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Construction of Dyeing Condition System for *Lithospermum erythrorhizon* by Applying Natural Dye and Mordants

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천연 염료와 매염제의 응용에 의한 *Lithospermum erythrorhizon*의 염색 조건 시스템 구축

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Abstract It was reported that a mobile application was designed to easily provide natural dyeing information such as natural dye related resources, colors and dyed fabrics in 2007. Since studies on the linkage, application, etc. between natural dye dyeing and IoT are still lacking, diversity of information on the change of dyeing pattern by natural dye dyeing is required. In this study, it was to construct dyeing information by natural dyes, e.g., *Lithospermum erythrorhizon,* on silk, which has been traditionally used as many fibers in Korea. The extraction of the dye from *L. erythrorhizon* was carried out under pH4. The dried root of *L. erythrorhizon* showed dark brownish purple. Silk fabric by a without a mordant typically showed a purple dyed pattern. In the staining by sodium tartrate plus citric acid, silk fabric was stained clear brown. Interestingly, the mordant of iron (II) sulfate, the silk fabric was dyed in a light gray color rather than black. When the mordant of aluminum potassium sulfate was treated with *L. erythrorhizon*-extracted dye, the results were almost the same as when the mordant was not treated. When the degree of dyeing was evaluated numerically, the treatment of the mordant of potassium dichromate was about 50% darker, and the dyeing by iron (II) sulfate was about 75% darker. These results will be helpful in the study of applying various dye colors using *L. erythrorhizon*, and it will provide information on dyeing controller and database system construction by dyeing parameters such as dyeing degree, pH concentration, and chromaticity change.

Key Words : *Lithospermum erythrorhizon*, Dye, Mordant, Silk, IoT

요 약 2007년에 천연 염료 관련 자원, 색상, 염색 원단 등 천연 염색 정보를 쉽게 제공 할 수 있도록 모바일 어플리케이션을 설계 한 것으로 알려졌다. 천연 염색과 IoT의 연관성, 응용 등에 대한 연구가 아직 부족한만큼 다양성 천연 염색에 의한 염색 패턴 변화에 관한 정보가 필요하다. 본 연구에서는, 천연 염료, 예를 들어 *Lithospermum erythrorhizon*가 한국에서 전통적으로 많이 사용되어온 실크에 대하여 염색 정보를 구축하고자 했다. *L. erythrorhizon*에서 염료의 추출은 pH4에서 수행되었다. *L. erythrorhizon*의 건조 된 뿌리는 짙은 갈색을 띤 자주색을 보였다. 매염제가 없는 실크 직물은 일반적으로 보라색 염색 패턴을 보였다. 타르타르산 나트륨과 구연산으로 염색 한 실크 직물은 투명한 갈색으로 염색되었다. 흥미롭게도 황산 철 (II)의 매염제 인 실크 직물은 검은 색이 아닌 밝은 회색으로 염색되었다. 알루미늄 포타슘 설페이트의 매염제를 *L. erythrorhizon* 추출 염료로 처리 한 결과 매염제를 처리하지 않았을 때와 거의 동일했다. 염색도를 수치로 평가 한 결과, 중크롬산 칼륨의 매염제의 처리는 약 50% 어둡고, 황산 철 (II)에 의한 염색은 약 75% 어두웠다. 이러한 결과는 *L. erythrorhizon*을 이용한 다양한 염색 색 적용 연구에 도움이되며, 염색 정도와 pH 농도, 색도변화라는 염색변수에 의한 염색 컨트롤러와 데이터베이스 시스템 구축에관한 정보를 제공 할 것이다.

