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# Utilization of factory tea (*Camellia sinensis*) wastes in eco-friendly dyeing of jute packaging fabrics

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ABSTRACT

Eco-friendly dyeing of jute packaging fabrics was evaluated using aqueous extraction of factory tea (*Camellia sinensis)* wastes. Jute and factory tea wastes are available in Bangladesh and jute bags are used for packaging of various exportable agricultural commodities. The extract of factory tea waste (FTW) is dark coffee colored and it was characterized by attenuated total reflection-fourier transform infrared (ATR-FTIR) and microbial analysis. Nontoxic, non-allergic and eco-friendly natural dyeing process of jute packaging fabrics using extract of FTW were developed and optimized. Metal mordants 10 % on the weight of fabric was used to get the fastness properties of dyed jute fabric. The methods of application of mordants were pre-mordanting, simultaneous or meta-mordanting and post-mordanting. The color fastness and tensile properties were measured for all jute packaging fabrics and it is found that the dyed jute packaging fabrics showed a slight decrease in tensile breaking force (N) than undyed jute packaging fabric. The highest color fastness obtained with the meta mordanting method with ferrous sulfate mordant in a shade of dark coffee. The results of the color fastness for light and washing showed an excellent value of grade 4–5.

## 1. Introduction

Coloring of textiles by natural dyes were used from ancient times to till the nineteenth century and started to decline the use of natural dyes after the invention of synthetic dyes in the second half of the nineteenth century [1]. In the textile industry, a constantly increasing interest in the revival of natural dyes has been aroused in recent years due to environmental awareness and health hazards associated with the use of synthetic dyes [1–3]. Natural dyes are considered eco-friendly as these are renewable, biodegradable, non-toxic, non-allergic and promote green revolution [1–5]. The sources of natural dyes are plant, animal, mineral and microbial origins. Dyes or colorants obtained from animal or vegetable matter without any chemical processing regarded as an ecological and sustainable but these have few draw backs such as, low colorfastness and non-availability of standard shade cards [4,5]. An inorganic process was used to extract tea dye from Longjing tea leaves, and after a variety of dying techniques was examined on cotton fabrics, it was discovered that tea dye has a significant potential market and may be used as a commercial natural dye [6].A study on the application of natural colorants derived from tea leaves for cotton fabric dyeing was conducted on Giza 86 Egyptian cotton fabric. The

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findings shown that tea leaves may be used as a dye to color fabrics, and that various chemical and natural mordants can be utilized to achieve varied color tones [7]. $Al3+$, $Cu2+$, $Cr3+$, $Co2+$, $Ni2+$, $Fe2+$, and so on are a few examples of chemical mordents. Environmental risks include substances such as $Cr3+$, $Cu2+$, $Ni2+$, and $Co2+$ [8,9]. Tea leaves were microwave-treated for 6 min in an aqueous medium to extract natural dye, which was then utilized to eco-friendly dye cotton fabric [10]. The safflower (Carthamus tinctorius L.) color source combined with biomordants (turmeric and henna) was used in a quick and environmentally friendly microwave technique for both natural color extraction and fabric dyeing [11]. The arjun bark colorant was extracted and silk fabric was dyed using a similar microwave technique [12]. When irradiated to microwave radiation, the seeds of Peganum harmala exhibit great potential for usage as a natural colorant for coloring cotton [13].

Tea (*C. sinensis*) is mostly composed of phenolic compounds, such as catechins and flavonoids and the flavonoid family contains therubigins and theaflavins, which are the principal natural dyeing agents used on natural fabrics [7,14–16].The main coloring pigment that gives fabrics their brown color is called catechin, which a naturally occurring pigment is found in tea leaves. Catechins include epicatechin (EC), epigallocatechin chin (EGC), epicatechingallate (ECG), and epigallocatechingallate (EGCG) [10,17,18]. The pigments of thearubigins are reddish-brown, whereas the pigments of theaflavins are yellowish-brown [16].

The goal of the current project is to extract colorants from FTW and utilize them to dye jute packaging materials. Jute fabric is eco-friendly and biodegradable. It can be found in many craft and decorative applications, as well as shopping bags, gift wrapping, flower arrangements, wedding decoration, food packaging etc. In Bangladesh, waste products from factories of tea plants constitute a plentiful supply of renewable natural resources. In this study, jute packaging materials were dyed using extracts from FTW. To increase the color fastness of dyed jute packaging materials, inorganic mordants were utilized as a dye fixing agent. Consequently, the items will be energy, time, and money-efficient. The characteristics and reports of extracted dyes and dyed jute packaging fabrics were investigated and provided.

