Indigenous Natural Indigo Dye for Denim Industry

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***Abstract*: Presently, denim industry is the fastest growing textile manufacturing sector in Pakistan and it has a promising local and international market for work and fashion clothings of all age groups. Synthetic indigo dye is widely used for denim dyeing which is toxic due to presence of aniline compounds. Due to increasing awareness and control for sustainable manufacturing, products and fashion brands, manufacturers and scientists have been looking for the alternatives. Natural indigo dye has been one of those alternative options. However, the natural indigo dye has been used only in the cottage industry so far. The present research includes the application of natural indigo dye extracted from indigenous indigofera tinctoria plants for dyeing of undyed denim fabric. The indigo dye extraction process was developed in collaboration with Archroma Pakistan Ltd. to make the dye suitable for dyeing procedures practiced in denim industry. The dyeing parameters of industrial scale dyeing procedure (i.e. pre-wetting, dyeing and rinsing) were optimized using the developed dye. The colour strength (K/S value) and colourfastness properties of the fabric dyed with the developed natural indigo dye were better than the synthetic dye.**

***Keywords*: Denim fabric, Dyeing, Indigofera tinctoria, Natural Indigo Dye, Sustainability.**

I. INTRODUCTION

At present, Pakistan is one of the largest exporters of denim fabric and is investing more capital in denim manufacturing and processing sector. More than forty denim industries such as Soorty denim, Artistic Fabric Mill, Crescent Bahuman, Denim International Siddique Sons, Al-Ameen Denim mill and more produce 584 million meters finished denim fabrics, yearly [1], [2]. Denim is twill-woven cotton fabric with warp usually dyed with synthetic indigo dye [3], introduced in 1897 by BASF AG (Germany) [4], and weft is undyed/white. The synthetic dye is 96% pure, with chemical formula of $C_{12}H_8O_2N_2$, synthesized from by-product of fossil fuels, example aniline or other aromatic derivatives [5], [6]. Textile industry being one of the most polluted sectors in world [7], contributes towards the wastewater pollution [8] that contains various unsafe chemicals and dyestuff. Synthetic colourants are major contributors because usually these are either non-biodegradable, toxic and/or carcinogenic [9]. The increase in consumer awareness towards sustainable material, products and manufacturing has revived the use of natural colourants due to their biodegradable nature and less toxic characteristics [10]. Thus, dyes extracted from natural sources are recognized as an important alternative to environmentally unfriendly synthetic dyes [11], [12]. Natural indigo is a dark blue crystalline powder that is being extracted from indigofera tinctoria plants since ancient times as the traditional craft. These plants are widely cultivated in Pakistan, belongs to the Indus River Belt, and hence, are indigenous source for natural indigo dye. The dye has been used only at cottage industry so far. Therefore, this work was undertaken to develop a natural indigo dye for denim dyeing at mass scale in collaboration with Archroma Pakistan Ltd. This paper presents dyeing results of the developed natural indigo and synthetic, such as colour strength (K/S value), colour coordinates (CIE L*a*b* values) and colourfastness properties.

II. MATERIALS & METHODS

A. Material

Desized and undyed 100% cotton fabric (2/1 twill weave denim construction) and synthetic liquid indigo (Denisol Indigo 30 liquid) were obtained from Archroma Pakistan Ltd. (APL). The natural indigo dye was developed in Colour Research Lab (CRL), Department of Textile Engineering, Mehran University of Engineering and Technology (MUET) Jamshoro. Sodium hydrosulfite (SHS) and sodium hydroxide of laboratory grade were used from CRL. Other commercial auxiliaries such as dispersing agent (Ekaline F) and wetting agent (Leonil-EHC) were provided by APL.

B. Methods 1) Extraction method

Indigo dye extraction was carried out initially by traditional aqueous extraction method [13]. Freshly cut plants were immersed in water for about 14- 19 hours. The resulted liquid was stirred with paddles to mix it with air to produce indigotin dye molecule. The upper layer of liquid was removed and the settled dye paste was then filtered over a sand bed to remove leftover water. The filtered dye was pressed into cakes and dried in sunlight for trade purpose.

The method was further optimized to make it more efficient in terms of extraction time and the colour strength of the dye. The optimized method and results are not shared herein due to protecting intellectual property and confidentiality disclosure agreement between MUET and APL. *2) Dyeing method*

Following industrial indigo-dyeing method [14] for warp yarn was followed and optimized for dyeing of undyed denim fabric.

a) Industrial practice of synthetic indigo

Pre-wetting: Substrate was immersed into solution of sodium hydroxide (10 g/L) and 5 g/L leonil-EHC (wetting agent) for 30 seconds, followed by squeezing, cold-rinse and squeezing.

Dyebath preparation: Synthetic indigo dye liquid was dispersed in water as per required shade followed by addition of 5 g/L SHS and 1.5 g/L sodium hydroxide (48ºBe) and stirring.

