

# Physico-Mechanical Properties of Industrial Tea Waste Reinforced Jute Unsaturated Polyester Composites

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## ABSTRACT

The industrial tea waste reinforced jute polyester composites (ITW-JPC) were prepared by hand lay-up method for six different wt% (0%, 3%, 6%, 9%, 12%, and 15%) at 115°C temperature. The effect of industrial tea waste filler on mechanical, physical, structural, and thermal properties in jute polyester composites were evaluated. It is found that tensile strength and flexural strength improved continuously with increasing filler loading up to 9wt% but decreased at 12wt% due to weak interfacial bonding and irregular distribution of filler and matrix. The maximum value of elongation at break (%) and Rockwell hardness were found in 0wt% and 15wt% composites respectively. The hardness increases when the resistance of the materials to the deformation increases. It is seen that water absorption and soil degradation are enhanced for all composites with the accumulation of filler content and time. The structural examination and functional group identification were investigated by using Fourier Transformation Infrared (FTIR) analysis. Thermal analysis of ITW-JPC showed that thermal degradation of composites started almost at the same time and the degradation of composites was occurring in three stages. Surface morphology and interfacial properties such as internal cracks, and fiber pull-out were examined through scanning electron microscopic (SEM) analysis.

Keywords: Tea Waste, Unsaturated Polyester Resin, Jute, Mechanical Properties, Bulk Density.



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## 1 Introduction

The industrialization has been a boon to the global economy, but it has come at a high cost. Unchecked and irresponsible development have disastrous effects, with our natural resources becoming contaminated to unimaginable degrees. As a result, developing and using biodegradable materials are necessary. The natural fiber is the first thing that comes to mind when considering biodegradable composites. The use of these fibers as reinforced materials improves biodegradability, lowers costs, and minimizes pollution and hazard to the environment. The use of natural fibers as reinforcement in both thermoplastic and thermosetting matrix composites, from renewable sources, gives favourable benefits in terms of ultimate disposability and utilization of raw materials. The focus of the scientist is now on developing polymer matrix composites reinforced with jute, coir, pineapple, sisal, etc. primarily to lower the price of raw materials [1].

Jute is considered to be a potential natural reinforcing material since it is comparatively cheap and commercially accessible in the necessary form among all-natural fibers. It has higher strength and modulus than plastic and may be used to replace traditional fibers in a variety of applications [2].

Unsaturated polyester (USP) resin is broadly applied as thermosetting resins across the world. They are a very viscous liquid polymer, and due to the existence of unsaturation, further cross-linking is feasible in polymer, which makes them hard. By mixing the resin with various reinforcements, fillers, and curing it with free-radical initiators, a wide range of thermoset products with a variety of mechanical and chemical characteristics may be fabricated [3].

Tea is the world's second most popular and widely consumed beverage after water [4]. They are known for their distinct flavours and antioxidant content, which can have health benefits. Bangladesh is the world's tenth-largest tea grower, and tea is the country's second most significant export cash crop after jute [5]-[6]. For the years 2019-2020, Bangladesh's total tea output and internal demand are 86.39 million kg and 57.06 million kg, respectively [7]-[8]. The majority of tea companies do not follow the Tea Board of Bangladesh's regulations for tea waste disposal. Every tea factory generates a significant amount of tea waste, yet tea waste buyers are limited in this region. As a result, a large volume of unusable tea waste is created.

Tea waste refers to the trash produced by tea manufacturers. Tea leaves, buds, and young stems of tea plants are among the waste. If tea waste is not properly disposed of, it can contaminate the environment, including the land, water, and air [9]. As a result, the focus of this research is on industrial tea waste, which will be referred to as tea waste in the article.

Tea waste has the same compounds as tea, with polyphenols and caffeine being the most significant substances in tea. Tea waste contains antioxidants, cellulose, catechins, flavanols, amino acids, non-soluble proteins, fiber, sugars, lignin, zinc, and tannic acid, all of which contribute to the flavor, texture, and nutritional value of tea. These trace elements are obtained by plants from growth mediums such as fertilizer solutions and soils. Instead of burning or burying the waste, it may be readily extracted, mixed, and shaped to generate value [10]-[12].

Tea waste is a lignocellulosic material that is likely to be utilized as a natural reinforcement or composite filler due to its advantages over inorganic materials, including fast recyclability, low energy, and low cost. Tea waste is one of the natural fibers

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that may be used as fillers in polymer composites with unique properties, both from an environmental and economic standpoint [13].

Furthermore, Tea waste is a plentiful resource that attracts researchers from a variety of areas due to its numerous adaptable characteristics. This is due to the increased usage of tea waste as nutrients, adsorptive membranes, animal feeds, fertilizer, biomass for fuel, bio-char, and bio-oil among other things [14]. In previous studies, Atiqah et al. investigate the mechanical characteristic of poly (lactic acid) (PLA) filled with household tea wastes (TW) green composites [15]. Zhang et al. synthesized amino hybrid biopolymer-decorated magnetic biochar composites derived from green tea waste [16].