## 2. Materials and methods

### 2.1. *Factory tea wastes and chemicals*

Factory tea wasteswere collected from the Sreemangal district and the jute packaging fabrics were supplied by Janata Jute Mills Ltd., Narsingdi. The quality of the jute fabric is 58 × 51/10 cm, yarn count is 10.60 lbs and weight is 425 gsm. Merck (Germany) provided the ferrous sulfate ($FeSO4$) (99 %), Copper sulfate ($CuSO4$) (99 %), sodium hydroxide ($NaOH$) ($\geq$99.8 %), and methanol ($CH3OH$) ($\geq$99.9 %).

### 2.2. *Preparation and extraction of dye from factory tea wastes*

FTW were cleaned and air-dried. Then the tea leaves were mechanically milled into a fine powder and kept in a dry, airtight container.The dye was extracted from the dried FTW and characterized. FTW (650 g) were dried and taken into distilled water (6.5 L) in a vessel and heated for 2 h at 100 °C. Extracted dye was collected by filtering and drying the filtrate. Dried extracted dye (94.25 g) was obtained from FTW(650 g).To compare the dye with water extraction, the same procedure was utilized to extract the dye using methanol. The extracted dye was characterized and stored at room temperature for dyeing the grey jute packaging fabrics.

### 2.3. *Desizing of jute packagingfabric*

There are four main types of desizing methods: oxidative, alkali, enzyme, and acid. In most cases, alkaline desizing was used [19]. To achieve the best desizing effect, a high desizing temperature is recommended. However, if better results are required, a longer term of treatment may surely be advantageous [20]. In this work, jute packaging fabric underwent alkaline desizing. Jet detergent powder (40 g), caustic soda (10 g) and soda ash (80 g)were applied to the jute packaging fabric for 3 h at 100 °C at a 1:10 jute fabric to liquor ratio. The desized jute packaging fabric was then completely cleaned with running water and allowed to soak in fresh water for half an hour. Before dying and mordanting, the desizedjute fabric was dried at room temperature.

### 2.4. *Dyeing and mordanting of jute fabric*

In this work, jute packaging fabric was dyed with extracted dyes from factory tea wastes using the conventional dyeing and mordanting procedure. There were two steps in the conventional dyeing and mordanting process: (i) one bath procedure where jute packaging fabric, dye, and mordant ingredients are all submerged in the same bath and (ii) double bath procedures. One of two methods was used for the double bath procedure: (i) dyeing first, then mordanting (post-mordanting); or (ii) mordanting first, then dying (pre-mordanting) [21,22].For each of these three pre-, meta-, and post-mordanting procedures, the same quantity of fabric, dye, and mordant was used. Out of the three processes, the meta-mordanting method produced the most vibrant shade of color in the dyed jute packaging materials.

### 2.5. *Meta-mordanting dyeing of jute packagingfabric*

To dye jute packaging fabric, the extracted dye (16 g) can be added directly to a dye bath made of distilled water (1 L). Alternatively, dried tea wastes (200 g) can be added to a dye bath made of distilled water (1 L) to dye desized jute packaging fabrics. The dye was extracted by boiling the dried FTW (100 g) for 2 h at 100 °C, followed by filtering. Ten grams of ferrous sulfate mordant were added to the dyeing bath filtrate. Now, the desized jute fabrics (100 g) were added into the dyeing bath. The proportions of dye and mordant added to the weight of the fabric (owf) were 16 % and 10 %, respectively. With occasional shaking, the temperature was raised to 100 °C gradually. The material was dyed at 100 °C for 2 h, with a fabric-to-liquor ratio of 1:10 and a pH of 5.5–6.5. After being dyed, jute packaging fabrics were rinsed with running water, allowed drying, and then had their fastness examined. To compare with dyed jute packaging fabric that had a mordant, the same process was utilized to dye jute packaging fabric without mordant.

### 2.6. *Color fastness test of dyed jute packaging fabric*

#### 2.6.1. *Washing fastness to comply ISO 105-C03:1989–12-15*

The color fastness of jute packaging fabrics that have been dyed was evaluated during washing and the results were recorded, in accordance with ISO standard.

