Ultrasound-assisted pre-treatment and dyeing of jute fabrics with reactive and basic dyes

Mohammad Mahbubul Hassan[1,*], Khaled saifullah[2]

[1] *Textile Science & Engineering Team, Food & Biobased Products Group, AgResearch Limited, Private Bag 4749, Christchurch 8140, New Zealand.*

[2] *Department of Mechanical Processing, Bangladesh Jute Research Institute, Manik Miah Avenue, Sher-E-Bangla Nagar, Dhaka 1207, Bangladesh.*

Abstract

Ultrasonic dyeing has been investigated as a means to increase the diffusion of the dye molecules into the fiber for the dyeing of various fibers. However, for scouring, bleaching, and dyeing of jute fabrics, the beneficial effect of sonication was never realized. In this work, we report the effect of sonicated scouring and bleaching of jute fabrics on their physicomechanical properties and the dyeability in the conventional dyeing with reactive and basic dyes. The sonicated scoured and bleached fabric showed higher whiteness index and weight loss but the tensile strength and yellowness index decreased compared to the conventionally scoured and bleached jute fabric. The

[*] Corresponding author: Tel.: +64-3-321-8755; fax: +64-3-321-8811

E-mail: Mahbubul.hassan@agresearch.co.nz (M M Hassan)

sonicated scoured fabric showed partial removal of lignin but the conventionally scoured fabric did not show any change in lignin content. It was found that in the case of conventional dyeing, the sonicated scoured and bleached fabric produced higher color strength than the jute fabric scoured and bleached at the same conditions but without sonication. Moreover, we also investigated the effect of ultrasound on the dyeing and color fastness properties of jute fabric dyed with two reactive and two basic dyes. It was found that the sonicated dyeing produced higher color strength compared to the fabrics dyed without sonication. Both conventional and sonicated dyed fabric showed very similar color fastness properties to light, washing, and rubbing indicating no degradation of dyes occurred during sonicated dyeing.

Key-words: Ultrasonic scouring; ultrasonic bleaching; ultrasonic dyeing; de-lignification; jute fiber; color strength

1. Introduction

Jute is an important biodegradable cellulosic fiber with a high moisture regain similar to the moisture regain of wool fiber. It falls under the bast fiber category as the fibers are collected from the bast or skin of the plant after the retting process. The fibers are located between the epidermises and an inner woody core of the jute plant. Within the stem, there are a number of fiber bundles, each containing individual fiber cells or filaments made up of cellulose. These fiber cells are bonded together by a matrix, which is composed of lignin, pectin, and hemicellulose. The pectin is removed during retting process to separate fibers from fiber bundles. A single jute fiber is consists of many cells bonded together by lignin. The length of each cell is averaging 2 to 6 mm and they are called ‘ultimate’ cells. Jute fibers are composed of 59-72% α-cellulose, 12-15.9% hemicelluloses, 11.8-14.2% lignin, 0.2-0.5% pectin, 0.3-0.5% waxes, 0.8-1.5% protein, and 0.6-1.2% mineral matters and nitrogenous substances [1,2]. As the jute fiber is composed of cellulose ultimate fibers cemented together by lignin cross-linking and hemicellulose [3,4], the complete removal of lignin is not beneficial as it makes the fiber useless. It was reported that the pre-treatment with chlorite increases lignin removal and therefore reduces the load on scouring [5], but industrially the chlorite treatment is not used as the tensile strength of the treated fiber is severely compromised by this treatment [6].

Cellular tissues and pectin surrounding the jute fiber bundles make the fiber stiff and un-spinnable. The fiber bundles are segregated to individual fibers by removing cellular tissues and pectin by carrying out a series of wet chemical processing, such as retting and then batching, to make the fiber soft and spinnable. Like cotton fiber, jute fiber is also needed to be scoured to remove hemicellulose, hydrophobic fatty matters and some levels of lignin from jute to improve their hydrophilicity and also need to bleach them to remove their natural color. Scouring is carried out with a cocktail of caustic soda, detergent and wetting agent but milder conditions are used compared to the alkali scouring of cotton. Other than natural impurities, jute fibers also contain jute batching oil (kerosene and other hydrocarbon oils) and a trace amount of that oil is retained in the woven jute fabrics that need to be removed during scouring. Scouring of jute is usually carried out with 2-6% caustic soda on the weight jute fiber (owf) and 2 g/l a non-ionic detergent at 80-90°C for 30 to 60 mins. Jute scouring process is less energy-hungry compared to the cotton scouring process but still consume a great level of energy. Therefore reduction of energy consumption in scouring is also desirable.

Ultrasonic waves of high-intensity ultrasound can form cavitation in a liquid that causes extreme effects. As the liquids are sonicated at high intensities, sound waves propagate into liquid media in alternating high-pressure compression and low-pressure expansion cycles. In the low-pressure cycle, the high-intensity ultrasonic waves produce a large number of microscopic vacuum bubbles, those bubbles explode causing in the high-pressure cycle causing localized high temperature and high-pressure shock wave and severe shear force that can disintegrate agglomerated particles, and also can cause milling, mixing and disintegration of cells. Ultrasound has been investigated to improve the exhaustion various dyes into textiles fibers [7-9]. It was reported that sonication improved the migration of dye molecules from dyebath to fiber and also improved level dyeing for the dyeing of a wide range of textile fibers including polyester [10], cotton [11], nylon [12, 13], cellulose acetate [14], and acrylic fibers [15]. Ultrasonication not only improved the migration of reactive dyes from dyebath to cotton fabrics but also improved their fixation by accelerating the reaction between the fiber and the fabric [16]. Ultrasonicated dyeing of cotton fabrics reduced dyeing time by accelerating the exhaustion of dyes into fiber [17]. Ultrasound was also investigated in the batching of cold pad-batch dyeing of cotton fabric with reactive dyes to enhance exhaustion and fixation of dyes [18]. It was reported that the dye-uptake also improved by the use of ultrasound in the dyeing of acrylic with a basic dye [15], and also in the dyeing of polyester with disperse dyes [19]. In the case of dyeing polyester with disperse dyes, ultrasonication decreased the size of dye particles [20]. The high affinity of dyes towards fibers causes a problem for dye molecules to diffuse into the fiber. A large number of dye molecules try to diffuse into fibers at the same time causing agglomeration of dyes onto fiber surface and can cause poor dyeing uniformity. Although certain dye auxiliaries can prevent agglomeration of dye molecules in a dyebath [21], they are not that much effective at ultra-low liquor ratios. It was reported that ultrasound in combination with a dye deagglomerating agent enabled ultra-low liquor ratio dyeing of wool, which prevented agglomeration of dye molecules in the dyebath [22]. Jute fabrics are usually dyed with reactive and basic dyes [23]. It is believed that jute fiber contains polyuronic acid, which helps exhaustion of basic dyes into jute fiber [24]. However, to the best of our knowledge, no research has been carried out that shows that ultrasonication could be beneficial for the scouring and bleaching of jute fabrics or for the dyeing of jute fabrics with basic and reactive dyes.

