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An Investigation of Color Fading of Sulfur-dyed Cotton Fabric by Plasma Treatment

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Abstract: Popularity of clothing with different kinds of faded color effects has been growing in recent years, leading to development of several technologies and techniques for imparting the vintage and old-fashion look. However, most color fading technologies and processes involve use of significant quantities of chemicals and water, raising concerns about environmental pollution, and other related problems. It is quite difficult to control the color fading effect on textile products. Plasma treatment has been used for color fading of cotton apparel quite successfully in the last few years. Air is used as the color fading agent and therefore no chemical effluents are generated. This study examines color fading of sulfur dyed cotton fabrics with plasma treatment. Cotton fabrics (yellow color) dyed with sulfur dyes were plasma treated under varying conditions and the resultant color fading effect was evaluated instrumentally. The color fading effect was found to be quite controllable if the treatment parameters were properly selected. Besides, evenness of the end product was excellent.

Keywords: Color fading, Sulfur dye, Cotton, Plasma

Introduction

Color is one of the most important considerations for consumers when buying apparel and textile products [1]. The young generation has developed a liking for color faded vintage and old fashion look in the last couple of decades [2]. This has resulted in manufacturers of textiles and apparel developing different technologies for imparting the faded color appearance to dyed fabrics [3,4]. This has been done mostly by using the conventional chemical treatments involving oxidation and enzymatic processes [5,6].

In the chemical treatments, oxidation method involves the use of sodium hypochlorite and potassium permanganate. In case of sodium hypochlorite, it is a bleaching agent for oxidizing the color in the dyed material at room temperature leading to significant color fading effect. The color fading result obtained from sodium hypochlorite depends on the dosage used and the strength of chemical. However, antichlor treatment by reducing agent is required to remove residual chlorine in the products, otherwise, the residual chlorine may cause irritation to human skin. In addition, if the product contains spandex materials, sodium hypochlorite treatment may reduce the strength of the spandex materials leading to reduced fabric strength. Other than sodium hypochlorite, potassium permanganate is another a popular oxidizing agent for color fading process of dyed textiles. When compared with sodium hypochlorite, potassium permanganate has a stronger oxidizing power and it could induce color fading effect in a faster rate than sodium hypochlorite. Similar to sodium hypochlorite, after color fading process with potassium permanganate, neutralisation process must be carried out to remove the residual chemical in order to prevent skin irritation. In fact, enzyme (cellulase) is an important sustainable color fading agent for dyed cotton textiles. The cellulase has a very specific reaction with the cellulose and hence remove the dyes from the cotton textiles for achieving color fading effect. However, the process parameters such as pH, temperature, and purity of water should be properly controlled in order to obtain the best enzymatic color fading effect. Although the sodium hypochlorite, potassium permanganate, and enzyme treatments are effective method for obtaining color fading effect in dyed textiles, these processes consume large quantities of chemicals, need highly skilled manpower and are generally time consuming. Besides, defining a standard process and reproduction of a given design is difficult in chemical treatments. The high consumption of water, chemicals and energy implies adverse environmental effects that result in health and environmental hazards for the workers and cause depletion of natural resources [7,8]. Safer and cleaner color fading processes are therefore needed.

We have found in our recent work [7] that plasma treatment is perhaps an alternative to the conventional way of achieving the color fading effect on fabric. In general, plasma treatment has proved to be an effective and good treatment for modification of surface of textile materials [9-12] and it can modify textile properties such as pilling, water vapour permeability, air permeability, and wettability [6,13,14]. It has been demonstrated clearly that surface of cotton textile products can be successfully altered by the use of plasma treatment, without affecting its bulk properties [6,13,14]. Environmental advantages of the plasma process for treating textile materials [10,11,15], compared with conventional wet processes [6] are quite obvious. In plasma treatment using air as reactive gas, the oxygen in the air will generate different radicals in the plasma process [16] such as O• and O_3. Ozone (O_3), a strong oxidizing agent [17,18], generated is dissolved readily in the moisture which in turn becomes

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an effective oxidant (•OH radical) for color fading [16]. When ozone is generated during plasma treatment with oxygen, UV light is also generated, as a by-product which involves generation of the •OH radical. The hydroxyl radical (•OH) is responsible for degrading and decolorizng the dye in colored textile materials [19-25] and thereby producing the color fading effect on dyed textiles.

We have successfully shown in previous studies [7,26,27] that plasma treatment can be used for color fading of reactive dyed cotton fabrics of different color depths. This study examines suitability of plasma treatment for sulfur dyed cotton materials. Sulfur dye is a dye class that accounts for about 30 % of worldwide consumption of textile dyes [28] of which 14 % is used for dyeing wearing apparel products, which are often subjected to color fading related processes [29]. Therefore, investigation of the use of plasma treatment for color fading of 100 % sulfur dyed cotton fabrics is important.

