REGULAR ARTICLE



Silk Textiles Dyeing by Plant-Derived Colorant in the Presence of Chitosan and Shellac

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Received: 28 November 2022 / Revised: 3 April 2023 / Accepted: 13 May 2023 / Published online: 27 June 2023 © The Author(s) 2023

Abstract

The research aimed to dye silk satin textiles with a plant-derived colorant in the presence of additives of natural origins, such as chitosan and shellac, to investigate whether they can dye silk textiles with satisfactory results. In this study, a series of mechanical properties (Young modulus, elongation at break, and tensile strength), and colorfastness following exposure to UVC irradiation and 6 months of storage of dyed silk textiles samples were tested. Colorimetric measurements and scanning electron microscopy (SEM) images were performed and FTIR spectra were registered. Results indicated that the plant-derived colorant used in this research had the potential to dye silk satin textiles with satisfactory results. Colorant with the addition of chitosan and shellac dyed silk textiles more intensely, shellac covering ensured their color stability following UVC irradiation and during storage while the addition of chitosan did not particularly affect the color stability under the influence of UVC irradiation. However, pre-treatment with low molecular weight chitosan improves colorfastness during storage. Both additives influenced the mechanical properties of dyed silk textiles.

Keywords Plant-derived colorant · Silk textiles · Natural dyeing · Chitosan · Shellac

1 Introduction

Silk is a fiber of animal origin consisting of two types of proteins: fibroin (70–80%) and sericin (20–30%). Fibroin is made of amino acids, such as glycine, serine, and alanine, and forms a β -sheet structure. Fibroin forms the basis of silk fiber and is surrounded by sericin acting as a binder. Silk, whose structure is linked by numerous hydrogen bonds, is one of the strongest naturally occurring fibers. In nature, it is a building material for cocoons and webs and can also be woven into fabrics. Silk textiles are characterized by smooth and soft texture, moderate elasticity, and resistance to breaking [1–3]. Nevertheless, UV radiation adversely affects silk structure and reduces its strength. Due to moisture absorbency and low conductivity, silk is

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widely used in the textiles industry to produce clothing, furniture, or various equipment. In addition due to biocompatibility and mechanical strength, silk is also desired as a biomaterial in tissue engineering and medicine. As textile production is indispensably connected with the dyeing process, silk coloration is of interest to both academic and industrial scientists. Sericulture is one of the oldest crafts that originated in China, thus silk dyeing has also a long and rich tradition. Nowadays, silk dyeing is dominated by synthetic dyes, but previously it was based on natural colorants obtained from numerous plants. There is a growing interest among scientists in researching the possibility of applying plant dyes in textiles coloration partly because of their environmentally friendly properties [4–6]. Many papers focus on alternatives to synthetic colorants in textile dyeing and the application of a wide range of plants that can be sources of efficient silk colorants [7-9]. Various natural colorants obtained from different plants, such as sophora flower bud [10], Cordyline fruticosa and Mussaenda erythrophylla [11], purple sweet potato [12], Vitex negundo [13], Quercus Robur and Salix Alba [14], Casuarina Equisetifolia [15], cumin seeds [16], grape skin powder [17], *Reinwardtia* flowers [18], croton leaves [19], coconut coir [20], hamelia and walnut (under acidic and alkaline conditions) [21], madder and gall oak [22], or Acacia auriculiformis [23], were used to dye silk fabrics and effects of the dyeing process were studied. Furthermore, numerous papers can be found concerning in particular the application of specific plant constituents such as anthocyanins in silk textile dyeing [24-31]. All of the conducted studies confirmed the potential of plantderived colorants to dye silk. Moreover, in most cases with very satisfying results and acceptable colorfastness or stability. Considering ecological silk dyeing, it is also worth mentioning that not only plant-derived colorants are gaining importance, but a new approach to dyeing fabrics is related to the use of microbiological pigments [32]. Plant dyeing of silk fabric is favored by the fact that protein-based fibers are dyed more easily than cellulosebased ones like cotton or linen. However, some colorants of plant origin are more complicated in application due to their low affinity to different kinds of fibers causing the low color intensity of dyed textiles; furthermore, they often are characterized by variability of shades and lower stability under various conditions. All of these factors contribute to the fact that additional substances enhancing their dyeability are used in textile dyeing. As an example, chitosan owns high potential as illustrated by numerous papers indicating the benefits of using this biopolymer. Once applied, it improves colorfastness, color strength, and dyeability, and ensures resistance to washing or antimicrobial properties [33–43]. Another natural material, which can improve selected parameters, can be shellac. In the past, shellac was used for protein fibers such as wool fabric to stiffen felt in the millinery industry or silk fabric as a shoe hardener for example in ballet points to increase material strength and ensure longevity and as a resinous binder in historic silk textiles.

