Indian Journal of Fibre & Textile Research
Vol. 47, June 2022, pp. 227-233

Eco-friendly dyeing of cotton fabric with tender green coconut soft husk

Sujata R Kawlekar[a], ASM Raja, Sujata Saxena & RM Gurjar

Chemical and Biochemical Processing Division, ICAR-Central Institute for Research on Cotton Technology, Adenwala Road, Matunga, Mumbai 400 019, India

Received 30 March 2021; revised received and accepted 16 September 2021

Tender coconut husk, a waste after consuming the coconut water, has been explored as a dye for the colouration of cotton fabric. It is abundant in polyphenolic compounds which are the main coloring agent. The aqueous extraction method is used for obtaining the dye. Different mordanting techniques, such as pre-mordanting, simultaneous mordanting and post mordanting, are used to dye cotton fabric. Tannic acid and alum are used as primary mordants, and ferrous sulphate and copper sulphate are used as secondary mordants. A range of shades is obtained by varying mordants and different mordanting methods. Overall fastness properties of these shades are found good. Dyed fabrics are also found to have an excellent UV protection property.

Keywords: Coconut husk, Colour strength, Cotton, Dyeing, Mordant, Natural dye

1 Introduction

Natural dyes, dyestuff and dyeing are as old as textiles themselves. Primitive dyeing techniques included sticking plants to fabric or rubbing crushed pigments into cloth. The methods became more sophisticated with time, and techniques included fabrics boiled with natural dyes from crushed fruits, berries and other plants. Natural colorants obtained from the plant parts have been a prominent part of the dyeing industry. But, they have slowly faded into oblivion, and hence synthetic dyes emerged as a new favorite during 1856-1900, as they imparted fast and reproducible shades along with the efficient reduction in cost of dyeing.

The textile processing industry is one of the major environmental pollutant generating industries, as the effluent from these industries contains a heavy load of chemicals including dyes used during textile processing. These effluents are a serious threat to aquatic ecosystems causing harmful ecological problems such as benthic photosynthesis and carcinogenicity[1]. It is estimated to be 10,000,000 tons per annum, causing serious health hazards and disturbing the eco-balance in nature[2]. As consumers have become more conscious towards the environment as well as the quality of the products to cater their needs, the textile industry has to revive the use of natural dyes to produce organic, non-toxic, and eco-friendly textiles. Currently, this has emerged as a new brand in the market. Thus, growing awareness of environmental problems coupled with the toxicity associated with synthetic dyes, is one of the reasons that made natural dye once again to hit the scenario of eco-friendly textile dyeing.

The natural dyes present in plants and animals are pigment molecules, which impart colour to the materials. These molecules, containing aromatic ring structure coupled with a side chain, are usually required for resonance and thus to impart colour. Resonance structure that causes displacement or appearance of absorption bands in the visible spectrum of light is responsible for colour[3]. There is a correlation of chemical structure with colour, and chromogen-chromophore with auxochrome[4]. Natural dyes as such are infamous for having limited shades, blending problems, and difficulty in reproducibility of results and being fugitive. Some of these problems have been overcome by using mordants. Mordants are metal salts that produce an affinity between the fabric and the dye[5]. Metal ions of mordants act as electron acceptors for electron donors to form coordination bonds with the dye molecule, making them insoluble in water[6].

Natural dyes can produce environmental friendly products with special aesthetic qualities imparting added value to textiles. In India, the rich biodiversity has provided us plenty of raw materials. Textiles in the form of apparels and accessories depending on its composition, construction and dyeing provide protection against UV radiation, which damages the

[a]Corresponding author.
E-mail: sujata.kawlekar@gmail.com

skin at molecular level. Ultraviolet protection factor (UPF) of fabrics dyed with synthetic dyes have been known very well and also reported too. Recently, UPF attributed to the textiles dyed with natural dyes have been reported as it was found that natural colorants can also increase UV protective property of the fabric.