In this study, for the first time, we are going to report the use of ultrasound for the pre-treatment of jute fabrics. The effect of sonication on the de-lignification of jute fabrics as well as whiteness, yellowness, tensile strength and hydrophilicity of jute fabrics, have been included in our study. The aims of this study are to enable the use of ultrasound in the scouring and bleaching of jute fabrics to make processing of jute sustainable. Moreover, the effect of sonication has been investigated on the dye-exhaustion in the dyeing of jute fabrics with reactive and basic dyes.

2. Experimental

2.1. Materials

The jute fabric used in this work was a 320 g/m^2 hessian cloth with 6 ends/cm and 5 picks/cm. The jute fabric was supplied by Latif Bawani Jute Mills Ltd (Dhaka, Bangladesh). The ultrasonic bath used was a Transonic Ultrasonic Cleaning Unit (35 kHz ultrasonic frequency, R.K. Transonic Engineers Pvt. Ltd., Noida, India). The reactive dyes used were Rifazol Golden Yellow RNL (C.I. Reactive Orange 107) and Rifazol Brilliant Blue R Special (C.I. Reactive Blue 19) supplied by Rifa Industrial Co., Ltd. (Korea). The two basic dyes used were Maxilon Red GRL (C.I. Basic Red 46) and Astrazon Blue FGGL (C.I. Basic Blue 41) supplied by Huntsman Chemicals (USA) and Dystar GmbH & Co. (Germany) respectively. Hostapal MRN (a non-ionic wetting agent) and Sandoclean PC (a detergent) were purchased from Clariant Chemicals (Singapore) Ltd. Acetic acid, citric acid, and all other chemicals were purchased from Sigma-Aldrich Limited (USA).

2.2 Scouring and bleaching of jute fabric

The jute fabric samples were scoured by traditional alkali scouring without sonication, and also with sonication. The treatment was carried out in a tall glass tube, which was placed in the ultrasonic bath. The liquor in the bath was heated by placing a coil-type immersion heater and the temperature of the bath was controlled by an electronic temperature controller. The fabric sample inside the bath was moved slowly by using an overhead stirrer. The same bath, chemical compositions, time, and temperature were used for the traditional as well as for the sonicated scouring and bleaching. Scouring was carried out using 5 g/l sodium carbonate, 1 g/l Sandoclean PC, and 0.5 g/l Hostapal MRN at 60 °C for 60 min. Bleaching of jute fabric samples was also carried out at 60 °C for 60 min using 8 g/l H_2O_2, 2 g/l Na_2CO_3, 1 g/l Sandoclean PC, 3 g/l Na_2SiO_3, and 0.25 g/l Hostapal MRN. After the completion of bleaching, the samples were washed several times with cold water and then dried.

2.3. Characterization of scoured and bleached jute fabrics

The surface of scoured and bleached jute fabric with and without sonication was examined using a JEOL scanning electron microscopy (Model JSM-6100, JEOL Corporation, Tokyo, Japan) with an accelerating voltage of 15 kV. All the samples were coated with gold before SEM testing to prevent from charging. The estimation of lignin content of the control, traditional scoured and ultrasound-assisted scoured jute fabrics was carried out by dissolving 2 g of jute fabric in 72% (w/w) H_2SO_4 and insoluble lignin were separated by centrifuging at 4000 rpm for 5 minutes. The recovered lignin was washed several times with water until the pH became neutral. The recovered lignin was dried, weighed and the percentage of lignin in the fabric was calculated. Fourier transform infra-red (FTIR) spectroscopy was also used to subjectively assess the lignin and hemicellulose content in various treated jute fabrics. Any change in lignin content in the fabric was assessed by using a Shimadzu FT-IR (Model Prestige 21, Shimadzu Corporation, Japan) with an attenuated total reflectance (ATR) attachment at a resolution of 4 cm^{-1} for 32 scans in the range from 650 to 4000 cm^{-1}. The ZnSe crystal was used to record ATR-FTIR spectra.

The tensile strength of jute fabrics was assessed by using an Instron Universal Tensile Testing Machine (Model 4402c, Instron Corporation, USA) at 20 °C and 65% relative humidity according to the ASTM Test Method D5035-06: *Standard Test Method for Breaking Force and Elongation of Textile Fabrics (Strip Method)*. The sample size was 25.4 × 152.4 mm, and the gauge length was 100 mm. The samples were conditioned at the above-mentioned temperature and humidity for 3 days. At least 10 samples were tested for each treatment and averages are reported here. A hand-held X-Rite spectrophotometer (X-Rite International, USA) was used to measure the whiteness index as well as yellowness index.

The wettability of the fabric was measured according to the AATCC Test Method 39-1998: *Evaluation of Wettability* at 20±2 °C and 65±2% relative humidity by placing a droplet of distilled water on the jute fabric samples from two cm above the fabric and the time taken to completely disappear the droplet was measured. The assessment of weight loss of jute fabric during traditional and sonicated scouring and bleaching was carried out by the gravimetric method. The oven dried (at 105 °C) jute fabric sample was weighed before and after the scouring and bleaching processes. The weight loss was calculated according to the following equation:

$$Weight\ loss=\frac{W_1-W_2}{W_1}\times 100 \qquad (1)$$

where W_1 and W_2 are the oven dry weight of jute fabric before and after the treatment respectively. The *L**, *a**, *b** values were measured by a hand-held X-Rite spectrophotometer (X-Rite International, USA). The assessment of whiteness was then deduced by the following equation:

$$Whiteness\ index=100-[(100-L^*)^2+(a^*)^2+(b^*)^2]^{0.5} \qquad (2)$$

2.4. Dyeing procedure used for the dyeing with reactive and basic dyes

2.4.1. Dyeing of jute fabrics with reactive dyes

All dyeings were carried out using the same glass tube and the ultrasonic bath set up that was used for the scouring and bleaching treatments, using materials to liquor ratio 1:30. The glass tube was filled with sufficient water, pre-dissolved dye solution and 0.5 g/L Antimussol SI. The electrolyte used was sodium sulfate at 10% owf. The fabric sample was introduced and treatment was continued for 20 min at 25°C followed by addition of 1/3 of sodium sulfate. The treatment was continued for another 10 min and the second 1/3 of the salt was added. After 10 min of treatment, the last 1/3 of sodium sulfate was added. The temperature was then raised to 60 °C and held for 10 min. The required quantity of sodium carbonate was added and held for another 60 min. The dye bath was then cooled to 45 °C, the liquor drained, and the dyed samples were rinsed with hot and cold water. They were then dried at 60 °C until completely dried.

2.4.2. Dyeing of jute with basic dye

The conventional dyeings of jute fabrics scoured and bleached without and with sonication were carried out with basic dyes using the same set up used for the scouring and bleaching of jute fiber using materials to liquor ratio 1:30. The dye pots were filled with sufficient water and then pre-dissolved sodium sulfate (10% owf), sodium acetate (3% owf), and 0.2 g/L Albegal POC were added to it. After introducing the fabric samples into the dye pots, the treatment was continued for 20 mins and the required quantity of pre-dissolved dye (2% owf) was then added to the pots. The temperature was raised to 60 °C and held for 60 min. After the completion of dyeing, the dye bath was then cooled to 45 °C and the liquor was drained. The dyed samples were rinsed with cold water and also hot washed. The samples were then dried at 60 °C for 30 minutes.