Research concerning natural silk coloration also focuses on the elimination of metal mordants; as an example, it was concluded that a combination of natural dyes and environmentally friendly mordants enhanced the color strength and UV protection of the silk examined [44]. Bio-mordants are becoming crucial and significant elements ensuring qualitative textiles coloring and excellent color strength thus providing a wide range of shades. Many studies focus on this issue and various bio-mordant are tested, among others extracts of turmeric, pomegranate, henna or acacia, and many more [45–49]. Furthermore, another direction of research aiming to eliminate mordants is also marked by enzymatic silk dyeing [50]. Due to increasing attention to environmental issues and sustainable production, the idea of using plant colorants, sometimes even obtained from agro or food by-products, for dyeing natural textiles meets the current industrial and social needs for sustainable and responsible use of raw materials.

For the same purpose to ensure sustainable energy consumption, one of the possibilities to decrease the amount of energy involved in dyeing can be conducting the process at a lower or even room temperature; cold dyeing could be one of a less energy-consuming technique of dyeing fabrics. There are reports concerning the application of low temperatures in silk and other textile dyeing. For example, Vankar et al. in their research, dyed silk, wool, and cotton fabrics with tea leaf extracts at the temperature of 40 °C [51], whereas in another study, silk textiles were dyed with Rubia cordifolia extract with enzyme addition at a temperature of 30–40 °C [52]. Furthermore, silk material was also efficiently dyed with Monascus at a temperature of 30 °C with dye uptake reaching almost 90% [53] and also at room temperature in the dyeing machine [54]. Similarly, for continuous improvement and excellence of the dyeing process, the reduction of water resource consumption should also be taken into consideration. On the other hand, dyeing involving more advanced techniques is also studied. For instance, ultrasonic treatment is applied as an ecological tool in the dyeing process allowing to use of sustainable conditions to extract plant colorants [55, 56] or microwave treatment enhancing the isolation of dyes from plant sources, improving colorants applicability under mild dyeing conditions and additionally favorably affect the quality of dyed fabric [57-61]. Moreover, ultraviolet or ionizing radiation is applicable in the natural dyeing procedures, where dye powder and natural fabric are exposed to UV or gamma radiation to modify the surface and as a result obtain permanent dyed material [62, 63].

This study aimed to dye silk satin textiles with a colorant of plant origin in the presence of chitosan and shellac to investigate the ability of a colorant to dye natural textiles and the impact of additives on the silk textiles' properties. The mechanical properties (Young modulus, elongation at break, and tensile strength) and colorfastness following exposure to UVC irradiation (0.5, 2, and 4 h) were measured. Moreover, colorimetric measurements of dyed silk samples were also performed following 6 months of storage. FTIR spectra were collected, and scanning electron microscopy (SEM) images were registered. We intended to propose a new process of silk dyeing for cleaner production of colored silk textiles.

2 Materials

2.1 Textiles

In the experiment, non-dyed silk satin (60 g/m²) from Jedwab Polski Sp. z o. o. (Milanówek, Poland) was used.

2.2 Colorants

Plant-derived food colorant from EXBERRY® by GNT (GNT Group B.V., Mierlo, The Netherlands) was used in the silk dyeing process (red colorant in liquid form). Commercially available food colorant was from vegetables, fruits, or other edible plant concentrates manufactured only through a physical process conducted with water.

2.3 Silk Dyeing Additives

In this study, two grades of chitosan were utilized: a low molecular weight chitosan powder was purchased from Sigma Aldrich (Poznań, Poland) and a cosmetic-grade chitosan powder from Calaya-natural cosmetic raw materials (Złotów, Poland). Shellac flakes (dewaxed, blonde) were from the local art conservation and restoration shop and food-grade rectified spirit (95%) was from Polmos SA (Warsaw, Poland). Citric acid monohydrate pure P.A. was bought from Avantor Performance Materials Poland SA (Gliwice, Poland).

3 Methods

3.1 Silk Dyeing

Silk satin was dyed red. Dye was first dissolved in distilled water at two different concentrations of 1% and 4% (wt%). Silk satin samples were cold-dyed (at room temperature) for 24 h with a liquor ratio of 1:25 under acidic conditions. In addition to colorants, to a dye bath, chitosan was added, or samples were pre-treated with chitosan and then dyed. The samples were treated with colorants and with a 1% (wt%) solution of cosmetic-grade chitosan in 3% (wt%) citric acid. Selected samples were pre-treated with two grades of chitosan: a 1% (wt%) solution of low molecular weight chitosan in 3% (wt%) citric acid (LMWCS/CA) or a 1% (wt%) solution of cosmetic-grade chitosan in 3% (wt%) citric acid (CGCS/CA) with a liquor ratio of 1:10 for one hour at room

temperature and, subsequently dyed. Moreover, the reddyed sample without any additives was covered with a thin layer of 20% (wt%) dewaxed blond shellac ethanol solution (SH). All data concerning the silk dyeing process are listed in Table 1.