Coconut comes from the family *Arecaceae* (*Palmae*) and subfamily *cocoidaeae*. It is an important oilseed crop that is cultivated in tropical and subtropical areas for its multiple uses. Coconut plays an important role in contributing to India's GDP. During 2019-20, approximately 23000 million coconut nuts have been produced in the country[7]. Tender coconuts account for 15 % of the total production in India, i.e. approximately 3450 million units. At global level, coconut palms are grown in more than 90 countries and territories of the world. Major part of the world production is grown in tropical Asia with Indonesia, Philippines and India accounting for over 72% of the world total[8]. The husk from the tender coconut is a waste material and also possesses disposal problems, as approximately 0.8-1.0 million tons of husks is produced every year. Tender coconut husk is a heavy stainer and a cheap raw material and this characteristic could be used to dye the cotton fabric. Kashyap *et al*.[9] have used coconut husk in powdered form as a raw material for the dyeing the cotton fabric. Sueli *et al.*[10] have extracted phenolic compounds from coconut husk. In the present study, attempt has been made to use fresh tender coconut husk for the extraction of dye and to apply it on cotton fabric to obtain various colors with the help of mordants. The aqueous extract of green coconut husk contains a huge amount of poly-phenolic materials[11] that may act as a coloring matter.

2 Materials and Methods

Ready for dyeing (RFD) grade 100% cotton cloth with EPI 135(43s), PPI 70(41s), and areal density of 123 GSM was purchased from M/s. Kiran Threads, Vapi, Gujarat. Laboratory grade chemicals, tannic acid, alum, copper sulphate, and ferrous sulphate used as mordants, were procured from Fischer Scientific Company. The raw material used as the dye (the fresh tender coconut shells) was bought from a coconut vendor in the local market. The tender coconut shell was immediately peeled and used fresh for the extraction of the dye. The exocarp and mesocarp parts of the shell were used as a raw material for the extraction of dye.

In a separate experiment, its moisture content was determined by drying the weighed sample at 103°C in an oven and the taking out difference between initial weight and oven dry weight of the sample. Similarly, water soluble matter of the green husk was determined by boiling the weighed sample with water in a flask connected to the condenser for 60 min. The flask was stoppered immediately and cooled to room temperature at 30°C after 60 min. After cooling, it was filtered immediately in a tared beaker and evaporated to dryness and finally the residue was dried to constant weight at 105°C. The percentage of water soluble matter was obtained by calculating the difference between weight of residue and initial weight of sample.

2.1 Extraction of Dye

Tender coconut husk was boiled in water, with material-to-water ratio of 1:2 for one hour while maintaining the volume of water. The extract was then filtered and used as such for dyeing of cotton textile.

2.2 Characterization of Tender Coconut Husk Extract

The *p*H of this extract was measured by a *p*H meter. The tannin content of the extract was estimated with spectrophotometric method[12] using Folin-Denis reagent, a solution of sodium tungstate and phospho-molybdic acid and phosphoric acid. First, a standard curve was plotted for aliquots of tannic acid after taking the absorbance measurements at 760 nm using UV visible spectrophotometer. Then using this standard curve, tannin content was calculated for the extract.

To characterize the poly-phenolic compounds present in the extract, it was evaporated and dried completely. This dried extract was pelletized using potassium bromide and its FTIR spectra were recorded in the range 4000 - 650 cm^{-1} at 4cm^{-1} resolution and 50 scans using Shimadzu IR Prestige FTIR spectrophotometer.

2.3 Primary Mordanting

The cotton fabric was pre-mordanted with tannic acid and alum (TAAl process). For this purpose, the ready for dyeing fabric was first treated with 5% (owf) tannic acid for 30 min at boiling condition and rinsed with water, and then further treated with boiling alum (5% owf.) till the alum solution became cold. The fabric was then rinsed with water and air dried. Since the tender coconut husk extract is supposedly abundant in tannins, this extract was also used in place of tannic acid for mordanting the fabric prior to mordanting with alum to study its self mordanting and dyeing capacity (CNAl process).

2.4 Dyeing and Optimization of Dyeing *p*H

Dyeing for both types of mordanted fabrics was carried out in launder-o-meter at 90°C for 1h with material-to-liquor ratio of 1:20. After dyeing was over, the dyed fabrics were thoroughly washed with cold water and air dried. For optimizing the dyeing *p*H, fabric swatches were dyed at different *p*H ranging from 7.0 to 10.0.

2.5 Secondary Mordanting

In this process, the RFD fabric mordanted with tannic acid and coconut husk extract respectively with and without further mordanting with alum was subjected to the secondary mordanting process. Inorganic salts, such as ferrous sulphate and copper sulphate, were used as secondary mordants to improve the colour yield. These salts were used before dyeing (pre-mordanting), after dyeing (post-mordanting) and during the dyeing (simultaneous mordanting). Experimental plan for secondary mordant application is given in Table 1. The concentration of secondary mordants used was 5% (owf). Experimental plan with secondary mordant is given in Table 1.

