



Research article

Waste natural fibers for polymer toughening and biodegradability of epoxy-based polymer composite through toughness and thermal analysis

Mohammad Salman Haque^{a,b}, M.A. Islam^{a,*}^a Department of Materials and Metallurgical Engineering, Bangladesh University of Engineering and Technology (BUET), Dhaka, 1000, Bangladesh^b Department of Materials Science and Engineering, Khulna University of Engineering and Technology (KUET), Khulna, 9203, Bangladesh

ARTICLE INFO

Keywords:

Waste natural fiber
Fracture surface
Polymer composite
Toughness
Thermal degradation
Microbial degradation

ABSTRACT

Polymeric materials are being increasingly used to replace many metallic components due to their beneficial properties such as higher strength-to-weight ratio and corrosion resistance. However, the widespread use of polymers poses a risk to the environment as they are not biodegradable. The addition of the waste jute fiber and sawdust fiber as reinforcement to the epoxy resin improved its toughness and induced the biodegradability of the polymer. To examine the effect of the jute fiber and sawdust fiber on biodegradability, the composites were then kept in the drainage system for one year, and the impact energy and fracture morphology of the as-cast and weathered samples were examined using a drop ball impact test and a Charpy impact test. During the weathering period, weight gain was initially observed due to the water absorption by the porous fibers, but after three months, the composites started to lose weight due to the degradation of the fiber by swelling and microbial attacks. Microorganisms in the drainage system used the fiber as their energy source, which resulted in the deterioration of the fiber and the production of CO₂. The production of CO₂ was identified by the FTIR analysis of the weathered composite samples. TGA analysis of the as-cast and weathered samples reveals the reduction of the onset thermal degradation temperature of the weathered composites due to the degradation of the composites. The fiber disintegrated through microbial attack and the fiber swelling caused by the absorption of water by jute fiber and sawdust fiber is identified through SEM imaging. The SEM image also reveals the formation of biofilms and the growth of microorganisms at the fibers. A higher growth rate of the microorganisms was observed in the jute fiber composite than in the sawdust fiber composite, as sawdust contains a high level of lignin that protects it from degradation. The results of this study suggest that both sawdust fiber and jute fiber composites induce biodegradability in the epoxy matrix, but jute fiber was more prominent in this regard. The discovery paves the way for using natural fibers in biodegradable polymer composites, reducing polymeric pollution in the environment.

1. Introduction

The engineering industry is experiencing a growing demand for materials that are lightweight yet offer high strength. Composite

* Corresponding author.

E-mail address: aminulislam@mme.buet.ac.bd (M.A. Islam).

materials are playing a crucial role in meeting this requirement [1]. Polymer-based composites have tremendous potential for use in a wide range of applications, including automobile bodies, energy production and storage, construction, packaging materials, and even space exploration. The automotive industry is increasingly adopting polymer composites for their fuel-efficient vehicles due to their low weight and high performance. Because of their easily adjustable qualities, including their viscoelasticity, chemical and mechanical resistance, and adaptability, polymers are used in a broad variety of applications. Furthermore, a vast array of synthetic techniques can be employed to generate polymeric materials with diverse structures and morphologies, such as films, elastomers, fibers, micro- and nanoparticles, branching, linear, and crosslink. Due to its adaptable architecture and ability to be modified into multifunctional responsive platforms that are sensitive to chemical and physical stimuli, the usage of these polymeric materials has gained interest in the creation of sensors, including radiation measurement systems, actuators, and biosensors [2,3].

Load-bearing components are frequently subjected to impact-type loading and unloading under regular service conditions. However, it should be noted that thermosetting polymers are susceptible to notches, which makes them vulnerable to the detrimental effects of impact-type loading [4,5]. According to the fracture mechanics hypothesis, an alternate approach to mitigate notch sensitivity is to improve the toughness of these polymers by creating composites [6,7]. Reinforcement fiber composites have been increasingly used in engineering applications for many decades, surpassing traditional materials such as concrete and steel. This is due to their high strength-to-weight ratio, remarkable resistance, and lightweight characteristics along the fiber direction. Composite fibers consist of strong fibers, such as carbon, glass, or aramid, and soft matrices, such as epoxy, polyester, vinyl-ester, and others. These materials exhibit remarkable properties such as excellent strength in tension and better absorption of energy [8].

Synthetic fibers possess unique properties that are unmatched by natural fibers. These include good thermal and electrical conductivity, higher stability, and lower moisture absorption, making them ideal for high-end applications such as wind turbines and aerospace. Additionally, they have a longer lifespan. However, these fibers are generated from finite sources like fossil fuels which mostly end up accumulating at landfill sites, posing a risk to the environment [9]. Plastics that have been contaminated can release toxic substances into the soil, which may eventually seep underground and pollute other water sources in the vicinity. This can have severe consequences for the organisms that rely on this water. Landfills are constantly accumulating huge amounts of plastic waste, which can include various types of plastics. Degradable substances are materials that can be easily decomposed or degraded in the natural environment. Biodegradable composites have the potential to revolutionize the upcoming era, as they are composed of biodegradable fibers and polymers that can be altered to meet the product specifications. Natural fibers are biodegradable and can be used in various product development processes, which are environmentally conscious [10,11]. Furthermore, natural fibers have several advantages, including a favorable fiber aspect ratio, low density, and high tensile and flexural moduli. Due to their availability and biodegradability, natural fibers such as wood sawdust and waste jute fibers are preferred over synthetic fillers in countries like Bangladesh [12,13].

In this experiment, composites based on epoxy were created using waste jute fiber and sawdust fibers to determine the effect of the natural fiber on biodegradation into the drainage system through drop ball and Charpy impact test. FTIR, TGA, and SEM analyses were

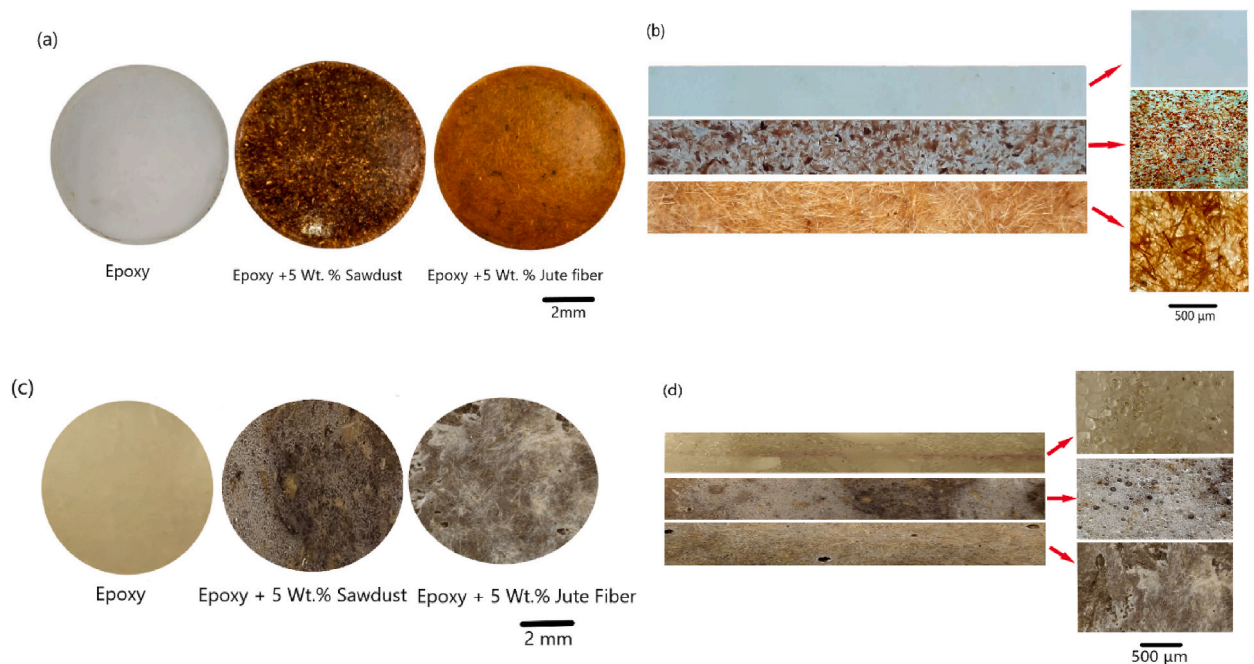


Fig. 1. a) As-cast epoxy, sawdust composite, jute fiber composite for drop ball impact test b) As-cast epoxy, sawdust composite, jute fiber composite for Charpy impact test c) 1-year weathered epoxy, sawdust composite, jute fiber composite for drop ball impact test d) 1-year weathered epoxy, sawdust composite, jute fiber composite for Charpy impact test.

also performed to validate the degradation by the microbial attacks presented in the drainage system. The novel aspect of this work is the assessment of the polymer's biodegradability using a composite of waste natural fiber to reduce the pollution caused by polymers in the drainage system. These waste natural fibers were utilized to lay the foundation for advanced and eco-friendly polymeric materials. The natural polymer addition increases the composites' toughness, making them suitable for usage in automotive body parts. Additionally, when the utilized structure is discarded into the environment, the natural fiber will aid in its degradation.