#### 2.6.2. *Light fastness to comply ISO 105-B02:1994–09-15*

A light fastness tester, 200 Trufade, James Heal, UK, evaluated and recorded the light fastness of dyed jute packaging fabrics in accordance with ISO standard.

#### 2.6.3. *Rubbing fastness*

The jute packaging fabrics that had been colored were vigorously rubbed against an undyed jute packaging fabric, and any color transfer that occurred was noted [21].

### 2.7. *Evaluation of the color strength (K/S) of dyed jutepackaging fabric*

Based on the color fastness results, the color strength (K/S), CIEL*a*b*, and C* h values of the colored jute packaging fabrics were evaluated using a Datacolor 850 spectrophotometer (USA).

### 2.8. *ATR-FTIR spectroscopic analyses*

Using an FT-IR/NIR spectrometer (Frontier, PerkinElmer, USA), the ATR-FTIR spectra of FTW, as well as its aqueous and methanol extracted dye, were recorded. The jute packaging fabric that had been colored was also examined and its ATR-FTIR spectra recorded. As ATR-FTIR spectra, the absorbance data was obtained.

### 2.9. *Antibacterial activity of extracted dye of FTW*

Gram-positive bacteria *Bacillus subtilis, Staphylococcus aureus, Enterococcus faecalis* and Gram-negative bacteria *Salmonella typhi, Shigella flexinery, Vibrio parahemolyticus, Escherichia coli* were employed for determination of antibacterial activity of the extracted dye of FTW.

#### 2.9.1. *Tensile properties of dyed and undyed jute fabric*

Computerized universal testing instrument (Model-Titan 5, Brand: James Heal, UK) was used to perform tensile tests on both colored and undyed jute packaging fabrics. To conduct the testing, the ASTM D 5035-06 was used at 300 mm per minute speed. The specimens had respective measurements of 150 mm in length and 25 mm in width. The distance between the jaws measured 75 mm. For every composition, six to seven specimens were assessed, and the average results were computed from a maximum of five values. The instrument produced the load vs. elongation graphs.

## 3. Results and discussion

### 3.1. *Extraction and characterization of dye from FTW*

In contrast to synthetic dyes, which are produced and applied with a great deal of waste and pose a major health risk as well as disrupt the natural equilibrium of the ecosystem, natural dyes are nontoxic, nonallergenic, noncarcinogenic, sustainable, and

**Table 1**
Physical propertiesof extracted dyes.

| Sample | FTW |
|---|---|
| Physical state | Solid |
| Colour | Dark Coffee |
| Extracted dye content in water | 14.5 % (approx.). |
| Extracted dye content in methanol | 12.1 % (approx.). |
| pH | 5.5–6.5 |

environmentally friendly [23]. Numerous plant species in nature produce dyes that can be used to color fabrics. These dyes can be collected from the roots, leaves, bark, flowers, and fruits of the plants, among other parts of the plants [18].

The dye was extracted from FTW for this work. Three percent was the yield of dye obtained by water extraction, and three and a half percent was obtained by methanol extraction. Physical properties (Table 1), antibacterial activity (Table 2), ATR-FTIR (Fig. 3 and Table 4), and other tests were used to analyze the extracted dyes.

In this work, the pH of the original FTW extract was utilized to color the jute packaging fabrics. Fig. 1(a–e) and Fig. 2 show the images from FTW used to color the jute packaging fabrics and the flowchart of the process.

The original tea extract's pH of 5.5 in the dye bath significantly affected the wool fabric's apparent color and color strength in addition to demonstrating exceptional antibacterial activity against *S. aureus* and *E. coli*. In the latter case, however, the tea polyphenols' oxidative polymerization in an acidic or alkaline dye bath led to a decline in the characteristic [24].

