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Natural Dyeing of Hair using Juglone

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주글론을 이용한 천연 모발염색

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Abstract

The purpose of this study is to investigate the efficacy of juglone, which shows high affinity to wool, for hair dyeing as a natural dye. Before dyeing hair with juglone, virgin hair was bleached by a conventional method using hair shops in the city. In order to study the dyeing properties of juglone on hair, the effects of dyeing conditions on dye uptake, color, tensile strength, and morphology were investigated. And the effect of Fe-mordanting on color change was also investigated. Dye uptake changed marginally as dye concentration increased in the range of 0.02-0.04g. Dye uptake increased progressively as dyeing time increased in the range of 10-20 min. Juglone produced YR colors on hair and the color of dyed hair got duller as juglone concentration increased. The hair was little damaged by bleaching, but cuticles were melted and stripped away by dyeing for longer time. Tensile strength retention decreased up to 65% at 20 min dyeing. Simultaneous mordanting with Fe did not improved dye uptake, but led to darker brown color compared with unmordanted hair.

Key words: Juglone, Hair dyeing, Dye uptake, Color, Tensile strength; 주글론, 모발염색, 염착량, 색상, 인장강도

I. Introduction

Hair dyeing represents an important aspect of cosmetology(Scarpi et al., 1998). Its interest has been growing due to the care taken in self appearance by both women and men. Currently, the interest in natural hair dyeing is dramatically increased because of the harmful influence to environment and toxicity of synthetic hair dyes as same as textile dyeing. Natural hair dyes have more compatibility with environment and biodegradability as well as anti-allergy and anti-cancer(Cho et al., 2003). Despite of those multi-functional features proven by hair dyeing, the lack of color range, the difficulties of securing and custody, low dye concentration, reproducibility, lightfastness, and other low all sorts fastness are critical problems. Additionally, the complexity of dyeing method gives practical difficulties. It is necessary to exploit the potential resources of natural dyes for hair. It is expected that a natural dye showing high affinity to protein fibers has potential to apply as a natural dye for hair.

In previous study(Shin & Moon, 2002), it was found that the colorants extracted from wallnut hull

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in water showed high affinity to wool and produced from yellowish brown to dark brown colors. Juglone (5-hydroxy-1,4-naphoquinone) is the main colorant of unripe wallnut hull(Roth 1998; Sweppe, 1992). It falls under quinone family and produces yellow to brown colors. So, the possibility of using the juglone as a natural hair dyes was expected to lead the same results on hair.

The present work is a basic study to investigate the efficacy of juglone using as a natural hair dye. In order to investigate the dyeing properties of juglone on hair, the effects of dyeing conditions on dye uptake, color, tensile strength, and morphology were studied. In addition, the effect of Fe-mordanting on color change was also evaluated because we found Fe-mordant gave a deeper shade on wool.

II. Experimental

1. Materials

Virgin hair was collected from 11 years old girl and used after scouring followed by bleaching. Juglone in powder form was purchased from Aldrich Chemicals Co. Welloxon Perfect, containing hydrogen peroxide(35%), and BLONOR, containing potassium persulfate as an active material, were obtained from Wella AG, Germany. Ferric sulfate($FeSO_4 \cdot 7H_2O$) as Fe mordant was used. All chemicals were reagent grade.

2. Methods

1) Bleaching

Virgin hair samples were treated with 1:1 mixture of commercial(Wella AG, Germany) bleaching products, potassium persulfate and hydrogen peroxide(9% solution), for 10 min at 50°C.

2) Dyeing and mordanting

Dyeing was carried out at a liquor ratio of 1:50, dye concentration 0.02~0.04 g(1~3% owb), 100°C, 10~20 min. Simultaneous mordanting was done during dyeing process by adding 0.02g(1% owb) of Fe mordant in dyeing solution. 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 Color Eye 3100 spectrophotometer at maximum absorption wavelength. H V/C values were obtained from L*a*b* data using CIE Munsell conversion program.

4) Tensile strength measurement

The tensile strength of a strand of hair was measured with a Rheo-meter(Compac-100II, Sai. Co., Japan) by following KS K 0521 method. Cross head speed was set at 10 mm/min. Average value was determined from measurements of five strands of hair sample. Retention of tensile strength was calculated from tensile strength before and after dyeing.