2. Experimental procedure

2.1. Collection of the reinforcements

The main components used in this investigation were natural fibers, which were utilized to improve the three-dimensional, cross-linked thermoset epoxy polymer's biodegradability. Waste Jute fibers and waste wood sawdust fibers were collected from an agricultural field and a carpentry shop, respectively, and used in this experiment. The jute fibers were then chopped to a length of 2 mm, and the sawdust fibers were screened to obtain fibers of that length.

2.2. Samples manufacturing

The initial quantity of epoxy resin (Bisphenol A-based) was added to a beaker and cured with methyl-tetrahydro phthalic anhydride at normal temperature using a volumetric ratio of 1:10 for hardener and resin [14]. To ensure a homogeneous mixture of hardener and resin, the mixture was stirred. Cast polymer products often contain defects such as blow holes, micro-cracks, and voids, which can negatively affect their mechanical qualities [15,16]. Thus the resin mixture underwent a 10-min vacuum degassing process to remove moisture before being cast into the circular mold. At a minimum 25 samples, each 5 mm thick, were created using the same technique (Fig. 1a).

The sawdust and jute fiber particles were treated with NaOH to remove wax and dust from the fiber, then dried in the sunlight to remove the moisture, and combined with epoxy resin [17,18]. The mixture was stirred to ensure homogeneity before adding a hardener (10% volume of the resin). Dissolved gases were removed before pouring the substance into the mold by vacuum degassing process. Fig. 1a depicts the 5 mm thick as-cast wood sawdust composite and 25 circular test samples for jute fiber produced with the same method. Waste jute and sawdust fibers were used to prepare the samples for Charpy impact samples by ASTM D6110-10 standard shown in Fig. 1b.

2.3. Weathering method

To understand the effect of natural fiber on the biodegradability of epoxy-based composite, the samples for the drop ball test and Charpy impact test were kept in the drainage system for about 1 year. During the period, the necessary data of the submerged samples and drainage water were collected regularly. The water parameter of the drainage water is given in Table 1. The color of the epoxy sample changed to brown, and numerous voids were observed at the jute and sawdust fiber composites suggesting microbial degradation (Fig. 1). The samples in Fig. 1(c) and (d) show significant texture alteration, severe damage, and numerous voids due to the thickness swelling and microbial attacks of the fibers.

Table 1

The average value of the swater parameter of the drainage water.

Immersion Time (Days)	Water Parameter (Average value)								
	Color	Odor	pH	Temperature °C	BOD (mg/L)	COD (mg/L)	DO (mg/L)	TDS (mg/L)	Appearance
0	Black	Present	7.4	34	462	1680	1.8	1345	Not Clear
15	Black	Present	8.0	36	530	1632	2.1	980	Not Clear
30	Black	Present	7.8	41	380	1578	1.5	1780	Not Clear
45	Black	Present	5.9	39	537	1674	2.4	2027	Not Clear
60	Black	Present	6.5	41	480	1890	1.2	1304	Not Clear
75	Black	Present	7.9	38	430	1764	3.1	1426	Not Clear
90	Black	Present	6.2	42	532	1527	2.7	1535	Not Clear
120	Black	Present	5.7	40	375	1696	2.9	1387	Not Clear
150	Black	Present	7.3	38	560	1858	1.6	2145	Not Clear
180	Black	Present	5.6	34	424	1952	2.4	937	Not Clear
210	Yellowish	Present	7.2	32	418	1860	1.5	1159	Not Clear
240	Black	Present	7.8	36	567	1685	3.1	1862	Not Clear
270	Black	Present	6.2	30	396	2021	2.8	1434	Not Clear
300	Yellowish	Present	7.5	18	580	1984	3.4	1256	Not Clear
330	Black	Present	5.9	26	490	1996	1.6	2053	Not Clear
345	Black	Present	7.4	24	372	1578	2.3	1625	Not Clear
360	Black	Present	7.8	30	434	1657	1.7	1793	Not Clear

2.4. Test methods

Polymers, polymer composites, and glassy materials are often subjected to the drop ball impact test to determine their toughness. This investigation used a drop ball impact test setup (Fig. 2a) according to the ASTM F3007-19 Standard to measure the toughness of the polymer and its composites by starting the cracking process at a weak point and spreading it throughout the sample until it shatters [18].

The following formula was used to determine the toughness values:

$$\text{Potential Energy } U = m \times g \times h \text{ ————— (1)}$$

Where the ball's mass is represented by m in kilograms, g is 9.8 m per second, and h is the height in meters at which the ball was dropped.

A 0.11 kg steel drop ball was used in the experiment. The initial drop height was selected by the trial and error process. For the pure epoxy sample, the initial height was selected at 1025 mm where two samples out of the five samples were broken. To break all samples in each group, the second set of five test samples was evaluated at a height greater than 25 mm of the initial drop height and increased the height continuously to find out the height at which all five samples were shattered. At, 1075 mm, all the tested samples were broken and the corresponding energy was 1.16 J (Equation (1)) that value has been considered as impact energy for the as-cast epoxy sample. The details of the drop ball impact energy determination for pure epoxy are presented in Table 2.

The Charpy impact test was also measured according to the ASTM D6110-10 standard [19] and the test setup is shown in Fig. 2b. Three samples were tested for each type and the average value was taken as the impact energy. The Charpy impact energy for the as-cast samples is shown in Table 2.

3. Results and discussion

3.1. Improvement of toughness by addition of natural fiber

The results of the drop ball impact energy and Charpy impact energy of the as-cast samples are shown in Fig. 3. For drop ball impact energy, the height and the corresponding energy that completely broke down the all tested samples is considered as the impact energy. The epoxy samples were the least tough of the three groups, cracking at the minimum drop height (1075 mm) due to the release of kinetic energy from the drop ball breaking the atomic bonds of the polymeric materials. On the other hand, the composite sample reinforced with chopped jute fibers had the highest toughness, indicated by its maximum drop height (1550 mm). For the Charpy impact test, the average energy for the break of the as-cast epoxy sample was found to be 3.73 J, and after the addition of the sawdust and jute fiber the impact energy was improved to 5.14 J and 6.5 J respectively (Fig. 3). Other researchers additionally reported similar kinds of improvements in toughness associated with the addition of natural fibers [20,21].

Fig. 3 illustrates how the addition of wood sawdust and chopped jute fibers as reinforcement enhances the toughness of pure epoxy-based polymer. Reinforcement with waste sawdust and waste-chopped jute fibers improved the impact resistance by around 31% and 44% respectively. When a composite was tested with a drop ball, the jute and sawdust fibers carried out the impact load, slowed down the crack growth, and considerably improved the composite's durability [20,21]. Jute fibers, due to their small diameter, are more suitable than wood sawdust particles for mechanical bonding and load distribution in the composite matrix [22]. The same kind of resistance by the jute fiber and sawdust fiber is also found for the Charpy impact test that's why the impact energy is improved with the fiber addition.

3.2. Effect of weathering in the drainage system

From Fig. 4, the drop ball impact energy of the weathered sample of pure epoxy decreased by 4.5% (1.16J–1.11 J) after one year of weathering in the drainage system. However, the degradation of pure epoxy is negligible compared to the degradation of jute fiber

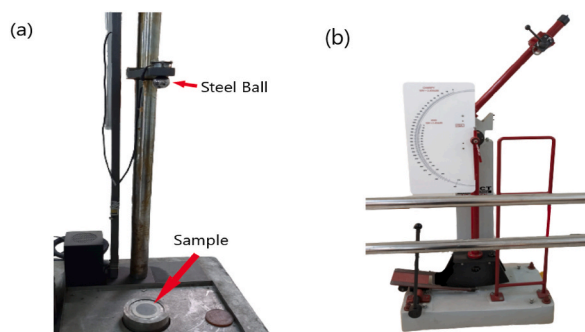


Fig. 2. a) Drop ball impact test setup b) Charpy impact test setup.

Table 2
Determination of drop ball impact energy for pure epoxy samples.