2.6 Evaluation of Colour Parameters, UPF Factor and Colour Fastness of Fabrics

The *K*/*S* and colour coordinates values of control and mordanted and dyed fabrics were determined using Premier Color-scan reflectance Spectrophotometer. UPF of un-dyed and dyed fabrics were measured using Labsphere UV transmittance analyzer as per AATCC: 183-2004.

The color fastness properties of the dyed fabrics were determined using standard methods. Color fastness to washing was done as per Test 2 of ISO: 105 C10:2006. Color fastness to light was carried out as per IS 2454:1985. Color fastness to rubbing and perspiration were carried out as per IS 766:1988and IS971:1983 respectively.

3 Results and Discussion

3.1 Moisture Content, Water Soluble Matter and Tannin Content

The moisture content of the tender coconut husk is found to be 82%. The water soluble matter content of the husk is found to be 5.9%. The testing of tannin content shows that the extract has tannin content of 3600ppm.

3.2 Extraction and Characterization of Dye

The FT-IR spectra of the tender coconut husk extract is given as Fig. 1. The spectrogram shows more than five major peaks attributing to the complex chemical constitution of the extract. The presence of distinctive absorption frequencies is the typical of a mixture of compounds[13]. Table 2 shows major peaks and their chemical constituents. It can be concluded that the extract might be a mixture of tannin, flavonoids as a major constituents.

3.3 Dyeing Potential of Coconut Husk Dye and Optimization of Dyeing *p*H

Aqueous extract of tender coconut husk is found to have a *p*H of 6.08. Very light colour is produced on cotton fabric when dyeing is carried out at this *p*H. The alkaline *p*H seems to be more suitable for dyeing with this dye. The *K*/*S* values of fabrics dyed at *p*H values ranging from 7.0 to 10.0 for both TAAl and CNAl processes are presented in Fig. 2. It is observed that both the mordanting processes show higher *K*/*S* values as dye-bath *p*H starts turning alkaline. CNAl process where the extract itself is used as tannin source shows highest *K*/*S* at 9.0 *p*H. These values decline thereafter and at *p*H 10 the *K*/*S* value becomes almost equal to the value at *p*H 7. In case of TAAl mordanting process where tannic acid is used as tannin source, highest *K*/*S* value of 5 is obtained at *p*H 9.5, although it is only marginally higher than the value obtained at *p*H 9. As the value of *p*H is increased further, the *K*/*S* value comes down in this mordanting method too. Regarding the comparative performance of both mordanting material, there is very little difference between the processes up to *p*H 8.0, but from *p*H 8.5 TAAl mordanting process shows higher colour strength with highest difference being observed at the *p*H value of 10. The differences observed may be due to the presence of various mineral and organic substances apart from tannins in coconut husk extract.

3.4 Primary Mordanting and Dyeing

The results of the dyeing experiment with coconut husk extract at the optimized *p*H of 9-9.5 without any mordant produce light pink colour. Primary mordanting with tannic acid + alum (TAAl) as well as extract + alum (CNAl) produces pinkish red colour on the fabric. The *K*/*S* value and colour coordinate values of the dyeing experiments with both type of mordanting processes at *p*H 9 and 9.5 is given in Table 3. The extract has been used as a self-mordant in CNAl process as it has tannin content; the idea is to avoid commercial tannic acid, in case the colour obtained with the extract as a mordant is better. Though the dyeing of the fabric mordanted with extract and alum also results in good colour, the depth of the shade is lesser as compared to fabric mordanted with tannic acid and alum, as indicated by the L,a,b values. Similarly, both the techniques impart 50+ UPF rating to the fabric, however the fabric mordanted with tannic acid and alum shows more UPF rating (Table 3).

