

# Effect of Nanomaterials on the Mechanical and Morphological Properties of Jute-GFRP Hybrid Nanocomposites



M S Rabbi<sup>1,\*</sup>, Snigdha Das<sup>1</sup>, Sanjida Sharmin<sup>1</sup>, Abdullah Al Mamun<sup>1</sup>

<sup>1</sup> Department of Mechanical Engineering, Chittagong University of Engineering & Technology, Chattogram-4349, Bangladesh



### **1. Introduction**

Composite is a multi-phase system in which the phase distribution and geometry are deliberately tailored to optimize one or more properties. Composite materials are comprised of at least two materials with physically detachable phases. One phase is usually continuous and is designated the "matrix", which holds reinforcement in aligned positions as separate entities and transfers the

**.** 

<sup>\*</sup> *Corresponding author.*

*E-mail address: rabbi@cuet.ac.bd (M. S. Rabbi)*

*E-mail of co-authors: u1503051@student.cuet.ac.bd, u1703100@student.cuet.ac.bd, amamun@cuet.ac.bd*



external load to the reinforcement. It also protects reinforcement from the external environment, slowing down or stopping crack propagation. The other phase (the "reinforcement") remains embedded in the matrix phase and carries the stress applied to the composite. The intention is to increase the mechanical properties of the matrix. Broadly, there are two fibrous composites: synthetic fiber-reinforced composites and natural fiber-reinforced composites.

In recent years, the development of natural fibers that may satisfy the needs of many engineering applications has been sparked by the growing global demand for lightweight, ecologically friendly, high stiffness, specific strength and as a possible alternative to synthetic fibers [1-3]. Due to their inherent benefits, such as renewability, biodegradability, affordability and adequate specific strength and stiffness, natural fiber-reinforced polymer composites have attracted a lot of attention. Notably, jute fibers, derived from the bast of the Corchorus plant, have emerged as a promising natural reinforcement due to their abundant availability and eco-friendly characteristics. It has been claimed that employing fiber composite parts can reduce vehicle weight and prices by 20 and 30 percent, respectively [4].

Natural fibers are generally composed of cellulose, hemicelluloses, lignin and waxes. Lignin helps keep the water in fibers, protects against the biological attack and strengthens stems. Cellulose content present in natural fiber mainly affects the mechanical properties of the fiber-reinforced composite. Fibrous composites are the most widely used polymer composites as they have found applications in various areas such as automotive, marine, and aerospace due to their high specific stiffness and strength. The fibers used are either continuous or discontinuous in the form of chopped fibers or short fibers [5-7]. Continuous fibers loaded to the fiber direction provide the best mechanical properties and exhibit considerable anisotropy.

Still, most natural fiber-based composites lack mechanical properties compared to synthetic fiber ones, as the mechanical properties of natural fibers are considerably poorer than synthetic fibers. Other drawbacks of natural fiber composites are their high susceptibility to moisture absorption, poor dimensional stability, and variability in properties, low thermal resistance and incompatibility with polymeric matrices resulting in deficient fiber/matrix interfacial adhesion. Combining natural and synthetic fiber in the same matrix is a feasible method of overcoming these drawbacks. According to Ashby, a hybrid material is a mixture of two or more materials whose size and shape have been set to carry out a given function as efficiently as possible [8]. A desirable method for creating goods with lower costs, a high specific modulus, strength, resistance to corrosion, and frequently great thermal stability is using hybrid composites [8,9]. The most widely used form of hybrid composites is when more than one kind of reinforcement (typically fibers) is used in the same polymer matrix, and this study will deal with this form. Hybridization of natural fiber composite by another natural fiber does not yield superior mechanical properties as hybridization by synthetic fiber, fiber metal laminates and filler particles. Hybridization of more robust, stiffer and more resistant to corrosion synthetic fibers (glass fibers) and biodegradable, cheaper natural fibers offer an equilibrium between degradability and functioning characteristics. Numerous research studies have been reported on developing hybrid polymer composites from both thermosets and thermoplastic by utilizing synthetic and natural fibers, and the combination of fibers with nanomaterials and their mechanical and thermo-mechanical properties elucidated [10]. Besides that, hybridizing the lignocellulosic fibers with a more robust and more corrosion-resistant synthetic fiber such as glass fiber can significantly improve the strength, stiffness, moisture and fire-resistant behavior of lignocellulosic composite.

Another way of increasing the properties of composites is the incorporation of nanofillers. Nanomaterials' mechanical, thermal, optical, electrical, magnetic and barrier properties differ from those of pure polymers[11-13]. Therefore, these particular properties of composites can be improved by incorporating nanofillers, enabling a wider range of applications [14,15]. Nanocomposites are



designed by adding nanoscale fillers to a polymer matrix to fulfill the expanding needs for specific qualities in a variety of industrial and practical applications [16,17]. Nanocomposite materials have at least one phase with a dimension of 100 m or less [18]. It also improves crystallinity, the formation process, the polymer chain's order, the polymer's optical and chemical properties, corrosion resistance and cost-effectiveness [19,20]. Due to the very small number of nanoparticles introduced in comparison to the bulk phase, the material weight also decreases [21]. Nanocomposite materials are considered potential substitutes to overcome the drawbacks of conventional and monolith polymer composites [22,23]. Two main approaches used for synthesizing nanomaterials are topdown approaches and bottom-up approaches. In top-down approaches, bulk materials are divided to produce nanostructured materials. Top-down methods include mechanical milling, laser ablation, etching, sputtering and electro-explosion. Bottom-up approaches include chemical vapour deposition, sol-gel processes, spray pyrolysis, laser pyrolysis and atomic/molecular condensation. Different types of nanofillers are available that improve material characteristics without sacrificing density, toughness, or processability [24,25].

Hybridization of natural fiber with synthetic fiber is the way to improve the composite's properties. The point of bringing two fiber types in a single matrix is to maintain both fibers' advantages and attenuate the disadvantages. Natural fibers are mainly hybridized with glass fibers because of their comparable properties and low cost. Vishnu *et al.* reported that a uniform dispersion of nanoparticles in the matrix promotes the formation of a good fiber-matrix interface and reduces fracture propagation in polymer composites [26]. Using nanocellulose-reinforced composites has always shown an improved performance. Khan *et al.* studied the effect of nanofibers and nano clay on kenaf/epoxy composites and found hybrid composites suitable for construction applications [27]. Softwood kraft pulp/epoxy nanocomposites were made up by the hand lay-up technique, and remarkable improvement was seen for the composite with 0.70 wt.% of nanocellulose [28].