Height (mm)	Number of Samples Tested	Number of Samples Broken	Energy (J)	Impact Energy (J)
1025	5	2	1.10	1.16
1050	5	4	1.13	
1075	5	5	1.16	

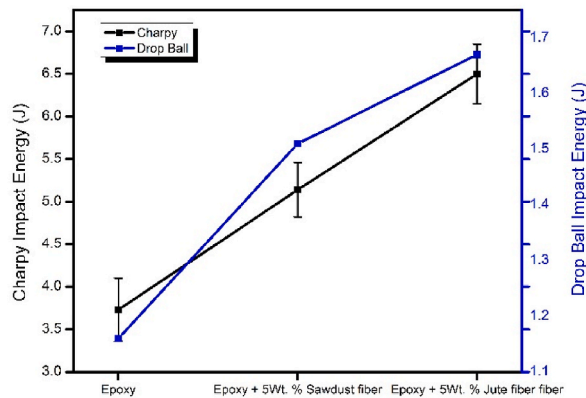


Fig. 3. Impact energy of the as-cast epoxy and composite samples.

composite and saw dust fiber composite (Fig. 4). The drainage system is full of pollutants (Table 1) and microorganisms such as bacteria and fungus causing a 45% fall in the impact energy of the jute fiber composite whereas impact energy for sawdust is decreased by 29%. The natural fiber from the composite serves as a source of energy for the microorganisms, breaking down the bonding between the matrix and the fiber, which significantly reduces the toughness [23].

It can be observed from Fig. 5 that the weathered samples experienced a decrease in the Charpy impact energy. The epoxy composite, which did not undergo microbial degradation, experienced a very slight degradation. On the other hand, the jute fiber and sawdust fiber composites experienced degradation of 67% and 32% respectively. A lower degradation rate is observed at the initial period as a lower reduction of the impact value during the 6-month weathering compared to the 1-year weathering. However, as time passed, microorganisms started to attack the composites at a faster rate and developed a zone to attack the fibrous materials [24,25].

Figs. 4 and 5 reveal that the incorporation of jute fiber has a more pronounced effect on decreasing impact energy than the sawdust composite when exposed to weathered conditions in drain water. Sawdust fiber composite is less susceptible to microbial degradation due to its higher lignin content compared to jute fiber composite [26,27]. When jute fiber is exposed to drain water for an extended period, its cellulose component gets oxidized and degraded, which eventually leads to a decrease in its toughness resistance [28]. On the other hand, sawdust, which has a higher concentration of lignin derived from wood, exhibits heightened resistance against weathering, resulting in enhanced stability and durability in the presence of various environmental factors. Lignin in the cellular matrix of wood acts as a safeguard for carbohydrates against microbial decomposition due to its intrinsic resistance to degradation by

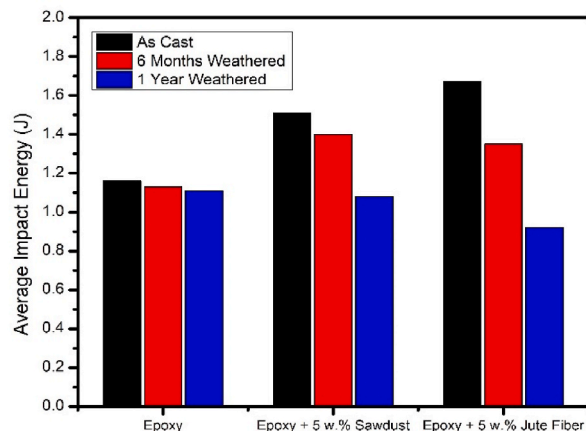


Fig. 4. Comparison of drop ball impact energy of as-cast and weathered samples.

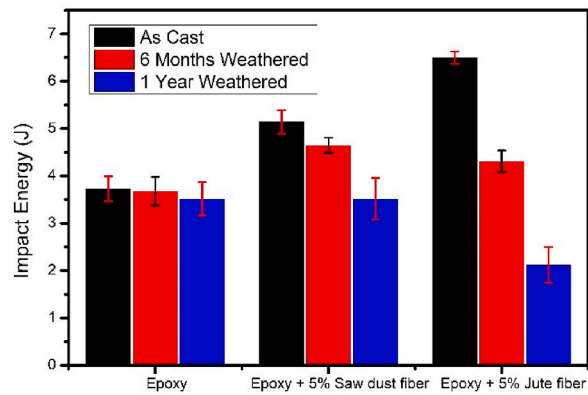


Fig. 5. Comparison of Charpy impact energy of as-cast and weathered samples.

most microorganisms. However, it is essential to note that the durability and robustness of sawdust can deteriorate over time when exposed to weather conditions, especially if it is not adequately protected from moisture [29,30]. Moisture in the area makes it susceptible to microbial degradation, oxidation, and deterioration when exposed to drainage water. This phenomenon can lead to the degradation and weakening of the toughness of the composites that incorporate sawdust over time [31].

3.3. Comparison of impact fracture patterns of the as-cast and weathered samples

Fig. 6 reveals the fracture pattern of the as-cast samples and weathered samples after the performing of the drop ball impact test. The as-cast epoxy sample was found to be susceptible to brittle fracture - when the drop ball hit the samples, they broke into several pieces due to the lack of resistance to failure shown in Fig. 6a. However, the natural fibers (both sawdust and jute) resisted crack formation during the instantaneous loading from the drop ball, making them tougher as seen in Fig. 6b and c.

As time went by (6 months and 1 year of weathering), the resistance of the natural fibers decreased due to the breakdown of the fiber matrix bonding caused by water absorption and microbial attack on the matrix wall. This is visible in the fracture pattern, as more fragmentations are formed, indicating no resistance during the failure shown in Fig. 6(d-i). Furthermore, it was observed that the jute fiber composite underwent more fragmentation compared to the sawdust fiber composite after one year of weathering (Fig. 6h and i). This suggests that the jute fiber composite experienced a higher level of degradation as compared to the sawdust fiber composite [32].

Fig. 7 shows the fracture surfaces of the as-cast and weathered samples after the Charpy impact test. The fracture surface of the as-cast epoxy sample is smooth and brittle with a river pattern shown in Fig. 7a. However, the addition of fiber changed the fracture pattern as the fiber resisted crack propagation (Fig. 7b and c). The fracture pattern of the weathered epoxy sample was quite similar to the as-cast epoxy samples (Fig. 7d and g), but the weathered natural fiber composites showed various types of porosity and holes, particularly in the jute fiber composite (Fig. 7e, f, 7h, and 7i). That's why the jute fibers in the weathered composite samples were not able to resist the load and reduced the impact energy of the composites after weathered in the drainage system.

Fig. 8 displays the TGA curve for both as-cast and weathered samples, which provides thermal degradation information. The onset thermal degradation temperatures for the as-cast epoxy, sawdust fiber, and jute fiber composite are found to be 330 °C, 298 °C, and

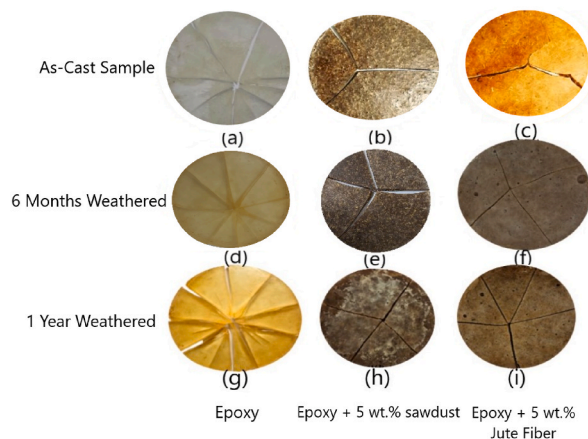


Fig. 6. Fracture pattern of Pure Epoxy Sample (a, d, g), Waste Saw Dust Epoxy Composite (b, e, h), and Waste Jute Fiber Composite (c, f, i) formed by drop ball impact test at 1550 mm.



Fig. 7. Fractured surface of Charpy impact test samples.