Experimental

Preparation of Sulfur Dyed Fabric

Knitted cotton fabric was dyed with 1.5 % (on weight of fabric, owf) sulfur dye (Diresul Yellow RDT-E, pre-reduced solubilised sulfur dye) with liquor-to-goods ratio of 10:1. The dyebath was heated to 60 °C and after 10 min, fabric was added into the dyebath. Sulfur dye was added 20 min later and temperature was maintained at 60 °C for a further 10 min. After that temperature was raised to 75 °C and then sodium sulphate (20 g/*l*) was added in three doses, at 5-min intervals and the temperature was maintained for a further 30 min [30]. This was followed by the oxidation process, at liquor-to-goods ratio of 10:1 at 50 °C for 20 min with pH 4-4.5 using 35 % hydrogen peroxide (2 % owf) and 80 % acetic acid (2 % owf). The fabrics were then rinsed with running water and soaping with detergent was conducted for 10 min at 90 °C. The samples were then dried by air, followed by conditioning (relative humidity: 65±2 %; temperature: 20±2 °C) for at least 24 h prior to further use [30].

Plasma Color Fading Treatment

Commercially available plasma machine (atmospheric pressure plasma type), G2 (Jeanologia, Spain), was used for treatment [31] (Figure 1). The machine has a maximum loading capacity of 50 kg and the power demand is 9 kW/h. The maximum ozone concentration generating is 180 g/N·m^3. The parameters used in the plasma color fading treatment are summarized in Table 1 [7].

Color Measurement

Color properties were measured by a spectrophotometer (GretagMacbeth Color-Eye7000A). Daylight D_{65} and 10° standard observer were the conditions during color measurement. The samples were conditioned for at least 24 h at 20±2 °C

Figure 1. G2, an industrial plasma machine for color fading (photo obtained with permission).

Table 1. Plasma process parameters [7,21]

Process parameter	Values
Air content (amount of air used inside the machine) (%)	10, 30, 50, 70
Water content (%)	35, 45
Treatment duration (min)	10, 20, 30

and relative humidity of 65±2 % and before measurement. Four measurements were obtained for each sample and the results were averaged. Reflectance curves, K/S_{sum} values and CIE $L^*a^*b^*$ values were determined. A lower K/S_{sum} value means lower color yield, i.e. better color fading effect. The color fading percentage (CF%) was calculated by equation (1) [32].

$$\text{Color fading percentage (\%)} = \frac{(K/S_{sum})_i - (K/S_{sum})_f}{(K/S_{sum})_i} \times 100\% \quad (1)$$

where $(K/S_{sum})_i = K/S_{sum}$ value before plasma treatment; $(K/S_{sum})_f = K/S_{sum}$ value after plasma treatment.

Color Levelness Measurement

The uniformity of color was measured by relative unlevelness index (RUI) according to Chong *et al.* [33] in which (i) RUI<0.2 means excellent levelness; (ii) RUI=0.2-0.49 means good levelness; (iii) RUI=0.5-1.0 means poor levelness, and (iv) RUI>1.0 means bad levelness.

Results and Discussion

Reflectance Measurement

Reflectance measurement, expressed as reflectance curve, is used for examining and shade change after the plasma color fading treatment and the results are shown in Figure 2. Figure 2 demonstrates the reflectance curves of cotton fabrics with different plasma treatment parameters, i.e. air content, water content and treatment duration. It is observed that all reflectance curves exhibit similar shape which means there is no peak shifting and all plasma treated fabrics have the same shade. Although no change in the shade after plasma color fading treatment, the reflectance percentages are varied for different fabrics. Lowest reflectance percentage is noted for fabric dyed with sulfur dye without plasma treatment. The reflectance curves of all plasma treated fabric samples show higher reflectance percentage. This increased reflectance percentage implies the plasma treated fabrics have a paler shade than the original sample. Thus plasma treatment decolorizes the dye, leading to effect of color fading.

Color Yield

Color yield of a colored fabric sample, measured by K/S_{sum} value, can be calculated by $K/S=(1-R)^2/2R$ (K/S_{sum} value is the summation of K/S values over the visible spectrum from 400 nm to 700 nm). A lower K/S_{sum} value denotes a lower color yield. Plasma treated fabric samples are expected to have a lower K/S_{sum} value than the original sulfur dyed fabric samples since the amount of color is reduced after the color fading process. The K/S_{sum} value of the original sulfur dyed fabric is 11.86 and Table 2 shows that K/S_{sum} values of plasma treated cotton fabrics are lower than 11.86. The K/S_{sum} values of the fabrics were obviously reduced after plasma treatment under different processing conditions. The lower K/S_{sum} values indicate color fading due to decolorization of dye in the cotton fabrics.

