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Dyeing and Color Fastness Properties of Natural Dyed Actual Size Hanji

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ABSTRACT

After manufacturing the natural dyed actual size Hanji using 11 kinds of plant natural dyeing materials and 2 kinds of animal natural dyeing materials, the color characteristics and color change and color fastness after post-mordanting were compared and discussed. The hues of 13 types of natural dyed Hanji were black, PB, and RP, each with 1 type, YP with 3 types, and Y with 7 types. Among the natural dyeing materials, Chinese ink, indigo and Lac showed high color yield and color difference, and violet-root cromwell and gardenia seeds showed low color yield. The color fastness of Hanji dyed with turpentine diluted Ottchil, Pagoda tree seeds, Chinese ink and indigo was excellent, while that of gardenia seeds and violet-root cromwell were very poor. After post-mordanting of natural dyed Hanji with Al, Cu, and Fe mordants, the hue changes were show up the Alnus firma, clove, lac and cochineal. In addition, the color difference was very diverse and was overall the most biggest due to Fe mordant. After 72 hr. of UV irradiation on post-mordanting natural dyed Hanji, hue change was observed in 3 types and color fastness was improved in 8 types by post-mordanting. The Hue and color fastness are significantly different depending on the type of natural dying materials and post-mordants. Therefore, when dyeing Hanji with natural dyes, it is necessary to dye with sufficient knowledges and informations about the desired color and fastness.

Keywords: Hanji, natural dyeing materials, post-mordanting, color difference, color yield, color fastness

1. INTRODUCTION

Paper was reportedly introduced to Korea between the 2nd and 7th centuries, since paper was manufactured systematically using fibers from the tree bark and hemp by Chaeryun around 105 AD. The culture and manufacturing technology of Korean ancestors for using paper and wood products were quite excellent (Han and Lee, 2021a, 2021b; Han *et al*., 2021; Yoon *et al*., 1996). Hanji (Korean traditional paper) from the Goryeo Dynasty is considered to be of a superior quality because the raw material of the paper is the fiber from tree mulberry and the mucilage from sunset hibiscus is used as a fiber dispersant. Other possible reasons include the bleaching method minimizing damage to the fibers, the water used during the Hanji making and washing processes, and a unique Hanji making method (Heullim tteugi), which leads the fibers' orientation in perpendicular to

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each other. Hanji quality has also significantly changed according to the times, functions, and purposes, and while records of hundreds of different types of papers have appeared in ancient documents, however, little data is available regarding their manufacturing methods.

In literature records and relics, different types of Hanji are called by different names depending on the material, use, color, manufacturing region, and processing method. In particular, a lot of Hanjis are dyed as a method to impart diversity, aesthetics, and functionality.

Among the colored papers that appear in the ancient documents, red-colored papers include Do-Wha-Ji (桃花紙), Dan-Mok-Ji (丹木紙), Hong-Jeo-Ju-Ji (紅楮注紙), Hong-Dang-Ji (紅唐紙), Hong-Pae-Ji (紅牌紙), and Mae-Hong-Ji (梅紅紙) , blue-to-green-colored papers include Cheong-Jeo-Ju-Ji (青楮注紙), Ah-Cheong-Cho-Ju-Ji (鴉青草注紙), Cheong-Saek-Ji (青色紙), Cheong-Dang-Ji (青唐紙), Cheong-Bak-Ji (青箔紙), Cheong-Seon-Ja-Ji (青扇子紙), Ah-Cheong-Ji (鴉青紙), Chi-Ji (翠紙), Cho-Rok-Ji (草綠紙), and Cho-Rok-Jeo-Ju-Ji (草綠楮注紙); , yellow-colored papers include Whang-Guk-Ji (黃菊紙), Whang-Jeo-Ju-Ji (黃楮注紙), Whang-Yeom-Cho-Ju-Ji (黃染草注紙), and Sang-Ji (橡紙), black-colored Hanji is Mook-Ji, gold-colored Hanji is Hanji coated with gold powder (冷金箋紙), and silver Hanji is coated with silver foil (銀箋紙). Unfortunately, although a large variety of colored Hanji used to exist, there are hardly any records of the natural dyes and methods that were used to manufacture them. As natural dyes for manufacturing colored Hanji, for red color, Safflower, Madder, dried Violet-root cromwell, and Sappan wood were used; for blue, Indigo and Ninam were used; for yellow, Pagoda tree, Amur cork-tree, Barberry root, Gardenia, and smoke tree were used. The dyes primarily used for black include gallnut, Amur maple, and Chinese ink (Meok).

Natural dyes used in the study for manufacturing natural dyed Hanji include Amur cork-tree (Choi, 2006), Clove tree (Jeon *et al*., 2006), Alder fruit (Choi *et al*., 2009), Gardenia (Yoo *et al*., 2011), Sappan wood, Gallnut (Park and Yoon, 2009, 2010, 2011), Pagoda tree (Yoo *et al*., 2009), Onion peel (Jeon, 2003), Mugwort (Jeon *et al*., 2006), *Neolitsea sericea* (Jo *et al*., 2007), Smoke tree (Lee *et al*., 2009), Persimmon juice (Yoo *et al*., 2010), Chestnut husk, Gallnut (Lee *et al*., 2009), etc. In addition, Gardenia and Sappan wood (Ju and Roh, 2020) have been used for regenerated wood fibers.

In general, the strength of Hanji depends on the fiber direction, the degree of lamination, and the coating, but the wet strength is weaker than that of the fabrics (Jeon, 1999, 2002). Therefore, most studies on natural dyeing have been mainly conducted on small-sized Hanji or fabric because Hanji, which has very weak wet strength compared to fabrics, is easily torn when immersed in a dye solution and dyed, and is rarely dyed once, and there are many cases of damage in the process of repeating several times. In addition, Hanji is more prone to damage when dyeing with a large one without cutting.

In this study, 13 types of natural dyed Hanji were manufactured using 11 types of natural plant ingredients and 2 types of natural animal materials that have previously not been applied to Hanji, and their color characteristics were compared and analyzed. Thereafter, color change due to post-mordanting, and color and fastness due to the type of natural dye used were reviewed.

