[Research Paper - Special Issues]

ISSN 1225-1151 (Print)
ISSN 2234-0793 (Online)
Journal of the Korean Society of Clothing and Textiles
Vol. 48, No. 6 (2024) p.1302~1319
https://doi.org/10.5850/JKSCT.2024.48.6.1302

Sustainable Dyeing of Silk Fabrics with Natural Extract from Seabuckthorn Leaves for UV-Protective and Antimicrobial Textiles

Sarmandakh Badmaanyambuu · Eunjou Yi[*†]

Taesan Industrial Corporation

[*]Department of Fashion and Textiles, Jeju National University/Faculty of Data Science for Sustainable Growth, Jeju National University

Received September 4, 2024; Revised October 10, 2024; Accepted October 28, 2024

Abstract

This study was performed to suggest a novel method of sustainably dyeing textiles with biofunctions for skin health by using natural seabuckthorn leaf extract. Mongolian seabuckthorn leaves were extracted in hot water to create a solid dye powder to be characterized by UV/Vis and FT-IR, and its biological activities were examined. A silk woven fabric was dyed with the seabuckthorn leaf extract under a variety of conditions to identify optimum dyeing conditions along with color fastness. To assess the skin-friendly functions of the dyed fabric, its UV-protective properties and antimicrobial activities were investigated quantitatively. As a result, the fabric showed maximum dye uptake (K/S) under the conditions of 80 °C and 60 min, which was improved under acidic conditions. The dyeing fastness mostly showed good and excellent grades in perspiration, rubbing, and dry cleaning, while it showed a fair grade in fastness to light. The silk dyed at 30% or higher concentrations showed superior UV-protective properties and provided excellent antimicrobial efficacy (99.9% bacterial reduction rate) against both S. aureus and K. pneumoniae. These results could be utilized in designing biofunctional textiles by natural dyeing with seabuckthorn leaf extract.

Key words: Sustainable dyeing, Silk, Seabuckthorn leaves, UV protection, Antimicrobial activity

I. Introduction

With the development of science and technology, the biggest change in the modern human life is the expansion of the range of application of functional materials together with sustainability centered on human comfort as well as environmental circulation. The global textile industry is responsible for having a serious environmental impact across the entire supply chain of clothing and textiles, especially for lots of water and air pollution from dyeing and finishing process (Pizzicato et al., 2023).

Natural dyes, which have come from plant, animal, or mineral sources, are becoming a good substitute for synthetic ones because natural dyes have been revealed as showing various biological efficacy to help improving human skin health as well as they're environmentally friendly and sustainable (Pranta & Rahaman, 2024). Natural dye can be obtained from different sources including plant, animal and minerals and they have been considered as not only safe and reliable, but exhibiting pharmacological activities such as antioxidant, anti-inflammatory, anti-cancer, anti-obesity, anti-microbial, and anti-viral effects. Traditionally, vegetable-originated natural dyes have been more widely obtained from various parts of plants including

[†]Corresponding author
E-mail: ejyi@jejunu.ac.kr

roots, bark, leaves, flowers, and fruits (Verma & Gupta, 2017). And recently leaves from plants have been utilized increasingly owing to their easy availability as by-products or wastes, relatively abundant quantities, and a variety of bio-functional components such as polyphenol, flavonoids, and tannin which could contribute to human skin health. Precisely numerous researches have been performed to introduce natural substances from plant leaves which might be used as dyes for textiles, for instances, in terms of banana leaves (Repon et al., 2016), henna leaves (Rahman Bhuiyan et al., 2018), Ipomoea batatas leaves (Fang et al., 2022), green tea leaves (An, 2016; Ren, 2016; Sukemi et al., 2019), Pomegranate fallen leaves (Haji et al., 2023), and Acorus gramineus Solander leaves (Yang & Yi, 2020) as newly suggested potentials for natural dyeing as well as indigo (Ebrahim et al., 2022; Lo, 2021; Oh & Ahn, 2011; Son, 2023) and Mugwort (Liu & Chen, 2012; Park, 2020; Rather et al., 2020) as in tradition employed in textiles.

Seabuckthorn is a deciduous shrub belonging to the family-Elaeagnacease, which grows in a wide area from Central Asia such as Mongolia to Europe and it contains many valuable nutrients and bioactive substances (Buyukokuroglu & Gulcin, 2009). Although seabuckthorn has been utilized mainly with its berries in folk medicine since ancient, it is nowadays widely utilized even in the area of food, beverage, nutritional supplements, and cosmetics (Bośko et al., 2024). Recently seabuckthorn berries and leaves have been considered to be a rich source of bioactive substances like isoflavones and flavonoids, which have various beneficial effects on health, such as anti-atherogenic, antioxidant, anticancer, and antibacterial effects (Criste et al., 2020). Nevertheless, the leaves of Seabuckthorn, as another abundant resource from seabuckthorn are not fully utilized (He et al., 2023). Moreover there has been little reports about its application into textile fields for dyeing and finishing compared with other plant resources except only a few previous studies. Yogendra Kumar et al. (2016) reported that ethanol extract seabuckthorn could be finished on aramid fabrics mainly consisted of Nomex using pad-dry-cure method finally to provide durable washability and antimictobial activity. Hot water extract of seabuckthorn as a natural dye was applied on cotton and nylon/PU woven fabric to optimize dyeing conditions and to examine antimicrobial activity and sun-protective property of dyed fabrics (Badmaanyambuu, 2022). However there has been rarely reported about other textiles treated or dyed with seabuckthorn leaves extract.

Therefore, in this study, seabuckthorn leaves extract was employed for dyeing a silk woven fabric in order to suggest a novel biofunctional textiles, Chemical characteristics and biological activities of seabuckthorn leaves extract were examined prior to dyeing and the optimum dyeing condition for the silk with seabuckthorn leaves extract was identified along with pH adjustment, color characteristics, and dyeing fastness. Finally biofunctional aspects of the dyed fabrics were evaluated in terms of UV protection properties and antimicrobial activity.

II. Experimental

1. Materials

As for a dye resource, seabuckthorn leaves grown in west Mongolia's central region were picked in early August and they were dried thereafter crushed to be brought to Korea. seabuckthorn leaves of 3.0 kg were soaked in 60 liters of distilled water, and they were three times extracted at 100°C for 3 hours using a low temperature extractor (COSMOS-660, KYUNGSED E&P, Korea). The extracted solution was concentrated to 30% around of the initial amount at 80°C and then it was powderized by a freeze dryer (SFDM 45L, Samwon Freezing Engineering Co., Korea) under the condition of –40°C for 3 days. The final yield rate of the powder was calculated as 14.72% as a solid dye. The fabric sample used for dyeing was 100% silk charmeuse and its specifications are presented in <Table 1>. For pH adjustment, the first grade reagents of citric acid and NaOH were used.

Table 1. Specification of fabric specimen

Fiber	Weave	Weight (g/m^2)	Thickness (mm)	Fabric count (warp×weft/in^2)
Silk 100%	satin	68.2	0.18	231×261

2. Characterization and Biological Assessment of Dye Extract

1) FT-IR Characterization

The FT-IR spectroscopy was employed to get its spectra from the extracted dye using the KBr method by measuring peaks in the range of 650～4000cm^{-1} using an infrared spectrophotometer (FT-IR 4600, JASCO, Japan). This analysis was carried out to assume the components contained in powder dye.

2) UV/visible Spectra Characterization

UV-responses of powder extract from seabuckthorn leaves were examined using an UV/visible spectrophotometer (UV/Vis spectrophotometer, Lambda25, PerkinElmar, USA) and its spectral ranges were given from 200 to 800nm of the wavelength.

3) Total Phenolic Content

The total polyphenolic contents of the hot water extract of seabuckthorn leaves were determined by the Folin-Denis method (Folin & Denis, 1912). The extract solution of 1mL was diluted with 75mL of distilled water and it was mixed with Folin-Denis reagent 0.5㎖ and 35% Sodium carbonate 1㎖ to be stored in the dark for 30 minutes. Tannic acid of 10mg was used as standard and the absorbance for the extracts was measured using a UV/Vis Spectrophotometer (X-ma 2000, Human Corporation, Seoul, Korea) at 760 nm. The experiments were carried out in triplicates and the results were expressed in terms of mg of tannic acid equivalents (TAE).

