

Research Article

Investigation of Chemical Treatments to Enhance the Mechanical Properties of Natural Fiber Composites

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A sustainable approach to composites is leading to the use of natural fibers rather than synthetic materials, like carbon or glass, for reinforcement. However, the higher moisture absorption of natural fibers impairs the composite's mechanical properties. Therefore, to improve the mechanical properties, some chemical treatments like silane and fluorocarbon can be performed to reduce the moisture absorption of natural fibers. In this study, flax was used as reinforcement, and epoxy was used as a matrix. In the first part of the study, flax reinforcement was treated with different concentrations of silane (20, 40, and 60 g/L) and fluorocarbons (80, 100, and 120 g/L). Moisture regains (MRs), absorbency, and tensile strength were measured at reinforcement levels. According to the results, reinforcements treated with 60 g/L silane (S3) and 120 g/L fluorocarbons (F3) exhibited the lowest MR values of 7.09% and 3.06%, respectively, whereas water absorbency was significantly reduced. The sample treated with 120 g/L fluorocarbons required 300 seconds extra time to absorb the water as compared with the untreated sample, whereas samples S3 and F3 showed an increase in tensile strength by 20.16% and 34.80% when compared with untreated reinforcement flax reinforcement. In the second part of the study, untreated and treated flax reinforcements were combined with an epoxy matrix for composite fabrication. MR and mechanical tests (tensile, flexural, and Charpy impact tests) were performed. Results revealed that treated flax-reinforced composites exhibited lower MR values 0.86% for F3 and 0.42% for S3, respectively. The tensile, flexural, and pendulum impact strengths of silane-treated reinforced composite sample C.S3 were increased by 15.07%, 117%, and 20.01%, respectively, compared with untreated reinforced composite samples. Consequently, both chemical treatments improve composite mechanical performance as well as service life.

1. Introduction

A composite is a specific material, that is, fabricated from two or more different constituents or components, this synergized material has the combined properties of both these components [1]. For the composite materials, there are two main components to make them include the reinforcement and matrix [2, 3]. The purpose of the matrix is to cement the reinforcement in composites. Desired properties and shapes also have a dependency on the matrix. This part of the composite can be from the thermoplastic and thermoset categories [1, 4]. During the manufacturing of composite materials, an interface is produced between the matrix and reinforced fibers, this interface is responsible for bonding in composites. Mechanical properties of manufactured

composites mainly depend on this induced bonding between fibers and matrix [5, 6]. Because of the tremendous characteristics of composite materials, these have a wide range of applications in many sectors of life [5] including aerospace, automotive, construction, medical, sports, defense, electrical, and electronics [7].

To obtain intelligent and distinct properties in composites, a wide range of fibers can be used as reinforcement, these fibers include natural as well as synthetic fibers [8]. As fiber-reinforced composite materials have different classes based on the fibers used, from these classifications the glass-fiber reinforced composites are considered the largest category because glass composites cover a maximum portion of the worldwide Fiber Reinforced Polymer market [9, 10]. As per the current scenario of worldwide sustainability, there is a very

high focus on two important aspects: (1) to reduce the usage of products and materials that originated from petroleum-based materials, and (2) to use such materials and products that are sustainable and friendly with the environment [11, 12].

In the continuity of eco-friendly and sustainable materials, there is an equal focus on eco-friendly composite materials. Such eco-friendly composite materials have examples of natural fiber-reinforced materials and bio-degradable polymer materials [13]. As natural fibers have two main distinct advantages of abundant availability and less cost over bio-degradable polymer materials, natural fibers have more usage suitability in eco-composite materials [14]. For the manufacturing of natural fibers-based composites, so-called "biocomposites," the most commonly used natural fibers are flax, jute, hemp, bamboo, and sisal. In these biocomposites rice husk may have its utilization as a filler component [15].

Natural fiber composites are one of the most important categories of composites, in which natural fibers are used as reinforcement components in different matrices [16]. Natural fiber composites not only have distinct applications in different work fields, but also they have a high potential to be used in the place of glass fiber-reinforced composites as a substitute [17, 18]. The composites made from natural fibers have certain strengths so that they can be used in many structural applications [19], for instance, the jute composites reinforced with polyester have modulus and tensile strength values of 7 GPa and 60 MPa, respectively [11, 20].

Natural fibers have wonderful distinct features over other polymeric materials, one of the most important features is their bio-degradability [21]. Meanwhile, natural fibers have some limiting factors that create hindrances in natural fibers composites manufacturing [22, 23]. Such limiting factors include the propensity to absorb water, fiber degradation by the attack of microorganisms, and exposure to sunlight. These factors eventually cause the composite to have reduced service life and less strength [11, 24].

Because of the moisture absorption of natural fibers, their usage in composites has been limited in many applications [25, 26]. To cope-up different challenges in natural fiber-based composites, fiber treatment is one of the commonly used alternative ways to modify the surface morphology by different techniques like physical, chemical, or mechanical methods [21, 27]. That is why, different researchers studied the impact of different chemical treatments on natural fibers to check their impact on surface morphology, moisture absorption, and interfacial adhesion of treated fibers with matrices [28, 29]. For fiber surface modification, different treatments are carried out to increase the interfacial adhesion with improved mechanical properties [30, 31]. A large number of chemical treatments, including sodium chlorite, silane treatment, mercerization, etherification, acetylation, fluorocarbon treatment, peroxide treatments, enzymatic treatment, plasma treatment, ozone treatments, and grafting have been carried out to enhance the mechanical properties and service life [32, 33].

Okra bast fibers were chemically treated with sodium chlorite. As per the observations, sodium chloride treatment had an impact on the improvement of their tensile properties,

that is, Young's modulus and tensile strength [11, 34]. Cantero et al. [35] conducted an esterification treatment of flax fibers with methacrylate to check its effect on the hydrophilicity and mechanical properties of composites. Results from these treated composites showed that tensile and as well as flexural strengths are improved [36]. Flax fibers were also treated with 20% methyl methacrylate; these treated flax fibers were incorporated into composites by using phenolic resin as a matrix. Treated flax-based composites showed low moisture regain (MR) value as compared with untreated fiber composites [37].

Ali et al. [38] studied the chemical treatments of jute fibers with different types of fluorocarbons and hydrocarbons to check their effect on the MR and mechanical properties of treated jute-reinforced composites. Jute fibers were treated with different concentrations of hydrocarbon, fluorocarbon, and hybrid fluorocarbon. Results showed that treated jute fibers had a considerable decrease in MR. Mechanical properties like tensile and flexural strengths were also measured; the results revealed that treated jute fiber composites had better mechanical properties [39]. Sepe et al. [25] carried out research on the treatment of hemp fibers with silane to check its effect on the flexural and mechanical properties of epoxy composites. During the silane treatment, the fiber surface is coated with the silane coupling agent. Results of mechanical tests exposed that hemp fibers treated with silane coupling agent had better tensile strength compared with the untreated hemp fibers [25, 40]. To modify the properties of flax fibers to increase hydrophobicity and to improve mechanical properties, acetylation treatment was carried out on flax fibers. In that study, flax fibers treated with acetic anhydride, the reaction takes place between the acetic anhydride and hydroxyl groups of cellulosic chains that cause the plasticization of cellulose. Results indicated that flexural and as well as tensile strengths are improved significantly [41].