2. MATERIALS and METHODS

2.1. Hanji

Hanji dyeing with natural dyes, has weak wet strength, must be conducted very carefully and is easy to fail unless Hanji prepared in the traditional way is used. It is more difficult to dye the big Hanji (66 cm × 94 cm) as it is manufactured. In this study, Hanji manufactured by a Hanji artisan with Heullim tteugi was purchased for natural dyeing, which was 66 cm × 94 cm in the size with the weight of 28–30 g/m^2.

2.2. Natural dyeing materials

As a natural dye for Hanji, plant materials such as Gardenia seeds, Amur cork-tree bark, *Alnus firma* fruit, Pagoda tree seed, Sappan wood, Violet-root cromwell, Indigo, Meok, Clove tree, and Ottchil (Oriental lacquer diluted with ethanol or turpentine) and animal materials such as Cochineal and Lac were used. Dyeing is a post-dyeing method in which a dye solution is extracted in consideration of the characteristics of the natural dye material, then immersed in the dye solution for a certain period, and repeated as needed. The type number, scientific or English name, and density of each dyed Hanjis are shown in Table 1. The density of Hanji was 0.35 g/cm^3, and after dyeing, only the density of Indigo-dyed Hanji slightly increased while the rest decreased.

2.3. Dyeing with natural ingredients

2.3.1. Ottchil (No. 2, 3)

Raw oriental lacquer was purchased and stored at 5℃ or lower before use. Ottchil solution (Park *et al*., 2020), typically used for coating wood products, was prepared by diluting 1,000 g of Ottchil in 10 L of turpentine (TP) and ethanol (ET). Each solution was placed in a separated stainless-steel container and Hanji was immersed in the solutions for 10 min., to prepare two types of Ottchil-dyed Hanji (TP, ET).

2.3.2. *Alnus firma* fruit (No. 4)

The seeds of *Alnus firma* tree, i.e., strobilus, which were hung after the leaves fell in winter were collected from Jinju, Gyeongsangnam-Do, and used further. 10 L of water was added to 500 g of strobilus, boiled over high heat, simmered over low heat for another 20 min., and finally the dye-extracted solution was filtered. Again, 10 L water was added to the strobilus and the extraction process was repeated four times. The final extract was mixed and used as a dyeing solution. For the dyeing process, the extracted dyeing solution was placed in a square stainless-steel container, and Hanji was immersed

Table 1. Type number, Korean name, and scientific or common name of natural dyeing materials used to dye the Hanji

Type no.	Korean name	Scientific or common name	Density of Hanji, after dyeing (g/cm^3)
1	원지, 한지	Control, Hanji	0.35
2	옻 TP(테레빈유)	Ottchil (Turpentine)	0.34
3	옻 ET(에탄올)	Ottchil (Ethanol)	0.32
4	사방오리나무 열매	*Alnus firma* Sieb. et Zucc., *Alnus firma* fruit	0.33
5	치자 열매	*Gardenia jasmininoides*, Gardenia seeds	0.32
6	괴화나무 열매	*Sophora japonica* L., Pagoda tree seeds	0.32
7	정향나무	*Eugenia caryophyllata*, Clove tree	0.32
8	황벽나무 수피	*Phellodendron amurense* Rupr., Amur cork-tree bark	0.32
9	먹	Meok, Chinese ink	0.32
10	쪽	*Persicaria tinctoria* (Aiton) H. Gross, Indigo	0.36
11	락	*Laccifer lacca, Kerria lacca*, Lac	0.31
12	자초 뿌리	*Lithospermum erythrorhizon* Sieb. et Zucc., Violet-root cromwell	0.31
13	코치닐	*Coccus cacti* L. Cochineal	0.31
14	소목	*Caesalpinia sappan* L., Sappan wood	0.31
15	복사 용지	Copy paper	0.70

twice at 50℃–60℃ for 15 min.

2.3.3. Gardenia seeds (No. 5)

Gardenia seeds (500 g) grown and dried in Jinju, Gyeongsangnam-Do were soaked in 10 L water for one day, boiled over high heat, simmered over low heat for about 20 min., and filtered to obtain an extract. Again, 10 L of water was added, and the extract obtained by repeating the same process thrice was combined and used as a dyeing solution for Gardenia seeds. Dyeing was performed for 15 min., by immersing Hanji in the dyeing solution at 50℃–60℃, and repeated twice.

2.3.4. Pagoda tree seeds (No. 6)

Pagoda tree (*Sophora*) seeds of 100 g that were purchased from oriental medicine store were soaked in 2 L water for approximately 1 hr. Thereafter, another 2 L of water was added, and the mixture was boiled over high heat, maintained over low heat for 15 min., and filtered to obtain an extract. Water (2 L) was added again, and the extract obtained by repeating the same method thrice was used as a dyeing solution. For the dyeing process, the extracted dyeing solution was placed in a square stainless-steel container, and Hanji was immersed twice at 50℃–60℃ for 15 min.

2.3.5. Clove tree (No. 7)

Clove tree (200 g) was bought from an oriental medicine store and washed with water. Thereafter, 10 L of water was added, boiled, simmered over low heat for another 20 min., and filtered down to obtain an extract. Again, 10 L water was added, and the extract obtained by repeating the same method thrice was used as a dyeing solution. For the dyeing process, the extracted dyeing solution was placed in a square stainless-steel container, and Hanji was immersed thrice at 50℃–60℃ for 15 min.

2.3.6. Amur cork-tree bark (No. 8)

Water (4 L) was added to 100 g of Amur cork-tree bark purchased at an oriental medicine store, boiled over high heat, and simmered over low heat for an additional 20 min. Thereafter, the extract was separated by filtration, 4 L of water was added, and the extract obtained by repeating the same process thrice was used as a dyeing solution. For the dyeing process, the extracted dyeing solution was placed in a square stainless-steel container, and Hanji was immersed thrice at 50℃–60℃ for 15 min.

2.3.7. Meok (No. 9)

A dye solution was prepared by mixing the Chinese ink purchased from a stationery store with water in a 1:10 ratio. For the dyeing process, the extracted dyeing solution was placed in a square stainless-steel container, and Hanji was immersed twice at 40℃–50℃ for 15 min.