4) Skin Cell Viability

Natural dyes and pigments can be used with caution as some of natural dyes are also toxic even though most are non toxic. Cell viability testing helps determine if the dye has any toxic effects on living cells, indicating whether it's safe for prolonged contact with human skin as performed in previous works (Bartolini et al., 2024; Räisänen et al., 2020). The skin cell viability of was assessed by MTT (3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyl-tetrazolium Bromide) assay. Immortalized HaCaT (human keratinocyte cell) was incubated in DMEM media with 10% of FBS added with 1% penicillin-streptomycin in 96-well plates at 37°C and in 5% CO^2 atmosphere. Cultured HaCaT cells (1×10^4 cells/well) were plated in 96-well plates for 24 hours and thereafter the extract of seabuckthorn leaves was treated with varied concentrations (0.02, 0.2, 2.0. 20.0, & 200 μg/mL) to each well to be incubated for another 24-hour term. After media was then removed, the plates were washed by PBS (Phosphate butter saline) and 5 mg/mL of MTT solution was added to each well (200 μL/well) to get incubated for 25 min at 37°C and under 5% CO_2. The medium was then collected and the wells were washed with 100 μl DMSO (Dimethyl sulfoxide). Using ELISA (Enzyme Linked Immunosorbent Assay) reader (VERSAMax, Molecular devices, USA), the absorbance was read at a primary wavelength of 570 nm and a reference wavelength of 630 nm.

3. Dyeing Methods

1) Dyeing Conditions

The dyeing of silk was carried out using an IR automatic dyeing machine (Perfect 24, Korea Science Co., Korea) under varied conditions including temperatures from 40 to 80°C, durations from 40 to 80 min, and dye concentrations in the ranges of 10～100% on the weight of the fiber (owf hereinafter). As for pH adjustment, each fabric sample was dyed under the pH levels from 2 to 11 adjusted by a pH meter (Starter 3100 pH Bench, Ohaus, USA) by applying the first-grade reagents of 1M citric acid and NaOH respectively.

2) Dye Uptake Measurement

As for dye uptake, the values of K/S were observed in the absorption wavelength range of 360-740λ using a spectrophotometer (CM-2500d, Minolta, Japan) on the surface of the dyed fabric. The color strength or dye uptake of a dyed fabric is usually expressed by its K/S value by measuring the surface reflectance (R) within the absorption wavelength range. The K/S value at a wavelength of maximum absorbance was calculated by Kubelka-Munk (Eq. 1) for assessing dye uptake depending on dyeing conditions. To identify the dye absorption behavior of seabuckthorn leaves extract on silk overall, the K/S SUM value at all wavelengths across the wavelength (Eq. 2) was calculated considering previous studies (Ferreira et al., 2023; Sarmandakh et al., 2017; Štěpánková et al., 2011; Tang et al., 2019),

$$K/S = (1-R_\lambda)^2/2R_\lambda \quad \cdots\cdots \text{ Eq. 1.}$$

K: Coefficient of absorption
S: Coefficient of scattering
R: Reflected light at wavelength
λ: wavelength of maximum absorbance

$$K/S\ SUM = \sum_{\lambda=360}^{\lambda=700} K/S_{(\lambda)} \quad \cdots\cdots \text{ Eq. 2.}$$

3) Color Measurements

Under D65 light source, the values of L*, a*, b*, C* of CIE at a 10° viewing angle were measured using a spectrophotometer (CM-2500d, Minolta, Japan). They were converted to Munsell notations of H V/C by SpectraMagic NX PC software. To examine color difference values according to ASTM E1347-06(2020) as shown in <Eq. 3>, CIEΔE* was obtained for each dyed sample. According to Practical Color Coordinate System (PCCS), tone was determined for each dyed fabric as well.

$$\triangle E = \sqrt{(\triangle L^*)^2 + (\triangle a^*)^2 + (\triangle b^*)^2} \quad \cdots\cdots \text{ Eq. 3.}$$

4. Color Fastness

The rubbing fastness of dyed silk samples was rated depending on KS K 0650 (Korean Agency for Technology and Standards [KATS], 2017) using the crock-meter in both wet and dry conditions, while the color fastness to perspiration was scored according to KS K ISO 105 E04 (KATS, 2019a) for four hours under 37±2°C. The color fastness to dry cleaning was measured based on KS K ISO 105-D01 (KATS, 2021a), and the color fastness to light was evaluated according to KS K ISO 105-B02 (KATS, 2021b) Xenon arc.

5. UV Protection Properties

The measurement regarding the UV protection properties of silk fabric dyed with hot water extracts from Ecklonia cava was done according to KS K 0850 (KATS, 2019b) using UV transmittance Analyzer (Labsphere Co., USA), with Xenon Arc as the light source, while the UV protection properties was presented in percentages by dividing the light source into UV-A (315 - 400㎚) and UV-B (290 - 315㎚) based on 315 nm UV light. The UV protection rates were calculated according to the <Eq. 4> and <Eq. 5>, as shown below.

$$\text{UV transmittance (\%)} = (T/B) \times 100 \quad \cdots\cdots \text{ Eq. 4.}$$

$$\text{UV protection (\%)} = 100 - \text{UV transmittance (\%)} \quad \cdots\cdots \text{ Eq. 5.}$$

T: UV transmittance through the fabric sample
B: UV transmittance without the fabric

6. Antimicrobial Activity

Klebsiella pneumoniae (MTCC 25922) and *Staphylococcus aureus* (MTCC 25293) as representative of Gram-negative bacteria and Gram-positive bacteria respectively were used for testing antimicrobial efficacy of seabuckthorn-dyed silk fabrics according to a quantitative measurement (KS K 0693:2006). The visible bacteria colonies on the agar plate were counted and the percentage reduction of the bacterium was calculated as follows (Eq. 6);

$$R(\%) = (B-A)/B \times 100\ (\%) \quad \cdots\cdots \text{Eq. 6.}$$

R : The percentage reduction of the bacterium
A : The number of bacterial colonies from test fabric
B : The number of bacterial colonies from control fabric

III. Results

1. Characterization and Biological Efficacy of Seabuckthorn Leaves Extract

1) UV/Visible spectra

The characteristics of UV/Visible spectra of seabuckthorn leaves extract was shown in <Fig. 1>. The maximum wavelengths were seen at both 246nm and 353nm. In general natural flavon and flavonol have been reported to have the maximum absorption wavelength at 220～400nm where those between 220～280 as Band I considered as associated with the presence of a B-ring cinnamoyl system while those between 280～400 as Band II absorption due to the A-ring benzoyl system (Cho, 2004). Kwon et al. (2017) stated that anti-inflammatory effect of seabuckthorn leaves might be caused by some flavonoids. Accordingly, the result showing the maximum wavelengths at both 246nm and 353nm form UV/Visible spectra of seabuckthorn leaves extract in this study confirmed that the extract might have natural flavonoid compounds as the main component which could contribute to biological efficacy dyed fabric with seabuckthorn leaves extract.

2) FT-IR

FT-IR transmission spectra of prepared seabuckthorn leaves extract was presented in <Fig. 2>. They exhibited main peaks at 3246.48cm^{-1}, 2916.56cm^{-1}, 1602.47cm^{-1}, 1493.12cm^{-1}, 1444.61cm^{-1}, 1326.55cm^{-1}, 1186.35cm^{-1}, 1047.36cm^{-1}, 916.331cm^{-1}, 827.025cm^{-1}, and 756.693cm^{-1}. A wide peak shown at 3246.48cm^{-1} indicated the existence of phenolic hydroxyl group (-OH) and a weaker peak centered at 2916.56cm^{-1} seemed to be characterized as C-H bands which are known as frequently contained in organic compounds (Yi & Yoo, 2010). A deeper absorption peak near 1602.47 cm^{-1} was assigned to C=O asymmetric stretching vibration as mentioned previously in a study about seabuckthorn leaves extract (Sharma & Deswal, 2018). Both absoption peaks at 1493.12cm^{-1} and 1444.61cm^{-1} may be caused by C＝C for aromatic groups and the

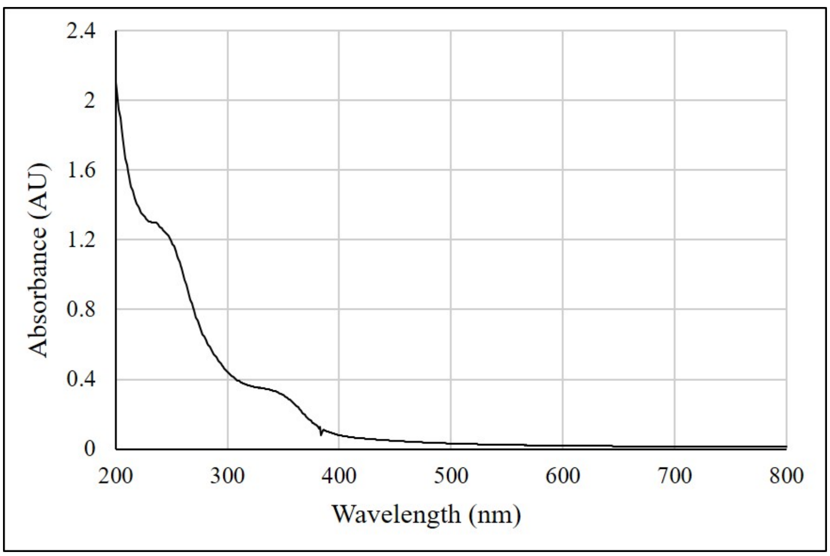


Fig. 1. Ultraviolet-visible spectra of seabuckthorn leaves extract.