3.5 Secondary Mordanting and Dyeing

Tannic acid and alum mordants impart a very good colour to the fabric. But, the fabrics mordanted with secondary mordants show higher *K/S* (Fig. 3). Fabrics

Table 1 — Experiments done with secondary mordants

Exp. No	Pre-mordanting	Simultaneous mordanting	Post mordanting
1	Tannic acid→ iron→ dyeing with husk Extract	–	–
2	–	Tannic acid→ iron+ dyeing with husk extract	–
3	–	–	Tannic acid→ dyeing with husk Extract→ iron
4	Tannic acid→ alum→ iron→ dyeing with husk Extract	–	–
5	–	Tannic acid→ alum→ iron+ dyeing with husk extract	
6	–	–	Tannic acid →alum→ dyeing with husk extract→ iron
7	Husk extract→ iron→ dyeing with husk extract	–	–
8	–	Husk extract→ iron+ dyeing with husk extract	–
9	–	–	Husk extract→ dyeing with husk extract →iron
10	Husk extract→ alum → iron→ dyeing with husk extract	–	–
11	–	Husk extract→ alum→ iron+ dyeing with husk extract	–
12	–	–	Husk extract →alum→ dyeing with husk extract→ iron
13	Tannic acid→ copper→ dyeing with husk extract	–	–
14	–	Tannic acid→ copper→ dyeing with husk extract	–
15	–	–	Tannic acid→ dyeing with husk extract→ copper
16	Tannic acid→ alum→ copper→ dyeing with husk extract	–	–
17	–	Tannic acid→ alum→ copper+ dyeing with husk extract	–
18	–	–	Tannic acid→ alum→ dyeing with husk extract→ copper
19	Husk extract→ copper→ dyeing with husk extract	–	–
20	–	Husk extract→ copper+ dyeing with husk extract	–
21	–	–	Husk extract→ dyeing with husk extract→ copper
22	Husk extract→ alum→ copper→ dyeing with husk extract	–	–
23	–	Husk extract→ alum→ copper+ dyeing with Tender coconut extract	–
24	–	–	Husk extract→ alum→ dyeing with husk extract→ copper

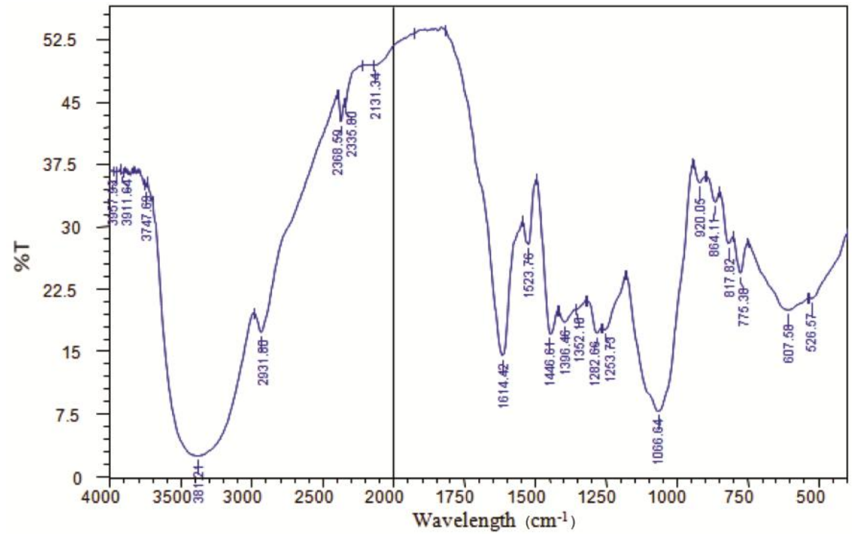

Fig. 1 — FT-IR spectra of coconut husk extract

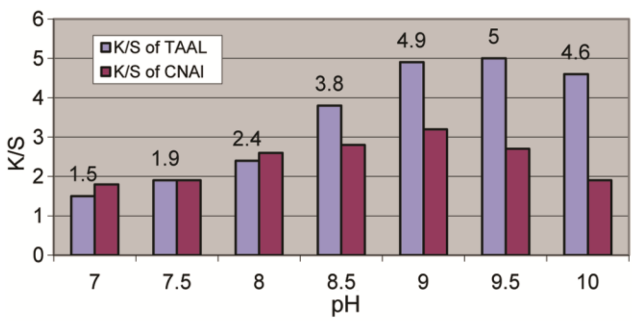

Fig. 2 — Optimization of pH of dye-bath (TA - tannic acid, Al -alum, CN -extract)

Table 2 — Major peaks in the FT-IR spectrogram of green coconut husk

Major peak areas, cm^{-1}	Possible chemical constituent
3380	-OH group stretching [14,15]
2931	Symmetric and asymmetric –C-H-stretching vibrations of CH2 and CH3 groups[15,16]
1614 - 1620	Axial deformation of C=C bond of aromatic ring[14,15]
1446	Stretching of aromatic ring vibrations
1352, 1282, 1252	-OH in-plane bend[16]
817and 920	Out of plane aromatic CH bending.