Kabir et al. [42] carried out silane, acetylation, and alkali treatments on hemp fibers to enhance interfacial bonding, improve mechanical properties, and decrease hydrophilicity. The mechanical strength of composites was superior for treated fibers than the untreated hemp fibers [42]. Coir fibers were also chemically treated with different concentrations of sodium hydroxide in the range of 2–10%. Then, these treated coir fibers were further combined with an epoxy matrix to fabricate natural fiber composites. Results showed that mechanical properties were improved as compared with untreated coir-reinforced epoxy composites [43].

In previous research studies, various chemical treatments have been applied to natural reinforcement to evaluate how they affect surface morphology, moisture absorption, and mechanical properties of natural fiber composites. According to the author's knowledge, no published data exists on the effects of silane and fluorocarbon treatments on flax reinforcement and their respective composite properties. Our research aimed to apply silane coupling agents and fluorocarbons to flax reinforcement and observe the effect of chemical treatments on the hydrophobicity of natural fibers. In addition, we investigate how surface treatments affect the mechanical properties of composites.

2. Materials and Methods

2.1. Materials. In this study, flax-based reinforcement was used for the fabrication of composites. The yarn used in the warp and weft direction of reinforcement has a linear density of 65.56 Tex, whereas the warp and weft densities were 15.35/cm. The areal density of reinforcement was 208 g/m². Before required functional chemical treatments over flax reinforcement, desizing was carried out to remove size material from the raw flax reinforcement surface. Enzyme-based desizing agent Beisol SED procured from CHT was used to desize the flax reinforcement under slightly acidic conditions. Desized flax reinforcement was used for the next functional chemical treatments. To investigate the chemical treatments on flax reinforcement, two types of treatments were decided to be carried out on desized flax reinforcement.

Silane and fluorocarbon treatments were carried out on desized flax reinforcement; these treated flax reinforcements were latterly used as reinforcement materials for the fabrication of natural fiber composites. Dynasylan®Glymo sourced from Evonik supplier and Ruco Guard® AFR6 from Rudolf were utilized for silane and fluorocarbon treatments, respectively. For the fabrication of flax reinforcement composites, epoxy resin was used as a matrix. Epoxy resin EPIKOTE™ 816 sourced from Hexion chemicals was used along with an epoxy hardener Epotec TH® 7301. Epotec TH® 7301 is sourced from Aditya Birla Chemicals and used to boost crosslinking of epoxy resin during the composite fabrication process.

2.2. Methodology

2.2.1. Reinforcement Treatment. To conduct this study, two experimental factors were decided for the treatment of flax reinforcements, that is, concentrations of silane coupling agent and fluorocarbons. For these two experimental factors, three different concentration levels were selected for the treatment of flax reinforcement. Factor levels for silane concentration were 20, 40, and 60 g/L, whereas factor levels for fluorocarbon concentrations were 80, 100, and 120 g/L. As the two types of experimental work were involved in this study, that is, treatment of flax reinforcement and composites fabrication, so two designs of experiments (DOE) were followed. DOE for the treatments of flax reinforcement is given in Table 1, treatment of flax reinforcement with three different concentration levels of both silane and fluorocarbons was applied accordingly. Five replicates were performed for the treatment of flax reinforcement.

In the study, the researchers employed specific experimental methods for the chemical treatments of the flax reinforcement. The desizing process was carried out using the exhaust processing method. Raw flax reinforcement was desized with the enzyme Beisol SED, provided by CHT. To achieve an acidic pH, lab-grade acetic acid was used, and a wetting agent called Sandopam was applied to enhance the fabric's wettability. The desizing process itself took place at a temperature of 70°C for 1 hour. Following desizing, the fabric underwent a hot wash at 90°C for 10 minutes, followed by cold washing and rinsing. The desized

TABLE 1: DOE for the treatments of flax reinforcement.

S. no.	Sample reference	Silane concentration (g/L)	Fluorocarbon concentration (g/L)
1	R0	0	0
2	S1	20	0
3	S2	40	0
4	S3	60	0
5	F1	0	80
6	F2	0	100
7	F3	0	120

fabric was then hydro extracted and dried in a tumbler steam dryer from Triveneta.

For the subsequent treatment of the desized flax reinforcement with the silane coupling agent and fluorocarbons, the researchers utilized the pad-dry-cure application method. The desized flax reinforcement was passed through a trough containing the necessary chemical solution, with concentrations determined based on the DOE parameters. As the fabric passed between rotating padder rollers, it achieved an approximate pick-up of 80–85% of the chemical solution. The padded fabrics were then secured onto pins of a stenter frame and passed through the stenter for drying purposes. Drying was conducted at a temperature of 120°C for 1.5 minutes. Subsequently, the dried fabrics were once again passed through the pin stenter frame, this time at 150°C for 3 minutes, to effectively cure the treated fabrics.

2.2.2. Composites Fabrication. After the fabric's treatment, these treated fabrics were used as reinforcement parts for the fabrication of composites with the usage of epoxy resin as the matrix. DOE for the composite fabrication is given in Table 2 for the second phase of this study. For composite fabrication, fiber volume fraction (V_f) was kept constant at 35%. Five replicates for each composite fabrication were planned and executed. For the composite fabrication of flax reinforcement, epoxy resin was used as a matrix. A hydraulic compression molding machine from the Carver Manufacturing Company was used for composite fabrication. Four plies of flax reinforcement were used for composite fabrication. Epoxy resin and hardener were mixed properly with the stirrer. Epoxy resin was applied on each ply of reinforcement by hand lap method. After applying resin on each ply, the next ply was placed on the previous ply, and the resin was again applied by the same hand lap method. Hand-lapped four plies wrapped in Teflon sheets were placed between the heating plates of the hydraulic compression molding machine. For composites fabrication, 120°C temperature was applied for 30 minutes, and the composite plate was cooled down for the proper curing process of epoxy resin. After cooling down, the composite plate was placed in the multifunctional oven for the post-curing process at 70°C for 40 minutes.

2.3. Characterizations

2.3.1. Reinforcement. As two DOEs were planned to conduct this study, that is, treatment of reinforcement and

fabrication of composites, characterization at both stages was conducted. For the characterization of flax-treated and untreated reinforcements, different types of tests were performed. These tests included MR, water absorbency, and tensile properties of the reinforcement's part.

The standard procedure ASTM D 2495 was followed to measure the MR of reinforcement. A fabric sample was taken, and its normal weight was measured by using a simple weighing scale, the same sample was placed in an oven to dry it completely until its constant weight was achieved. To compute the MR, two essential parameters were noted. These parameters were MW that is, the mass of absorbed water in the specimen, and MD, that is, the mass of a dry specimen. MR was calculated in units of percentage by using the below formula:

$$MR = MW/MD \times 100. \quad (1)$$

To check the absorbency of treated and untreated fabrics, the standard test method AATCC-79 was used. The fabric was a mountain in the embroidery hoop and placed under a burette having a distance of 10 mm between the burette tip and fabric surface. From the burette, a drop of distilled water was dropped on the surface of the fabric, and the stopwatch was started immediately. Time in seconds was noted until the water drop was absorbed completely and lost its reflectivity. For the measurement of tensile strength, the standard test method ASTM D 5035 was used. Universal testing machine "Lloyd tensile tester" of LRX Plus, USA was used. The sample size for the test was 254 mm × 25.4 mm (length × width). The specimen was fastened within the jaws of the testing machine, and 5 N pre-tension force was applied to the specimen. The specimen's tensile strength was measured under applied maximum force.