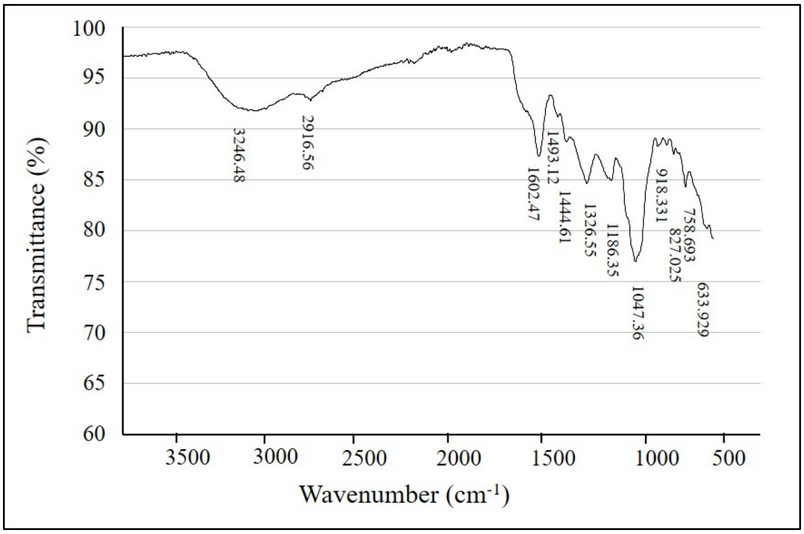


Fig. 2. FT-IR spectra of seabuckthorn leaves extract.

deeper absorption peaks between 1326.55～1047.36 cm^{-1} were attributed to the stretching vibration of the bonds formed between C–O in CO-H or C-O-C, which was supported by a previous work dealing with seabuckthorn leaves extract (Liu et al., 2023). A wide range of weak bands between 918.331～633.929cm^{-1} was thought as corresponding to C–H stretching and bending region of flavonoids rings in seabuckthorn leaves extract. These results indicate that the extract for dyeing silk as a natural in this study might contain some organic compounds like flavonoids and phenol which have been known as consisted of seabuckthorn leaves. Moreover the organic compounds were expected as playing a role as any effective ingredients of the dye contributing to bio-functions of dyed silk.

3) Total Phenolic Content

The amount of total phenolic contents of seabuckthorn leaves extract was determined as 29.08mg/g, which was found as richer in total phenolics than a previous research reporting the contents as 2.47mg/g in fat-soluble green tea extracts (An, 2016) while similar to the quantity in eriobotrya japonica Lindl. of 28.91 mg/g (Jeong et al., 2009). This result leads to a conclusion that the total phenolic contents of seabuckthorn leaves extract could contribute to biological activities of the dyed silk fabrics.

4) Skin cell viability of seabuckthorn leaves extract

Effects of seabuckthorn leaves extract on cell viability of by MTT assay using immortalized HaCaT incubated in DMEM were examined as shown in <Fig. 3>. At 0.02μg/mL of seabuckthorn leaves extract concentration the viability rates of DMSO cell was clearly higher than that of control, which indicates that seabuckthorn leaves extract were not cytoxic at the concentration. From the concentration of 0.2μg/mL to 20μg/mL the cell viability showed slight decrease but still similar to the rate of control. At 200μg/mL of concentration the cell viability rate was obviously reduced showing much lower rate than that of the control. In summary, it seems that extract of seabuckthorn leaves might be dermal cytotoxic at least at much higher concentrations such as at 200 μg/mL. This result implies that seabuckthorn leaves extract would not be toxic or allergic to human skin, which is crucial to confirm the extract's safety prior to dyeing a fabric with it. Furthermore it could leads to the assumption that any dyed fabric with seabuckthorn leaves extract might neither be harmful nor have any potential toxicity.

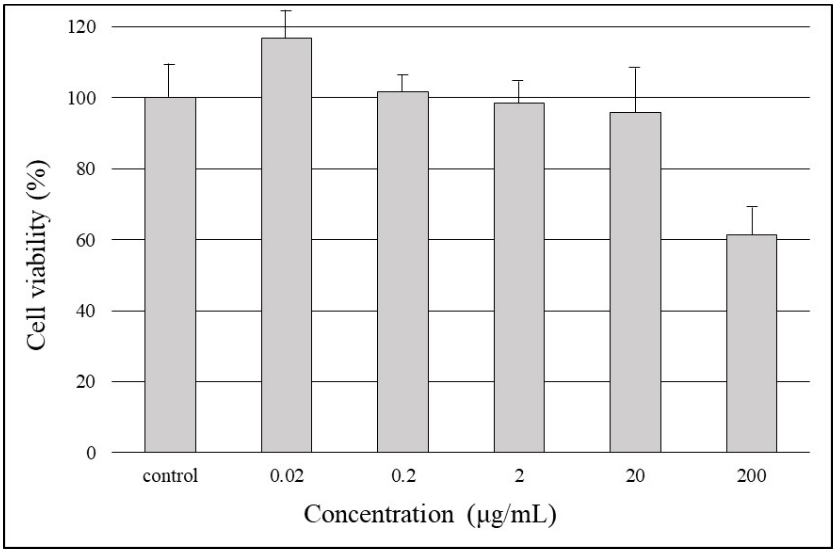


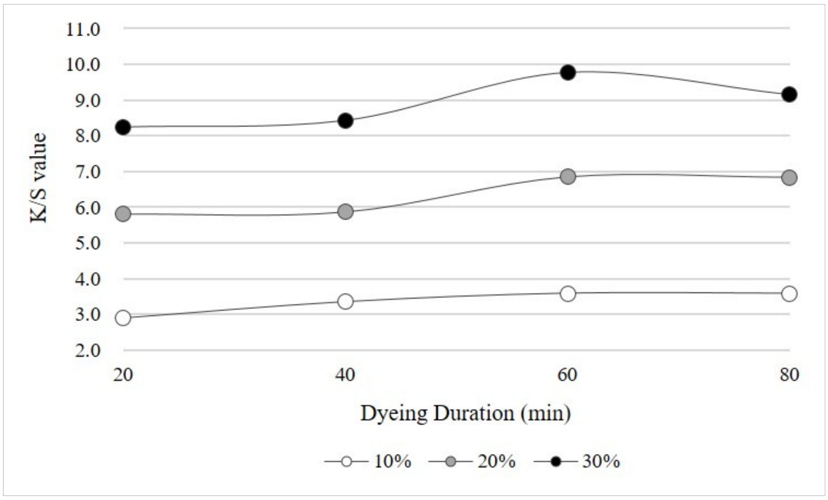
Fig. 3. Effects of seabuckthorn leaves extract on cell viability of DMSO.

