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A STUDY OF SUSTAINABLE COLORATION OF LYOCELL FABRICS USING EXTRACTS OF TROPICAL ONION SKINS

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Abstract. Lyocell is considered as a new fiber that represents a milestone in the development of environmentally sustainable textiles. Following the concept of lower the environmental impact of fashion clothing, this study aims to determine the suitable natural dyes recipes with the color extracting from tropical onion skins. Colorants were extracted by dissolving crushed dry onion skins in boiled water at 100 ºC for 20-25 minutes. The extracting ratios are 1:25 and 1:30, respectively. The optimal dyeing conditions were found out at 80 ºC, 45 minutes with 75 % v/v. In addition, a variety of the most common mordants including Potassium aluminum sulfate, Copper (II) sulphate and Iron (II) sulphate was used. The dyeing efficiency was evaluated via color strength (K/S) and color values CIE L*a*b*. The results showed that mordants do not only change the color shade but also improve color fastness to washing, crocking and light, which can be applied in commerce.

Keywords: lyocell, natural dyes, onion, colors, mordant.

Classification numbers: 1.3.4, 2.7.2.

1. INTRODUCTION

In the field of regenerated cellulosic fibres, lyocell fiber (U.S. brand name Tencel), has been the breakthrough textile material over the last 20 years, receiving several awards for its environmental-friendly process. Also made from wood pulp, but lyocell is only made by dissolving wood pulp in amine oxide solution of hot N-Methylmorpholine-N-oxide (NMMO), a cyclic amine oxide that has proved to be an excellent solvent for cellulose [1]. In addition, scientific investigation showed that superior moisture management of lyocell is essential as it keeps the body dry and generates a kind of natural air conditioning [2]. Lyocell fiber is famous for its better strength than any other man-made cellulosic fibres in wet state [3]. Lyocell fabric is very stable in washing and drying, easy to dye to deep vibrant colours and very comfortable to wear [4, 5]. People added more natural values to lyocell by using natural dyes for colouring fabrics. Although the use of metallic mordant is not always eco-friendly, the pollution problems created by metallic mordants are of very low order and can be easily overcome. Many scientists obtained several significant results in dyeing cellulosic fibers, even this work need more attention in pre-treatment, processing and mordanting [6]. Among cellulosic fibers, alum mordanted cotton fabric exhibited the highest colour yield, while flax required more dyestuff for the same depth of shade as for cotton [7]. Dyeing with thyme and pomegranate peel extracts found significant bacterial reduction for dyeing of unmordanted cotton fabrics. Moreover, the pre-mordanting process increased the color efficiency for both thyme and pomegranate fruit peel dyed cotton fabrics [8].

However, there are only a few studies on dyeing lyocell with natural colorants and less experiences in processing compared to traditional cotton natural dyeing, especially in Vietnam where lyocell is still new and advanced fiber in textile industry and commercial usage. A study on dyeing Tencel fiber with Turkish red pine bark extract found the low colour strength at 14.4 and 5.9 [9]. A research on dyeing with extracts pomegranate peel in Tencel fabrics indicated the importance of using ferrous sulphate as mordants, resulting in light fastness, washing fastness and rubbing fastness properties through premordanting methods [10, 11]. Another approach to colour lyocell fabrics by screen printing using extracts of leaves and bark from eucalyptus found the important role of mordanting pretreatment of the lyocell with alum [12]. Following the attempt to colour lyocell fabric with natural colorants, this study aims to determine the optimal dye recipes with the colour extracting from Vietnamese onion skin waste. Onion skin extract can provide several brown hues to natural fibers and it is popular waste in Vietnam. It was identified that quercetin is the most abundant flavonoid found in many onion species [11], therefore we only focused on finding optimal extracting and dyeing conditions, and dyeing process with mordants. Colorants were extracted by dissolving crushed dry onion skins in boiled water. The results were confirmed by measuring colour strength (K/S), color values (CIE L*a*b*) and colour fastness to washing, crocking and light following ISO standards.