Vempaty et al. developed and evaluate the functionalized tea waste ash-clay composite [17]. Meena et al. fabricated polyaniline-coated porous and fibrous nanocomposite with granular morphology using tea waste [18]. Majid et al. developed tea waste/kapok fiber composite [19], Wesley et al. fabricated Tea waste biochar composite with nickel phthalocyanine as a potential supercapacitor electrode material [20].

However, considerable attention was not paid to investigating the characteristics of industrial tea waste filler-based USP polymer composites. In addition, no research has been done to see how tea waste filler affects the properties of jute polyester composites. In this study, the different techniques for utilizing tea wastes and their proper management by using as filler in polymeric matrix composites were explored and the influence of industrial tea waste filler on the mechanical, physical structural, and thermal properties of jute polyester composites was investigated.

## 2 Experimental

### 2.1 Materials and Methods

For the present research, the USP resin was purchased from (3262P-DC, SHCP, Singapore) and was used as a matrix material. Methyl ethyl ketone peroxide MOLPEROX F60, Turkey) was applied as a hardener within the matrix. The woven jute fabric mat (collected from the local market, Rajshahi) and industrial tea waste (collected from Bangladesh Tea Research Institute, Sylhet) were used as reinforcement. Sodium hydroxide, NaOH (Merck, India) was used for the treatment of jute mat.

### 2.2 Preparation of Industrial Tea Waste Powder

The collected industrial tea waste was cleaned with excessive amounts of distilled water to remove non-cellulosic components and then dried for 48 hours at 60°C to reduce water content before being ground in a blender. Finally, it was hand-sieved to obtain the fine tea waste powder of 1-250 micron.

### 2.3 Treatment of Jute Mat (JM)

In this research NaOH solution at a concentration of 5% was used to treat the jute mat. The treatment was done for 1 hour at room temperature, although it has been observed that overtreatment of natural fibers with NaOH might have a detrimental effect on the base fiber characteristics, to neutralize the NaOH solution, the jute mat was cleaned multiple times with distilled water and dried at 60°C for 24 hours [21]-[22].

### 2.4 Fabrication of Composites

The composites were fabricated using a hand lay-up technique [23]-[24]. Before putting the matrix material on the jute mat, it was blended in a magnetic stirrer hot plate thoroughly with USP resin, industrial tea waste powder, and 1% MEKP to obtain a homogenous solution. One layer of jute mat was sandwiched between two layers of mixed matrix solution to make the composites. The weight ratios of industrial tea waste filler varied between 0, 3, 6, 9, 12, and 15 wt%, while the weight ratio of jute fabric remained fixed at 10 wt % in composites. A steel mold with dimensions of 150x120x5mm was made. Silicon spray is applied to the inner wall of the mould for the easy ejection of composites. The mix matrix solution was applied to the mold. The jute mat was then placed on top of the mixed matrix, and the jute mat was appropriately rolled over with a roller to remove the air bubbles from the composite matrix mixture. Again, applied the mixed matrix layer on the jute mat and closed the mold. A hydraulic lamination heat press machine was used to press this sandwich at 115°C for 20 minutes under a pressure of 5 tons, resulting in composites. After that, the composites were allowed to cool to ambient temperature. Finally, the composites were separated from the two steel molds, cut to size according to ASTM requirements, and stored in desiccators.



Fig. 1 Industrial tea waste powder (a) before preparation (b) after preparation

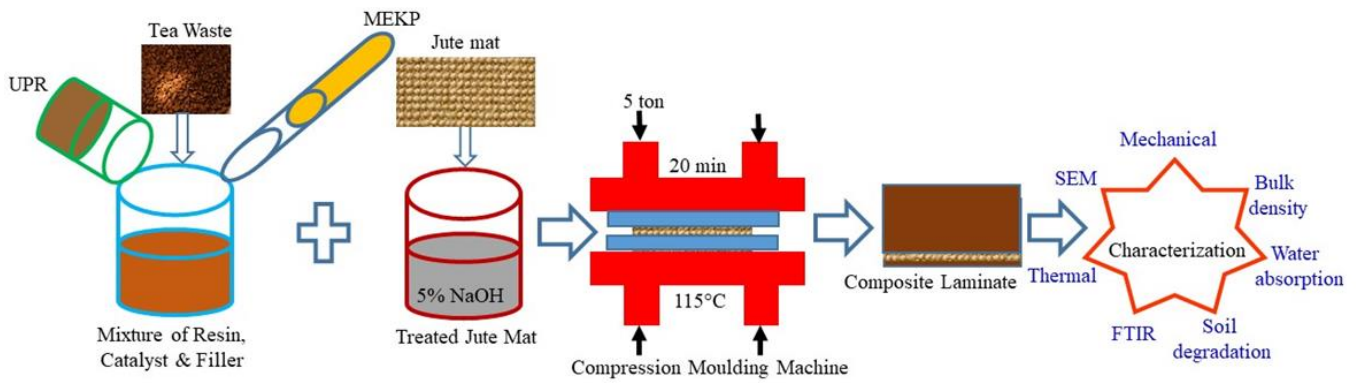


Fig. 2 Schematic illustration of the fabrication process and characterization proposed in this study

