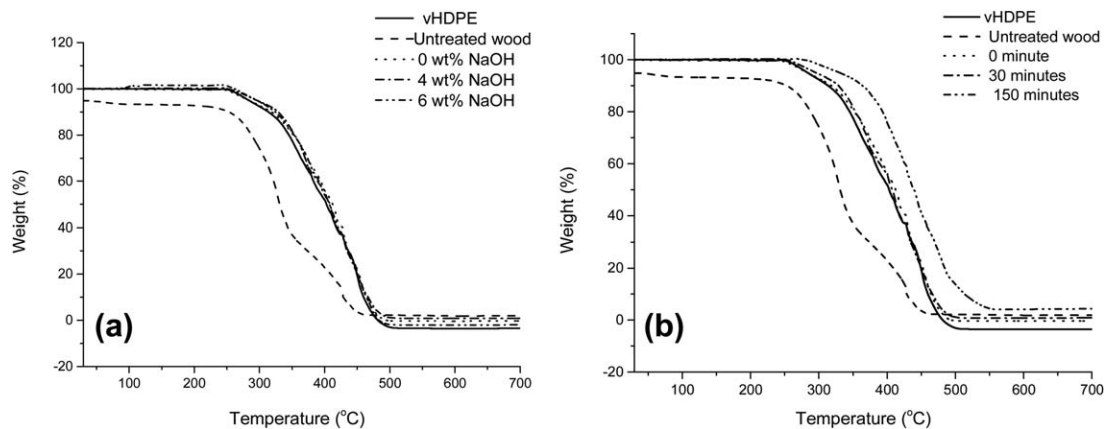


FIG. 10. TGA of vHDPE, untreated-unreinforced wood, untreated WPC, and treated WPCs at varying (a) concentration and (b) time of treatments in NaOH solution.

treated with NaOH concentrations lower than 4 wt% or for treatment times less than 150 min, the physical and mechanical properties of WPCs deteriorated (Figs. 5 and 6) due to ineffective removal of non-cellulosic materials (Fig. 7b–c). However, when wood flour was treated with NaOH concentrations above 4 wt%, the physical and mechanical properties of WPCs gradually decreased, possibly as a result of the increasing brittleness of the wood fiber [9]. The main structural components of the wood fiber were attacked, resulting in the depolymerisation of native cellulose and also excessive delignification of the wood fiber which affects its strength.

### Thermal Stability

Figure 10 shows the thermogravimetric analysis (TGA) of the vHDPE, the untreated-unreinforced wood, and the WPCs reinforced with untreated and NaOH treated wood flour. The thermograms for the untreated-unreinforced wood and all the composites in Fig. 10 exhibit two mass loss peaks. The first mass loss peak occurs below 100°C, which has been attributed to the evaporation of moisture from the wood fibers in all the WPC samples. The second peak, within the range of 205–380°C, could be ascribed to the degradation of hemicellulose, cellulose, and lignin in all the WPC samples as a result of the oxidation of char residue generated in the first mass loss peak [36]. According to Bryne and Nagle [37], Yang et al. [38] and Lafia-Araga and co-investigators [39], lignocellulosic materials which are chemically active, decompose thermo-chemically in the range of 150–500°C, whereas hemicellulose degrades between 150°C and 350°C, cellulose between 240°C and 350°C, and lignin between 250°C and 500°C. The TGA analysis of vHDPE reveals a single-mass loss step with peak degradation temperature at 455°C suggesting the thermal and oxidative decomposition of saturated and unsaturated carbon atoms in PE.

It is clear from Fig. 10 that WPCs (240°C and 515°C) present a degradation temperature higher than that of their individual components: untreated-unreinforced wood

(350–490°C) and vHDPE (220–410°C) which decompose with a lower initial mass loss. This finding implies the occurrence of an anti-degradation synergy between the vHDPE and the alkaline treated *D. Oliveri*. Hence, the alkaline treated wood flour and the polymer matrix protected each other mutually since the degradation of the WPCs is delayed (Fig. 10) in comparison to the degradation kinetics of the individual components [36]. Meanwhile, improved thermal stability of the composites could also be accounted for by the development of a char skin-layer at the earlier stage of degradation which acted as barrier to the onset of thermo-oxidation mechanism. This barrier effect is elucidated by the mass loss delay noted in Fig. 10 for the WPCs which resulted from the slow-down of diffusion of volatile thermo-oxidative products and oxygen to the polymer. Furthermore, the onset temperature ( $T_{\text{onset}}$ ) and degradation temperature values of treated composites increased appreciably over that of the untreated composites and untreated wood flour (Table 1) with the untreated WPCs degrading with a lower mass loss in comparison to treated composites (Fig. 10). This indicates that the incorporation of alkaline treated wood flour particles into vHDPE resulted in the formation of composites with thermal stability better than that of the untreated composites and untreated unreinforced wood flour. In agreement with Kotalainen et al. [40]; the reported improvement in thermal stability of alkaline treated WPCs may also be ascribed to the removal of hemicellulose from the wood fiber by alkaline treatment, which is envisaged to have possibly improved its thermal properties by reducing its heat transfer co-efficient [41].