2.3.2. Composites. To complete this study, the characterization of fabricated composites was also conducted. For the characterization of flax-reinforced composites, different types of tests were performed including MR and mechanical (tensile, flexural, and pendulum impact) tests of composites.

The MR tests were carried out on composites by following ASTM D 5229 standard. Composite samples were cut and polished, and to remove any dirt from the surface of the sample these were cleaned by using methanol, then dried in an oven at 60°C. During the drying step, the weight of the composite sample was measured at several intervals; and drying continued until the sample weight remained constant. After drying, the weight of the sample was noted, and the sample was instantaneously placed into a standard conditioning environment with a temperature of 20 ± 2°C and relative humidity of 65 ± 5%. The weight change was measured with a weighing balance. The specimen's weights were used to calculate the MR of composite samples.

For the measurement of composite tensile strength, the test method ASTM D 3039 was used. ZwickRoell Universal Testing machine (ZwickRoell, Z100) was used. Test specimens for tensile strength had length and width dimensions of 250 and 25 mm, respectively.

The testing method employed for the measurement of composite flexural strength was ASTM D 7264. This test method was a three-point bending test. A universal testing machine (ZwickRoell, Z100) was used to investigate the flexural properties. Test specimens had length and width dimensions of 120 and 13 mm, respectively.

To calculate the pendulum (Charpy) impact strength of composites, ISO 179 test method was applied. A pendulum impact tester (ZwickRoell, HIT50P) was used for Charpy impact testing. The test specimen had length and width dimensions of 100 and 10 mm, respectively.

3. Results and Discussion

3.1. Reinforcement

3.1.1. Moisture Regain. MR results of flax reinforcement treated with different concentrations of silane coupling agent are shown in Figure 1. There is a significant difference between the silane-treated and untreated fabrics. Furthermore, values of MR decreased with an increase in the concentration of silane coupling agent. The hydrophilic nature of natural fibers is one of the main key problems that create a hindrance to better mechanical properties. During chemical treatment, the silane coupling agent reacts with hydroxyl groups present on natural fibers and reduces the number of hydrophilic hydroxyl groups, hence the tendency of water absorbance is decreased. Thus, surface modification of flax fibers led to reducing MR of silane-treated flax reinforcements.

The effect of fluorocarbon treatments on MR is also shown in Figure 1. Results showed that MR values decreased with the application of fluorocarbons on natural fibers. It also showed a decreasing trend in MR with the increase in fluorocarbon concentrations. Fluorocarbons are molecules that have fluorine and carbon atoms in their molecular structure that bind themselves with fabric surfaces in such a way that hydrophilicity character is decreased. Due to such surface modification of flax reinforcement, the water-loving tendency was reduced, hence MR was reduced as compared with untreated natural flax reinforcement. Compared with silane treatment, fluorocarbons reduced MR more since they engaged more hydroxyl groups during their chemical reaction. MR was reduced due to a lower number of hydroxyl groups present. In summary, both the silane and fluorocarbon treatments effectively reduce MR in flax reinforcements. The silane treatment reduces MR by reacting with hydroxyl groups, whereas fluorocarbon treatment decreases MR s by modifying the surface hydrophilicity. The physical interpretations provided elucidate the mechanisms behind the observed results and emphasize the role of surface modifications in reducing the MR of the treated flax reinforcements.

3.1.2. Water Absorbency. The effect of silane treatment on water absorbency is shown in Figure 2. Results showed that there was slower water absorbency for silane-treated flax reinforcements as compared with untreated reinforcements. With the increase of silane coupling agent concentration, water absorbency was reduced gradually. Sample number S3 with 60 g/L silane concentration has the highest hydrophobicity

TABLE 2: DOE for the composite fabrication.

S. no.	Sample reference	Silane (g/L)	Fluorocarbon (g/L)
1	C.R0	0	0
2	C.S1	20	0
3	C.S2	40	0
4	C.S3	60	0
5	C.F1	0	80
6	C.F2	0	100
7	C.F3	0	120

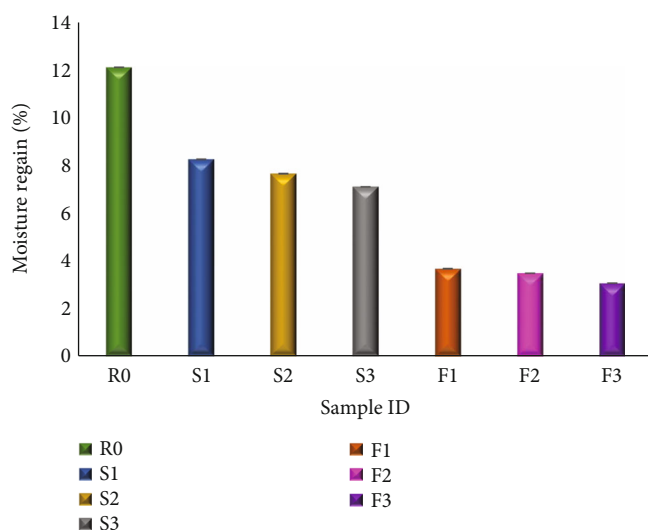


FIGURE 1: MR for silane- and fluorocarbon-treated flax reinforcements.

level as it showed the least water absorbency among other silane-treated samples.

Natural fibers contain micro-level pores in their structure when the silane coupling agent reacts with cellulosic fibers it creates a coating on the fiber surface. Moreover, after silane treatment, stable covalent bonds have formed that increase the hydrophobicity, and hence water absorbency is decreased.

The physical interpretation of these findings lies in the interaction between the silane coupling agent and the cellulosic fibers of the flax. Natural fibers, including flax, possess micro-level pores within their structure. When the silane coupling agent reacts with the cellulosic fibers, it creates a coating on the surface of the fibers. This coating, along with the formation of stable covalent bonds, increases the hydrophobicity of the fiber surface. As a result, water absorbency is decreased because the hydrophobic surface does not readily allow water to penetrate the fibers. The gradual reduction in water absorbency with an increase in the silane concentration further supports the enhanced hydrophobicity achieved through the treatment.

The effect of fluorocarbons on the water absorbency of natural-based flax reinforcements is also shown in Figure 2. Results revealed that water absorption time increased with

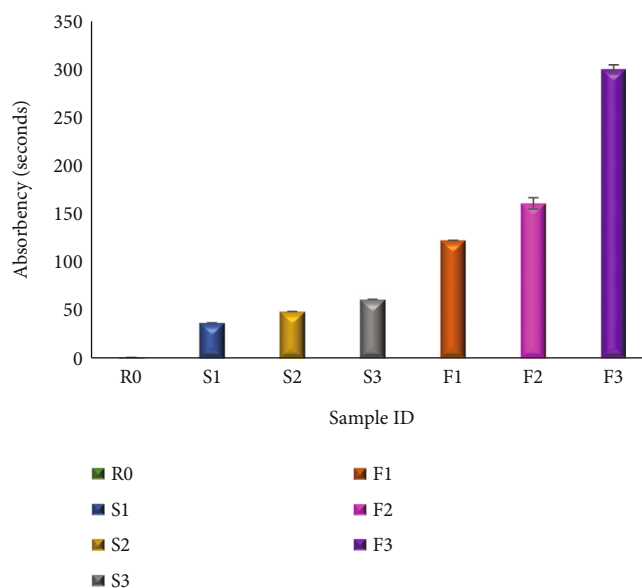


FIGURE 2: Water absorbency for silane- and fluorocarbon-treated flax reinforcements.