Dyeing process: Pre-wet substrate was immersed into the prepared dyebath for 30 seconds, padded/squeezed evenly at 75% pick-up on a Mathis horizontal laboratory padder HF. Immediately after padding, the substrate was subjected to air oxidation (aeration) for 90 seconds. Padding and aeration steps were repeated four times followed by rinsing with tap water and drying.

b) Dyeing with the developed natural indigo dye

Pre-wetting of substrate and dyeing procedures are same as that for synthetic indigo dye liquid. However, following experimentation was performed for dyebath preparation.

The developed indigo dye was applied on undyed cotton denim fabric. The number of dyeings were carried out for varied dyeing temperature (20, 30, 40 and 50ºC), dipping time (15, 30, 45 and 60 sec), concentration of Ekaline F (dispersing agent) in dyebath (0, 0.5, 1, 1.5 and 2 g/L), sodium hydrosulphite concentration (10, 20, 30, 40 and 50 g/L), concentration of sodium hydroxide of 48ºBe (5, 10, 15, 20 and 25 g/L) and dye concentration (10, 15, 20, 25, 30 and 35 g/L). Finally, dyed samples were cold rinsed for 20 seconds.

C. Testing 1) Colour strength (K/S value)

Each dyed fabric sample was measured for colour strength (K/S) value on a Datacolor 850 Spectrophotometer. Each measurement was made at four different places of the sample and averaged. The K/S value was determined by using the Kubelka-Munk equation (Equation 1).

$$K/S = \frac{(1-R)^2}{2R} \quad (1)$$

Where, R is reflectance value of the dyed fabric at maximum absorption, K is absorption coefficient and S is scattering coefficient.

2) Colorimetric coordinates (CIE L a* b* values)*

In order to monitor the shade, CIE L* a* b* values were also measured on a Datacolor 850 Spectrophotometer.

3) Colourfastness assessment

Colourfastness to washing (ISO 105-C03) was carried out on a washing fastness machine. Colourfastness to light (BS 1006: 1990 UK-TN) was carried out on a Mercury lightfastness tester (SDL Atlas, USA) and colourfastness to rubbing (ISO 105-X12) was done on a Crockmeter (James H. Heal Co., UK).

III. RESULTS

A. Effect of dyebath temperature

Fig 1 shows the increase in colour strength of dyed fabric with increasing dyebath temperature. At 40ºC (usual working temperature in industry), maximum colour strength value was obtained. Further increase in temperature reduced the colour strength. This is because SHS is degraded at higher temperature [14], [15] that results in premature consumption of SHS causing lesser dye reduction.

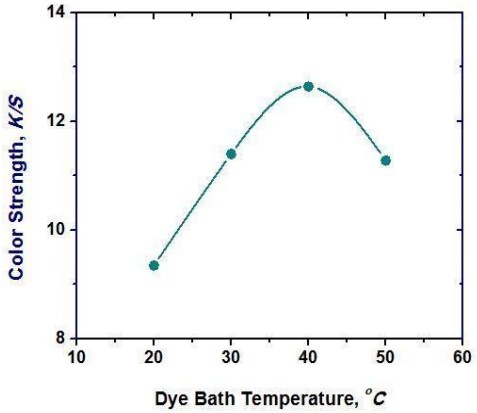

Fig 1: Effect of dyebath temperature on colour strength

B. Effect of fabric dipping time

Fig 2 shows the increase in colour strength of dyed denim fabric with increase in dipping/immersing time of the fabric in dyebath up to 45 sec, keeping dyebath temperature optimized, i.e. 40ºC. The colour strength reduced with immersion time longer than 45 sec. This is because dye penetrates more into the core of fibre [4], [13], [16]. Same happens when number of dips are increased.

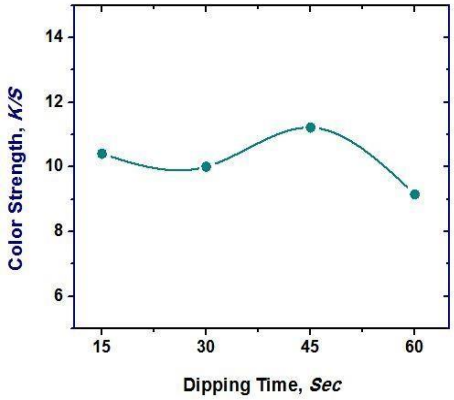

Fig 2: Effect of fabric dipping time on colour strength

C. Effect of dispersing agent

The developed indigo dye is in powder form compared to the available synthetic liquid dye which is a pre-reduced 30% solution. Therefore, natural dye is dispersed in water and dispersing agent could support the proper dispersion. However, Fig 3 shows that with no use of dispersing agent, maximum colour strength was obtained, and with even colour. The colour strength of dyed fabric reduced with increase in concentration of the dispersing agent.