5) Scanning electron microscopy(SEM)

Scanning electron microscope(JSM 5400, Japan) was used to observe morphological change of the dyed hair with 1000 times of magnification.

III. Results and Discussion

1. Dye uptake and color

<Fig. 1> shows the effect of juglone concentration on dye uptake(K/S). Dye uptake increases as dye concentration increases up to 0.02 g and then is not

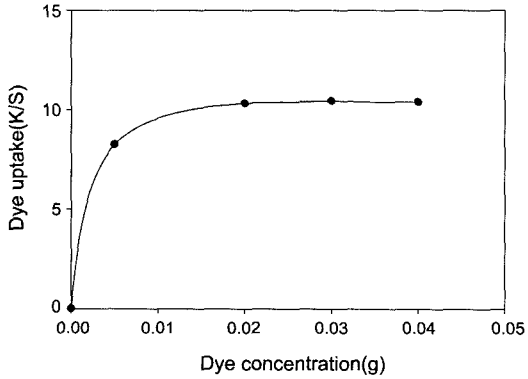

Fig. 1. Effect of dye concentration on dye uptake.

changed. It is considered that accessible dye sites are saturated even at 0.02 g concentration. Compared with the results for wool fiber reaching up to 18-20

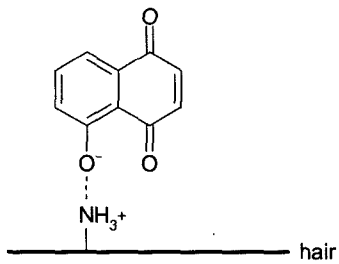

Fig. 2. Ionic bond between hair and juglone.

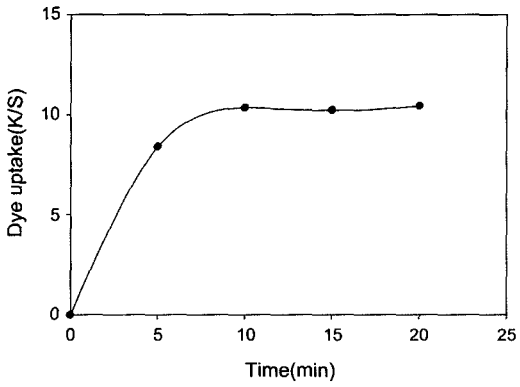

Fig. 3. Effect of dyeing time on dye uptake.

of K/S value(Shin and Moon, 2002), hair shows lower affinity than wool. The adsorption isotherm, as shown in <Fig. 1>, is known as the Langmuir type (Trotman, 1970). This is obtained where there are a definite nimber of dyeing sites within the fibers which can enter into combination with the dyes. Therefore, it is speculated that amino groups in hair bind with dye anion, as shown in <Fig. 2>.

<Fig. 3> shows that dye uptake increases for initial 10 min and then reaches equilibrium. Considering the characteristic of hair dyeing, the dyeing time is the better the shorter.

<Tables 1 and 2> show the effect of dyeing time and concentration on CIELAB color values. 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 & Saltzman, 1981).

As shown in <Table 1>, L*, a* and b* values decrease as dyeing time increases up to 10 min, indicating the change of color to darker, duller, and less red and yellow shade. Thereafter they are not much changed up to 20 min. Hue, value and chrome values are changed progressively with dyeing time up to 10

Table 1. Effect of dyeing time on L*, a*, b*, & H V/C of the dyed hair

Dyeing time(min)	L*	a*	b*	H	V/C
0(bleached)	70.269	3.849	18.855	9.95YR	6.86/3.02
5	57.851	2.877	10.750	9.35YR	5.60/1.77
10	25.649	0.866	0.975	4.83YR	2.46/0.22
15	25.362	0.302	0.272	3.36YR	2.43/0.07
20	25.377	0.600	0.638	4.51YR	2.43/0.15

dye conc.; 0.02 g

Table 2. Effect of juglone concentration on L*, a*, b*, & H V/C of the dyed hair

Dye conc.(g)	L*	a*	b*	H	V/C
0(bleached)	70.269	3.849	18.855	9.95YR	6.86/3.02
0.005	61.068	3.441	13.227	9.35YR	5.92/2.18
0.02	25.649	0.866	0.975	4.83YR	2.46/0.22
0.03	25.269	0.479	0.593	5.56YR	2.42/0.13
0.04	25.055	0.073	0.096	6.01YR	2.40/0.02

dyeing time; 10 min

min and then not much changed.