WPCs treated with 4 wt% NaOH concentration for 150 min have the highest onset degradation temperature ( $T_{\text{onset}}$ ) value of 375°C in comparison to untreated-unreinforced wood, untreated WPCs, or WPCs treated with NaOH concentration and time other than 4 wt% and 150 min (Table 1). The optimization of  $T_{\text{onset}}$  is indicative of the maximization of the improvement in the thermal stability of the WPCs produced with specified treatment parameters [42]. Table 1 further reveals that both  $T_{50\%}$

TABLE 1. TGA parameters of neat vHDPE, untreated-unreinforced wood, untreated and treated composites at varying concentration and time of treatment of NaOH..

Entry	Material	Treatment time (min)	Onset temperature ( $T_{\text{onset}}/^{\circ}\text{C}$ )	Peak temperature ( $T_{\text{p}}/^{\circ}\text{C}$ )	$T_{50\%}$ ( $^{\circ}\text{C}$ )	Degradation temperature ( $^{\circ}\text{C}$ )
1	vHDPE	0	425	460	450	350–490
2	Untreated Wood	0	220	340	340	210–410
3	0 wt% NaOH WPC	0	350	465	445	310–485
4	4 wt% NaOH WPC	30	365	490	470	250–510
5	6 wt% NaOH WPC	30	360	485	465	255–500
6	4 wt% NaOH WPC	150	375	495	480	240–515

and  $T_{\text{p}}$  increased up to 4 wt% NaOH concentration, then decreased when WPCs were treated with NaOH concentration above 4 wt%. This further reinforces the fact that 4 wt% NaOH concentration may be the optimum concentration at which the thermal stability of NaOH treated WPCs produced from *D.niella oliveri* wood flour is highest. Hence, it may be inferred that reinforcing vHDPE with alkaline treated wood has increased the thermal resistance of the neat matrix [43]. However, as the concentration of NaOH increases above 4 wt%, the reduction noted in  $T_{\text{onset}}$ ,  $T_{50\%}$  and  $T_{\text{p}}$  could be attributed to the reduced thermal stability of the treated wood flour compared with optimal composite sample [44]. Hence, non-optimization of the thermal properties of WPCs at NaOH concentrations, other than 4 wt% could partly be explained by the non-effective removal of non-cellulosic materials at concentrations lower than 4 wt% and depolymerisation of the native cellulose resulting in excessive delignification when wood fiber was treated with NaOH concentrations greater than 4 wt%.

Furthermore, it is evident that increasing the time of treatment up to 150 min appears to impact more positively on the thermal stability of the composites at the optimum NaOH concentration of 4 wt% (Table 1). Possible explanation for this behavior is that lignin; the most thermally stable component in wood flour will not degrade appreciably when treated with 4 wt% NaOH for 150 min. Consequently, alkaline treatment of wood flour at 4 wt% NaOH concentration for 150 min, which could significantly degrade other components of wood, seems to have little or no decomposition effects on lignin [40]. As a result, wood treated at 4 wt% NaOH for 150 min has higher lignin content and leads to more thermally stable composites (Table 1).

DSC thermograms, corresponding to the second heating and cooling scans of vHDPE, untreated WPC, and treated WPCs at varying NaOH concentration and time are presented in Fig. 11. The relative percentage crystallinity ( $X_{\text{c}}$ ) of WPCs was determined using Eq. 3.

$$X_{\text{c}} = \frac{\Delta H_{\text{m}}}{(\Delta H_{\text{m}} W_{\text{m}})} \times 100\% \quad (3)$$

where  $X_{\text{c}}$  is the degree of crystallinity,  $\Delta H_{\text{m}}$  the heat of fusion,  $W_{\text{m}}$  the weight fraction of vHDPE in the compos-