Two natural and one synthetic fiber-reinforced hybrid composites made using the hand lay-up method have been used to enhance their mechanical properties. Arup Mandal *et al.* studied nanocomposites based on PVA and nanocellulose isolated from waste sugarcane bagasse which demonstrated a notable improvement in tensile strength, percent elongation at break, yield force and toughness in both linear PVA and crosslinked PVA [29]. The impact of bamboo nanoparticles in kenaf fiber-polyester composite, as investigated by Rosamah *et al*. [30], results in 3% addition of nanofillers contributed to a strong bonding and increased wettability with matrix, which gives superior mechanical properties and thermal properties of the composite.

In the study of an epoxy composite reinforced with glass/nylon fibers and filled with alumina, Dilip *et al.* observed that the nanocomposite's flexural properties are improved by the addition of 2 wt.% alumina nanofillers to epoxy [31]. Akash Mohanty *et al.* have investigated the effect of alumina nanoparticle in hybrid carbon/glass-reinforced epoxy composite and found that the elastic modulus of the composite has been significantly improved, whereas tensile strength also enhanced [32]. Özdemir *et al.* demonstrated incorporating lead oxide nanoparticles into a synthetic elastomer called Ethylene Propylene Diene Monomer (EPDM) polymer improves the thermal, mechanical, and radiation properties of polymer matrix composites [33]. Madhu P. *et al.* investigated glass/silk/Ca<sub>2</sub>SiO<sub>4</sub> reinforced hybrid epoxy composites by hand lay-up method with the hybrid composites of glass fiber (50 wt.%), silk fiber (10, 9, 8 & 7 wt.%), Ca<sub>2</sub>SiO<sub>4</sub> nano filler (with 0, 1, 2 and 3 wt.%) reinforced with epoxy (40 wt.%) were scrutinized and results revealed that the composite laminates with 3 wt.%  $Ca<sub>2</sub>SiO<sub>4</sub>$  show better tensile, flexural and impact strength properties [34]. In the study conducted by Shukla *et al.*, the fracture toughness and higher crack velocity in nano-filler nanocomposites using TiO<sub>2</sub> (35 nm) and  $Al_2O_3$  (14 nm) nanoparticles were improved when compared to the neat epoxy resin [35].



Abenojar *et al.* demonstrated that increasing the number of nanoparticles in polymer composites affects the curing process and glass transient temperature [36].

Jute fibers exhibit reasonable tensile strength and stiffness-to-weight ratios. Incorporating jute into a hybrid composite can enhance or balance these properties in the final material. Swain and Biswas [37] have studied ceramic-filled jute/epoxy hybrid composites implying Al<sub>2</sub>O<sub>3</sub> filler at wet and dry conditions. In their study, composite samples were fabricated with varying proportions of fiber loading, epoxy, and filler particles. The water absorption characteristics of these composites were found to be dependent on the fiber content and filler composition. Interestingly, an increase in fiber loading led to a corresponding enhancement in the hardness of the composite [37]. However, it was also observed that all mechanical properties experienced a decrease following water absorption. In a separate investigation, Gujjala et al., mentioned in Raghavendra et al. [38], examined a jute/glass fiber-reinforced epoxy composite. Their findings revealed that the inclusion of 4 wt.% alumina nanofillers resulted in optimal tensile and flexural properties.

Though much work has been done on a wide variety of hybrid nanocomposites, very little has been reported on the hybridization of three types of fiber to obtain the required characteristics of composites. Several articles cover the effect of fillers in a polymer composite, the most used hybrid reinforcement (natural/synthetic fiber) with micro or sometimes nanosized filler. Some researchers also reported the effect of fiber treatment in a polymer composite. Still, a notable lack of research exists on the combined effect of nanofillers addition and fiber surface treatment to improve the performance of hybrid composites. However, the possibility that incorporating nanomaterials with natural fibers with GFRP in polymer could provide a synergism in terms of improved performance has not been adequately addressed so far. This study intends to develop a hybrid composite using both jute and banana fiber incorporation with glass, which is expected to have better mechanical characteristics than conventional two fiber-based hybrid composites. This study mainly focuses on nanofillers' influence on surface-treated natural fiber polymer properties. It shows the effect of three different types of nanomaterials, consecutively,  $Al_2O_3$ , ZnO and TiO<sub>2</sub> in jute-glass. The main objective of this study is to study the influence of nanomaterials on the mechanical and morphological properties of hybridization with the natural fibers in the matrix. Moreover, the influence of three types of nanomaterials on natural-synthetic fiber-based hybrid composites is investigated. Furthermore, the influence of the sequence of laminates on the mechanical behaviour of the composite is evaluated. In addition, the surface morphology is studied using Scanning Electron Microscopy (SEM) analysis.

This pioneering study aims to establish a comprehensive framework for harnessing the potential of naturally occurring fibers in the commercial production of composite materials. The scope of this work extends beyond the present, envisioning future adaptations that involve the strategic reconfiguration of the hybrid composite's layout by varying the orientations of the constituent fibers. Moreover, the research contemplates the introduction of an additional fiber layer sandwiched between the existing banana and jute layers, with a subsequent exploration of its unique properties.

### **2. Materials and Methods**

### *2.1 Materials*

This study is centered around the comprehensive analysis of two primary constituents that form the core elements of the examined composites: the reinforcement components (banana fiber, jute fiber, and E-glass fibers) and the matrix constituents (resin). The jute fiber mats were procured from local markets located in Chittagong, Bangladesh. The commercial-grade epoxy resin and the requisite hardener were sourced from a local supplier. A bidirectional woven mat of glass fiber, weighing



300gsm, was employed for fabricating the specimens. Aluminium oxide, zinc oxide, and titanium oxide were procured from a local supplier, while chemicals like sodium hydroxide and distilled water were sourced from a local chemical supplier.

