

# Banana fiber reinforced low-density polyethylene composites: effect of chemical treatment and compatibilizer addition

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**Abstract** In this study, an attempt has been made to utilize banana fiber (a natural fiber from agricultural waste) as reinforcement for low-density polyethylene (LDPE) to develop environmental friendly composite materials. LDPE/banana fiber composites were fabricated at different fiber loadings (10, 15, 20, 25, and 30 wt %) using compression molding technique. The composite with the composition of 25 wt % banana fiber was observed to be optimum on the basis of biodegradability and mechanical properties. Further, the effect of banana fiber surface treatment (alkali and acrylic acid) on the mechanical properties, morphology and water absorption behavior of the LDPE/banana fiber composites in the absence and presence of compatibilizer (maleic anhydride grafted LDPE, MA-g-LDPE) was comparatively studied. The alkali and acrylic acid treatment of the banana fibers led to enhanced mechanical properties and water resistance property of the composites, and these properties got further improved by the addition of the compatibilizer. The addition of compatibilizer to the acrylic acid treated banana fiber composites showed the most effective improvement in the flexural and impact strength and also, exhibited a reduction in the water absorption capacity. However, the tensile strength of the compatibilized composites with treated fibers resulted in slightly lower values than those with untreated fibers, because of the degradation of fibers by chemical attack as was evidenced by scanning electron microscopy (SEM) micrographs. SEM studies carried out on the tensile fractured surface of the specimens

showed improved fiber-matrix interaction on the addition of compatibilizer.

**Keywords** Banana fiber · LDPE composites · Acrylic acid · MA-g-LDPE · Mechanical properties

## Introduction

Over the past few years, natural fibers reinforced thermoplastic composites have attracted much attention for number of applications, particularly as non-structural building materials and automotive components [1]. This is due to the appreciable mechanical properties offered by the resultant bio-composites with added environmental and economic benefits [2]. Natural fibers such as banana, coir, hemp, sisal, kenaf and flax fibers are better component for fiber based polymer composites because they are biodegradable, renewable, and easily accessible.

Banana (*Musa Sapientum*) is one of the most well-known fruit crops, cultivated in many tropical parts of the world. After harvesting fruits, huge quantity of biomass is left over as waste. These wastes can provide additional income to the cultivators as the extracted fibers from it can be put to use, thus helping improve the rural economy of the country. Banana fiber has high cellulose content (64 wt %) and low microfibrillar angle (11°) which make it suitable for use as a reinforcing material [3]. Banana fiber possesses good tensile strength, specific flexural strength and rotting resistance (which is comparable to that of the glass fibers) [4].

However, like other natural fiber, banana fibers are also hydrophilic in nature; making it incompatible with the hydrophobic thermoplastic matrices, causing weak interfacial bonding, and leading to poor mechanical properties.

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In addition, incomplete wetting between the fiber and the matrix leads to generation of micro-gaps and flaws, resulting in swelling because of absorption of water and moisture from the surroundings into these micro-spaces of the composites. This swelling can distort the dimensions of the finished products, leading to micro-cracks, thus, degrading the mechanical properties of the composites [5].

The interfacial adhesion between the fiber and polymer matrix can be improved, usually, via two principal methods: chemical treatment of fiber and/or addition of a compatibilizer. Chemical treatment methods involve modification of the fiber and/or the matrix by silanization, alkalinization, addition of maleated coupling agents, acetylation, benzoylation, acrylation and acrylonitrile grafting, etc., [6, 7]. Among chemical treatment techniques for natural fibers, alkali treatment using sodium hydroxide (NaOH) is one of the most effective and economical technique [8, 9]. This treatment helps to extract the noncellulosic components (hemicellulose, lignin, pectins, waxes and impurities) and some part of amorphous cellulose which are responsible for poor surface wetting and inefficient fiber-matrix interaction. Removal of these components increases the surface roughness and reduces its hydrophilic nature, thus leads to better mechanical interlocking between fibers and matrix which provides better mechanical properties [10].

Acrylic acid treatment is found to be one of the best techniques to improve mechanical properties, storage modulus, and glass transition temperature as well as to reduce water absorption of the composites [11–13]. This method reduces the hygroscopicity of the natural fiber by generating an ester bond when carboxylic group of acrylic acid reacts with the cellulosic hydroxyl group [14]. Not many studies have been done on acrylic acid treated natural fiber reinforced composites.

Compatibilizers are generally graft or block copolymers obtained by modification of polymer matrix on grafting with monomers such as stearic acid, acetic anhydride, maleic anhydride and methyl isocyanate. These copolymers modify not only the fiber surface, but also the polymer matrix, which facilitates better mechanical interlocking between fiber and matrix [15]. Among all the above mentioned compatibilizers, maleic anhydride-grafted polymer is most widely used compatibilizer to improve interfacial adhesion of the composites of natural fibers and polymer [16, 17].

Incorporation of maleated coupling agents into natural fiber/thermoplastic composites generates ester linkages between the hydroxyl groups of the natural fiber and the anhydride groups of the compatibilizer whilst another end of the compatibilizer entangles with the thermoplastic matrix and thereby improves fiber-matrix adhesion [18]. The addition of maleated polypropylene (MA-g-PP)

improves the interaction between the banana fiber and polypropylene composites leading to improvement in mechanical properties [19, 20]. Some researchers used maleated polyethylene (MA-g-PE) as a compatibilizer to improve the adhesion between the natural fiber and thermoplastic matrix. The addition of MA-g-PE led to the improved mechanical properties, thermal stability, and water resistance of the composites [21–23]. However, there is no literature available on the application of MA-g-PE in LDPE/banana fiber composites.

It is found that the acrylic acid treatment of the natural fiber and addition of compatibilizer (MA-g-PE) have a positive effect on the composite properties. In this paper, an attempt has been made to investigate and compare the effect of banana fiber surface treatment using acrylic acid and most common alkali (NaOH) on the physico-mechanical properties of the banana fiber reinforced LDPE composites in the absence and presence of a compatibilizer (MA-g-LDPE). The main objective of fiber treatment and use of compatibilizer is to improve the bonding between the banana fiber and the LDPE matrix, which will consequently, reduce the microvoids in the composites and hydrophilic nature of banana fiber as well, thus leading to improvement in the mechanical properties and reduction in water absorption capacity of the composites.

SEM studies were also carried out on the tensile fractured surface of the specimens, to understand the effect of different modification techniques on the fiber/matrix interfacial bonding. Furthermore, thermal and crystallization behavior of either fibers and composites were analyzed using thermogravimetric analysis (TGA) and X-ray diffractometer (XRD).