To evaluate the effect of ultrasound on the dyeing of jute fabrics with basic dyes, the jute fabrics were scoured and bleached by the traditional method (without sonication) as mentioned before but the dyeing was carried out in a laboratory dyeing machine (Roaches Laboratory Dyeing Machine, Model: Pyrotec 2000, Roaches International Ltd., Birstall, England). The other dyeing conditions were same for the both methods except the dyeings were also carried out at 98 °C instead of 60 °C that was used for the dyeing of jute fabrics with basic dyes to compare the effect of sonication on scouring and bleaching of jute fabrics.

2.5. Measurement color and various fastness properties

The color measurements of various dyed samples were carried out by measuring the reflectance values (at the appropriate wavelength of maximum absorption for each dye) of the dyed samples using a Datacolor Spectraflash 600 spectrophotometer interfaced to a personal computer [25]. The color strength (K/S) was calculated according to the following equation:

$$\frac{K}{S} = \frac{(1-R)^2}{2R} \qquad [3]$$

where R = reflectance values at the wavelength of maximum absorption of a particular dye, K = absorption coefficient, and S = scatting coefficient. The reflectance measurements were carried out under illuminant D65, using a 10° standard observer with UV component and specular excluded. Each of the samples was folded twice to realize a total of four layers of fabric. A total of four measurements were made on each sample, from which the average value was calculated. The levelness of the shade produced was spectrophotometrically measured by measuring CIE *L**, *a**, *b** color difference (ΔE) between two areas of the same dyed sample by a by a hand-held X-Rite spectrophotometer (X-Rite International, USA). The value of ΔE lower than 1 is perceived as acceptable color difference as the human eye cannot identify the color difference lower than 1. For the assessment of dye fixation, the reactive dyed fabric samples were soap washed with 1 ml/l Sandoclean PC at 60 °C for 15 min and again cold washed and hot washed. The samples were then dried at 60 °C for 30 min. The dye fixation percentage was measured by measuring K/S values of the dyed fabrics before and after the soaping operation.

The color fastness to light and washing of the dyed fabrics were assessed according to the AATCC Test Method 16.3-2011 *Colorfastness to Light: Xenon-Arc* and the wash fastness was measured according to the ISO Test Method 105-C03 1987: *Textiles -- Tests for color fastness – Part C03: Color fastness to washing: Test 3* by washing in a Gyrowash (Model 415/8) using the phosphate-free standard SDC detergent. In both cases, fastness grades were assessed by comparing with the 3M Grey Scale. The rubbing fastness was measured according to the AATCC Test Method 13-2016, *Colorfastness to Crocking: Crockmeter Method* by using a crock meter (Crock Master, Model 670, James Heal Limited, Halifax, UK) but only at dry condition and the fastness grade was assessed by comparing with the 3M Grey Scale.

2.6. Measurement of UV transmittance through the dyed fabrics

Varian UV-VIS Spectrophotometer (Model: CARY 3E, Varian Inc., Palo Alto, USA) with Cary 1/3E Diffuse Reflectance Measurement attachment was used to assess the percent transmission at wavelength intervals up to 5 nm in the 290–400 nm spectral span.

3. Results and discussions

3.1. Effect of sonicated scouring and bleaching on physicochemical properties of scoured and bleached jute fabrics

It was observed that ultrasound irradiation not considerably affected scouring and bleaching performance of jute fabrics but also their other physicochemical properties compared to the scouring and bleaching without sonication.

3.1.1. Effect on fiber morphologies

Fig. 1 shows SEM micrographs of raw jute fibers and also the jute fibers scoured and bleached at the same conditions with and without sonication. The SEM images of raw jute show multicellular structure (as shown by an arrow in Fig. 1A) but ultimate cells are invisible because of covering and cementing by lignin. The contaminant particles visible on the surface of raw jute fibers could be mostly lignin crystals. It is evident that scouring with or without sonication both removed the hemicellulose-based cellular structure and ultimate cells are clearly visible by the removal of hemicellulose. It can be seen that traditional scouring progressively cleaned the surface of the fiber and made fiber surface smooth but some lignin crystals are still visible on the fiber surface. It can be seen that even after bleaching, particles of lignin are still visible on the bleached fiber surface. On the other hand, the sonicated scouring considerably cleaned the fiber surface as hardly any lignin particles are visible on the scoured fiber surface but produced micro gaps or grooves (as shown by arrows in Fig. 1C) on the fiber surface and also made the fiber surface rough indicating removal of surface bound lignin. The number of grooves produced on the fiber increased after the sonicated bleaching (as shown in Fig. 1E), indicating further removal of lignin during the sonicated bleaching treatment. The fiber damage was further enhanced in the bleaching stage but no fibrillation occurred and ultimate bundles were intact. It is evident that ultrasonic scouring removed the lignin particles adhered to the surface of the fiber and ultrasonic bleaching further cleaned the surface of fiber through the removal of hemicellulose and lignin without causing any major fibrillation. The ultrasound irradiation dislodged most of the lignin particles adhered to the fiber surface.

3.1.2. Effect on scouring and bleaching efficiency

For ease of bleaching and dyeing, especially to improve the whiteness and hydrophilicity of jute fiber surface, the residual lignin content in jute needs to be reduced. Scouring of jute removes hemicellulose thereby improving the color and whiteness of jute fiber. Table 1 shows the scouring performance of jute fabrics including their lignin content, whiteness, yellowness, wettability and mechanical properties, scoured without and with sonication. It can be seen that the effect of sonication on the reduction of the lignin content of jute fiber is minimal as the lignin content of the fiber only marginally reduced. The lignin content of untreated jute was 14.3±0.25%. No reduction in lignin content was observed for the jute fabric scoured without sonication but in the case of ultrasound-assisted scouring the lignin content reduced to 12.8±0.3%. It is not expected that ultrasound will considerably increase lignin removal as its solubility in water will not be affected by sonication. However, the reduction in lignin content achieved for the sonicated scouring could be due to some of the lignin from the surface of fiber was dislodged by strong cavitation force produced by ultrasound. It can be seen that more weight and tensile strength loss of jute fiber occurred in the case of sonicated scouring compared to the traditional scouring. The weight loss for the traditional scouring and bleaching was 5.0 and 7.1% respectively. However, for the US-assisted scouring and bleaching, the weight loss increased to 6.4 and 8.9% respectively. Scouring and bleaching without and with sonication both had a great effect on the reduction of yellowness of jute fabric but the greatest effect was observed for the sonicated scouring and bleaching. The yellowness index for the control fabric was 63.38, which decreased to 55.18 and 50.23in the case of traditionally scoured and bleached jute fabric. However, in the sonicated scouring, the yellowness index decreased to 53.21, which further decreased to 29.3 for the sonicated bleaching. Scouring and bleaching had a great effect on the wettability of wool. Traditional and ultrasound-assisted scouring and bleaching both showed excellent wettability because of the removal of fatty matters and hemicellulose.

