Mordants Application and Data Establishment for Natural Dye Standardization and Accuarcy

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천연염색 표준화와 정확성을 위한 매염제의 적용 및 데이터 확립

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Abstract Natural dyeing has traditionally been used in many countries around the world, and as natural dyes are diversified, the diversity of dyeing patterns is also expanding. This study tried to establish standardization by providing numerical values that could provide quantified information to the Internet of Things by more accurately analyzing the color changes of dyes and mordants for the four natural dyes. The addition of copper acetate, iron II sulfate and potassium dichromate to the dye extracted from *Juglans regia* Linn changed the original color of brown to other colors of purple, khaki and dark brown, respectively. Except for potassium dichromate added to *Sophora japonica* L. or *Phellodendron amurense Ruprecht*, the concentration of other mordants was reduced, but the color difference of the dyed silk was very large. However, although there is a difference in degree, copper acetate and iron sulfate induced color changes of 35% and 15%, respectively. In summary, it was confirmed that the highest color change was induced when 15 grams of copper acetate was added to *J. regia* Linn, *S. japonica* L. and *P. amurense Ruprecht* and 150 grams of iron to *Phytolacca americana*. The results of this study suggested that the accurate color change by various mordants can be utilized as important information that enables more accurate color induction by dyes and mordants.

Key Words : IoT, Color change, Mordant, Natural dyeing

요 약 천연염색은 전통적으로 세계 여러 나라에서 사용되어 왔으며, 천연염색이 다양해짐에 따라 염색 패턴의 다양성도 확대되고 있다. 본 연구는 4가지 천연염료에 대한 염료와 매염제의 색상 변화를 보다 정확하게 분석하여 사물인터넷에 정량화된 정보를 제공할 수 있는 수치를 제공함으로써 표준화를 마련하고자 했다. *Juglans regia* Linn에서 추출한 염료에 구리 아세테이트, 철 II 황산염 및 중크롬산 칼륨을 첨가하면 갈색의 원래 색상이 각각 보라색, 카키색 및 암갈색의 다른 색상으로 변경되었다. *Sophora japonica* L. 또는 *Phellodendron amurense Ruprecht*에 첨가된 중크롬산 칼륨을 제외하고는 다른 매염제의 농도는 감소되었지만 염색된 실크의 색상 차이는 매우 컸다. 그러나 정도의 차이는 있지만 초산구리와 황산철은 각각 35%와 15%의 색변화를 유도하였다. 종합해볼때, *J. regia Linn, S. japonica L.*과 *P. amurense Ruprecht*에는 copper acetate 15 gram을 *Phytolacca americana*에는 150 gram의 iron이 정확히 첨가된 것이 가장 높은 색상변화를 유도할 수 있다고 확인되었다. 본 연구의 결과는 다양한 매염제에 의한 정확한 색상 변화가 염료 및 매염제에 의한 보다 정확한 색상 유도를 가능하게 하는 중요한 정보로 활용될 수 있음을 시사하였다.

주제어 : 사물인터넷, 색변화, 매염제, 천연염색

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1. Introduction

Natural dyeing has traditionally been used in many countries around the world, and as natural dyes are diversified, the diversity of dyeing patterns is also expanding. Although many such various information and results are available, the exact dosage and methods of mordant for dyes are not well formulated. The term natural dyes refer to plants, insects and can be extracted from animals and can be used indirectly or with mordants [1]. According to previous studies, natural dyes have been applied to silk fibers and used to derive soft and glossy colors. One of the biggest advantages of natural dyes is that they can be used as good materials to minimize the damage caused by the use of synthetic dyes, for example, to avoid the possibility of allergies [2].