## 2.5 Characterization

Tensile, flexural, and Rockwell hardness tests of the manufactured neat jute polyester (0wt% filler) composites (JPC) and industrial tea waste-jute polyester composites (ITW-JPC) were investigated. Five specimens were prepared for each test, and the average values were calculated. The tensile test was performed using a Universal Testing Machine (WDW-50, Serial No- 180067, China) according to ASTM D638. For the flexural test, the specimens were prepared following the ASTM D790-98. Each test was carried out until the failure occurred. The hardness of the prepared composites was measured using (HR-150DT Rockwell Hardness Tester, China) according to ASTM D785. The hardness test is performed with a diamond indenter (Rockwell C scale) with an applied load of 100. The bulk density of the samples was measured according to the ASTM C-134-76 and the bulk density of the specimen was calculated using the following formula:  $D = W_s/V$ , where  $D$  is the bulk density,  $W_s$  is the weight,  $V$  is the volume of the sample. Water absorption of the composites was measured according to ASTM: C-67-91. The test specimens were cut in a size of 6 cm in length, 2 cm in width, and 0.5 cm in thickness. The samples were dried for 24 hours at around 80°C in the oven, then cooled in a desiccator and immediately weighed with a microbalance. Then it was placed in a jar of distilled water (25°C) for 24 hours [21]. The specimens were then removed one at a time, cleaned with tissue paper, and instantly weighted using a microbalance. Let the initial weight be  $W_i$  and after water absorption the weight is  $W_f$ . Then the amount of water absorption was calculated by the following formula:  $\text{Water absorption (\%)} = [(W_f - W_i)/W_i] \times 100\%$ . The biodegradability of the composites was investigated using a soil burial test in the laboratory, as reported by Goudar et al. [25]. The fresh soil was obtained from the Rajshahi University Campus garden in Rajshahi, Bangladesh. To obtain the initial dry weight, the samples were cut into  $2 \times 2 \text{ cm}^2$  and dried at 40°C ( $W_1$ ). The samples were buried in the soil at a depth of approximately 8–10 cm below the surface. Spraying water on the soil's surface kept the moisture in the soil from evaporating. After 4 weeks, the weight of the composites was measured by taking them from the soil, washing them, and then drying them in an oven ( $W_2$ ). The Soil degradation was calculated using the formula:  $\text{Soil degradation (\%)} = [(W_1 - W_2)/W_1] \times 100\%$ . The chemical composition of the industrial tea waste (ITW) powder, JPC, ITW-JPC, and the bonding nature of matrix and reinforcement inside the composites were investigated using (Perkin-Elmer Frontier FTIR/MIR Spectrometer, USA). The transmittance range of the scan was  $4000 \text{ cm}^{-1} - 225 \text{ cm}^{-1}$ . Using a DTA/TGA (Perkin Elmer Simultaneous, STA-8000, USA),

thermal Analyzer, melting and degradation temperatures of ITW powder, JPC, and ITW-JPC were studied. Under nitrogen gas flow, the tests were carried out from 30 to 700°C at a heating rate of  $20^\circ\text{Cmin}^{-1}$ . The melting and degradation temperatures are derived from the exotherm vs temperature curves on the DTA traces. JEOL USER 7610F Scanning Electron Microscope, Japan was used to evaluate the surface morphology and filler's dispersion of the composite samples.

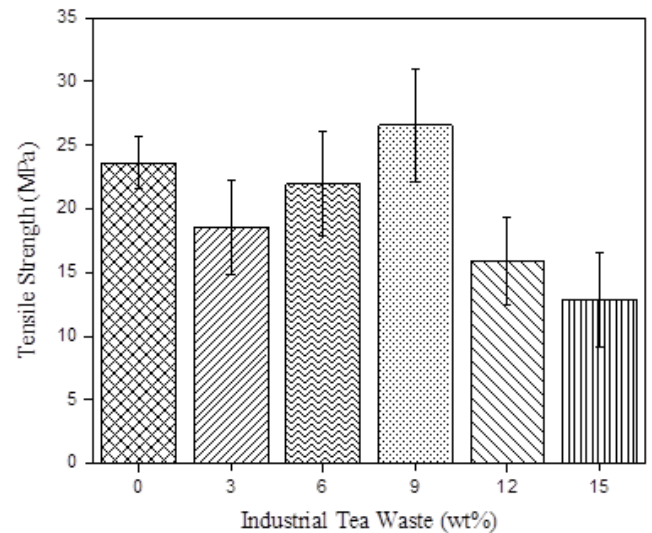


Fig. 3 Effect of filler loading on tensile strength of industrial tea waste-jute polyester composites.