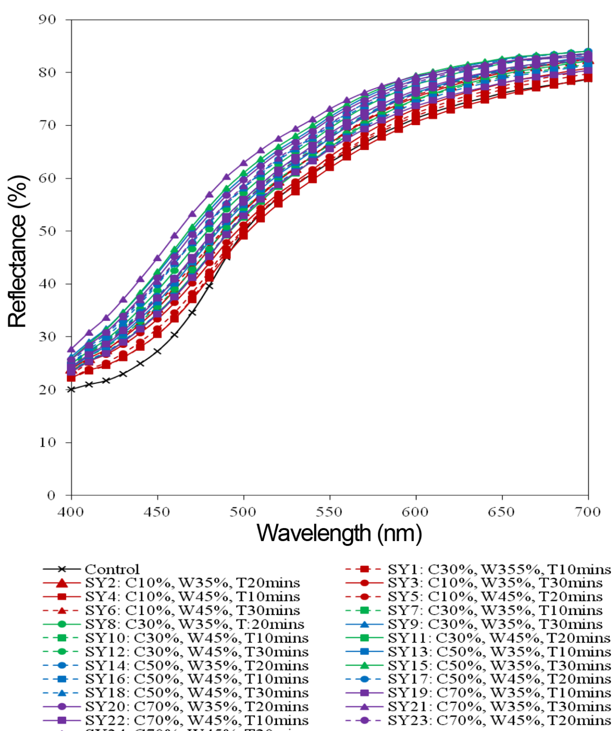

Figure 2. Reflectance curve of plasma treated cotton fabric samples (sample code: (i) SY=sulfur yellow; (ii) C30%=air content of 30 %, etc.; W45%=water content 45 %, etc. and T20mins= treatment duration of 20 min., etc.).

Table 2. K/S_{sum} and color fading percentage values

Air content (%)	Treatment duration (min)	Water content (%)	
		35	45
10	10	10.19 (14.08 %)*	10.35 (12.73 %)
	20	8.90 (24.96 %)	9.79 (17.45 %)
	30	8.08 (31.87 %)	8.13 (31.45 %)
30	10	7.99 (32.63 %)	8.52 (28.16 %)
	20	6.93 (41.57 %)	7.50 (36.67 %)
	30	6.00 (49.41 %)	7.38 (37.77 %)
50	10	7.68 (35.24 %)	8.48 (28.50 %)
	20	6.69 (43.59 %)	7.44 (37.27 %)
	30	5.97 (49.66 %)	6.58 (44.52 %)
70	10	7.40 (37.61 %)	8.44 (28.84 %)
	20	6.25 (47.30 %)	7.24 (38.95 %)
	30	5.64 (52.45 %)	6.42 (45.87 %)

*The value inside bracket represents the color fading percentage (CF%).

Table 2 shows relationship between air content and the K/S_{sum}. It is noted that as the air content increases from 10 % to 70 %, K/S_{sum} values decrease generally. This is due to the fact that at a higher air content, more oxygen is supplied and hence more oxygen molecules are contained in the plasma treatment. Therefore, it will generate more hydroxyl radicals under the influence of plasma, leading to more of oxidation effect on the dye resulting in higher degree of color fading (decolorization). If the treatment duration is taken into consideration, it is seen that K/S_{sum} values decrease correspondingly with the increase of treatment duration from 10 min to 30 min. This can be explained by a longer treatment duration will provide a longer contact duration between active species in plasma and dye in the fabric [7] and therefore, more dyes are oxidized and decolorized leading to color fading (resulting in lowered K/S_{sum} values). The experimental results indicate 35 % water content in fabric could have a better color fading effect than 45 % water content in fabric because higher water content would dilute the bleaching effect.

Color Fading Percentage (CF%)

Color fading percentage (CF%) denotes the degree of color fading [26]; the greater the CF% value, the greater is the color fading effect [32]. Table 2 illustrates the results of CF% of the sulfur dyed cotton fabric under the plasma treatment with different combination process parameters (i.e. air content, treatment duration and water content). Based on the results in Table 2, it is clearly noted that as the color fading effect increases when air content and treatment duration are increased. However, the increase in water content in plasma treatment does not introduce any enhancement of color fading because of the dilution of the bleaching agent generated if the fabric has a higher water content and this result agreed with the reactive dye dyed fabrics [7].

