Innovated Banana Fiber Nonwoven Reinforced Polymer Composites: Effects of Pre- and Post-treatments on Physical and Mechanical Properties

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Research Article

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Abstract

Four types of nonwovens were prepared from different sections of the banana tree e.g., outer bark (OB), middle bark (MB), inner bark (IB) and midrib of leaf (MR) by wet laid web formation. They were reinforced on two different types of matrices e.g., epoxy (E) and polyester (P) to make eight variants of composites. Different concentration (5–15%) of NaOH and water repellent (WR); and different doses (100-500krd) of gamma radiation were applied in different stages of process. The properties like water absorbency, tensile strength (TS), flexural strength (FS) and elongation at break (Eb%) were investigated. OB composites were exhibited higher water absorbency, TS and FS but lower Eb% than other types of composites. Epoxy composites were found to have 16% lower water absorbency, 41.2% higher TS and 39.1% higher FS than polyester composites on an average. Alkali treatment reduced the water absorbency by 32%; improved the TS by 71%; improved the FS by 87% on an average at 15% NaOH. Water repellent treatment (on alkali treated composites) decreased the absorbency by 63% at 10% WR but increased 6.3% at 15% WR. Gamma radiation improved the TS of 30% and FS of 35% on an average at a dose of 100krd for IB and 200krd for other composites. Further increment of dose reduced both the FS and TS.

1. Introduction

From the last century, traditional materials like wood, metal, ceramic, glass are being rapidly replaced by the polymer matrix composite (PMC) materials with the reinforcement of synthetic fibers due to a lot of advantages like light weight, easy processing, low cost, and high productivity. However, this rapid increase of non-biodegradable PMC also created a lot of dangerous and alarming problems like environmental pollution from plastics, burning of fossil fuels, increasing global warming potential, etc. Consequently, these problems are creating a harmful and unsafe environment for humans, animals, and marine lives (Malviya et al. 2019). For these reasons, researchers are paying attention to alternative eco-sustainable materials now-a-days. Natural fiber reinforcement can be a potential alternative in polymer composite due to their numerous advantages including biodegradability, low relative density, economical, ease of handling, availability, light weight, high impact resistance, high flexibility, low specific gravity, recyclability, low carbon emission, good thermal and acoustic insulation and so on (Wu et al. 2018; Keya et al. 2019; Nayak et al. 2020; Kerni et al. 2020; Santhanam et al. 2020). Natural fiber reinforced composites (NFRCs) are becoming more attractive in many areas of engineering applications with a wide range of properties (Lotfi et al. 2021). Natural fibers those are used as a reinforcement in NFRCs can be obtained from plants, animals, and minerals. However, over the years, most of the natural fibers investigated as a reinforcement have been from agricultural plant, byproducts, or wastes.

There are numerous sources of plant fibers all over the world especially in tropical regions. They can be categorized by the parts of the plant from where they are extracted like leaf fibers (pineapple, sisal, abaca), bast fibers (jute, banana, flax, hemp) and seed fibers (cotton, coir, kapok) (Zwawi 2021). Most of the plant fibers are mainly consists of cellulose, hemicellulose, and lignin (Santhanam et al. 2020). Banana fiber is one of the cellulosic fibers extracted from the stem of banana tree (Musa acuminata).
Banana fiber contains 71.08% of cellulose, 12.61% of hemicellulose and 7.67% of lignin in their chemical composition with a diameter of 138 µm and density of 1.28 g/cm³ (Kenned et al. 2020a). In a tropical country like Bangladesh, banana plants are considered as agricultural crops, growing abundantly due to favorable climate conditions. After harvesting fruits, banana plants are cut at their lower section and the whole cutting portions are considered as a complete waste including pseudo stem and leaf those can be utilized as a source of natural fibers for the manufacturing of NFRCs, textiles, nonwoven, packaging materials, wiping materials and so on (Adeniyi et al. 2019). Therefore, the fibers can be used in the industry without any additional expenses for the cultivation (Balaji et al. 2020). Moreover, banana fibers exhibit good mechanical properties competently with other cellulosic fibers that makes them a potential reinforcing material for varieties of engineering applications (Gholampour and Ozbakkaloglu 2020; Komal et al. 2020; Srinivasan et al. 2020; Lotfi et al. 2021).

However, the performance of the NFRCs depends on fiber orientation, fiber content, length, shape and their interfacial bonding with the matrix (Al-Oqla and Salit 2017). Fiber orientations are also varied with different forms of reinforcement like chopped fiber reinforcement, continuous fiber (filament) reinforcement, woven fabric reinforcement and nonwoven reinforcement. Woven fabrics are generally produced by interlacing the yarns usually at the right angles by following a regular pattern. The strength of woven fabrics can be increased by increasing the twist angle of the yarns up to a certain limit. However, this twist angle plays an opposite role in case of composites. Increase of twist angle decreases the permeability of matrix to the fiber which results poor fiber-matrix adhesion and low mechanical properties (Peças et al. 2018). In case of filament reinforcement, mechanical properties are much lower in transverse direction of fibers that is also a limitation for different applications. To avoid these problems, nonwoven reinforcement can be a great option. Nonwovens are prepared in a flat structure with different thickness without interloping or interlacing. Fibers are chopped, uniformly distributed, and bonded together by chemical, mechanical or thermal treatment. They don’t have preferential strength direction and can produce in large scale due to availability and low cost (Al-Oqla and Sapuan 2014).

Thermosetting resins are most widely used in the composite industry as a polymer matrix. Among them, epoxy and polyester resin are most applied matrix in the composites. Epoxy resin, also known as polyepoxides, have good adhesion properties with natural fibers. Other key features are low moisture absorption, high chemical resistance, low shrinkage, and simple processing. These excellent properties make them superior in market with wide range of applications (Oliveira et al. 2019). However, unsaturated polyester resin, also known as polyhydric alcohols, have satisfactory mechanical properties and enough adhesion properties with the natural fibers which are reported in several studies. The main advantage of polyester resin is, they are cheaper, available and can be used in wide ranges of applications (Sreekumar and Thomas 2008).