2. MATERIALS AND METHODS

2.1. Materials

The commercial 100 % lyocell fabric were supplied by Viet Thang Co, Viet Nam (plain woven, 147 g/m^2, warp density 68 threads cm^{-1}, weft density 44 threads cm^{-1}, scoured and bleached). Aluminum potassium sulfate ($AlK(SO_4)_2.12H_2O$), ferrous sulfate ($FeSO_4.7H_2O$), and copper(II) sulfate pentahydrate ($CuSO_4.5H_2O$) supplied by AR, China. Onion skin was collected from a domestic market in Ho Chi Minh city, Viet Nam.

2.2. Dye extraction from onion skin

Onion skins were dried in sunlight and then crushed to convert into powder form. That powder was the raw material for dye extraction. The dried onion skin powder (5 g) was extracted with 125 ml, 150 ml, 175 ml of distilled water with liquor ratios of 1:25, 1:30, 1:35 at 100 ºC for 20-25 minutes. They were then filtered to obtain dyeing solution.

2.3. Dyeing process

The dyeing processes were carried out in Laboratory Mesdan S.p.A IR dyeing machine and a drying machine (Italy) was used for the drying of the dyed fabrics. Four concentrations of dried onion skin extracting solutions were chosen: 25, 50, 75 and 100 % v/v. The concentration of 75 % v/v dyeing solution was chosen to investigate the effect of different mordants, temperatures (60 ºC, 70 ºC, 80 ºC), times (30 mins, 45 mins, 60 mins) and pH (4, 5, 7, 8) on the dyeing process. Liquor ratio of dyeing was 1:30. After dyeing, the samples were washed with deionized water to remove any unfixed dye and then dried at 60 °C.

2.4. Mordanting process

In this experiment, three types of mordants with the different concentrations and meta mordanting techniques were applied. These laboratory–grade mordants were used: aluminum potassium sulphate ($AlK(SO_4)_2.12H_2O$), ferrous sulfate ($FeSO_4.7H_2O$), and copper(II) sulphate pentahydrate ($CuSO_4.5H_2O$). Mordanting concentration were 1 % w/v, 5 % w/v, and 10 % w/v.

2.5. Colour measurement and colour fastness testing

CIE L*a*b* values were measured using spectrophotometer (Xrite Color i5). The L* value shows luminance in CIELab colour space and its values from 0 (black) to 100 (white). The +a* value indicates red and -a* value indicates green, while +b* value indicates yellow and -b* value indicates blue. The colour strength of the dyed samples was evaluated using the K/S values generated by the spectrophotometer (Xrite Color i5). K/S is a function of colour depth and is calculated by the Kubelka–Munk equation, $K/S = (1 - R)^2/2R$, where R is representative of surface reflectance, K is light absorption coefficient, and S is the light scattering coefficient. The colour fastness to washing and rubbing of the dyed samples was determined according to ISO 105-C06 A1S: 1994 and ISO 105-X12: 2001, respectively.

3. RESULTS AND DISCUSSION

3.1. Effect of extraction rate and productivity of onion peels extracted in distilled water to the colour of fabric

Samples after dyeing with different extraction rates were washed, dried at room temperature and measured color values (CIE L*a*b*). The measurement results in Table 1 showed that the color intensity K/S of fabric after being dyed with different extraction solutions decreased following the declination of extraction rate. In particular, K/S data of 1:25 extraction rate achieved the best color intensity. The highest extraction efficiency reached 84 % when extracted at 1:35 and the lowest was 76.80 % when extracted at 1:25. The evaporation and dehydration of 3 extraction times are nearly equal to each other with no significant difference.

Table 1. Effect of extracting rates on the colour of fabric.