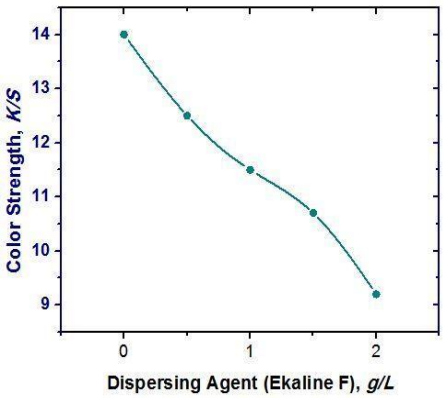

Fig 3: Effect of dispersing agent on colour strength

D. Effect of SHS concentration

Fig 4 shows increase in colour strength with increase in SHS concentration with 20 g/L sodium hydroxide (48ºBe) at dyebath temperature and optimized dipping time. However, a sudden higher value was obtained at 40 g/L SHS. Whereas, further increase in SHS concentration reduced the colour strength.

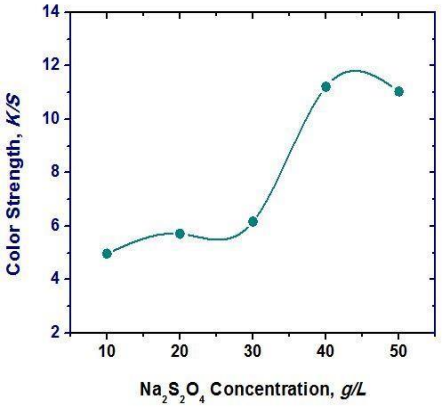

Fig 4: Effect of Sodium hydrosulphite concentration on colour strength

E. Effect of sodium hydroxide concentration

Sodium hydroxide (alkali) assists SHS to reduce the dye molecules [17]. Fig 5 shows that the maximum colour strength was obtained at 20 g/L. Further increase in the concentration decreased the colour strength. This may be attributed to the increased dyebath pH causing more penetration of the dye into the fibre [4], [13].

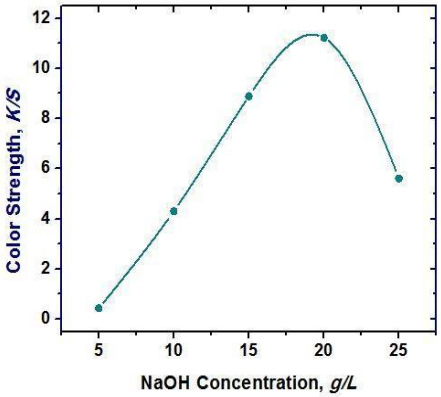

Fig 5: Effect of Sodium hydroxide concentration

F. Effect of dye concentration

Fig 6 shows that colour strength increased with the increasing indigo dye concentration in the dyebath. However, it limits up to 30 g/L due to limited available dye sites on the fibre [13]. Moreover, that the higher indigo dye concentration in the dyebath requires higher amounts of reducing agent and the alkali to reduce the dye properly.

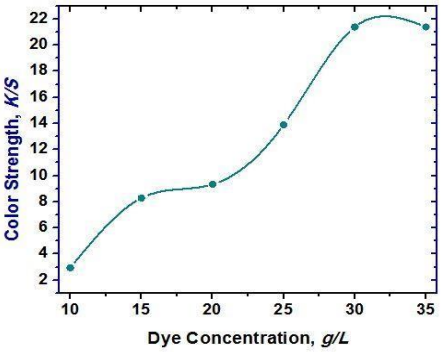

Fig 6: Dye concentration effect on colour strength

G. Comparison of natural and synthetic indigo dyed fabrics

Fabric samples were dyed with 10 g/L shade at optimized dyeing conditions. Table 1 shows colour strength and colorimetric values of the denim fabrics dyed with developed natural and synthetic indigo dyes. The table shows that colour strength (K/S value) and CIE L*a*b* values of the developed natural indigo dye were better than synthetic indigo liquid dye.

Table 1: Colour strength (K/S) and CIE L*a*b* values of dyed fabric

Indigo dyed samples	L*	a*	b*	C*	h*	Color strength (*K/S* values)
Developed Natural Indigo	13.51	3.22	-6.53	7.28	296.22	36.964
Synthetic Indigo Liq	15.48	0.74	-11.36	11.38	273.71	33.674

Colourfastness results

Table 2 shows the acceptable rubbing and washing fastness with excellent light fastness results of fabrics dyed with both natural and synthetic indigo dyes.

Table 2: Colourfastness assessment of indigo dyed fabric

Indigo dyed samples	Lightfastness rating	Rubbing fastness rating (dry)	Washing fastness (colour change rating)
Developed Natural Indigo	7	3/4	4/5
Synthetic Indigo Liq	7	4	4/5

IV. CONCLUSIONS

This paper concludes successful indigenous natural indigo dyeing as a better alternative to synthetic indigo dye for continuous dyeing process for denim. The natural indigo exhibited higher colour strength and colorimetric parameters closer to true indigo at optimized conditions. And, acceptable rubbing and washing fastness along with excellent light fastness results were obtained. The developed dye and the dyeing procedure can be proposed for industrial scale dyeing of warp yarn with natural indigo for production of denim fabric.

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