Table 3 — Colour parameters and UPF values of fabric dyed with husk extract

Mordant	*p*H	*K*/*S*	L*	a*	b*	UPF (rating)
Tannic acid→ alum	9.0	4.0	52.9	18.4	25.6	365
Extract→ alum	9.0	3.0	49.9	20.7	27.7	186
Tannic acid→ alum	9.5	4.7	55.1	17.6	24.3	427
Extract→ alum	9.5	2.6	47.9	22.5	29.8	151
Control	-	-	-	-	-	6

mordanted with ferrous sulphate are dyed to different shades from grey to black. Similarly, those mordanted with copper sulphate are dyed to different shades of brown. Tannins and metal salts can form coordination complexes with dye and bind it to the cotton fabric. Secondary mordanting results in increase in colour depth as compared to single mordanting. This could possibly be due to the reason that the metal salts bind the maximum number of molecules of natural dye to the fabric. Mordants help binding of dyes to fabric by forming a chemical bridge from dye to fiber and forming an insoluble compound of the dye within the fibre[17]. Thus, a complex is formed among the fibre, mordant and dye system[18] by the metal salt on which one site is with the fibre and the other site is with the dye.

It is observed that ferrous sulphate mordanted fabric has higher *K*/*S* when it is pre-mordanted with tannic acid without alum, whereas in case of copper sulphate it is the other way round. In case of coconut extract pre-mordanting, samples with alum show higher *K*/*S* in case of both ferrous sulphate and copper sulphate. From all the three mordanting methods, the simultaneous mordanting method gives excellent results. Simultaneously mordanted and dyed fabrics display the best shades among all the samples.

3.6 UV Protection Factor

UPF of un-dyed fabric is 6.22. UPF of all the samples mordanted, dyed and secondary mordanted is found 50+. The results in Table 3 indicate that the tender coconut husk, apart from colouring the fabric, also provides additional functional property of UV protection.

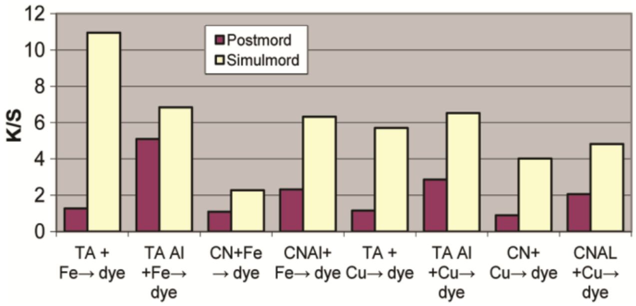

Fig. 3 — Effect of mordanting on *K/S* (TA —tannic acid, Al —alum, CN —extract, Fe — ferrous sulphate, and Cu — copper sulphate)

3.7 Colour Fastness Properties

Colour fastness to washing for the samples pre-mordanted with tannic acid+alum and extract+alum is found very good with negligible stain on adjacent fabrics (4/5). Colour change in case of tannic acid+alum is found towards darker shade. Wet rubbing rating is 4. In case of colour fastness to light the change in colour shows grade 4. For fastness to perspiration the staining is found negligible (4/5) and shade change is found towards darker. The fabric mordanted with tannic acid + alum as well as extract+alum has over all very good fastness properties.

In case of fabrics mordanted with secondary mordants, similar trend in fastness properties is observed. The wash fastness rating is found very good (4/5) in almost all the samples with negligible staining, except for fabric post-mordanted with iron which shows change in colour rating to 2. Fastness to light is also found 4 in all the samples. Both dry and wet rubbing are very good (4/5) in case of all the fabrics, except the fabrics mordanted with copper in the simultaneous mordanting, where the rating of dry rubbing is found in the range of 4/5 - 4 and wet in the range of 3/4 - 3. In case of fastness to perspiration, the staining on adjacent fabrics is 4/5 in most of the cases, but in case of copper mordanted fabric in simultaneous mordanting it is in the range of 4 - 3. The change in colour is in the range of 4 - 2 for alkaline perspiration for both iron and copper mordanted fabrics in post mordanting technique. Copper mordanted fabrics in many cases show change in colour towards darker.