2. Optimum Dyeing Conditions and Color Characteristics

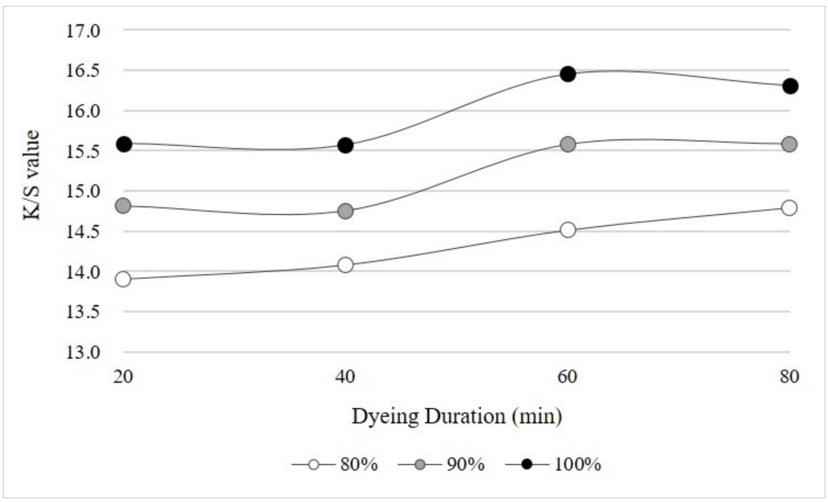
To optimize dyeing conditions of seabuckthorn leaves extract, the silk was dyed under a variety of concentrations of dye bath, time intervals and temperatures. The effects of dyeing durations on K/S at 360λ for both lower levels of 10%, 20%, and 30%, and higher ones of 80%, 90%, and 100% (owf) under the condition of 80°C were presented in <Fig. 4-a> and <Fig. 4-b>. In <Fig. 4-a>, K/S values as dye uptake showed increase at the slowest rate from 20min up to 60min and seemed to reach equilibrium for 60min and 80min at both 10% and 20% of dye concentrations while as for 30% K/S value decreased slightly from 60 to 80 min. Under higher dye concentrations including 80%, 90%, and 100% in <Fig. 4-b> there were more obvious increases of dye uptake from 40 to 60 min showing the value of 16.46 of K/S for 100% while any further increase was not exhibited over 60 min except under 80%. This result means that the silk fabric reached the range of tolerance to dyeing with seabuckthorn leaves extract at around 60 min.

The impact of dyeing temperature from 40 to 80 °C on dye uptake was also investigated as shown in <Fig. 5-a> and <Fig. 5-b>. The lower dye concentrations such as 10%, 20%, and 30% did not exhibit any clear differences in terms of K/S values along the temperature levels as given in <Fig. 5-a>. On the contrary, under higher concentrations including 80%, 90%, and 100%, the dye uptake values showed a tendency of gradual increase as the dyeing temperature got higher up to 80°C. This result may be caused by the fiber swelling effects owing to higher temperature levels that improve the dye diffusion and better dye uptake. Overall, dyeing temperature seems to be more influential on dye uptake under higher concentrations rather than under lower ones. From these results, stronger dye uptake was assumed as obtained at 80°C but any saturation was not confirmed, which means more dye uptake could be expected at temperatures higher than 80 °C. Nevertheless much higher temperature than 80°C have been rarely employed in natural dyeing fields. By that reason, 80°C was concluded as the optimum dyeing temperature of silk with seabuckthorn leaves extract in this study.

Finally, K/S values at 360λ as the wavelength of the maximum absorbance and the sum of K/S (K/S SUM) of all wavelengths between 360λ and 700λ were examined respectively depending on dye concentrations



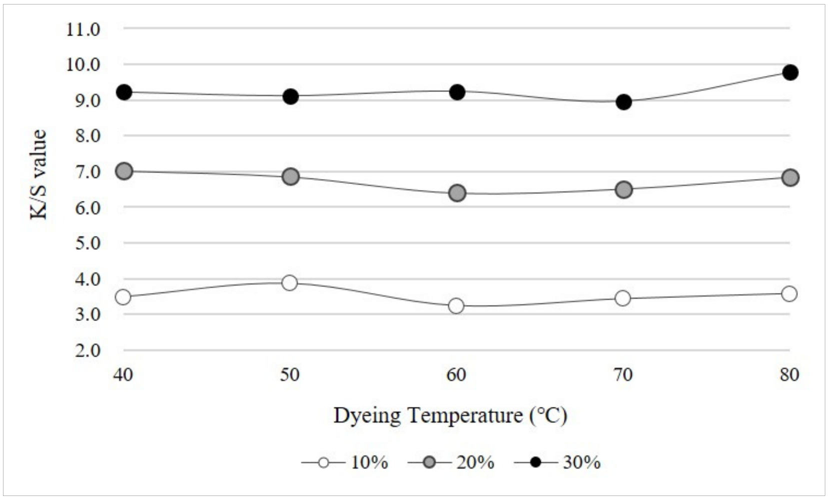
(a) lower dye concentrations



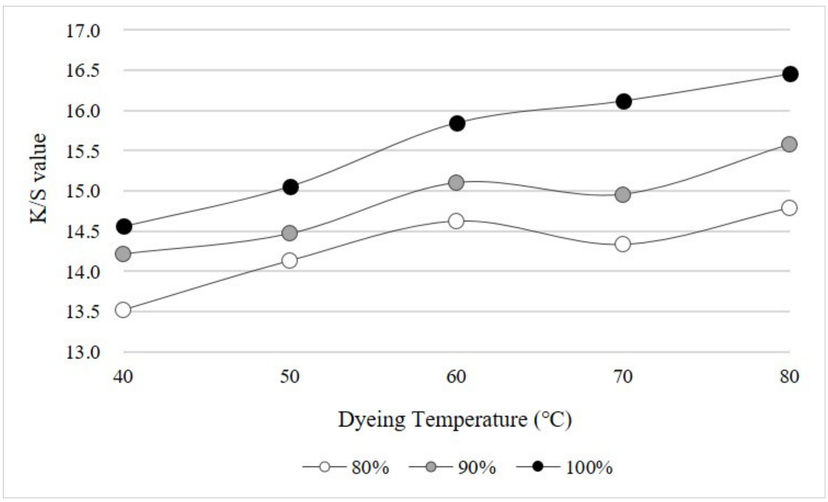
(b) higher dye concentrations

Fig. 4. Effects of dyeing duration on dye uptake (K/S) depending on dye concentrations (80°C).

from 10 to 100 % o.w.f. as given in <Fig. 6-a> and <Fig. 6-b>. It is clear that the dye uptake of K/S values increased with higher concentrations of dye bath. In details, as dye concentrations increased from 10% up to 40%, K/S values at 360λ rose up faster as seen in <Fig. 6-a>. However after 40%, the dye uptake value increased less rapidly up to 16.46 at 360λ. The sum of K/S would reflect general color intensity considering throughout shades which might be caused from reflectance at all visible wavelength as depicted in some previous works (Ferreira et al., 2023; Sarmandakh et al., 2017; Štěpánková et al., 2011; Tang et al., 2019). As given in <Fig. 6-b>, the increase along with concentrations was seen similarly to the increase tendency at 360λ in that steeper increase up to 40% and slower thereafter. Therefore, in this study, seabuckthorn leaves extract might provide relative even shades to silk woven and overall it could be confirmed that dye uptake was processed proportionally to dye concentrations little with equilibrium even up to 100%, which



(a) lower dye concentrations

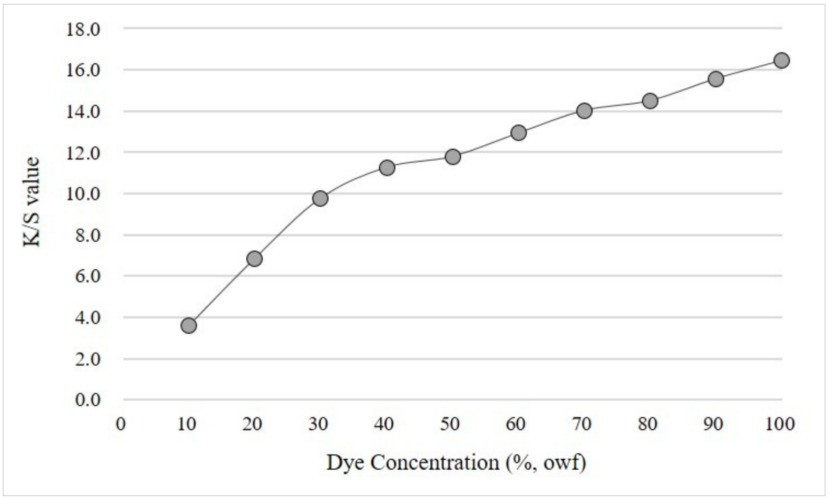


(b) higher dye concentrations

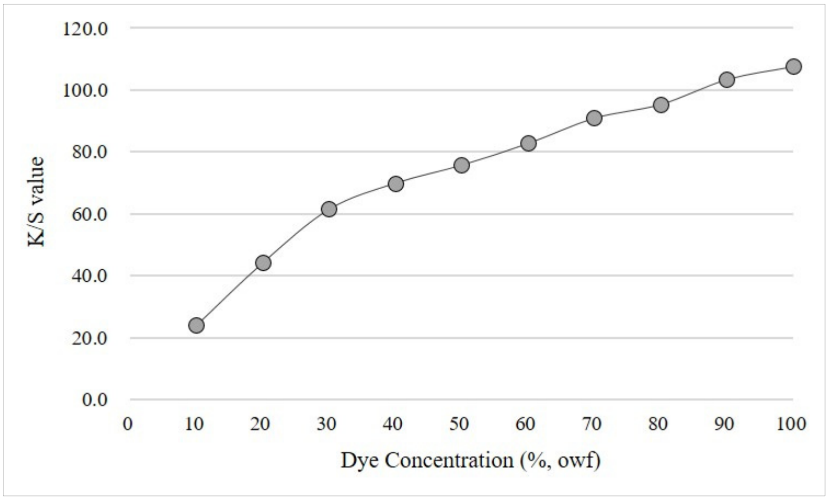
Fig. 5. Effects of dyeing temperature on dye uptake (K/S) depending on dye concentrations (60min).

leads to the conclusion that dye absorption behavior of seabuckthorn leaves extract on silk might be in accordance to Freundlich isotherm (Elemen et al., 2012). Consequently, the optimum dyeing conditions of the silk fabric were determined as 80°C and 60 minutes. The optimum dyeing conditions were employed for further dyeing with pH adjustment, color fastness, and biological assessments.