## 3 Results and Discussion

### 3.1 Mechanical Properties of Composites

#### 3.1.1 Tensile Strength

The tensile strength of industrial tea waste-jute polyester composites for different wt% of filler loading is represented in Fig. 3. This figure shows that the tensile strength of the prepared composites is decreasing drastically with the addition of industrial tea waste filler in composites. It was observed that tensile strength decreased sharply for 3wt% and 6wt% composites, however, after 6wt% composition tensile strength increased slightly for 9wt% composition then the value decreased again for further addition of filler content. The maximum tensile strength was obtained for 9wt% filler content composite with a measured value of 26.58 MPa, whereas the tensile strength was only 23.62 MPa for pure (0wt% filler) jute polyester composite. The improved tensile strength was found due to the good



interface and strong bonding between the tea waste filler and resin matrix. However, further increase in filler content, a decrease in tensile strength was observed. This is because the higher volume of industrial tea waste promotes agglomeration that initiates cracks within the USP resin matrix. As a result, proper stress transfer is hampered and lowers the tensile strength [26]. According to Neher et al., tensile strength dropped until it reached 10wt% sawdust content, after which it began to rise (up to 15wt%). After 15wt% it again decreased up to 20wt% compositions [21]. Rahman et al. also showed an increase in tensile strength with increasing filler content up to 10wt% and further it decreased in addition of filler [27]. It was surprising to see how well our findings were supported by both pieces of literature.

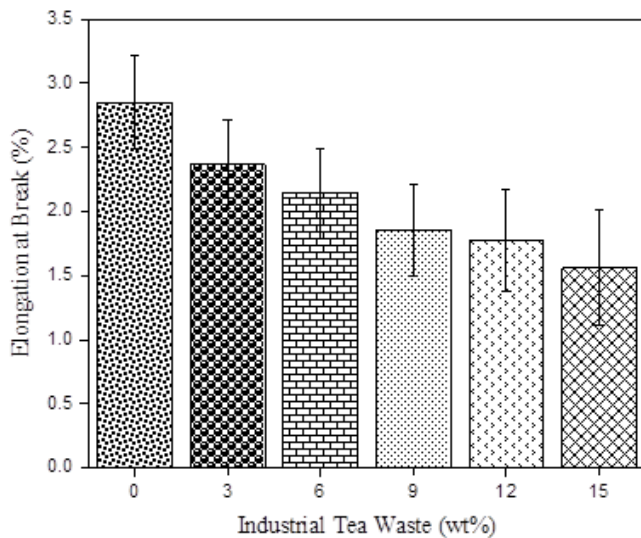


Fig. 4 Effect of filler loading on elongation at break of industrial tea waste-jute polyester composites.

### 3.1.2 Elongation at Break

The elongation percentages at the break of industrial tea waste and jute fabric reinforced polyester composite specimens are presented in Fig. 4. The finding shows that the pure jute polyester composite exhibit higher elongation at break than the corresponding value of industrial tea waste- jute polyester composites at any tea waste content. The elongation at break showed a decrease as the filler content increases from 0 to 15 wt% in the composites. This is due to the hindrance by filler to molecular mobility or deformability of polyester matrix [27]-[28]. The elongation at break for 3 wt%, 6 wt%, 9 wt%, 12 wt% and 15 wt% filler loading were 2.37 %, 2.14 %, 1.86%, 1.78%, and 1.56% respectively. The results agreed with the trend line of elongation at the break of bio-composites observed by other researchers [29]-[30].

### 3.1.3 Flexural Strength

Fig. 5 shows the flexural strength of jute polyester composites with different wt(%) of industrial tea waste. From the figure, it was observed that flexural strength decreased drastically with 3wt% filler content. Then it was increased for 9wt% filler and again was decreased for 15wt% filler reinforced composites. For the pure jute polyester composite sample the flexural strength was 34.23 MPa, for 3wt%, 6wt%, 9wt%, 12wt%, and 15wt% filler content in composites was 26.57 MPa, 31.97 MPa, 40.58 MPa, 26.94 MPa, and 22.83 MPa respectively.

Flexural strength value was decreased due to the agglomeration and improper dispersion of filler within the matrix [31]-[32].

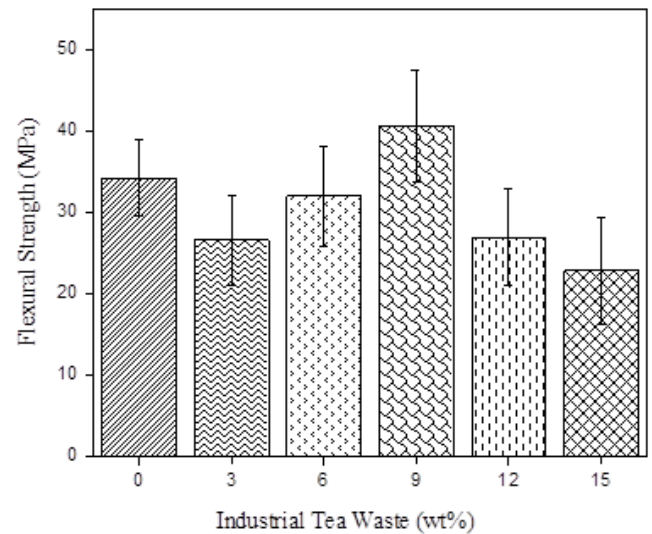


Fig. 5 Effect of filler loading on flexural strength of industrial tea waste-jute polyester composites.