<Table 2> shows color change with dye concentration. L*, a*, and b* values decrease as dye concentration increases up to 0.02 g, getting darker and duller color with less redness and yellowness. At higher concentration, color values are changed marginally.

2. Morphology

<Fig. 4> shows SEM pictures of the virgin, bleached, and dyed hairs with different dyeing time. Cuticles on the surface of virgin hair(a) are well developed, and are not damaged by bleaching(b). The severe damages of cuticles are observed in the hair dyed for 10 min(c). Whereas, cuticles on the hair dyed for 20 min(d) are completely stripped away, appearing smooth surface and thinner thickness. Keratin fiber would be hydrolyzed in dyeing in acidic condition with high temperature, resulting the breakdown of S-S linkages and causing damage on hair fiber. This contributes to the decrease in hair strength.

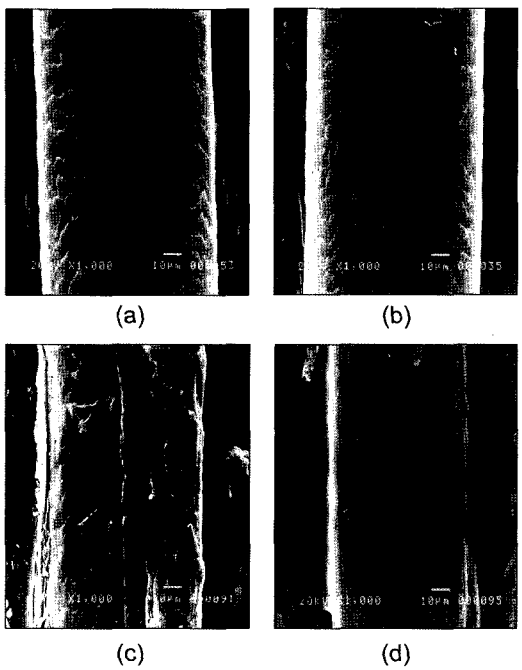

Fig. 4. SEM pictures of dyed hairs; (a) virgin hair, (b) bleached, and dyed; (c)10 min, (d) 20 min.

Bae(2003) carried out morphological study on hair damage by permanent dyeing and found that dyeing caused splaying edge, splitting, tearing off, and finally dissolving of hair cuticle, leading to decrease hair thickness. It is known that the repeated dyeing causes more severe damages than temperature factor.

3. Tensile Strength

<Fig. 5> shows tensile strength retention depending on dyeing time. As dyeing time increases, tensile strength retention decreases up to 65% at 20 min dyeing. In juglone solution of pH 5.3 with high temperature for prolonged periods, as shown in chemical reaction below, the disulfide linkage of hair is hydrolyzed to give a thiol group(II) as well as a sulfenic acid(I), which can break down to give an aldehyde and liberate hydrogen sulfide(Peters, 1963). The cleavage of disulfide linkage leads to the weakening of hair.

$$\underset{}{RS\text{-}SR} \rightarrow \underset{(I)}{RS\text{-}OH} + \underset{(II)}{R\text{-}SH} \rightarrow R\text{-}CHO + H_2S + R\text{-}SH$$

Also, hydrolysis of peptide groups would occur to some extent during dyeing, resulting in the degradation of hair and thereby the loss of tensile strength. It has been known that wet tensile properties of keratin fibers are related to the disulfide bonds, whereas the dry tensile properties are influenced more by peptide bonds(Robbins, 1988).