Table 1 also shows mechanical properties of control and various scoured and bleached jute fabrics. Scouring and bleaching affected the mechanical properties of jute fabrics and progressively reduced tensile strength. The tensile strength shown by the untreated fabric is 34.74±2.11 kgF, which reduced to 31.87±1.32 and 30.45±0.75 kgF after traditional scouring and bleaching respectively. The corresponding figures for US-assisted scouring and bleaching were 28.3±1.27 and 27.8±0.87 kgF respectively. The sonicated scoured and bleached jute fabric showed considerably higher tensile strength loss compared to the tensile strength loss observed for jute fabric scoured and bleached by the conventional method. From SEM images (Fig. 1), it is evident that sonicated scouring produced grooves on the surface of the fiber, which was possibly formed by the removal of surface bound lignin, resulting in causing tensile strength loss. The damage was further enhanced in the sonicated bleaching, which caused a further loss of tensile strength. However, the levels of tensile strength loss during sonicated scouring and bleaching were quite low indicating only small levels of removal of lignin.

3.1.3. Effect on whiteness and Color

Table 2 shows whiteness and CIE L*a*b* color values of control and also the scoured and bleached fabric with sonication and without sonication. It can be seen that in the case of scouring without sonication, compared to the untreated fabric, the lightness value (L*) and blueness/redness value (b*) decreased but the yellowness/greenness value (a*) slightly increased resulting in the decrease in whiteness index. After the bleaching stage, the whiteness index slightly increased. On the other hand, ultrasound-assisted scouring slightly increased the whiteness index of the fabric, which considerably improved by the sonicated bleaching process.

3.1.4. Effect on UV absorption

Lignin mostly absorbs UV at 360 and 300 nm which fall under UVA and UVB range of UV absorption. It can be expected that the removal of lignin will reduce absorption of UV and thereby increased the transmittance. Fig. 2 shows UV transmittance of various treated jute fabrics over 290 to 400 nm. It can be seen that the untreated control showed the lowest UV transmittance. The jute fabrics scoured and bleached without sonication showed similar UV transmittance. However, in the case of sonicated scouring and bleaching, the UV transmittance considerably increased indicating that lignin content of the fabric decreased by the sonicated scouring and bleaching. The results achieved are consistent with the surface morphologies of the various treated jute fabrics.

3.1.5. FTIR

Fig. 3 shows the ATR-FTIR spectra of untreated jute fabric, and also the scoured and bleached jute fabrics with and without sonication. The spectra showed prominent peaks in the fingerprint regions of 650 to 1600 cm^{-1}. The spectrum of untreated jute shows a typical spectrum of jute fiber containing lignin and hemicellulose. The peak at 1508, 1418 and 1374 cm^{-1} represent aromatic skeletal in lignin, C-H deformation and C-H deformation in polysaccharides, respectively [26]. The peak at 1233 cm^{-1} could be attributed to the –OH groups and the peak at 1158 could be attributed to C–O–C vibration in the polysaccharide. The other peaks are shown at 1047 and 896 cm^{-1} could be associated with C-O stretch in polysaccharide and C-H deformation in cellulose out of plane bending, respectively. We used FTIR data to qualitatively measure the lignin content of the scoured jute fabrics without and with sonication. Zhu et al., successfully used FTIR to quantify the lignin content of wood [27] by considering the peak height at 1505, 1458 and 1420 cm^{-1} as they found that the intensity of the peak at 1505, 1458 and 1420 cm^{-1} increased with an increase in the lignin content in wood. Similarly, in Fig. 3 it can be seen that in the case of jute fabric scoured with sonication the intensity of the peak at 1505, 1458 and 1420 cm^{-1} is considerably lower compared to the intensity shown in the spectrum of scoured jute fabric at those wavenumbers. In summary, ATR-FTIR spectra of jute fabrics scoured with and without sonication suggest that the lignin content reduced in the case of sonicated scouring. It can be assumed that only loosely bound lignin rather than the covalently bonded lignin are removed by sonicated scouring.

3.3. Dyeing

In the case of dyeing, we looked mainly two things, the effect of removal of lignin in scouring and bleaching on the dyeability of jute fabrics as well as the effect of sonication on the migration of dye molecules from dyebath in the dyeing of jute fabrics.

3.3.1. Effect of sonicated scouring and bleaching on the dyeability of jute fabrics

Fig. 4 shows reflectance spectra of jute fabrics (those were scoured and bleached without and with sonication) dyed with reactive and basic dyes. The color strength (K/S) for the non-sonicated scoured and bleached fabric dyed with Maxilon Red RNL and Rifazol Golden Yellow RNL was 18.83 and 17.88 respectively but the corresponding values for the jute fabric scoured and bleached with sonication are 19.64 and 18.25, respectively. It is evident that in the case of both basic and reactive dyes, the sonicated scoured and bleached fabric produced better color strength than the color strength produced by the jute fabric scoured and bleached without sonication. The dyeing with basic dyes produced higher color strength compared to the dyeing with reactive dyes. It is known that the removal of lignin increases the hydrophilicity of jute fabric and also increases its contents of –CHO and –COOH groups [28]. As the content of –COOH groups in jute fiber increases along with the increase of surface hydrophilicity with an increase in the removal of lignin, the absorption of cationic basic dyes increases. However, the absorption of anionic reactive dyes should decrease with the removal of lignin, but we found that the color strength of the dyed fabric slightly increased in the case of sonicated scoured and bleached fabric. In this case, the increase in hydrophilicity of the fabric positively affected the dye exhaustion countering the negative effect of the lignin removal. However, the level of improvement in color strength is not high representing that only a fraction of lignin content of jute was removed by sonication.

Table 3 shows CIE L*a*b* values of ultrasound-assisted scoured and bleached jute fabrics dyed with a reactive and also a basic dye. The L*, a*, b* values are related to the lightness, greenness/redness and blueness/yellowness /darkness of the fabric, respectively. It can be seen that in the case of ultrasound-assisted scoured and bleached fabric dyed with Maxilon Red GNL, the value of L* and a* was higher but the value of b* was considerably lower compared to the traditionally scoured and bleached fabric dyed with the same dye at the same depth of shade. Similarly, in the case of Rifazol Blue R dye, the US-assisted scoured and bleached fabric showed lower L* value but higher a* and b* values compared to the traditionally scoured and bleached fabric. In the case of both of the dyes, the ultrasound-assisted scoured and bleached fabric produced deeper shade compared to the traditionally scoured and bleached fabric.

The dye fixation (%), color fastness to light, washing, and rubbing of jute fabric dyed with reactive and basic dyes by the conventional method are shown in Table S1 (Supplementary Information). It can be seen that the reactive dyed fabric samples showed better wash and rubbing fastness compared to the basic dyed fabric for both conventional and sonicated scoured and bleached fabric. It can be seen that the conventional scoured and bleached dyed fabric showed dye fixation and fastness properties similar to the dyed fabric that was sonicated scoured and bleached.