303 °C respectively (Fig. 8a, b, and 8c). The addition of fiber caused a decrease in the thermal degradation on-set temperature due to the lower thermal stability of the jute fiber and sawdust fiber [33,34]. However, after one year of weathering, the onset temperature for the epoxy only slightly degraded to 323 °C. In contrast, the onset temperature for the weathered jute fiber and sawdust fiber

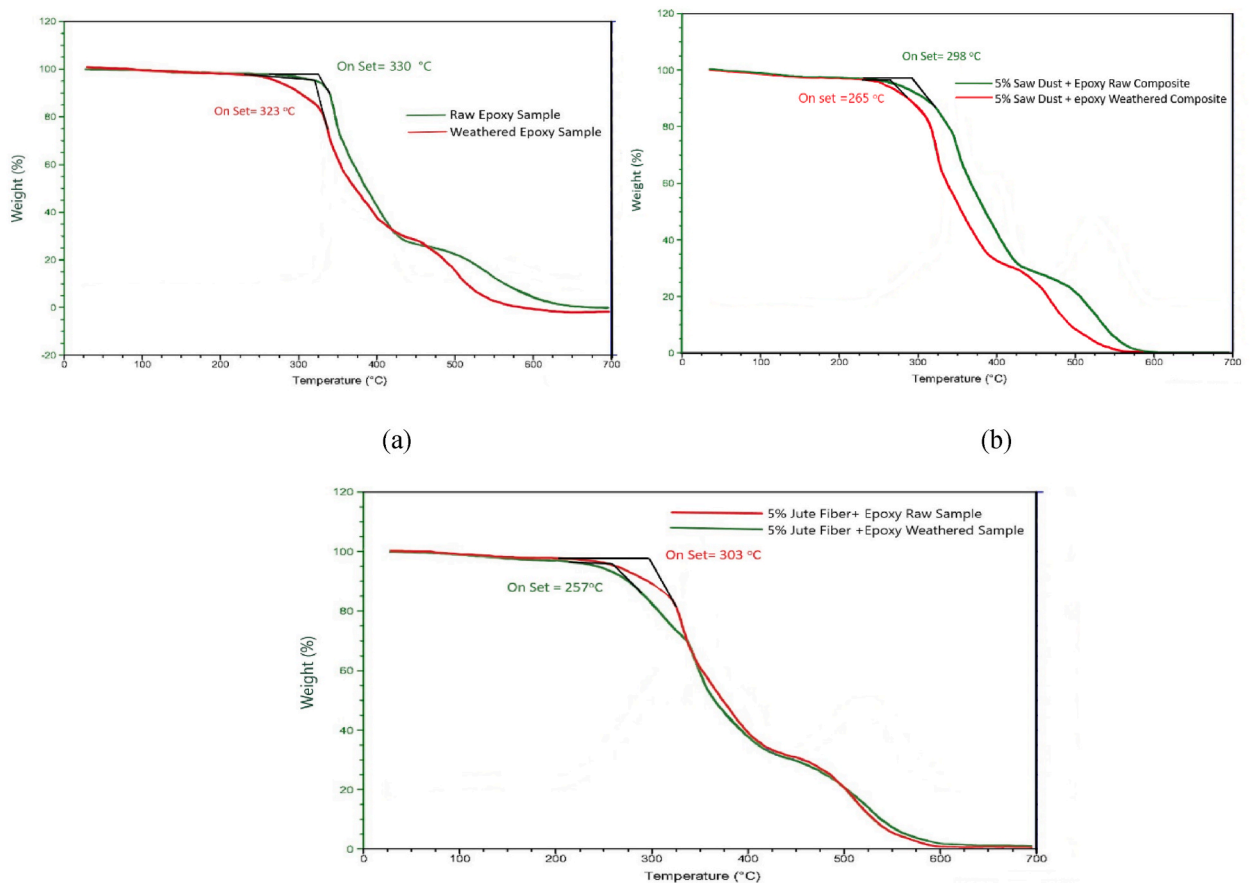


Fig. 8. Thermo-gravimetric analysis of a) As-cast and weathered epoxy samples b) As-cast and weathered sawdust epoxy composites c) As-cast and weathered jute fiber epoxy composite.

composites significantly decreased as the degradation of the fibers occurred by the microorganisms. Notably, the degradation of jute fiber was more significant compared to the sawdust fiber composite as more degradation occurred in the jute fiber composite. Fig. 9 displays a comparison of the onset of thermal degradation temperature of the as-cast and weathered samples.

Microbial organisms, including bacteria and fungi, are known to cause biological oxidation in natural fiber epoxy composites, rendering them susceptible to damage. Microbial enzymes such as lipases and proteases can break down the matrix material, jute fibers, and sawdust fibers present in composites [34,35]. Thus, thermal degradation occurred for the weathered composite samples due to the lack of bonding between the fiber and matrix that is responsible for the stability of the composite materials [36].

3.4. Degradation by thickness swelling

Fig. 10a and b show that both jute fiber and sawdust fiber are porous, making the composites susceptible to water absorption when immersed in the drainage system. This results in a reduction of the bond strength between the fiber and matrix, as well as thickness swelling of the composite, as seen in Fig. 11. Thickness swelling caused by water absorption leads to the disintegration of the fiber and weakens it, decreasing the efficacy of stress transfer at the fiber-matrix interface [37].

Fig. 11 reveals that pure epoxy experienced minor thickness swelling during the initial period due to the presence of air bubbles during casting, which created voids in the sample. However, jute fiber and sawdust fiber absorbed more water and swelled during the same period. However, the thickness swelling stopped as the porous fiber was filled with drain water within two months. Eventually, the fibers disintegrated and weakened the bond with the matrix leading to the degradation of the composites, causing a decrease in their toughness.

The graph in Fig. 12 shows the change in mass over time. In the initial period, the samples gained some weight due to water absorption, with jute fiber being better at absorbing water than sawdust fiber because of its lack of lignin content [27]. However, after three months, the composite samples began to lose weight due to microbial attack in the drainage system. The drainage system is full of microorganisms, and some of them were observed using computer-based light microscopy as shown in Fig. 13.

Microbial enzymes like lipases and proteases can break down the matrix material, jute fibers, and sawdust fibers in composites, resulting in significant degradation of the mechanical and structural properties of lingo-cellulosic materials [38,39]. The consumption of degraded organic components by microorganisms for energy also leads to degradation of the mechanical properties of the composite material when submerged in drain water.

3.5. SEM image of the fracture surface

The scanning electron microscopy (SEM) technique was employed to conduct a microanalysis of the fractured surface of both as-cast and weathered composites. Fig. 14 reveals that the formation of cracks is attributed to the presence of an air pocket, while the propagation of these cracks leads to a brittle fracture that exhibits a distinct river pattern for the as-cast epoxy and weathered epoxy samples [40,41]. The as-cast samples' fractured surfaces displayed a relatively even texture, as illustrated in Fig. 14a. After being exposed to drain water, the samples displayed observable signs of surface roughness, voids, and Degradation (Fig. 14b and c).

The ability of jute and sawdust fibers to carry loads makes them resistant to deformation during the impact test. Figs. 15a and 16a show that the crack caused by the impact force was terminated by the sawdust and jute fibers. The bonding between the fiber and matrix appears to be good for both the as-cast sawdust sample and the jute fiber sample. However, in Fig. 15b and c, the distortion of the fiber due to swelling and microbial attack can be observed. When exposed to drain water, the jute and sawdust fiber-epoxy composites underwent absorption process where water molecules infiltrated the porous fibers, resulting in their subsequent expansion and deterioration of the adhesive properties of epoxy, leading to the detachment of fibers from the matrix [42]. Fig. 15c shows that

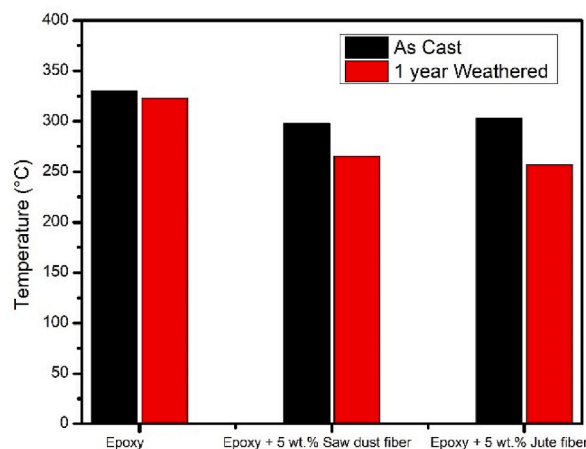


Fig. 9. Comparison of on-set thermal degradation temperature of as-cast and weathered samples.

