Original Research Article

Development of Brewed Tea Waste Reinforced Jute Polyester Composites: Physical, Mechanical, Structural and Thermal Characterizations

Abstract

The effect of brewed tea waste filler on mechanical, physical, structural and thermal characteristics were evaluated.Results indicate that mechanical strength improved maximum at 6wt% of filler, and thereafter declined because of the poor bonding between filler and polyester matrix.The composites with the highest values of elongation and hardness were those containing 0wt% and 15wt% filler, respectively. With increasing hardness effective resistance to deformation is also increased due to the strong adhesion between the filler and matrix.The bulk density showed an improvement as the filler content was increased.Moreover, tests for water absorption and soil degradation were used to examine the tendency of composites for moisture and biodegradation.FTIR spectroscopy was used to determine the existence of chemical compounds and functional groups in order to provide a comprehensive characterization of the material. Thermal analysis reveal that 15wt% composites exhibit maximum thermal stability and degradation temperature.SEM analysis was used to perform surface morphological characterization of composites, which displayed fiber pullout, voids, and other fracture mechanisms. Moreover, this finding indicated the

strong interfacial adhesion between the filler and matrix at 6wt%, which enhanced mechanical characteristics the composites.

Keywords: Brewed tea waste, Jute fiber, Polyester resin, Bulk density, Thermal properties

1.Introduction

Due to pollution and changing weather patterns, environmental consciousness has recently become an alarming global issue. Researchers worldwide have been prompted to create environmentally friendly composites of natural fiber as part of the fight to save our climate. Natural fibers appeal to researchers and manufacturers because of their biodegradability, improved mechanical properties, ease of manufacture, and overall cost efficiency. Despite a few drawbacks, replacing synthetic fibres for natural fibres as reinforcing material in composites may give a green, renewable approach for 'Go Green' technological applications. Jute, a widely available and one of the most common lignocellulosic reinforcement materials in Bangladesh [1,2]. The main benefits of utilizing jutefiber as a reinforcing materialin making polymer composites are its biocompatibility, cost effectiveness, non-abrasive nature, light weight, lower energy consumption, broad variety, and most importantly, its ability to create an agriculture-based economy [3].

Unsaturated polyester (UPE) resins are some of the most essential and commercially useful thermosetting materials owing to their low cost, easy manufacturing, and highly efficient characteristics,they are used in a variety of functions, including automobiles, naval, transport, building materials, and electronic equipment. The ability to chemically modify UPEs with crosslinkers makes them useful. They are often reinforced with multiple types of filler materials ranging from nanoscale to macro size in order to enhance their mechanical properties and tribological characteristics. [4]. The use of natural waste filler to reinforcementpolymers as a replacement for synthetic fillers is receiving attention for not onlylowering the cost but also reducing negative environmental impacts.

Camellia sinensis, a green species or tree of the family Theaceae, is the source of the leaves used to brew tea.Bangladesh is the 10th largest tea producer country in the world and after jute, tea is Bangladesh's second most important export cash crop. The sector generates 3% of global tea supply and accounts for 1% of national GDP of Bangladesh. In 2020 to 2021,

Bangladesh will produce 86.39 million kg of tea, while domestic consumption would equal 57.06 million kg.Increased amounts of tea waste are being produced as a consequence in Bangladesh [5,6].

Tea sweepings, tea dust, tea thread, tea stems, and any other material claiming to be tea that does not satisfy the tea criteria set out in the Prevention of Food Adulteration Act of 1954 are all considered as tea waste[7]. Green tea and green tea stems are not included in this definition.Therefore, the brewed tea leaves that are discarded after being used are the subject of this study and hereafter referred to as tea waste.

Tea waste has almost the similar ingredient as tea and the most significant compounds contained in tea seems to be the polyphenols and caffeine.The antioxidants, catechins, flavonoids, cellulose, amino acid, non-soluble proteins, fiber, sugars, lignin, zinc, and tannic acid that characterize tea so flavorful and nutritious are all present in the waste that is left over after brewing.Soil and nutritional solutions are two examples of growth medium from which plants absorb these trace elements.Rather of burning or burying the garbage, it may be readily processed to remove, blend, and mould them into something of worth. [8,9,10].

It is possible that tea waste, a lingo-cellulosic product, will be employed as a natural reinforcing material or composite filler because to its advantages over inorganic resources, including its rapid reusability, low energy requirements, and cheap cost. In view of the environmental perspective and cost effectiveness, tea waste is a promising candidate for use as a filler in polymer composites [7]. Moreover, Tea waste seems to be an abundant material that draw scientists in diverse fields with several versatile properties. This is because tea waste is now being used for a wider variety of purposes, such as minerals, adsorptive coatings, livestock feed, fertilizer, fuel bioenergy, bio-char, organic-oil, and other similar things. In addition, caffeine nutrient has a wide demand with a significant export potential in tea waste[11,12].

In previous studies,Bagheriet al. fabricated tea waste reinforced polypropylene and nanoclay hybrid composites [13], Rasoolet al. utilized black tea as a partial replacement for cement [14], Gokulkumaret al. investigatedtea leaf filler based pineapple leaf/glass fiber composites [15], Theja et al. used green tea waste for removal of heavy metal from waste water[16], Radhika et al. extracted carbon from tea waste to develop nanocomposites[17],Khalid et al. prepared tea waste reinforced PLA based polymer biocomposites[18], Hussinet

al.fabricatedbrewed tea leaf /polyester composites [7] . But, a little concentration was given to investigate several properties of brewed tea waste filler based UPE polymercomposites.We have worked on industrial tea waste reinforced jute unsaturated polyester composites in recent past.[19].To the best of our knowledge, there have been no reports on brewed tea waste reinforced jute unsaturated polyester composites.In the present study, composites containing jute, UPE and brewed tea waste with various compositions were fabricated by hand lay-up method. The samples were characterized by various experimental techniques, and the impact of brewed tea waste filler on the mechanical, physical, structural, and thermalproperties of jute polyester composites were investigated.