Jute is a bast fiber obtained from two herbaceous annual plants, white Corchorus capsularis (white jute) from Asia and Corchorusolitorius (Tossa jute) from Africa. The white jute (Corchorus capsularis) is the best known among over thirty Corchorus species [34,39]. Jute takes nearly three months to grow to a height of 12-15 ft, during the season and then cut, bundled and kept immersed in water for the "retting" process, where the inner stem and outer, get separated, and the outer plant gets 'individualized', to form a fiber. After drying, the fiber is taken to jute mills to convert it into jute yarn and hessian. Tables 1 and 2 show jute fibre's mechanical properties and chemical composition, respectively.

#### **Table 1**

#### Mechanical properties of jute fiber [40]



#### **Table 2**

#### Physical and chemical composition of jute fiber [41]



Fiberglass is an immensely versatile material due to its lightweight, inherent strength, weatherresistant finish and variety of surface textures [42]. It is a common type of fiber-reinforced plastic using glass fiber, also called glass fiber-reinforced plastic. The fibers may be randomly arranged, flattened into a sheet called a chopped strand mat, or woven into glass cloth. The plastic matrix may be a thermoset polymer matrix. Fiber-reinforced plastic (FRP) composites possesses interesting properties like high specific strength and stiffness, good fatigue performance and damage tolerances, low thermal expansion, non-magnetic properties, corrosion resistance and low energy consumption during fabrication [43].

Epoxy compounds are characterized by an oxirane or epoxy ring and represented by a threemember ring containing an oxygen atom that is bonded with two carbon atoms already united in some other way. The epoxy resin consists of monomers or short-chain polymers with an epoxide group at either end. Epoxies are available in liquid and solid forms and cured into the finished plastics by a catalyst. They are cured at room temperatures as well as elevated temperatures.

For any epoxy, it requires a hardener to initiate curing. It is also called the catalyst, the substance that hardens the adhesive when mixed with resin. When resin and hardener react together, the hardener opens the C—O—C rings, and the bonds are rearranged to join the monomers into a threedimensional network of crosslinked chain-like molecules. The hardener consists of polyamine monomers. Polyamine and acid anhydride are the most widely used curing agents because they can provide very active groups.

Three types of inorganic oxide particles were used in this work: Al2O3, ZnO, and TiO<sub>2</sub>. They were purchased from Taj Scientific Stores, Chittagong, Bangladesh.

Aluminum Oxide: The hardest form of alumina, aluminium oxide (Al<sub>2</sub>O<sub>3</sub>), is typically employed as a filler to enhance polymers' mechanical and thermal properties [44].



- Zinc Oxide: ZnO improves the composite's thermal, mechanical, UV resistance, and other pertinent qualities [45]. Since the materials have a straight and wide band gap, they may play a significant role in energy interaction and conversion. Its near-UV spectral area has a direct and large band gap, which is a benefit that might be employed for photoelectronic activities [46].
- Titanium Oxide: Titanium oxide nanoparticles attract increasing attention not only for their unique properties but also for their potential applications in industries such as pigments, cosmetics, catalysts, photocatalysts, etc. [47].

Ball milling machine was used to prepare the nanoparticles for each oxide. The average particle size was found to be  $40 - 70$  nm.

### *2.2 Chemical Treatment of Fiber*

Within the scope of this research, jute woven fabrics and Glass Fiber Reinforced Plastic (GFRP) materials were procured and meticulously tailored to match the desired specimen length. The precise quantification of these materials was achieved by employing a precision balance. Afterwards, the fibers underwent thorough cleansing using pressurized water, eliminating undesired organic residues that might have been present on their surfaces. The treated fibers were then subjected to an eighthour solar drying period to eliminate residual moisture content.

Jute fibers are recognized for bearing hydroxyl groups attributed to the presence of cellulose and lignin. This hydroxyl group is involved in hydrogen bonding within the cellulose, thereby reducing the activity towards the matrix. As a result, when the hydrophilic fiber is reinforced with hydrophobic resin fiber swelling within the matrix occurs, causing weak bonding between fiber and matrix, dimensional instability, matrix cracking and poor mechanical properties of the composites [48].

Fiber surface modification with different chemical treatments is done to enhance the effectiveness of interfacial bonding and to reduce moisture absorption. Chemical treatment of reinforcing fiber activates the fiber structure using a hydroxyl group that changes the fiber element's composition to interact with the matrix [49]. As a result, adhesion between the fiber surface and polymer matrix is improved, resulting in increased mechanical properties and strength of the fiberreinforced composite.

Chemical treatment with NaOH removes moisture from the fibers, increasing its strength. Also, it enhances the flexural rigidity of the fibers, clears all the impurities adjoining the fiber material, and stabilizes the molecular orientation. It also helps reduce the fiber diameter, increasing the aspect ratio.

In alkalization, the Jute fibers were soaked in a 5% (w/v) aqueous alkaline (NaOH) solution separately at room temperature, maintaining fibers, solution ratio of 1:15 (w/w) for 24 hours with intermittent stirring for proper de-polymerization of cellulose, hemicellulose, pectin and lignin. Alkali treatment swells the amorphous regions of the fibers and in doing so, causes the removal of approximately 41% of the hemicellulose cementing material in the fiber structure. After soaking, the fibers were rinsed several times with distilled water to remove the excess NaOH sticking on their surface. The neutrality of the fibers was verified using a pH paper followed by thorough rinsing with distilled water until they were alkali-free. The final pH was maintained at 7. Finally, the fibers were dried in open air/sun for hours and dried in the oven at 100°C for 8 hours. Chemical treatment with alkalis increases the surface roughness of the NF's. This improves the agglutination between the polymer and fiber. Untreated and treated jute fiber mats are depicted in Figure 1.





**Fig. 1**. (a) untreated and (b) alkalized jute mat

### *2.3 Fabrication of Specimen*

The technique employed for manufacturing the composite specimens is known as the hand layup method, encompassing entirely manual procedures. Throughout this experiment, fiber-reinforced hybrid composites were synthesized under ambient conditions, marked by a room temperature setting and an average relative humidity of 65%.

The initial preparation of all the materials and tools that will be used is a fundamental standard procedure when working with composites. The term "mold" herein refers to either a male or female mold utilized for composite part creation or a flat surface where fibers are impregnated. A wooden mold was precisely fashioned in this endeavour, boasting dimensions of 500 mm  $\times$  500 mm  $\times$  25 mm.