주제어 : *Lithospermum erythrorhizon*, 염료, 매염제, 실크, 사물인터넷

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1. Introduction

There is few case in the world that has applied information on natural staining and natural dyes to IoT(Internet of things) yet. However, there was one report about IoT to provide natural dyeing information service in 2017 [1]. It was designed a mobile application to easily provide natural dyeing information such as natural dye related resources, colors and dyed fabrics. Since studies on the linkage, application, etc. between natural dye dyeing and IoT are still lacking, diversity of information on the change of dyeing pattern by natural dye dyeing is required. In fact, on the Internet search site, descriptions about dyeing with natural dyes are simplified. However, if more studies on the effect of various mordants on the dyeing pattern are added, it will be an opportunity to be applied to IoT having more information. The IoT environment, people connected to the network, object, everything such as space (Things) generates data in real time [2]. In this study, it was to construct dyeing information by natural dyes on silk, which has been traditionally used as many fibers in Korea. One of the key elements of this study was to analyze the pattern of dyeing change by treating various mordants and quantitatively measure the change value of the color. In connection with these studies, the data on which the research team previously confirmed the pattern of dyeing change by mixing various dyes and mordants were the basis.

L. erythrorhizon, called redroot lithospermum of red pigment, is a plant that grows mainly in far-east Asian countries and has been applied as a natural color to various fields such as food and textiles [3, 4]. *L. erythrorhizon* is a Chinese medicine that has various antiviral effect to human immunodeficiency virus type 1 (HIV-1) [5] and biological activities of inflammation, wounds, etc [6].

The main ingredients of the root contain acetylshikonin, shikonin, alkannan, isobutyryl-shikonin, β-dimethyacrylshikonin, β-hydroxy isovaloryl shikonin, teracrylshikonin which are various shikonin and ester derivatives [7, 8].

In Korea, it is called Jacho, because the root of *L. erythrorhizon* is purple. In particular, it is widely used as natural purple dye and is also used to make red wine [3, 9]. The main pigment component of *L. erythrorhizon*, shikonin, is enantiomer to alkannin, the main pigment component of *Alkanna tinctoria* belonging to its same family.

A mordant is a material used to bind the dye onto the fabric by forming a coordination complex with the dye, which is attached to the fabric [10]. In addition, the mordant can induce color changes in the dye. For example, aluminum potassium sulfate made silk fabric purple to dark blue by the use of *Vitis coignetiae* Pulliat-extracted dye [11].

In this study, silk dyeing was observed using the dye extracted from *L. erythrorhizon*, and changes in silk dyeing by various mordants were observed. These results will contribute not only to the evaluation of a mordant that can induce more various color changes, but also to the evaluation of the commercialization potential of a dye. In particular, this study will provide information on dyeing controller and database system construction by dyeing parameters such as dyeing degree, pH concentration, and chromaticity change.

2. Materials and Methods

2.1 Dye extraction for silk dyeing from *L. erythrorhizon*

The extraction of the dye from *L. erythrorhizon* was carried out under acidic conditions, e.g., pH4. The results of silk dyeing in neutral or alkaline conditions were unsuitable for visual observation (data not shown). Briefly, *L. erythrorhizon* was purchased from a traditional market, foreign substances were removed from running tap water, washed three times with distilled water (DW), and then dried completely in air. Four hundred grams of *L. erythrorhizo*n and 2 liters of distilled water were added, boiled for 30 minutes, and then the dye was extracted at room temperature for 2 days for maximal extraction. When a was sufficiently wet with water, it was filtered using a 0.45 ㎛ syringe filter (Sartorius, Goeltingen, Germany) to obtain pure dye. Thus, about 1.5 liters of dye were extracted and prepared for silk dyeing.

2.2 Selection of various mordants and dye application to silk fabric

In this study, the following five types of mordants were used, e.g., copper acetate, aluminum potassium sulfate, sodium tartrate plus citric acid, iron (II) sulfate and potassium dichromate. The volume of the mordant and the treatment time for silk dyeing are described in Table 1. Another reason for the treatment time was to change the treatment time to observe a clearer color change.

〈Table 1〉 Volume of mordants and their treatment time to a silk fabric.