3.2 Colorimetric Measurements

Colorimeter (Colorimeter CL 400, Courage, Khazaka, Köln, Germany; core measuring area: 5 mm; illuminated area: approximately 17 mm; a range of emitted wavelengths: 440–670 nm; measurement uncertainty: $\pm 5\%$) was used to evaluate the color of dyed silk textiles samples, dyed silk textiles samples following 6 months of storage (with limited light exposure) and dyed silk textiles samples following exposure to UVC irradiation for 0.5, 2, and 4 h. The color parameter $L^*a^*b^*$ (the mean values of 3 measurements) was used to calculate the ΔE value using the equation $\Delta E = (\Delta L^2 + \Delta a^2 + \Delta b^2)^{0.5}$, where $\Delta L = L - L_0$, $\Delta a = a - a_0$, $\Delta b = b - b_0$, and L_0 , a_0 , b_0 are values of reference samples. The Nix Sensor color converter tool (Nix Sensor Ltd., Hamilton, Canada) was applied to visualize differences in colors of dyed silk textiles samples using the values of L, a, and *b* [64].

3.3 UV Irradiation

Samples of dyed silk textiles in different variants were irradiated for 30 min, 2 h, and 4 h at a distance of 5 cm from the UV lamp (ULTRAVIOL NBV 15, Ultra-Viol, Zgierz, Poland). The radiation intensity was 21.5 W/m² and the wavelength was 254 nm (the lamp used was emitting mainly UVC irradiation).

3.4 ATR-FTIR (Attenuated Total Reflection-Fourier Transform Infrared) Spectroscopy

ATR-FTIR spectra were registered for undyed samples, dyed samples, and for dyed samples following exposure to UVC irradiation using a spectrophotometer Nicolet iS10 with ATR

Table 1Parameters of dyeingprocess of silk textiles

Type of fabric	Silk textiles (60 g/m ²)							
Liquor ratio	1:25							
Type of dyeing	Cold dyeing							
Color of dye	Red							
Dye concentration [%]	1	1	1	1	4			
Pre-treatment prior to dyeing	no	LMWCS/ CA	CGCS/CA	no	no			
Additives in the dyeing process	CGCS/CA	no	no	no	no			
pH of dye bath	2.09	3.04	2.81	3.58	3.64			

accessory equipped with diamond crystal (Thermo Fisher Scientific, Waltham, MA, USA). Background scanning was registered prior to sample analysis. OMNIC software was applied to process data. The parameters of the spectro-photometer were as follows: the range of $400-4000 \text{ cm}^{-1}$, absorption mode at 4 cm⁻¹ intervals, and 64 times scanning.

3.5 Mechanical Properties

The mechanical testing machine (Z.05, Zwick and Roell, Ulm, Germany) was used to study the mechanical properties of dyed silk textiles samples, which were cut into rectangular shapes, $5 \text{ cm} \times 1 \text{ cm}$ (4–6 measurements were performed for each sample). Mechanical testing machine parameters were as presented: initial force was 0.1 MPa; speed starting position was 50 mm/min; the speed of the initial force was 5 mm/min. TestXpert II 2017 program was used to collect data and results were gathered as average values with standard deviation (SD). Statistical analysis using the Q-Dixon test was performed.

3.6 Scanning Electron Microscopy (SEM)

Three silk textiles samples were imaged by a scanning electron microscope (LEO Electron Microscopy Ltd., Cambridge, UK): undyed samples, dyed samples pre-treated with LMWCS, and dyed samples pre-treated with LMWCS following 4 h of UVC irradiation. Samples were covered with a gold layer prior to analysis. Images were recorded at $150 \times \text{and} 1500 \times \text{times}$ magnification.