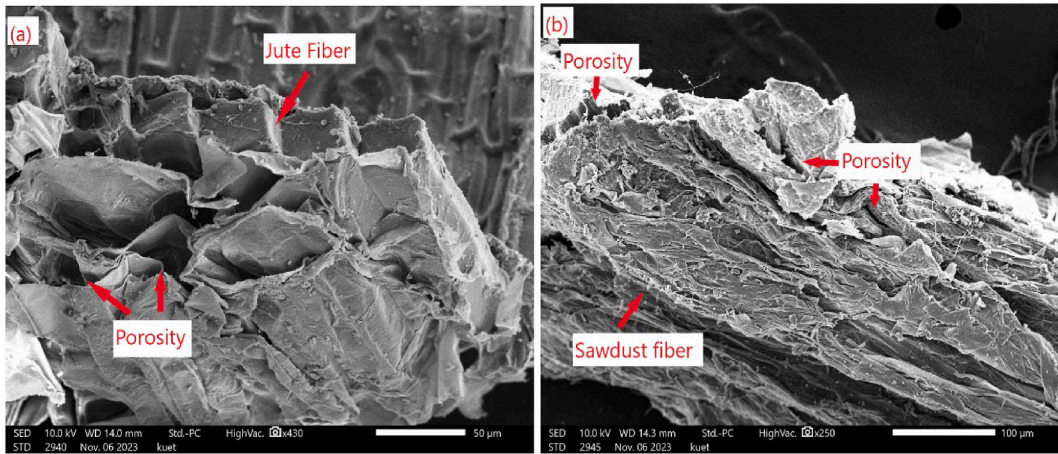


Fig. 10. a) Porosity in single jute fiber and b) Porosity in the single sawdust fiber.

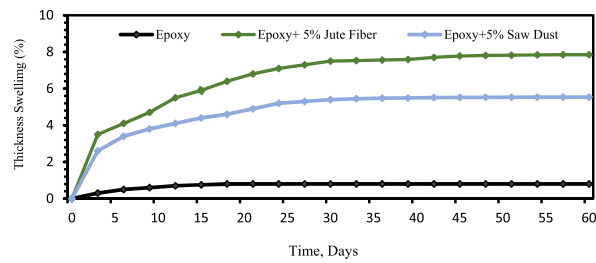


Fig. 11. Thickness swelling of the weathering samples for two months.

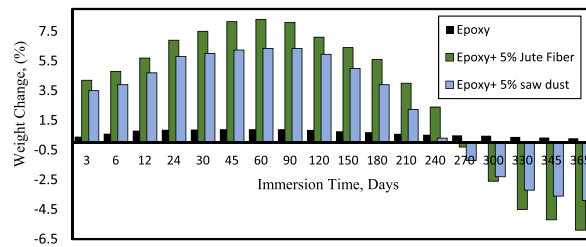


Fig. 12. Weight change of the samples in terms of the immersion days.

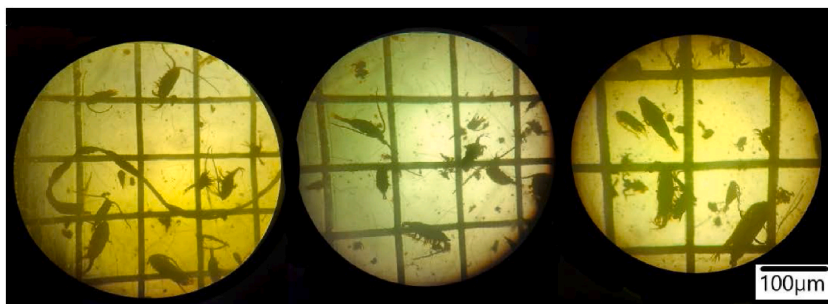


Fig. 13. Microorganisms found in the drainage system.

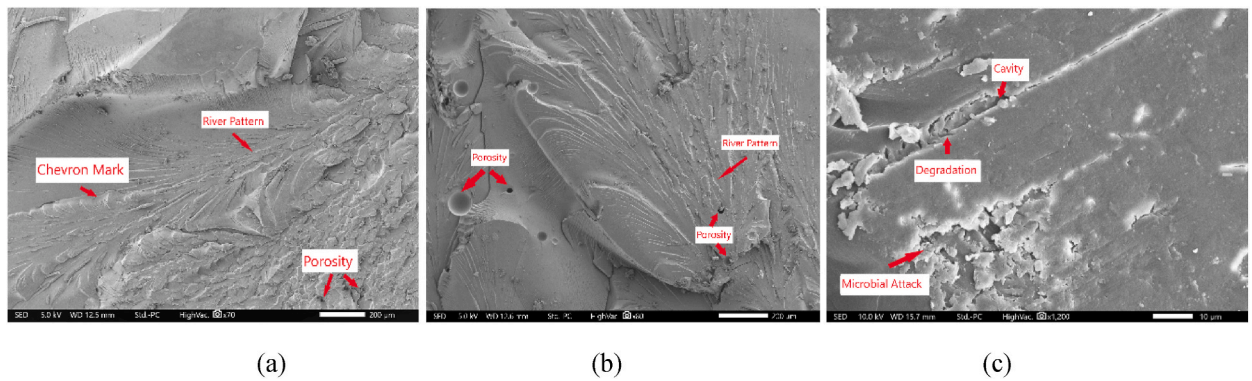


Fig. 14. SEM image of the fractured surface of the (a) non-weathered epoxy sample (b) 6 month weathered epoxy sample and (c) 1-year weathered epoxy sample.

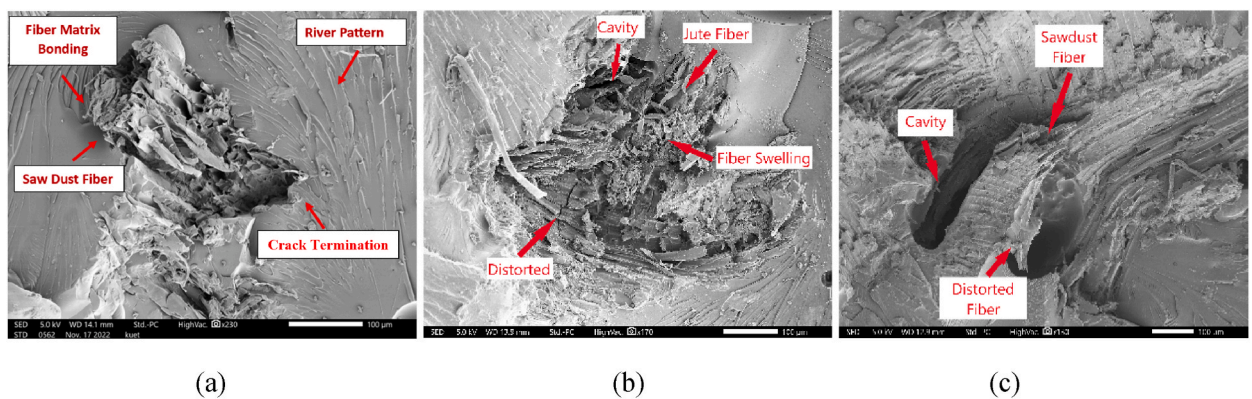


Fig. 15. SEM image of the fractured surface of the (a) non-weathered saw dust fiber composite (b) 6 months weathered saw dust fiber composite and (c) 1-year weathered saw dust fiber composite.

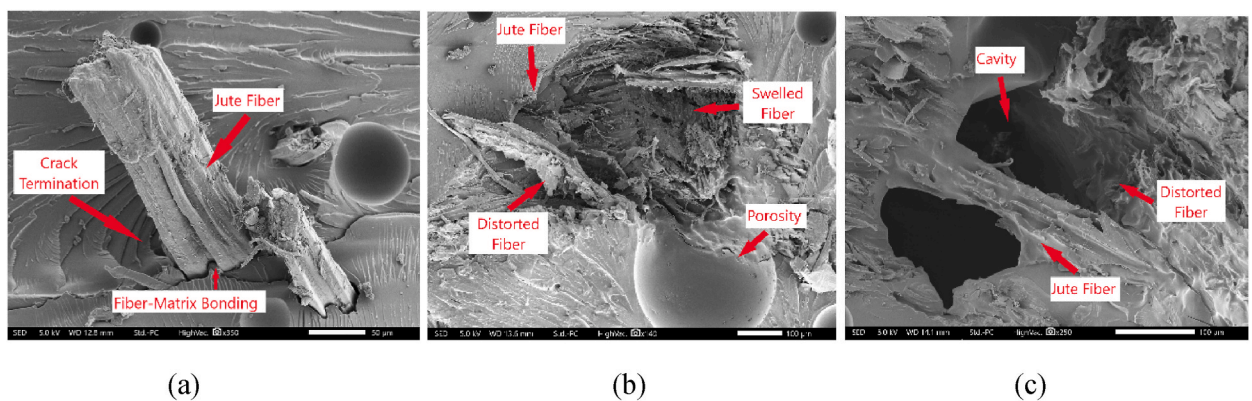


Fig. 16. SEM image of the fractured surface of the (a) non-weathered jute fiber composite (b) 6 months weathered jute fiber composite and (c) 1-year weathered jute fiber composite.

after a year of weathering, the sawdust fibers were severely damaged, and large cavities were formed due to microbial attack. The weight loss observed in Fig. 12 for the weathered composite samples can be explained by the degraded fiber, which microorganisms used for energy and deteriorated the composites. Thus, the impact energy was degraded due to weathering into the drainage system over time. However, the jute fiber composites underwent notable severe degradation after 6 months and 1 year of weathering due to the lack of protection by lignin content (Fig. 16b and c). Therefore, the degradation of jute fiber composites was greater than sawdust fiber composites due to higher fiber swelling and cavity formation.