There is no doubt that, natural fiber brings a lot of potentiality for their unique properties and environmental friendliness, but they have some drawbacks like high moisture absorption, low compatibility with the commercial resins, poor adhesion between fiber and matrix, less homogenous like
overcome by different types of physical and chemical treatments. Chemical treatments of the fibers including alkaline, saline, acetylation, benzoylation and many more can improve the adhesion between fiber and matrix (Faruk et al. 2014). Alkali treatment is one of the simplest and cheapest method which is easily applicable to natural fibers by immersing them into the solution of NaOH. After the alkali treatment, fibers become more uniform by removing all the impurities. Therefore, physical and mechanical properties of the composites are improved consequently (Gholampour and Ozbakaloğlu 2020). Several studies have been reported the improvement of physical and mechanical properties of the composites by alkali treatment of natural fibers (Yan et al. 2012; Mohd Nazarudin et al. 2013; Manalo et al. 2015; Preet Singh et al. 2017; Wijianto et al. 2019; Binti Mohd Hafidz et al. 2021). To overcome the high-water absorbency problem of NFRCs, surface treatment of natural fibers with water repellent is a potential way. The water repellent makes a coating on the fiber surface and resist the water to penetrate inside. There is no relevant study yet regarding the water repellent treatment on natural fibers to improve the hydrophobicity of NFRCs.

Physical treatments like x-ray, ultraviolet (UV) ray, gamma ray, plasma, corona are applied to the composites to improve the fiber-matrix adhesion. Due to less time consumptions, high productivity, low environmental pollution, structural availability and easy application, gamma radiation becomes popular day by day (Noura et al. 2018). Gamma radiation is a powerful ionizing radiation which can penetrate inside the polymeric structure of the composites and produce reactive sites to make more oriented polymeric structures and improve the mechanical properties of the composites (Masudur Rahman et al. 2018). Many researchers have been applied this radiation and reported the improvement of mechanical properties of the composites (Khan et al. 2009; Haydaruzzaman et al. 2009; Masudur Rahman et al. 2018; Martínez-Barrera et al. 2020). However, the studies also reported that, gamma radiation improves the mechanical properties up to a certain level of gamma radiation dose after that it alters the properties. Therefore, an optimum dose must be maintained.

Numerous studies have been noted regarding the properties of banana fiber reinforced composite materials. Majority of them used banana fiber or pseudo stem mat as a reinforcing material (Jordan and Chester 2017; Mohan and Kanny 2019; Balaji et al. 2020; Komal et al. 2020; Srinivasan et al. 2020). Only a few of them studied about banana nonwoven reinforced composite materials. Kenned J et al. studied thermo-mechanical and morphological characterization of needle punched nonwoven banana fiber reinforced polymer composites (Kenned et al. 2020b). The properties of needle punched nonwoven from banana fiber was also studied by Sengupta et al (Sengupta et al. 2020). Thilagavathi et. al. developed needle punched banana nonwovens for the application of noise control car interiors (Thilagavathi et al. 2010). But no research has been reported the development of banana fiber nonwoven by wet laid web formation technique from various parts of banana tree and the properties of their reinforced composites. Moreover, no study has been found regarding the surface modification of such nonwovens and their composites.

The aim of this study is to develop an ecofriendly composite material from a natural source to replace the existing environmentally destructing, carcinogenic, synthetic composites materials those are being used
vastly in different areas like packaging, households, building materials, automobiles, technical textiles and so on. To fulfil the objectives, four types of banana fiber nonwovens were developed from different parts of the banana tree e.g., outer bark, middle bark, inner bark of banana stem and mid rib of banana leaf in our previous study (Motaleb et al. 2020). This study is the continuation of the last work. Nonwovens were prepared by following the previously developed wet laid web formation technique from the extracted fibers. The prepared four types of nonwovens were reinforced on two different types of matrices e.g., epoxy resin and polyester resin to make eight variants of composites. Surface treatments were applied in three stages, i) fiber stage – alkali (NaOH) treatment, to improve the mechanical properties and hydrophobicity, ii) nonwoven stage - water repellent treatment, to improve the hydrophobicity and, iii) composites stage – gamma radiation, treatment to improve the mechanical properties. Water absorbency of the composite samples and the improvement of hydrophobicity by the surface modification were inspected. Mechanical properties like tensile and flexural strength and the influence of physical and chemical treatment on those properties were also analyzed in this study. A comparative study between the composites of epoxy matrix and polyester matrix are elaborated in different aspects throughout the study.

2. Materials And Methods

2.1 Materials

Banana trees were collected from a banana plantation as a waste material (after harvesting banana fruit) in Gazipur, Bangladesh. Epoxy resin, hardener HY-951, polyester resin and methylethylketone peroxide (MEKP) were bought from a European chemical supplier. Caustic soda and water repellent (perfluoroalkyl acrylic) were purchased from Archroma International Ltd.

2.2 Methods

2.2.1 Banana Fiber Extraction

Banana trees were segregated into four different sections such as 1) outer layers of banana bark designated as outer bark (OB), 2) middle layers of banana bark designated as middle bark (MB), 3) inner layers of banana bark designated as inner bark (IB) and 4) middle rib of banana leaves designated as midrib (MR). All the sections are mentioned in Fig. 1. The raw materials of each section were then pressed by a metal tube squeezer for removing the inside water as much as possible. After that, they were dried in sunlight for about 15 days.