2.Experimental

2.1 Materials and Method

Unsaturated polyester (UPE) resin (3262P-DC, SHCP, Singapore) was employed as the polymer matrix, and woven jute cloth mat was used as the reinforcing material (collected from local market, Rajshahi).For the treatment of jute mat, methyl ethyl ketone peroxide (MEKP,MOLPEROX F60, Turkey) was utilized as the hardener material, and sodium hydroxide NaOH (Merck, India) was employed for pre-treatment of jute. The tea waste (also known as TW) that was utilized in this investigation was obtained from several tea vendors located in the University of Rajshahi.

2.2 Preparation of Tea Waste Powder

Waste from brewing tea was collected, cleaned thoroughly with running water to eliminate any remaining non-cellulosic materials, dried in an oven for 48 hours at 60°C to remove any remaining moisture, and finally milled for 5 minutes.To get the fine powder of tea waste, it was then manually sieved through a 250micron sieve.

2.3 Treatment of Jute Mat (JM)

The jute mat has been given an alkaline treatment with a 5% NaOH solution. Since it was stated that prolonged contact with NaOH might compromise the structural integrity of natural fibers, the treatment was carried out for a single hour at room temperature. Because of this, the NaOH solution was removed from the jute mat by repeatedly washing it with distilled water and then drying it at 60˚C for 24 hours[20,21, 22].

2.4 Fabrication of Composites

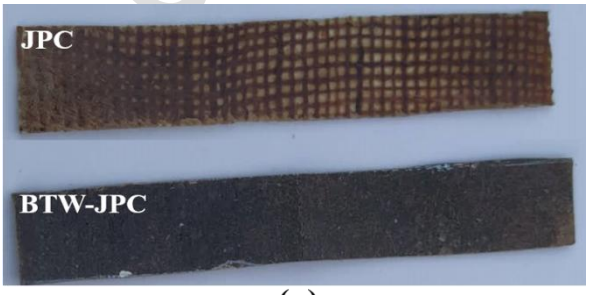
For the fabrication of the composite specimens, a conventional hand lay-up method was used. Before putting it to the jute mat, the matrix material was prepared by thoroughly combining unsaturated polyester resin, brewed tea waste powder, and 1% MEKP in a magnetic stirrer hot plate. One layer of jute mat was made up of two layers of mix matrix solution to construct the composites. In composites, we experimented with varying the percentage of tea wastes filler of 0,3,6,9,12 and 15% by weight while keeping the percentage of jute mat remain constant of 10 wt%. A steel mold measuring 150 mm × 120 mm × 5 mm in thickness was made. The base of the mold was covered with melot paper. On milot paper a mixed matrix solution was placed. The jute mat was then spread on top of the blended matrix and rolled out appropriately. The jute mat had another layer of mixed matrix solution added to it. After that, a melot paper is applied and the mould is closed. This sandwich was then pressed in a hydraulic lamination heat press machine at 95°C for 15 minutes while under a pressure of 5 tons to fabricate the composites. Then, composites were brought down to room temperature. Finally, two steel molds were removed, the composites were separated, they were trimmed to size in accordance with ASTM requirements, and they were stored in desiccators.

2.5 Testing Methods

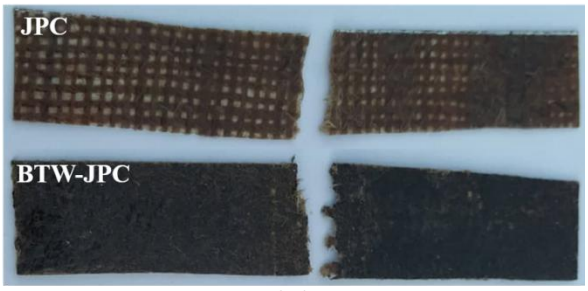
2.5.1 Mechanical Properties

(i) Tensile and Flexural testing

Both neat jute polyester (0% filler) composites (JPC) and brewed tea waste jute polyester composites (BTW-JPC) were subjected to tensile and flexural testing utilizing a Universal Testing Machine (WDW-50, Serial No- 180067,China) in accordance with ASTM D 638 and ASTM D790-98 respectively. Without an extensometer, tensile tests were conducted with a clamping length of 15 mm for each specimen on each jaw and to ensure accuracy, testing were conducted using a crosshead speed of 5 mm/min. For flexural test the speed was 10 mm/min. Each recorded result is the mean of five independent measurements.



(a)



(b)

Figure1Tensile test specimen (a) before fracture (b) after fracture.

(ii)Hardness Test

The Rockwell Hardness Tester (HR-150DT,China) used to evaluate the hardness of fabricated composite samples. The Rockwell hardness test was performed in accordance with the ASTM D785 standards with a 150 applied load, a diamond indenter (Rockwell C scale) was used for the hardness test. Each provided value is also the mean of five independent measurements.

2.5.2 Physical Properties

(i) Bulk Density

Composites were tested for bulk density using ASTM C-134-76. Bulk density was determined after the samples had been dried in an oven at 60˚C for 24 hours.Using the average values for length, width, thickness, and weight of five samples, the bulk density of composites was calculated using the following Equation (2.1):

$$\text{Bulk Density, D} = \frac{W_s}{V} = \frac{W_S}{L \times W \times H} \quad \text{..... (2.1)}$$

Where, (Ws = weight, L = length, W = width, and H = height of sample), respectively.