3.6. FTIR analysis

Fig. 17 shows the FTIR analysis of the as-cast samples. It reveals that the fibers absorbed moisture from the environment [43,44]. However, for the weathered jute and sawdust fiber samples, a new peak emerged at the 1700 cm^{-1} wavenumber, representing the C=O group that is absent for weathered epoxy. The C=O functional group indicates CO₂ formation [43,44]. The CO₂ formation provides information that suggests microorganisms used the fibers as their energy sources and degraded them. As a result, microorganisms in the drainage system helped to degrade the natural fiber composites, making the sawdust and jute fiber composites biodegradable.

3.7. Microbial activities within the natural fiber

Microorganisms have been detected on the outer surface of weathered composite materials made of jute and sawdust fibers, as illustrated in Fig. 18a and c. Scanning electron microscope (SEM) images of the weathered composite samples show that fungi and bacteria are growing on the surfaces of both jute and sawdust fibers. The filaments and clusters of these bacteria are seen adhering to the fibers of the matrix. The images demonstrate how the growth of microorganisms inside the natural fibers leads to the development of biofilms and colonies. Some disintegration of the jute and sawdust fibers has also been observed due to these microbial attacks. According to the EDS analysis (Fig. 18b and d), there were no trace metals found from the drainage system except for Au, which came from the gold coating used during SEM sample preparation. The presence of oxygen represents the fiber's oxidation process during the weathering period by the microorganism [45,46].

Fig. 19a and c depict the growth of microorganisms and biofilm on jute and sawdust fibers. These microorganisms attach themselves to the fibers, which they utilize as their energy source and for their growth. The size distribution of the microorganisms' growth is shown in Fig. 19b and d. It can be observed that the microorganisms grow more rapidly on jute fibers than on sawdust fibers, as they can easily degrade the fibers and use them as an energy source. This makes jute fiber composites more degradable than sawdust fiber composites.

4. Conclusions

The study aimed to examine the effects of adding 5% each of waste jute and wood sawdust fibers to epoxy-based polymer composites on toughness and fracture behavior through the drop ball impact test and Charpy impact test. The addition of waste sawdust and chopped jute fibers improved the impact resistance of the polymer by slowing down the crack propagation. However, weathering in drain water degraded the sawdust fiber composite and jute fiber composite reduced their toughness energy, and altered the fracture patterns. The textures and color of the epoxy and composite samples changed with numerous voids after one year of weathering in the drainage system. Weight gain occurred in the initial period, but weight loss happened after three months due to the degradation of the fiber. Microorganisms present in the drainage system used the fiber as their energy source deteriorated the fibers and produced CO₂. The production of CO₂ was identified by the FTIR analysis of the weathered composite samples. TGA analysis of the as-cast and weathered samples shows that the onset thermal degradation temperature of the weathered composites was reduced due to composite

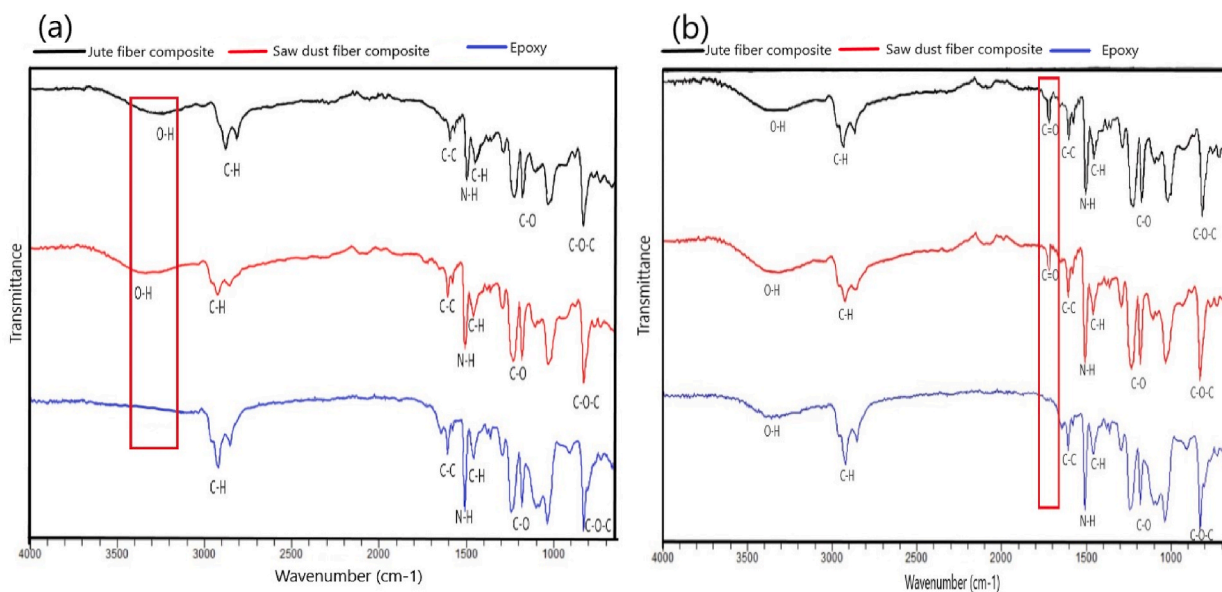


Fig. 17. FTIR analysis of the (a) as-cast samples and (b) 1-year weathered samples.

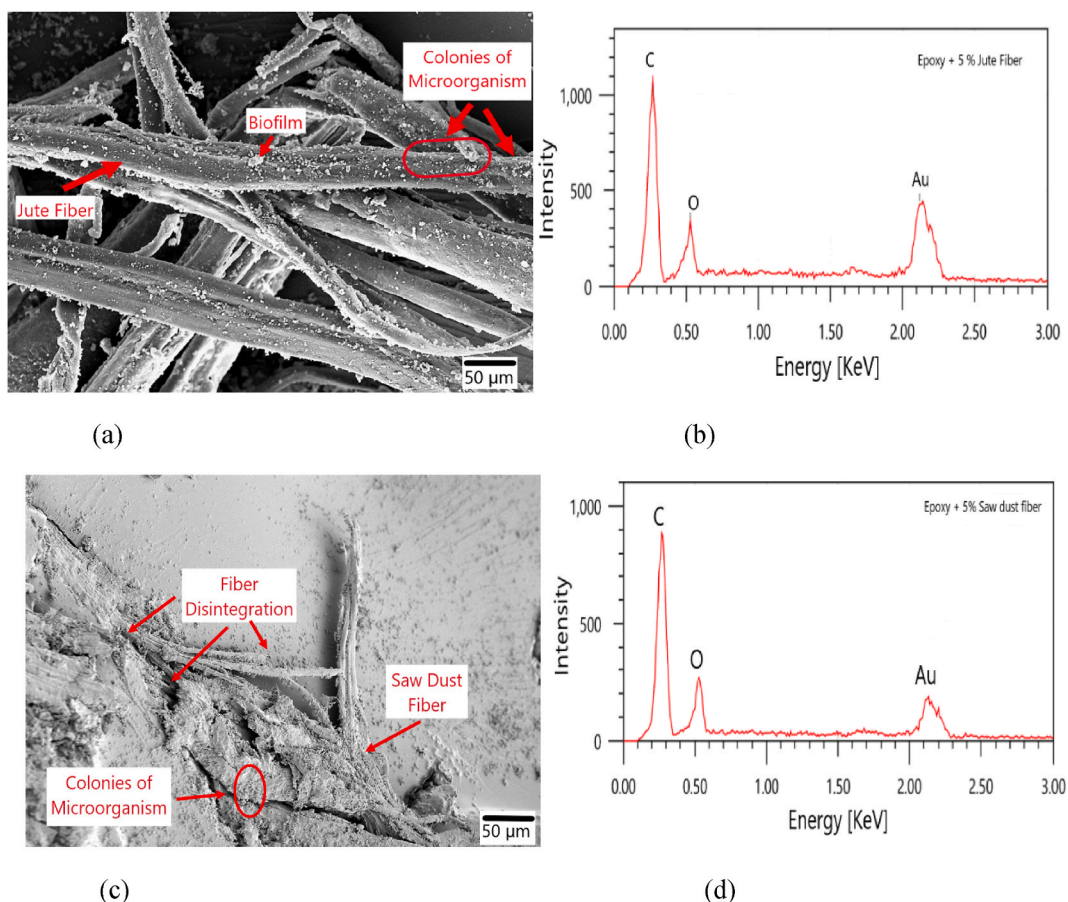


Fig. 18. a) Microbial activity at the jute fiber surface of the weathered sample; b) EDS analysis of the weathered jute fiber composite; c) Microbial activity at the sawdust fiber surface of the weathered sample; d) The weathered saw dust composite d) EDS analysis of the weathered jute fiber composite.

degradation. One year of weathering in the drainage system disintegrated the fiber through microbial attack and fiber swelling by the absorption of water by the porous jute fiber and sawdust fiber was identified by the SEM image. A higher growth rate of the microorganisms was found in the jute fiber composite than the sawdust fiber composite as sawdust contains a high level of lignin that protects it from degradation. All the findings suggest that both sawdust fiber and jute fiber composites induced biodegradability to the epoxy matrix, but jute fiber was more prominent in this regard. Thus, the addition of the waste natural fibers laid the foundation for producing more advanced and eco-friendly polymeric materials.