The first step of matrix preparation is the amalgamation of resin, nanofillers, and, finally, the hardener. Epoxy resin and nanoparticles were continuously and thoroughly mixed with a beater for 30 minutes and then prescribed quantity of hardener was added to the mixer. The epoxy to hardener ratio selected was 10:1 as recommended by the supplier and the percentage of nanofillers were 3%. The mixture was poured onto the mylar paper and spread over the mold with a paint brush. Then one ply glass fabric was placed on to the epoxy resin mixture and rolled with hand roller. The second layer of fiber, i.e. jute mat was added at this stage and special care was taken to eliminate all air bubbles possible. Before the resin got dried, the subsequent layers were filled using the corresponding fibers. In a lay-up, the heavier the reinforcement material, the greater the amount of resin needed to saturate the material. An optimal fiber to resin ratio is typically 60 percent fiber to 40 percent resin mixture. 2 layers GFRP and 2 layers of Jute are placed. Finally, it was sandwiched with two pieces of mylar paper, and a squeegee or flexible plastic spreader was used to distribute the resin evenly and remove excess resin until the material was saturated. The finished composite was closed by wooden reapers and was clamped at all possible ends. After curing at room temperature for 24 - 48 hours, the mold was opened and the developed composite part was taken out and further processed. After fabrication, the composite specimen was kept in sunlight for several hours to remove the moisture content. The sequences of the fiber layers were different for different samples. A total of 12 samples were prepared, as follows:

Sample 1 (Without nanoparticles)

- i. Jute + Glass + Jute + Glass (JGJG)
- ii. Jute + Glass + Glass + Jute (JGGJ)
- iii. Glass + Jute + Jute + Glass (GJJG)



#### Sample 2 (with nano  $Al_2O_3$ )



# *2.4 Characterization Methods 2.4.1 Tensile Test*

The tensile strength of the composite depends on how well the stress can be transferred from the broken to the surviving fibers through shear in the resin at the interface and the amount of stress a sample can withstand before failure occurs [50]. The testing process involves fixing the test specimen in a testing machine and slowly extending it by applying load until it fractures. The tensile properties of the composite samples were evaluated according to ASTM D3039, using rectangularshaped samples.



**Fig. 2.** (a) Cut-out specimens for the tensile test, (b) Specimen under tensile load

A universal testing machine (UTM) with a maximum load of 400 KN was used for testing. A crosshead speed of 3 mm/min and a gauge length of 127 mm was maintained. The longitudinal unidirectional fiber embeds in the matrix experienced the axial load, and fracture occurred when the material lost its elastic nature and extended plastically. Three different composite specimens with different fiber stacking sequences were tested, and the tensile properties, such as tensile strength (TS) elongation at break percentage (EB%) and Young's modulus (Y) of the prepared composites were then evaluated. The prepared specimens and the machine used are depicted in Figure 2.



# *2.4.2 Flexural Test*

The three-point static flexural test is the most common flexural test and was used in this experiment. This test determines the specimen's behaviour when subjected to simple beam loading the maximum stress induced in the outermost fiber. The prepared flexural test samples were tested by applying a three-point bending load with the aid of a UTM machine. The bending was carried out on rectangular-shaped flexural specimens of 150 × 12.7 × 6 mm according to the ASTM D790 standard at a strain rate of 0.5 mm/min. The distance between the two supports was maintained at 60 mm. Flexural strength and flexural modulus can be calculated using standard relationship among applied load, span length width of the specimen, the thickness of the specimen and deflection produced during testing. The prepared specimens and the machine used are depicted in Figure 3.



**Fig. 3.** (a) Test set up for three-point bending, (b) Specimen after flexural test

### *2.4.3 Hardness Test*

The hardness properties of the composites are studied by applying an indentation load normal to fibers diameter and normal to fiber length. In this experiment, The Rockwell test determines the hardness by measuring the depth of penetration of an indenter under a force of 187.5 kgf using the L scale. Test specimens were made according to the ASTM D 785. The diameter of the ball indenter used was 2.5 mm and the maximum load applied was 187.5 kg as per the standard L-scale of the tester. All the readings were taken 10 s after the indenter made firm contact with the specimen. Top hardness numbers that are obtained from hard materials indicate a shallow indentation while low numbers found with soft materials indicate deep indentation.

# *2.4.4 FE-SEM Analysis*

Field emission scanning electron microscopy (FE-SEM) generates images of the materials' microstructures that are three to six times crisper, less electrostatically distorted and with spatial resolution down to nanometers than conventional SEM. FE-SEM pictures can be used to identify defects such as internal cracks, voids, and interfacial behavior. The specimen was cut into small pieces measuring 2 mm<sup>2</sup> to 3 mm<sup>2</sup> approximately prior to analysis. The failing surface of the tensile testing materials underwent morphological examination using the Sigma 300 (Carl Zeiss).



# **3. Results and Discussion**

### *3.1 Tensile Properties*

The specimens of different combinations of hybrid composite laminates of GFRP and jute fibers, i.e. JGJG, JGGJ and GJJG, were subjected to tensile testing using a universal testing machine. The mechanical properties, such as maximum load, strain, ultimate tensile strength, and modulus, are presented in Table 3.

### **Table 3**



The graphical representation of different tensile properties is illustrated in Figure 4(a) and (b). In these graphs, sample 1 denotes composites created without nanoparticles. Sample 2 signifies composites infused with 3% nano  $Al_2O_3$ . Sample 3 characterizes composites infused with 3% nano ZnO, and sample 4 features  $3%$  nano TiO<sub>2</sub>. The results demonstrate that incorporating  $3%$  nano ZnO in the epoxy composite, along with jute fiber and GFRP in a stacking sequence of Jute/Glass/Jute/Glass, yielded the highest tensile strength and modulus values of 56.32 MPa and 3546.35 MPa, respectively. Additionally, across all four samples, the tensile strength is highest for Jute/GFRP/Jute/GFRP stacking sequence, except for sample 2 where the highest tensile strength is observed for Jute/GFRP/GFRP/JUTE sequence with incorporated ZnO.