The dried materials were scratched by a metal comber to make like ribbon and cut them with a length of 3 cm. For the initial fiber extraction, these small pieces were taken in a big metal pot and treated with 5 (w/v) % of NaOH with a temperature of 90ºC for about 30 minutes until they became soft. They were rinsed properly for removing unwanted materials and dried subsequently. Thus, the raw banana fibers from different parts of the banana trees were extracted.
2.2.2 Alkali Treatment on Fibers

The extracted raw fibers were still contained various impurities like fat, wax, pectens and so on. To clean off these impurities, the fibers were immersed into a solution of NaOH at different concentrations i.e., 5, 10 and 15 (w/v) % for 24 hours at a temperature of 23 ± 2ºC. The fibers were rinsed and dried again after the treatment.

2.2.3 Nonwoven Formation

Firstly, the alkali treated banana fibers were blended with water to make a uniform pulp mixture. Then, they were rinsed properly to remove the leftover of NaOH and dried once again. The prepared banana pulp was taken in the blender again with the fiber (pulp)/water ratio of 1:50. After blending, the mixture was poured in a mold prepared with wooden frame and mesh fabric. The fibers were distributed uniformly by immersing the complete mold on a tab of water to make a sheet from the fiber webs according to the wet laid web formation technique. The sheet, usually known as nonwoven, was then moved to a plastic plate, and pressed with wiping paper to remove extra water. Finally, they were dried in sunlight and straighten with an electrical iron in case of rough surface. Similar procedure was followed to make all types of banana fiber nonwovens. The average thickness of the nonwovens were found 0.75 ± 0.05 mm.

2.2.4 Water Repellent Treatment on Nonwovens

Before making composites by reinforcing the nonwovens, some of them were treated with a water repellent (WR) chemical (perfluoroalkyl acrylic) to improve the hydrophobicity of the composites. WR was applied at three different concentrations i.e., 5%, 10% and 15% to find out the appropriate dose to decrease the water absorbency by keeping up the strength. The nonwovens were immersed into the solution of WR and kept for couple of minutes. The wet nonwovens were then squeezed by a padding roller to remove excess solution. They were dried and cured in an oven with a temperature of 160–170ºC for 30 minutes.

2.2.5 Composite Formation

The composites were prepared with hand layup technique. The already prepared nonwoven from four different section of banana tree i.e., OB, MB, IB, MR were used as reinforcing material and two types of resin i.e., epoxy (E) and polyester (P) were used as matrix. In total, eight variants of composites were prepared, which are designated as OB/E, MB/E, IB/E, MR/E, OB/P, MB/P, IB/P and MR/P with all possible combination of nonwovens and resins. Two metal plates were used as top and bottom surface of the mold with a size of 35×35 cm. The metal plates were wrapped with Teflon (PTFE) paper to avoid the sticking difficulties during the composite peel off. Three layers of nonwovens were reinforced for all types of composites. At first, the nonwovens were cut with a size of 30×30 cm. Three pieces of nonwoven sheets were weighted together by a precise scale. According to the weight of nonwovens, a certain amount of resin mixture was prepared with the addition of appropriate catalyst by maintaining a
constant fiber/resin weight ratio of 30:70 for all the composites. 10% HY951 was used for epoxy resin and 2% MEKP was used for polyester resin as a hardener. The bottom metal plate was placed in a suitable flat surface. To begin the fabrication of composite, ¼ of the resin mixture was poured on the bottom metal plate and spread them uniformly with a brush according to the size of the nonwovens. Then the first nonwoven layer was placed on them and pressed with a hand roller in such a way that the resin penetrated throughout the nonwoven. Again, ¼ of the resin mixture was poured on the first nonwoven layer and repeated the same process to reinforce second and third nonwoven layer. The rest ¼ of the resin mixture was poured on the third nonwoven layer, the top metal plate was placed on them to make a complete sandwich structure. A dead weight of 20kg was laid on the top metal plate and kept them 24 hours for curing. Finally, the dead weight was removed, and the composite was separated from metal plates. Similar procedure was followed for making all the composites. The overall thickness of the composites was found 3 ± 0.5 mm.

2.2.6 Sampling

In total eight types of composites were prepared. Samples for each treatment or test were prepared separately according to the prescribed standards. All types of samples are described in Table 1. Some prepared samples for the tensile tests are presented in Fig. 2.

| Types | Description                                      | Designation |
|-------|--------------------------------------------------|-------------|
| 01    | Outer bark nonwoven reinforced epoxy composites  | OB/E        |
| 02    | Middle bark nonwoven reinforced epoxy composites | MB/E        |
| 03    | Inner bark nonwoven reinforced epoxy composites  | IB/E        |
| 04    | Midrib nonwoven reinforced epoxy composites      | MR/E        |
| 05    | Outer bark nonwoven reinforced polyester composites | OB/P     |
| 06    | Middle bark nonwoven reinforced polyester composites | MB/P     |
| 07    | Inner bark nonwoven reinforced polyester composites | IB/P     |
| 08    | Midrib nonwoven reinforced polyester composites  | MR/P        |

2.2.7 Gamma Radiation on Composites
The composite samples were irradiated with different doses of gamma radiation. A capsule type of gamma irradiator Co-60 was used which has remote controlled electromechanical system with a capacity of 65Kci. Five different doses of gamma radiation i.e., 100krd, 200krd, 300krd, 400krd and 500krd were applied for each type of sample.

2.2.8 Water Absorbency

Samples were prepared and tested according to the standard ASTM D570-98. Before immersing into the water, they were conditioned in an oven for 24 hours at 50°C, cooled in a desiccator, and weighted immediately to have the dry weight of each sample. The conditioned samples were then put in a beaker of water, maintained the temperature of 23 ± 2°C. The samples were taken out for maximum 2 minutes for measuring weight after every hour for the first four hours and then after every 4 hours over the 24 hours. Before measuring weight, the samples were wiped off every time to remove surface water. The water absorbency by weight percentage was calculated by the following Eq. (1).

\[
Water Absorbency(\%) = \frac{W_w - W_c}{W_c} \times 100
\]  

Where \(W_w\) is wet weight after water immersion and \(W_c\) is conditioned weight.