The colorimetric characteristics of the dyed fabric under the optimum dyeing condition, were given along with dye concentrations as shown in <Table 2>. The values of a* and b* indicating redness and yellowness respectively in CIE color space, showed both positive values along the all dye concentrations, which led to Yellow Red (YR) of hue notation in Munsell for all of dyed fabrics and furthermore their YR shades seemed to be nearer to Yellow rather than to Red. Consequently, silk fabrics dyed with seabuckthorn were thought to develop YR color series at all concentrations from 10 to 100% owf. Owing to the increase of both a* and b*



(a) K/S values at 360λ



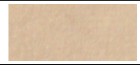
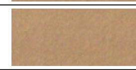
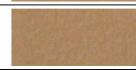
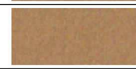
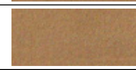
(b) K/S SUM values between 360λ and 700λ

Fig. 6. Effects of dye concentrations on dye uptake (K/S) (80°C, 60min).

values with an increase in the dye concentrations, the increases of both CIE C* and Munsell C representing color saturation were obtained. It means that higher dye concentrations could result in colorfulness rising. Nevertheless the colorfulness by CIE C* and Munsell C did not go over 30 and 50 respectively, which means that the dyed silk might be still in lower saturation similar to other fiber substrates dyed with some plant leaves sources. On the other hands, the values of both CIE L* and Munsell V, which signifies brightness, got lower as the concentration of dye bath increased with colors being darken. Since the values ranged from 75.27 to 59.54 for CIE L* and from 7.38 to 5.79, this can be depicted as a medium brightness.

Accordingly, PCCS tone was also determined for each dyed sample with compared visually to PCCS tone chart. The lowest dye solution gave pale tone to silk while highly concentrated solutions caused deeper and darker tones. Accordingly the range of dye concentrations from 10% to 100% was revealed as provid-

Table 2. Color characteristics of dyed silk fabric depending on dye concentrations

Dye conc. (%, owf)	CIE				Munsell			ΔE*	PCCS tone**	Photo of dyed fabric
	L*	a*	b*	*C**	H	V	C			
10	75.27	3.23	16.21	16.53	9.34YR	7.38	2.57	29.16	p	
20	68.82	4.24	19.09	19.56	9.35YR	6.72	3.06	35.73	ltg	
30	65.42	4.73	20.80	21.33	9.43YR	6.37	3.35	39.40		
40	64.67	4.73	21.44	21.95	9.54YR	6.30	3.43	40.37	sf	
50	63.02	4.91	21.76	22.31	9.54YR	6.13	3.50	41.82		
60	62.16	4.98	22.19	22.74	9.59YR	6.05	3.56	42.75		
70	61.19	5.16	22.91	23.48	9.63YR	5.95	3.68	43.96		
80	60.43	5.30	23.19	23.79	9.58YR	5.87	3.73	44.73	d	
90	59.57	5.42	23.75	24.36	9.65YR	5.79	3.82	45.76		
100	59.54	5.44	24.13	24.74	9.70YR	5.79	3.87	46.03		

* color difference from undyed silk fabric
** p: pale, ltg: light grayish, sf: soft, d: dull

ing four different PCCS tones including pale, light grayish, soft, and dull on silk. In summary, all of the dyed silk fabrics with seabuckthorn were of a medium brightness and low saturation, and they belonged to YR series, while tones were varied from pale to dull depending on dye concentrations.

3. Effects of pH on Dyeing Properties and Color Characteristics

In order to examine the effect of pH on dye uptake, dyeing according to pH was carried out under the dyeing concentrations of 10%, 30%, 50%, and 90% (owf), as representatives chosen as for each tone (Fig. 7). Prior to adjusting pH of dye solution, the unadjusted pH value of the solution by seabuckthorn leaves extract was found as between 5 and 5.3, that is, slightly acidic. The results showed that the higher the pH adjusted, the lower the K/S values were shown on silk, while the lower the pH, the higher the K/S values. This result was partially supported by some previous works dealing with protein fiber materials dyed with natural plant dye sources (Fang et al., 2022; Ren et al., 2016; Yang & Yi, 2020), which could be concerned as due to the amino group (-NH_3) among dye solutions coming to have cationic properties which makes ionic bonding stronger with anions from seabuckthorn leaves extract resulting greater dye absorption. On the contrary K/S values decreased rapidly as alkalinity of dye solution got stronger at pH 7 over, which implies that ionic bonding force between fibers and seabuckthorn leaves extract seemed to get less strong. In summary, seabuckthorn is assumed as a proper acidic dye for protein fibers such as silk from the result that the dye uptake of silk is stronger under acidic dye solutions.

Color characteristics of silk dyed under the optimized conditions and a variety of pH adjustment were given in <Table 3>. Even when the dye solution is

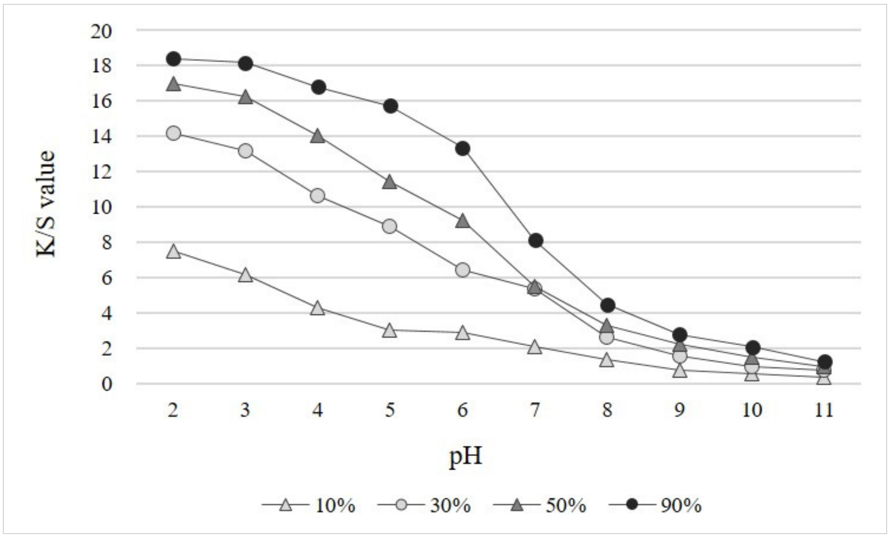


Fig. 7. Effects of pH adjustment on dye uptake depending on dye concentrations.