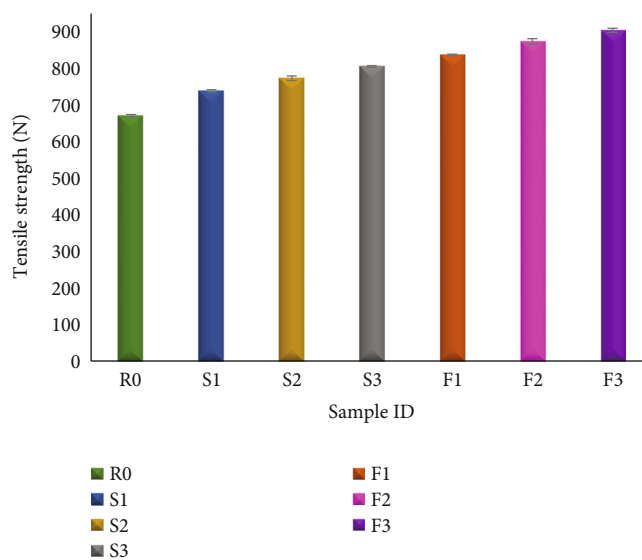


FIGURE 3: Tensile strength for untreated and treated reinforcement fabrics.

the application of fluorocarbons on flax reinforcements as compared with untreated reinforcement. As fluorocarbon concentration was increased, water absorbency was decreased by a considerable amount. F3 sample with a concentration of 120 g/L showed maximum hydrophobicity. Fluorocarbon treatment causes the fiber surface to have lower surface energy that does not allow water to absorb on the surface. Fluorocarbons provide water and oil-repellent functionality on cellulosic-based fibers that make the surface superhydrophobic. Due to lower surface energies and hydrophobic surface water absorbency decreased considerably. The physical interpretation of these findings is attributed to the lower surface energy of the fiber surface caused by the fluorocarbon treatment. Fluorocarbons

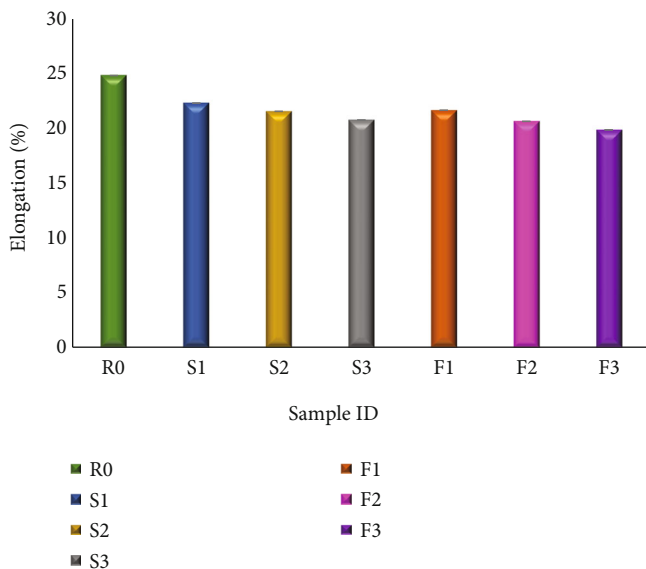


FIGURE 4: Elongation for untreated and treated reinforcement fabrics.

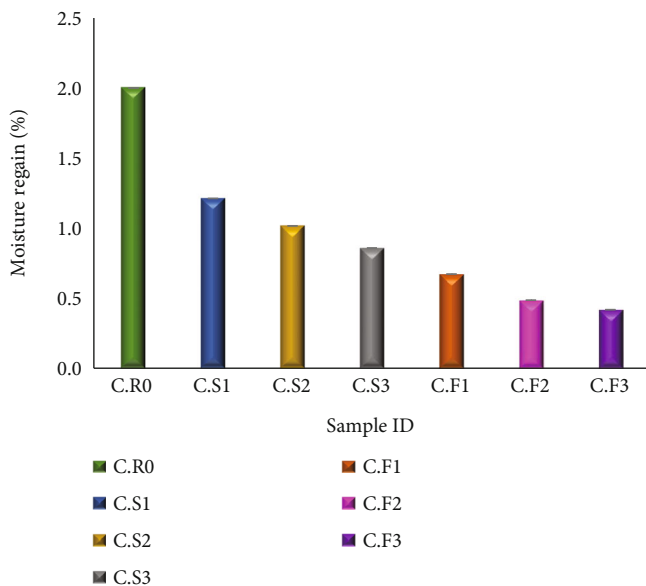


FIGURE 5: MR for silane- and fluorocarbon-treated reinforced composites.

provide water and oil-repellent functionality to cellulosic-based fibers, resulting in a superhydrophobic surface. The lower surface energy and hydrophobic nature of the treated surface significantly reduce water absorbency. Water is unable to readily penetrate the fibers due to the repellent properties of the fluorocarbon-treated surface.

Both the silane and fluorocarbon treatments contribute to the formation of hydrophobic barriers on the flax reinforcement. These barriers lower the surface energy and alter the surface wettability of the fibers. With reduced surface energy, water molecules face hindrance during their interaction with the reinforcement, resulting in lower MR and decreased water absorbency.

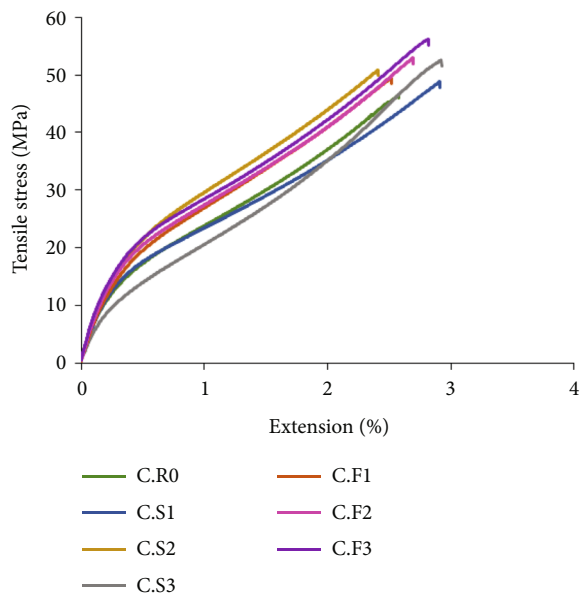


FIGURE 6: Tensile stress versus extension curves of treated reinforced composites.

TABLE 3: Tensile testing results of composites.

Sample code	Tensile strength (MPa)	Tensile modulus (GPa)
C.R0	45.66 ± 3.4	2108.76 ± 95.2
C.S1	48.76 ± 4.1	2394.48 ± 101.5
C.S2	50.89 ± 3.1	3433.07 ± 99.8
C.S3	52.54 ± 4.5	4055.66 ± 110.7
C.F1	49.44 ± 3.9	2399.12 ± 98.7
C.F2	52.89 ± 4.3	3866.00 ± 103.3
C.F3	56.17 ± 3.8	4180.35 ± 108.2

In summary, the silane and fluorocarbon treatments modify the fiber–matrix interface by reducing the hydrophilic nature of the flax reinforcement. Through the formation of stable covalent bonds and the creation of hydrophobic barriers, these treatments improve surface wettability and reduce moisture absorption. The observed improvements in MR and water absorbency can be attributed to these mechanisms influenced by the silane and fluorocarbon treatments on the flax reinforcement.