### *3.2. Anti-Bacterial activity studiesof dye from FTW*

Tea is widely known around the world and is highly valued for its antioxidant qualities. A class of chemicals found in tea, especially catechins, is well-known for their effects on the body as antioxidants [16,25,26]. Green tea had the highest levels of antioxidant and antibacterial properties against food-borne and oral infections, indicating its potential use as a natural preservative or functional ingredient in food products that promote health [26]. In this study, seven different gram-positive and gram-negative bacterial pathogens were used to assess the dye extract's possible antibacterial qualities. Against every tested microbe, the dye extract showed antibacterial properties. In this study, seven different gram-positive and gram-negative bacterial pathogens were used to screen the possible antimicrobial activity of dye extract. Dye extract exhibited antibacterial activity against all tested microorganisms. When it came to Gram-positive bacteria like *Bacillus subtilis, Staphylococcus aureus,* and *Enterococcus faecalis*, as well as Gram-negative bacteria like *Salmonella typhi*, *Shigellaflexinery*, *Vibrio parahemolyticus*, and *Escherichia coli*, the dye had good antibacterial effectiveness (Table 2).

### *3.3. The method of dyeing and visual inspection of the colored jute packaging fabric*

The testing results showed that the meta mordanted jute packaging fabrics were darker than the other two pre- and post-mordanted jute packaging fabrics. The mordant $FeSO_4$ produces good color under all jute packaging fabric dyeing circumstances. Darker color hues and better color fastness were obtained when dye concentrations, temperatures, and dyeing times rose, according to the results. Using aqueous extracted dye, the dyeing process has been optimized and established (Table 3).

### *3.4. ATR-FTIR characterization of dye from FTW and jute packaging fabric before and after dyeing*

Fig. 3 displays the ATR-FTIR spectra of FTW and extracted dye in methanol and aqueous media.The broad and strong absorption peaks at around 3305-3200 $cm{-}^{1}$ are attributed to O–H stretching vibrations resulting from hydrogen bonding between and within polymeric molecules (macromolecular associations), including alcohols, phenols, and carboxylic acids found in lignin, pectin, and cellulose.The symmetric and asymmetric C–H stretching vibration of aliphatic acids is responsible for the peak at 2921-2850 $cm{-}^{1}$. Aromatic C=C bending and asymmetric stretching vibrations of C=O are responsible for the peak at 1739-1450 $cm{-}^{1}$. Similar findings are documented in the literature [24,27].The dye extracted with methanol showed similar ATR-FTIR spectra to the dye extracted with water (Fig. 3 and Table 4).

Fig. 4 displays the ATR-FTIR spectra of both dyed and undyed jute packaging fabrics. Fig. 4 shows that the dyed jute packaging fabrics include the same functional group of dyes at 1600–1500 $cm\text{-}^{1}$ ranges, while the undyed jute packaging fabric does not contain this functional group.The new peak that appeared at 1033 $cm^{-1}$on dyed jute fabric, suggesting that tea polyphenols were dyed into fabric, can be attributable to the C–*O*–C bond of polyphenols [24].

The peak assignments of the absorption bands corresponding to various groups from Figs. 3–4 are summarized in Table 4. Table 4 presents an overview of the peak assignments for the absorption bands corresponding to the various groups shown in Figs. 3–4.

**Table 2**
Antimicrobial activity against bacterial pathogens of extracted dyes.

| Test organism | Zone of Inhibition (mm) |
|---|---|
| Gram-positive bacteria | |
| *Bacillus subtilis* | 22.5 ± 0.22 |
| *Staphylococcus aureus* | 21.5 ± 0.24 |
| *Enterococcus faecalis* | 22.0 ± 0.30 |
| Gram-negative bacteria | |
| *Salmonella typhi* | 20.0 ± 0.32 |
| *Shigellaflexinery* | 22.5 ± 0.22 |
| *Vibrio parahemolyticus* | 21.5 ± 0.24 |
| *Escherichia coli* | 22.0 ± 0.30 |

**Table 3**
Meta-mordanting process for dyeing of jute packaging fabrics using extracted dye of FTW.

| Dye (owf) | Mordant (owf) | Material: Liqour | Temp. | Time | Shade |
|---|---|---|---|---|---|
| 16 % | 10 % | 1:10 | 100 °C | 2 h | Dark coffee |

**Table 4**
ATR-FTIR data of extracted dye and jute packaging fabrics before and after dyeing.

| Position/cm$^{-1}$ | Assignment | References |
|---|---|---|
| 3305–3200 | Alcohol/Phenol O–H Stretch | 24, 27 |
| 2921–2850 | Alkyl C–H Stretch | 24, 27 |
| 1739–1450 | Asymmetric stretching vibrations of C═O and aromatic C═C bending | 24, 27 |
| 1384–1218 | Carboxylic acids (C–O bonds) | 4, 21 |
| 1050–1028 | Polyphenols (C–*O*–C bonds) | 24 |