Data availability statement

No data was used for the research described in the article.

Additional information

No additional information is available for this paper.

CRediT authorship contribution statement

Mohammad Salman Haque: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **M.A. Islam:** Writing – review & editing, Supervision, Resources, Project administration, Conceptualization, Formal analysis, Investigation, Methodology, Validation, Visualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to

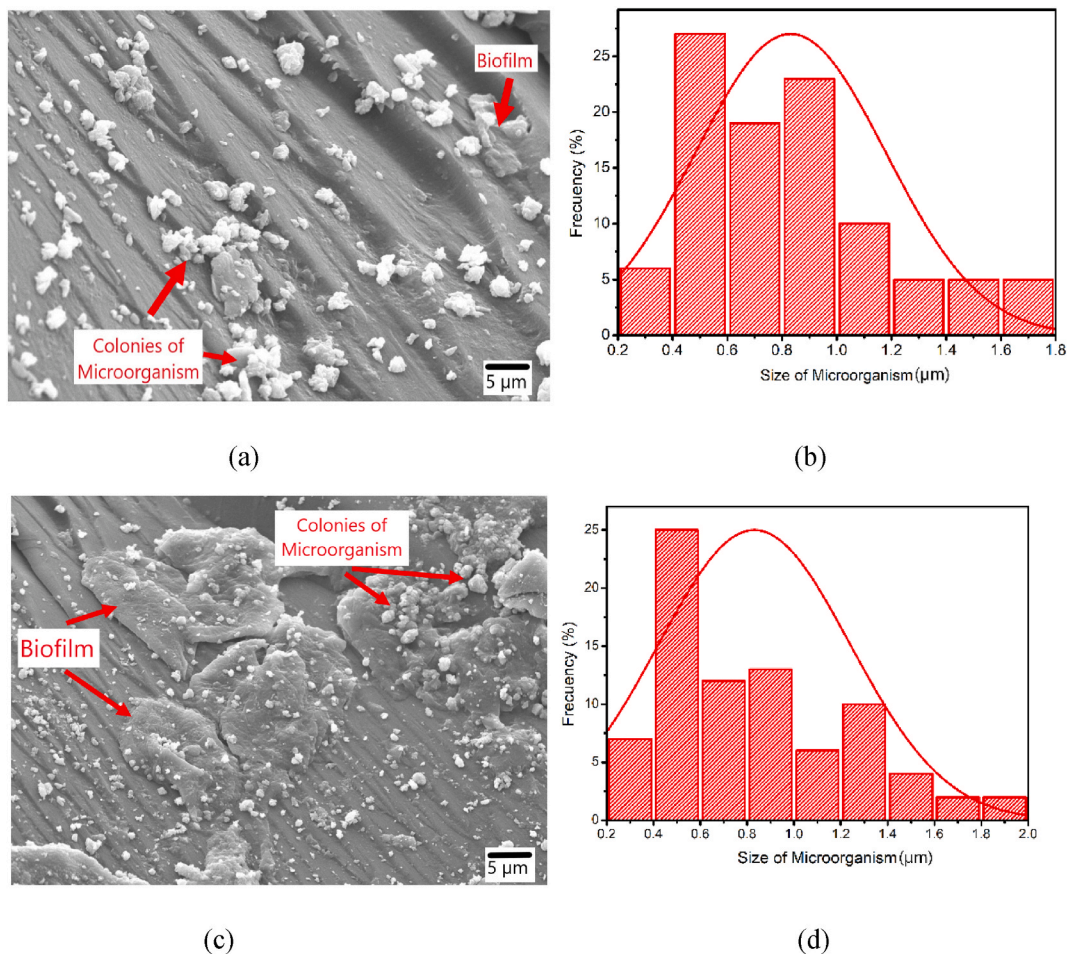


Fig. 19. a) Growth of the microorganisms at the jute fiber surface of the weathered sample; b) Size distribution of the microorganism at the jute fiber; c) Growth of the microorganisms at the jute fiber surface of the weathered sample; c) The weathered saw dust composite d) Size distribution of the microorganism at the sawdust fiber.

influence the work reported in this paper.

Acknowledgments

For their assistance with and support of this research, the authors would like to thank the Materials and Metallurgical Engineering (MME) Department of the Bangladesh University of Engineering and Technology (BUET) and the Department of Materials Science and Engineering of the Khulna University of Engineering and Technology (KUET).

References

- [1] T. McNally, Introducing functional composite materials, *Functional Composite Materials* 1 (2020) 1, <https://doi.org/10.1186/s42252-020-00007-9>.
- [2] P. Yao, et al., Review on polymer-based composite electrolytes for lithium batteries, *Front. Chem.* 7 (2019), <https://doi.org/10.3389/fchem.2019.00522>.
- [3] Mohammad Salman Haque, Modassher Nomani, Azmery Akter, Istiak Ahmed Ovi, Synergistic effect of Mg addition on the enhancement of the mechanical properties and evaluation of corrosion behaviors in 3.5 wt% NaCl of aluminum alloys, *Heliyon* (2024) e25437.
- [4] Kanok Md Mehedi Hasan, Shifa Silvina Siddika, Haque Mohammad Salman, Akter Azmery, Comparative study on mechanical properties of banana-glass fiber reinforced epoxy hybrid composites, *Mater. Today: Proc.* (2023). <https://doi.org/10.1016/j.matpr.2023.08.356>.
- [5] M. Winnacker, B. Rieger, Bio-based polyamides: recent advances in basic and applied research, *Macromol. Rapid Commun.* 37 (2016) 1391–1413, <https://doi.org/10.1002/marc.201600181>.
- [6] J. Njuguna, et al., Natural Fiber-Reinforced Polymer Composites and Nanocomposites for Automotive Applications, *Cellulose Fibers: Bio- and Nano-Polymer Composites*, 2011, pp. 661–700, https://doi.org/10.1007/978-3-642-17370-7_23.
- [7] Mohammad Salman Haque, SM Nasim Rokon, Mohammad Salim Kaiser, Strengthening and softening behavior of non-heat-treatable aluminum alloys subject to deformation and annealing treatment, *Mater. Today: Proc.* 82 (2023) 151–157.
- [8] S.-J. Tan, et al., Recent advancements in polymer-based composite electrolytes for rechargeable lithium batteries, *Electrochem. Energy Rev.* 1 (2) (2018) 113–138, <https://doi.org/10.1007/s41918-018-0011-2>.