Color Levelness

RUI is used for defining the color levelness after plasma color fading treatment in this study. The RUI was measured by spectrophotometer which may provide color levelness with objective and quantitative meanings. The degree of color levelness of sulfur dyed cotton samples before and after plasma treatment and the RUI values of different fabric samples are summarized in Table 3.

According to the definition of RUI, fabric sample with RUI lesser than 0.2 can be classified to have excellent levelness [33]. It is clearly shown in Table 3 that the color levelness of all plasma treated fabrics is excellent (i.e. unlevelness is not detectable instrumentally) but has different degrees of color fading, also reflected by K/S_{sum} and color fading percentage values (Table 2). Thus, plasma treatment can provide excellent levelness after the color fading process in a controlled manner, which cannot be achieved easily with conventional chemicals [7,32].

Table 3. RUI values

Water content (%)	Air content (%)	Treatment duration (min)	RUI	Degree of levelness
	Original		0.06	Excellent
35	10	10	0.05	Excellent
		20	0.06	Excellent
		30	0.09	Excellent
	30	10	0.06	Excellent
		20	0.07	Excellent
		30	0.12	Excellent
	50	10	0.06	Excellent
		20	0.08	Excellent
		30	0.18	Excellent
	70	10	0.08	Excellent
		20	0.10	Excellent
		30	0.19	Excellent
45	10	10	0.05	Excellent
		20	0.05	Excellent
		30	0.08	Excellent
	30	10	0.05	Excellent
		20	0.06	Excellent
		30	0.10	Excellent
	50	10	0.05	Excellent
		20	0.08	Excellent
		30	0.16	Excellent
	70	10	0.06	Excellent
		20	0.09	Excellent
		30	0.17	Excellent

CIE $L^*a^*b^*$ Measurement

Besides color yield and color levelness, CIE $L^*a^*b^*$ values of untreated and plasma treated fabric samples were also measured (Table 4). In the CIE system, L^* value represents the lightness of the shade. If the fabric sample has a high L^* value, the fabric sample shade is pale. L^* value of the original sulfur dyed fabric is 82.40 but in case of plasma treated colored fabrics, L^* values are higher. Also, the water

Table 4. CIE $L^*a^*b^*$ values

Water content (%)	Air content (%)	Treatment duration (min)	L^*	a^*	b^*	$\Delta E^{\#}$
	Original		82.40	0.62	36.20	-
35	10	10	83.04	0.53	35.09	1.36
		20	83.15	0.43	32.61	3.75
		30	83.63	-0.35	32.18	4.39
	30	10	83.44	0.50	31.66	4.74
		20	84.97	-0.20	30.64	6.26
		30	86.01	-0.62	28.88	8.33
	50	10	84.09	0.43	31.62	4.96
		20	85.35	-0.30	30.12	6.90
		30	86.27	-0.73	28.81	8.53
	70	10	84.76	-0.82	30.95	6.01
		20	86.05	-0.97	29.47	7.89
		30	86.62	-1.45	28.35	9.22
45	10	10	82.49	0.55	35.28	1.01
		20	82.93	0.50	34.16	2.19
		30	83.53	-0.18	32.67	3.87
	30	10	83.11	0.52	32.68	3.67
		20	84.10	-0.15	31.01	5.59
		30	84.15	-0.56	30.76	5.91
	50	10	83.74	0.49	32.58	3.94
		20	84.30	-0.25	30.89	5.78
		30	85.20	-0.68	29.56	7.40
	70	10	84.03	-0.58	32.20	4.56
		20	84.38	-0.72	30.76	6.02
		30	85.39	-1.32	29.53	7.64

#Remark: Color difference $(\Delta E)=[(\Delta L^*)^2+(\Delta a^*)^2+(\Delta b^*)^2]^{1/2}$ (where $\Delta L^*=L^*_{sample}-L^*_{original}$; $\Delta a^*=a^*_{sample}-a^*_{original}$; and $\Delta b^*=b^*_{sample}-b^*_{original}$).

content (if the air content and treatment duration are kept in same condition) in the fabric samples demonstrates an important effect on the L^* values, i.e. 35 % water content could lead to have a higher L^* value when compared with 45 % water content. This reduction in L^* values in 45 % water content is due to dilution of bleaching agent generated during the plasma treatment [7]. L^* value increases as both air content and treatment duration increase. If a higher air content is used, the larger number of oxygen molecules generate more hydroxyl free radicals for oxidizing and decolorizing the dyes, leading to greater color fading effect [7]. The treatment duration refers to the contact duration between the plasma active species and the dyes [7]. The longer treatment duration results in more dye getting oxidized and decolorized leading to greater color fading effect. Therefore, the lightness of the fabric shade can be controlled by controlling parameters of the plasma treatment process.