Conversely, the lowest tensile strength (33.51 MPa) is witnessed for Jute/GFRP/Jute/GFRP sequence in sample 2. In all samples, the specimens from sample 3 with 3% ZnO exhibit superior tensile strength compared to those with 3% nano Al<sub>2</sub>O<sub>3</sub> and 3% nano TiO<sub>2</sub>. Furthermore, the hybrid composites with nano  $Al_2O_3$  consistently display the lowest average tensile strength, slightly lower than composites without nanoparticles. This trend might be attributed not to the nanoparticles interacting with the fiber and matrix but to increasing particle-to-particle interaction due to a higher filler content. Consequently, particle agglomeration leads to inadequate dispersion of the nanofiller within the matrix, potentially compromising the interfacial bonding between the filler and matrix and consequently diminishing the tensile strength of the fabricated laminates [51]. An increase in the



filler content may result in an increase in the micro-spaces between the filler and the matrix. Samples with 3% nano TiO<sub>2</sub> has moderate tensile strength (between 40 MPa to 50 MPa). However, in case of tensile modulus, samples with  $3\%$  nano TiO<sub>2</sub> have higher modulus on average than the other samples and GFRP/Jute/Jute/GFRP has lowest tensile modulus.



**Fig. 4**. Graphical representation (a) Tensile strength and (b) Tensile modulus of biocomposites reinforced with various nanoparticles

### *3.2 Flexural Properties*

The flexural properties of tested specimen of chemically treated hybrid laminates are tabulated in Table 4. The flexural properties such as maximum load, flexural strain and flexural strength of JGJG, JGGJ and GJJG composites are presented in Figure 5(a) and (b). The basic bending theory proposes that under bending load, the top layer is subjected to compression, the bottom layer is subjected to tension, and the middle layer is subjected to shear. This theory also suggests that the prominent loadcarrying members are external layer in sandwich composite, and hence outer layers should be made of high-strength fibers to withstand high load and thus improve the properties of the composite [52].

The addition of GFRP as the external layer increases the strength and stiffness of the composites. It was found that GFRP/Jute/Jute/GFRP with nano TiO<sub>2</sub> in sample 4 has the highest flexural strength and modulus, and in sample 2, the flexural strength and modulus of GFRP/Jute/Jute/GFRP stacking sequence is 57.69 MPa which is higher than the Jute/GFRP/Jute/GFRP and the Jute/GFRP/GFRP/Jute sequence. However, GFRP/Jute/Jute/GFRP with nano ZnO has the lowest flexural strength and modulus and GFRP/Jute/Jute/GFRP without nanofiller has less flexural strength and modulus than Jute/GFRP/Jute/GFRP which may be due to irregular dispersion of nanofiller and weak filler-matrix



interfacial bonding respectively. Samples with nano ZnO (Jute/GFRP/Jute/GFRP and GFRP/Jute/Jute/GFRP) also have significant flexural strengths and modulus. Overall, specimens of sample 4 incorporated with nano  $TiO<sub>2</sub>$  have higher flexural strength and modulus on average than other samples.

### **Table 4**



### *3.3 Hardness*

The hardness of a material is the measurement of its resistance to deformation due to a compressive force caused by a sharp object. The result of the hardness test is depicted in Figure 6. It shows that the hardness is maximum for the stacking sequence GFRP/Jute/Jute/GFRP in sample 3 with 3% nano ZnO. With the addition of nanomaterials, the hardness of the composites was increased except for the stacking sequence Jute/GFRP/Jute/GFRP in sample 2. Among the three types of samples with added nanomaterials, composites with 3% nano  $Al_2O_3$  are lower than the other two types. Literature reveals a similar trend for incorporating nanoparticles in investigating the hardness [51,52].





**Fig. 5.** Graphical representation (a) Flexural strength and (b) Flexural modulus of biocomposites reinforced with various nanoparticles



**Fig. 6.** Rockwell hardness of the fabricated composite specimens

### *3.4 Morphological Analysis*

FE-SEM is a useful tool for morphological analyses of composite materials. The dispersion and compatibility of the fillers with the matrix can be observed using FE-SEM analysis. The morphological analysis was done on the tensile-tested specimens using SIGMA 300 FE-SEM for evaluating the fiber failure and interfacial bonding between the matrix and the fiber. Figure 7(a) shows the FE-SEM images of fracture surface morphology of a sample without nanoparticles at a magnification of 500X.



A significant number of fibers pull-out and voids are observed in the images due to the poor adhesion between the fibers and matrix.



Fig. 7. FE-SEM images of composites (a) without nanomaterial (b) nano Al<sub>2</sub>O<sub>3</sub> (c) with nano ZnO (d) with nano TiO<sup>2</sup>

In Figure 7(b), more fiber breakage and cracks are observed which may be attributed to the better bonding between the fiber and matrix because of  $3\%$  Al<sub>2</sub>O<sub>3</sub> nanoparticles. Figure 7(c) shows the FE-SEM images of a sample with 3% nano ZnO at a magnification of 1000X. Here, very less void and fiber pull-out are seen, which may be due to the uniform dispersion of Zinc oxide nanoparticles in epoxy matrix, and resulted in good interfacial adhesion and enhanced strength of the sample. In Figure 7(d), with 3% nano TiO<sub>2</sub> and at 300X magnification, more fiber breakage is observed than fiber pull out. Higher fiber breaking indicated a significant interaction between the fiber and matrix [53].



# **4. Conclusion**

In this study, we have conducted experimental investigations on some new types of fiber reinforcement composites. The chemically treated Jute-GFRP fiber reinforced epoxy composites were successfully fabricated using the hand lay-up method. The effect of layering sequence and type of nanomaterial used on the mechanical and morphological properties of the Jute-GFRP reinforced hybrid nanocomposite has been studied.

It is observed that the layering pattern significantly affects the tensile, flexural properties of the composite. The Jute/GFRP/Jute/GFRP sample is more suitable for applications encountering tensile and flexure loads. The tensile strength is maximum for the stacking sequence Jute/GFRP/Jute/GFRP in sample 3 with 3% nano ZnO, and after adding nanomaterials, the tensile properties have enhanced except for nano  $Al_2O_3$  in sample 2, which may be due to poor dispersion of the filler and matrix resulting in a weaker strength. The flexural strength is maximum for GFRP/Jute/Jute/GFRP with nano TiO<sup>2</sup> in sample 4, which is 137. 07 MPa. Adding GFRP as the external layer may act as a factor for the higher-strength composites. The SEM test shows that samples without nanomaterials have more fiber pull-out and voids than those with nanomaterials. In samples with 3% nano ZnO composites, very less void and fiber pull out are observed. This may be due to the uniform dispersion of Zinc oxide nanoparticles in epoxy matrix, resulting in good interfacial adhesion and enhanced strength of the sample. Better bonding between the fiber and matrix because of 3% Al<sub>2</sub>O<sub>3</sub> nanoparticles added is seen in sample 2. In the SEM image with 3% nano  $TiO<sub>2</sub>$  more fiber breakage is observed than fiber pull out which is due to increased interaction between the fiber and matrix.