(ii) Water Absorption Test

As specified by the ASTM in their standard C-67-91, the water absorption rates of the samples were determined. Experiment samples were sized at 6 cm in length, 2 cm in width, and 0.5 cm in thickness. The samples were heated in an oven at 80°C for 24 hours. It was weighed as soon as it was taken out of the oven. Let this weight be W_i. After that, the samples were then kept for 24 hours in distilled water at 25°C. It was removed from the water, cleaned with a cloth, let to air dry, and then weighed. Let refer to this weight as W_f.

The following formula was then used to determine the total water absorbed:

$$\text{Water absorption (\%)} = \left[\frac{W_f - W_i}{W_i}\right] \times 100\% \quad \text{..... (2.2)}$$

(iii) Soil Burial Test

According to the procedure outlined by Goudar et al. [23], the biodegradability of the prepared composites were investigated using a soil burial test in a controlled laboratory setting. Fresh soil was gathered from a garden on the campus of Rajshahi University in Rajshahi, Bangladesh. In order to get the initial dry weight, the samples were sliced into 2 × 2 cm^2 and dried at 40°C (W_1).The samples were placed 8-10 cm below the soil surface. Water was sprayed over the soil's surface to keep it wet. After 20 days, the composites were dug out of the soil, washed, and dried in an oven, and their weight was recorded (W_2). Equation (2.3) was used to determine soil deterioration.

$$\text{Soil degradation (\%)} = \left[\frac{W_1 - W_2}{W_1}\right] \times 100\% \quad \text{..... (2.3)}$$

2.5.3 Structural Properties

FTIR Analysis

Perkin-Elmer Frontier FTIR/MIR Spectrometer,USA was used to analyze the infrared spectra of brewed tea waste (BTW) powder, JPC, and BTW-JPC.The scan's transmittance range was 4000 cm^{-1} to 225 cm^{-1}.

2.5.4 Thermal Properties

Differential Thermal Analysis *(*DTA)and Thermogravimetric Analysis (TGA)

Using a DTA/TGA (Perkin Elmer Simultaneous, STA-8000,USA), thermal analyzer, the melting and degradation temperatures of brewed tea waste powder, JPC, and BTW-JPC were studied. The temperature range covered by the measurements was from 30 to 700 ° C, with a heating temperature of 20°C per minute.Melting and degradation temperatures are calculated using exotherm versus temperature curves, which are shown as DTA traces.

3. Results and Discussion

3.1. Mechanical Properties of Composites

3.1.1 Tensile and Flexural Strength

The variation of mechanical strength with varying different brewed tea waste filler content is presented in Figure2.With adding only 3wt% filler to UPE resin matrix, the mechanical strength immediately drops. The possible explanation is the irregular distribution of filler at low filler loading.When filler material was added after the 3wt% composition, the mechanical strength improved considerably until 6wt% composition, but subsequently declined again following further increments of filler content. The maximum tensile and flexural strength value was achieved for 6wt% filler content and that was 25.45 MPa, 37.56 MPa respectively.However, the addition of 6twt% filler loading enhanced the fibre-matrix adhesion to a great level, which ultimately resulted in higher mechanical strength in the material. It is possible that the mechanical strength is also influenced by the chemical as well as its interior structure of the filler. Yet, further loading of filler caused a decrease in tensile strength may be traced back to the aggregation of filler particles inside the matrix of polymer molecules. Because of this, it can be deduced that the interfacial bonding between the matrix and the filler is not capable of withstanding significant amounts of mechanical stress.Neher et al. found that mechanical strength decreased up to a sawdust content of 10wt%, after which it increased to a maximum of 15wt %. Again, it dropped down to compositions of 20wt% [24].Rahman et al. showed that the tensile and flexural strength was increased with filler content up to 10wt%[25], but thereafter to deteriorate with subsequent addition of filler. It was interesting to learn that both parts of literature validated our results.

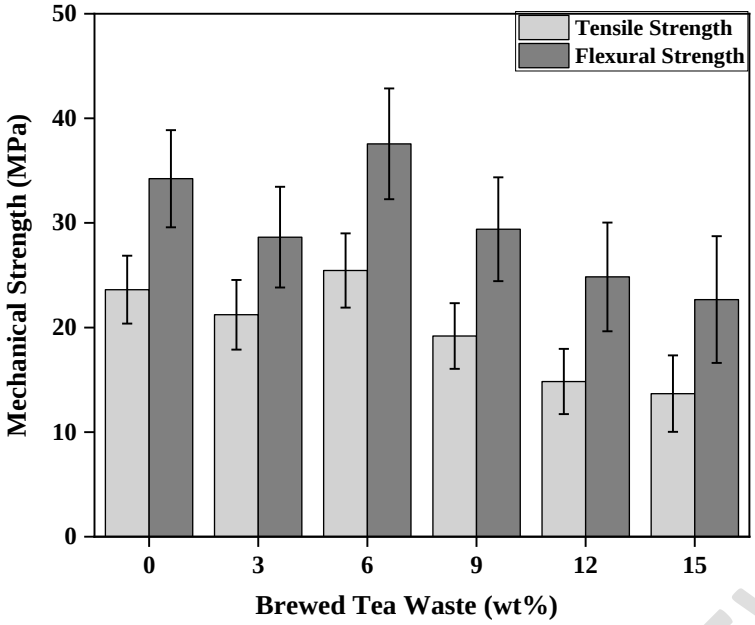


Figure2The variations of the mechanical strength as a consequence of the filler content of jute polyester composites.