4 Results

4.1 Colorimetric Measurements

For the dyed silk textiles samples, colorimetric measurements were performed. ΔE values for all of the dyed silk textiles with various additives are presented in Tables 2 and 3. Visualized colors of dyed silk textiles using a color converter are presented in Table 4. According to the ISO 11664–4:2019 standard, ΔE value greater than 5

Table 2 Values of $L^* a^* b^*$	Type of fabric	Silk textiles (60 g/m ²)							
dyed silk textiles samples (the mean values of three	Color of dye	Non- dyed	Red	Red	Red	Red	Red	Red	
uncertainty $\pm 5\%$)	Dye concentration [%]	_	1	1	1	1	1	4	
	Pre-treatment prior to dyeing	_	No	CGCS/CA	LMWCS/CA	No	No	No	
	Additives in the dyeing process	_	CGCS/CA	No	No	No	No	No	
	pH of dye bath	_	2.09	2.81	3.04	3.58	3.58	3.64	
	SH covering	_	-	-	_	-	SH	-	
	Values of parameter L	78.04	46.02	46.36	48.08	46.22	33.75	31.85	
	Values of parameter a	- 0.18	34.11	24.11	25.73	18.2	17.75	23.09	
	Values of parameter b	4.07	5.04	0.76	1.10	- 2.38	2.21	2.24	
	ΔE	-	46.93	40.06	39.72	37.31	47.82	51.75	

Table 3 ΔE values for dyed silk textiles samples, dyed silk textiles samples following exposure to UVC irradiation, and dyed silk textiles samples following 6 months of storage (the mean values of three measurements; measurement uncertainty $\pm 5\%$)

Type of fabric		Silk textiles (60 g/m ²)							
Color of dye	Non- dyed	Red							
Dye concentration [%]	_	1	1	1	1	1	4		
Pre-treatment prior to dyeing	_	No	CGCS/ CA	LMWCS/CA	No	No	No		
Additives in the dyeing process	_	CGCS/CA	No	No	No	No	No		
pH of dye bath	_	2.09	2.81	3.04	3.58	3.58	3.64		
SH covering	_	-	_	_	-	SH	-		
ΔE [0.5 h irradiation]	1.07	3.54	4.78	3.38	4.08	1.33	5.84		
ΔE [2 h irradiation]	2.31	7.11	8.71	9.54	6.96	2.97	7.35		
ΔE [4 h irradiation]	3.99	9.64	13.67	11.94	9.75	4.91	10.41		
ΔE [6 months of storage]	-	5.67	5.57	4.75	5.12	2.96	4.37		

Table 4Visualized colors ofdyed silk textiles samples by thecolor converter

Non-dyed silk textiles								
Dved silk textiles								
Red colorant (4%)	Red colorant (1%)	Red colorant (1%)/ SH	Red colorant (1%)/ CGCS addition	Red colorant (1%)/ CGCS pre-treatment	Red colorant (1%)/ LMWCS pre-treatment			

is considered as a significant color change. Based on this state, the color change following 30 min of UVC irradiation was only observed for samples dyed with a colorant of 4% concentration. Following 2 h of UVC irradiation, the color changed for the dyed silk sample (1% dyed concentration), the dyed silk sample with the addition of CGCS, and the dyed silk sample pre-treated prior to dyeing with both grades of chitosan. The color of dyed samples covered with shellac solution did not change significantly also following 4 h of UVC irradiation. Following 6 months of storage, the color was the most stable for dyed samples pre-treated with LMWCS, dyed samples with a colorant of 4% concentration, and for dyed samples covered with shellac solution.

4.2 ATR-FTIR Spectroscopy

Infrared spectra were registered for either undyed silk, and dyed silk samples with various additives following 0, 0.5, 2, and 4 h of exposure to UVC irradiation. The ATR-FTIR spectra of the silk textiles samples examined are shown in Figs. 1 and 2.

Wavenumbers for IR characteristic bands of undyed and dyed silk samples following 0, 0.5, 2, and 4 h of exposure to UVC irradiation are presented in Table 5. FTIR spectra had shown that as a result of dyeing with red colorant in two different concentrations, characteristic bands' positions remained unaltered. For all of the dyed silk samples with chitosan and shellac, the amide II band position was slightly shifted to lower wavenumbers, also for dyed silk with CGCS addition, CH₂ and amide III bands positions were little shifted (to higher and lower wavenumbers respectively). As a result of UVC irradiation for dyed silk (1% colorant concentration), the amide I band was shifted to higher wavenumbers and the CH₂ band to lower wavenumbers, while for dyed silk with CGCS addition, amide I and CH₂ bands were shifted to lower wavenumber and for dyed sample pre-treated with CGCS the position of CH₂ band was shifted to higher wavenumbers. The dyed sample pre-treated with LMWCS showed that amide A band position was shifted to lower wavenumbers, and the dyed sample with SH covering amide A and amide II bands positions was shifted to lower and higher wavenumbers, respectively. Only silk textiles samples dyed with a 4% red colorant concentration characteristic bands positions remained unaltered following UVC exposure.