Overall, a comparison between the properties of all the laminates revealed that the hybrid laminate with 3% nano ZnO balances the properties and cost well. Jute fibers are abundantly available from agricultural resources; they are cheaper than conventional synthetic fibers. Moreover, they have high mechanical properties, as discussed above; hence, the hybrid of these natural fibers can be used for various applications. It is suggested that the chemically treated Jute-GFRP hybrid nanocomposites can be used instead of synthetic fibers in polymer composites.

### **Acknowledgment**

This research was funded by a grant from University Grants Commission of Bangladesh and approved by Directorate of Research & Extension, CUET.

### **References**

- [1] M.M. Billah, M.S. Rabbi and A. Hasan, "A Review on Developments in Manufacturing Process and Mechanical Properties of Natural Fiber Composites," Journal of Engineering Advancements 2, no. 1 (2021): 13-23. <https://doi.org/10.38032/jea.2021.01.003>
- [2] M.S. Rabbi, T. Islam and G.S. Islam, "Injection-Molded Natural Fiber-Reinforced Polymer Composites–A Review," International Journal of Mechanical and Materials Engineering 16 (2021): 1-21. <https://doi.org/10.1186/s40712-021-00139-1>
- [3] M.E.A. Razzaq, S.E. Moma and M.S. Rabbi, "Mechanical Properties of Biofiber/Glass Reinforced Hybrid Composites Produced by Hand Lay-Up Method: A Review," Materials Engineering Research 3, no. 1 (2021): 144-155. <https://doi.org/10.25082/MER.2021.01.003>
- [4] M.S. Huda, L.T. DrzaL, D. Ray, A.K. Mohanty and M. Mishra, "Natural-fiber Composites in the Automotive Sector," in *Properties and Performance of Natural-Fibre Composites*, ed. K.L. Pickering (Elsevier, 2008): 221-268. <https://doi.org/10.1533/9781845694593.2.221>
- [5] M.M. Billah, M.S Rabbi and A. Hasan, "Injection Molded Discontinuous and Continuous Rattan Fiber Reinforced Polypropylene Composite: Development, Experimental and Analytical Investigations," Results in Materials 13 (2022): 100261. <https://doi.org/10.1016/j.rinma.2022.100261>

30



- [6] A. Hasan, M.S. Rabbi and M.M. Billah, "Making the Lignocellulosic Fibers Chemically Compatible for Composite: A Comprehensive Review," Cleaner Materials 4 (2022): 100078. <https://doi.org/10.1016/j.clema.2022.100078>
- [7] M.M.H. Parvez, S.M.N. Rupom, M.M. Adil, T. Tasnim, M.S. Rabbi and I. Ahmed, "Investigation of Mechanical Properties of Rattan and Bamboo Fiber Reinforced Vinyl Ester Composite Material for Automotive Application," Results in Materials 19 (2023): 100437. <https://doi.org/10.1016/j.rinma.2023.100437>
- [8] A.B.A. Hariharan and H.A. Khalil, "Lignocellulose-based Hybrid Bilayer Laminate Composite: Part I-Studies on Tensile and Impact Behavior of Oil Palm Fiber-Glass Fiber-Reinforced Epoxy Resin," Journal of Composite Materials 39, no. 8 (2005): 663-684.

<https://doi.org/10.1177/0021998305047267>

[9] L.A. Pothan, P. Potschke, R. Habler and S. Thomas, "The Static and Dynamic Mechanical Properties of Banana and Glass Fiber Woven Fabric-Reinforced Polyester Composite," Journal of Composite Materials 39, no. 11 (2005): 1007- 1025.

<https://doi.org/10.1177/0021998305048737>

- [10] M.V. Ramana and S. Ramprasad, "Experimental Investigation on Jute/Carbon Fibre Reinforced Epoxy Based Hybrid Composites," Materials Today: Proceedings 4, no. 8 (2017): 8654-8664. <https://doi.org/10.1016/j.matpr.2017.07.214>
- [11] M.M. Shokrieh, A.R. Kefayati and M. Chitsazzadeh, "Fabrication and mechanical properties of clay/epoxy nanocomposite and its polymer concrete," Materials & Design 40 (2012): 443-452. <https://doi.org/10.1016/j.matdes.2012.03.008>
- [12] C.I.W. Calcagno, C.M. Mariani, S.R. Teixeira and R.S. Mauler, "The Role of the MMT on the Morphology and Mechanical Properties of the PP/PET Blends," Composites Science and Technology 68, no. 10-11 (2008): 2193-2200. <https://doi.org/10.1016/j.compscitech.2008.03.012>
- [13] H.Y. Kordkheili, M. Farsi and Z. Rezazadeh, "Physical, Mechanical and Morphological Properties of Polymer Composites Manufactured from Carbon Nanotubes and Wood Flour," Composites Part B: Engineering 44, no. 1 (2013): 750-755.

<https://doi.org/10.1016/j.compositesb.2012.04.023>

- [14] A. Amjad, H. Awais, A. Anjang Ab Rahman and M.S.Z. Abidin, "Effect of Nanofillers on Mechanical and Water Absorption Properties of Alkaline Treated Flax/PLA Fibre Reinforced Epoxy Hybrid Nanocomposites," Advanced Composite Materials 31, no. 4 (2022): 351-369.
- <https://doi.org/10.1080/09243046.2021.1993563> [15] T. Hassan, A. Salam, A. Khan, S.U. Khan, H. Khanzada, M. Wasim, M.Q. Khan and I.S. Kim, "Functional nanocomposites and their potential applications: A review," Journal of Polymer Research 28, no. 2 (2021): 36.
	- <https://doi.org/10.1007/s10965-021-02408-1> [16] N. Saba, M. Jawaid, O.Y. Alothman and M.T. Paridah, "A Review on Dynamic Mechanical Properties of Natural Fibre Reinforced Polymer Composites," Construction and Building Materials 106 (2016): 149-159. <https://doi.org/10.1016/j.conbuildmat.2015.12.075>
	- [17] N. Saba, M. Jawaid, O.Y. Alothman, M.T. Paridah and A. Hassan, "Recent Advances in Epoxy Resin, Natural Fiber-Reinforced Epoxy Composites and Their Applications," Journal of Reinforced Plastics and Composites 35, no. 6 (2016): 447-470.