3.1.2 Elongation at Break

Elongation at break is a measure of the toughness of a composite material. The impact of changing the concentration of filler on the elongation-at-break is seen in Figure 3. The elasticity of a polymer composite may be measured by its ability to undergo elongation. Figure 4 demonstrates that the material's elongation at break decreases as the filler content increases from 0% to15%. This is due to the fact that an increase in filler loading also improves the stiffness of the composite. According to the reinforcing process, molecular mobility decreases as filler loading increases due to the creation of physical bonds between filler particles. Therefore, larger filler loading leads to failure. Asimilaroutcome was found in the research published in [26, 27].As the percentage of filler in the polyester composites increased, the research found that elongation decreased.

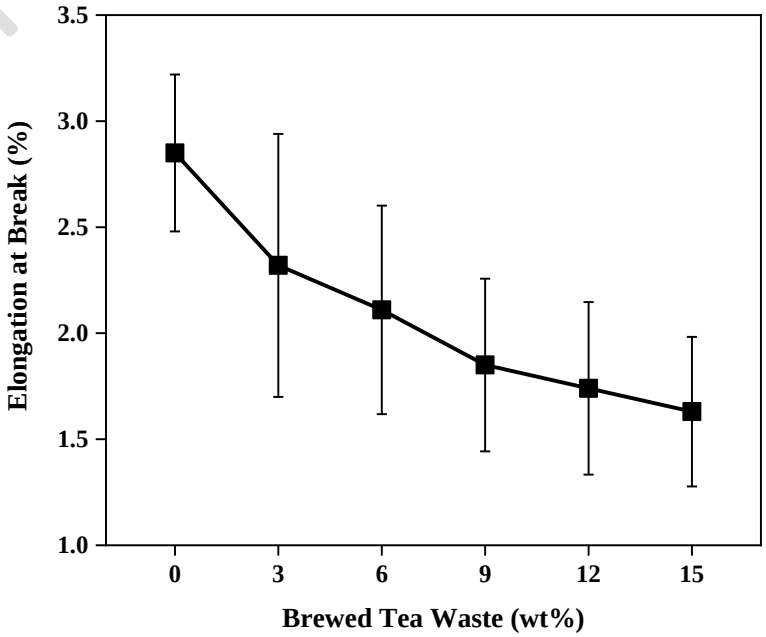


Figure3 The variations of the elongation at break as a consequence of the filler content of jute polyester composites.

3.1.3 Tensile Modulus

The tensile modulus is utilized since it provides a standard by which the stiffness of polymeric materials may be evaluated. Figures 4 describe the impact of brewed tea waste filler integration with UPE matrix on the tensile modulus of the resulting composites According to the Figure 5, the tensile modulus of 0wt% compositewas measured to be 850 MPa. The highest tensile modulus 1463 MPa was found in 6wt% filler content.The increase in tensile modulus may be due to the more even distribution of filler, which reduce the mobility of polymer chains in response to external loading.Additionally, the polymer matrix and filler successful interaction and efficient adhesion is caused the tensile modulus value to rise.There is a decrease in modulus when the filler content is more than 6 wt%.This is likely due to the agglomeration of tea waste filler in the UPE matrix and the creation of an organic–inorganic interpenetrating network. Moreover, the inhomogeneous dispersion of filler within the matrix, which led to poor contact between the filler and polymer matrix. As a result, the tensile modulus of the remaining composites decreased[28,29].

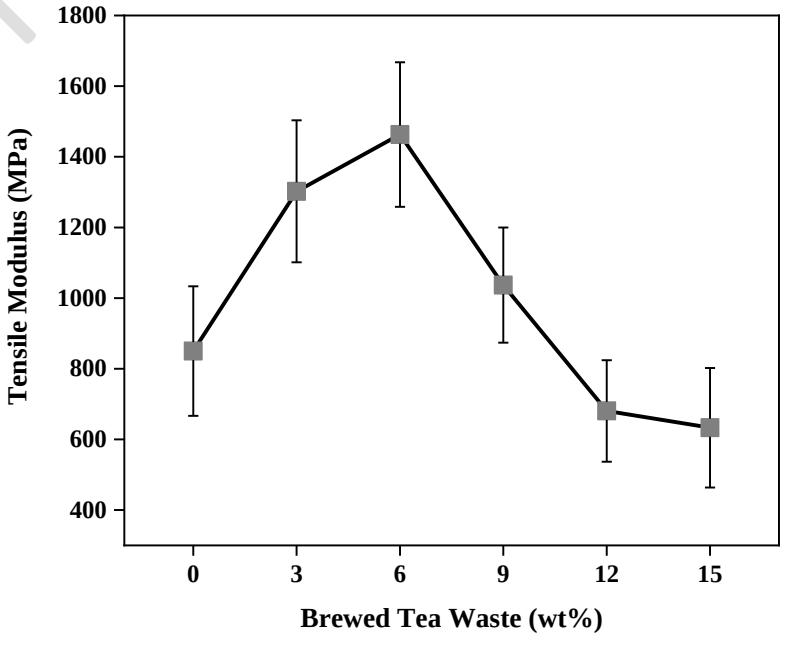


Figure 4 The variations of the tensile modulus as a consequence of the filler content of jute polyester composites.