2.2.9 Mechanical Tests

Tensile properties like tensile strength (TS) and elongation at break percentage (Eb%) were tested according to the standard ASTM D638-14. A universal testing machine (UTM) from the brand Zwick was used for testing the samples at Laboratory of Materials Engineering, Kaunas University of Technology (KTU), Kaunas, Lithuania. Samples were prepared with according to the standard size of 165mm×13mm. A gauge length of 50mm was maintained. Load was applied at a constant rate of motion 10 mm/min of grips. The tensile strength and elongation at break were calculated by the following equations (2) and (3) respectively.

\[
TS = \frac{F_{\text{max}}}{A}
\]  

Where \(F_{\text{max}}\) is maximum load and \(A\) is cross-sectional area of the sample.

\[
Eb\% = \frac{\Delta l_b}{l_0} \times 100
\]  

Where \(\Delta l_b\) is elongation at breaking point and \(l_0\) is initial length of the sample.
Flexural property was tested with the same UTM according to the standard ASTM D790-03 to determine the flexural strength (FS) of the composites. Samples were prepared according to the standard and placed on the two supports with a span length of 16 times the thickness of the samples. Load was applied on the midspan with a constant deflection speed of 0.10 mm/mm/min until breaking. The flexural strength was calculated by the Eq. (4).

\[
FS = \frac{3FL}{2bd^2} \quad (4)
\]

Where \( F \) is breaking load, \( L \) is length of support span, \( b \) is width and \( d \) is thickness of the sample.

3. Results And Discussions

3.1 Water Absorbency

3.1.1 Effect of Alkali Treatment on Water Absorbency

Figure 3 (a) demonstrates the water absorbency of different types of banana nonwoven composites after 24 hours of water immersion. The effect of alkali treatments on the water absorbency of the composites are presented in Fig. 3 (b). Among the untreated composites, MR/E showed the lowest water absorbency of 15.14% and OB/P showed the highest absorbency of 28.63%. OB and MB composites exhibited higher absorbency and MR composites exhibited lower absorbency for both polyester and epoxy matrix. In comparison of epoxy and polyester composites, epoxy always showed lower absorbency than polyester composites. For instance, OB/E, MB/E, IB/E and MR/E was found 20.7%, 9.28%, 25.43% and 9.45% lower water absorbency than OB/P, MB/P, IB/P and MR/P respectively.

The water absorbency of the composite were varied with different types of banana fibers. This may be due to the different chemical compositions of the banana fibers from different parts of the banana trees. From the earlier studies, the chemical compositions of the banana stem fibers are also varied notably with different percentages of cellulose content such as 71.08% (Kenned et al. 2020b), 60–65% (Alavudeen et al. 2015), 57.6% (Mostafa and Uddin 2016), 43.46 (Wijianto et al. 2019). The other constituents like lignin and hemicellulose are also varied remarkably on these studied. This variation may occur due to different species of banana trees as well different sections of banana trees. OB nonwovens may contain more cellulose than other types. Thus, more hydrophilic sites of cellulose absorbed more water. The composites with epoxy matrix showed less absorbency than polyester matrix. This may be due to the better interfacial adhesion between fiber and epoxy matrix than polyester (Oliveira et al. 2019). Which results better covering of hydrophilic fibers by the hydrophobic resins and make them more watertight. Better adhesion also leads to remove the amorphous regions and porosity in the fiber-matrix interface thus, water absorbency reduces.
Alkali treatment improves the hydrophobicity of the composite samples which are clearly visible in Fig. 3 (b). The water absorbency of the composites were decreased significantly with the concentration of NaOH applied to the banana fibers. For example, at lowest concentration of 5% NaOH treatment, water absorbency was decreased by 12.5%, 12.9%, 17.7% and 17.9% for OB/E, MB/E, IB/E and MR/E respectively. In the same way, 24.1%, 17.1%, 13.6% and 17.2% decreases were found for OB/P, MB/P, IB/P and MR/P composites respectively. Even more influence was found at 10% of NaOH treatment. For instance, 27.5%, 24.8%, 23.7%, 25.6% decrease of water absorbency was found for OB/E, MB/E, IB/E, MR/E composites and 32.7%, 35.2%, 25.0%, 23.7% for OB/P, MB/P, IB/P, MR/P composites respectively in compared to untreated composites.

Water absorbency was continued to decrease at 15% NaOH as well. From all the 2nd order polynomial curve, it is evident that, the influence is lower in 15% than 10% concentration regarding the amount of NaOH. Between Epoxy and Polyester composites, Polyester composites were influenced more by the NaOH than Epoxy composites. For example, at 10% of NaOH treatment, water absorbency was decreased approximately 30% for Polyester composites and 22% for Epoxy composites in an average.

The hydrophobicity of the composites were improved with the various concentration of alkali treatments. This is because, alkali treatment removes unwanted materials like pectin, oil, wax, lignin, hemi-cellulose, and other impurities to a certain amount (Binti Mohd Hafidz et al. 2021). That results better adhesion between the fiber and matrix. Therefore, the strong interfacial bonding makes the fibers firmly protected with the hydrophobic matrix and decreases the water absorbency (Mohd Nazarudin et al. 2013). The application of NaOH also reduce the hydroxyl groups of cellulose (the main responsible group to absorb water) by ionizing them into alkoxides (Li et al. 2007; Reddy et al. 2018).

Water absorbency flows described in Fig. 4, shows that water absorbency rate is very fast in first couple of hours. It was observed that, approximately 40–50% water was absorbed in first two hours of 24 hours of complete observation. The rate can be considered medium during the time of 3–8 hours. Near about 80% water is absorbed after 8 hours of 24 hours. Then all the flows were become slow up to 20 hours and very slow up to 24 hours.