Fig. 1 Infrared spectra (from the top) of undyed silk textiles, dyed silk textiles with a colorant concentration of 1%, and dyed silk textiles with colorant's concentration of 4% from 4000 to 500 cm.⁻¹



Fig. 2 Infrared spectra (from the top) of dyed silk textiles with CGCS addition, dyed silk textiles pre-treated with CGCS, dyed silk textiles pre-treated with LMWCS, and dyed silk textiles covered with shellac solution from 4000 to 500 cm. $^{-1}$

Table 5 Wavenumbers for characteristic bands position of undyed and dyed silk textiles samples following 0, 0.5, 2, and 4 h exposure to UVC irradiation

Sample	Time of UVC radiation	Characteristic bands (cm ⁻¹)					
		Amide A	CH ₂	Amide I	Amide II	δ(CH ₂)	Amide III
Non-dyed silk	0 h	3279	2925/2853	1621	1516	1442	1228
	0.5 h	3276	2920/2849	1622	1515	1441	1229
	2 h	3281	2917/2851	1626	1512	1445	1229
	4 h	3279	2929	1624	1511	1444	1227
Red-dyed silk (1% of dye)	0 h	3276	2929	1617	1514	1441	1226
	0.5 h	3277	2926/2854	1622	1509	1443	1227
	2 h	3273	2923/2852	1621	1512	1441	1229
	4 h	3272	2921/2851	1625	1513	1442	1229
Red-dyed silk (4% of dye)	0 h	3278	2922/2855	1622	1514	1441	1228
	0.5 h	3275	2924/2853	1624	1509	1443	1228
	2 h	3280	2921/2852	1626	1509	1441	1230
	4 h	3280	2924/2851	1621	1514	1442	1228
Red-dyed silk (CGCS/CA)	0 h	3277	2931	1625	1511	1442	1223
	0.5 h	3277	2926/2855	1616	1515	1440	1221
	2 h	3274	2925/2853	1615	1514	1440	1230
	4 h	3274	2923/2855	1617	1515	1440	1226
Red-dyed silk (pre-treatment CGCS/CA)	0 h	3277	2924/2851	1623	1509	1446	1228
	0.5 h	3273	2923/2852	1620	1515	1441	1228
	2 h	3275	2928/2854	1621	1509	1443	1226
	4 h	3275	2926/2857	1623	1509	1444	1228
Red-dyed silk (pre-treatment LMWCS/CA)	0 h	3283	2922/2852	1623	1511	1442	1228
	0.5 h	3274	2921/2851	1622	1514	1440	1229
	2 h	3274	2920/2849	1621	1514	1442	1223
	4 h	3273	-	1623	1509	1443	1227
Red-dyed silk (SH covering)	0 h	3282	2927/2863	1624	1508	1442	1230
	0.5 h	3277	2927/2859	1623	1514	1445	1229
	2 h	3278	2929/2852	1618	1514	1441	1229
	4 h	3273	2926/2860	1620	1515	1441	1229



Fig. 3 Young modulus of undyed and dyed silk textiles samples



Fig. 4 Elongation at break of undyed and dyed silk textiles samples

4.3 Mechanical Properties

Young modulus values for dyed silk textiles samples are shown in Fig. 3. For all of the samples tested, the results obtained along the warp direction were significantly higher than along the weft direction. Compared to the undyed samples, slightly higher Young modulus values were obtained for dyed silk textiles samples without any additives and dyed samples with the addition of chitosan, whereas lower ones were for samples pre-treated with both types of chitosan and samples covered with shellac solution. Values of elongation at break parameter for dyed silk textiles samples are shown in Fig. 4. Compared to the undyed samples, dyed samples and dyed samples pre-treated with both types of chitosan were characterized with higher values for elongation at break. Whereas for dyed samples with the addition of chitosan and those covered with shellac solution, there were no significant differences among results, and all of the obtained values were lower than for undyed samples. For undyed and pre-treated with both types of chitosan samples, slight differences between values along warp and weft directions could be observed.



Fig. 5 Tensile strength of undyed and dyed silk textiles samples

Results for the tensile strength parameter for dyed silk textiles samples are shown in Fig. 5. Values of tensile strength are significantly higher along the warp direction for all of the silk textiles samples tested. When analyzing results, one could observe that for dyed samples without any additives, values were significantly higher than those obtained for undyed ones, whereas for undyed samples and dyed samples with the addition of chitosan, the obtained values were similar. For both pre-treated dyed samples, the values of tensile strength were similar to each other and lower than for undyed samples. Lower values compared to the undyed samples were also obtained for dyed samples covered with shellac solution.

4.4 SEM

To study the fiber morphology of undyed silk textiles sample, dyed silk textiles sample pre-treated with LMWCS and dyed silk textiles sample pre-treated with LMWCS following 4 h of exposure to UVC irradiation SEM microscopy were performed. In Fig. 6, SEM images of the tested samples are presented $(150 \times \text{and } 1500 \times \text{times of magnification})$.