3.3.2. Effect of sonication in increasing exhaustion of dyes

Fig. 5 shows reflectance spectra of jute fabrics dyed with two reactive and two basic dyes with and without ultrasonication. It is evident that the dyeing carried out at 60 °C produced lower color strength compared to the dyeing carried out at 98 °C for basic dyes (as shown in Table 3) as it is known that the dyeing temperature has a great effect on the dye exhaustion in the dyeing with reactive and basic dyes [29,30]. It can be seen that for both classes of dyes, sonicated dyeing produced considerably higher color strength (K/S) compared to the color strength produced by the dyeing without ultrasound. In the case traditional dyeing, the color strength of jute fabrics dyed with Astrazon Blue FGGL and Maxilon Red RNL was 14.93 and 15.8, the corresponding values for the dyeing with ultrasound were 17.5 and 16.87. In the case of dyeing with reactive dyes, jute fabrics dyed with Rifazol Golden Yellow RNL and Rifazol Brilliant Blur R Spc without ultrasound produced color strength 6.22 and 4.74 but in the case of dyeing with sonication, the color strength increased to 7.25 and 5.49 respectively. The findings are consistent with the phenomenon observed for the dyeing of other textile fibers with reactive and basic dyes [12, 31]. Probably, ultrasound increases the mobility of dye particles in the dye bath and also breaks down agglomeration of dyes on fiber surfaces that ease higher migration and diffusion of dye molecules from dyebath to the fiber surface and from fiber surface to the interior of fibers. As a result, deep shades are produced. Therefore, ultrasonication is very useful to reduce energy consumption by reducing dyeing temperature and also reduces the effluent load on effluent treatment plant by reducing dyes remain in the effluent.

Table 4 shows CIE L*a*b* values of traditionally scoured and bleached fabric dyed with two reactive and also two basic dyes with and without ultrasonication. It is evident that for all dyeings, the ultrasound-assisted dyed fabrics showed lower L* compared to the fabrics dyed by a conventional dyeing method, indicating producing deeper shades compared to the shades produced by the fabric dyed by the conventional dyeing method.

The dye fixation (%), color fastness to light, washing, and rubbing of jute fabrics dyed with reactive and basic dyes without and with sonication are shown in Table S2 (Supplementary Information). It can be seen that the reactive dyed fabric samples showed better wash and rubbing fastness compared to the basic dyed fabric for both conventional and sonicated dyeing. The sonicated dyed fabrics showed dye fixation (%) and fastness properties almost similar to the dye fixation (%) and fastness properties shown by the fabrics dyed without sonication indicating no degradation of dye molecules occurred during sonicated dyeing.

4. Conclusions

We have demonstrated that sonication could be beneficial for the pre-treatment as well as for the dyeing of jute fabrics, as the pre-treatments and dyeing can be carried out at much lower temperature compared to the conventional scouring, bleaching, and dyeing. Ultrasonic scouring provided partial removal of lignin compared to the conventional scouring, in which case no lignin was removed. Sonicated scouring and bleaching improved the whiteness of the fabric and reduced its yellowness and tensile strength. Similarly, sonicated dyeing also showed improved dye exhaustion and produced deeper shades compared to the fabric dyed with the same dyes at the same conditions but without ultrasonication. The sonicated dyeing did not affect color fastness properties of dyed jute fabrics. Moreover, in the case dyeing with reactive dyes, ultrasound only marginally affected dye fixation(%). The developed method enhances the sustainability of jute fiber processing by reducing energy consumption for the pre-treatment and dyeing of jute fabrics.

References

[1] C. Roul, The International Jute Commodity System, The Northern Book Centre, New Delhi, India, 2009, p. 22.

[2] N. N. Das, S. C. Das, A. K. Mukherjee, Origin of acidity in jute fiber, Text. Res. J. 54 (1984) 166–171.

[3] D. P. Chattopadhyay, Introduction, chemistry and preparatory processes of jute, Colourage 5 (1998) 23–35.

[4] N. C. Pan, A. Day, K. K. Mahalanabis; Chemical composition of jute and its estimation, Man-made Textiles In India 9 (1999), 467–473

[5] F. Khan, S. R. Ahmad, Chemical modification and spectroscopic analysis of jute fiber, Polym. Degrad. Stab. 52 (1996) 335-340.

[6] W. –M. Wang, Z.-S. Cai, J.-Y. Yu, Study on the chemical modification process of jute fiber. J. Eng. Fiber. Fabric. 3 (2008) 1–11.

[7] M. M. Kamel, R. M. El-Shishtawy, H. L. Hanna, N. S. E. Ahmed, Ultrasonic-assisted dyeing: I. Nylon dyeability with reactive dyes. Polym. Int. 52 (2003) 373-380.

[8] E. Öner, I. Başer, K. Acar, Use of ultrasonic energy in reactive dyeing of cellulosic fabrics, J. Soc. Dyer. Colour. 111 (1995) 279-281.

[9] G. A. Grande, M. Giansetti, A. Pezzin, G. Rovero, S. Sicardi, Use of the ultrasonic cavitation in wool dyeing process: Effect of the dye-bath temperature, Ultrason. Sonochem. 35 (2017) 276-284.

[10] L.Wang, H. Zhao, J. Lin, Studies on the ultrasonic-assisted dyeing of poly(trimethylene terephthalate) fabric, Col. Technol. 126 (2010) 243–248.

[11] N. D. Tissera, R. N. Wijesena, K. M. N. De Silva, Ultrasound energy to accelerate dye uptake and dye-fiber interaction of reactive dye on knitted cotton fabric at low temperatures. Ultrason. Sonochem. 29 (2016) 270-278.

[12] N. Merdan, M. Akalin, D. Kocak, I. Usta, Effects of ultrasonic energy on dyeing of polyamide (microfiber)/Lycra blends, Ultrasonics 42 (2004) 165–168.

[13] R. El-Shishtawy, M. Kamel, H. Hanna, N. Ahmed, Ultrasonic-assisted dyeing: II. Nylon fiber structure and comparative dyeing rate with reactive dyes, Polym. Int. 52 (2003) 381–388.

[14] C. Udrescu, F. Ferrero, M. Periolatto, Ultrasound-assisted dyeing of cellulose acetate, Ultrason. Sonochem. 21 (2014) 1477–1481.

[15] M. Kamel, H. Helmy, H. Mashaly, H. Kafafy, Ultrasonic assisted dyeing: Dyeing of acrylic fabrics C.I. Astrazon Basic Red 5BL 200%, Ultrason. Sonochem. 17 (2010) 92–97.

[16] D. Sun, Q. Guo, X. Liu, Investigation into dyeing acceleration efficiency of ultrasound energy, Ultrasonics 50 (2010) 441–446.

[17] D. Sun, Q. Guo, X. Liu, Investigation into dyeing acceleration efficiency of ultrasound energy, Ultrasonics 50 (2010) 441–446.

[18] A.A. Babar, M.H. Peerzada, A.K. Jhatial, N. Bughio, Pad ultrasonic batch dyeing of causticized lyocell fabric with reactive dyes. Ultrason. Sonochem. 34 (2017) 993–999.

[19] A.N. Saliogram, S.R. Shukla, M. Mathur, Dyeing of polyester fibers using ultrasound. Col. Technol. 109 (1993) 263–266.