Extracting rates	L*	a*	b*	C	ΔE	K/S	Extracting productivity (%)
Standard samples	90.30	-0.51	2.44	2.49	-	0.07	
1:25	-31.86	10.78	15.11	17.84	26.05	17.13	76.80
1:30	-29.05	10.03	14.46	16.90	24.52	16.74	80.67
1:35	-28.00	11.02	15.22	18.06	25.39	16.44	84.00

3.2. The effect of dyeing conditions (concentration, temperature, time and pH) on colour of lyocell dyed fabric.

To assess the effect of dyeing conditions (concentration, temperature, time and pH), the changing parameters were assessed via colour values (CIE L*a*b*) and color strength values (K/S). The colour differences ΔE compared to standard samples in turn are shown in Table 2 and Table 3. The clear colour changes could be seen via ΔE values following the changes in concentrations and temperatures, especially at 70 °C.

Table 2. Effect of dyeing concentrations on the colour of dyed fabric.

Concentration (v/v)	L*	a*	b*	C	ΔE
Standard sample	90.74	-0.11	3.09	3.10	-
100 % v/v	-27.29	9.16	18.15	19.99	27.11
75 % v/v	-21.49	6.83	18.67	19.67	25.91
50 % v/v	-14.65	3.36	18.52	18.75	24.01
25 % v/v	-8.99	0.92	17.84	17.82	22.46

Table 3. Effect of dyeing temperature on the colour of dyed fabric.

Temperature (°C)	L*	a*	b*	C	ΔE
Standard samples	89.58	-0.10	2.91	2.90	-
60 °C	-18.56	6.78	17.75	18.80	24.19
70 °C	-20.67	2.11	25.01	25.08	31.57
80 °C	-20.03	7.17	19.83	20.90	26.79

Figure 1 showed a decrease in the K/S value when reducing the concentration from 100 % v/v to 25 % v/v, the highest K/S is about 14.54 at 100 % v/v. However, the use of 100 % v/v concentration did not have a significant colour difference from the concentration of 75 %, leading to the use of 75 % concentration to both achieve colour value and material saving. The temperature of 80 °C showed the best dyeing effect. We did not raise the temperature to 90 °C - 100 °C because this is the temperature that could affect the colorants in natural dyes [6, 8].

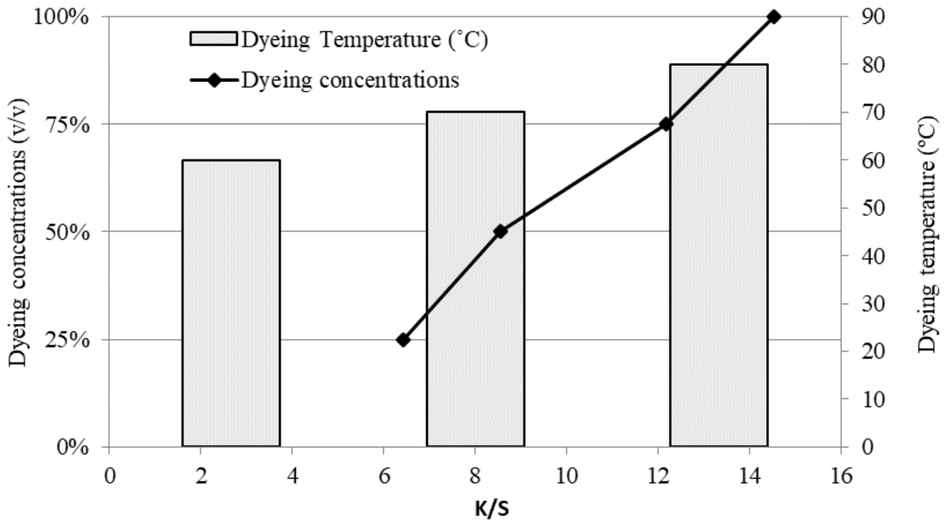

Figure 1. Effect of dyeing concentrations and temperatures on colour strength of fabric.

Table 4. Effect of dyeing time on the colour of dyed fabric.

Time	L*	a*	b*	C	ΔE	K/S
Standard sample	90.18	-0.11	2.72	2.72	-	0.08
30 mins	-19.74	4.80	20.64	21.10	27.21	11.31
45 mins	-22.76	7.48	19.96	21.12	27.67	11.52
60 mins	-17.35	4.14	22.63	22.95	29.91	9.23

Table 4 showed that dyes tend to attach good colour to lyocell fabric when dyeing at 30-45 mins at 80 °C, but the long dyeing time could reduce the effect of colouring on the fabric due to the fact that water-soluble colours are affected at certain temperatures and times. Moreover, the colour stain observed on the fabric surface for dye samples in the 60 minutes' period clearly showed this observation.