Table 3. Color characteristics of dyed silk fabric depending on pH

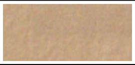
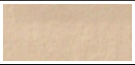
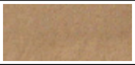
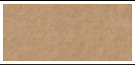
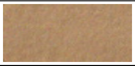
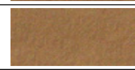
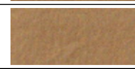
Dye conc. (%, owf)	pH	CIE				Munsell			ΔE*	PCCS tone	Photo of dyed fabric
		L*	a*	b*	*C**	H	V	C			
10	2	64.77	5.19	20.45	21.09	9.07YR	6.31	3.35	12.60	sf	
	3	68.93	4.37	19.57	20.05	9.36YR	6.73	3.14	8.41	sf	
	4	73.84	3.54	17.18	17.54	9.34YR	7.23	2.73	2.91	ltg	
	5	76.20	3.24	15.50	15.84	9.12YR	7.47	2.48	-	p	
30	2	56.00	6.10	22.62	23.43	9.19YR	5.43	3.71	11.24	d	
	3	59.61	5.92	23.17	23.91	9.22YR	5.79	3.80	7.97	sf	
	4	64.69	4.77	21.41	21.94	9.54YR	6.30	3.43	2.52	sf	
	5	66.83	4.48	20.12	20.61	9.46YR	6.52	3.22	-	ltg	
50	2	54.37	6.38	24.73	25.54	9.41YR	5.27	4.00	10.10	d	
	3	56.64	5.82	24.02	24.72	9.60YR	5.50	3.87	7.67	d	
	4	61.14	4.99	22.37	22.92	9.65YR	5.94	3.59	2.81	sf	
	5	63.82	4.77	21.54	22.06	9.57YR	6.21	3.45	-	sf	

Table 3. Continued

Dye conc. (%, owf)	pH	CIE				Munsell			ΔE*	PCCS tone	Photo of dyed fabric
		L*	a*	b*	C*	H	V	C			
90	2	51.05	6.76	25.28	26.17	9.41YR	4.95	4.08	9.54	g	
	3	54.24	6.46	25.72	26.52	9.53YR	5.26	4.14	6.51	g	
	4	57.81	5.66	24.79	25.42	9.74YR	5.61	3.97	2.75	g	
	5	60.37	5.32	23.85	24.43	9.68YR	5.87	3.83	-	d	

* color differences from dyed silk fabric without pH adjustment
** p: pale, ltg: light grayish, sf: soft, d:dull, g: grayish

strong acidic, the shades on the dyed silk were all shown as YR. As for brightness and saturation, more acidic state had a tendency to give lower CIE L* and Munsell L while to show higher CIE C* and Munsell C, which means deeper and more saturated shades might result from stronger acidity. PCCS tone differed according to pH even under an identical dye concentration. Precisely, more variety of tones were obtained under lower dye concentrations by pH adjustment.

4. Dyeing Fastness

<Table 4> shows the results of dyeing fastness of dyed silk fabrics selected for each PCCS tone in the way that an unadjusted dye solution was first concerned and any pH-adjusted one was invited secondly for each tone. As for the weakest tone, pale, only 10% owf of dye solution without pH adjustment was employed for testing color fastness. The fastness to perspiration was found to be excellent, mostly at grade 4 or higher for every dyed fabric in terms of both changes in color and the contamination of silk and cotton, except for the grayish-toned silk at 90% (pH 4) showing grade 3-4 for both color changes due to alkaline and the contamination of cotton by alkaline sweat. The fastness to rubbing under humid and dry conditions was shown as excellent with grade 4 or higher for both pale-toned under 10% and grayish-toned under 90%; The grades of lightly grayish-toned silk under 30% were 4; those of silk fabrics under 30%, 50%, and 90% of dye without any adjustment of pH had 3-4 grades for dry rubbing while they did 4 for wet rubbing. In terms of color fastness to light, grades 4 depicting good fastness were given to both paled and light grayish-toned fabrics. Even though a lower grade of 2 was assigned to grayish-toned silk under 90% of dye at pH 4, dyeing fastness of silk with in this study seemed as better when compared to the results by other naturally dyed fabrics in some previous studies including silk and cotton without any additives like a mordant (Repon et al., 2016; Yang & Yi, 2020). Regarding to the fastness to dry cleaning, the most of dyed fabrics were excellent in general with 4-5 grades except for two different dull-toned silk (grades 3-4) and a grayish-toned one (grades 2-3), which implies darker toned fabrics are likely to be poor in color fastness to dry cleaning.

In summary, perspiration and rubbing fastness were mostly graded as 4 or 4-5, which implies good and excellent for silk fabrics dyed with seabuckthorn even under various dye solutions and pH adjustments. In the case of color fastness to rubbing, light, and dry cleaning, a few dyed fabrics were scored as fair (3-4 or 3) and especially the most dark-toned one as grayish was thought to need an improvement for the fastness to light and the color changes by dry cleaning because it got a poor grade of 2 and 2-3 respectively. As for light grayish, soft, and dull, pH-adjusted fabrics under lower dye concentrations were found as showing little differences in dyeing fastness from unadjusted counter-

Table 4. Dyeing fastness properties of dyed silk fabric (80°C, 60min)

Dyeing fastness properties			PCCS tone & dyeing conditions	p	ltg		sf		d		g
				-	pH4	-	pH4	-	pH3	-	pH4
				10	10	30	30	50	50	90	90
Perspiration	Acidic	Color change		4-5	4	4-5	4	4-5	4	4-5	4-5
		Stain	Cotton	4-5	4-5	4-5	4-5	4	4	4-5	4-5
			Silk	4-5	4-5	4-5	4-5	4	4	4-5	4-5
	Alkaline	Color change		4-5	4	4	4	4	4	4-5	3-4
		Stain	Cotton	4-5	4-5	4-5	4-5	4-5	4-5	4-5	3-4
			Silk	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5
Rubbing	Dry			4-5	4	3-4	4	3-4	4	3-4	4-5
	Wet			4-5	4	4-5	3-4	4-5	3-4	4-5	4
Light	Color change			4	4	4	3	4	3	4	2
Dry cleaning	Color change			4-5	4-5	4-5	4-5	4-5	3-4	3-4	2-3
	Stain	Acetate		4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5
		Cotton		4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5
		Polyamide		4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5
		Polyester		4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5
		Acrylic		4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5
		wool		4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5

part in each identical tone. This result means that pH-adjustment could be useful to impart higher color intensity even with lower dye concentrations without concerns of getting dyeing fastness degraded.

5. UV Protection Properties

The silk fabrics dyed with 10%, 30%, 50%, and 90% of dye solutions respectively were evaluated in terms of UV protection performances. As presented in <Table 5>, while undyed silk showed 10.8 for UPF indicating the UV blocking effect grade is zero(0), this figure increased significantly to 47.8 (very good) for pale-toned silk under 10% of dyes and the other silk fabrics with light grayish, soft, and dull of tones exhibited all over 100 of UPF (excellent) with the grades of 50+, representing very excellent UV protection performance. As for UV cut ratio %, all of dyed silk fabrics presented 96% or higher for UV-A, and 98% or higher for UV-B. The silk fabric used in current study was slightly thicker than other woven types of silk, which might result in higher values of both UPF and UV cut ratio than others. Nevertheless the undyed silk fabric failed to have a practicable values for sun-pro-

Table 5. UV protection properties of dyed silk fabrics dependong on PCCS tones

PCCS tone	Dye conc. (%, owf)	UV protection factor		UV cut ratio (%)	
		UPF	Grade	UV-A	UV-B
-	Undyed	10.8	0	83.8	94.3
p	10	47.8	40	96.3	98.5
ltg	30	136.6	50+	98.7	99.4
sf	50	211.5	50+	99.2	99.6
d	90	266.9	50+	99.4	99.7

tective textiles. Owing to dyeing with seabuckthorn leaves extract, UV-blocking efficacies of the dyed silk were obviously improved. These results first seemed as caused by darker and more intense shades by dyeing with seabuckthorn leaves extract. Furthermore it could be noted that flavonoids in the extract might contribute to improving UV protection properties considering some previous works (Ferreyra et al., 2021; Laoué et al., 2022) which have reported some natural effective components such as flavonoids, tannins, and anthraquinones have strong UV-absorbing property.

6. Antimicrobial Activity

Antimicrobial activities of silk fabric dyed with seabuckthorn under 10% and 30% of dye solution respectively were evaluated using *S. aureus* and *K. pneumoniae* as given in <Table 6>. As for 10% dyed silk fabric, bacterial reduction rate against S. aureus was 99.9% while 91.4% against K. pneumoniae, of which both are fairly effective bacterial reduction rates against the two bacteria. Antimicrobial activity of 30%-dyed fabric was improved in that bacterial reduction rate against K. pneumoniae raised up to 99.9% identical to its rate against S. aureus, which leads to an assumption that higher dye concentrations than 30% might provide excellent antimicrobial efficacy. From these results we could grant antimicrobial activity on silk fabric regarded as fairly acceptable as a bio-functional textiles by dyeing it under the conditions of more than 10% o.w.f. dye concentration of seabuckthorn leaves extract. In addition, considering that fastness rates of 10% dyed fabric was good in general as discussed above, it can be concluded that 10 % of dye concentration for seabuckthorn is acceptable to prepare an antibacterial silk fabric. However in order to obtain both antimicrobial activity and UV protective performance from seabuckthorn-dyed silk, 30% of dye concentration could be recommended because 30%-dyed fabric showed very excellent UV protection properties.