In addition, cases of applying natural dyeing and natural dyeing results to the Internet of Things (IoT) are still rare in the world [3]. In the IoT environment, since everything connected to the network, such as people, objects, and spaces (objects) [4], can generate data in real time, it will be possible to use the information on dyes and mordants in this study to combine it with IoT. In this study, I tried to obtain accurate information that can be applied to IoT by quantifying silk dyeing patterns for four dyes, which the researcher has previously used as natural dyes, e.g., green husk of *Juglans regia* Linn, *Phytolacca americana* L.-berries, *Sophora japonica* L. *Phellodendron amurense Ruprecht*. Briefly, J. regia bark and leaf extracts are mainly used to verify biological effects [5]. It has been used in *Mycobacterium tuberculosis* for the biological effect [6] and its wood has also been used in luxury flooring, guitars, furniture, and handles. Although *P. americana* L. is rarely used as a natural dye, it was first applied in our previous experiments [7], but it is a plant that has traditionally been widely used in Korea as a diuretic [8]. *S. japonica* L. is a tree species belonging to the subfamily Faboideae of the pea family [9] and its leaves are pinnate, and there are 9-21 leaflets, and the flowers form a raceme with double layers similar to that of a black grasshopper [10]. In particular, it can be used as a material for building and crafting sculptures [11]. *P. amurense Ruprecht*, commonly called Amur cork tree, has been used as an analgesic in oriental medicine, and is mainly known as a material for biological evaluation [12]. In order to use natural dyes, mordants are generally used [13]. The mordant plays a role in adsorbing well to the fiber by forming a complex with the natural dye [13]. Moreover, there is an advantage that a wide variety of colors can be induced by the mordant.

However, the exact method for color induction by mordant is not well established. Depending on how or at what concentration the mordant is actually treated, a wide variety of colors can be induced. Therefore, in this study, the color change in silk dyeing was quantified by applying the treatment time and concentration of the mordant. It is thought that this numerical value can be a data for standardization of natural dye-mordant-color change. As such, the higher the utilization of the mordant that induces various colors, the higher the utilization of natural dyes will be. This study aimed to provide data that can provide quantified information to IoT by more accurately analyzing the color changes of dyes and mordants for four natural dyes.

2. Analysis of Staining Patterns

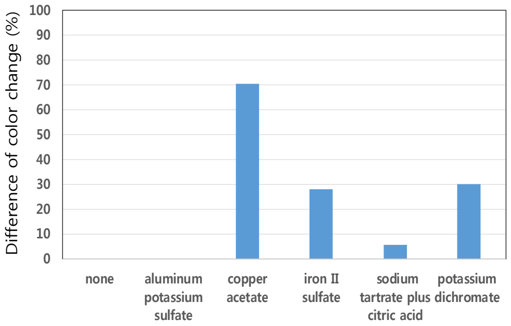
2.1 Numerical value of silk-staining by *J. regia* Linn

Generally, the mordants, e.g., copper acetate, aluminum potassium sulfate, sodium tartrate plus citric acid, iron II sulfate and potassium dichromate were applied to silk fabric [14]. The natural dye of *J. regia* Linn was extracted after boiling 500 g of green peel by adding 900 ml of water except for the mordant of sodium tartrate plus citric acid with 800 ml, and the pH of the added water was maintained at 5.5. The table 1 below described the colors of silk fibers dyed using different mordants and *J. regia* Linn-extracted dye [14]. Aluminum potassium sulfate mordant induced little color change. However, even sodium tartrate and citric acid showed little color change, while the dyed silk was found to be more lustrous than the mordant mordant alone or aluminum potassium sulfate. Interestingly, copper acetate, iron II sulfate and potassium dichromate altered the original color of brown to another color of purple, khaki and dark brown, respectively (Table 1).

〈Table 1〉 Effect of mordants and *J. regia* Linn-extracted dye to silk fabric [14].

Mordants	Volume of mordants (gram)	Volume of distilled water (ml)	Color of silk fabric
none	0	900	brown
copper acetate	15	900	purple
aluminum potassium sulfate	15	900	brown
sodium tartrate plus citric acid	160	800	brown
iron (II) sulfate	150	900	khaki
potassium dichromate	15	900	brown

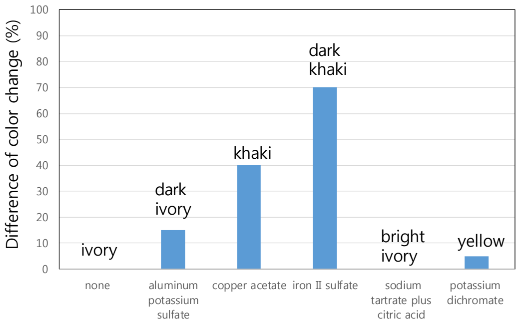
Therefore, the aluminum potassium sulfate and a negative control without any mordant could be expressed as a value of 0. The color change of silk dyeing by the other four types of mordants above and *J. regia* Linn-extracted dye was explained by setting a as 0 (Fig. 1). By adding copper acetate to *J. regia* Linn-extracted dye, the color change was the highest. Therefore, it suggested that a standardization could be established to add 15 gram of copper acetate to 900 ml of water.