3.8 Final Scale-up Trial

The scale-up trial has been carried out with the same cotton fabric. Cotton fabrics of 4m x 14.5 inches size are dyed in rope dyeing machine and pressurized vessel. The scale up trials are undertaken firstly with dyeing of the tannic acid + alum as well as husk extract + alum mordanted fabrics with coconut extract. Further, as the fabric dyed using simultaneous mordanting and dyeing methods displays more prominent colors, method numbers 2, 17 and 23 from experiments (Table 1) are selected for the scale up trials using secondary mordants. The scale-up trials produced amazing results as the myriads of bright colours are obtained on the fabrics. The dyed fabrics were subjected to different fastness tests. The results obtained have followed the trend displayed by the samples dyed at lab-scale.

4 Conclusion

The results have demonstrated the potency of tender coconut husk as a source of natural colouring agents for dyeing of cotton textiles. In this study, the husk extract produces very good shades on the cotton fabric having reasonably better fastness properties and imparted it with excellent UPF functionality as well. There are several phenolic compounds in coconut shell which can be isolated and identified through the simple isolating process. A systematic study of extraction, characterization and improving the dyeing technique can be devised with minimum cost investment, yield maximization and dye purity. The dyes are eco-friendly and safe only when they are easily biodegradable, having no health hazard effects. In this case, the extracted husk can be easily degraded and utilized as manure.

References

1 Adeel S, Ali S, Bhatti I A & Zsila F, *Asian J Chem*, 21(5) (2009) 3493.
2 Goodarzian H & Ekrami, *World Appl Sci J,* 8(11) (2010) 1387.
3 Mahapatra N N, *Textile Dyes* (Woodhead Publishing Pvt Ltd, New Delhi, India), 2016.
4 Purohit A, Mallick S, Nayak A, Das N B, Nanda B & Sahoo S, *Current Sci*, 92(12) (2007) 1681.
5 Papadaki S G, Krokida M C, Economides D, Vlyssides A G & Koukios E G, *Chem Eng Transact*, 29 (2012) 7.
6 Mongkholrattanasit R, Kryštůfek J, Wiener J & Viková M, *Fibres Text East Eur*, 86(3) (2011) 94.
7 https://economictimes.indiatimes.com/markets/commodities/news/coconut-output-drops-10-to-lowest-in-4-years/articleshow/72505025.cms?utm_source=contentofinterest&utm_medium=text&utm_campaign=cppst, ET Bureau (accessed on December 13, 2019).
8 Raghavi M D, Sakthi Balaa M, Surender S, Lokesh P & Kalidas K, *Int J Res Business Management*, 7(1) (2019) 1.
9 Kashyap R, Sharma N, Sharma L& Divya, *Int J Sci Technol Eng*, 3 (04) (2016) 92.

10 Sueli R & Gustavo A S P, *J Food Eng*, 80 (2007) 869.
11 Akhter A, Zaman S, Umar Ali M, Ali M Y & Jalil Miah M A, *J Sci Res*, 2 (1) (2010)186.
12 *AOAC Official Method 952.03*, 17th edn, edited by W Horwitz, Vol. II (AOAC International, *Gaithersburg*, M D, USA) 2000, 17.
13 James I, Gabriel O & Olubunmi A, *Res J Appl Sci*, 4(6) (2009) 217.
14 Morbeck F L, Lelis R C C, Schueler M V E, Santos W A, Sampaio D A, Couto da Silva B, Morais R deM & Santana G M, *Matéria (Rio J)*, 24(3) (2019) 1. https://doi.org/10.1590/S1517-707620190003.0748.
15 Pantoja-Castroa A M & González-Rodrígueza H, *Rev. Latinoamer Quím*, *3*9(3) (2011) 107.
16 Nandiyanto A B D, Oktani R & Ragadhita R, *Indonesian J Sci Technol*, 4(1) (2019) 97.
17 Vankar P S, *Resonance*, October (2000) 73.
18 Samanta A K, Konar A, Chakraborti S & Datta S, *Indian J Fibre Text Res*, 36(1) (2011) 63.