### 3.1.1 Effect of Water Repellent Treatment on Water Absorbency

The water absorbency of untreated (0% WR + 15% NaOH) composites after 24 hours of immersion into water is presented in Fig. 5 (a). OB/P composite showed the highest 15.55% and MR/E composite showed the lowest 10.16% of water absorbency. OB composites exhibit higher absorbency than all other three types of composites (MB, IB and MR) for both Epoxy and Polyester composites. Between the two types of matrices, Epoxy composites showed significantly lower water absorbency than Polyester composites except for MB composites where the absorbency is close to each other. For instance, OB/E, MB/E, IB/E and MR/E was found 13.44%, 0.43%, 21.16% and 17.03% lower water absorbency than OB/P, MB/P, IB/P and MR/P respectively.
For the further improvement of hydrophobicity, alkali treated nonwovens were treated again with water repellent chemical. Figure 5. (b) details the effects of WR on the water absorbency of the composites. It is evident that, water absorbency is decreased remarkably by the WR treatment for all types of composites. For instance, only at 5% concentration of WR treatment, water absorbency is reduced by 45.7%, 53.8%, 56.3%, 49.9% for OB/E, MB/E, IB/E, MR/E composites and 48.3%, 40.4%, 43.5%, 42.4% for OB/P, MB/P, IB/P, MR/P composites respectively in compared to untreated composites. Water absorbency was continued to decrease at 10% WR application. An overall 60–70% decrease of water absorbency was found at 10% WR in compared to untreated composites. However, from the 2nd order polynomial curves, the influence is lower at 10% WR when compared to 5% WR.

On the other hand, water absorbency started to increase at 15% WR for all the composites. The absorbency was increased by 7.7%, 3.5%, 0.9%, 5.0% for OB/E, MB/E, IB/E, MR/E and 7.1%, 11.6%, 6.9%, 8.0% for OB/P, MB/P, IB/P, MR/P composites respectively from the absorbency at 10% WR. WR treatment improves the hydrophobicity of the composites dramatically to a certain level of concentration. The WR chemical which was used in this study is perfluoroalkyl acrylic. This WR create a surface coating on the materials and consequently resist water molecules to enter inside of the material. WR may also cross-links with the cellulose to make them harder and rougher the surface. This rougher surface of the ber creates air trap on the surface that makes them more hydrophobic (Bae et al. 2009; Chowdhury 2018). However, at 15% WR, the water absorbency started to increase which is due to the thicker coating of the fiber surface. That weaken the interfacial fiber-matrix bonding and creates porosity between them. Therefore, water can penetrate on those perforated structures easily.

From the absorbency flows over the soaking times presented in Fig. 6, water absorbency was very fast in first couple of hours like the absorbency flows of alkali treated composites. About 50–60% of water was absorbed in first two hours and about 75–85% was in first eight hours. After that all the flows were become very slow. Moreover, at 24 hours, they seemed quite stable and absorbed the maximum amount of water. The main difference between the flows after alkali treatment (Fig. 2) and after WR treatment (Fig. 4) is, the curves look more stable after WR treatment than after alkali treatment at 24 hours. That gives an assumption of further water absorbency of the composites after the period of 24 hours. The composites after alkali treatment will have the possibility to take water over a long period of time whereas, the composites after WR treatment will have the possibility to stop taking water in a short time after the period of 24 hours.

### 3.2 Mechanical Properties

Mechanical properties like Tensile Strength (TS), Flexural Strength (FS) and Elongation at Break (Eb%) were analyzed in this study. The effects of alkali treatment, water repellent treatment and gamma radiation on these mechanical properties were also investigated for all the composites.

#### 3.2.1 Effects of Alkali Treatment
3.2.1.1 Effect of Alkali Treatment on Tensile Strength

Tensile strength of the untreated (0% NaOH) composites are illustrated in Fig. 7 (a). It is shown that, OB/E exhibits the highest TS (15.78 MPa) and IB/P exhibits the lowest TS (8.45 MPa) among all types of composites. Study revealed that, there was an influence of different fiber types which were used for making the nonwovens and composites subsequently. Among them, tensile strength was found with a sequence of OB > MB > MR > IB for both epoxy and polyester matrix composites where, 15.78 MPa, 14.56 MPa, 13.78 MPa, 12.23 MPa was found for OB/E, MB/E, MR/E, IB/E and 12.25 MPa, 10.11 MPa, 9.35 MPa, 8.45 MPa was found for OB/P, MB/P, IB/P, MR/P composites respectively. Among two types of matrices, epoxy composites demonstrated higher tensile strength than polyester composites. It is evident that, OB/E, MB/E, IB/E and MR/E composites were found 28.8%, 44.1%, 44.7% and 47.4% higher TS than OB/P, MB/P, IB/P and MR/P composites respectively.

There is apparent influence of alkali treatment on the tensile strength of the composites which is presented in Fig. 7 (b). It is evident that, TS was increased with the increase of NaOH concentration. At 5% of NaOH treatment, TS was increased by about 35% where, about 60% increase was found at 10% NaOH and 75% increase was found at 15% NaOH treatment in an average for all types of composites. For instance, at maximum 15% NaOH treatment, TS were increased by 71.4%, 67.1%, 74.8%, 72.9% for OB/E, MB/E, IB/E, MR/E composites and 63.3%, 75.7%, 69.5%, 75.8% for OB/P, MB/P, IB/P, MR/P composites respectively. The 2nd order polynomial curves prove that the impact of alkali is more in 5% and 10% than that of 15% of NaOH concentration.