3.1.4 Hardness

Figure 5 displays the Rockwell hardness of a jute polyester composite at varying compositions of brewed tea waste filler.Over time, the hardness of jute polyester composites enhanced when filler was included into the material. With increasing filler composition from 0wt% to15wt%, Rockwell hardness showed an increase.Composites with 15wt% filler had the highest hardness. Similar results were found in the studies by Girimuruganet al. [29], Kumar et al. [31].The incorporation of tea waste filler into the polymer matrix led to an increase in thehardness of the fabricated composites. The load was transferred from the polymer matrix to the filler due to the successful interaction between the two. As a result, the filler particles reduced the applied load, and the hardness value improved. In addition, the harder surface is the result of a more uniform distribution of filler throughout the polyester matrix. However, aggregation of filler inside the polymer matrix reduced the hardness of composites.

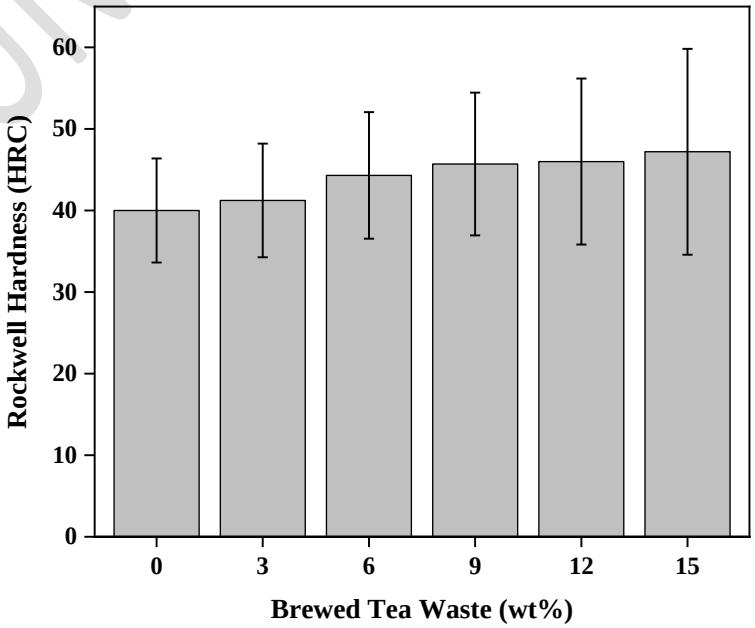


Figure5 The variations of the Rockwell hardnessas a consequence of the filler content of jute polyester composites.

3.2 Physical Properties of Composites

3.2.1 Bulk Density

The consequence of filler changes on the bulk density of jute reinforced polyester composites is shown in Figure 6. The bulk density improved, as tea waste filler content was increased from 0wt% to15wt%. Composites with0wt% filler had a density of 0.92 gm/cc, whereas, 15wt% filler had density increased to 1.19 gm/cc.Due to the porous nature of the composite, the mass of the added filler increased at a significantly faster rate than the volume, causing the composite's total bulk density to rise. An increase in bulk density indicates that the composite has become more dense. Therefore, composite with 15wt% filler is better suited for high load applications.Fariaet al. [32] noticed a similar pattern of behaviour.

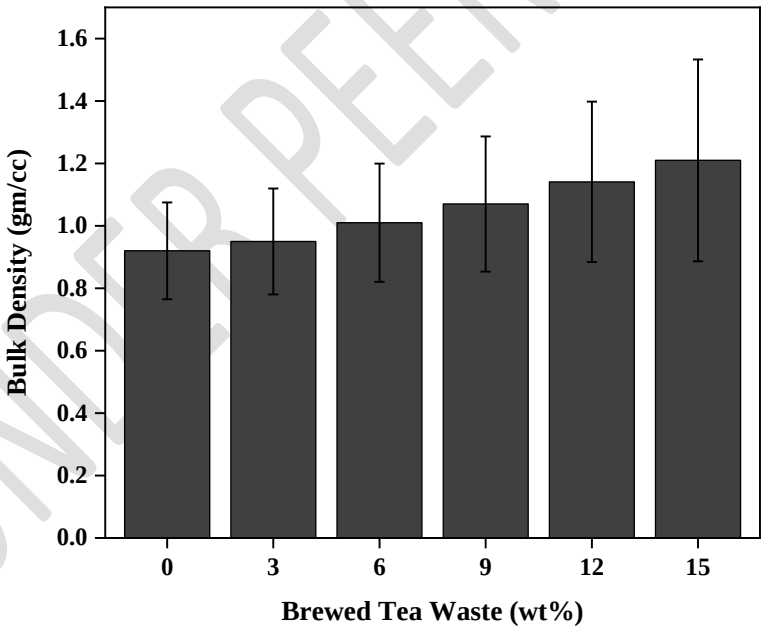


Figure6 The variations of the bulk density as a consequence of the filler content of jute polyester composites.

3.2.2 Water Absorption

The water absorption test seems to be an essential test for determining a material's degradation rate in moist environment. Figure 7 depicts the proportion of water absorbed by

the brewed tea waste-jute polyester composites. It is considered that the water absorption of the composites increased steadily over time. The greatest value, 4.62%, was observed in the jute polyester composites reinforced with 15wt% of brewed tea waste filler; the lowest value, around 3.80%, was observed in the 0wt% filler. After 6 days of immersion the water absorption of the composite samples remained constant and reached to its saturation point. The majority of natural fillers have almost identical structures and components. One of the biggest issues with using these filler is their susceptibility to moisture. Because of cellulose and hemicellulose compose the majority of natural fillers, the molecules that comprise the cell wall of those fillers are able to form hydrogen bonds with one another.The ratio of hydroxyl (OH) to carbon (C) in cellulose and hemicellulose is quite high. Additionally, cellulose has an area that is very crystalline and may not be accessible to water molecules. Water molecules, however, enter the amorphous areas of cellulose and hemicellulose and dissociate intermolecular hydrogen bonds there[33,34].