### 3.1.4 Hardness

Rockwell hardness for different wt% of industrial tea waste filler loading in jute polyester composites is shown in Fig. 6. With the addition of filler in jute polyester composites, the hardness of the composites increased gradually. However, after a 0% combination with the increase of the filler loading Rockwell hardness for other combinations was increased. Moreover, it was also seen that industrial tea waste -jute polyester composites are harder than pure ones. The optimum hardness for the composites was obtained at 15wt% filler. The findings also agreed as reported in Sivarao et al. [33], Cao et al. [34]. The hardness increases when the resistance of the materials to the deformation increases. This happens when more filler is added; the composite becomes harder and the materials' hardness improves. The filler layer provides greater resistance to plastic deformation in the filler's transverse direction [33].

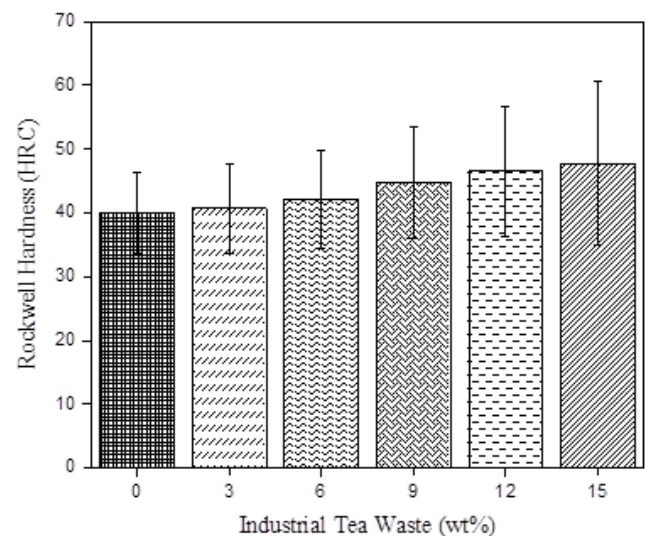


Fig. 6 Effect of filler loading on Rockwell hardness of industrial tea waste-jute polyester composites.

### 3.2 Physical Properties of Composites

#### 3.2.1 Bulk Density

Fig. 7 shows the effect of variation of industrial tea waste filler in the bulk density of jute polyester composites. The bulk density of the composites was increased with the increment of tea waste filler. The figure indicates that bulk density varied in the range of 0.92 -1.24 gm/cc. The highest bulk density was found for 15wt% of industrial tea waste-jute polyester composites. Due to the presence of pores in composite, the increment of filler loading mass increased more rapidly than its volume, which therefore increases the overall density of composites. Bulk density increment means the composite becomes denser. Jahan et al. also showed a similar kind of observation [28].

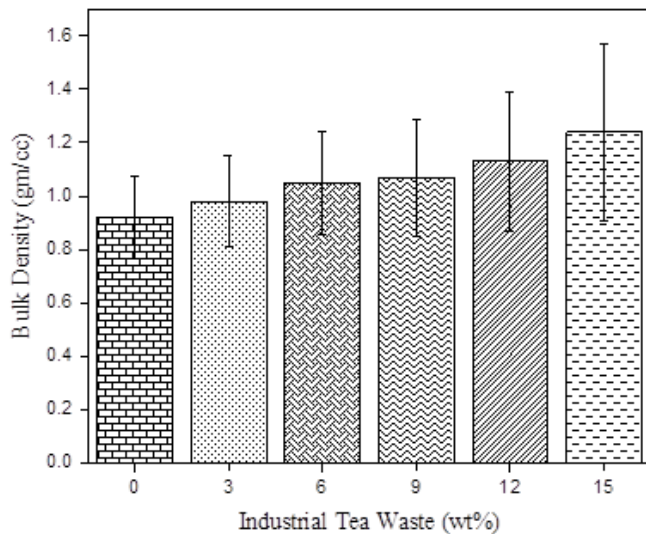


Fig. 7 Effect of filler loading on bulk density of industrial tea waste-jute polyester composites.

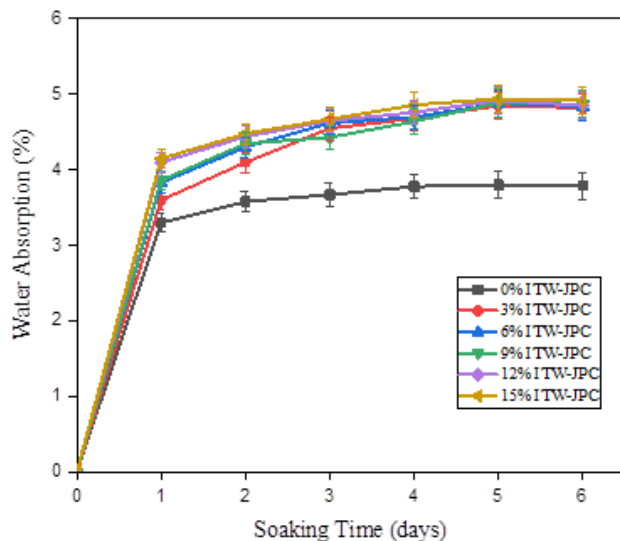


Fig. 8 Water absorption of different compositions of industrial tea waste-jute polyester composite as a function of time.