2.3.8. Indigo (No. 10)

For Indigo, an imported Indigo powder was purchased and used. Indigo dye solution contained 100 g powdered Indigo dissolved in 300-mL water at 60℃–70℃ and 200 g potassium carbonate in 300 mL water at 60℃–70℃ in a stainless-steel container containing 1,100 mL water at 50℃–60℃. The prepared solution and 200 g hydrosulfite dissolved in 300 mL water at 60℃–70℃ were sequentially added, followed by mixing. Further, Hanji was dip dyed in an Indigo dye solution at 50℃–60℃ for 15 min., followed by oxidative color development in air for 20 min. This process was repeated five times, and finally washed with water thrice.

2.3.9. Lac (No. 11)

Lac from a dye material dealer was purchased and used. 50 g of Lac resin finely ground with a grinder was placed in 10 L of water with 100 mL of vinegar, boiled over high heat, simmered over low heat for 15 min., and then filtered to obtain a solution. Thereafter, water was added again, and the solution obtained in the same way was combined and used as the Lac dye solution. For the dyeing process, the extracted dyeing solution was placed in a square stainless-steel container, and Hanji was immersed twice at 50℃-60℃ for 15 min.

2.3.10. Violet-root cromwell (No. 12)

Domestic grown Violet-root cromwell was purchased, dried, and used. Dried Violet-root cromwell (200 g) was washed and added to 4 L of water at 40℃ with 30 mL of vinegar, and it was soaked for 20 min. and scooped out. Thereafter, the Violet-root cromwell removed with a wooden bat was beaten, put in a burlap bag, and rubbed in water at 40℃-50℃ to extract the dye solution. Further, the Violet-root cromwell residue was put back into water, extracted the same way thrice, and the obtained solution was mixed and used as the Violet-root cromwell dyeing solution. For the dyeing process, the extracted dyeing solution was placed in a square stainless-steel container, and Hanji was immersed at 40℃-50℃ for 15 min., dried, and this process was repeated thrice.

2.3.11. Cochineal (No. 13)

Cochineal was purchased from a dye material supplier. Finely grounded with a grinder, 100 g of cochineal was put into 10 L of water with 100 mL vinegar, boiled over high heat, simmered over low heat for 15 min., and filtered to obtain a solution that was used as the dyeing solution. For the dyeing process, the extracted dyeing solution was placed in a square stainless-steel container, and Hanji was immersed at 50℃-60℃ for 15 min., dried, and this process was repeated twice.

2.3.12. Sappan wood (No. 14)

Sappan wood was purchased from an oriental medicine store. For the Sappan wood dye solution, 100 g of Sappan wood was pre-soaked in about 2 L of water for 1 hr., thereafter, more 2 L of water was added, boiled over high heat, simmered over low heat for 15 min., and filtered to obtain an extract. Thereafter, 4 L of water was added, and the extract obtained by repeating extraction thrice in the same way was mixed and used as the Sappan wood dyeing solution. For the dyeing process, the extracted dyeing solution was placed in a square stainless-steel container, and Hanji was immersed at 50℃-60℃ for 15 min., dried, and this process was repeated thrice.

2.3.13. Copy paper (No. 15)

Copy paper used for comparison was white paper with a basis weight of 75 g/m^2.

2.4. Mordanting of natural dyed Hanji

For post-mordanting, aluminum ammonium sulfate dodecahydrate, Copper (II) acetate anhydrous, Iron(II) sulfate heptahydrate (Extra pure, Daejung) were added at 5% (O.W.F) of the Hanji weight and 50:1 liquid ratio as mordants, treated at 40℃ for 30 min., and washed with distilled water thrice.

2.5. Measurement of color characteristics

The surface color of natural dyed Hanji was measured using Minolta CM-3800d (Tyoko, Japan). The color difference of Hanji treated here was calculated based on the color of Hanji before dyeing for the dyed Hanji, dyed Hanji before mordanting for the mordanted dyed Hanji, and dyed and/or mordanted Hanji before UV irradiation for UV irradiated dyed and/or mordanted Hanji, respectively. The K/S value was calculated by the Kubelka-Munk equation after measuring the reflectance coefficient of each dyed sample.

2.6. Evaluation of color fastness

The color fastness of the natural dyed Hanji was measured using an accelerated weathering tester (QUV/SP, USA) before and after UVB-313 lamp, illuminance of 0.67 W/m^2, and UV irradiation treatment at 50℃ for a certain period. The color difference was evaluated in the CIE Lab color space, typically used to indicate the standard color space for wood products (Hadi *et al*., 2020; Lee and Lee, 2021; Priadi *et al*., 2020).

3. RESULTS and DISCUSSION

3.1. Color characteristics and fastness of natural dyed Hanji

3.1.1. Color characteristics

The hue (H) of Hanji before dyeing was 4.3Y, and the L* and V values were very high, indicating a yellowish color that was close to white. Table 2 and Fig. 1 show the color characteristics of the actual size Hanji, dyed with 13 natural dyes. Among all the natural dyes, Chinese ink, Indigo, and Lac showed high dyeing yield and color difference, and Lac showed significantly high color difference compared to the dyeing yield. Conversely, the dyeing yield was less than 1 for Violet-root cromwell and Gardenia seeds, and the color difference was less than 10.0 for Pagoda tree seeds, Clove tree, and turpentine-diluted Ottchil.

Among the Hanjis dyed with the 13 dyes, Hanji dyed with two types of Ottchil, *Alnus firma* fruit, Gardenia tree seeds, Pagoda tree seeds, and Clove tree presented a yellow color (H) of 1.2–4.7Y. Amur cork-tree bark expressed a yellow color close to green at 9.8Y. The dyeing yield values of these seven natural yellow dyes was in the range of 1.0 to 4.19, the highest in *Alnus firma* fruit and the lowest in Gardenia tree seeds extract. The color difference of post- dyeing Hanji was in between 5.5 and 19.3, which was high in ethanol-diluted Ottchil, *Alnus firma* fruit, Gardenia tree seeds, and Amur cork-tree bark in the order, and was low in the order of Pagoda tree seeds, Clove tree, and terpentine-diluted Ottchil. Chinese ink-dyed Hanji showed extremely high dyeing yield and color difference and its Munsell’s value was V 2.82 and C 0.42.