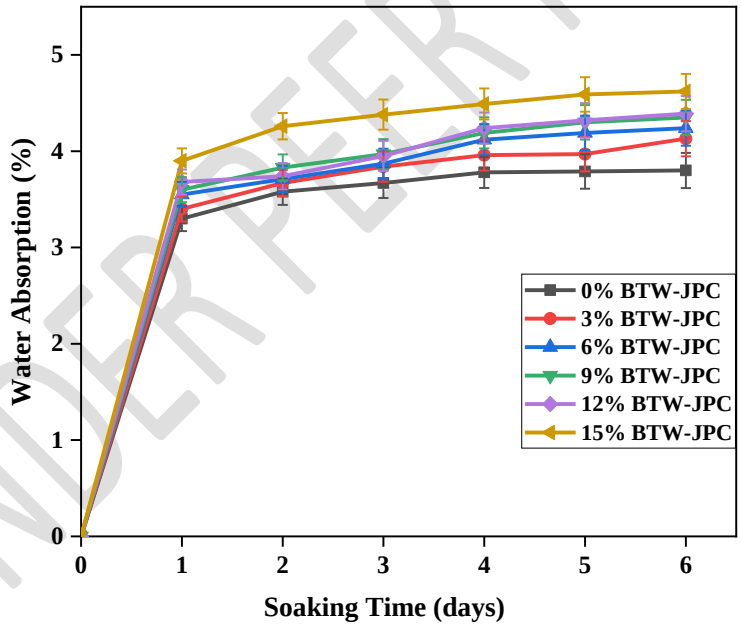


Figure7Variation in water absorption rate as a function of time for varied compositions of brewed tea waste - jute polyester composites.

3.2.3 Soil Degradation

The soil degradation of brewed tea waste- jute polyester composites have been measured by using the weight loss after 20 weeks. Figures 8 shows the soil degradation of various composites as a function of burial time. According to the graph, all composites exhibit an accelerated rate of deterioration during the buried process. At 20 weeks, a composite with a 15wt% filler had the most weight loss, at 25.34%, while a composite with a 0wt% filler experienced the least, at 17.14%. This is probably because moisture got into the composites and caused the surfaces and interfaces to break down. In addition, given that natural fibers and filler are often subjected to attack by microbes, are responsible for the reduction in the mechanical properties of materials[35].

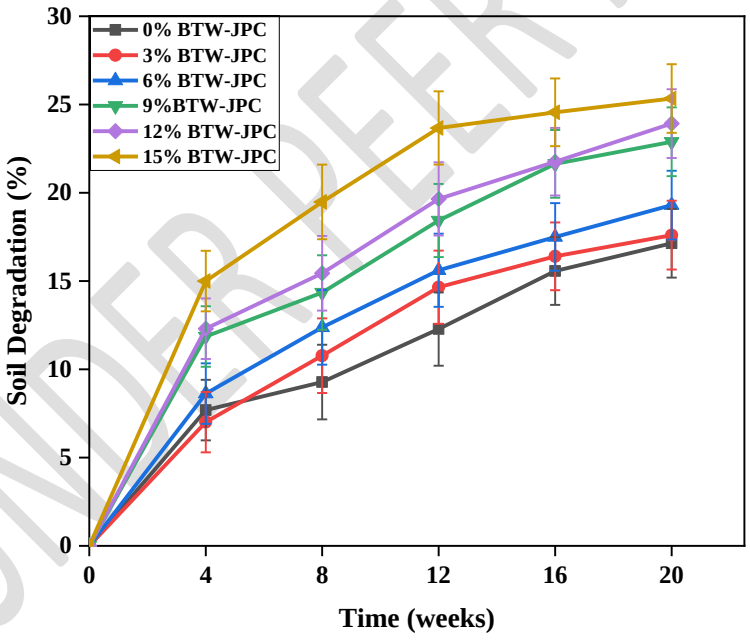


Figure8Variation in soildegradation rate as a function of time for varied compositions of brewed tea waste - jute polyester composites.

3.3 Structural Properties of Composites

3.3.1 FTIR Analysis

FTIR analysis of brewed tea waste (BTW) powder is illustrated in Figure 9.Transmittance measurements have shown a variety of prominent peaks, most notably at the wave number 3150 cm^{-1}.This peak corresponding to O-H stretching, because of the presence of alcohols and phenols in BTW powder. The C=O stretch of acidhalaide is involved for the small peak at 1840 $cm^{-1.}$ Additionally, the N-H bend and O-H bend peaks at 1625 cm^{-1} and 1400 cm^{-1} include valuable information on the amine and carboxylic acid group respectively. The other noticeable peak at 1050 cm^{-1} is due to the C-O stretching of alcohol. The detected peaks are in excellent agreement with earlier research by Hayeemasae et al. [36].