#### 3.2.2 Water Absorption

The water absorption test is crucial for evaluating the degradability of materials under wet conditions. The water absorption percentages of industrial tea waste-jute polyester

composites are shown in Fig. 8 for different soaking times. Water absorption increased with increasing weight percent of industrial tea waste filler. The jute polyester composites that are reinforced with 15 wt. % of industrial tea waste filler had the highest value which was 4.92% on the other hand, the lowest figure was found in the reference sample, which was about 3.3%. This was due to the availability of more hygroscopic cellulose and hemicellulose in the tea waste which promoted moisture absorption from the surroundings and increased water absorbance. The water absorbance (%) of the composites depends on the water absorption properties of the reinforcing fibers and fillers and the degree of matrix-reinforcement adhesion. Natural fibers containing hydroxyl (–OH) group in their chemical composition has the tendency to absorb water quickly [35]. Marvin et al. showed the increase of water absorption in particleboard made from tea waste and wood particles [36].

#### 3.2.3 Soil Degradation

The industrial tea waste-jute polyester composites were exposed to soil degradation at ambient conditions for up to 20 weeks. In Fig. 9, it is clearly shown that the mass loss of the composites was increased slowly with the extent of degradation time. After 20 weeks the maximum mass loss occur with a value of 28.34% for 15wt% of composites and the lowest value was 17.14% for 0wt% of composites. It is already reported that natural fibers and fillers are hydrophilic in nature. Water entered the fiber and filler edges of the composites during their immersion in the wet soil medium, and thus degraded the fiber and filler slowly inside the composites. Furthermore, since degradable fibers and filler are often attacked by microbial activity, bacterial action may be to blame for the loss of mechanical characteristics [37].

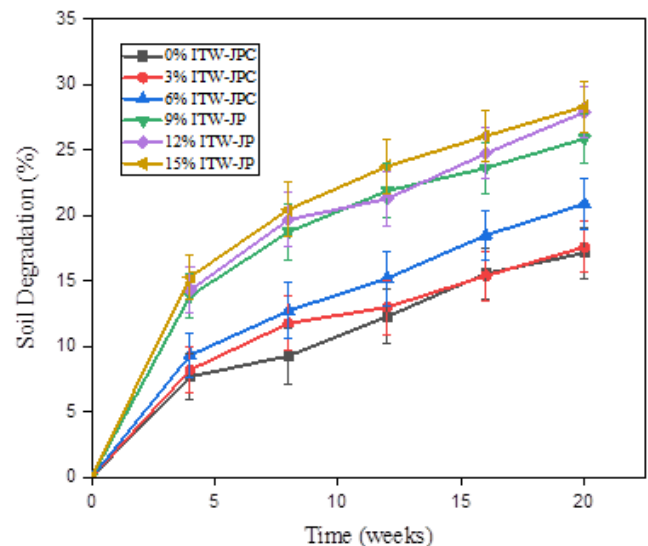


Fig. 9 Soil degradation of different compositions of industrial tea waste-jute polyester composite as a function of time.

#### 3.2.4 Fourier Transform Infrared (FTIR) Spectroscopy Analysis

The FTIR spectra of industrial tea waste (ITW) powder and industrial tea waste – jute polyester composites with 0wt%, 3wt%, 6wt%, 9wt%, 12wt%, and 15 wt% of industrial tea waste content are shown in Fig. 10 and Fig. 11 respectively. In the spectrum of industrial tea waste powder, the absorption bands around 3258 cm<sup>-1</sup> was due to stretching vibrations of O–H groups

in water, alcohol, and phenols and N–H stretching in amines. The peaks at  $2995\text{cm}^{-1}$  was associated with the C–H stretching in alkanes. The strong band at  $1621\text{ cm}^{-1}$  was attributed to the C=C stretch in the aromatic ring and C=O stretch in polyphenols. The other prominent peaks were due to S=O stretching, C–N stretching  $1415$ , and  $1095\text{cm}^{-1}$  respectively [38]–[39].

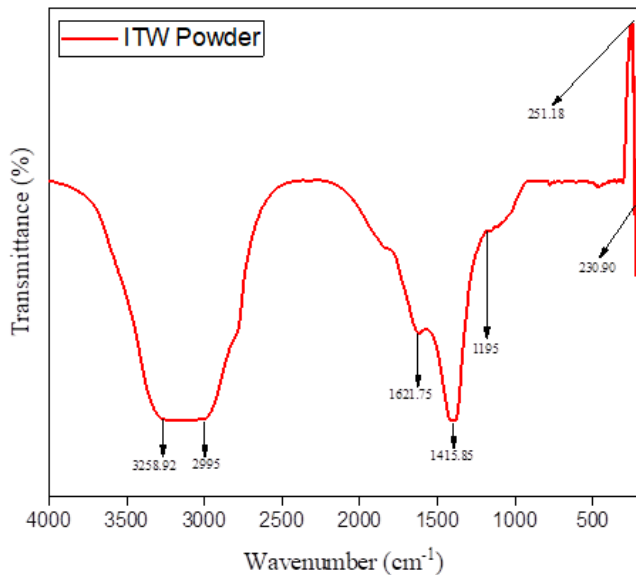


Fig. 10 FTIR Spectrum of industrial tea waste powder.