## Experimental

### Materials

The banana fiber, used as a reinforcing fiber, was purchased from M/s Resha Enterprises, Bihar, India. It comprises of 63–64 wt % cellulose, 5 wt % lignin and 19 wt % hemicellulose [3]. The fibers were chopped to about 10 mm length to ensure their proper distribution in polymer matrix and easy blending with it [24]. Low-density polyethylene (LDPE), used as a matrix material, was procured from M/s Rapid Engineering Company Private Limited, New Delhi, India in the form of powder. It had a melt flow index (MFI) of 34 g/10 min (2.16 kg at 230 °C) and a density of 0.930–0.945 g/cm<sup>3</sup>. The compatibilizer, LDPE functionalized with maleic anhydride (MA-g-LDPE, OPTIM-142<sup>®</sup> functionalized with 0.5–0.8 % maleic anhydride) was procured

from M/s Pluss Polymers Private Limited, Gurgaon, India. Sodium hydroxide (NaOH) and acrylic acid used for the treatment of banana fiber were purchased from Merck, India and LobaChemie, India, respectively.

### Treatment of banana fiber

#### Alkali treatment

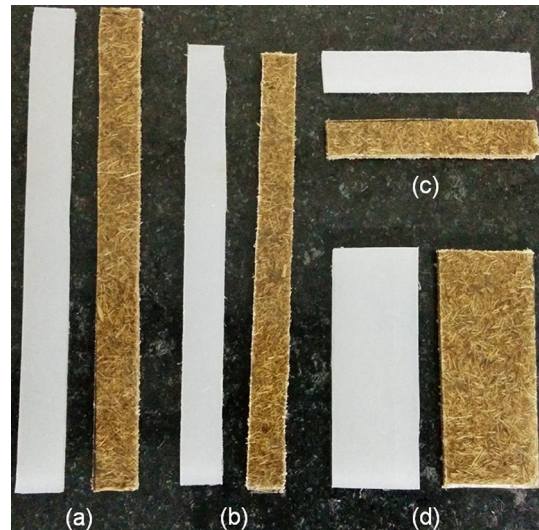
Chopped banana fibers were first washed with water to remove impurities. After that, banana fibers were soaked in 1 % NaOH solution maintaining fiber/solution ratio of 1:15 (w/v). The fibers were kept in the solution for 30 min at ambient temperature [10]. After that, fibers were washed with tap water followed by distilled water until the last traces of alkali were removed (reflected by the pH level of the used washing solution reaching approximately 7). Finally, the fibers were dried in an air oven at 70 °C for 48 h to obtain alkali treated fibers.

#### Acrylic acid treatment

Alkali treated banana fibers were soaked in a 1 % acrylic acid solution maintaining fiber/solution ratio of 1:15 (w/v) for 20 min [11]. Then, the fibers were washed thoroughly and dried in an air oven at 70 °C for 48 h to obtain acrylic acid treated fibers.

#### Composites fabrication

Before preparing composite samples, both banana fiber, and LDPE were dried in an air oven to avoid voids formation. The required amount of fiber and matrix for different weight fraction were weighed and placed in the beaker. Fiber and matrix were thoroughly mixed using mechanical stirrer and the mixture was put into the mold between two Teflon sheets for the easy removal of the sample. Randomly oriented short banana fiber composite sheets were prepared by placing the mold in the compression molding machine at around 180 °C temperature and 20 MPa pressure for 10 min. Mold was taken out after complete cooling at room temperature. Initially, untreated LDPE/banana fiber composite with different fiber loading (10, 15, 20, 25, and 30 wt %) were prepared, to find the optimum fiber-matrix ratio. Later, to improve composites properties, untreated (with compatibilizer) and chemically treated (with or without compatibilizer) LDPE/banana fiber composites at optimum fiber-matrix ratio, were prepared. All the compatibilized composites were prepared with 3 wt % of MA-g-LDPE based on the total composite weight [23]. Upon



**Fig. 1** Typical test specimens for: **a** tensile test, **b** flexural test, **c** impact test, and **d** water absorption test

completion of cooling, composite sample was removed from the mold and the test specimens were cut for the characterization (Fig. 1).

### Characterization

#### FTIR

An FTIR spectra of untreated and treated banana fiber samples was examined using a FTIR spectrometer (Nicolet 6700 series, UK). Potassium bromide (KBr) was used as reference substance. The samples were analyzed over the range of 4000–600  $\text{cm}^{-1}$  with a spectrum resolution of 4  $\text{cm}^{-1}$ .

#### XRD

X-ray diffraction (XRD) was used to study the crystallinity of the untreated and treated fibers and composite containing these fibers. The XRD patterns were recorded using a Bruker AXS D8 diffractometer (Germany) operating with Cu-K $\alpha$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ) at 40 kV and 30 mA in the range of  $2\theta = 10\text{--}50^\circ$  with a scan speed of 0.02°/s. The crystalline thickness ( $L$ ) was calculated according to the Scherrer's equation as follows:

$$L = \frac{K\lambda}{\beta \cos\theta}$$

where,  $K$  is the Scherrer's constant normally taken as 0.89,  $\lambda$  is the wavelength of the X-ray radiation

(1.5406 Å for Cu),  $\beta$  is the full width at half maximum (FWHM) of diffraction peak in radian and  $\theta$  is the Bragg angle.

### TGA

Thermal analysis of untreated and treated fibers and composite containing these fibers was carried out using EXSTAR TG/DTA 6300 equipment (Hitachi, Tokyo, Japan). Samples of approximately 8–10 mg were heated steadily at a heating rate of 10 °C/min from 50–600 °C under the nitrogen atmosphere.

### Mechanical testing

Tensile test was conducted as per standard ASTM D3039 using a universal testing machine (Instron Model 5982, USA). The test was performed at a crosshead speed of 2 mm/min. Flexural strength tests were conducted according to the ASTM D790 using the same testing machine and crosshead speed as mentioned above. The Izod impact strength was determined using un-notch specimens according to the ASTM D256 standard using pendulum impact testing machine, TINIUS OLSEN Model impact 104. The average value was taken for test results of three specimens.

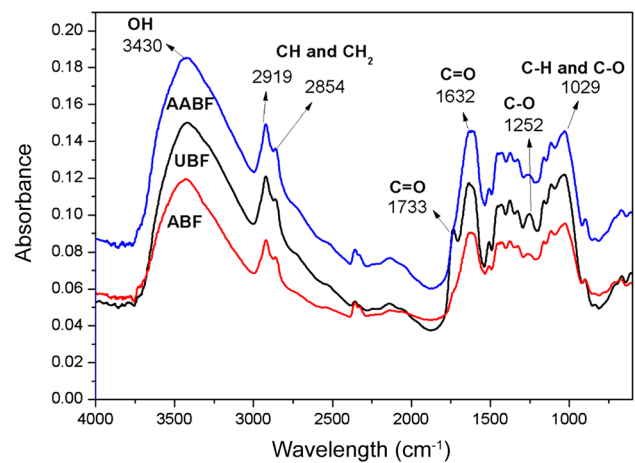
### SEM

Scanning electron microscope (LEO-435VP, USA) with an acceleration voltage of 10 kV was used for the study of the morphological behavior of the untreated and chemically modified fiber surface. The fractured surface of composites after the tensile test was also studied to assess the modes of failure. For this purpose, samples were pre-coated with gold prior to SEM analysis.