In order to assess the influence of the dyeing environment (pH) on the ability to colour the lyocell fabric, the dyeing process is performed with the parameters: 1:25 extraction rate, 1:30 dyeing ratio, 75 % v/v concentration, temperature 80 °C, 45 minutes dyeing time in turns of dyeing environments including acid environment (pH ≤ 4), neutral acid (pH 4.5 ÷ 5.5), neutral (pH ≈ 7) and basalt environment (pH ≥ 9). The results are shown in Table 5 and Figure 2.

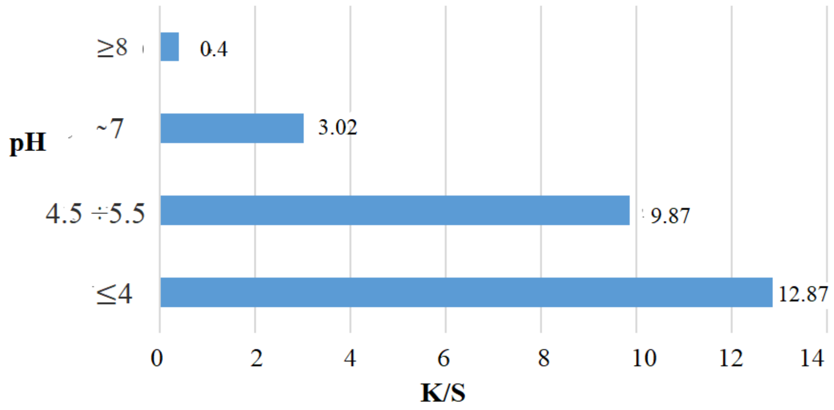

Figure 2. Effect of pH condition on colour of dyed fabric.

Table 5. Effect of dyeing pH on colour of dyed fabric.

pH	L*	a*	b*	C	ΔE	K/S
Standard sample	89.89	-0.21	3.05	3.06	-	0.07
pH ≤ 4	-24.53	8.26	17.24	18.77	24.77	12.87
pH 4.5 ÷ 5.5	-19.40	7.93	20.36	21.59	27.38	9.87
pH ≈ 7	-14.80	6.60	15.44	16.51	21.06	3.02
pH ≥ 9	-6.96	3.96	6.11	6.84	9.24	0.40

Table 5 showed that the colour absorption on the fabric is very susceptible to the pH environment, the changing pH would easily change the colour intensity. The higher the pH, the lower the colour intensity. The highest colour intensity (K/S = 12.87) is in the acidic environment and the lowest colour intensity is in basalt medium with pH ≥ 8 (K/S = 0.4).

3.3. Evaluate the effect of mordants on colours of dyed fabrics

The influence of mordants on the dyeing absorbency was investigated with fixed conditions: concentration of 75 % v/v, 80 ºC, 45 minutes, pH ≤ 4. The simultaneous mordanting method were used with different mordants ($KAl(SO_4)_2.12H2O$, $CuSO_4.5H_2O$, and iron sulphate ($FeSO_4.7H_2O$), and comparing to non-mordant samples.

Table 6. Effect of mordanting on colour of dyed fabric.