5 Discussion

The colorant of plant origin used in this research dyed the silk textiles with satisfactory results in both applied concentrations and with additives. The addition of chitosan during the dyeing process, in general, did not improve the colorfastness of dyed silk textiles samples following exposure to UVC irradiation, but dyed samples pre-treated with low molecular weight chitosan were characterized by improved colorfastness during storage, whereas shellac covering improve color stability of irradiated and stored samples.

Registered FTIR spectra indicated that the red colorant did not affect the chemical structure of silk textiles. In the spectra of dyed samples with the addition of chitosan and shellac, amide II, amide III, or CH_2 bands positions were shifted, and under the influence of UVC irradiation, characteristic band wavenumbers were also altered. In Fig. 7, a scheme presenting the potential interaction of silk textile with chitosan and colorant of plant origin is shown.

The results of mechanical properties measurements indicated that the dyeing process with colorant caused greater flexibility and tensile strength of silk textiles. There were no significant differences in mechanical properties of dyed samples pre-treated with two types of chitosan and those samples were characterized by lower tensile strength and Young modulus but higher elongation at break than undyed silk textiles and thus were more flexible. However, differences were observed among chitosan applications; the addition of the chitosan to dye bath during the dyeing process caused greater stiffness and tensile strength of silk textiles than pre-treatment with chitosan prior to the dyeing process. Shellac covering also influenced mechanical properties; samples were stiffer but presented lower tensile strength than undyed ones.

It is commonly known that the maintaining colorfastness and functionality of the fabrics dyed with natural colorants had been the major constraint of natural dye in modern textile practices. The research results published Fig. 6 SEM images of silk textile samples (on the left- $150 \times \text{times}$ of magnification; on the right- $1500 \times \text{times}$ of magnification), from top to bottom: (a) and (b) undyed sample; (c) and (d) red-dyed sample pretreated with LMWCS; (e) and (f) red-dyed sample pre-treated with LMWCS after 4 h of exposure to UVC irradiation



by Wang et al. [65] show that still, several research groups try to find new possibilities to dye textiles in an ecofriendly way. For example, a natural dye was extracted from wall nut tree wood and it was used for dyeing silk textiles. It was found that natural wall nut tree wood dye also showed excellent UV protection properties. Next, curcumin was studied as a colorant for silk fibers. The dyeing of silk fabric carried with curcumin, extracted from turmeric was studied by Palaskar et al. [66]. The curcumin-dyed silk samples showed antioxidant activity and good ultraviolet protection depending on the percentage of curcumin used. Excellent dyeing properties of a natural dye extracted from the leaves of *Phoenix dac-tylifera* Linn on cotton and silk fabrics have been shown by Hossain et al. [67]. Comparing our results with the existing knowledge about the applications of natural Fig. 7 Scheme presenting the potential interaction of silk textile with chitosan and colorant of plant origin



colorants to dye silk textiles, there is no doubt that our results could be attractive from a practical point of view for eco-friendly dyeing.

6 Conclusion

Natural colorant applied in this study has the potential to dye silk textiles with satisfactory results. In the presence of chitosan and shellac gums, the colorant more intensely dyed silk as demonstrated through colorimetric measurements. Shellac and low molecular weight chitosan improve the color stability of dyed silk textiles samples. However, the colorfastness under the influence of UVC irradiation and during storage needs to be improved. In addition, the selected additives positively influence the mechanical properties of silk textile samples. Nevertheless, the use of appropriate natural additives on the one hand, and the development of suitable dyeing methods increasing colorfastness and stability, on the other hand, seem to be essential for plant-derived colorants to be considered as a relevant alternative to the use of synthetic ones. As environmental issues have nowadays become important, there are strong expectations to reduce

polluting materials used in textile production; thus, one can expect that new routes for dyeing textiles should be created.

Author contributions Conceptualization; PB and AS Data curation; PB Formal analysis; PB, MG and AS Funding acquisition; AS., PB Investigation; PB Methodology; PB and AS. Project administration MG and AS Resources; AS, PB Software; PB, AS Supervision MG and AS Validation; PB, MG and AS Visualization; PB Roles/Writing-original draft; PB Writing-review & editing AS and MG.