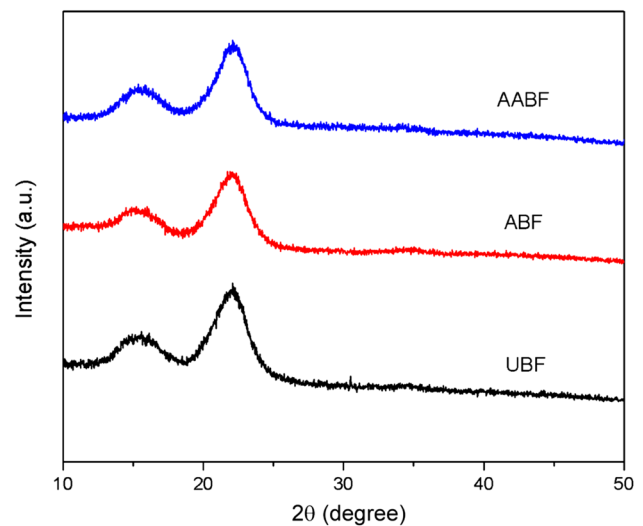
### Water absorption tests

The water absorption of composite materials was conducted according to ASTM D570-98 standards. Specimens were oven dried at 50 °C until a constant weight was obtained. Thereafter, composite samples were dipped in the distilled water and taken out after regular time intervals, wiped with tissue paper, and weighed in a high precision balance. The water absorption capacity of samples was calculated by the weight difference. The water absorption percentage was calculated using the following equation:

$$\text{Water absorption (\%)} = \frac{\text{weight of wet sample} - \text{weight of oven dry sample}}{\text{weight of oven dry sample}} \times 100$$



**Fig. 2** FTIR spectra of the banana fibers: untreated (UBF), alkali treated (ABF), and acrylic acid treated (AABF)



**Fig. 3** X-Ray diffractograms of the banana fibers: untreated (UBF), alkali treated (ABF), and acrylic acid treated (AABF)

## Results and discussion

### FTIR analysis of the fibers

FTIR spectra of banana fibers (untreated, alkali treated, and acrylic acid treated) are shown in Fig. 2. The strong absorbance band observed in the region 3600–3100  $\text{cm}^{-1}$  is attributed to the bonded O–H stretching vibration and hydroxyl groups present in the cellulose and hemicellulose

components [25]. In case of alkali treated banana fibers, the absorbance band at about  $3430\text{ cm}^{-1}$  is observed to be slightly reduced since alkali treatment removes some hemicellulose from the fiber surface. However, the absorbance band widens again with the acrylic acid treatment, because of the hydroxyl group of the acrylic acid. The absorbance band at about  $2919$  and  $2854\text{ cm}^{-1}$  appeared to be slightly stronger for the acrylic acid treated fiber than alkali treated fibers, corresponding to the introduction of  $-\text{CH}-$  and  $-\text{CH}_2-$  groups by the acrylic acid treatment.

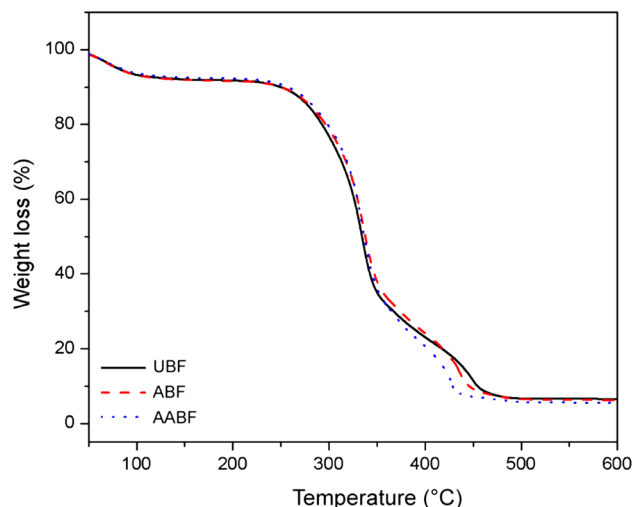
The absorbance band at about  $1733\text{ cm}^{-1}$  in the spectrum of untreated banana fibers (UBF) corresponds to the  $\text{C}=\text{O}$  (carbonyl) of hemicellulose, pectin and wax, but this peak is no longer observed in the spectra of alkali and acrylic acid treated fibers. The disappearance of the peak clearly indicates that the chemical treatment significantly removed the hemicellulose content, pectin and wax from the fiber surface. The peak near  $1632\text{ cm}^{-1}$  for acrylic acid treated fiber is found to be broader, indicating that some ester groups were grafted on it. The change in the peak near  $1252\text{ cm}^{-1}$  corresponds to the  $\text{C}-\text{O}$  stretching vibration of acetyl groups of lignin, indicating the removal of lignin with the alkali and acrylic acid treatments.

### XRD analysis

X-ray diffraction (XRD) patterns were recorded for both untreated and treated fibers to verify any alternation in their crystallinity due to chemical treatments. Figure 3 shows the X-ray diffraction patterns of untreated, alkali and acrylic acid treated banana fibers, respectively. Both untreated and treated banana fibers display a similar diffraction pattern. The diffractograms show two main reflections, corresponding to  $2\theta$  values of around  $15^\circ$  and  $22^\circ$ : the broad angle reflection at  $\sim 15^\circ$  corresponds to the amorphous part and sharp and intense reflection at  $\sim 22^\circ$  corresponds to the crystalline part of the fiber. The intense peak at about  $22^\circ$  is related to the (002) lattice plane of the cellulose [16]. The crystallite size of untreated, alkali treated, and acrylic acid treated banana fibers was calculated using the Scherrer's equation and found to be 2.77, 2.86, and 3.08 nm, respectively. The crystallite size of the treated fibers was higher than that of the untreated fibers. This can be ascribed to the removal of amorphous constituents such as hemicelluloses and lignin from the fibers and rearrangement of the crystalline regions such that the treated fibers exhibit more crystalline nature.