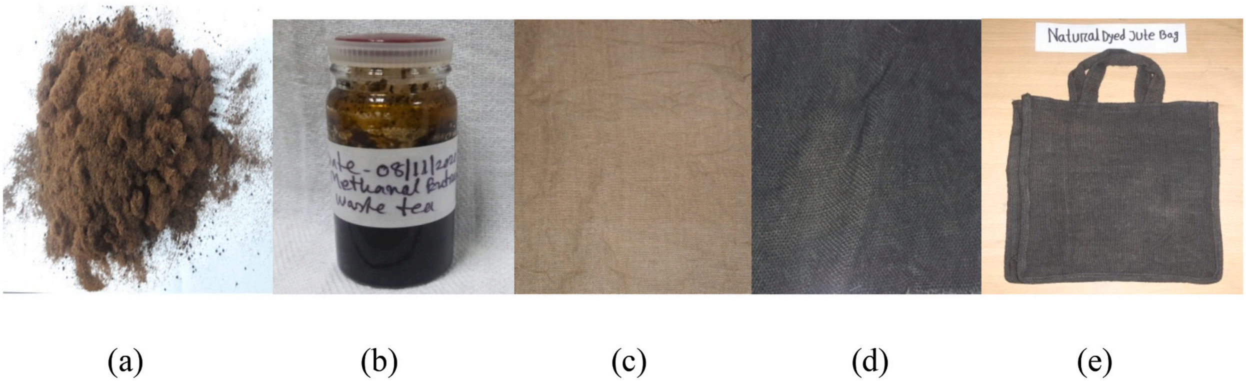


**Fig. 1.** (a) Factory tea waste (b) Extracted dye of FTW (c) Undyed jute packaging fabric (d) Dyed jute packaging fabric and (e) Dyed jute bag.