3.1.3. Tensile Test. The impact of silane treatment on flax reinforcement is shown in Figure 3. Results revealed that with the silane treatment of flax reinforcement, tensile strength was improved as compared with untreated flax reinforcement fabric. Silane treatment created coating on the treated fabric and stable covalent bonds were formed between the fabric surface and the silane agent. These stable covalent bonds were formed upon the reaction of silanol with hydroxyl groups of cellulosic chains, thus forming stable covalent bonds as well as three-dimensional (3D) cross-linked networking that was responsible for higher tensile strength. With the increase of silane concentration, the value

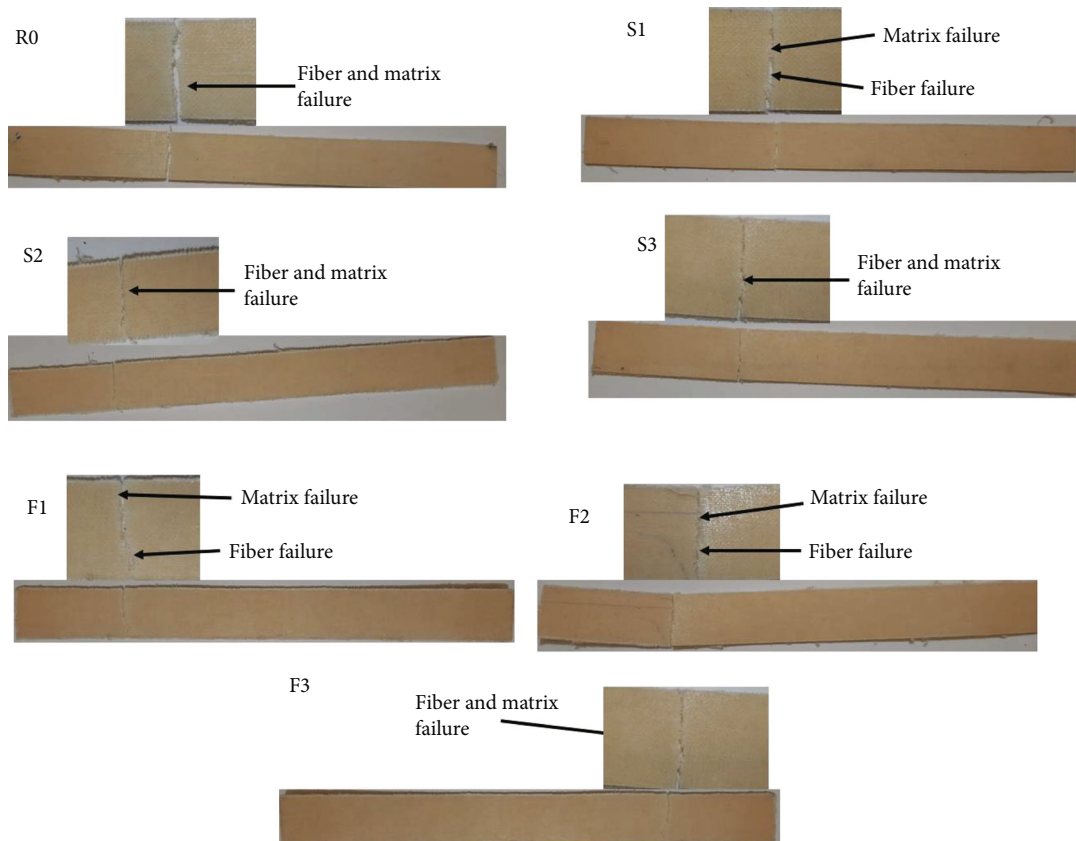


FIGURE 7: Tested samples of tensile test.

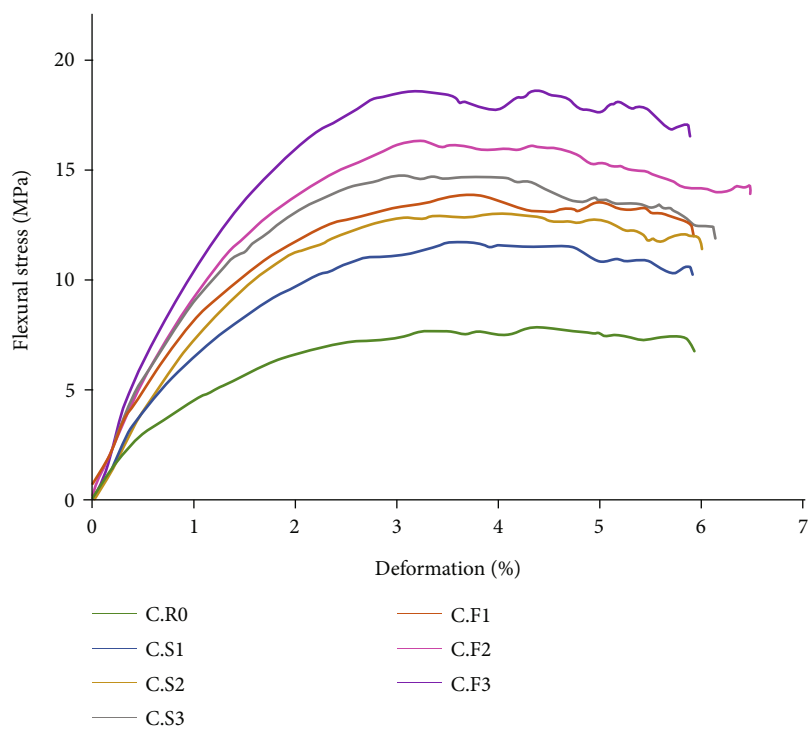


FIGURE 8: Flexural stress versus deformation curves of treated reinforced composites.

TABLE 4: Flexural testing results of composites.

Sample code	Flexural strength (MPa)	Flexural modulus (GPa)
C.R0	7.03 ± 0.41	472.00 ± 34.2
C.S1	10.53 ± 0.49	563.71 ± 37.1
C.S2	12.71 ± 0.53	733.95 ± 42.6
C.S3	15.26 ± 0.38	885.81 ± 41.9
C.F1	12.69 ± 0.42	643.89 ± 35.6
C.F2	15.42 ± 0.33	869.58 ± 39.2
C.F3	17.57 ± 0.31	948.93 ± 42.2

of tensile strength increased also attributed to increased covalent bond networking. Sample S3 had the highest strength among other silane-treated fabrics because it creates the highest number of covalent bonds and 3D networking structure, thus giving improved tensile strength.

The effect of fluorocarbons on the tensile strength of flax reinforcement is also shown in Figure 3. Obtained results discovered that fluorocarbons had increased the tensile strength of treated flax reinforcements when compared with untreated fabric. Improved tensile strength is attributed to the chemical reaction that took place between the reactive groups of fluorocarbon and hydroxyl (-OH) groups of cellulosic fabrics. This chemical reaction upon curing formed a strong 3D cross-linked structure that ultimately increases tensile strength. Sample F3 treated with 120 g/L of fluorocarbons had the highest tensile strength as compared with F1 and F2 samples. This high strength was due to the maximum involvement of cellulosic hydroxyl groups with fluorocarbons, hence maximum covalent bonds were formed giving a strong 3D network structure.