Table 6. Antimicrobial activities of dyed silk fabric under 10% and 30% of dye concentrations

Dye Conc. (%, owf)	Bacterial reduction rate (%)	
	S. aureus	K. pneumoniae
Control	 -	 -
10	 99.9	 91.4
30	 99.9	 99.9

IV. Conclusions

In this study, seabuckthorn leaves extract was explored in terms of its potential use as a biofunctional dye for a silk woven fabric. The extract was examined for their chemical characterization and biological assessment such as cell viability and total phenolic contents and its optimum dyeing conditions, the effects of pH adjustment, dyeing fastness for the silk were investigated. Furthermore UV protection properties and antimicrobial activity of the dyed silk were evaluated. The results and their implications are as follows;

1. Through UV/Vis spectral observation and FT-IR characterization, some of flavonoids compounds were expected as contained in seabuckthorn leaves extract, which was quantitatively confirmed in total phenolic contents of 29.08mg/g while the extract was identified as non-toxic by cell viability.

2. The silk fabric was optimized for maximum dye uptake as 80°C for temperature and 60 minutes for du-

ration and all of the dyed fabrics were colored with a medium brightness and low saturation, and hue of Yellow Red series, while tone were varied from pale to dull depending on dye concentrations. Furthermore, the dye uptake of silk was found as improved under acidic conditions with stronger saturations and darkness while staying in the shade of Yellow Red.

3. The dyeing fastness mostly showed good and excellent grades of 4 or 4-5 in perspiration and rubbing fastness, and fair grade (3-4 or 3) in fastness to rubbing, light, and dry cleaning for silk fabrics dyed with seabuckthorn leaves extract even under various dye solutions and pH adjustments, which leads to the conclusion that various tones such as pale, light grayish, soft, and dull could be obtained by varying the conditions of dye concentrations and pH adjustment without concerns of fastness degradation. However the fastness to light needed an improvement in a future for the most dark-toned silk because it received a poor grade of 2 around.

4. As biofunctional features for skin health, UV protection properties and antimicrobial activity of the dyed silk were evaluated depending on dye concentrations. The dyed silk at 30% or higher concentrations showed superior UV protection properties (a UV protection factor of 50 or higher). In terms of quantitative antimicrobial activity, 30% and higher concentrations were seen to provide excellent antimicrobial efficacy of 99.9% as bacterial reduction rate against both S. aureus and K. pneumoniae, which leads to the conclusion that higher dye concentrations than 30% of seabuckthorn leaves extract might provide excellent antimicrobial efficacy.

These results could contribute to designing biofunctional textiles in respect of sustainability by employing seabuckthorn leaves extract for silk for high-valued sustainable textile products in providing UV-protection and antimicrobial efficacy. In a future, natural mordanting and any biologically compatible additives to improve dye fastness need to be considered. Washing durability of UV protection and antimicrobial activity is another area of interest.

1. Acknowledgement

This research was supported by the 2023 scientific promotion program funded by Jeju National University.

2. Ethics and consent

Not applicable.

3. Availability of data and materials

The datasets in used during the current study are available from the corresponding author on reasonable request.

4. Conflicting interests

Not applicable.

5. Funding

This research was supported by the 2023 scientific promotion program funded by Jeju National University

6. Authors' contributions

SB was responsible for data collection, experiments and data analysis and drafted initial manuscript. EY supervised the findings of this work and responsible for final manuscript completion, and funding acquisition including the overall management of the study. All authors have read and approved the final manuscript.

7. Authors' information

Sarmandakh Badmaanyambuu

Design Development, Taesan Industrial Corporation, Team Leader/Dept. of Fashion and Textiles, Jeju National University, Ph.D.

Eunjou Yi

Dept. of Fashion and Textiles, Jeju National University/ Faculty of Data Science for Sustainable Growth, Jeju National University, Professor

References

An, S. J. (2016). *The analysis of main ingredients influencing the skin and the inhibitory effects on cytotoxicity and nitric oxide(no) production in fat-soluble green tea extracts* [Unpublished master's thesis]. Nambu University.

Badmaanyambuu, S. (2022). *A study of skin health functional textiles by treatment with Sea Buckthorn extract* [Unpublished doctoral dissertation]. Jeju National University.

Bartolini, D., Pallottelli, L., Sgargetta, D., Varfaj, I., Macchiarulo, A., Galli, F., Romani, A., Sardella, R., & Clementi, C. (2024). Natural cellulosic biofunctional textiles from onion (Allium cepa L.) skin extracts: A sustainable strategy for skin protection. *Industrial Crops and Products*, *212*(6), 118295. https://doi.org/10.1016/j.indcrop. 2024.118295

Beśko, P., Biel, W., Witkowicz, R., & Piątkowska, E. (2024). Chemical composition and nutritive value of Sea Buckthorn leaves. *Molecules*, *29*(15), 3550. https://doi.org/10.3390/molecules29153550

Buyukokuroglu, M. E., & Gulcin, I. (2009). In vitro antioxidant and antiradical properties of Hippophae rhamnoides L. *Pharmacognosy Magazine*, *5*(19), 189–195.

Cho, K. R. (2004). *Natural dyes and dyeing*. Hyungseul.

Criste, A., Urcan, A. C., Bunea, A., Pripon Furtuna, F. R., Olah, N. K., Madden, R. H., & Corcionivoschi, N. (2020). Phytochemical composition and biological activity of berries and leaves from four Romanian sea buckthorn (Hippophae rhamnoides L.) varieties. *Molecules*, *25*(5), 1170. https://doi.org/10.3390/molecules25051170

Ebrahim, S. A., Mosaad, M. M., Othman, H., & Hassabo, A. G. (2022). A valuable observation of eco-friendly natural dyes for valuable utilisation in the textile industry. *Journal of Textiles, Coloration and Polymer Science*, *19*(1), 25–37. https://doi.org/10.21608/jtcps.2021.97342.1090

Elemen, S., Kumbasar, E. P. A., & Yapar, S. (2012). Modeling the adsorption of textile dye on organoclay using an artificial neural network. *Dyes and Pigments*, *95*(1), 102–111. https://doi.org/10.1016/j.dyepig.2012.03.001

Fang, J., Meng, C., & Zhang, G. (2022). Agricultural waste of Ipomoea batatas leaves as a source of natural dye for green coloration and bio-functional finishing for textile fabrics. *Industrial Crops and Products*, *177*, 114440. https://doi.org/10.1016/j.indcrop.2021.114440

Ferreira, F. L., de Campos, É. A., Santos, W. L. F., & da Silva, M. G. (2023). Wool fabric dyeing with natural dye extracted from cinnamomum verum J. presl fruit. *Fibers and Polymers, 24*(12), 4301–4309. https://doi.org/10.1007/s12221-023-00370-x

Ferreyra, M. L. F., Serra, P., & Casati, P. (2021). Recent advances on the roles of flavonoids as plant protective molecules after UV and high light exposure. *Physiologia Plantarum*, *173*(3), 736–749. https://doi.org/10.1111/ppl.13543

Folin, O., & Denis, W. (1912). On phosphotungstic-phosphomolybdic compounds as color reagents. *Journal of Biological Chemistry*, *12*(2), 239–249. https://doi.org/10.1016/S0021-9258(18)88697-5

Haji, A., Shahmoradi Ghaheh, F., & Indrie, L. (2023). Pomegranate fallen leaves as a source of natural dye for mordant-free dyeing of wool. *Coloration Technology*, *139*(2), 165–170. https://doi.org/10.1111/cote.12651

He, Q., Yang, K., Wu, X., Zhang, C., He, C., & Xiao, P. (2023). Phenolic compounds, antioxidant activity and sensory evaluation of sea buckthorn (Hippophae rhamnoides L.) leaf tea. *Food Science & Nutrition*, *11*(3), 1212–1222. https://doi.org/10.1002/fsn3.3155

Jeong, Y.-S., Jung, H.-K., Youn, K.-S., Kim, M.-O., & Hong, J.-H. (2009). Physiological activities of the hot water extract from Eriobotrya japonica Lindl. *Journal of the Korean Society of Food Science and Nutrition*, *38*(8), 977–982. https://doi.org/10.3746/jkfn.2009.38.8.977

Korean Agency for Technology and Standards. (2017, December 13). *KS K 0650-1 Test method for color fastness to rubbing: Crock meter method*. Korean Standards & Certifications. https://e-ks.kr/streamdocs/view/sd;streamdocsId=72059197970371628