<https://doi.org/10.1177/0731684415618459>

- [18] P. Ramu, C.J. Kumar and K. Palanikumar, "Mechanical Characteristics and Terminological Behavior Study on Natural Fiber Nano Reinforced Polymer Composite–A Review," Materials Today: Proceedings 16 (2019): 1287-1296. <https://doi.org/10.1016/j.matpr.2019.05.226>
- [19] A.J. Crosby and J.Y. Lee, "Polymer Nanocomposites: the "Nano" Effect on Mechanical Properties," Polymer Reviews 47, no. 2 (2007): 217-229. <https://doi.org/10.1080/15583720701271278>
- [20] S.M.R. Billah, "Composites and Nanocomposites," in *Functional Polymers*, ed. M.A.J. Mazumder, H. Sheardown, A. Al-Ahmed (Switzerland: Springer Nature, 2019): 1-67. [https://doi.org/10.1007/978-3-319-92067-2\\_15-1](https://doi.org/10.1007/978-3-319-92067-2_15-1)
- [21] M. Roy, "Nanocomposite Films for Wear Resistance Applications," in *Surface Engineering for Enhanced Performance against Wear*, ed. M. Roy, (Vienna: Springer, 2013): 45-78. [https://doi.org/10.1007/978-3-7091-0101-8\\_2](https://doi.org/10.1007/978-3-7091-0101-8_2)
- [22] N. Saba, P. Md Tahir and M. Jawaid, "A Review on Potentiality of Nano Filler/Natural Fiber Filled Polymer Hybrid Composites," Polymers 6, no. 8 (2014): 2247-2273. <https://doi.org/10.3390/polym6082247>



- [23] N. Saba, M. Jawaid and M. Asim, "Recent Advances in Nanoclay/Natural Fibers Hybrid Composites," in *Nanoclay Reinforced Polymer Composites*, ed. M. Jawaid, A.E.K. Qaiss and R. Bouhfid (Singapore: Springer, 2016): 1-28. [https://doi.org/10.1007/978-981-10-0950-1\\_1](https://doi.org/10.1007/978-981-10-0950-1_1)
- [24] S.B. Hosseini, "Natural Fiber Polymer Nanocomposites," in *Fiber-Reinforced Nanocomposites: Fundamentals and Applications,* ed. B. Han, S. Sharma, L. Li, K.S. Bhat and T.A. Nguyen (Elsevier, 2020): 279-299. <https://doi.org/10.1016/B978-0-12-819904-6.00013-X>
- [25] P.C. LeBaron, Z. Wang and T.J. Pinnavaia, "Polymer-layered Silicate Nanocomposites: An Overview," Applied Clay Science 15, no. 1-2 (1999): 11-29. [https://doi.org/10.1016/S0169-1317\(99\)00017-4](https://doi.org/10.1016/S0169-1317(99)00017-4)
- [26] V. Prasad, M.A. Joseph and K. Sekar, "Investigation of Mechanical, Thermal and Water Absorption Properties of Flax Fibre Reinforced Epoxy Composite with Nano TiO2 Addition," Composites Part A: Applied Science and Manufacturing 115 (2018): 360-370. <https://doi.org/10.1016/j.compositesa.2018.09.031>
- [27] A. Khan, A.M. Asiri, M. Jawaid and N. Saba, "Effect of Cellulose Nano Fibers and Nano Clays on the Mechanical, Morphological, Thermal and Dynamic Mechanical Performance of Kenaf/Epoxy Composites," Carbohydrate Polymers 239 (2020): 116248. <https://doi.org/10.1016/j.carbpol.2020.116248>
- [28] N. Saba, F. Mohammad, M. Pervaiz, M. Jawaid, O.Y. Alothman and M. Sain, "Mechanical, Morphological and Structural Properties of Cellulose Nanofibers Reinforced Epoxy Composites," International Journal of Biological Macromolecules 97 (2017): 190-200. <https://doi.org/10.1016/j.ijbiomac.2017.01.029>
- [29] A. Mandal and D. Chakrabarty, "Studies on the Mechanical, Thermal, Morphological and Barrier Properties of Nanocomposites Based on Poly (Vinyl Alcohol) and Nanocellulose from Sugarcane Bagasse," Journal of Industrial and Engineering Chemistry 20, no. 2 (2014): 462-473. <https://doi.org/10.1016/j.jiec.2013.05.003>
- [30] E. Rosamah, A. Khalil HPS, S.W. Yap, C.K. Saurabh, P.M. Tahir, R. Dungani and A.F. Owolabi, "The Role of Bamboo Nanoparticles in Kenaf Fiber Reinforced Unsaturated Polyester Composites," Journal of Renewable Materials 6, no. 1 (2018): 75-86.

<https://doi.org/10.7569/JRM.2017.634152>

- [31] K.D. Kumar, "Enhancement of Flexural Strength of Glass-Nylon Fibre Hybrid Composites using Alumina Nano Filler," Materials Research Innovations 25, no. 6 (2021): 338-341. <https://doi.org/10.1080/14328917.2020.1816368>
- [32] A. Mohanty and V.K. Srivastava, "Effect of Alumina Nanoparticles on the Enhancement of Impact and Flexural Properties of the Short Glass/Carbon Fiber Reinforced Epoxy Based Composites," Fibers and Polymers 16 (2015): 188-195.

<https://doi.org/10.1007/s12221-015-0188-5>

[33] T. Özdemir, A. Güngör, I.K. Akbay, H. Uzun and Y. Babucçuoglu, "Nano Lead Oxide and Epdm Composite for Development of Polymer Based Radiation Shielding Material: Gamma Irradiation and Attenuation Tests," Radiation Physics and Chemistry 144 (2018): 248-255.