3.2.1.2 Effect of Alkali Treatment on Flexural Strength

Figure 8 (a) shows the flexural strengths (FS) of the untreated composites and the effect of alkali treatment on flexural strengths are shown in Fig. 8 (b). Flexural strength was found in a sequence of OB > MB > MR > IB for both epoxy and polyester matrix composites. The highest 29.47 MPa FS was found from OB/E composite and the lowest 13.32 MPa FS was found from IB/P composite. Study revealed that, OB/E composites exhibited 56.1% higher FS than IB/E and OB/P composites exhibited 60.1% higher FS than IB/P composites for instance. That clearly defines the effects of fiber types from different parts of banana trees on the FS of the composites. Like TS, epoxy composites showed better FS than polyester composites. OB/E, MB/E, IB/E and MR/E composites were found 37.5%, 42.4%, 41.7% and 34.7% higher TS than OB/P, MB/P, IB/P and MR/P composite respectively.

The effect of alkali treatment on flexural properties of the composites followed the similar trend as tensile properties described earlier. Flexural strength was increased with increase of NaOH concentration. For example, FS were improved by 47.3%, 63.3%, 80.8%, 64.1% for OB/E, MB/E, IB/E, MR/E composites and 72.4%, 80.8%, 85.2% 65.2% for OB/P, MB/P, IB/P, MR/P composites respectively at a concentration of 10% NaOH than untreated composites. The improvement was even more at a concentration of 15% NaOH. From the 2nd order polynomial curve, impact is more at 10% NaOH than 15% by considering the amount of NaOH.
3.2.1.3 Effect of Alkali Treatment on Elongation at Break

Elongation properties of the composites shows exactly opposite trend of TS and FS which is presented in Fig. 9 (a). The highest Eb% was found from IB/P and lowest was from OB/E. In comparison of different nonwoven reinforcements, OB always showed lowest and IB always showed highest Eb% where MB and MR exhibited medium Eb% for both polyester and epoxy matrix composites. The composite with polyester matrix showed higher Eb% than epoxy matrix. Study found that, OB/P, MB/P, IB/P and MR/P demonstrated 11.8%, 32.1%, 25.8% and 29.5% of higher Eb% than OB/E, MB/E, IB/E and MR/E composites respectively.

The effects of NaOH on Eb% of the composites are illustrated in Fig. 9 (b). Alkali treatment reduces the Eb% to a small extent. The maximum 21.0%, 19.8%, 20.2%, 20.1% reduction of Eb% were found for OB/E, MB/E, IB/E, MR/E composites and 19.9%, 22.5%, 18.5%, 18.1% for OB/P, MB/P, IB/P, MR/P composites respectively at a concentration of 15% NaOH.

After analyzing the mechanical properties i.e., TS, FS and Eb% of the composites, the results can be summarized as, OB composites showed higher mechanical properties like TS and FS but lower Eb% than other type of nonwoven composites. Where IB composite exhibited lower TS and FS but higher Eb%. MB and MR can be considered medium in all the case. As discussed above, there may be a variation of chemical compositions of different types of banana fibers. OB nonwoven may contain higher percentages of cellulose that leads them to achieve better mechanical properties. The cellulose percentage may gradually decrease from the outer bark (OB) to the inner bark (IB) of the banana tree. As a result, lower cellulose content makes the IB fibers weaker and consequently lower mechanical properties in composites. Also, outer bark of the banana tree is found harder as a raw material that can make stronger materials than other layers of the banana stem. The midrib also found harder but surprisingly became soft after chemical extraction. The previous study also proved the higher mechanical properties of OB as a nonwoven material (Motaleb et al. 2020). Epoxy composite always found higher mechanical properties i.e., higher value of TS and FS but lower Eb% than polyester composites. This because of better interfacial bonding between fiber and epoxy that leads very good adhesion between them. As a result, the applied load can be distributed properly though the fiber and matrix which leads to bear higher loads. Similar results were found in some earlier studies (Rohen et al. 2018; Oliveira et al. 2019; Sivakandhan et al. 2020).

The alkali treatment demonstrated the improvement of mechanical properties like TS and FS but decrease the Eb%. As discussed above, alkali treatment eliminates some unwanted materials including lignin and hemicellulose. This elimination creates rough fiber surface that helps better mechanical interlocking among the fibers. By cleaning the impurities, the cellulose content of the fibers is increased which may increase the reactive sites and create strong bonding with the matrix. Therefore, the mechanical properties like TS and FS were improved. Due to the same reason, Eb% of the composites were decreased. As better as the adhesion between the fiber and matrix, the material become more solid.
and hard thus the elongation property is declined (Li et al. 2007; Mohd Nazarudin et al. 2013; Manalo et al. 2015; Preet Singh et al. 2017).

### 3.2.2 Effects of Water Repellent

#### 3.2.2.1 Effect of Water Repellent Treatment on Tensile Strength

Figure 10 reveals the effects of WR on the tensile strength of the composites. There is no doubt that, the hydrophobicity of the composites were improved to a great extent by the WR treatment. On the other hand, this treatment drew a negative effect on the tensile properties of the composites. Study found that, the maximum 27.3%, 25.1%, 31.4% and 25.4% decrease of TS were found for OB/E, MB/E, IB/E and MR/E composites where, 67.5%, 59.4%, 67.3% and 67.5% for OB/P, MB/P, IB/P and MR/P composites in compared to untreated composites respectively. However, the deterioration is very low (about 4–14%) at 5% WR concentration. For instance, the TS of OB/E, MB/E, IB/E and MR/E composites were decreased by 5.3%, 4.4%, 7.0% and 6.2% where, the TS of OB/P, MB/P, IB/P and MR/P composites were decreased by 6.7%, 6.0%, 12.9% and 13.2% respectively at a concentration of 5% WR. At 10% WR, the TS of epoxy composites were reduced by approx. 10% where the TS of polyester composites were reduced by approx. 34% in an average. Likewise, at 15% WR, polyester composites were also showed higher reduction (approx. 65%) than epoxy composites (approx. 27%) in an average.