Korean Agency for Technology and Standards. (2019a, December 17). *KS K ISO105-E04 Textiles - Tests for colour fastness - Part E04: Colour fastness to perspiration*. Korean Standards & Certifications. https://e-ks.kr/streamdocs/view/sd;streamdocsId=72059197887273701

Korean Agency for Technology and Standards. (2019b, December 30). *KS K 0850 Test method for ultraviolet blocking and sun protection factor of textiles*. Korean Standards & Certifications. https://e-ks.kr/streamdocs/view/sd;streamdocsId=72059197723529206

Korean Agency for Technology and Standards. (2021a, July 1). *KS K ISO105-D01 Tests for colour fastness - Part D01: Colour fastness to dry cleaning using perchloroethylene solvent*. Korean Standards & Certifications. https://e-ks.kr/streamdocs/view/sd;streamdocsId=72059219239491253

Korean Agency for Technology and Standards. (2021b, July, 21). *KS K ISO105-B02 Textiles - Tests for colour fastness - Part B02: Colour fastness to artificial light: Xenon arc fading lamp test*. Korean Standards & Certifications. https://e-ks.kr/streamdocs/view/sd;streamdocsId=72059219161355820

Kwon, E.-Y., Lee, J., Kim, Y. J., Do, A., Choi, J.-Y., Cho, S.-J., Jung U. J., Park, Y. B., & Choi, M.-S. (2017). Seabuckthorn leaves extract and flavonoid glycosides extract from seabuckthorn leaves ameliorates adiposity, hepatic steatosis, insulin resistance, and inflammation in diet-induced obesity. *Nutrients*, *9*(6), 569. https://doi.org/10.3390/nu9060569

Laoué, J., Fernandez, C., & Ormeño, E. (2022). Plant flavonoids in mediterranean species: A focus on flavonols as protective metabolites under climate stress. *Plants*, *11*(2), 172. https://doi.org/10.3390/plants11020172

Liu, Y. S., & Chen, L. (2012). Application of montmorillonite in dyeing on silk fabric with mugwort. *Advanced Materials Research*, *550*, 1435–1437. https://doi.org/10.4028/www.scientific.net/AMR.550-553.1435

Liu, Y., Yao, W., Qin, F., Zhou, L., & Zheng, Y. (2023). Spectral classification of large-scale blended (Micro) plastics using FT-IR raw spectra and image-based machine learning. *Environmental Science & Technology*, *57*(16), 6656–6663. https://doi.org/10.1021/acs.est.2c08952

Lo, C.-H. H. (2021). Degumming silk by CO_2 supercritical fluid and their dyeing ability with plant indigo. *International Journal of Clothing Science and Technology*, *33*(3), 465–476. https://doi.org/10.1108/IJCST-06-2019-0072

Oh, J., & Ahn, C. (2011). A Study on the current status and dyeing characteristics of natural indigo powder dye. *Journal of the Korean Society of Clothing and Textiles*, *35*(7), 736–747. https://doi.org/10.5850/JKSCT.2011.35.7.736

Park, Y. (2020). The dyeing properties of Mugwort (Artemisia princeps) extract using nano-cellulose. *Textile Coloration and Finishing*, *32*(3), 142–149. https://doi.org/10.5764/TCF.2020.32.3.142

Pizzicato, B., Pacifico, S., Cayuela, D., Mijas, G., & Riba-Moliner, M. (2023). Advancements in sustainable natural dyes for textile applications: A review. *Molecules*, *28*(16), 5954. https://doi.org/10.3390/molecules28165954

Pranta, A. D., & Rahaman, M. T. (2024). Extraction of eco-friendly natural dyes and biomordants for textile coloration: A critical review. *Nano-Structures & Nano-Objects*, *39*, 101243. https://doi.org/10.1016/j.nanoso.2024.101243

Rahman Bhuiyan, M. A., Ali, A., Islam, A., Hannan, M. A., Fijul Kabir, S. M., & Islam, M. N. (2018). Coloration of polyester fiber with natural dye henna (Lawsonia inermis L.) without using mordant: A new approach towards a cleaner production. *Fashion and Textiles*, *5*(2), 1–11. https://doi.org/10.1186/s40691-017-0121-1

Räisänen, R., Primetta, A., Nikunen, S., Honkalampi, U., Nygren, H., Pihlava, J.-M., Berghe, I. V., & Wright, A. v. (2020). Examining safety of biocolourants from fungal and plant sources-examples from Cortinarius and Tapinella, Salix and Tanacetum spp. and dyed woollen fabrics. *Antibiotics*, *9*(5), *266*. https://doi.org/10.3390/antibiotics9050266

Rather, L. J., Zhou, Q., Ali, A., Haque, Q. M. R., & Li, Q. (2020). Valorization of natural dyes extracted from Mugwort leaves (Folium artemisiae argyi) for wool fabric dyeing: Optimization of extraction and dyeing processes with simultaneous coloration and biofunctionalization. *ACS Sustainable Chemistry & Engineering*, *8*(7), 2822–2834. https://doi.org/10.1021/acssuschemeng.9b06928

Ren, Y., Gong, J., Wang, F., Li, Z., Zhang, J., Fu, R., & Lou, J. (2016). Effect of dye bath pH on dyeing and functional properties of wool fabric dyed with tea extract. *Dyes and Pigments*, *134*, 334–341. https://doi.org/10.1016/j.dyepig.2016.07.032

Repon, M. R., Mamun, M. A. A., & Islam, M. T. (2016). Eco-friendly cotton coloration using banana (Musa sapientum) waste: Optimization of dyeing temperature. *Universal Journal of Engineering Science*, *4*(1), 14–20. https://doi.org/10.13189/ujes.2016.040102

Sarmandakh, B., Lee, A. R., & Yi, E. (2017). Dyeing properties and bio-function of cotton fabrics with Ecklonia cava extract. *Textile Science and Engineering*, *54*(5), 386–396. https://doi.org/10.12772/TSE.2017.54.386

Sharma, B., & Deswal, R. (2018). Single pot synthesized gold nanoparticles using Hippophae rhamnoides leaf and berry extract showed shape-dependent differential nanobiotechnological applications. *Artificial Cells, Nanomedicine, and Biotechnology*, *46*(sup2), 408–418. https://doi. org/10.1080/21691401.2018.1458034

Son, K. (2023). Expansion of color space in Hanji fabrics by using sustainable natural dyeing: Focused on natural indigo dyeing and combination dyeing. *Textile Coloration and Finishing*, *35*(2), 82–96. https://doi.org/10.5764/TCF.2023.35.2.82

Štěpánková, M., Wiener, J., & Rusinová, K. (2011). Decolourization of vat dyes on cotton fabric with infrared laser light. *Cellulose*, *18*(2), 469–478. https://doi.org/10.1007/s10570-011-9494-2

Sukemi, Pratumyot, K., Srisuwannaket, C., Niamnont, N., & Mingvanish, W. (2019). Dyeing of cotton with the natural dye extracted from waste leaves of green tea (Camellia sinensis var. assamica). *Coloration Technology*, *135*(2), 121–126. https://doi.org/10.1111/cote.12381

Tang, A. Y., Lee, C. H., Wang, Y. M., & Kan, C. W. (2019). A study of PEG-based reverse micellar dyeing of cotton fabric: Reactive dyes with different reactive groups. *Cellulose*, *26*, 4159–4173. https://doi.org/10.1007/s10570-019-02340-0

Verma, S., & Gupta, G. (2017). Natural dyes and its applications: A brief review. *International Journal of Research and Analytical Reviews*, *4*(4), 57–60.

Yang, Y.-A., & Yi, E. (2020). Dyeing properties, UV protection, and deodorization of silk fabric using a hot- water extract of an Acorus gramineus Solander Leaf. *Textile Science and Engineering*, *57*(1), 31–41. https://doi.org/10.12772/TSE.2020.57.031

Yi, E., & Yoo, E. S. (2010). A novel bioactive fabric dyed with unripe *Citrus grandis* Osbeck extract Part 1: Dyeing properties and antimicrobial activity on cotton knit fabrics. *Textile Research Journal*, *80*(20), 2117–2123. https://doi.org/10.1177/0040517510373633

Yogendra Kumar, M. S., Raghu, T. S., Varghese, F. V., Shekar, R. I., Kotresh, T. M., Rajendran, R., Babu, K. M., & Padaki, V. C. (2016). Application of seabuckthorn (Hippophae rhamnoides L.) leaf extract as antimicrobial finish on aramid fabric. *Journal of Industrial Textiles*, *45*(5), 1115–1127. https://doi.org/10.1177/1528083714553689