### TGA analysis

Thermogravimetric study of banana fibers was done to compare the thermal degradation behavior of chemical treated fibers with that of the untreated fiber. The thermogravimetric analysis (TGA) curves of the untreated and



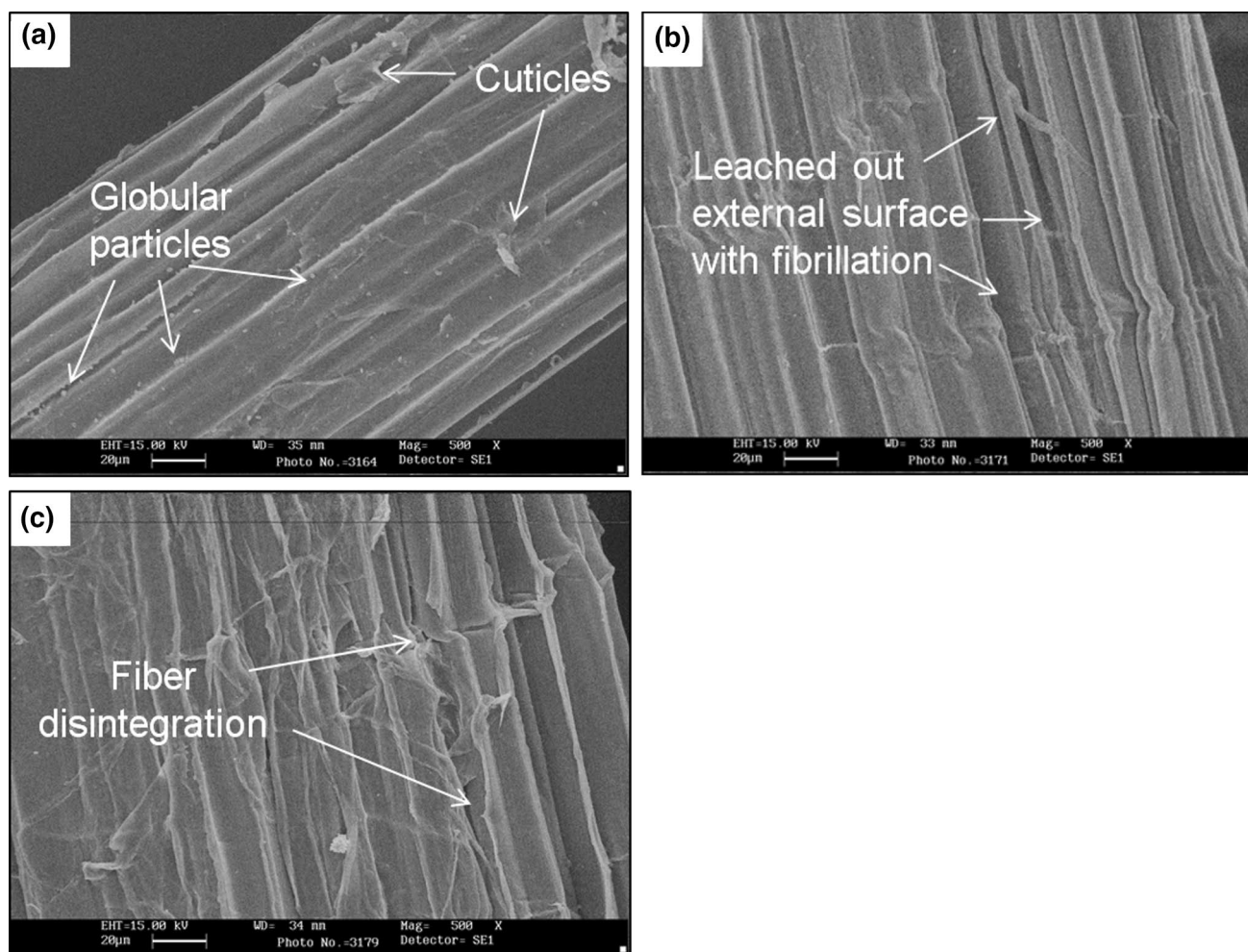
**Fig. 4** Thermogravimetry (TG) curve of the banana fibers: untreated (UBF), alkali treated (ABF), and acrylic acid treated (AABF)

chemical treated banana fibers are depicted in Fig. 4. The TGA curves of both untreated and treated fibers show negligible weight loss below  $200\text{ }^\circ\text{C}$  though initial degradation occurs between  $30$  and  $150\text{ }^\circ\text{C}$  with mass loss of approximately  $8\%$ , owing to the evaporation of the absorbed moisture. Above this temperature, the thermal stability of the fiber decreases gradually due to the decomposition of the major components of the fibers such as hemicellulose, cellulose, and lignin. The weight loss in the temperature range of  $220$ – $315\text{ }^\circ\text{C}$  is ascribed to decomposition of pectin and hemicellulose. The major decomposition corresponding to decomposition of cellulose is observed between  $300$  and  $390\text{ }^\circ\text{C}$ , whilst the degradation of the lignin takes place in broad temperature range from  $200$  to  $500\text{ }^\circ\text{C}$  [26, 27].

Figure 4 indicates the marginal difference in the thermal stability of untreated and treated fibers. The degradation temperature corresponding to  $20\%$  weight loss shifts from  $293\text{ }^\circ\text{C}$  (untreated) to  $297$  and  $299\text{ }^\circ\text{C}$  for alkali and acrylic acid treated fibers, respectively, indicating treated fibers are thermally more stable. Untreated fiber shows lower thermal stability due to the presence of thermally unstable fiber constituents such as hemicellulose and pectin, whereas treated fiber is thermally more stable due to the removal of these constituents. The residual char content is reduced from  $6.45\%$  (untreated) to  $6.19$  and  $5.45\%$  on the alkali and acrylic acid treatment, respectively.

### Effect of fibers surface treatment on their morphology

Figures 5a–c show the SEM micrographs of the untreated, alkali treated and acrylic acid treated banana fibers. These micrographs reveal that the morphology of the banana fibers was changed after the chemical treatment. The SEM



**Fig. 5** SEM micrographs of the banana fibers: **a** untreated, **b** alkali treated, and **c** acrylic acid treated

micrograph of the untreated banana fibers surface (Fig. 5a) clearly shows the presence of impurities, globular particles, wax and fatty substances.

However, the alkali treated banana fiber surface (Fig. 5b) is clean with individual ultimate fibers were slightly separated. This is attributed to the removal of the impurities and less stable non-cellulosic components from the fiber. The acrylic acid treated banana fiber surface (Fig. 5c) appear to be rough with the slight disintegration of the fiber along with fibrillation, which is attributed to the removal of cementing materials (lignin and hemicellulose) through dissolution in NaOH and acrylic acid during the treatment stage.

#### Effect of fiber loading on mechanical properties of composites

Effect of untreated banana fibers on the mechanical properties (tensile strength, Young's modulus, elongation-at-break, flexural strength and impact strength) of the

reinforced LDPE composites with respect to different fiber loading (10, 15, 20, 25, and 30 wt %) are illustrated in Table 1.