In light of the above discussion, both silane coupling agents and fluorocarbons had an impact on flax reinforcement tensile strength, but fluorocarbons had a high impact as compared with silane treatment. Samples S3 and F3 showed an increase in tensile strength by 20.16% and 34.80% when compared with untreated reinforcement flax reinforcement.

During the fabric tensile strength testing, the elongation percentage was also measured for both chemical treatments. The effects of the silane coupling agents and fluorocarbons on flax reinforcement fabrics are shown in Figure 4. Results in both chemical treatments showed that elongation was reduced in treated fabrics when compared with untreated fabrics. As in both chemical treatments, strong covalent bonds and 3D crosslinking networks were formed between chemical reactive groups and hydroxyl groups of cellulosic chains. This networking structure binds the chemicals with each other as well as with the fibers, hence restricting the elongation of fibers.

The above Figures 3 and 4 showed that samples S3 and F3 have minimum elongations among their other respective chemical treatments. Treated flax reinforcement samples S3 and F3 showed elongation of 20.77% and 19.87%, respectively.

3.2. Composites

3.2.1. Moisture Regain. MR values for untreated and silane-treated reinforced composites C.R0, C.S1, C.S2, and C.S3 are shown in Figure 5. Graphical presentation of these results showed a drastic decrease in MR values in all fabricated composites. MR values of silane-treated composites were less as compared with untreated reinforced composites, but this difference was not very significant. When reinforcement was lapped with epoxy resin then after curing molecular chains of epoxy reacted with each other, with reactive groups of silane coupling agent and with hydroxyl groups of cellulosic substrate. As a very small number of sites were available for interaction with moisture, MR values went down drastically. Furthermore, silane treatment caused lower surface energy of reinforcement also responsible for lowering the MR of composites. Due to the lower surface energies, water molecules faced hindrance during their interaction with reinforcement and composites, which is why very lower values of MR were observed. The physical interpretation lies in the interactions that occur during the fabrication process. When the reinforcement is combined with epoxy resin and cured, the molecular chains of the epoxy react with each other, with the reactive groups of the silane coupling agent, and with the hydroxyl groups of the cellulosic substrate. As a result, a smaller number of sites are available for interaction with moisture, leading to a drastic reduction in MR values. The silane treatment also lowers the surface energy of the reinforcement, which further contributes to the reduction in MR. The lower surface energy hinders the interaction of water molecules with the reinforcement and composites, resulting in significantly lower MR values.

The effect of fluorocarbon treatment on MR of fabricated composites is also shown in Figure 5. The same downward trend in MR was observed. Fluorocarbon-treated reinforcement was covered within the epoxy resin. After the curing reaction, reactive sites were not available that are essential for moisture absorption, hence the downstream level of MR was observed. In addition, lower surface energies of fluorocarbons over reinforcement caused hydrophobicity in their fabricated composites, which play a vital role in lower values of MR in composites. From the above results and discussion, we can see that composites fabricated from S3 and F3 reinforcements showed the lowest values of MR among the remaining treated reinforced composites.

3.2.2. Tensile Test. The effect of silane and fluorocarbon treatments on tensile properties of fabricated composites are shown in Figure 6 having tensile stress versus extension curves obtained from composites tensile testing. Figure 6 showed that tensile stress was increased for silane- and fluorocarbon-treated reinforced composites as compared with untreated reinforced composites. Tensile stress was also increased with the increase in the concentration of silane and fluorocarbons. Composite samples C.S3 and C.F3 showed the highest tensile stress values than other fabricated composites. These C.S3 and C.F3 composites were fabricated with reinforcements S3 and F3, respectively, that were treated with

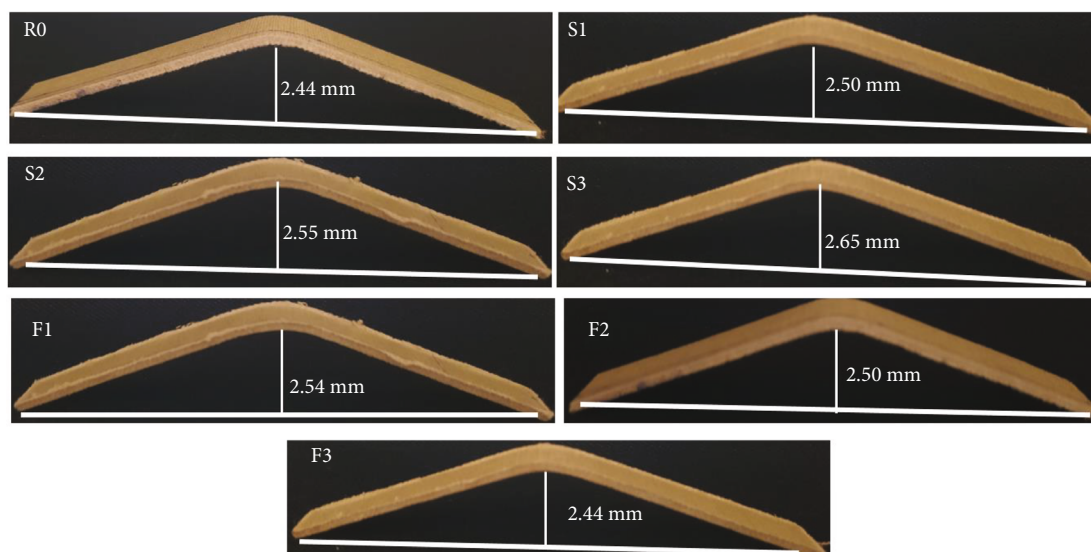


FIGURE 9: Tested samples of the flexural test.

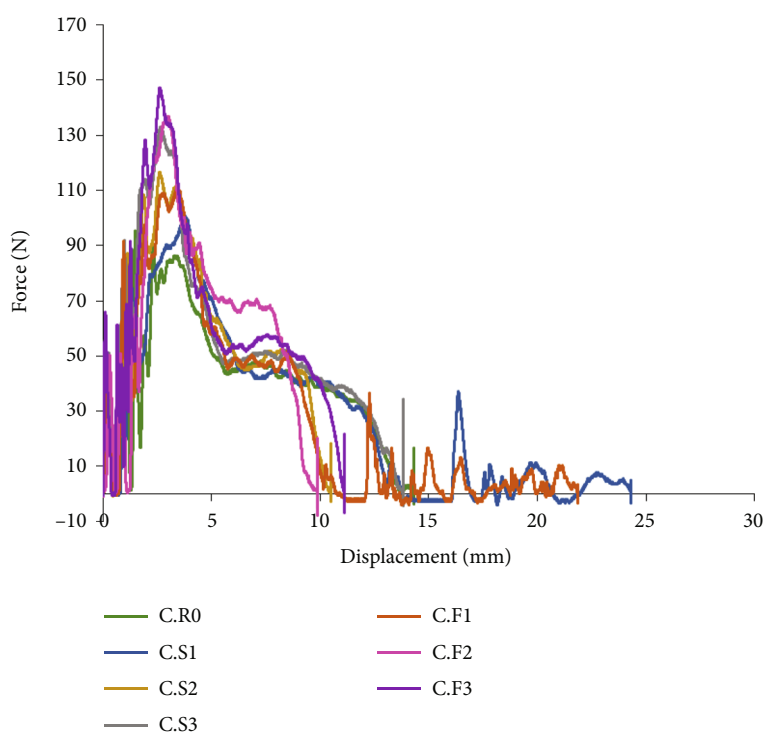


FIGURE 10: Impact force versus displacement curves of treated reinforced composites.