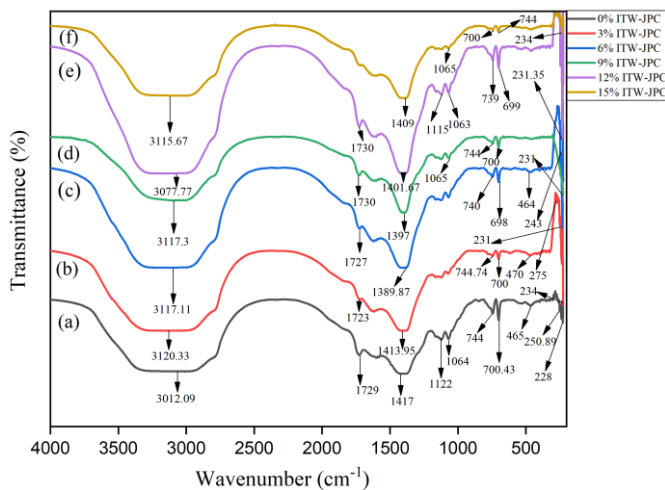


Fig. 11 FTIR Spectrum of industrial tea waste-jute polyester composites with 0, 3, 6, 9, 12, and 15 wt % of industrial tea waste content.

As it has been seen, the FTIR spectrum of industrial tea waste powder, and ITW-JPC composites are different. The peak shape of ITW-JPC composites with increasing tea waste content, was nearly the same because the functional groups remain the same even as the filler weight percent rises. For the ITW-JPC composites, 3000-3150  $\text{cm}^{-1}$  was assigned to the C-H asymmetric stretching vibration from USP resin. The peaks at 1720-1730  $\text{cm}^{-1}$  was attributed to the C=O stretching in ester. The C=O was observed in ITW-JPC composites due to the covalent bonding in industrial tea waste having taken place through an esterification reaction between filler OH and USP COOH groups. The other major peaks were due to O-H bending, C-O stretching, and C-H bending 1389-1410, 1065-1125 and 700-744  $\text{cm}^{-1}$  respectively. The results agreed with the trend line of

FTIR spectra of tea waste-based composites observed by other researchers [13].

### 3.2.5 Thermal Properties Analysis

**Fig. 12** and **Fig. 13** show the Thermogravimetric Analysis (TGA) and Differential Thermal Analysis (DTA) curves of industrial tea waste powder and industrial tea waste-jute polyester composites respectively. The decomposition process consisted of three regions. They were 30-300°C, 300-500°C, and 500-600°C, respectively. In the TGA curve, the first weight loss indicated the evaporation of absorbed water. The second stage consisted of the decomposition of polymers like hemicellulose, cellulose, and partial lignin and the third stage indicated the decomposition of all the residue from the second stage.

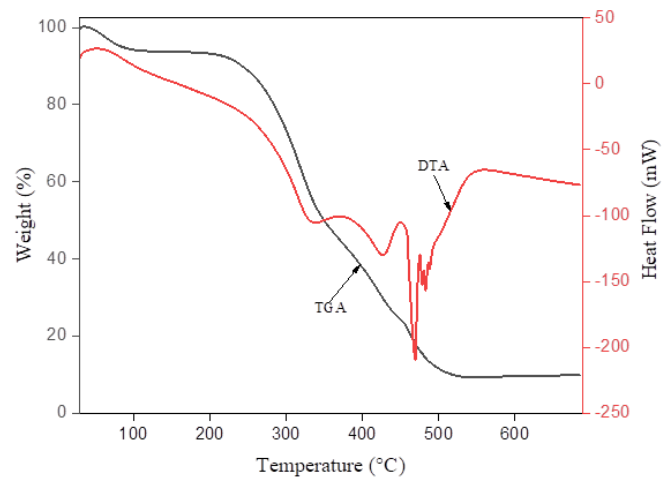


Fig. 12 TGA and DTA curves of industrial tea waste powder.