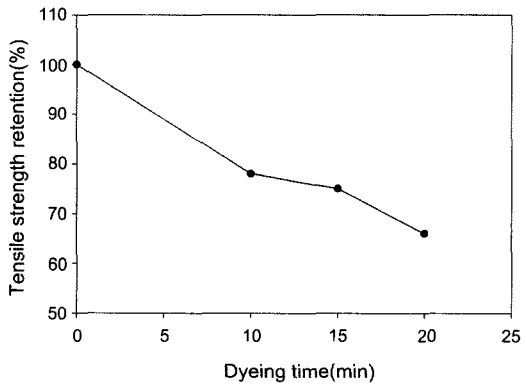

Fig. 5. Effect of dyeing time on tensile strength retention.

Table 3. Effect of Fe mordanting on L*, a*, b*, & H V/C of the dyed hairs

Sample	K/S	L*	a*	b*	H	V/C
Control	10.036	25.649	0.866	0.975	4.83YR	2.46/0.22
Mordanted	10.437	24.967	0.315	0.414	6.02YR	2.39/0.09

4. Effect of Fe-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. The influence of mordanting on K/S and color values of the dyed fabrics are compared in <Table 3>. From the results, it is observed that mordanting is not an effective method for improving dye uptake in the case of juglone. Some of dye is lost in simultaneous mordanting method, because of the formation of insoluble complex in the dyebath itself. This phenomenon leads to a decrease in the effective dye concentration in the dyebath(Deo & Desai, 1999).

L* value decreases slightly by mordanting, meaning that shade of the samples gets darker. a* and b* values decrease, indicating the decrease in redness and yellowness. Irrespective of mordanting, the samples show YR colors. It did not show significant change in value, but chroma decreases. Compared with the unmordanted hair, duller and darker brownish colors are obtained by mordanting. It is speculated that different mordant types would produce different depth and shade of YR colors, such as orange brown, yellowish brown, reddish brown, dark brown, etc., referring to the results of wool dyeing with juglone(Shin & Moon, 2002)

From the results obtained, It is considered that juglone is possible to use a natural hair dye for producing YR colors. Color variations within YR colors could be achieved by mordanting.

IV. Conclusion

The dyeing properties of juglone on hair were investigated. The effect of dyeing conditions and mordanting were evaluated in terms of dye uptake (K/S value) and CIELAB color values. Morphological changes were investigated by SEM. And the effect of dyeing time on tensile strength was evaluated.

Dye uptake increased as dye concentration increased up to 0.02 g and then almost saturated. The adsorption isotherm was the Langmuir type. Therefore, it was speculated that amino groups in hair bind with dye anion. Dye uptake increased for initial 10 min and then reached equilibrium. The dyed hair with juglone showed YR color. L*, a* and b* values decreased as dyeing time increased up to 10 min, indicating change of color to darker, duller, and less red and yellow shade. L*, a*, and b* values decreasd as dye concentration increased up to 0.02 g, getting darker and duller color with less redness and yellowness. The severe damages of cuticles were observed in the dyed hairs, especially in the hair dyed for 20 min shows complete stripping of cuticles. Tensile strength retention decreased up to 65% at 20 min dyeing. Fe-mordanting changed the color of hair to duller and darker brownish shade.

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요 약

모섬유에 높은 염착성을 가진 호두외피의 주색소인 주글론을 모발염색에 적용 가능성을 알아보기 위해 주글론의 모발에 대한 염색성을 조사하였다. 모발염색을 하기 위해서 먼저 시중 헤어전문점에서 사용하는 방법으로 모발을 표백하였다. 모발염색의 특성상 염색시간을 10-20분으로 섬유염색보다 훨씬 짧게 설정하였으며 염착성, 인장강도, 전자현미경 분석을 하였다. 염색시간이 증가함에 따라 염착성은 증가하는 경향을 보였으며 인장강도는 떨어져 모발 손상이 일어난 것을 알 수 있었다. 모발손상은 전자 현미경 사진으로 확인 되었다. 주글론 염색에 의해 모발에 YR 계열의 색상을 낼 수 있엇다. 동시매염 방법으로 철매염제를 사용한 결과 염착성 증진은 크지 않았으나, YR 계열 내에서 더 진한 색상을 얻을 수 있었다. 천연 모발염색에 주글론을 사용할 수 있을 것으로 사료되며, 염색조건에 따라 다양한 갈색계열 색상을 부여할 수 있다.