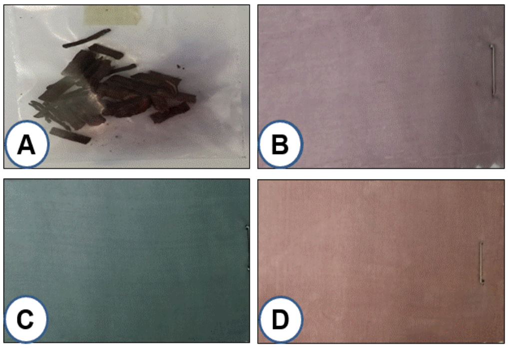
Mordants	Volume of mordants (gram)	Volume of distilled water (ml)	Treatment time (min)
copper acetate	10	600	15
aluminum potassium sulfate	10	600	10
sodium tartrate plus citric acid	30 + 90	600	15
iron (II) sulfate	20	600	15
potassium dichromate	10	600	5

As mentioned above, the prepared dye and mordant were added with the silk fabric, and dyeing proceeded while maintaining 40℃. Dyeing was performed for 30 minutes, hand-wringing the dye uniformly to adsorb on the silk fabric. After checking the degree of dyeing and dyeing was completed, it was washed well with water to remove excess dye, and the silk was slowly dried in the shade without twisting.

3. Results

3.1 Dried root of *L. erythrorhizon* and staining of silk fabric by *L. erythrorhizon*-extracted dye

The silk fabric dyeing by *L. erythrorhizon*-extracted dye was observed, and how the silk fabric dyeing by the mordants of copper acetate and sodium tartrate plus citric acid was changed was also observed. As shown at Fig. 1, the dried root of *L. erythrorhizon* showed dark brownish purple. Silk fabric by a without a mordant typically showed a purple dyed pattern in “None”.



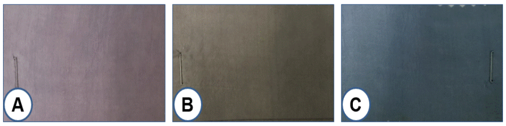
[Fig. 1] Dried root of *L. erythrorhizon* (A) and staining patterns of silk fabric. Staining f silk-fabric was performed by *L. erythrorhizon*-extracted dye (B) and then several mordants of copper acetate (C) sodium tartrate plus citric (C)

On the other hand, when a mordant of copper acetate was treated simultaneously with *L. erythrorhizon*-extracted dye, the silk fabric had a green color different from that of purple. In the staining by sodium tartrate plus citric acid, silk fabric was stained clear brown. These results suggested that the dyeing pattern by copper among the components of the mordant could be very different.

3.2 Color changes by several mordants and *L. erythrorhizon*-extracted dye

Here, it was observed how the silk fabric dyeing pattern by several mordants, e.g., aluminum potassium sulfate, iron (II) sulfate and potassium dichromate changed. The results by the three mordants mentioned here rather than the results of the above dyes mainly led to a dark color.

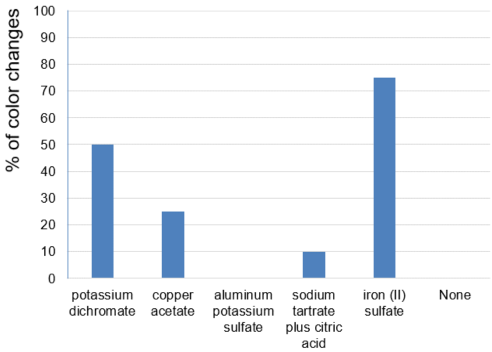
In particular, by the mordant of iron (II) sulfate, the silk fabric was dyed in a light gray color rather than black (Fig. 2a). When the mordant of aluminum potassium sulfate was treated with *L. erythrorhizon*-extracted dye, the results were almost the same as when the mordant was not treated (Fig. 2b). Interestingly, the silk fabric was dyed in a dark blue color completely different from the result of *L. erythrorhizon*-extracted dye dyed purple with the mordant of potassium dichromate (Fig. 2c).



[Fig. 2] Color changes by mordants. Staining of silk fabric was performed as mentioned at Methods, and several mordants of aluminum potassium sulfate (A), iron (B) and potassium dichromate (C)

The degree of dyeing was evaluated numerically in order to more clearly evaluate the silk fabric dyeing pattern visually confirmed (Fig. 3).

