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Natural Dyeing using the Colorants extracted from American Fleabane (Part II)
- Dyeing properties on cotton -

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개망초 추출물을 이용한 천연염색 (제2보)
- 면섬유에 대한 염색성 -

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Abstract

The purpose of this study was to investigate dyeing properties of cotton fabrics by American Fleabane extract. Effect of dyeing condition on dye uptake and effect of mordanting on dye uptake, color change and colorfastness were explored. Its affinity to cotton fiber was considerably lower than wool, and its hydrogen bonding was involved in the absorption of American fleabane colorants to cotton fiber. K/S value of pre-mordanting was higher than sim-mordanting or post-mordanting. Except that Sn mordanted showed high K/S value, it did not significantly increase. American fleabane produced mainly yellowish color on mordants and mordanting method. Cotton fabrics showed generally high colorfastness except colorfastness to washing.

Key words: American fleabane colorants, Adsorption isotherm, Mordanting, Dye uptake, Fastness; 개망초 색소, 등온흡착곡선, 매염, 염착량, 견뢰도

I. Introduction

Green technology is one of important concern in textile industries. It has been realized that the number of dyes being used in the textile industries contain toxic and carcinogenic substances. Germany banned the use of azo dyes containing toxic aromatic amine. Sweden and India imposed a ban on dyes based on benzidine(Lokhande et al., 1998). It is generally accepted that natural dyes are more eco-friendly and so safer than synthetic dyes. It is imperative to exploit dyes from natural and renewable resources as alternatives to synthetic dyes. However, there are few works reported on exploiting new potential resources of natural dyes.

American fleabane can be found very commonly in the roadside as well as fields in summer. We exploited it as a new resource of natural dye. In previous study, the dyeing properties of colorants extracted from American fleabane on wool were investigated. We confirmed the high affinity of American fleabane colorants on wool without mordanting and relatively good colorfastness(Shin and Cho, 2003).

The present study focuses on the dyeing of cotton with American fleabane colorants. Three different mordanting methods, i.e. pre-mordanting, simultaneous mordanting, and post-mordanting are used. The purpose is also to investigate the dyeing and fastness properties of the colorants on cotton. Comparisons between dyeing with and without mordanting were made, and the depths of shade were evaluated in terms of K/S and CIELAB color difference values of the dyed samples.

II. Experimental

1. Materials

A scoured and bleached 100% cotton fabric was used(plain weave, 29×26/cm^2, 100g/m^2, 0.23mm thickness). American fleabane plant was dried after removing roots for 2 weeks, and used to extract for dyeing. Mordants including aluminium ammonium sulfate($Al_2(SO_4)_3(NH_4 \cdot 24H_2O)$, ferric sulfate($FeSO_4 \cdot 7H_2O$), cupric sulfate($CuSO_4 \cdot 5H_2O$), potasium dichromate($K_2Cr_2O_7$), and stannic chloride ($SnSl_2 \cdot 2H_2O$) were used. All chemicals were reagent grade.

2. Methods

1) Preparation of colorants

Aqueous extracts of American fleabane were prepared by adding 50g dried plants to 1500ml distilled water. The mixture was held at 100°C for 60min, filtered and dried into powder. Yield of colorants was about 25%.

2) Dyeing and mordanting

Dyeing was done at a liquor ratio of 50 : 1, colorants concentration 0.5-5%(o.w.b.), 40-120°C, 30-150min. The pH of dyeing solution was adjusted to 3-11 by the addition of acetic acid and sodium hydroxide. Three different methods of pre-mordanting, simultaneous mordanting and post-mordanting were employed. Mordanting was carried out in 1%(o.w.f.) mordant solution at a liquor ratio of 50:1 by holding at 60 for 30min. In the simultaneous-mordanting, mordants were applied during the dyeing process. An automatic laboratory dyeing machine(Ahiba Nuance, Datacolor International, USA) were used for dyeing and mordanting.

3) Color measurement

Color values were evaluated in terms of K/S values and CIE L*a*b* data(Illuminant D_{65}, 10° Observer) with a Macbeth Coloreye 3100 spectrophotometer at 420nm, maximum absorption wavelength. H V/C values were obtained from L*a*b* data using CIE Munsell conversion program.