The tensile strength of the composite decreased considerably with the addition of untreated banana fibers. This is due to the hydrophilic nature of the banana fiber, which is incompatible with the hydrophobic LDPE matrix which led to reduction of interaction between banana fibers and LDPE matrix, resulting in poor stress transfer between the fibers and polymer matrix [28]. The composite with 10 wt % fiber loading, shows a tensile strength of 8.83 MPa which is 22.6 % less than that of the pure LDPE matrix. However, in comparison to 10 wt % fiber loading composite, an improvement in the tensile strength value is noticed with further increase in banana fiber loading from 10 to 30 wt %. Because, at 10 wt % fiber loading, the amount of fibers is not sufficient to restrain the matrix, and as a result, large stress develops at low strain which leads to non-uniform stress distribution [28]. The composite with

**Table 1** Mechanical properties of LDPE/untreated banana fiber composites at different fiber loading

Fiber loading (wt %)	Tensile strength (MPa) Mean $\pm$ SD	Young's modulus (MPa) Mean $\pm$ SD	Elongation-at-break (%) Mean $\pm$ SD	Flexural strength (MPa) Mean $\pm$ SD	Impact strength (kJ/m <sup>2</sup> ) Mean $\pm$ SD
0	11.4 $\pm$ 0.4	325 $\pm$ 11	121 $\pm$ 31	21.9 $\pm$ 1.9	26.8 $\pm$ 0.5
10	8.8 $\pm$ 0.8	591 $\pm$ 77	5 $\pm$ 0.4	14.1 $\pm$ 2.2	8.2 $\pm$ 0.7
15	10.3 $\pm$ 0.7	642 $\pm$ 79	5 $\pm$ 1.6	15.7 $\pm$ 1.4	10.2 $\pm$ 0.8
20	9.7 $\pm$ 1.4	933 $\pm$ 72	4 $\pm$ 0.5	16.0 $\pm$ 2.2	10.8 $\pm$ 0.7
25	9.8 $\pm$ 2.5	1050 $\pm$ 68	4 $\pm$ 0.5	19.0 $\pm$ 1.0	11.5 $\pm$ 0.9
30	9.5 $\pm$ 1.6	1160 $\pm$ 82	3 $\pm$ 0.7	18.5 $\pm$ 0.4	9.9 $\pm$ 0.4

SD standard deviation

15 wt % fiber loading, showed the highest tensile strength value of 10.27 MPa which is 16.26 % higher than the value of 10 wt % fiber loading composite. Even though, it is still 10 % lower than the value of tensile strength of the LDPE matrix.

Further, as the fiber loading increased from 20 to 30 wt %, more or less a constant trend in the tensile strength of the composites is observed. Incorporation of 20, 25 and 30 wt % fiber loading with the LDPE matrix shows increment of approximately 10.48, 10.73, and 7.37 %, respectively, in comparison with 10 wt % fiber loading composites. As the fiber loading in the composite increases, fiber distribution throughout the composite is uniform, thus, shows a constant tensile strength for the composites [29]. Similar trend results have been also reported by Velmurugan et al. [30] and Geethamma et al. [31].

Like tensile strength, the flexural strength of the composites was also observed to decrease considerably with the addition of untreated banana fiber compared to that of the LDPE matrix. This could be attributed to the poor interaction and wetting of the untreated banana fiber with the LDPE matrix. A flexural strength of 14.04 MPa was observed for the composite with 10 wt % fiber loading, which is nearly 36 % less than that of the pure LDPE matrix. However, with further increase in fiber loading from 15 to 30 wt %, an improvement in the flexural strength values was observed. At 25 wt % fiber loading composite, the value of the flexural strength is maximum which shows nearly 35.4 % higher value as compared to 10 wt % fiber loading composite.

The impact strength of the composites also exhibits a decrease with the addition of untreated banana fiber in comparison with that of the pure LDPE matrix. An impact strength of 8.20 kJ/m<sup>2</sup> is observed for the composite with 10 wt % fiber loading, which is nearly 69.4 % less than that of the of pure LDPE matrix. However, with further increase in fiber loading from 15 to 30 wt % an improvement in the

impact strength values was observed. The composite with 25 wt % fiber loading, showed the highest impact strength value which is approximately 40.6 % higher than that of 10 wt % fiber loading composite.

On the other hand, a consistent increase in Young's modulus of the LDPE matrix is observed with the addition of untreated banana fibers. Pure LDPE matrix showed Young's modulus of 325.5 MPa which was increased to 591.49 MPa at 10 wt % fiber loading, thereby showing an increase of about 81.7 % compared to that of the pure LDPE matrix. Similar increase of approximately 97.4, 186.9, 227.6 and 256.4 % in the Young's modulus values were observed over pure LDPE matrix with 15, 20, 25 and 30 wt % fiber loading, respectively. This may be due to the much higher modulus of banana fibers compared to that of the pure LDPE matrix. Thus, incorporation of fiber into the polymer matrix improved the Young's modulus or the stiffness of the composites.

The percent of elongation-at-break of the banana fiber reinforced LDPE composite decreased consistently with the increase of fiber loading. The value of elongation-at-break almost 97 % decreased from 121 % for pure LDPE matrix to 3.5 % for composites when banana fiber content was increased from 10 to 30 wt %. This is attributed to the addition of fiber into the polymer matrix which reduced the matrix mobility. As a result, Young's modulus increased and percent of elongation-at-break decreased for the composites by increase in the fiber loading [32].

The composite containing 25 wt % fiber loading was observed to have maximum impact and flexural strength. In case of tensile strength, the same composite only lags behind the one with 15 wt % fiber loading composite, by 4.8 %. Also, the composite containing 25 wt % fiber loading has highest composition of biodegradable content except for the one with 30 wt % fiber loading. Owing to all these factors, the 25 wt % fiber loading composite was used for further study.

**Table 2** Mechanical properties of LDPE/banana fiber composites (at 25 wt % fiber loading) as a function of chemical treatment and compatibilizer

Composite samples code	Tensile strength (MPa)	Young's modulus (MPa)	Elongation-at-break (%)	Flexural strength (MPa)	Impact strength (kJ/m <sup>2</sup> )
	Mean $\pm$ SD	Mean $\pm$ SD	Mean $\pm$ SD	Mean $\pm$ SD	Mean $\pm$ SD
LDPE/UBF	9.8 $\pm$ 2.5	1050 $\pm$ 68	4 $\pm$ 0.4	19.0 $\pm$ 0.9	11.5 $\pm$ 0.8
LDPE/ABF	10.9 $\pm$ 1.9	1154 $\pm$ 97	5 $\pm$ 0.6	19.2 $\pm$ 0.5	11.8 $\pm$ 0.5
LDPE/AABF	10.7 $\pm$ 1.2	969 $\pm$ 70	6 $\pm$ 2.0	19.9 $\pm$ 1.9	12.3 $\pm$ 0.4
LDPE/MA-g-LDPE/UBF	12.6 $\pm$ 1.4	1126 $\pm$ 98	3 $\pm$ 0.4	21.1 $\pm$ 1.0	11.8 $\pm$ 0.5
LDPE/MA-g-LDPE/ABF	12.2 $\pm$ 1.5	1203 $\pm$ 78	4 $\pm$ 1.1	21.9 $\pm$ 0.7	13.2 $\pm$ 0.4
LDPE/MA-g-LDPE/AABF	12.4 $\pm$ 1.7	1119 $\pm$ 83	5 $\pm$ 1.6	23.3 $\pm$ 0.9	13.8 $\pm$ 0.5

SD standard deviation

### Effect of modification methods on mechanical properties of composites

The mechanical properties of the untreated and chemically treated (alkali and acrylic acid) banana fiber-LDPE composites (at 25 wt %), with or without compatibilizer, (MA-g-LDPE) are illustrated in Table 2.