ite, and  $\Delta H_{\text{m}}^*$  the heat of fusion for the fully crystalline vHDPE, taken as 293.6 J/g [45]. According to the data extracted from the DSC thermograms (Table 2), the cold crystallization ( $T_{\text{c}}$ ) and melting ( $T_{\text{m}}$ ) temperatures of vHDPE are 113.34 $^{\circ}\text{C}$  and 127.70 $^{\circ}\text{C}$ , respectively. Hence, the thermal behavior of vHDPE was significantly influenced with the incorporation of untreated *D. oliveri* flour as the  $T_{\text{c}}$  of the matrix increased to 118.50 $^{\circ}\text{C}$ , while the  $T_{\text{m}}$  increased slightly. This is attributed to the restricted chain mobility within the vHDPE matrix as a result of physical and/or chemical interactions at the wood flour/polymer interphase [46]. Moreover, the  $T_{\text{c}}$  of all the treated WPCs are slightly lower than that of the untreated WPCs while there are no significant changes in the  $T_{\text{m}}$  of treated WPCs irrespective of treatment parameters (Table 2). The reduction in the  $T_{\text{c}}$  of treated WPCs suggests that the by-products of the chemical reaction between NaOH and *D. oliveri* wood flour possess the capability to plasticize or act as nucleating sites for impurities within the vHDPE matrix [46]. Therefore, this indicates a resulting decrease in the cohesive energy density in the amorphous phase of the WPCs in comparison with untreated WPCs due to the rupture of the hydrogen bonds in vHDPE resulting from the interaction between the vHDPE matrix and the treated wood flour. This eventually is responsible for the rubbery properties and the increase of the mobility of the macromolecules which exist at lower temperatures [46, 47].

The degree of crystallinity of vHDPE in WPCs, determined from second melting enthalpy values (Table 2), increases with the alkaline concentration up to 4 wt% NaOH, whereas it reduced at higher concentrations (Table 2). Furthermore, the degree of crystallinity increases as the time of treatment of wood flour also increases. Therefore, optimum crystallinity (72.54%) was obtained when *D. oliveri* wood flour was treated with 4 wt% NaOH for 150 min. In addition, the degree of crystallinity in the untreated WPCs (49.16%) was lower than that of the treated WPCs (63.48–72.54%), while the degree of crystallinity of vHDPE was 58.77%. Therefore, untreated wood flour is capable of inhibiting a heterogeneous nucleation effect during the crystallization of vHDPE while the crystallization rate may be improved during phase transition as a result of alkaline treatment of wood flour incorporated