[20] K.W. Lee, J.P. Kim, Effect of ultrasound on disperse dye particle size, Text. Res. J. 71 (2001) 395–398.

[21] M. M. Hassan, M Bhagvandas, Sustainable low liquor ratio dyeing of wool with acid dyes: Effect of auxiliaries on agglomeration of dye molecules in a dyebath and levelness of shade produced on fabrics, *J. Clean. Prod.*, 152 (2017) 464-473.

[22] M. M. Hassan, M. Bhagvandas, Sustainable ultrasound-assisted ultra low liquor ratio dyeing of wool textiles with an acid dye, ACS Sust. Chem. Eng. 5 (2017) 973–981.

[23] F. Bouatay, N.Meksi, S. Adeel, F. Salah, F. Mhenni, Dyeing behavior of the cellulosic and jute fibers with cationic dyes: process development and optimization using statistical analysis. J. Natur. Fiber. 13 (2016) 423–436.

[24] P. D. Sarkar, H. Chatterjee, A. K. Majumdar, K. B. Pal, The acid nature of vegetable fibers in relation to basic dye absorption, J. Soc. Dyers Col. 63 (1947) 229–231.

[25] M. M. Hassan, S J Leighs, Effect of surface treatments on physicomechanical, stain-resist, and UV protection properties of wool fabrics, Appl. Surf. Sci. 419 (2017) 348-356 .

[26] W. Collier, T. Schultz, V. Kalasinsky, Infra-red study of lignin: Re-examination of aryl-alkyl ether C-O stretching peak assignments, Holzforschung 46 (1992) 523–528.

[27] G. Zhu, G. Taylor, A. Polle, FTIR-ATR-based prediction and modeling of lignin and energy contents reveals independent intra-specific variation of these traits in bioenergy poplars, Plant Methods 7 (2009) 9.

[28] P.K. Ganguly, S. Chanda, Dyeing of jute: Effect of progressive removal of hemicellulose and lignin, Indian J. Fibre Text. Res. 19 (1994) 38–41.

[29] C.L. Bird, G.P. Stancey, The Effect of Temperature and pH on the Adsorption of Basic Dyes by Wool. Col Technol. 77 (1961) 244–246.

[30] A.K. Samanta, S. Chakraborty, T.K. Guha Roy, Dyeing of jute with reactive dyes: Optimization of the process variables and assessment of color fastness characteristics, J. Inst. Eng. (India) 93 (2012) 15–24.

[31] A. Sanislav, M. Fogorasi, M. D. Stanescu, S. Muntean, M. Dochia, Ultrasound effect on dyeing wool fibers with two anthraquinone dyes, Fiber. Polym. 16 (2015) 62-66.

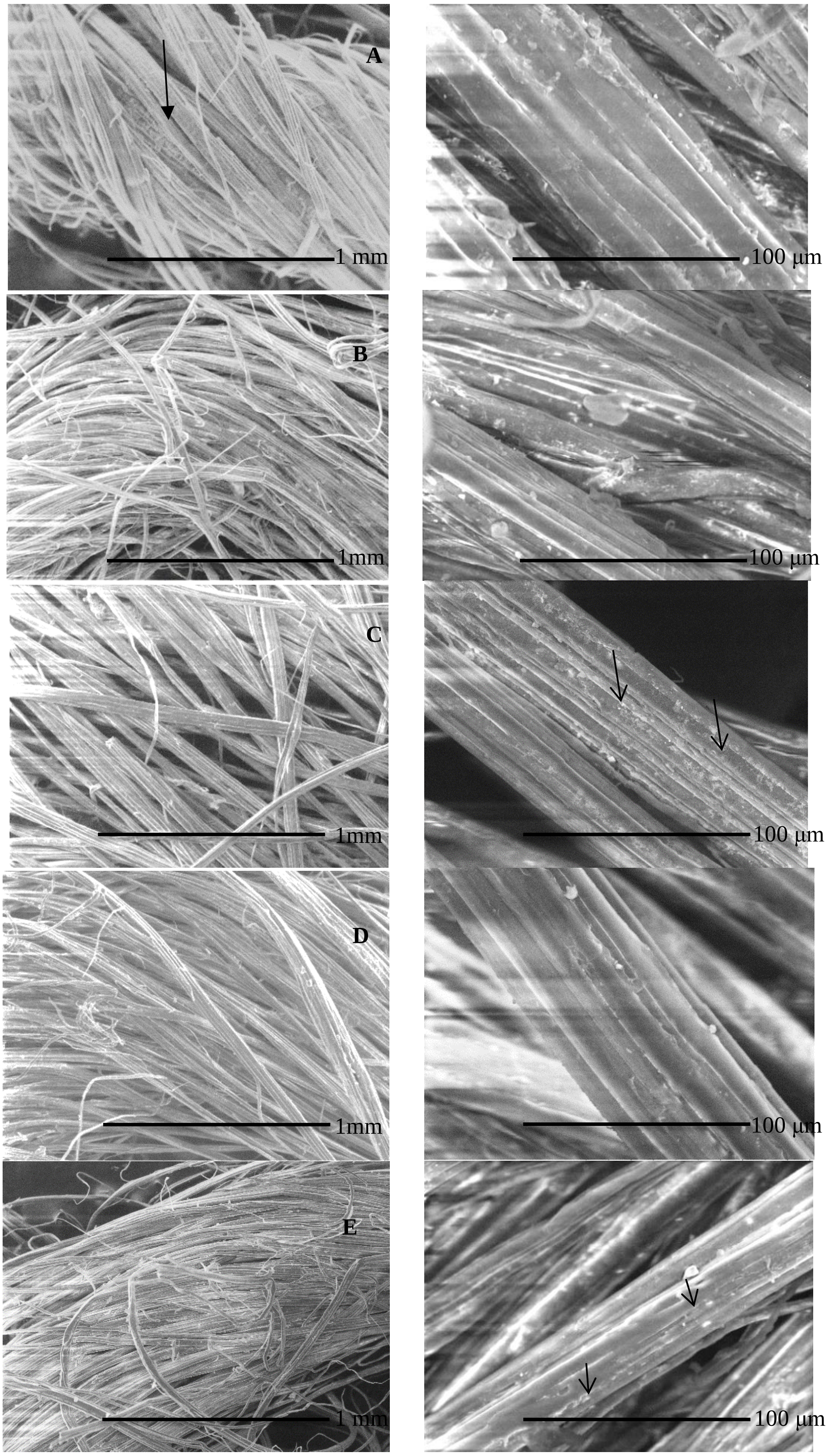

Fig. 1. Low (left) and highly (right) magnified SEM images of jute fibers scoured and bleached with and without ultrasound. A = control jute; B = scoured without sonication; C = sonicated scoured; D = bleached without sonicated; E = sonicated bleached.