Indigo-dyed Hanji (5 times dyed), which was dyed more 2–3 times than the others, showed 6.04 in the dyeing yield and 53.3 in the color difference so that it ranked in the second highest place after Meok. Its hue was a blue type (3.7PB) and a* and b* were −2.64 and −14.32, respectively.

The hue of Hanji dyed with Lac, a natural animal dye, was 7.7RP and a* was as large as 19.6 and b* was 1.1, indicating red purple. The hues of Hanji dyed with plant dye Violet-root cromwell and Sappan wood and an animal dye cochineal were 5.8YR, 3.2YR, and 8.2YR, respectively, indicating a yellow red. The dyeing yield

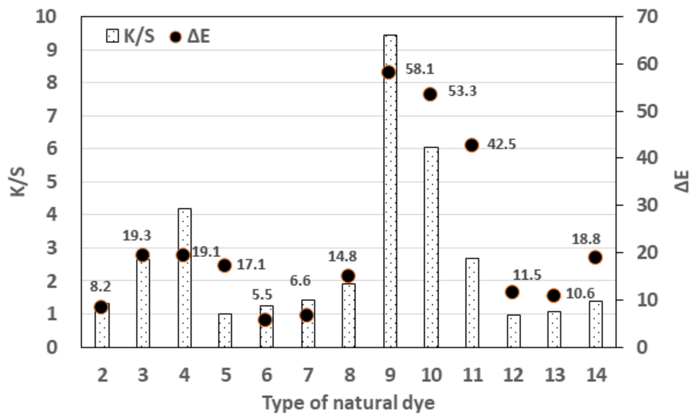

Fig. 1. Dyeing yield (K/S) and color difference (△E) of various natural dyed Hanjis.

Table 2. Color characteristics of various natural dyed Hanjis

Type of natural dye	λmax (nm)	K/S	CIE Lab				Munsell's value		
			L*	a*	b*	△E	H	V	C
1	360	0.49	86.81	−0.30	7.96		4.3Y	8.58	0.94
2	360	1.32	84.56	−0.09	15.86	8.21	4.0Y	8.37	2.02
3	360	2.66	75.33	4.27	22.78	19.25	1.2Y	7.45	3.44
4	360	4.19	77.11	2.83	24.20	19.14	2.3Y	7.63	3.50
5	360	1.00	84.33	0.52	24.82	17.06	4.7Y	8.39	3.41
6	360	1.23	85.40	−0.33	13.29	5.50	3.9Y	8.45	1.64
7	360	1.40	85.45	−0.28	14.44	6.61	4.2Y	8.46	1.80
8	360	1.91	86.38	−4.31	22.18	14.79	9.8Y	8.57	2.75
9	370	9.44	28.92	0.68	2.57	58.05		2.82	0.42
10	650	6.04	38.30	−2.64	−14.32	53.34	3.7PB	3.67	3.45
11	530	2.69	49.74	19.58	1.10	42.54	7.7RP	4.84	4.93
12	360	0.96	76.07	4.01	7.84	11.49	5.8YR	7.49	1.57
13	360	1.05	78.26	5.81	8.82	10.57	3.2YR	7.71	2.15
14	360	1.39	76.93	8.63	21.23	18.77	8.2YR	7.62	3.75
15	360	0.94	91.70	2.78	−12.83	21.59		9.02	3.94

increased in the order of Sappan wood, cochineal, and Violet-root cromwell.

3.1.2. Color fastness

Table 3 shows the color fastness after 72 hr. UV irradiation on the undyed Hanji and dyed Hanji with each natural dye. Even with the harsh UV irradiation at 50℃ for 72 hr., the small color difference implied good color fastness. The color difference of undyed Hanji after 72 hr. of UV irradiation was 2.23, indicating even undyed Hanji also underwent color change by UV rays. Among these 13 natural dyed Hanji, the ones with less color difference than the undyed Hanji were turpentine-diluted Ottchil, Pagoda tree seeds, Meok, and Indigo-dyed Hanji. The color difference of Meok-dyed Hanji was 0.03. It implies the color of Meok-dyed Hanji could be maintained without fading away even after 72 hr. of UV irradiation. In addition, the color difference of Pagoda tree seeds and Indigo-dyed Hanji was also less than 1, indicating an excellent color fastness where color change can only be prevented by dyeing. Conversely, the color difference of Gardenia-dyed Hanji after the UV irradiation was very high at 15.2, followed by 11.6 of Violet-root cromwell, considered a dye material with poor color fastness. Amur cork-tree bark, Cochineal, ethanol-diluted Ottchil, and Sappan wood were used as dyes with a color difference of 6 to 9.

UV irradiation decreased the dyeing yield on most of the dyed Hanjis, but for Meok-dyed Hanji, it slightly increased. The color value of most of the natural dyed Hanjis increased, while those of *Alnus firma*, Amur cork-tree, and Meok-dyed ones decreased. After 72 hr. of UV irradiation, most of the natural dyed Hanji did not change the color series, but Violet-root cromwell dyed Hanji changed from 5.8YR to 2.7Y, and Sappan wood changed from 8.2YR to 0.2Y.