4) Fastness testing

Fastness of the dyed samples was evaluated by standard procedures; wash fastness by KS K 0430-A; rubbing fastness by AATCC method 116-1989; perspiration fastness by KS K 0715. Fastness was assessed by using gray scale for color change and chromatic transference scale. Light fastness was assessed in terms of color difference(ΔE) according to AATCC method 16-1998 with Fade-Ometer(Atlas Electric Devices Co, USA). Color differences were measured with a Macbeth Coloreye after irradiating for 5, 10, 20, and 40 hours.

III. Results and Discussion

1. Effect of Dyeing Conditions on Dye Uptake and Color

<Fig. 1-4> show the effect of colorants concentration, dyeing temperature and time, and pH on dye

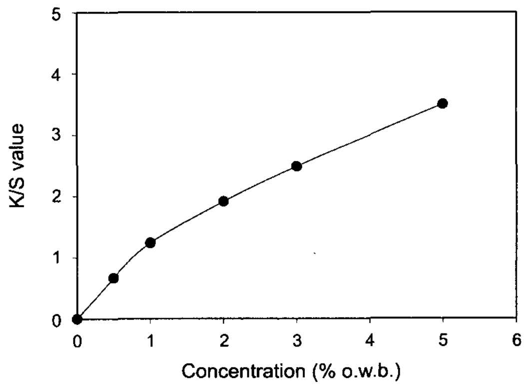


Fig. 1. Effect of Dyeing Concentration on the Dye Uptake of Cotton Fabric (100°C, 60min).

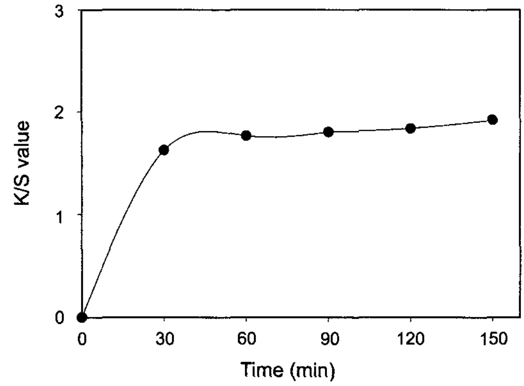


Fig. 2. Effect of Dyeing Time on the Dye Uptake of Cotton Fabric (2% o.w.b., 100°C).

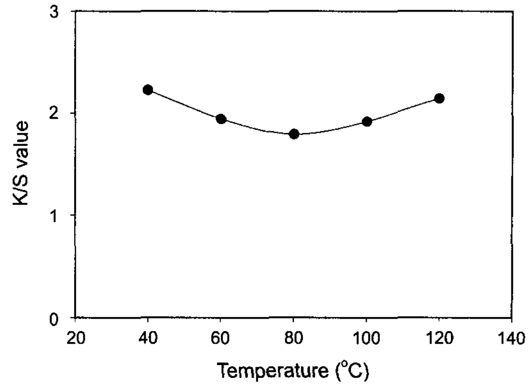
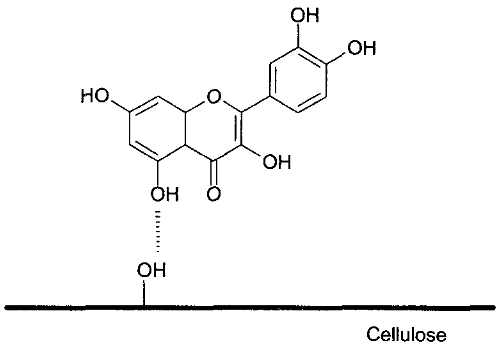


Fig. 3. Effect of Dyeing Temperature on the Dye Uptake of Cotton Fabric (2% o.w.b., 60min).



Scheme 1. Hydrogen Bonding between Cotton and Quercetine.

uptake(K/S). Dye uptake increases progressively as colorant concentration increases. The adsorption isotherm presented in <Fig. 2> is considered as Freundlich type, indicating that adsorption of the colorants occurs mainly by hydrogen bonding(Kim and Lee, 1998). It is known that the colorants contain quercetin, apigenin-7-glucoside, gallic acid, tannic acid, chlorogenic acid, d-limonene , etc(Roth et al., 1992; Schweppe, 1992; Yook, 1997). Most of them have hydroxyl groups in their structure. As an example, hydrogen bonding between cotton and quercetin is shown in (Scheme 1).