#### 3.2.2.2 Effects of Water Repellent Treatment on Flexural Strength

Effect of WR on the flexural properties are evident in Fig. 11. Similar negative trend was found for all the composites like TS as it declined the FS to a large extent of about 40–50% (in an average) after treating with 15% WR in compared to untreated composites. But the effect was much lower at 5% and 10% WR. For examples, the FS of OB/E, MB/E, IB/E and MR/E composites were declined by 4.4%, 3.9%, 7.5% and 6.0% while, the FS of OB/P, MB/P, IB/P and MR/P composites were declined by 4.1%, 2.6%, 11.0% and 8.8% respectively at 5% WR in compared to untreated composites.

#### 3.2.2.3 Effects of Water Repellent Treatment on Elongation at Break

Elongation of the composites were increased with the increase of WR% that is clearly defined in Fig. 12. The maximum increases of Eb% were found at 15% WR. For example, OB/E, MB/E, IB/E and MR/E composite exhibited 63.8%, 66.7%, 62.0% and 61.3% of increment at 15% WR than untreated composites. At 10% WR, the effect was lower, Eb% were increased by 25% (approx.) on an average considering all types of composites. Furthermore, at 5%WR, the effect was very low as Eb% was increased by about 10% maximum from the untreated composites. The 2nd order polynomial curves also prove this trend.

The application of WR on the nonwoven surface, declined the mechanical properties like TS and FS but as coating on the fiber surface and resist water
to penetrate inside the fiber thus improve the hydrophobicity. However, because of this coating or polymer blockage, mechanical properties can be reduced. The fiber-matrix interface can be disrupted by this type of coating that results poor adhesion between the fiber and matrix thus the poor mechanical properties of the composites. The good thing is, this effect is negligible at lower concentration like 5% WR. Study revealed that the deterioration of TS and FS in about 10% maximum for all types of composites at a concentration of 5% WR. Whereas the hydrophobicity was increased by 40–50% (in an average) at the same concentration of WR. Therefore, it is recommended to apply the WR with a concentration of 5% to balance the water absorbency and mechanical properties.

### 3.2.3 Effects of Gamma Radiation

Effects of gamma radiation on mechanical properties like tensile strength, flexural strength and elongation at break were investigated in this study. The results are described in 2nd order polynomial curves because the mechanical properties were influenced by gamma radiation in two opposite factors.

#### 3.2.3.1 Effect of Gamma Radiation on Tensile Strength

Figure 13 depicts the influence of gamma radiation on tensile properties of the composites. All the curves demonstrate that, gamma radiation improves the mechanical properties significantly to a certain level of dose. The TS of OB/E, MB/E and MR/E composites were improved by maximum 31.2%, 33.3% and 37.7% at a gamma radiation dose of 200krd where, the TS of IB/E was improved by 20.1% at a dose of 100krd respectively in compared to non-irradiated composites. However, the TS of the composites were decreased to a large extent by further increasing of gamma radiation dose. For instance, the TS of OB/E, MB/E, MR/E composites were decreased by 8.5%, 16.3%, 13.1% at 300krd and the TS of IB/E was decreased by 7.4% at 200krd respectively from the maximum value. Similar trend was found for Polyester composites as increased the TS by 37.7%, 41.4%, 30.0% for OB/P, MB/P, MR/P composites at 200krd and 24.9% for IB/P composite at 100krd but after that the TS was decreased with the increase of radiation dose. At higher dose like 500krd, TS was drastically by 60% in an average for all types of composites from the maximum value of TS, which is even less than half of the TS of nonirradiated composites. Both the polyester and epoxy composites were influenced by the gamma radiation in the same ways though the TS of epoxy composites were improved slightly higher in percentage than polyester composites.

#### 3.2.3.2 Effect of Gamma Radiation on Flexural Strength

The effect of gamma radiation on the flexural strength of the composites are presented in Fig. 14. Similar tendency like TS was found in this case as well; the FS of the composites were enhanced noticeably to a certain level of dose and reduced to a large amount after that certain level. It is evident that, FS were improved by 37.8%, 39.2%, 39.8% for OB/E, MB/E, MR/E composites and 37.3%, 34.4%, 40.8% for OB/P, MB/P, MR/P respectively at a gamma radiation dose of 200krd. IB composite such as IB/E and IB/P showed the maximum 21.1% and 31.2% improvement of FS at a radiation dose of 100krd. Further increment of gamma dose, for example at 300krd, FS were decreased by 12.9%, 14.5%, 25.0%, 17.6% for OB/E, MB/E, IB/E, MR/E composites and 22.3%, 17.2%, 29.4%, 14.3% for OB/P, MB/P, IB/P, MR/P.
composites respectively. FS were fallen dramatically at higher dose like 500krd. FS were found 32.89, 27.69, 14.34 and 23.92 MPa for OB/E, MB/E, IB/E and MR/E composites; 22.38, 20.79, 6.67 and 18.36 MPa for OB/P, MB/P, IB/P and MR/P composites respectively at a gamma dose of 500krd which is almost half of the FS of nonirradiated composites.

The improvement of TS and FS through gamma radiation is mainly due to the improvement of polymeric bonding among the intra-chain of fiber and matrix by cross-linking each other. Which leads more oriented polymeric structures and better fiber-matrix adhesion, therefore increase the mechanical properties like TS and FS (Masudur Rahman et al. 2018; Gnatowski et al. 2020). Gamma radiation is a powerful ionizing radiation which has an ability to penetrate the materials and influence the polymeric structure by producing reactive sites like free radicals, ions, and peroxides. Consequently, these reactive species can cross-link or, bind to each other to form long polymeric chains or, large molecules and leads to change the mechanical properties of the materials. It is also evident from several studies that, gamma radiation can break the C = C bond and generate free radicals, subsequently improve the mechanical properties (Haydaruzzaman et al. 2009; Masudur Rahman et al. 2018; Martínez-Barrera et al. 2020). Gamma radiation may also extract the inside moisture of the composites which is also a possible reason for improving the properties (Khan et al. 2009; Martínez-Barrera et al. 2020).