Table 3. Color characteristics of various natural dyed Hanjis after 72 hr. UV radiation

Type of natural dye	UV radiation (hr.)	λmax (nm)	K/S	CIE Lab				Munsell's value		
				L*	a*	b*	△E	H	V	C
1	0	360	0.49	86.81	−0.30	7.96		4.3Y	8.58	0.94
	72	360	0.49	87.36	−0.63	10.06	2.23	5.1Y	8.64	1.19
2	0	360	1.32	84.56	−0.09	15.86	8.21	4.0Y	8.37	2.02
	72	360	0.82	85.80	−0.58	14.99	1.24	4.8Y	8.5	1.86
3	0	360	2.66	75.33	4.27	22.78	19.25	1.2Y	7.45	3.44
	72	360	1.59	80.85	2.41	21.88	6.33	2.3Y	8.01	3.14
4	0	360	4.19	77.11	2.83	24.20	19.14	2.3Y	7.63	3.50
	72	360	3.96	74.10	4.44	25.52	3.17	1.6Y	7.33	3.83
5	0	360	1.00	84.33	0.52	24.82	17.06	4.7Y	8.39	3.41
	72	360	0.41	87.15	−0.47	10.89	15.22	5.1Y	8.63	1.32
6	0	360	1.23	85.40	−0.33	13.29	5.50	3.9Y	8.45	1.64
	72	360	0.86	86.19	−0.34	13.34	0.67	4.2Y	8.53	1.65
7	0	360	1.40	85.45	−0.28	14.44	6.61	4.2Y	8.46	1.80
	72	360	0.77	87.20	−0.41	11.68	3.06	4.5Y	8.63	1.42
8	0	360	1.91	86.38	−4.31	22.18	14.79	9.8Y	8.57	2.75
	72	360	0.73	85.77	−0.05	15.39	8.42	4.1Y	8.5	1.96
9	0	370	9.44	28.92	0.68	2.57	58.05		2.82	0.42
	72	370	9.69	28.67	0.68	2.63	0.03		2.8	0.43
10	0	650	6.04	38.30	−2.64	−14.32	53.34	3.7PB	3.67	3.45
	72	650	5.82	39.59	−3.19	−14.36	0.98	3.3PB	3.79	3.53
11	0	530	2.69	49.74	19.58	1.10	42.54	7.7RP	4.84	4.93
	72	530	2.40	50.78	17.43	1.72	3.14	8.3RP	4.94	4.43
12	0	360	0.96	76.07	4.01	7.84	11.49	5.8YR	7.49	1.57
	72	360	0.57	85.38	0.61	12.23	11.59	2.7Y	8.45	1.59
13	0	360	1.05	78.26	5.81	8.82	10.57	3.2YR	7.71	2.15
	72	360	0.70	82.34	2.89	10.62	6.67	8.8YR	8.13	1.71
14	0	360	1.39	76.93	8.63	21.23	18.77	8.2YR	7.62	3.75
	72	360	1.19	79.99	4.54	18.75	5.79	0.2Y	7.92	2.93
15	0	360	0.94	91.70	2.78	−12.83	21.59		9.02	3.94
	72	370	0.37	92.01	0.55	−5.45	7.37		9.08	2.18

Fig. 2 shows the comparison of color differences of the natural dyed Hanji after exposure to UV irradiation for 24, 48, and 72 hr. Regardless of the UV irradiation time, the color difference in turpentine-diluted Ottchil, Pagoda tree, Meok, and Indigo-dyed Hanjis was little, on the other hand the color difference in Gardenia, Violet-

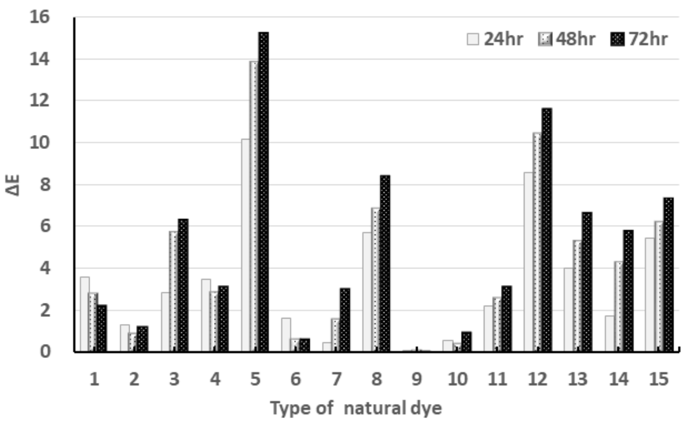

Fig. 2. Comparison of △E according to UV radiation time for each natural dyed Hanjis.

root cromwell, and Amur cork-tree-dyed Hanjis was big. For most of the natural dyed Hanji, prolongation of UV irradiation time resulted in increase of the color difference and gradual color discoloration. However, turpentine-diluted Ottchil, *Alnus firma*, and Pagoda-dyed Hanji showed that the color difference at 24 hr. UV irradiation was smaller than that at 72 hr. UV irradiation.

3.2. Color characteristics and color fastness of the natural dyed Hanji after post-mordanting

3.2.1. Color characteristics of the natural dyed Hanji after post-mordanting

Table 4 and Fig. 3 show the results of post-mordanting Hanji dyed with 13 natural dyes using Al, Cu, and Fe mordants. The color difference of the post-mordanted dyed Hanji by the three mordants was very big. Even none-dyed Hanji showed color difference of 0.29, 2.58, and 10.32 by Al, Cu, and Fe mordants, respectively. Thus, there was almost no difference after Al mordant, but a clear difference for Cu, and in Fe mordants, there was a very big color difference due to a decrease in L*

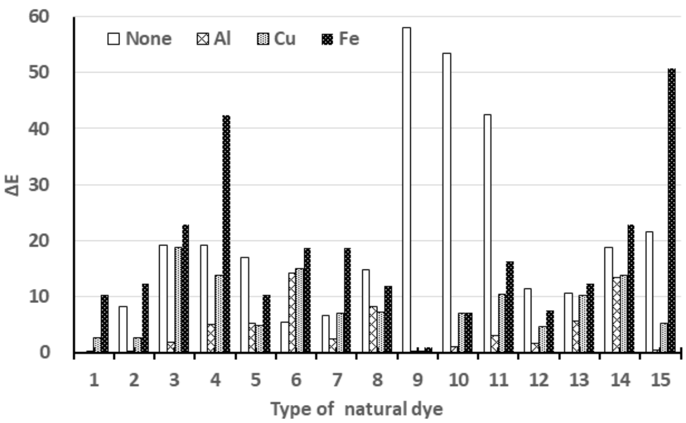

Fig. 3. Color difference of various natural dyed Hanjis after post-mordanting.