Dye uptake increases abruptly for initial 30min and then reaches equilibrium at 60min, as seen in (Fig. 3). Dye uptake decreases up to 80°C and then increases. Maximum dye uptake is observed at strong acidic condition, pH 3.0. The dye uptake decreases with the increase of pH to alkaline. In alkaline condition, electrostatic repulsion between anionic colorants and cotton fibers occurs and this leads to decrease in dye uptake. pH 5.5 of colorants solution was not adjusted because strong acidic conditions may cause the strength loss of cotton(Choi and Cho, 2001; Lee et al., 1998; Lee et al., 2000).

From the obtained results, dyeing conditions are fixed 2%(o.w.b.) concentration, pH 5.5, 40°C, and 60min in the latter experiments.

The influence of pH on color is shown in Table 1. The L* value indicates perceived lightness in CIELAB color space. The L* scale runs from 0(black) to 100(white); the higher the L reading, the lighter the color. The a* value indicates red(+a*) and green(-a*) while the b* value indicates yellow(+b*) and blue(-b*)(Billmeyer and Saltzman, 1981). As pH increases, L* values increase and then decreases, meaning that the shade is getting lighter up to pH 5.5 and then darker in alkaline condition. a* decreases and then increases, which relates with the reduction of reddish shade as it approaches to neutral from acidic and then increase of reddish shade in alkaline condition. And b* increases up to pH 7.0 and then decreases, indicating that yellowish shade decreases in alkaline condition. Although all of the dyed samples show Y color irrespective of pH, hue increases up to pH 7.0 indicating the reduction of yellowish shade. Value increase up to pH 5.5 and then decreases. On the other hand, chroma increases up to

Table 1. Effect of pH on L*, a*, b*, & H V/C Values of the Dyed Cotton Fabrics.

pH	L*	a*	b*	H	V/C
3.0	79.107	-0.625	15.156	3.13Y	7.77/2.06
5.5	81.650	-1.669	15.333	4.39Y	8.04/2.00
7.0	81.153	-1.669	16.418	4.62Y	7.99/2.15
9.0	79.294	-1.348	14.346	4.14Y	7.79/1.89
10.5	74.127	-0.048	10.614	2.52Y	7.26/1.48

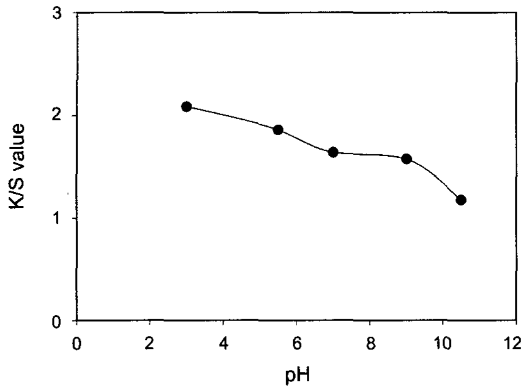


Fig. 4. Effect of pH on the Dye Uptake of Cotton Fabric (2% o.w.b., 100°C, 60min)

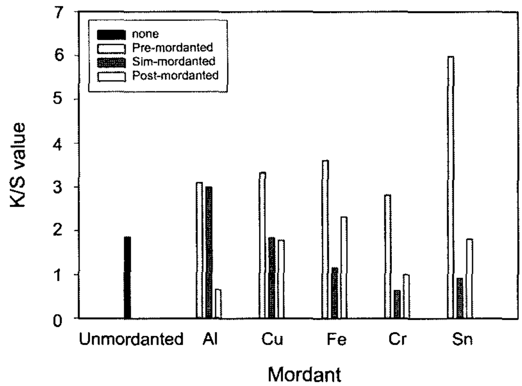


Fig. 5. Effect of Mordants on Dye Upteke of Cotton Fabric.

pH 7.0 and then decreases. The shade of dyed fabrics is darker and duller in alkaline condition.

2. Effect of Mordanting on Dye Uptake and Color

Mordants play an important role in natural dyeing because they usually have substantivity for both the fiber and the colorants. They form coordination bonds with fiber and at the same time, form insoluble chelate with the dye.