the highest concentrations of silane (60 g/L) and fluorocarbons (120 g/L). The effects of silane and fluorocarbon treatments on the tensile properties of fabricated composites are also shown in Table 3. It shows that the tensile strength of composites increased with the silane-treated reinforcement. It was also clearly shown that the value of tensile strength increased with the increasing concentration level of the silane coupling agent. During the curing reaction in the composite fabrication process, a strong 3D network structure formed

between the matrix and silane-treated reinforcement contributes to the increased tensile strength of fabricated composites. As the amount of silane coupling agent increased, the tensile strength of flax reinforcement also imparted a significant role in the increased tensile strength of composites. Composite sample C.S3 showed the highest tensile strength among other concentration levels, this increase is directly associated with a greater number of reaction sites engaged among silane coupling agent, reinforcement, and epoxy matrix.

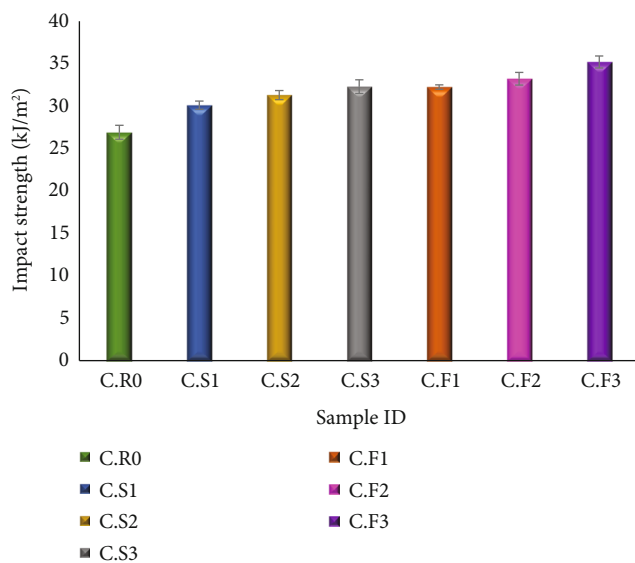


FIGURE 11: Pendulum impact strength for silane-treated reinforced composites.

Results from Table 3 also showed that tensile strength for treated reinforcement composites C.F1, C.F2, and C.F3 was higher as compared with untreated reinforced composite sample C.R0. It is a fact that the strength of composites generally depends on the reinforcement strength and as already discussed the strength of treated flax reinforcement was higher than that of untreated reinforcement. When treated reinforcements F1, F2, and F3 were lapped with epoxy resin, the reactive sites of fluorocarbons interacted with the epoxy having hydroxyl groups due to which a better interface was formed. Furthermore, during the curing step, there was better adhesion between the treated reinforcement and matrix, which imparted its role in improving the tensile strength of composites. Composite sample C.F3 showed the highest tensile strength among other composite samples.

From the above discussion, we saw that composites fabricated from reinforcements S3 and F3 showed the highest values of tensile strength among other treated reinforced composites. Composite samples C.S3 and C.F3 showed an increase in tensile strength by 15.07% and 23.01% when compared with untreated reinforcement flax reinforcement. Figure 7 showed the images of tested samples.

The tensile modulus for fabricated composites is also shown in Table 3. The tensile modulus for fabricated composites was increased with the increase in the concentration of silane and fluorocarbons over treated reinforcements. The highest values of tensile modulus were exhibited by composite samples C.S3 and C.F3 containing flax reinforcements that were treated with the highest concentrations of silane (60 g/L) and fluorocarbon (120 g/L), respectively.

3.2.3. Flexural Test. The effect of silane- and fluorocarbon-treated reinforced composites on flexural bending strength is shown in Figure 8 having flexural stress versus deformation curves obtained from composites flexural

testing. Figure 8 showed that flexural stress was increased for silane- and fluorocarbon-treated reinforced composites as compared with untreated reinforced composites. Flexural stress was also increased with the increase in the concentration of silane and fluorocarbons. Composite samples C.S3 and C.F3 showed the highest flexural stress values compared with other fabricated composites. These C.S3 and C.F3 composites were fabricated with reinforcements S3 and F3, respectively, that were treated with the highest concentrations of silane (60 g/L) and fluorocarbons (120 g/L).

The effect of silane- and fluorocarbon-treated reinforced composites on flexural bending strength is also shown in Table 4. Results from Table 4 showed that the flexural strength of the composite was increased with the increase of the silane concentration level of flax reinforcements. Composite samples C.S1, C.S2, and C.S3 exhibited flexural strength values of 10.53, 12.71, and 15.26 MPa, respectively. As in C.S3 composite sample, reinforcement was treated with the highest level of silane coupling agent, so properties achieved in reinforcement S3 played a very important role in the improved flexural strength of its fabricated composite. When the load was applied to the testing specimen during a flexural bending test, two types of forces act on the specimen, that is, compressive forces at the top of the sample and tensile forces below the sample. As discussed earlier, composite tensile strength was improved with treated reinforcements, so the flexural strength of fabricated composites was also improved. Figure 9 showed the tested images of the flexural test.

Results from Table 4 also showed that composite samples C.F1, C.F2, and C.F3 exhibited flexural strength values of 12.69, 15.42, and 17.57 MPa, respectively. Results elaborated that the value of flexural strength was increased with the increase in the concentration level of fluorocarbons in reinforcement treatment. In composite sample C.F3, reinforcement was treated with the highest level of fluorocarbon concentration, that is, 120 g/L, hence its improved tensile strength played an important role in the improved flexural bending strength of fabricated composites.

The flexural modulus for fabricated composites is shown in Table 4. The flexural modulus for fabricated composites was increased with the increase in the concentration of silane and fluorocarbons during reinforcement treatment. The highest values of flexural modulus were demonstrated by the highest values of silane- and fluorocarbon-treated reinforced composites, that is, C.S3 and C.F3.

3.2.4. Pendulum (Charpy) Impact Test. Figure 10 showed the impact force versus displacement curves of respective composite samples. The effect of treated reinforced composites on the pendulum (Charpy) impact properties of composites was observed during this test. The impact force of the composite was increased with an increase in the concentration of silane and fluorocarbons. It is also shown that the highest concentration of silane (60 g/L) and fluorocarbons (120 g/L) gave the highest impact force in composites. This increase in impact force showed that more energy was absorbed by composite samples showing increased pendulum impact strength.