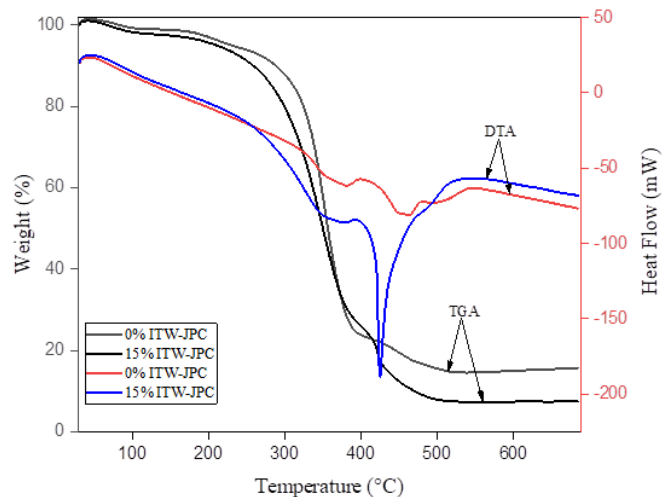


Fig. 13 TGA and DTA curves of industrial tea waste-jute polyester composites with 0 and 15wt % of industrial tea waste content.

DTA curves of ITW powder and ITW-JPC showed three endothermic peaks at 330-370°C, 420-460°C, and 490-540°C to remove moisture, the second peak was due to lighter material and the third peak corresponded to major degradation respectively [40]-[41].

### 3.2.6 Scanning Electron Microscopy (SEM) Analysis

The cross-sectional views of the fabricated composite material consisting of jute fabric, USP resin, and different wt%



of industrial tea waste, are presented in Fig. 14 (a) and (b). The SEM images were taken to observe the interfacial properties, internal cracks, and the internal structure of the fractured surfaces of composite materials. These figures show the SEM photographs of surfaces of different composite materials investigated in the present work fractured under the tensile loading. The figures clearly indicated that there was a considerable difference in the fiber-matrix interaction between neat jute polyester and industrial tea waste reinforced jute polyester-based composite. Fractographic observation suggested the fracture behaviour be brittle in nature. Fiber pull-out phenomena were observed for both the cases, but for industrial tea waste-based jute polyester composites pull-out was observed as individual fiber, but in neat jute polyester composite, there was an agglomeration of fibers into a bundle. This was a significant change of morphology which was effective for better mechanical bonding between fiber, filler, and polymer matrix. From the SEM of the industrial tea waste-based jute polyester composite, it can be clearly said that the reinforcement matrix adhesion between the jute fabric, industrial tea waste, and polyester matrix was higher with respect to the neat jute polyester composites. This may be the reason for the slightly higher mechanical properties and the smoother surface of industrial tea waste-based jute polyester composites [13],[19].

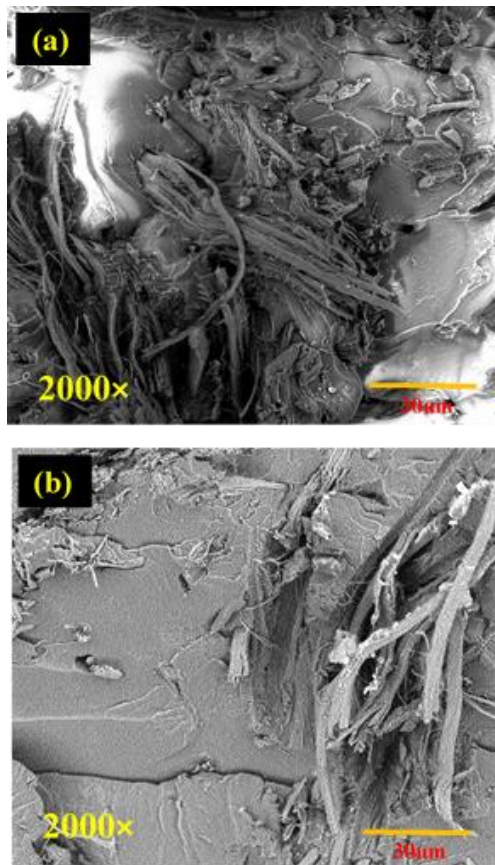


Fig. 14 SEM images of (a) neat jute polyester composite (JPC) and (b) 9wt% industrial tea waste- jute polyester composite (ITW-JPC).

#### 4 Conclusion

In the present work, the mechanical, physical, thermal, and structural behaviour of industrial tea waste-jute polyester composites (ITW-JPC) were studied. The tensile strength and flexural strength of the composites initially decreased with the

increase of the filler content but later increased with increasing filler. For tensile and flexural strength, the maximum value was obtained at 9wt% composites. The percentage of elongation was maximum for 0wt% composite, and then it decreased with the increase of the filler loading in ITW-JPC. However, the Rockwell hardness of the composites increased gradually with the increase of the filler loading. This study showed that maximum bulk density was found for 15wt% composites. Water absorption and soil degradation showed an increase with the addition of filler and the maximum value in both cases was obtained at 15wt% composites. FTIR analysis proved the presence of O-H, C-H, and C=O in the ITW powder and ITW-JPC. From TGA and DTA curves, it was found that the degradation of composites occurred in three stages. The thermal stability of 0wt% ITW-JPC and 15wt% ITW-JPC were quite similar. SEM analysis proved the presence of industrial tea waste filler led to good adhesion between the matrix and fiber, which also increased the mechanical strength of ITW-JPC when 9wt % tea waste was incorporated into the composite.

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