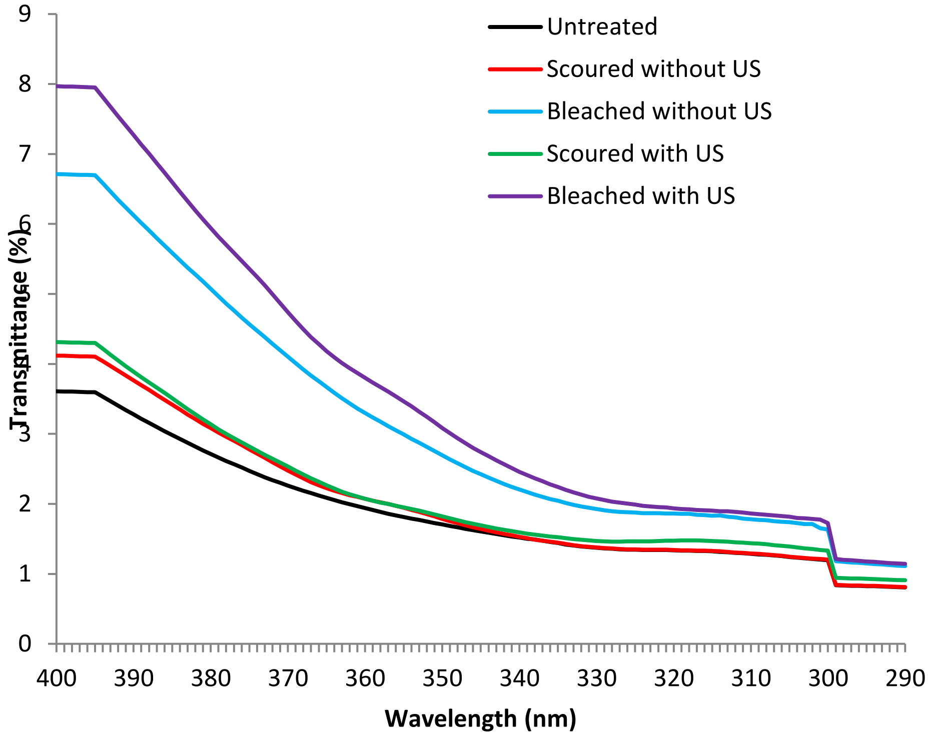

Fig. 2. UV transmittance spectra of untreated jute fabric, and also scoured and bleached jute fabric without and with sonication. US = ultrasound.

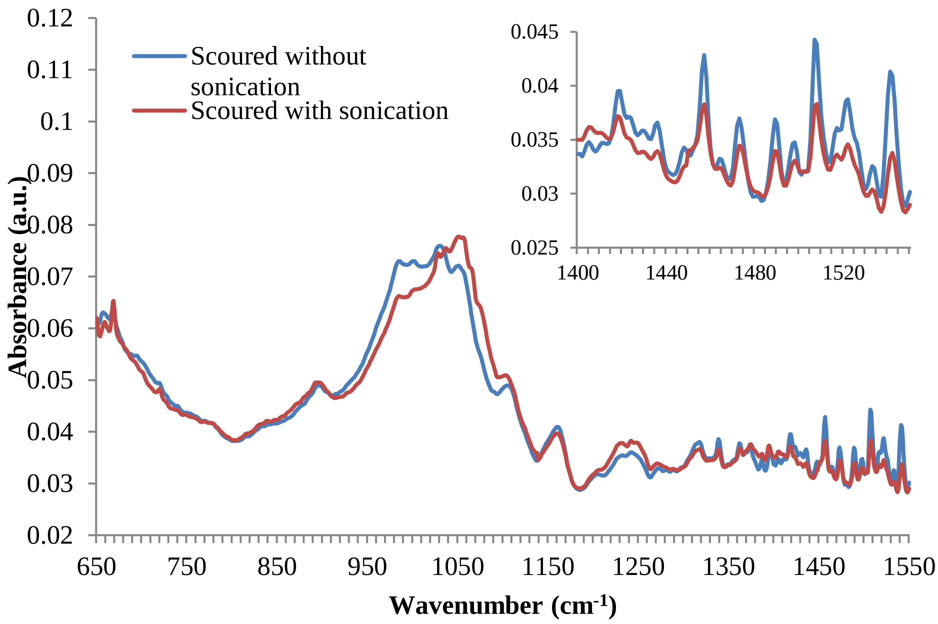

Fig. 3. ATR-FTIR spectra of jute fabrics scoured with and without sonication.

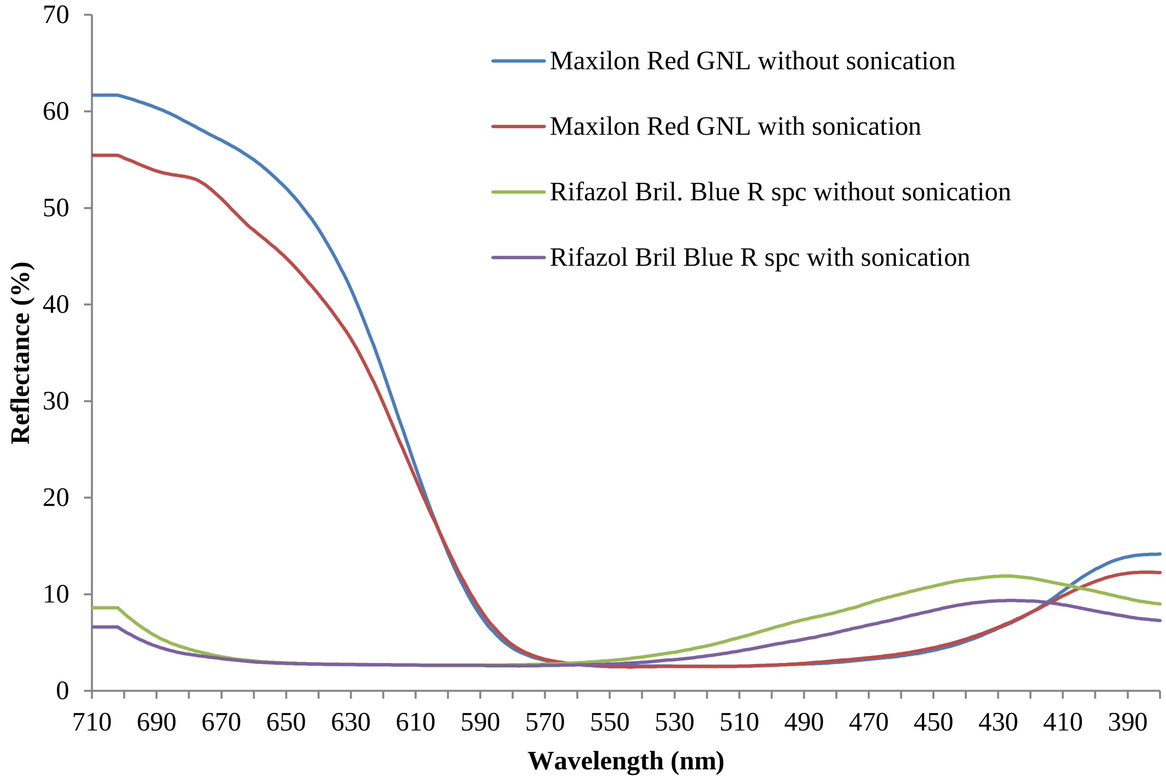

Fig. 4. Wavelength vs reflectance curves of traditional and sonicated scoured and bleached fabrics dyed with reactive and basic dyes by the conventional dyeing method.