Table 4. Color characteristics of post-mordanted natural dyed Hanjis

Type of natural dye	Type of post-mordanting	λmax (nm)	K/S	CIE Lab				Munsell's value		
				L*	a*	b*	△E	H	V	C
1 Control (Hanji)	None	360	0.49	86.81	−0.30	7.96		4.3Y	8.58	0.94
	Al	360	0.43	86.99	−0.44	7.78	0.29	4.8Y	8.60	0.91
	Cu	360	0.58	85.44	−1.72	9.64	2.58	8.8Y	8.44	1.10
	Fe	360	1.36	80.34	2.45	15.52	10.32	1.2Y	7.94	2.26
2 Ottchil (Turpentine)	None	360	1.32	84.56	−0.09	15.86	8.21	4.0Y	8.37	2.02
	Al	360	1.27	84.73	−0.28	15.81	0.27	4.3Y	8.39	2.00
	Cu	360	1.43	82.24	−0.55	16.95	2.60	4.9Y	8.14	2.18
	Fe	360	2.32	73.70	4.80	19.09	12.33	0.2Y	7.28	2.98
3 Ottchil (Ethanol)	None	360	2.66	75.33	4.27	22.78	19.25	1.2Y	7.45	3.44
	Al	360	2.74	73.69	3.96	21.94	1.86	1.4Y	7.28	3.29
	Cu	360	3.23	59.56	2.35	12.91	18.70	2.0Y	5.83	1.91
	Fe	360	3.88	56.26	1.02	10.74	22.79	3.2Y	5.49	1.53
4 *Alnus firma*	None	360	4.19	77.11	2.83	24.20	19.14	2.3Y	7.63	3.50
	Al	360	4.38	74.30	2.89	28.47	5.11	2.9Y	7.35	4.09
	Cu	360	4.99	63.64	5.77	24.50	13.79	0.8Y	6.27	3.82
	Fe	360	6.04	40.75	1.47	2.53	42.35	5.4YR	3.96	0.48
5 Gardenia seeds	None	360	1.00	84.33	0.52	24.82	17.06	4.7Y	8.39	3.41
	Al	360	0.73	85.72	−0.88	20.00	5.21	5.8Y	8.51	2.59
	Cu	360	0.96	83.31	−1.75	20.55	4.94	7.3Y	8.26	2.66
	Fe	360	1.73	76.05	4.17	19.93	10.28	1.0Y	7.52	3.04
6 Pagoda tree	None	360	1.23	85.40	−0.33	13.29	5.50	3.9Y	8.45	1.64
	Al	360	0.95	84.17	−2.25	27.41	14.30	6.6Y	8.36	3.59
	Cu	360	1.11	79.34	0.40	27.06	15.06	4.4Y	7.87	3.77
	Fe	360	2.30	67.81	3.69	18.33	18.74	1.2Y	6.67	2.77
7 Clove tree	None	360	1.40	85.45	−0.28	14.44	6.61	4.2Y	8.46	1.80
	Al	360	1.12	85.34	−1.57	16.57	2.49	6.0Y	8.45	2.01
	Cu	360	1.45	80.84	−0.60	19.69	7.00	4.9Y	8.00	2.59
	Fe	360	2.55	67.48	5.09	15.32	18.78	9.1YR	6.63	2.53
8 Amur cork-tree	None	360	1.91	86.38	−4.31	22.18	14.79	9.8Y	8.57	2.75
	Al	360	0.92	86.47	−2.27	14.16	8.28	8.0Y	8.56	1.67
	Cu	360	1.26	84.49	−2.75	15.39	7.22	9.8Y	8.36	1.84
	Fe	360	1.89	78.02	4.17	21.06	11.96	0.9Y	7.72	3.20
9 Chinese ink	None	370	9.44	28.92	0.68	2.57	58.05		2.82	0.42
	Al	370	9.46	28.74	0.62	2.34	0.29		2.81	0.39
	Cu	360	9.54	28.99	0.65	2.61	0.09		2.84	0.42
	Fe	360	10.90	28.49	0.71	3.33	0.87		2.79	0.54
10 Indigo	None	650	6.04	38.30	−2.64	−14.32	53.34	3.7PB	3.67	3.45
	Al	650	6.22	38.09	−3.06	−13.27	1.16	3.1PB	3.65	3.20
	Cu	640	6.62	34.63	−1.52	−8.55	6.93	3.4PB	3.34	1.90
	Fe	630	6.98	33.71	−1.92	−8.99	7.08	3.3PB	3.25	2.01
11 Lac	None	530	2.69	49.74	19.58	1.10	42.54	7.7RP	4.84	4.93
	Al	360	3.34	50.09	22.29	2.31	2.99	8.6RP	4.88	5.52
	Cu	360	3.44	48.38	9.29	1.70	10.40	9.3RP	4.70	2.42
	Fe	360	4.20	45.34	3.81	1.76	16.38	5.1R	4.40	0.92

Table 4. Continued

Type of natural dye	Type of post-mordanting	λmax (nm)	K/S	CIE Lab				Munsell's value		
				L*	a*	b*	△E	H	V	C
12 Violet-root cromwell	None	360	0.96	76.07	4.01	7.84	11.49	5.8YR	7.49	1.57
	Al	360	0.80	75.78	3.83	6.20	1.68	4.1YR	7.45	1.42
	Cu	360	1.09	73.11	0.51	7.24	4.62	1.8YR	7.18	0.99
	Fe	360	1.82	69.97	3.60	12.39	7.62	9.5YR	6.88	1.98
13 Cochineal	None	360	1.05	78.26	5.81	8.82	10.57	3.2YR	7.71	2.15
	Al	360	0.84	77.41	10.83	6.26	5.69	5.8YR	7.62	3.17
	Cu	360	1.36	69.32	1.23	6.70	10.26	1.3Y	6.79	0.98
	Fe	360	2.19	66.64	2.27	10.77	12.30	0.5Y	6.53	1.62
14 Sappan wood	None	360	1.39	76.93	8.63	21.23	18.77	8.2YR	7.62	3.75
	Al	360	1.34	71.23	13.95	10.36	13.38	8.0YR	7.00	3.86
	Cu	360	1.59	68.19	8.90	10.60	13.77	2.2YR	6.69	2.72
	Fe	360	3.13	58.11	3.94	9.15	22.84	7.9YR	5.67	1.62
15 Copy paper	None	360	0.94	91.70	2.78	−12.83	21.59	9.4PB	9.02	3.94
	Al	370	0.93	91.65	2.42	−12.45	0.52	9.1PB	9.02	3.96
	Cu	360	0.90	90.91	−0.37	−8.83	5.15	6.7PB	8.95	3.43
	Fe	360	5.55	73.39	13.04	33.43	50.79	8.3YR	7.29	5.82