### Effect of chemical treatment on properties of the composite without compatibilizer

It is evident that the alkali and acrylic acid treated banana fibers improved bonding with the LDPE matrix. Since, alkali treatment resulted in the removal of impurities, wax, and part of hemicellulose and lignin, which led to the fiber fibrillation and rough surface, these composites showed improved tensile strength, Young's modulus, elongation-at-break percent, flexural and impact strength. This treatment generates a larger contact surface area, offering better mechanical interlocking with LDPE matrix [6]. Venkateshwaran et al. [10] reported that alkali treated banana fiber reinforced epoxy composites showed better mechanical properties than the untreated fiber composites.

For the acrylic acid treatment method, an improvement in the fiber wetting and bonding with LDPE matrix was noticed due to the generation of strong ester linkage with the hydroxyl groups of the fibers, made their surface more hydrophobic in nature. In addition, esterification raises the surface roughness of the fibers, resulting in better mechanical interlocking with the polymer matrix. However, in comparison with alkali treated fiber composites, acrylic acid treated fiber composites showed reduced tensile strength and Young's modulus values but slightly improved flexural and impact strength, and elongation-at-break percent. The Young's modulus of the acrylic acid treated fiber composites has even lower values than that of the untreated fiber composites. These results are in

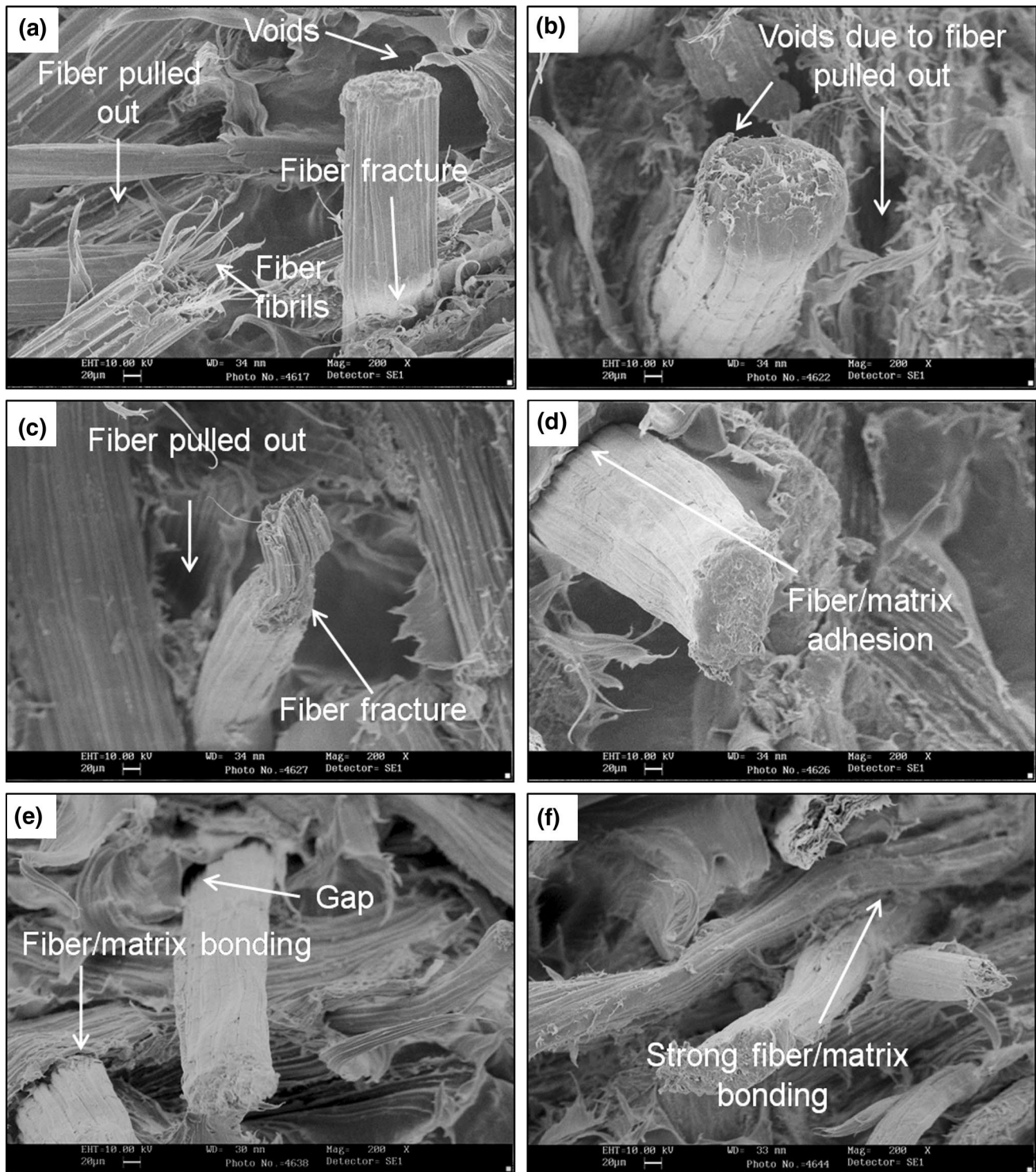
contradiction with the results obtained by Li et al. [13] and Patel et al. [33] which showed improved mechanical properties after acrylic acid treatment. This unlike phenomenon has probably happened due to the combined treatment of alkali and acrylic acid which had a lasting effect on the banana fibers.

### Effect of chemical treatment on properties of the composite with compatibilizer

Incorporation of MA-g-LDPE into untreated and treated banana fiber composites leads to improved interfacial bonding between banana fiber and LDPE matrix. Addition of MA-g-LDPE showed improvement of approximately 29, 11 and 16.5 % in tensile strength, 7, 4 and 15 % in Young's modulus, 10.7, 14.5 and 16.7 % in flexural strength, and 2, 11.1 and 11.9 % in impact strength for untreated, alkali and acrylic acid treated banana fiber composites, respectively as compared to the same composite formulation without MA-g-LDPE. This is due to the addition of MA-g-LDPE which generates strong covalent bonds between the maleic anhydride groups of MA-g-LDPE and hydroxyl groups of fibers surface while the LDPE of MA-g-LDPE is compatible with LDPE matrix, as a result wetting and dispersion of the fibers improved within the LDPE matrix and, thereby, enhanced the mechanical properties [22].