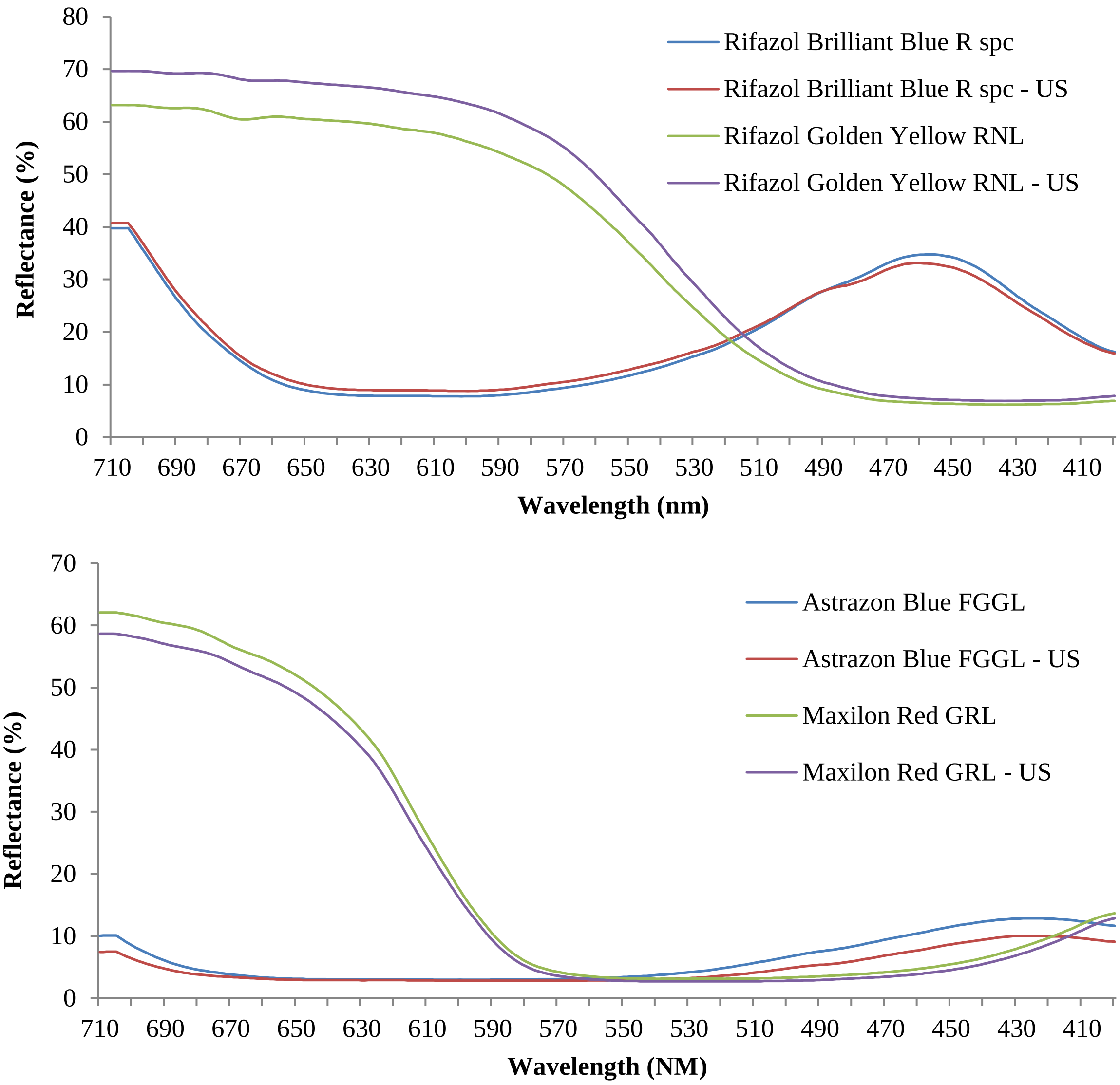

Fig. 5. Wavelength vs reflectance curves of conventionally scoured and bleached jute fabrics dyed with reactive (top) and basic (bottom) dyes with and without ultrasound (US).

Table 1.

Effect of ultrasound (US)-assisted scouring and bleaching on various properties of jute fabric.

Parameters	Lignin content (%)	Yellowness index (D1926)	Weight loss (%)	Wettability (s)	Peak force (kgF)	Breaking force (kgF)	Elongation (%)
Control fabric	14.3±0.15	63.38	-	248.00	34.74±2.11	1.13±0.04	8.67±0.96
Scoured without US	14.3±0.20	55.18	5.0±0.12	0.27	31.87±1.32	4.74±0.53	4.99±0.77
Bleached without US	-	29.3	7.1±0.15	0	30.45±0.75	8.44±1.70	13.57±0.31
Scoured with US	12.5±0.30	53.21	6.4±0.14	0.27	28.3±1.27	6.24±0.69	16.28±2.27
Bleached with US	-	50.23	8.9±0.11	0	27.8±0.87	9.44±1.32	16.26±3.38

Table. 2.

*CIE L**, *a**, *b** values of scoured and bleached jute fabric samples with and without sonication.

	L*	a*	b*	WI	ΔE with control
Control	59.49	6.77	22.93	52.96	0
Scoured	53.42	6.82	19.54	49.02	7.6
Bleached	71.33	4.94	22	63.53	11.54
Ultrasonic scoured	57.03	5.6	16.83	53.51	9.48
Ultrasonic bleached	74.12	5.03	21.99	65.7	13.24

Table 3.

Colour strength and CIE L*a*b* values of traditional and ultrasound-assisted scoured and bleached fabric dyed with reactive and basic dyes by the conventional dyeing method.

Shade	Conventional					Ultrasound-assisted				
	L*	a*	b*	ΔE within a sample	Colour strength (K/S)	L*	a*	b*	ΔE within a sample	Colour strength (K/S)
3% Maxilon Red RNL	32.65	50.94	15.27	0.57	18.83	31.15	50.36	18.36	0.45	19.64
3% Rifazol Brilliant Blue R	19.29	7.00	-27.74	0.69	17.88	16.48	9.06	-24.55	0.63	18.25

Table 4.

Dyeing performance of conventionally scoured and bleached jute fabrics dyed with reactive and basic dyes by conventional and ultrasound-assisted dyeing.

Dyes used	Conventional dyeing					Sonicated dyeing				
	L*	a*	b*	ΔE in same samples	Colour strength (K/S)	L*	a*	b*	ΔE in same samples	Colour strength (K/S)
2% Maxilon Red RNL	32.16	50.67	17.37	0.86	14.93	29.93	48.05	16.62	0.62	17.50
2% Astrazon Blue FGGL	18.57	8.82	-27.34	0.97	15.80	16.91	8.34	-26.08	0.75	16.87
1% Rifazol Golden Yellow RNL	67.1	22.79	68.80	0.79	6.22	66.69	23.655	69.47	0.51	7.25
1% Rifazol Brilliant Blue R.	37.46	-3.265	-30.25	0.93	4.74	36.91	-3.22	-30.21	0.95	5.49