The a^* value is a measure of redness or greenness color properties of a fabric sample. Generally speaking, a redder shade has a more positive a^* value. On the other hand, a negative a^* value implies a greener shade. The original sulfur dyed fabric has a^* value of 0.63 while all plasma treated fabrics have lower values, which means plasma treated fabrics are greener than the original fabric. When both treatment duration and air content are increased, a^* values decline correspondingly. 35 % Water content influences a^* values more than 45 % water content if other process parameters such as air content and treatment duration are kept constant.

A more positive b^* value indicates a more yellowish shade. On the other hand, a negative b^* value refers to a bluish shade. The b^* value of the original sulfur dyed fabric is 36.20 and a relatively lower b^* is observed after plasma color fading treatment (Table 4). The lower b^* value indicates that plasma treatment makes the fabric less yellowish in shade in couple with the increasing of both treatment duration and air content. Similar to the L^* and a^* values, the water content in fabric samples also plays an important in affecting the b^* values of plasma treated fabric samples. Samples treated with water content of 35 % water content have a relatively smaller b^* value than samples treated with 45 % water content.

Table 4 show that the color difference (ΔE) results increase as air content and treatment duration are increased. However, 35 % water content results in greater color difference than 45 % water content 45 % indicating that 35 % water content can provide better color fading effect. CIE $L^*a^*b^*$ and ΔE values show that if the process parameters of plasma treatment can be properly controlled, desirable color fading effect in sulfur dyed cotton fabrics can be obtained.

Correlation of Different Color Properties

An attempt was made to correlate properties of different specimens to provide a better understanding of the correlations between different properties [34-37]. Table 5 summarizes the correlations and combinations of different properties. Linear relationships between properties of each pair were measured by calculating the correlation coefficient (r) and the correlation of determination (R^2) for the different correlation combinations [34]. Since r values are spread over a wide range, it is desirable to test its significance. The null hypothesis postulates that there is no correlation between the two properties. The null hypothesis can be tested by equation (2) [34,38].

Table 5. Relationship between different properties and their R^2, $|r|$ and t_o values

Correlation combinations	R^2	$\|r\|$	t_o	$t_o > t$ (2.074), null hypothesis rejected	Linear correlation
K/S_{sum} and CF%	0.9998	0.9999	331.629	Yes	Linear
K/S_{sum} and RUI	0.1251	0.2613	1.310	No	Non-linear
K/S_{sum} and ΔE	0.9777	0.9888	31.058	Yes	Linear
CF% and RUI	0.1255	0.2587	1.298	No	Non-linear
CF% and ΔE	0.9773	0.9886	30.776	Yes	Linear
RUI and ΔE	0.1135	0.2114	1.053	No	Non-linear

$$t_o = \frac{|r|\sqrt{n-2}}{\sqrt{1-R^2}} \tag{2}$$

The t_o has a t distribution with (n−2) degrees of freedom and its significance can be tested by comparing t_o with values in a t-distribution table [35,39]. In this study, the degree of freedom is 24-2=22. With reference to the t-distribution table with a 95 % confidence level using a two-tailed test, the t value is 2.074 [39]. Table 5 shows t_o values of the different relationships. If t_o is greater than t, the null hypothesis is rejected. Table 5 shows that K/S_{sum}, CF% and ΔE are statistically related. Although RUI does not have significant statistical relationship to K/S_{sum}, CF% and ΔE, RUI values of plasma treated fabrics are acceptable, with excellent levelness, as indicated in Table 3.

Conclusion

In this study, we investigated the effect plasma color fading process on the color properties of sulfur dyed cotton fabrics. In the plasma color fading process, three process parameters, which were air content, water content and treatment duration, were used for treating the sulfur dyed cotton fabrics. After the plasma color fading process, the color properties such as reflectance, K/S_{sum}, color fading percentage, CIE $L^*a^*b^*$ and levelness of the fabric samples were evaluated instrumentally. Experimental results revealed that the degree of color fading in sulfur dyed cotton fabric can be controlled by the process parameters. Generally speaking, a longer treatment duration and a higher air content could increase the color fading effect considerably. However, 35 % water content achieved a better color fading effect than 45 %. Unlike conventional color fading process, plasma treated fabrics could achieve excellent degree of color levelness. It was also observed that K/S_{sum}, color fading percentage and ΔE are statistically correlated. Thus, it can be concluded that plasma treatment, which is more environment-friendly than the conventional process, constitutes a feasible alternative for color fading in a controllable manner, in textile and apparel industries.

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