<https://doi.org/10.1016/j.radphyschem.2017.08.021>

- [34] P. Madhu, K.N. Bharath, M.R. Sanjay, G.R. Arpitha and D. Saravanabavan, "Effect of Nano Fillers on Glass/Silk Fibers Based Reinforced Polymer Composites," Materials Today: Proceedings 46 (2021): 9032-9035. <https://doi.org/10.1016/j.matpr.2021.05.383>
- [35] A. Shukla, V. Parameswaran, Y. Du and V. Évora, 'Dynamic Crack Initiation and Propagation in Nanocomposite Materials," Reviews on Advanced Materials Science 13, no. 1 (2006): 47-58.
- [36] J. Abenojar, J. Tutor, Y. Ballesteros, J.C. Del Real and M.A. Martínez, "Erosion-wear, Mechanical and Thermal Properties of Silica Filled Epoxy Nanocomposites," Composites Part B: Engineering 120 (2017): 42-53. <https://doi.org/10.1016/j.compositesb.2017.03.047>
- [37] P.T.R. Swain and S. Biswas, "Influence of Fiber Surface Treatments on Physico-Mechanical Behaviour of Jute/Epoxy Composites Impregnated with Aluminium Oxide Filler," Journal of Composite Materials 51, no. 28 (2017): 3909- 3922.

<https://doi.org/10.1177/0021998317695420>

[38] G. Raghavendra, S. Ojha, S.K. Acharya and S.K. Pal, "Influence of Micro/Nanofiller Alumina on the Mechanical Behavior of Novel Hybrid Epoxy Nanocomposites," High Performance Polymers 27, no. 3 (2015): 342-351. <https://doi.org/10.1177/0954008314550889>



[39] Z. Dumanoğlu, "A Study on the Determination of Some Physical and Physiological Properties of Seeds of Two Different Jute Varieties (Corchorus capsularis L. and Corchorus olitorius L.)," ISPEC Journal of Agricultural Sciences 5, no. 2 (2021): 456-462.

<https://doi.org/10.46291/ISPECJASvol5iss2pp456-462>

- [40] S. Pujari, A. Ramakrishna and M.S. Kumar, "Comparison of Jute and Banana Fiber Composites: A Review," International Journal of Current Engineering and Technology 2, no. 2 (2014): 121-126. <http://dx.doi.org/10.14741/ijcet/spl.2.2014.22>
- [41] S.B.R. Devireddy and S. Biswas, "Physical and Mechanical Behavior of Unidirectional Banana/Jute Fiber Reinforced Epoxy Based Hybrid Composites," Polymer composites 38, no. 7 (2017): 1396-1403. <https://doi.org/10.1002/pc.23706>
- [42] N.P. Cheremisinoff and P.N. Cheremisinoff, Fiberglass Reinforced Plastics: Manufacturing Techniques And Applications (New Jersey: Noyes Publications, 1995), 1-38.
- [43] M. Jawaid, H.A. Khalil, A.A. Bakar and P.N. Khanam, "Chemical Resistance, Void Content and Tensile Properties of Oil Palm/Jute Fibre Reinforced Polymer Hybrid Composites," Materials & Design 32, no. 2 (2011): 1014-1019. <https://doi.org/10.1016/j.matdes.2010.07.033>
- [44] C. Li, S. Qi and D. Zhang, "Thermal Degradation of Environmentally Friendly Phenolic Resin/Al2O3 Hybrid Composite," Journal of Applied Polymer Science 115, no. 6 (2010): 3675-3679. <https://doi.org/10.1002/app.31469>
- [45] K.D. Kumar, "Enhancement of Flexural Strength of Glass-Nylon Fibre Hybrid Composites Using Alumina Nano Filler," Materials Research Innovations 25, no. 6 (2021): 338-341. <https://doi.org/10.1080/14328917.2020.1816368>
- [46] T.G. Smijs and S. Pavel, "Titanium Dioxide and Zinc Oxide Nanoparticles in Sunscreens: Focus on Their Safety and Effectiveness," Nanotechnology, Science and Applications (2011): 95-112. <https://doi.org/10.2147/NSA.S19419>
- [47] W.N. Wang, I.W. Lenggoro, Y. Terashi, T.O. Kim and K. Okuyama, "One-step Synthesis of Titanium Oxide Nanoparticles by Spray Pyrolysis of Organic Precursors," Materials Science and Engineering: B 123, no. 3 (2005): 194-202.

<https://doi.org/10.1016/j.mseb.2005.08.006>

- [48] M.M. Kabir, H. Wang, T. Aravinthan, F. Cardona and K.T. Lau. "Effects of Natural Fibre Surface On Composite Properties: A Review" *1st International Postgraduate Conference on Engineering, Designing and Developing the Built Environment for Sustainable Wellbeing (eddBE 2011)*, University of Southern Queensland, 2011.
- [49] S. Harish, D.P. Michael, A. Bensely, D.M. Lal and A. Rajadurai, "Mechanical Property Evaluation of Natural Fiber Coir Composite," Materials Characterization 60, no. 1 (2009): 44-49. <https://doi.org/10.1016/j.matchar.2008.07.001>
- [50] V. Chaudhary, P.K. Bajpai and S. Maheshwari, "An Investigation on Wear and Dynamic Mechanical Behavior of Jute/Hemp/Flax Reinforced Composites and Its Hybrids for Tribological Applications," Fibers and Polymers 19 (2018): 403-415.

<https://doi.org/10.1007/s12221-018-7759-6>

[51] T. Mishra, P. Mandal, A.K. Rout and D. Sahoo, "A State-of-The-Art Review on Potential Applications of Natural Fiber-Reinforced Polymer Composite Filled with Inorganic Nanoparticle," Composites Part C: Open Access 9 (2022): 100298.

<https://doi.org/10.1016/j.jcomc.2022.100298>

- [52] A. Atmakuri, A. Palevicius, A. Vilkauskas and G. Janusas, "Review of Hybrid Fiber Based Composites with Nano Particles—Material Properties and Applications," Polymers 12, no. 9 (2020): 2088. <https://doi.org/10.3390/polym12092088>
- [53] D. dos Santos Rosa and D.M. Lenz, "Biocomposites: Influence of Matrix Nature and Additives on the Properties and Biodegradation Behaviour," in *Biodegradation: Engineering and Technology*, ed. R. Chamy (Croatia: InTech, 2013): 433-475.