Data availability Not applicable.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

- 1. T.R. Sethy, J. Ahi, J. Nat. Fibers 19, 11492 (2022)
- A.U. Ude, R.A. Eshkoor, R. Zulkifili, A.K. Ariffin, A.W. Dzuraidah, C.H. Azhari, Mater. Des. 57, 298 (2014)
- S.U.D. Wani, S.P. Gautam, Z.L. Qadrie, H.V. Gangadharappa, Int. J. Biol. Macromol. 15, 2145 (2020)
- T. Bechtold, A. Turcanu, E. Ganglberger, S. Geissler, J. Cleaner Prod. 11, 499 (2003)
- L. Nambela, L.V. Haule, Q. Mgani, J. Cleaner Prod. 246, 119036 (2020)
- Y. Lam, S. Fan, Y. Chae, L. Wong, L. He, B. Fei, J.H. Xin, J. Cleaner Prod. 332, 130010 (2022)
- S. Baliarsingh, A.K. Panda, J. Jena, T. Das, N.B. Das, J. Cleaner Prod. 37, 257 (2012)
- A.K. Prusty, T. Das, A. Nayak, N.B. Das, J. Clean. Prod. 18, 1750 (2010)
- N. Rungruangkitkrai, R. Mongkholrattanasit, P. Phoophat, N. Chartvivatpornchai, S. Sirimungkararat, K. Wongkasem, P. Tuntariyanond, N. Nithithongsakol, R. Chollakup, I.O.P. Conf, Ser. Mater. Sci. Eng. **773**, 012027 (2020)
- 10. S. Yan, P. Shanshan, J. Junling, Nat. Prod. Res. 32, 308 (2018)
- 11. T.J. Manicketh, M.S. Francis, G. Joseph, Nat. Prod. Res. 36, 1 (2021)
- 12. Y. Yin, J. Jia, T. Wang, C. Wang, J. Cleaner Prod. 149, 673 (2017)
- V. Narayana Swamy, K.N. Gowda, R. Sudhakar, J. Inst. Eng. (India): E 97, 31 (2016)
- S.M. Geelani, S.J.A. Bhat, G. Qazi, S. Qasba, S.N. Qureshi, A.A. Parray, P.K. Mishra, S. Ara, A.H. Sofi, P.A. Khan, J. Ind. Pollut. Control 31, 227 (2015)
- V. Narayana Swamy, K.N. Gowda, R. Sudhakar, Int. J. Pharm. Biol. Sci. 4, B94 (2013)
- 16. P. Tayade, R. Adivarekar, Int. J. Chemtech Res. 5, 699 (2013)
- 17. J. Moses, N. Ravi, Man-Made Text. India 46, 295 (2003)
- S. Yadav, N.M. Rose, S.S.J. Singh, Asian J. Home Sci. 8, 250 (2013)
- 19. R. Tiwari, S. Paul, Int. Dye. 197(1), 35 (2012)
- S. Kiran, S. Adeel, M.S. Yousaf, N. Habib, A. Hassan, M.A. Qayyum, M. Abdullah, Ind. Textila 71, 227 (2020)
- 21. E. Grover, A. Sharma, B. Rawat, S. Paul, S. Jahan, Int. Dye. **190**(10), 9 (2005)
- L. Meyanci Özer, R. Karadag, E. Torgan, Tekstil ve Mühendis 23(103), 197 (2016)
- L. Chakraborty, P. Pandit, S. Roy Maulik, J. Cleaner Prod. 245, 118921 (2020)
- 24. H. Wang, P. Li, W. Zhou, J. Text. 2014, 587497 (2014)
- 25. Ph. Punyachareonnon, V. Deerattrakul, K. Luepong, Prog. Color. Color. Coat. 14, 179 (2021)
- W. Haddar, M. Ben Ticha, N. Meksi, A. Guesmi, Nat. Prod. Res. 32, 141 (2018)
- 27. T. Matsubara, H. Inoue, A. Satoh, J. Text. Eng. 65, 73 (2019)
- 28. P.S. Vankar, D. Shukla, J. Appl. Polym. Sci. 122, 3361 (2011)
- A. Yasukawa, A. Chida, Y. Kato, M. Kasai, Text. Res. J. 87, 2379 (2017)
- C.K. Yen, L.Y. Peng, C.X. Wang, Y.Y. Chen, Adv. Mater. Res. 418–420, 585 (2011)
- L. Peng, H. Mao, Y. Xiu, K. Zhang, C. Wang, Adv. Mater. Res. 441, 371 (2012)
- Y. Ren, J. Gong, R. Fu, J. Zhang, K. Fang, X. Liu, J. Cleaner Prod. 201, 648 (2018)