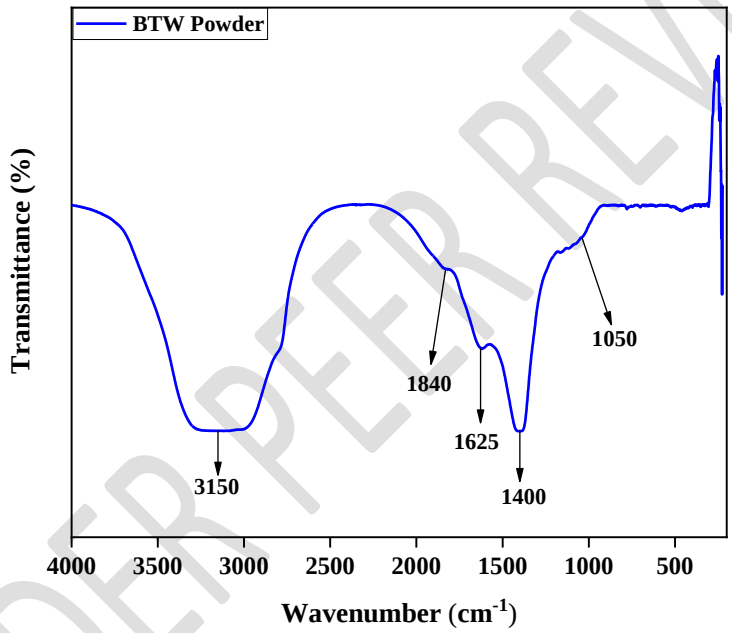


Figure9FTIR spectrum of brewed tea waste powder.

Figure 10 depicts the FTIR spectra of five distinct tea waste filler compositions of BTW-JPC.In the 3130 cm^{-1} broad absorption band because of O–H stretch and H–bonded structure, found in primary functional groups of phenols, alcohols, and water. It was determined that the C-O stretching bond structures, which comprise a functional group of aldehyde and ester, are responsible for the small peaks at 1736-1742 cm^{-1}.These characteristic peaks of C-O shifts because of the interaction of filler and matrix. A medium band at 1404 cm^{-1} is described as the functional group of carboxylic acid and alcohol group in a O-H bending structure. This peak move from 1404-1410 cm^{-1} due to the reaction between reinforcement and matrix.The other major peaks owing to C-O stretching of alcohol and C-H bending at 1080 and 745, 700 cm^{-1} respectively[27].

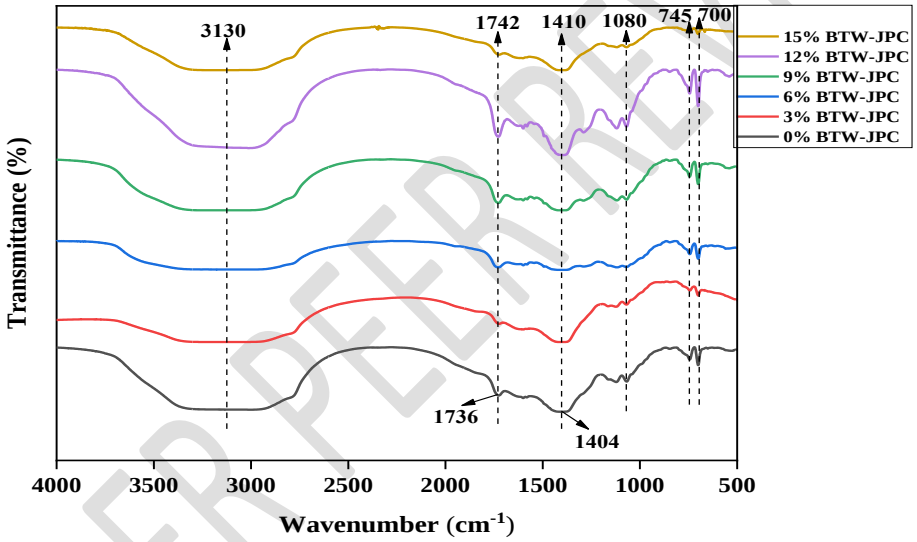


Figure10FTIR spectra of different brewed tea waste-jute polyester composites.

3.4 Thermal Properties of Composites

3.4.1 Thermogravimetric analysis (TGA) and Differential Thermal Analysis (DTA)

The TGA and DTA graphs for brewed tea waste powder and brewed tea waste –jute polyester composites are presented in Figure 11and 12. In the first stage,the range of temperatures that corresponded to 30 to 250 °C was responsible for the weight loss of brewed tea waste powder, which was caused by the evaporation of water at the low temperature in the first stage. The second stage was considered as devolatilization stage from 250 to 540°C in which the greater decomposition of volatiles that resulted from the breakdown of organic substances such as hemi-celluloses, partly lignin, and other molecular compounds, in addition to the reduced fixed carbon content of the sample. The last stage (> 540 °C) seemed to be the result of the gradual thermal decomposition of residuals such as chars, minerals, and ash, which constituted the remaining solid residues.