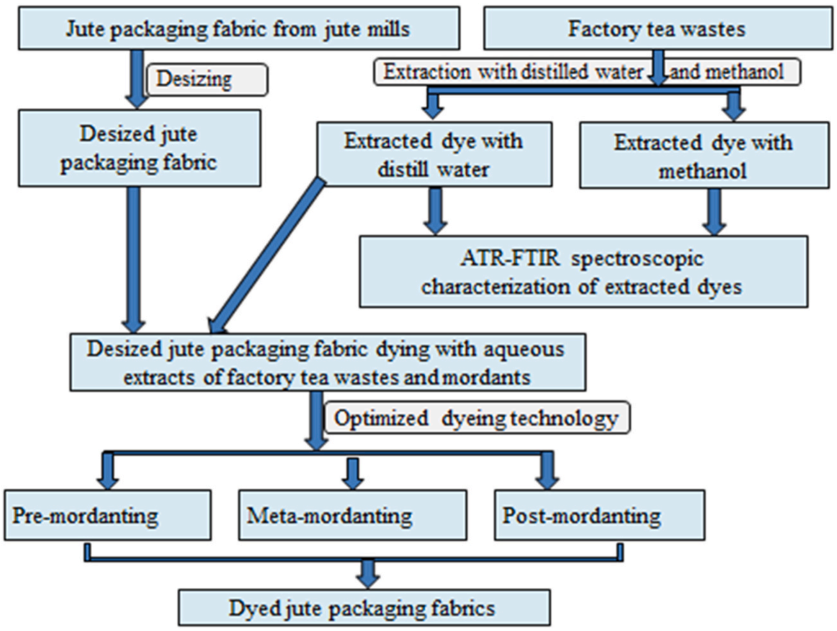


**Fig. 2.** The flowchart of the methodology of dyeing jute packaging fabric with the extracted dye of FTW.

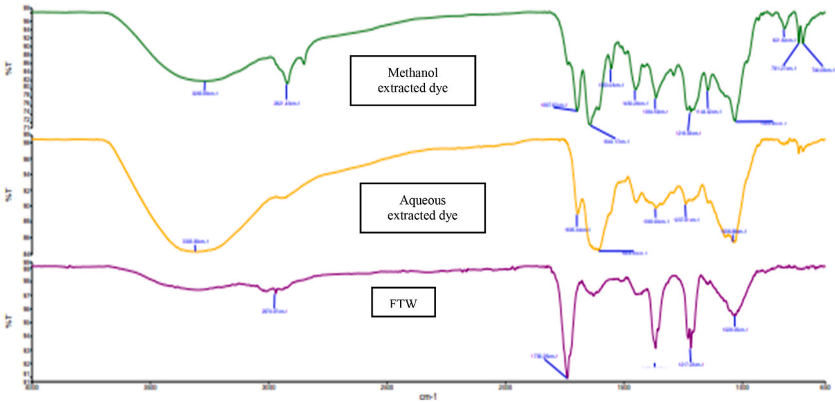


**Fig. 3.** ATR-FTIR spectra of FTW, aqueous extracted dye and methanol extracted dye.

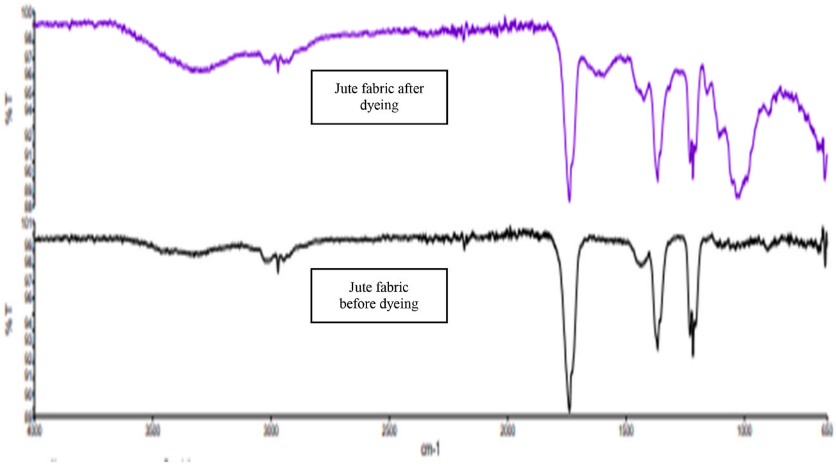


**Fig. 4.** ATR-FTIR spectra of jute packaging fabric before and after dyeing.

### 3.5. *Color fastness and strength of color in jute packaging fabric dyed with extract of FTW*

The color fastness test results for colored jute packaging fabrics are shown in Table 5. The color developed and color strength (K/S) value, CIE L* a* b*, and C* h values of all dyed jute packaging fabrics are evaluated and presented in Table 6.

The color strength (K/S) value for jute packaging fabrics by meta mordanting process is good, as Table 6 demonstrates, suggesting that the dyeing chemicals used have strong bindings. This is because the dye molecules in the jute packaging fabrics have formed coordination bonds with the metal mordants [21,28]. The relationships between the fibers, dye, and mordant are shown in Fig. 5. Fig. 5 illustrates the mordant's capacity to react with the hydroxyl groups of the extracted dyes as well as the jute packaging fabric. Similar results were observed when natural colors were applied to fabrics using Albizia procera sawdust, Arjun bark, Rheum emodi root etc [21,28,29]. However, the reason for the weak color intensity can be the dye and mordant aggregating together on the fabric [9,21].

### 3.6. *Tensile properties of dyed and undyed jute packaging fabric*

The observed tensile properties, such as elongation (%) and breaking force (N), of dyed and undyed jute packaging fabrics are shown in Table 7. The maximum force encountered by the undyed jute packaging fabric is 631.10 N at the warp and 530.56 N at the weft places, as shown in Table 7. The corresponding extensions at the warp and weft positions are 5.44 % and 12.08 %, respectively. It is also discovered that a specimen of undyed jute packaging fabric requires more force to break than a specimen of de-sized and dyed jute packaging fabric.The tensile strength of dyed cotton samples is lowered by the majority of dyeing formulations that contain natural extracts of *Albizia procera* sawdust and *Alkanna tinctoria* [21,30]. But when CuSO4 is utilized as the mordant for dying cotton fabric with the extracted *Albizia procera* sawdust [21], an increase in strength was also noted for certain of the dye recipes. Table 7 further shows that compared to all other fabrics, the dyed jute packing fabric has a larger elongation (%) at the warp location. This might be because the weft direction shrinks during the heating phase of the dyeing process, increasing the number of warp threads and raising the breaking elongation of the warp direction [31].