[Fig. 1] Color changes by mordants and *J. regia* Linn-extracted dye. Five mordants were applied to silk fabric. None indicated a negative control of the dye alone without any mordant.

2.2 Numerical value of silk-staining by *P. americana* L.-berries

The concentration of mordant added to the dye may affect the degree of dyeing. The different gram numbers of mordants treated in the aforementioned *J. regia* Linn-extracted dye were actually treated for fear of chemical toxicity. Mordants for dyes extracted from *P. americana* L.-berries were treated in same amounts to *J. regia* Linn staining [15]. These results could explain more staining information, that is, the degree of staining. Silk fibers were stained ivory by a negative control that was not treated with a mordant (Fig. 2). Sodium tartrate plus citric acid was very similar to the negative control, but a slightly lighter ivory color appeared in the silk fibers. In addition, a color similar to ivory but close to yellow was dyed on silk fibers by potassium dichromate. On the other hand, the most different staining was caused by copper acetate and iron II sulfate, which induced khaki and dark khaki, respectively. When comparing the staining patterns by *J. regia* Linn, although there was a slight difference in the degree of staining by aluminum potassium sulfate, staining results very similar to those of the negative control were derived.

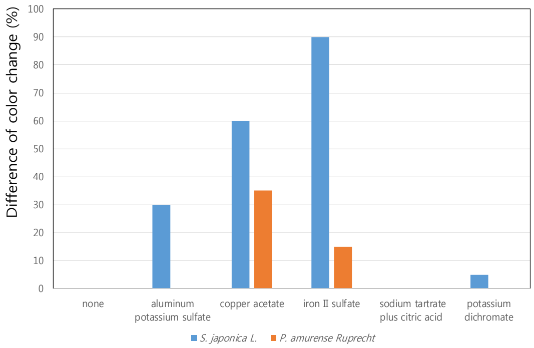


[Fig. 2] Color changes by mordants and *P. americana* L.-berries-extracted dye. Five mordants were applied to silk fabric. None indicated a negative control of the dye alone without any mordant.

The dye was extracted from the husk of *J. regia* Linn, and the dye was extracted from the berries of *P. americana*. Although the initial color of the dye appeared in the initial dye extraction process was different, the color change by the mordant was obvious. The addition of iron II sulfate to *P. americana* L.-berries could induce the highest color change. However, 150 gram iron, which was 10 times higher than copper acetate added to *J. regia* Linn, was added. If it was treated with the same amount of copper acetate, it was assumed that the change in dyeing would have been reduced to some extent.

2.3 Numerical value of silk-staining by *S. japonica* L. and *P. amurense* Ruprecht

Both *S. japonica* L. and *P. amurense Ruprecht* were dyes extracted from hard wood material [15]. For these two ingredients, about 600 ml of water was added to the previously mentioned *J. regia* Linn and *P. americana* L. with a little less water. In particular, the amount of mordants added to *S. japonica* was treated with a different concentration than the other three dyes, and 10 grams of copper acetate, aluminum potassium sulfate and iron II sulfate were added. On the other hand, sodium tartrate plus citric acid was added in a rather small amount of 120 grams, and potassium dichromate was added in a slightly higher amount of 20 grams. This slight change was to analyze the effect of the mordant concentration as well. Since *S. japonica* L. and *P. amurense Ruprecht* induced a yellowish color, which was almost the same color, when no mordant treatment was applied, this result was intended to compare the two. The effect of the mordants treated on *S. japonica* L. induced a very pronounced color change (Fig. 3). Except for potassium dichromate, although the concentration of the other mordants was reduced, the difference in the color of the dyed silk was very large. The effect of aluminum potassium sulfate showed a distinct color difference in the staining by *S. japonica* L., although it was almost colorless in the negative control, but when treated with aluminum potassium sulfate, the silk fibers were dyed yellow. This yellow color also showed a difference from the staining by sodium tartrate plus citric acid, which was almost colorless. The biggest difference was staining by iron II sulfate, which induced a dark brown color that was completely different from that of the negative control. Staining by *P. amurense Ruprecht* hardly induced color change when no mordant was