<Fig. 5> shows dye uptake depending on mordant type and mordanting method. Among three mordanting methods, pre-mordanting gives higher depth of shade(K/S), compared with the fabrics dyed using the other two methods. Especially, Sn improves dye uptake substantially with pre-mordanting method. This may be due to the greater complex-forming ability of the metal ions with the dye molecules in this method. With the exception of Al, simultaneous and post-mordanting methods give lower depth of shade than the unmordanted sample. In the case of simultaneous mordanting method, some of dye is lost because of the formation of an insoluble complex in the dyebath itself. On the other hand, during post-mordanting some of dye is stripped out in the bath and subsequently forms an insoluble complex with metal ions in the solution. These phenomena lead to a decrease in the effective dye concentration in the dyebath(Deo and Desai, 1999). From the results, it is observed that mordanting is not an effective method for improving dye uptake in case of Americane fleabane. So, it is speculated that surface modification is needed to create dye sites on cotton fiber. There has been a number of attempts to modify the cotton fiber using cationic agents, such as quarternary ammonium salts, polymeric amines or amides, chitosan, etc., for improving dye exhaustion(Lim and Hudson, 2004).

The influence of mordanting on color values of the dyed fabrics are compared in (Table 2). L* shows (–) value in most of the mordanted samples except for the

Table 2. Effect of Mordanting on L*, a*, b* & H V/C Values of the Dyed Cotton Fabrics.

None		L*	a*	b*	H	V/C
		81.650	-1.669	15.333	4.39Y	8.04/2.00
Mordants	Mordanting method	ΔL*	Δa*	Δb*		
Al	Pre-mordant	-3.112	-3.441	18.680	6.61Y	8.33/2.43
	Sim-mordant	-2.520	-6.105	29.409	7.97Y	8.39/3.94
	Post-mordant	-3.109	-2.291	13.285	5.99Y	8.38/6.71
Cu	Pre-mordant	-5.682	-2.727	22.920	5.03Y	8.06/3.15
	Sim-mordant	-3.954	-4.650	25.212	7.01Y	8.34/3.37
	Post-mordant	-10.507	-0.708	20.006	3.68Y	7.56/2.85
Fe	Pre-mordant	-11.854	0.029	15.270	2.72Y	7.42/2.19
	Sim-mordant	-9.160	-1.044	8.812	4.84Y	7.69/1.12
	Post-mordant	-20.555	0.172	13.740	2.84Y	6.53/1.98
Cr	Pre-mordant	-3.651	-3.044	18.757	5.92Y	8.27/2.47
	Sim-mordant	-2.376	-1.745	10.923	5.55Y	8.40/1.42
	Post-mordant	-6.587	-0.272	15.373	2.84Y	7.97/2.15
Sn	Pre-mordant	-6.189	-2.783	27.025p	4.82Y	8.01/3.79
	Sim-mordant	0.313	-2.313	7.627	9.50Y	8.68/0.97
	Post-mordant	-4.138	-1.438	27.570	3.97Y	8.23/3.90

Table 3. Colorfastness of the Dyed Cotton Fabrics.

Mordant	Washing			Perspiration (acidic)			Perspiration (alkaline)			Rubbing	
	Color change	Stain		Color change	Stain		Color change	Stain		Dry	Wet
		Cotton	Wool		Cotton	Wool		Cotton	Wool		
None	2	5	5	3	4/5	5	4/5	4	4/5	5	5
Al	2	5	5	3/4	4/5	5	4/5	4	4/5	5	4/5
Cu	2	5	5	4	4/5	5	4/5	4	4/5	5	5
Fe	2	5	5	4	4/5	5	4/5	4	4/5	5	4/5
Cr	2	5	5	3/4	4/5	5	4/5	4	4/5	5	4/5
Sn	3	5	5	3/4	4/5	5	4	4	4/5	5	4/5

sample simultaneous mordanted with Sn, meaning that shade of the samples gets lighter by mordanting. a* show (–) value except for the sample mordanted with Fe, indicating the decrease in reddish shade, while b* show (+) value, indicating the increase in yellowish shade. Irrespective of mordant type and mordanting method, the samples show Y color and did not show significant change in value and chroma.