Mordants	L*	a*	b*	C	ΔE	K/S
Standard samples	89.86	-0.44	3.03	3.06	-	0.07
$KAl(SO_4)_2.12H_2O$	-28.25	-0.12	37.89	37.94	47.26	11.88
$CuSO_4.7H_2O$	-26.64	6.95	31.71	32.29	40.44	6.64
$FeSO_4.5H_2O$	-28.36	-0.72	9.39	9.41	15.14	2.39
Non-mordanting	-22.19	7.17	19.40	20.36	26.29	12.75

As shown in Table 6, the highest K/S value for $KAl(SO_4)_2.12H_2O$ mordant is 11.88, while the K/S of non-mordant dyed fabric is 12.75. The difference in delta E between the dyed sample and the standard sample reached the highest value of 47.26 when using $KAl(SO_4)_2.12H_2O$. The reason is that when extracting red onion peels in distilled water containing tannin, this compound tends to change the colour of the dyes. Especially, the colour sample with iron salt $FeSO_4.7H_2O$ will be coloured from grey to black. The Fe^{2+} ions reacted with tannin to form iron (II), which is dark green and turns in black when exposing to air. Mordant with $CuSO_4.5H_2O$ gave a yellowish brown colour, brighter and evenly coloured, as they formed insoluble complexes with the dyes, giving a lighter colour and raising the colour tone to a more yellowish direction. Meanwhile, mordant alum salt helps to obtain the best brightness. The brightest colour compared with other mordants is due to the acting of Al^{3+} ions with tannin, leading to yellow green colour. The use of mordant with $KAl(SO_4)_2.12H_2O$ helped to improve colour stability and colour intensity.

3.4. Colour fastness to washing, to crocking and to light of lyocell fabric dyed with purple onion peel extract

Table 7. Colour fastness of fabrics dyed with extracted solutions of onion peels.

<table>
<tr><th rowspan="2">Standards</th><th rowspan="2">Testing condition</th><th colspan="2">Mordanting</th></tr>
<tr><th>No mordant</th><th>$KAl(SO_4)_2.12H_2O$</th></tr>
<tr><td>Color fastness to light ISO 1302:2014</td><td>Up to grade 4</td><td>3-4</td><td>4</td></tr>
<tr><td rowspan="8">Color fastness to washing ISO 105 C06:2010
Detergent: 0.4 % ECE (B); 0.1 % $NaBO_3.4H_2O$</td><td>40 ^oC, 30 mins, 10 steel balls</td><td>3</td><td>3-4</td></tr>
<tr><td colspan="3">Color staining on multi-fiber</td></tr>
<tr><td>Di-acetate</td><td>4-5</td><td>4-5</td></tr>
<tr><td>Cotton</td><td>4-5</td><td>4-5</td></tr>
<tr><td>Polyamide</td><td>4-5</td><td>4-5</td></tr>
<tr><td>Polyester</td><td>4-5</td><td>4-5</td></tr>
<tr><td>Acrylic</td><td>4-5</td><td>4-5</td></tr>
<tr><td>Wool</td><td>4-5</td><td>4-5</td></tr>
<tr><td rowspan="2">Color fastness to Crocking ISO 105 X16:2016</td><td>Dry</td><td>4-5</td><td>4-5</td></tr>
<tr><td>Wet</td><td>4</td><td>3-4</td></tr>
</table>

Colour fastness is assessed generally by comparing any staining of specified adjacent fabrics during the test with a set of standard ‘grey scales’. A numerical grading is given on a scale of 1–5, where 1 is very poor and 5 is excellent. The results in Table 7 exhibited the colour fastness to all important impacts: light, washing and crocking. The tabled confirmed that samples reached the grade of 4 to 4-5, which is deemed acceptable for commercial use. However, care should be taken to natural dyes fabrics; those are very sensitive for colour fading.

4. CONCLUSION

Our study showed that the optimal conditions for natural dyeing of lyocell fabric with extract from purple onion peels in distilled water are: extraction rate is 1:25 (g/ml), dyeing ratio 1:30, concentration of 75 % v/v, temperature of 80 ºC, 45 minutes and using mordant of $KAl(SO_4)_2.12H_2O$ by simultaneous mordanting method. When using a dyeing solution from distilled water to dye lyocell fabric with a non-mordant method, the colours of the samples are pink, and became greenish gold. Mordants are not only improve the darkness and colour, but also bring many different colour shades. Colour fastness to all important criteria of textile products such as washing, crocking and light achieved a good average of 3-4 or 4-5, indicated that they could be used in commerce. However, natural coloured products always have a relatively short life-time compared to synthetic dyed products.

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