In relation to the compatibilized composite containing untreated fiber, compatibilized composites with surface treated fibers showed better flexural strength. The better flexural strength indicates that the incorporation of compatibilizer provides better compatibility between surface treated fiber and LDPE matrix. The maximum flexural strength value was observed with compatibilized composites containing acrylic acid, with almost 22.7 % improved value than that of the untreated fibers composite without compatibilizer. However, the tensile strength of the





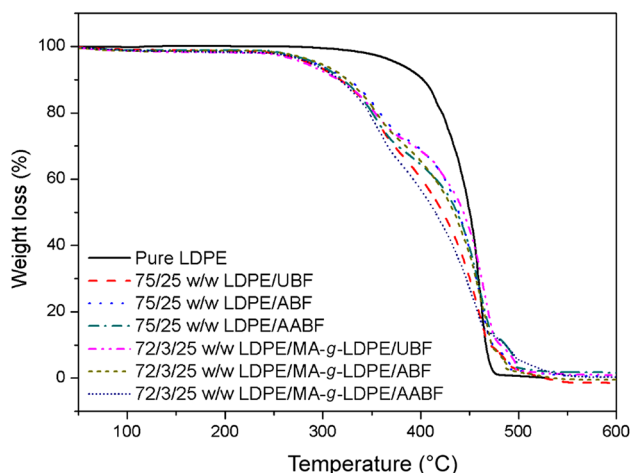
**Fig. 6** SEM micrographs of tensile fractured surfaces of the 25 wt % composites of: **a** LDPE/UBF, **b** LDPE/ABF, **c** LDPE/AABF, **d** LDPE/MA-g-LDPE/UBF, **e** LDPE/MA-g-LDPE/ABF, and **f** LDPE/MA-g-LDPE/AABF

compatibilized composites with treated fibers lowers in value, which is paradoxical to the fact that good interfacial bonding improves the tensile strength of the composites. It

may be due to the degradation of cellulose fibrils by chemical treatments which reduced composite tensile strength, as a result.

## Morphology of tensile fractured surfaces

For a clear understanding of mechanical behavior, the morphological behavior of the tensile fractured surface of modified and unmodified composites at 25 wt % fiber loading was investigated by SEM technique which has been shown in Fig. 6a–f. It can be clearly observed that the modified composites possess better interlocking between fiber and polymer matrix compared to unmodified composites. Figure 6a shows the SEM micrograph of untreated fiber composite without compatibilizer, in which fibers seems to be detached from the LDPE matrix and show relatively large pullout and voids, indicating weak interfacial bonding between fiber and matrix. Whereas, alkali and acrylic acid treatment to the banana fiber provided relatively better interlocking between the fiber and the polymer matrix as shown in Fig. 6b and c, respectively. On the other hand, micrographs of MA-g-LDPE incorporated composite in Fig. 6d–f manifest better interfacial adhesion with a lower number of fiber pulled outs and reduced voids



**Fig. 7** Thermogravimetry (TG) curves of pure LDPE and LDPE/banana fiber composites

**Table 3** Degradation temperature obtained from TGA curves

Composite sample code	$T_o$ (°C)	Mass loss (%)	$T_f$ (°C)	Mass loss (%)	Residue left (%)
Pure LDPE	324	1.72	478	98.94	1.06
LDPE/UBF	245	1.72	465	81.95	18.05
LDPE/ABF	250	1.88	468	82.56	17.44
LDPE/AABF	248	1.52	470	85.49	18.51
LDPE/MA-g-LDPE/UBF	246	2.26	474	85.55	14.45
LDPE//MA-g-LDPE/ABF	243	1.50	471	86.99	13.01
LDPE//MA-g-LDPE/AABF	241	1.72	461	82.62	17.38

$T_o$  onset degradation temperature,  $T_f$  final degradation temperature

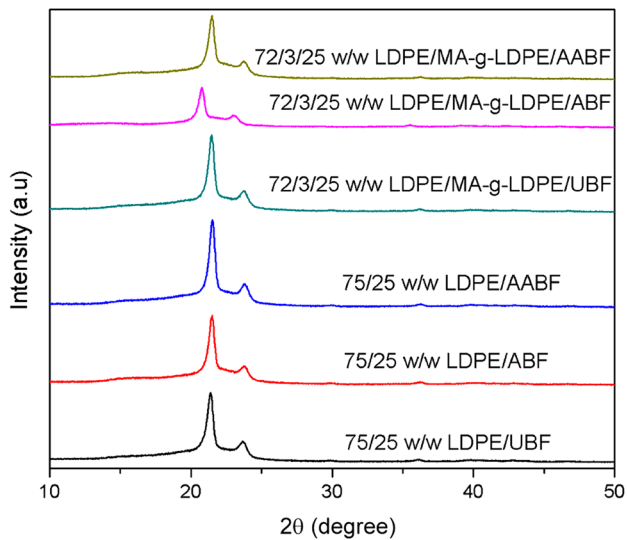
formation in the cases of both untreated and treated fiber composites.

## Thermal and crystallization behavior of composites

Figure 7 shows the effect of untreated and treated banana fibers on the thermal behavior of the LDPE in the presence and absence of compatibilizer. The TGA curve of pure LDPE matrix is also shown for the comparison. The onset temperature, final degradation temperature ( $T_f$ ) and corresponding mass loss are presented in Table 3. The onset degradation temperature ( $T_o$ ) indicates the thermal stability of the composites, since above this temperature, degradation occurs at a faster rate. From TGA results, it has been observed that both  $T_o$  and  $T_f$  values of the LDPE matrix decreased with the addition of the fibers. This is owing to the thermal stability of the banana fiber which is much lower than that of the pure LDPE matrix. However, for the treated fiber composites, due to the removal of thermally unstable fiber constituents such as hemicellulose and pectin the values of  $T_o$  and  $T_f$  were slightly improved.

The compatibilizer seems to have little influence on the thermal degradation of the composites. In the case of untreated fiber composite, addition of compatibilizer improved both the  $T_o$  and  $T_f$  values. But, concerning treated fiber composites, the compatibilizer caused reduction in  $T_o$  value. This is possible because addition of compatibilizer improved the interfacial interaction between the fibers and the matrix by generating strong ester linkages between them. This improved interaction also enhanced the interaction between the degradation processes of the two components, i.e., the degradation of one component may accelerate the decomposition of the other component.