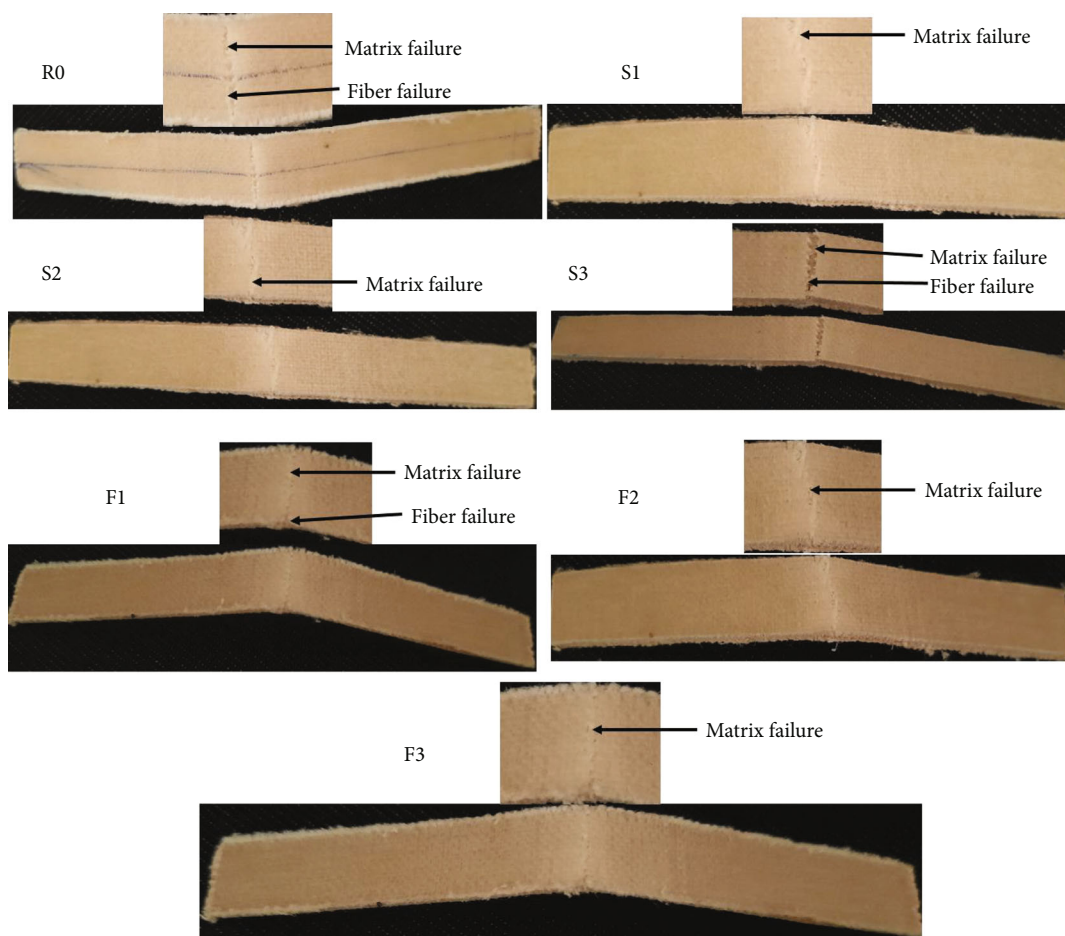


FIGURE 12: Tested samples of Charpy pendulum impact test.

The effect of silane- and fluorocarbon-treated flax-reinforced epoxy composites on its pendulum impact strength is also shown in Figure 11. Results revealed that silane treatment of reinforcement had a positive influence on pendulum impact strength. The pendulum impact strength was increased with the increase of the silane concentration level. During the curing step of the composite fabrication process, epoxy resin cross-linked with the silane groups present on the treated surface, thus giving a more stable structure. Furthermore, silane-treated reinforcement itself had improved strength that also imparted its role to increase overall pendulum impact strength. During the chemical reaction, strong covalent bonds were formed between the reinforcement and epoxy resin that led towards a more stabilized 3D structure, hence giving a positive influence on pendulum impact strength. As already discussed in the reinforcement testing section, MR was also reduced, which also played an important role in improving composite strength.

Results from Figure 11 also showed that fluorocarbon treatment had a positive influence on composite pendulum (Charpy) impact strength. Strength was increased with the increase of the fluorocarbon concentration level over the reinforcement. This positive impact on Charpy strength was also due to the strong 3D bonding network of fluorocarbons with

epoxy resins. Strong covalent bonds were also equally responsible for improved pendulum impact strength.

The low MR value of fluorocarbon-treated flax reinforcement also contributed to its advanced strength, as described in the silane effect segment. This increase in strength states that fluorocarbons had an encouraging impact on the composite's strength and ultimately its service life. Among all three levels of silane and fluorocarbons, composites fabricated with S3 and F3 reinforcement samples showed the highest level of pendulum strength, whereas composite samples C.S3 and C.F3 showed an increase in pendulum strength by 20.01% and 30.79% when compared with untreated reinforcement flax reinforcement. Figure 12 showed the tested images of the Charpy (pendulum) impact test.

4. Conclusions

In this study, flax fabric was utilized as reinforcement for composite fabrication. Silane and fluorocarbon treatments were applied to the flax reinforcement at different concentration levels. The results demonstrate that the MR of the treated reinforcements is significantly reduced compared with the untreated reinforcement. Specifically, reinforcements treated with 60 g/L silane and 120 g/L fluorocarbons

exhibited the lowest MR values of 7.09% and 3.06%, respectively. The water absorbency test revealed that treated reinforcements displayed lower water absorbency than untreated samples, and higher concentration levels of silanes and fluorocarbons correlated with decreased water absorbency. Comparing the two treatments, fluorocarbon-treated samples exhibited lower water absorbency than those treated with silane. Furthermore, both silane and fluorocarbon treatments resulted in improved tensile strength of the reinforcements. Notably, the highest concentration values of silane and fluorocarbons (S3 and F3) yielded the highest tensile strength values in the reinforcements, with fluorocarbon-treated reinforcements demonstrating higher strength values compared with those treated with silane.

Composites fabricated with treated reinforcements also showed lower MR values. MRs for composites fabricated with the highest concentration of silane (60 g/L) and fluorocarbon (120 g/L) was 0.86% and 0.42%, respectively, whereas the MR value for fluorocarbon treatment is less than silane treatment. Compared with untreated reinforced composites, composites with treated reinforcements have improved mechanical properties (tensile strength, flexural strength, and pendulum impact strength). The tensile, flexural, and pendulum impact strengths of silane-treated reinforced composite sample C.S3 were increased by 15.07%, 117%, and 20.01%, respectively, compared with untreated reinforced composite samples. In the same way, the fluorocarbon-treated reinforced composite sample C.F3 showed 23.01%, 149%, and 30.79% higher tensile, flexural, and pendulum impact strengths than the reference sample. As a result, mechanical properties increase with increasing concentration levels of both chemical treatments. Fluorocarbon-treated flax reinforcement and their respective composites showed better properties than those treated with silane.

Furthermore, the improved mechanical properties, including increased tensile strength, flexural strength, and impact resistance offer potential applications in various industries. The treated reinforcements and composites can find utility in various sectors, such as automotive, aerospace, construction, and marine, where high-performance and durable materials are crucial. The enhanced strength properties make the composites suitable for structural applications, such as load-bearing components and reinforcement elements. The future scope of the study involves optimization of treatment parameters, characterization of interfacial properties, assessment of long-term durability, comparative studies with other treatments, exploration of additional properties, and scale-up for industrial application.

Data Availability

Data supporting this research article are available from the corresponding author or first author upon reasonable request.

Conflicts of Interest

The author(s) declare(s) that they have no conflicts of interest.

Authors' Contributions

Farooq Rauf and Tehseen Ullah executed the experimental work and wrote the manuscript. Dr. Muhammad Umair and Dr. Khubab Shaker helped in the Characterization of the composites. Dr. Yasir and Dr. Sheraz reviewed the manuscript and guided the overall project.

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