[Fig. 3] Color changes by mordants and *S. japonica* L. or *P. amurense* Ruprecht-extracted dye. Five mordants were applied to silk fabric. None indicated a negative control of the dye alone without any mordant.

treated and when treated with aluminum potassium sulfate, sodium tartrate plus citric acid, and potassium dichromate. However, although there was a difference in degree, copper acetate and iron II sulfate induced a color change of 35% and 15%, respectively (Fig. 3).

Taken together, the smallest changes in staining caused by mordants were aluminum potassium sulfate and sodium tartrate plus citric acid. The most interesting fact was the fact that the staining change by iron II sulfate mordant was the greatest. When copper acetate was added to *S. japonica* L. and *P. amurense Ruprecht*, the staining change was not higher than that of iron II sulfate. However, considering the amount of 150 grams, it was confirmed that 15 gram of copper acetate was the most suitable amount of mordant for *S. japonica* L. and *P. amurense Ruprecht* dyeing.

3. Discussion

Extraction and utilization of natural dyes may be more stable in humans or animals than chemical dyes [1, 3, 16-20]. This extracted component can be used for staining and evaluation of biological usefulness by filtration with a 0.45 μm-sized syringe filter.

In this study, it was attempted to express with a relatively accurate numerical value how the color change induced by the various mordants used in combination with the four natural dyes that this researcher studied previously.

Mordants are classified into components including metal salts, alkalis, and acids [21]. Aluminum, iron, chromium, copper, and tin are widely used as metal components. Alkali-containing mordants include potassium carbonate and calcium hydroxide, and acid-containing mordants include acetic acid and citric acid. In this study, most interestingly, the change in staining was greatest when iron II sulfate was treated. For this reason, it was confirmed that the iron component had a significant effect on the staining change. As shown in the previous study results, it was confirmed that the degree of staining by iron (II) sulfate mordant was different than that of the negative control by 75% [3]. On the other hand, sodium tartrate plus citric acid did not induce any color change when compared to the negative control. Compared with the negative control, the aluminum potassium sulfate mordant showed a slightly more color change (about 15%), which was thought to be because it was a mordant containing aluminum and tended to slightly brighten the color.

In addition to the mordant, in order to extract the essential dye for the dyeing process, it is generally necessary to add water and boil it. All four natural dyes mentioned above were boiled for 30 minutes by adding water. Of course, if it was boiled for a longer period of time, it was thought that the extraction of a more concentrated dye was possible, but the time of 30 minutes was found to be sufficient time as shown by the staining results by negative controls. In addition, the pH of the solvent was added while maintaining it from 5.5 to 7.0. Although there are still few clear reports, it was thought that the color of the extracted dye would change if the change in pH was increased. However, sodium tartrate plus citric acid used in this study was a mordant containing acid, and it was assumed that the effect of the mordant could be different depending on the pH of the solvent.

By expressing the color change of dyeing numerically, an environment could be prepared in which the set-up of an accurate protocol that can be grafted to the IoT network could perform the dyeing more accurately [17]. According to the most recent research report, real-time dyeing of textiles was applied using a dyeing method and IoT-based application cases [22]. The operator was able to accurately identify the end point of dyeing and maintain the dyeing conditions constant. In addition, a module that measured dyeing parameters such as dye absorption rate, pH concentration, and color change of dyed fibers over time in real time and monitor software changes was produced. Taken together, it was evaluated that it was most important to quantify the results showing the highest change in dyeing according to the concentration of mordant, pH, and treatment time. Therefore, it was confirmed that 15 grams of copper acetate in *J. regia* Linn, *S. japonica* L. and *P. amurense Ruprecht* and 150 grams of iron in *P. americana* were precisely added to induce the highest color change.

Therefore, it was thought that the results of this study would be used as important information to enable the accurate color change by various mordants to induce more accurate color by dyes and mordants in building the IoT platform.

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