3. Fastness

Colorfastness of the pre-mordanted and unmordanted samples is compared in (Table 3). The unmordanted sample shows comparatively good fastness properties except for color change after washing. The mordanted samples also show low rating in color change after washing. Colorfastness ratings of less than grade 3 indicate considerable change in color after washing. The change in color after washing in alkaline condition can be attributed to the loss of colorants as a result of conversion of the colorants to soluble salts and other soluble products(Popoola, 2000). In practical point of view, the results imply the importance of alkalinity of solu-

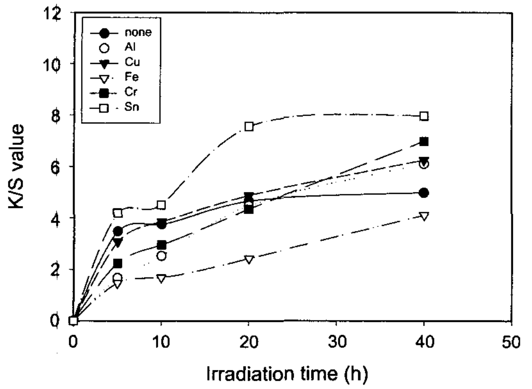


Fig. 6. Effect of Irradiation Time on the Color Difference of Cotton Fabric Dyed with American Fleabane Colorants.

tions used for washing fabrics dyed with natural colorants(Taylor, 1986). The ratings of wash fastness in terms of the degree of staining are excellent for both of the mordanted and unmordanted samples. Rubbing fastness is relatively good regardless of mordanting. Alkaline perspiration gives a slightly lower rating than acidic perspiration. Pre-mordanting has little effect on the fastness properties.

<Fig. 6> shows color differences of the dyed samples depending on irradiation time. As expected, photofading of the dyed samples increases as irradiation time increases, especially the Sn-mordanted sample shows more severe fading. Fe mordant improves light fastness. The improvement of light fastness by mordanting with Fe is attributed to the stabilizing effect of chelate formed with colorants (Yoon and Kim, 1993). Most natural dyes have poor light fastness on cotton because it is hygroscopic. The moisture present in the fiber catalyzes the photochemical oxidation of dyes on the fiber, leading to poor light fastness(Taylor, 1986).

IV. Conclusions

The dyeing and fastness properties of the colorants extracted from American fleabane on cotton were investigated. Effect of dyeing conditions and mordanting were evaluated in terms of dye uptake(K/S value) and CIELAB color. Also, effect of mordanting on fastness was assessed.

Dye uptake increases continuously as concentration increases. It is considered that the adsorption of colorants on cotton occurs mainly by hydrogen bonding because it shows Freundlich type isotherm. Dye uptake decreases up to 80 and then increases. Dyeing equilibrium reaches at 60 min. The dye uptake decreases with the increase in pH from neutral to alkaline. The dyed samples show Y color regardless of pH and mordants. Pre-mordanting is more beneficial with regard to getting higher color depth compared with other two methods. The samples with mordanting and without mordanting show relatively good fastness properties except for color change after washing. Mordanting has a marginal effect on the improvement of dye uptake and fastness.

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요 약

개망초로부터 추출한 색소의 면 섬유에 대한 염색성을 조사하였다. 염색조건과 매염처리가 염착량(K/S 값)및 CIELAB 색상에 미치는 영향을 평가하였으며, 또한 염색한 시료의 견뢰도를 측정하여 실용성을 확인하였다. 개망초 색소의 면 섬유에 대한 친화력은 높지 않았으며, Freundlich형의 등온흡착곡선을 보여 염착이 주로 수소결합에 의해 이루어지는 것으로 나타났다. 동시 또는 후매염처리 방법보다는 전매염처리 방법이 염착량 증진에 더 효과적이었으나, 주석매염제를 제외하고 매염제에 의한 염착량 증진효과는 크지 않았다. 매염처리에 관계없이 모든 시료는 Y 계열의 색상을 나타내어 큰 변화는 없었다. 모든 시료의 세탁견뢰도는 낮았으며, 땀 및 마찰견뢰도는 매우 양호하였으며 매염 처리가 견뢰도 증진에 미치는 영향은 크지 않았다.