The effect of untreated and treated banana fibers on the crystallization behavior of the LDPE in the presence and absence of compatibilizer was also studied using X-ray diffraction which has been depicted in Fig. 8. The diffraction pattern of pure LDPE matrix is also shown for the comparison. The peak positions of the major reflection crystalline plane (100) of LDPE and its composites are listed in



**Fig. 8** X-Ray diffractograms of pure LDPE and LDPE/banana fiber composites

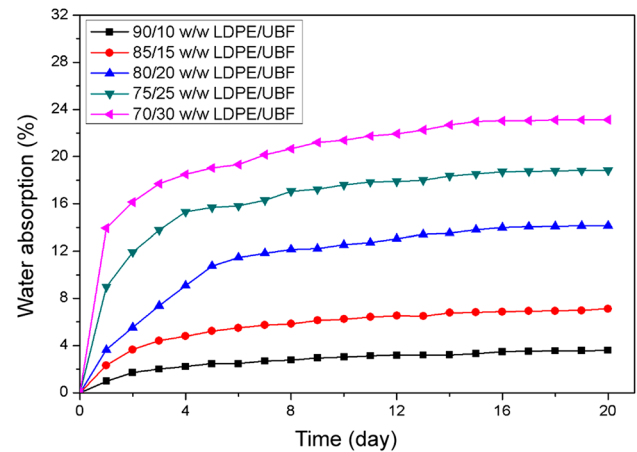
**Table 4** Crystalline peaks position and crystal thickness of pure LDPE and LDPE/banana fiber composites by XRD

Composite sample code	Peak position $2\theta$ ( $^{\circ}$ )	Crystal thickness (nm)
Pure LDPE	21.17	11.14
LDPE/UBF	21.37	14.58
LDPE/ABF	21.51	15.42
LDPE/AABF	21.49	14.85
LDPE/MA-g-LDPE/ UBF	21.47	15.42
LDPE//MA-g-LDPE/ ABF	20.75	15.73
LDPE//MA-g-LDPE/ AABF	21.49	15.73

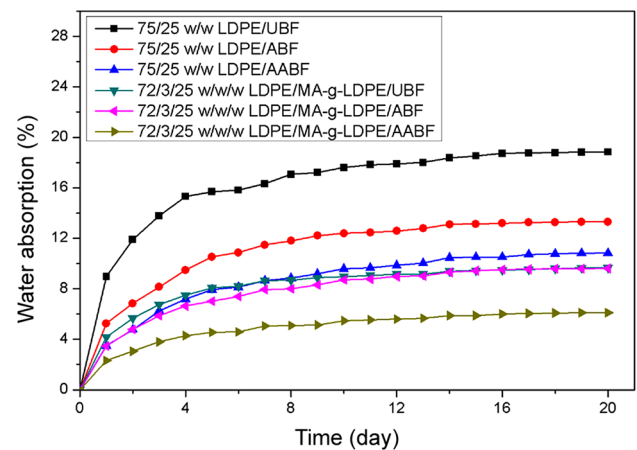
Table 4. The results show that adding fiber to LDPE matrix does not change the characteristics of the peak position; however, fiber increased the crystalline thickness of the LDPE system. This is possible due to the nucleating ability of the natural fibers which partially increased the crystallization rate of the LDPE matrix. The results also showed that the alkali and acrylic acid treatments to the banana fibers led to enhanced crystalline thickness of the composites, which was further improved by the addition of the compatibilizer, indicating better interfacial adhesion between fibers and matrix.

### Water absorption

The water absorption behavior of the unmodified and modified composites was studied and the results are shown in



**Fig. 9** Water absorption behavior of LDPE/untreated banana fibers (UBF) composites at different fiber loading



**Fig. 10** Water absorption behavior of LDPE/banana fiber composites with different modifications

Figs. 9 and 10, respectively. The effect of different fiber loadings on the water absorption behavior of the LDPE/untreated banana fiber composites is shown in Fig. 9. From the graph, it is evident that the water absorption capacity of the composite increased by increasing fiber loading from 10 to 30 wt %. This behavior is explained on the fact that banana fibers are hydrophilic in nature. Cellulose, which is the main constituent of banana fibers, has free hydroxyl groups which create hydrogen bonding with the molecules. Another reason is the incompatibility between natural fibers and polymer matrix which increased microvoids formation in the composites.

The effect of different modifications techniques on the moisture absorption behavior has been studied at 25 wt % fiber loading (Fig. 10). It is revealed that the water absorption capacity of the modified composites reduced

considerably as compared to the untreated banana fiber composites. The best result has been obtained for the acrylic acid treated fiber composite with the incorporation of MA-g-LDPE. This is due to the replacement of hydroxyl groups with hydrophobic ester groups by acrylic acid treatment [34]. Another reason is the incorporation of MA-g-LDPE that generates chemical linkage between acrylic acid treated fibers and LDPE matrix which reduces the microvoids formation.

## Conclusion

In this research, banana fibers (an agricultural waste) was utilized as a reinforcement for LDPE matrix to develop cost-effective eco-friendly composite material. To improve the compatibility between the banana fiber and the LDPE matrix, banana fiber was chemically treated with sodium hydroxide (alkali) and acrylic acid along with MA-g-LDPE as a compatibilizer. Chemically treated fibers showed slightly higher crystalline thickness and onset degradation temperature. The fiber treatment and the use of compatibilizer improved the mechanical properties of the LDPE composites. Alkali and acrylic acid treated LDPE composites without compatibilizer showed almost similar mechanical properties. This suggests that the alkali treatment to the banana fiber has improved its compatibility with the LDPE matrix by removing impurities, wax, fatty acid, and part of hemicellulose and lignin from the fiber surface. But, further treatment with acrylic acid over alkali treated fiber leads to fiber disintegration resulting in slightly lower tensile properties and higher flexural strength than that of the simple alkali treated fiber composites. In corporation of MA-g-LDPE resulted in the most effective enhancement of the mechanical properties for both treated and untreated fiber/LDPE composites, as compared to the same composite formulation without MA-g-LDPE. SEM micrographs manifested improved fiber-matrix interfacial bonding after use of compatibilizer. The fiber surface treatment and use of compatibilizer led to reduced water absorption capacity of the LDPE composites. The addition of MA-g-LDPE with acrylic acid treated fiber composite exhibited the highest water resistance property. Physico-mechanical studies carried out for the LDPE/banana fiber composites showed that these composites had lower strength properties and were sensitive to the moisture due to hydrophilic nature of lingo-cellulosic fibers. In spite of these limitations, the banana fibers can serve as an appropriate alternative to synthetic fibers and has a lot of potential as low cost polymeric composite material for various engineering applications. Recycling (including burning) characteristics is an attractive future research that will provide socio-economic benefits.

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