and a large increase in a* and b*. In addition, the color difference by Fe mordant was the biggest in all the dyed Hanji, and that of Cu mordant was bigger than Al mordant, except for *Alnus firma* and Amur cork-tree dyed Hanji. However, the color difference of Meok-dyed Hanji was less than 1, and almost no color change was observed for post-mordanting with the three mordants. The color difference for Al mordant of Hanji was 0.32, which was very small, but among the post-mordanted natural dyed Hanji by Al, Hanji with a color difference of 5.0 or higher were Pagoda tree (14.3), Sappan wood (10.4), Amur cork-tree (8.3), Cochineal (5.7), Gardenia (5.2), and *Alnus firma*-dyed Hanji (5.1). Natural dyed Hanji with a color difference of 10.0 or more were ethanol-diluted Ottchil (18.7), *Alnus firma* (13.8), Pagoda tree (15.1), Sappan wood (13.8), Lac (10.4), and Cochineal (10.3) dyed Hanji treated with Cu mordant. According to Fe mordant, the color difference of *Alnus firma* (42.4) dyed Hanji was the biggest, followed by Sappan wood (22.8), ethanol-diluted Ottchil (22.8), Clove tree (18.8), and Pagoda tree (18.7) dyed Hanji. Violet-root cromwell (7.6) and Indigo (7.1)-dyed Hanji were smaller than none-dyed Hanji.

Almost no hue change was observed even when each natural dyed Hanji was post-mordanting with three types of mordant. However, the Fe mordant of *Alnus firma*-dyed Hanji (2.3Y→5.4YR 4.0/0.5), the Fe mordant of Clove tree (4.2Y→9.1YR 6.6/2.5), the Fe mordant of Lac (7.7RP→5.1R 4.4/0.9), and the Cu and the Fe mordants of Cochineal (Cu 3.2YR→1.3Y 6.8/1.0, Fe 3.2YR→0.5Y 6.5/1.6) showed hue changes. Except for Cu mordant of Cochineal, hue change was observed in Fe mordant. It was discolored to almost black by Fe mordant of *Alnus firma* and Lac. Although the color Value differed depending on the mordants, it was similar to that of one before mordant for Al mordant, but in Cu or Fe mordant, most of them decreased and progressed to a rather dark color.

3.2.2. Color fastness of post-mordanted natural dyed Hanji

Fig. 4 shows the color fastness of Hanji dyed with natural dyes after post-mordanting with three mordants and exposure to 72 hr. of UV irradiation. There was slight

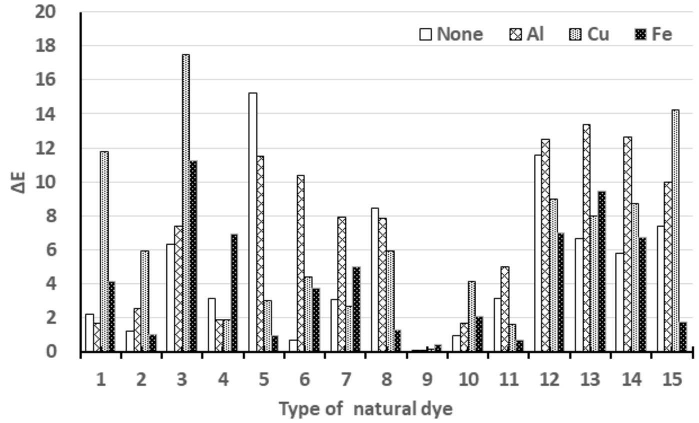

Fig. 4. Color difference of post mordanting natural dyed Hanjis after 72 hr. UV radiation.

change in hue after 72 hr. of UV irradiation on post-mordanting Hanji base paper (Control). Al post-mordanting Hanji demonstrated a smaller color difference than Hanji itself, but the one Cu post-mordanted showed a significantly big color difference of 11.8.

Meok-dyed Hanji showed color difference of less than 1.0 even after 72 hr. of UV irradiation for all three post-mordants. Among the 12 types of natural dyed Hanji excluding Meok-dyed Hanji, those with a color difference of less than 1.0 after 72 hr. of UV irradiation on Hanji without post-mordanting were *Alnus firma* and Indigo-dyed Hanji with excellent color fastness. However, there were cases where the color difference of dyed Hanji between after post-mordanting and after 72 hr. UV irradiation was smaller than that between before post-mordanting and after 72 hr. UV irradiation. Fe mordant on ethanol-diluted Ottchil-dyed Hanji (1.2→1.0), Al, Cu mordants on *Alnus firma*-dyed Hanji (3.2→1.9), Al, Cu, and Fe mordants on Gardenia-dyed Hanji (15.2→11.5, 3.0, 1.0), Cu mordant on Clove tree-dyed Hanji (3.1→2.7), Al, Cu, and Fe mordants on Amur cork-tree-dyed Hanji (8.4→7.9, 5.9, 1.3), Cu and Fe mordants on Lac-dyed Hanji (3.1→1.6, 0.7), Cu and Fe mordants on Violet-root cromwell dyed Hanji (11.6→9.0, 7.0), and Al, Cu, and Fe mordants on Gardenia-dyed Hanji (15.2→11.5, 3.0, 1.0) drove improvement in color fastness of them. Especially, Gardenia-dyed Hanji, which has a tendency to discolor easily, might be prevented the discoloration by Fe post-mordanting.