In spite of that, mechanical properties like TS and FS were started to decrease after a certain level of dose. This reduction is because of another aspect, usually known as chain scission or, chain degradation which is completely opposite of chain cross-linking. At higher gamma radiation dose, the main polymeric chains are destroyed into small particles. Thus, the mechanical properties like TS and FS are decreased (Masudur Rahman et al. 2018).

3.2.3.3 Effect of Gamma Radiation Treatment on Elongation at Break

Effect of gamma radiation on Eb% of the composites are revealed in Fig. 15. The Eb% was reduced by the gamma radiation to a small amount up to a certain level of irradiation then increased gradually. At a dose of 200krad, Eb% were decreased by 20% (maximum) from the nonirradiated composites, considering all types of composites except IB composites. They were exhibited the lowest Eb% at 100krd. After this certain level of doses, EB% were increased with the increment of gamma radiation doses. The highest Eb% were found at a radiation dose of 500krd as 1.59, 1.78, 2.18 and 1.92% for OB/E, MB/E, IB/E and MR/E composites while, 1.66, 1.94, 2.86 and 2.57% for OB/P, MB/P, IB/P and MR/P composites respectively, the values are 80–90% higher than nonirradiated composites.

As described above, gamma radiation leads strong cross-linking among the intra-chains of fibers and matrices that ensures better adhesion between them. The resultant polymeric structures become more crystalline and limit the movement of polymer chains which leads lower Eb% of the materials (EL-Zayat et al. 2019). In other words, the more oriented structure makes the materials more solid and harder that reduce the elongation properties. However, at higher dose the main polymer chains and fiber-matrix...
bonding may destroy into small pieces. That results severe disorder of polymeric structure which leads higher elongation.

## 4. Conclusions

The current study reveals the developments of an innovative natural composite materials by reinforcing different banana fiber nonwovens which were developed by a special manual technique of wet laid web formation. The outcome of this study can be summarized by the following points.

1. OB composites showed higher mechanical properties (TS and FS) and higher water absorbency than other nonwoven composites. Between the two matrices, polyester composites exhibited higher absorbency and lower mechanical properties than epoxy composites.

2. The hydrophobicity and mechanical properties of the composites were improved significantly by the alkali treatment due to the better fiber-matrix adhesion which is achieved by removing unwanted materials from the fibers through this treatment. For instance, about 32% decrease of water absorbency, 71% increase of TS and 87% increase of FS was found on an average at a concentration of 15% NaOH.

3. The hydrophobicity was continued to improve remarkably by the water repellent treatment on the nonwovens by creating a surface coating on the materials. On the other hand, the mechanical properties were decreased by disrupting the fiber matrix bonding through this treatment. But the good thing is, this declination is less than 10% approximately at a concentration of 5% WR with the significant improvement of hydrophobicity by 47.5% on an average. Therefore, this study recommends applying the WR with the maximum concentration of 5% to balance the water absorbency and mechanical properties.

4. Gamma Radiation improved the mechanical properties like TS, FS and decreased Eb% due to more oriented polymeric structure achieved by this radiation. Maximum 30% of TS and 35% of FS were increased at a radiation dose of 200krd but further increasing of dose decreased the properties due to breaking of main polymeric chains at higher radiation. Thus, this study recommends gamma radiation dose of maximum 200krd.

Based on the achieved results, it is evident that, banana fiber nonwoven reinforced composites are well developed by different physical and chemical treatments on their pre and post manufacturing stages. The developed material demonstrates excellent hydrophobicity and comparable mechanical properties which can replace the existing non-biodegradable, carcinogenic and synthetic materials on the market.

## 5. Declarations

### Funding
Not applicable.

### Conflicts of interest
The authors declare that they have no conflicts of interest.
Consent to participate Not applicable.

Consent for publication Not applicable.

Availability of data and material Not applicable.

Code availability Not applicable.

Permission for collecting samples The authors clarify that the plant samples were collected with appropriate permissions from the owner of the plant.

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Figures
Figure 1

Banana tree and cross-section of banana stem.
Figure 2

Prepared composite samples for the tensile test (from the left: OB/P, OB/E, MB/P, MB/E, IB/P, IB/E, MR/P and MR/E).
Water absorbency percentage of composites after 24 h water immersion, (a) Water absorbency of untreated (0% NaOH) composite samples; (b) Effect of alkali treatment on the water absorbency of the composite samples.

![Graph showing water absorbency percentage over time for different composite samples treated with various alkali concentrations.](image)

**Figure 4**

Water absorbency flows of 15% NaOH treated composite samples by the soaking time up to 24 hours.

![Bar chart showing water absorbency percentage for different composite samples.](image)

(a) Composite Samples

(b) Water Repellent (%)

**Figure 5**
Water absorbency percentage of composites after 24 h water immersion, (a) Water absorbency of untreated (0% WR, 15% NaOH) composites; (b) Effect of WR treatment on the water absorbency of the composites.

Figure 6

Water absorbency flows of 10% WR treated composite samples by the soaking time up to 24 hours.

Figure 7
(a) Tensile strength of untreated (0% NaOH) composites; (b) Effect of alkali treatment on the tensile strength of the composites.

Figure 8

(a) Flexural strength of untreated (0% NaOH) composites; (b) Effect of alkali treatment on flexural strength of the composites.

Figure 9

(a) Eb% of untreated (0% NaOH) composites; (b) Effect of alkali treatment on Eb% of the composites.
Figure 10

Effect of water repellent on tensile strength of the composites.
Figure 11

Effect of water repellent on flexural strength of the composites.
**Figure 12**

Effect of water repellent on flexural strength of the composites.
Figure 13

Effect of gamma radiation on tensile strength of the composites.
Figure 14

Effect of gamma radiation on flexural strength of the composites.
Figure 15

Effect of gamma radiation on elongation at break (%) of the composites.

Supplementary Files

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