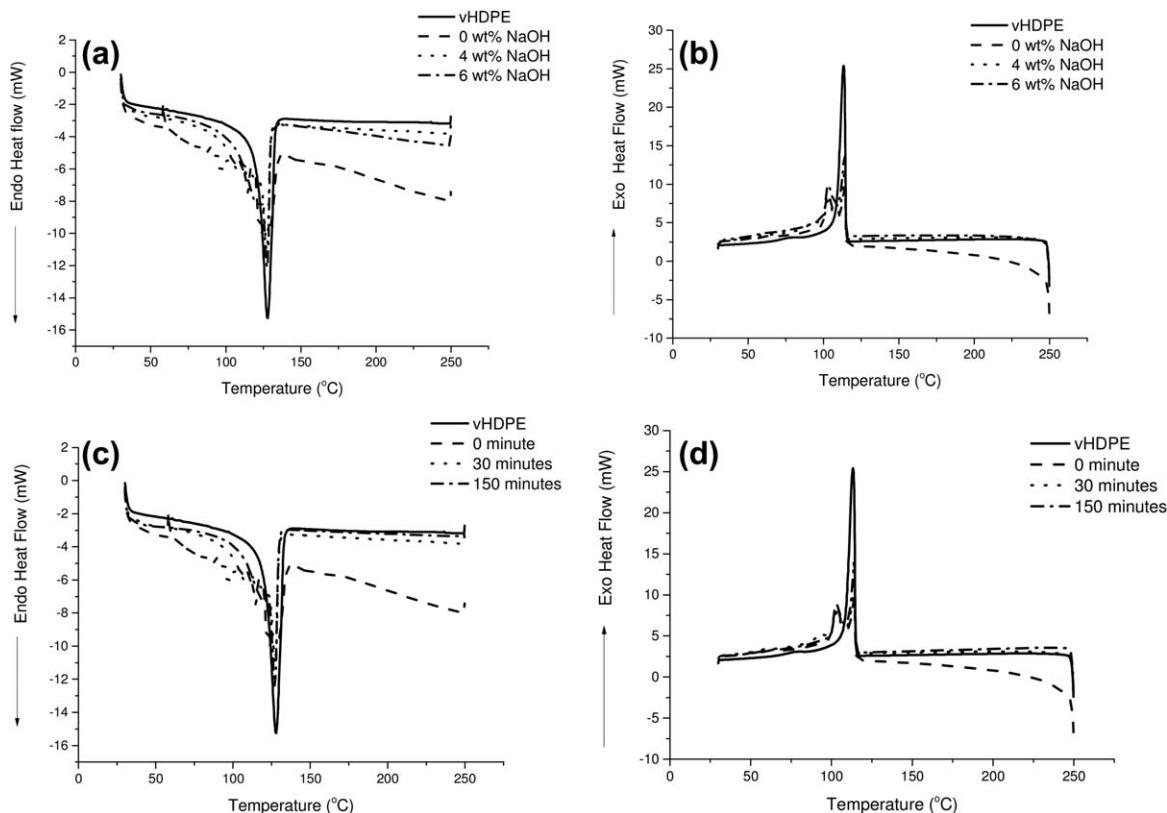


FIG. 11. (a, c) Heating and (b, d) cooling DSC thermograms of vHDPE, untreated WPC, and treated WPCs at varying NaOH concentration (a, b) and time of treatment (c, d) of NaOH solution.

into the matrix of vHDPE. According to Koo and Jang [48] and Shaha et al. [49], this could also explain the improved mechanical properties of treated WPCs over those of untreated WPC samples, because the mechanical properties of semi-crystalline vHDPE are a function of its degree of crystallinity. Higher fusion enthalpies ( $H_m$ ) obtained for treated WPCs over untreated WPCs points to the difficulty encountered by the untreated wood flour in forming the crystal nucleus which influences the phenomenon of crystal growth [46]. Reduced crystallinity observed at treatment parameters other than 4 wt% and 150 min shows that the treated wood flour act as diluents to the vHDPE matrix, and thus restricts crystal growth as confirmed by reduction in mechanical properties of WPCs [46]. This also accounts for increasing value of ( $H_c$ ) when wood flour was treated with NaOH concentrations and times other than 4 wt% and 150 min, indicating that higher energy is released during crystallite formation.

## CONCLUSIONS

Biodegradable WPCs produced from a vHDPE matrix reinforced with *D. oliveri* wood flour have been developed. The effect of NaOH concentration and time of treatment on the physical, mechanical, microstructural, and thermal properties of these biodegradable WPCs have been studied. The following conclusions can be drawn from this study:

1. Alkali treatment of the *D. oliveri* wood fibers induced the removal of hemicellulose and lignin from its fiber as evident from the FTIR spectra.
2. Alkali treatment of wood fiber increased the interfacial bonding and the wet-ability of the wood fiber by vHDPE leading to the enhancement in densification of the treated composites in comparison to the untreated samples.
3. The density, WAC and mechanical properties of the WPCs containing treated wood fiber increased with increasing

TABLE 2. DSC parameters of vHDPE, untreated and treated composites at varying concentrations and time of treatment with NaOH solution.

Entry	NaOH conc. (wt%)	Treatment time (min)	$T_m$ (°C)	$T_c$ (°C)	$\Delta H_m$ (J/g)	$-\Delta H_c$ (J/g)	$X_c$ (%)
1	vHDPE		127.70	113.34	172.56	154.34	58.77
2	0	0	128.00	118.50	72.17	24.85	49.16
3	4	30	127.38	116.00	93.16	19.16	63.48
4	6	30	127.67	115.55	82.55	21.84	56.23
5	4	150	127.15	116.90	106.50	25.62	72.54

NaOH concentrations up to 4 wt%, but at higher concentrations, the properties of the biodegradable composite decreased. Moreover, these functional properties also increased with increasing time of treatment.

4. Thermal decomposition stability of the WPCs was increased relative to the vHDPE, with the alkaline treated WPCs having higher stability in comparison to the untreated WPCs. In addition, the TGA analysis confirms that alkaline treatment improves the thermal properties of *D. oliveri* wood fibers, as a result of the degradation of hemicelluloses and modification of the lignin component in wood.
5. The DSC analysis establishes that alkaline treatment of wood fibers has no effect on the melting behaviors of the WPCs, whereas it influences its cold crystallization temperature. Moreover, alkaline treated vHDPE/*D. oliveri* composites are characterized by improved compatibility and higher degree of crystallinity.
6. The experimental results suggest that useful composite with optimum density, WAC and mechanical properties could be successfully developed with treatment parameters of 4 wt% NaOH and 150 min time of treatment for tape rack applications.

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## ABBREVIATIONS

BS	Bending strength
DSC	Differential scanning calorimetry
FM	Modulus of Flexure
FTIR	Fourier transform infrared
MOE	Modulus of Elasticity
PBS	Poly(butylene succinate)
PE	Polyethylene
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinyl chloride
SEM	Scanning electron microscopy
TGA	Thermo-gravimetric analysis
TS	Tensile strength
WAC	Water Absorption Capacity
WPC	Wood polymer composite

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