Post-mordanting turpentine-diluted Ottchil Hanji had a hue change by Fe post-mordanting (0.2Y→9.9YR). However, even after 72 hr. of UV irradiation, all three types of post-mordants showed a relatively small color difference. In particular, the color difference of Fe post-mordanting Hanji was very small at 1.0 despite the change in hue, exhibiting excellent color fastness. Post-mordanting ethanol-diluted Ottchil staining Hanji showed a big color difference after 72 hr. of UV irradiation for all three types of post-mordants, indicating low color fastness. Hue of *Alnus firma*-dyed Hanji was changed from 5.4YR to black due to Fe mordant, and the color difference increased. For Al and Cu mordants, color fastness improved as the color difference decreased after 72 hr. of UV irradiation. The Gardenia-dyed Hanji showed very poor color fastness, but it was greatly improved by the post-mordanting as in Gardenia-dyed Hanji, which showed superior color fastness. The Pagoda-tree dyed Hanji without post-mordanting treatment showed very good color fastness, but when post-mordant treatment was applied, the color difference of all three types of mordants increased significantly, especially Al post-mordanting. Post-mordanting Clove tree-dyed Hanji showed almost the same color difference as Clove tree-dyed Hanji before mordants treatment in Cu and Fe mordants even after 72 hr. of UV irradiation, but Al mordant was significantly increased. Post-mordanting Amur cork-tree-dyed Hanji showed a smaller color difference than that of Amur cork-tree-dyed Hanji before mordant treatment in all three types of mordants even after 72 hr. of UV irradiation. The hue also changed from 0.9Y to 9.8YR by Fe mordant. The color fastness of Indigo-dyed Hanji before mordanting was very good, but the color difference slightly increased due to post-mordanting. In Lac-dyed Hanji, the color difference decreased by Cu and Fe post-mordanting, while it increased in Al post-mordanting. The color fastness observed in Fe post-mordanting Lac dyeing Hanji was superior. In the case of Lac-dyed Hanji's Cu post-mordanting, the hue changed from 9.3RP to 1.3R. The hue of Violet-root cromwell dyed Hanji was changed from 4.1YR to 3.5Y due to Al post-mordanting, and although color fastness was poor overall, it was slightly improved with Fe and Cu post-mordanting. Both Cochineal and Sappan wood-dyed Hanji showed a color difference of 6.7, which was increased by the post-mordanting, especially for Al post- mordanting.

As shown above, the color and color fastness were significantly different depending on the type of natural dye and the presence or the absence of post-mordant treatment. Therefore, when dyeing Hanji with natural dyes, it is recommended to dye with sufficient knowledge and information about the desired color and fastness.

4. CONCLUSION

The results of comparing and examining the color characteristics and changes in color and color fastness after post-mordanting on 13 types of natural dyed Hanji, which were big as it was manufactured manually without cutting and were dyed with 11 types of plant natural dyes and two types of animal natural dyes are as follow:

1) Before dyeing, the hue of Hanji was 4.3Y, with considerably high L* and V values, indicating a yellowish color that was close to white.
2) Meok, Indigo, and Lac showed high dyeing yield and color difference among 13 natural dyeing materials, and Violet-root cromwell and Gardenia seeds showed low dyeing yield (less than 1).
3) Hanji dyed with two types of Ottchils, *Alnus firma* fruit, Gardenia seeds, Pagoda tree seeds, and Clove tree displayed a yellowish color (H) of 1.2–4.7Y while Amur cork-tree bark expressed a yellow color close to green with 9.8Y.
4) Meok-dyed Hanji showed considerably high dyeing yield and color difference, which was black, and had a V of 2.82 and a C of 0.42.
5) Indigo-dyed Hanji showed a big color difference and displayed a blue-ish hue with 3.7PB. The hue of Hanji dyed with Lac, a natural animal dye, was purple at 7.7RP. The hues of Hanji dyed with the plant Violet-root cromwell, Sappan wood, and animal Cochineal were 5.8YR, 3.2YR, and 8.2YR, respectively, and all displayed an orange-ish color.
6) The color difference after exposure to 72 hr. of UV irradiation was less than that of the undyed Hanji with excellent color fastness with turpentine-diluted Ottchil, Pagoda, Meok, and Indigo-dyed Hanji. Gardenia and Violet-root cromwell dyed Hanji showed substantially poor color fastness.
7) The color difference of natural dyed Hanji after post-mordanting by Al, Cu, and Fe mordants was diverse. Overall, the color difference due to Fe mordant was the biggest, and Cu mordant was bigger than Al mordant, except for *Alnus firma* and Amur cork tree-dyed Hanji. Changes in hue are as follows: *Alnus firma* Fe mordant (2.3Y→5.4YR), Clove tree Fe mordant (4.2Y→9.1YR), Lac's Fe mordant (7.7RP→5.1R), and Cochineal's Cu and Fe mordants (Cu 3.2YR →1.3Y, Fe 3.2YR→0.5Y). However, the color difference of

Meok-dyed Hanji was less than 1, and almost no color change was observed as the post-mordanting by the three mordant dyes.

8) There was no color change observed for post-mordanting Hanji base paper (Control) after exposure to 72 hr. of UV irradiation, and while Al post-mordanting Hanji base paper had a smaller color difference than the Hanji base paper itself, Cu post-mordanting showed a considerable color difference with 11.8. Meok-dyed Hanji showed excellent fastness with less than 1.0 color difference even after exposure to 72 hr. of UV irradiation for all three types of post-mordanting.
9) Fe mordant on ethanol-diluted Ottchil-dyed Hanji (1.2→1.0), Al, Cu mordants on *Alnus firma*-dyed Hanji (3.2→1.9), Al, Cu, and Fe mordants on Gardenia-dyed Hanji (15.2→11.5, 3.0, 1.0), Cu mordant on Clove tree-dyed Hanji (3.1→2.7), Al, Cu, and Fe mordant on Amur cork-tree-dyed Hanji (8.4→7.9, 5.9, 1.3), Cu and Fe mordant on Lac-dyed Hanji (3.1→1.6, 0.7), Cu and Fe mordants on Violet-root cromwell dyed Hanji (11.6→9.0, 7.0), and Al, Cu, and Fe mordants on Gardenia-dyed Hanji (15.2→11.5, 3.0, 1.0) drove improvement in color fastness of them. As a result, color fastness was improved by post-mordanting, especially, Gardenia-dyed Hanji having tendency to discolor easily, was almost prevented by Fe post-mordanting.

CONFLICT of INTEREST

No potential conflict of interest relevant to this article was reported.

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