- 33. M. Chairat, S. Wisessombat, J. Text. Inst. 112, 207 (2020)
- A. Chimprasit, S. Hannongbua, P. Saparpakorn, J. Mol. Graph. Model. 106, 107934 (2021)
- 35. N.H. Erdawati, J. Fithriyah, Basic. Appl. Sci. Res. 3(1), 5 (2013)
- 36. T. Kako, A. Katayama, J. Seric. Sci. Jpn 66, 330 (1997)
- N. Kandasamy, K. Kaliappan, T. Palanisamy, Ind. Crops Prod. 171, 113969 (2021)
- 38. R.M. Kotb, J. Eng. Fibers Fab. 12, 60 (2017)
- 39. Y. Park, Text. Color. Finish. 31, 268 (2019)
- A.A. Shahin, S.A. Mahmoud, A.A. Ragheb, H.M. El-Hennawi, Egypt. J. Chem. 63, 3199 (2020)
- 41. L. Sheng, Adv. Mater. Res. 821–822, 369 (2013)
- 42. D. Shukla, P.S. Vankar, Fibers Polym. 19, 1913 (2018)
- 43. N. Talib, M.R. Ahmad, M.I.A. Kadir, K. Ismail, Int. J. Recent Technol. Eng. 8, 6897 (2019)
- 44. Y. Chao, T. Ho, Z. Cheng, L. Kao, P. Tsai, Fibers Polym. 18, 1523 (2017)
- S. Adeel, S. Kiran, M. Shahid, S. Habib, N. Habib, M. Hussaan, Environ. Sci. Pollut. Res. 29, 564 (2022)
- N. Amin, F. Rehman, S. Adeel, T. Ahamd, M. Muneer, A. Haji, Environ. Sci. Pollut. Res. 27, 6851 (2020)
- F. Batool, N. Iqbal, M. Azeem, S. Adeel, M. Ali, Pol. J. Environ. Stud. 28, 3081 (2019)
- N. Habib, S. Adeel, F. Ali, N. Amin, S. Khan, Environ. Sci. Pollut. Res. 28, 54041 (2021)
- N. Habib, W. Akram, S. Adeel, N. Amin, M. Hosseinnezhad, E. Haq, Environ. Sci. Pollut. Res. 29, 35048 (2022)
- 50. P.S. Vankar, R. Shanker, A. Verma, J. Cleaner Prod. 15, 1441 (2007)
- 51. P.S. Vankar, D. Shukla, S. Wijayapala, J. Textile Eng. Fashion Technol. 2, 274 (2017)
- P.S. Vankar, D. Shukla, S. Wijayapala, Pigment Resin Technol. 46, 296 (2017)
- M. Yang, Y. Yu, H. Zhai, J. Sheng, T. Xing, G. Chen, J. Text. Inst. 109, 1329 (2018)
- N. Mauliza, Y. Mardiyati, B. Purwasasmita, I.O.P. Conf, Ser. Mater. Sci. Eng. 105, 012035 (2016)
- S. Adeel, F. Rehman, M. Khosa, H. Aslam, M. Zuber, J. Ind. Eng. Chem. 77, 317 (2019)
- S. Adeel, K.M. Zia, M. Abdullah, F. Rehman, M. Salman, M. Zuber, Nat. Prod. Res. 33, 2060 (2019)
- 57. S. Adeel, F. Rehman, A. Hameed, N. Habib, S. Kiran, K.M. Zia, M. Zuber, J. Nat. Fibers **17**, 1 (2018)
- S. Adeel, M. Salman, S.A. Bukhari, K. Kareem, F. Rehman, A. Hassan, M. Zuber, J. Nat. Fibers 17, 635 (2020)
- S. Adeel, F. Rehman, K.M. Zia, M. Azeem, M. Kiran, M. Zuber, M. Irfan, M.A. Qayyum, J. Nat. Fibers 18, 136 (2019)
- S. Adeel, F. Rehman, M.K. Khosa, S. Rajab, K.M. Zia, M. Zuber, F. Batool, J. Nat. Fibers 19, 4684 (2021)
- M. Zuber, S. Adeel, F. Rehman, F. Anjum, M. Muneer, M. Abdullah, K.M. Zia, J. Nat. Fibers 17, 1410 (2019)
- T. Gulzar, S. Adeel, I. Hanif, F. Rehman, R. Hanif, M. Zuber, N. Akhtar, J. Nat. Fibers. 12, 494 (2015)
- F. Rehman, S. Adeel, R. Hanif, M. Muneer, K.M. Zia, M. Zuber, M.A. Jamal, M.K. Khosa, J. Nat. Fibers 14, 63 (2016)
- Nix Color Convertor. https://www.nixsensor.com/free-color-conve rter/ Accessed 4 August 2022
- S. Wang, L. Wang, M. Wu, K. Song, Z. Yu, J. Nat. Fibers 19, 11181 (2022)
- S.S. Palaskar, R.D. Kale, R.R. Deshmukh, Color Res. Appl. 46, 1301 (2021)
- S. Hossain, M.A. Jalil, A. Kader, S.A.B. Kamal, J. Nat. Fibers 19, 9803 (2021)