- [9] N. Vignesh, N. Hynes, P.V. Shenbaga, R. Pravin, A survey on characterization of natural fibers, in: *Advances in Basic Science*, AIP Conference Proceedings, vol. 2142, 2019 150014, <https://doi.org/10.1063/1.5122563>, 1-5.
- [10] H.A.J. Sadiqsha, P.P. Patil, "A review on natural fiber composite", *J. Xidian Univ.* 14 (2020) 2884–2889, <https://doi.org/10.37896/jxu14.6/332>.
- [11] Haque, Mohammad Salman, Istiak Ahmed Ovi, Farah Samsi Prome, and Anika Hossain. "Impact of water absorption on tensile strength: A Comparative Study of Jute Fiber Composites with Various Fiber Content."
- [12] Y. Zhao, M. Cao, H.X. Tan, M. Ridha, T.E. Tay, Hybrid woven carbon-dyneema composites under drop-weight and steel ball impact, *Compos. Struct.* 236 (2020) 111811, <https://doi.org/10.1016/j.compstruct.2019.111811>.
- [13] S. Kalnaus, et al., Analysis of composite electrolytes with sintered reinforcement structure for Energy Storage Applications, *J. Power Sources* 241 (2013) 178–185, <https://doi.org/10.1016/j.jpowsour.2013.04.096>.
- [14] N. Navaranjan and T. Neitzert, "Impact Strength of Natural Fiber Composites Measured by Different Test Methods: A Review. MATEC Web of Conferences, vol. 109, pp.01003, <https://doi.org/10.1051/mateconf/201710901003>.
- [15] Y. Zhang, et al., Excellent energy storage performance and thermal property of polymer-based composite induced by multifunctional one-dimensional nanofibers oriented in-plane direction, *Nano Energy* 56 (2019) 138–150, <https://doi.org/10.1016/j.nanoen.2018.11.044>.
- [16] G. Zhao, et al., Polymer-based nanocomposites for heavy metal ions removal from Aqueous Solution: a Review, *Polym. Chem.* 9 (26) (2018) 3562–3582, <https://doi.org/10.1039/c8py00484f>.
- [17] J. Shesan, O, et al., "Improving the mechanical properties of natural fiber composites for structural and biomedical applications," *Renewable and Sustainable Composites [Preprint]* (2019) <https://doi.org/10.5772/intechopen.85252>.
- [18] S. Ma, X. Zhuang, X. Wang, Particle distribution-dependent micromechanical simulation on mechanical properties and damage behaviors of particle reinforced metal matrix composites, *J. Mater. Sci.* 56 (11) (2021) 6780–6798, <https://doi.org/10.1007/s10853-020-05684-2>.
- [19] M.R. Hossain, et al., Tensile behavior of environment friendly jute epoxy laminated composite, *Procedia Eng.* 56 (2013) 782–788, <https://doi.org/10.1016/j.proeng.2013.03.196>.
- [20] R.M. Hossain, et al., Effect of fiber orientation on the tensile properties of jute epoxy laminated composite, *J. Sci. Res.* 5 (1) (2012) 43–54, <https://doi.org/10.3329/jsr.v5i1.10519>.
- [21] M.F. Hossain, M.K. Islam, M.A. Islam, Effect of chemical treatment on the mechanical and physical properties of wood saw dust particles reinforced polymer matrix composites, *Procedia Eng.* 90 (2014) 39–45, <https://doi.org/10.1016/j.proeng.2014.11.811>.
- [22] M.F. Hossain, S.N. Shuvo, M.A. Islam, Effect of types of wood on the thermal conductivities of wood saw dust particle reinforced composites, *Procedia Eng.* 90 (2014) 46–51, <https://doi.org/10.1016/j.proeng.2014.11.812>.
- [23] Shraboni Sarker, Mohammad Salman Haque, Md Saddin Azad Alvy, Md Mehedi Hasan Abir, The effects of solution heat treatment on the microstructure and hardness of an aluminum-4% copper alloy with added nickel and tin, *Journal of Alloys and Metallurgical Systems* 4 (2023) 100042.
- [24] A. Majumder, et al., Physical and mechanical characteristics of as-cast jute fibers, threads and diatoms, *Construct. Build. Mater.* 326 (2022) 126903, <https://doi.org/10.1016/j.conbuildmat.2022.126903>.
- [25] N. Nandihalli, C.-J. Liu, T. Mori, Polymer-based thermoelectric nanocomposite materials and devices: fabrication and characteristics, *Nano Energy* 78 (2020) 105186, <https://doi.org/10.1016/j.nanoen.2020.105186>.
- [26] K. Varaprasad, et al., Alginate-based composite materials for wound dressing application: A mini-review, *Carbohydr. Polym.* 236 (2020) 116025, <https://doi.org/10.1016/j.carbpol.2020.116025>.
- [27] M.W. Islam, et al., Fabrication and mechanical characterization of bagasse, rice husk, saw dust reinforced epoxy composites, *AIP Conf. Proc.* (2019), <https://doi.org/10.1063/1.5115957>.
- [28] D. Zhou, et al., Performance and behavior of LLZO-based composite polymer electrolyte for lithium metal electrode with high capacity utilization, *Nano Energy* 77 (2020) 105196, <https://doi.org/10.1016/j.nanoen.2020.105196>.
- [29] F. Zhang, Y. Feng, W. Feng, Three-dimensional interconnected networks for thermally conductive polymer composites: design, preparation, properties, and Mechanisms, *Mater. Sci. Eng. R Rep.* 142 (2020) 100580, <https://doi.org/10.1016/j.mser.2020.100580>.
- [30] M.A. Mohd Abdah, et al., Review of the use of transition-metal-oxide and conducting polymer-based fibers for high-performance supercapacitors, *Mater. Des.* 186 (2020) 108199, <https://doi.org/10.1016/j.matdes.2019.108199>.
- [31] B. Fan, et al., Polymer-based materials for achieving high energy density film capacitors, *Prog. Polym. Sci.* 97 (2019) 101143, <https://doi.org/10.1016/j.progpolymsci.2019.06.003>.
- [32] X. Tong, et al., Recent advances in natural polymer-based drug delivery systems, *React. Funct. Polym.* 148 (2020) 104501, <https://doi.org/10.1016/j.reactfunctpolym.2020.104501>.
- [33] D.Q. Tan, Review of polymer-based nanodielectric exploration and film scale-up for advanced capacitors, *Adv. Funct. Mater.* 30 (18) (2019) 1808567, <https://doi.org/10.1002/adfm.201808567>.
- [34] S. Chen, et al., Polymer-based dielectric nanocomposites with high energy density via using natural sepiolite nanofibers, *Chem. Eng. J.* 401 (2020) 126095, <https://doi.org/10.1016/j.cej.2020.126095>.
- [35] A.M. Maan, et al., Recent developments and practical feasibility of polymer-based antifouling coatings, *Adv. Funct. Mater.* 30 (32) (2020) 2000936, <https://doi.org/10.1002/adfm.202000936>.
- [36] S. Chen, et al., Poly(vinylene carbonate)-based composite polymer electrolyte with enhanced interfacial stability to realize high-performance room-temperature solid-state sodium batteries, *ACS Appl. Mater. Interfaces* 11 (46) (2019) 43056–43065, <https://doi.org/10.1021/acsami.9b11259>.
- [37] R. Gobi, et al., Biopolymer and synthetic polymer-based nanocomposites in wound dressing applications: a Review, *Polymers* 13 (12) (2021) 1962, <https://doi.org/10.3390/polym13121962>.
- [38] D. Zhao, et al., Poly(lactic-co-glycolic acid)-based composite bone-substitute materials, *Bioact. Mater.* 6 (2) (2021) 346–360, <https://doi.org/10.1016/j.bioactmat.2020.08.016>.
- [39] S. Xu, et al., Conducting polymer-based flexible thermoelectric materials and devices: from mechanisms to applications, *Prog. Mater. Sci.* 121 (2021) 100840, <https://doi.org/10.1016/j.pmatsci.2021.100840>.
- [40] M. Naseri-Nosar, Z.M. Ziara, Wound dressings from naturally-occurring polymers: a review on homopolysaccharide-based composites, *Carbohydr. Polym.* 189 (2018) 379–398, <https://doi.org/10.1016/j.carbpol.2018.02.003>.
- [41] M.R. Hossain, et al., Quantitative analysis of hollow lumen in Jute, *Procedia Eng.* 90 (2014) 52–57, <https://doi.org/10.1016/j.proeng.2014.11.813>.
- [42] H. Huang, et al., Nano-SiO₂-embedded poly(propylene carbonate)-based composite gel polymer electrolyte for lithium-sulfur batteries, *J. Mater. Chem. A* 6 (20) (2018) 9539–9549, <https://doi.org/10.1039/c8ta03061h>.
- [43] R.A. Surmenev, et al., Hybrid lead-free polymer-based nanocomposites with improved piezoelectric response for biomedical energy-harvesting applications: a Review, *Nano Energy* 62 (2019) 475–506, <https://doi.org/10.1016/j.nanoen.2019.04.090>.
- [44] T.K. Das, P. Ghosh, N.C. Das, Preparation, development, outcomes, and application versatility of carbon fiber-based Polymer Composites: a Review, *Adv. Compos. Hybrid Mater.* 2 (2) (2019) 214–233, <https://doi.org/10.1007/s42114-018-0072-z>.
- [45] E. Avcu, et al., Electrophoretic deposition of chitosan-based composite coatings for biomedical applications: a Review, *Prog. Mater. Sci.* 103 (2019) 69–108, <https://doi.org/10.1016/j.pmatsci.2019.01.001>.
- [46] M. Fazeli, M. Keley, E. Biazar, Preparation and characterization of starch-based composite films reinforced by cellulose nanofibers, *Int. J. Biol. Macromol.* 116 (2018) 272–280, <https://doi.org/10.1016/j.ijbiomac.2018.04.186>.