The intensity of the color of the silk fabric by the *L. erythrorhizon*-extracted dye and different mordants was expressed as a percentage through the percentage values. Zero percent meant only *L. erythrorhizon*-extracted dye was applied to the silk fabric without a mordant. As mentioned earlier, the most color change was highly caused by the mordant of iron (II) sulfate mordant. The silk fabric dyeing by the mordant of sodium tartrate plus citric acid was about 10% higher than that treated only with *L. erythrorhizon*-extracted dye and aluminum mordant, and about 25% higher when treated with copper acetate. On the other hand, the treatment of the mordant of potassium dichromate was about 50% darker, and the dyeing by iron (II) sulfate was about 75% darker than "None".



[Fig. 3] Numerically evaluated staining colors by *L. erythrorhizon*-extracted dye. In particular, Y bar indicated percentages of color changes or color darkness as compared with the silk staining by *L. erythrorhizon*-extracted dye alone

4. Discussion

In this study, silk fabric dyeing by dye extracted from *L. erythrorhizon* was observed, and dyeing patterns by various mordants were comparatively analyzed. As shown in the results of this study, it was observed that the silk fabric dyeing pattern by the mordant of iron (II) sulfate was very different, and when expressed as a numerical value, there was a difference of about 75% than when only dye was treated.

As mentioned above, the dye was mixed in an acidic solvent, which mainly showed good clarity of dyeing in an acidic solvent. In the previous study, dyeing was performed using a solvent of about pH 4 to pH 6. For example, the dye extracted from Sappan wood induced brown color [12] and another dye from Korean chestnut induced light grey color [12]. For example, certain white materials can be colored during washing, and the colored material can get a new color due to the movement of the dye from the original dyed material known as dyeing [13, 14]. The establishment of the protocol obtained from this method and the results can be said to correspond to an IoT network that can provide an environment for anyone to apply dyeing [15].

It is well known that the use of plants or natural dyes is more environmentally safe and harmless to humans than using chemical dyes [12, 14, 16, 17, 18]. On the other hand, it has been used biochemically and medically by evaluating biological activity by extracting single components from natural substances.

It was known that copper ion was required to biosynthesize shikonin of dye substances of *L. erythrorhizon*, and polyphenol oxidase could play an important role in the shikonin biosynthesis [19]. Although it was somewhat difficult to compare the biological role of copper with the results of this study, copper acetate, used as the mordant of copper acetate, was able to induce a completely different color from dyeing with *L. erythrorhizon* dye by combining with shikonin, the main component of a dye shown at Fig. 1. There was a recent report on real-time measurement of textile dyeing using IoT in dyeing methods and IoT-based application cases [20]. Based on the module analysis value, the operator can accurately identify the timing of drug input and the end of dyeing, and keep the dyeing conditions constant. The research team created an all-in-one module capable of real-time measurement of three major dyeing parameters, such as dyeing absorption rate, pH concentration, and chromaticity change, dyed on fabric over time and monitoring the change in software. In another report on dyeing related to IoT, the future development of the dyeing processing intelligent factory system is based on IoT-based collection and control of the entire production process from fabric receipt to dyeing processing, QC, and delivery, through a manufacturing site operation system and an intelligent prediction system [21]. Meanwhile, in relation to the dyeing industry, the problem that about 15-20% of re-dyeing occurs, meaning the work of re-dying by finding defective products in the fabric being produced [22]. In the results of this study, it is believed that quantitative indicators of dyes, methods, and mordants will provide information that can be applied to IoT systems, a platform that can reduce the occurrence of the above problems.

The result of this study was to understand the characteristics of the main component of the dye of *L. erythrorhizon*, and to analyze the change of the dyeing pattern through silk dyeing. In addition, changes in silk dyeing by various mordants were analyzed. These results will be helpful in the study of applying various dye colors using *L. erythrorhizon*, and in the future, it will be also helpful for researchers trying to understand the biochemical properties of *L. erythrorhizon* dye.

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