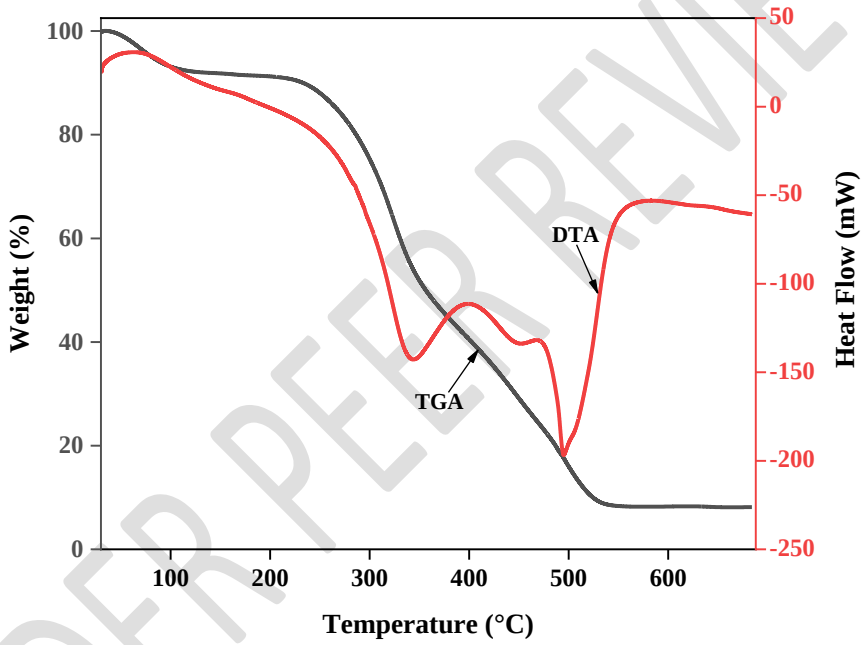


Figure 11TGA and DTA curves of brewed tea waste powder.

From DTA curve of brewed tea waste powder, it can be seen three endothermic peaks at 350,450 and 500 °C. The first peak describes the loss of moister and second and third peaks wasdue to the degradation of polymeric substances[37,38].

Fromthe Figure 12, it may observed that early weight loss happened between 50 to 270 °C. This reflected the loss of moisture or solvents from the composite. At 270 °C and above, the composite decomposed at a quicker pace, with the 15wt% composites decomposing at a somewhat higher temperature than the 0wt% composites. During the second stage, which occurs between 270°C and 500°C for 15wt% composites and 270°C and 450°C for 0wt% composites, the composite drops significant weight and three primary components

(hemicelluloses, lignin,cellulose) of the composites were degraded in this second stage of deterioration. The third stage describe the degradation of residual elements. Using the steep downward slope, it can be observed that treated 15wt% composites exhibit less weight loss at higher temperatures than 0wt% composites. This clearly showed that the 15wt% composite was more thermally stable than the 0wt% version.

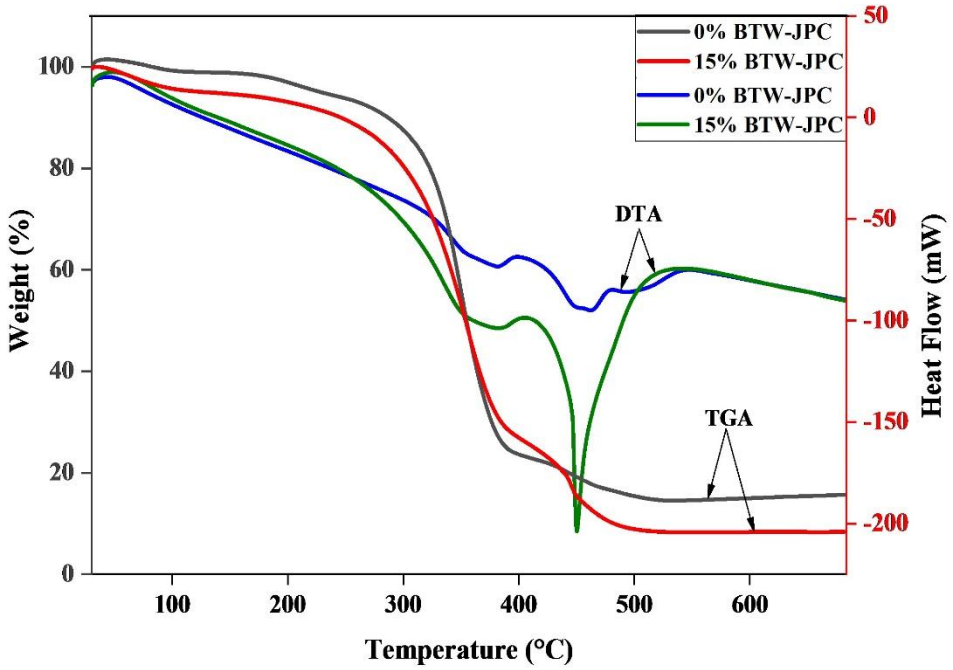


Figure 12TGA and DTA curves ofdifferent brewed tea waste-jute polyester composites.

For brewed tea waste-jute polyester composites, the first endothermic peak at temperature ranged from 380°C owing to moisture, solvents, hemicelluloseof the composites. Second endothermic peak occurred at 450°C for composites.Lignin,cellulose and other solid residues in the composites were degraded in this second stage of degradation. As can be seen from their peaks in Figure 12,15wt% composites had greater decomposition temperatures than the 0wt% composites[39,40].

4 Conclusions

In this study, natural filler brewed tea waste was reinforced with jute polyester composites by hand lay-up technique in varied weight percentages.The mechanical, physical, structural, thermal andmorphological characteristics of composites wereanalysed.According to the observations, the maximum tensile strength, flexural strength and tensile modulus were 25.45 MPa, 37.56MPa, and 1463 MPa respectively found for the 6wt% brewed tea waste

composites (BTW-JPC) in the experiments. Further increase in filler content decreased the mechanical strength.The degree of elongation was at maximum when the composite had 0wt% of filler, and then it began to drop as the filler content increased in BTW-JPC. Despite this, the Rockwell hardness of the composites steadily improved when the filler amount was increased. Based on the findings of this investigation, the composites with a 15wt% filler content had the highest bulk density. With the addition of filler, water absorption and soil degradation both increased, with composites at 15wt% filler achieving the highest value in both situations.Following the outcome of the FTIR investigation, there was good interaction between the polyester resin, jute fabric and the tea waste filler.TGA and DTA both showed that the 15wt% composites exhibit higher thermal stability and thermal decomposition temperature than 0wt% composites.

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