**Table 5**
Color fastness of dyed jute packaging fabrics.

| Sample | Wash fastness | Light fastness | Rubbing |
|---|---|---|---|
| Dyed jute packaging fabric by pre-mordanting process | 4 | 4 | 4 |
| Dyed jute packaging fabric by meta-mordanting process | 4–5 | 4–5 | 4–5 |
| Dyed jute packaging fabric by post-mordanting process | 4 | 4 | 4 |
| Dyed jute packaging fabric without mordant | 4 | 4 | 4 |

**Table 6**
Color strength (*K/S*), CIE$L^*a^*b^*$ and $C^*h$ values of color fast dyed jute packaging fabrics.

| Name of the Sample | *K/S* at 400 nm | *C** | *h* | CIE L* a* b* system | | |
|---|---|---|---|---|---|---|
| | | | | *L** | *a** | *b** |
| Dyed jute packaging fabric by pre-mordanting process | 17.47 | 9.43 | 69.70 | 28.25 | 3.27 | 8.84 |
| Dyed jute packaging fabric by meta-mordanting process | 24.23 | 4.61 | 61.44 | 19.17 | 2.20 | 4.05 |
| Dyed jute packaging fabric by post-mordanting process | 19.57 | 10.15 | 70.83 | 29.20 | 3.33 | 9.59 |
| Dyed jute packaging fabric without mordant | 20.30 | 24.32 | 62.05 | 36.01 | 11.40 | 21.48 |

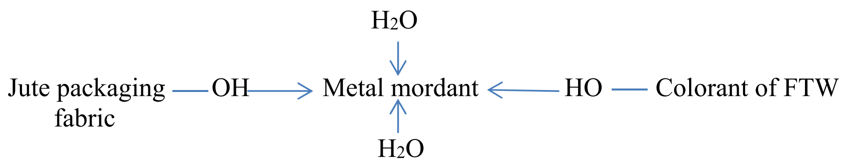


**Fig. 5.** Interaction of dye, mordant and jute packaging fabric.

**Table 7**
Tensile properties of undyed and dyed jute packaging fabrics.

| SN | Sample Name | Force (N) | | Elongation (%) | |
|---|---|---|---|---|---|
| | | Warp | Weft | Warp | Weft |
| 01 | Sized jute packaging fabric | 631.10 | 530.56 | 5.44 | 12.08 |
| 02 | De-sized jute packaging fabric | 492.48 | 457.78 | 9.92 | 15.55 |
| 03 | Dyed jute packaging fabric | 394.04 | 386.47 | 16.19 | 11.34 |

## 4. Conclusion

Eco-friendly, cost effective and sustainable natural dyeing process has been developed and optimized for jute packaging fabrics using aqueous extracts of FTW. ATR-FTIR spectroscopic analyses showed the presence of dye in the extracted dye of FTW and dyed jute packaging fabrics. Microbial analysis of extracted dyes showed the antimicrobial activity against bacterial pathogens. Around 14.5 wt % dye was obtained from the aqueous extraction of factory tea wastes. Dark coffee shades were obtained with ferrous sulfate mordant by optimized dyeing technology of pre-, post- and meta-mordanting process respectively. It was observed that the fastness properties of the dyed jute packaging fabrics improved with the application of mordants. Among the three mordanting methods, simultaneous or metamordanted dyed jute packaging fabrics evaluated as very good in performance of fastness properties. Tensile properties of dyed jute packaging fabrics were decreased than undyed jute packaging fabrics. It can be concluded that natural dyeing of jute fabric by aqueous extract of factory tea wastes are safe for packaging materials including food products.

## Data availability

The datasets generated and analyzed in the present study are available from the corresponding author upon reasonable request.

## CRediT authorship contribution statement

**Md Khabir Uddin Sarker:** Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Data curation. **Mohammad Majedul Haque:** Validation, Methodology, Investigation, Formal analysis, Data curation. **Md Rashed Hasan:** Validation, Methodology, Investigation, Formal analysis, Data curation. **Shahin Sultana:** Writing – review & editing, Validation, Supervision, Resources, Methodology, Investigation, Conceptualization. **Swapan Kumer Ray:** Validation, Methodology, Investigation, Formal analysis. **Md Aftab Ali Shaikh:** Writing – review & editing, Supervision, Resources, Project administration.

## Declaration of competing interest

The authors declare that they have no conflict of interest.

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