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Effects of various colorants on self-dyed silk properties: aspects of color, thermal stability, morphology, and degumming

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Abstract. The dyeing process in the textile industry consumes significant water and generates substantial wastewater. This study aims to develop and characterize self-dyed silks through feeding techniques using various natural and synthetic colorants, reducing environmental risks and enhancing silk's value. Bombyx mori was fed mulberry leaves dyed with Nanocurcumin from turmeric, dyestuffs from Caesalpinia sappan, Acid Red 88, and Basic Red 13. The colorimetric, morphological, and thermal properties of pristine and degummed self-dyed silks were analyzed using CIELab color, color strength (K/S), scanning electron microscopy (SEM), and thermal gravimetric analysis (TGA). Acid Red 88 showed superior coloring performance compared to natural and basic dyes. The study revealed that pigments were mainly retained in the sericin layer rather than fibroin after degumming, with notable changes to the silk's surface morphology and thermal properties. By proposing a waterless and sustainable silk coloration method, this research addresses environmental and health concerns in dyeing while promoting greener solutions for long-term development. The impacts of degumming on self-dyed silks were also comprehensively evaluate.

Keywords: self-dyed silk, mulberry, degumming, characterization, colorant.

Classification numbers: 1.4.8, 2.3.2, 2.9.3.

1. INTRODUCTION

Silk, a natural fiber composed of the proteins fibroin and sericin, has been used in textiles for at least 5000 years and is a commonly used and color-required textile material [1]. The silk is a valuable substance with numerous beneficial properties that is utilized for a range of applications, most notably textiles, and fashion. The Silk is frequently colored to improve aesthetics using several ways such as printing or dyeing, with dyeing being the most commonly used and popular. More than 90 % of commercially manufactured silk is actually spun by the domesticated silkworm Bombyx mori, a monophagous insect whose diet consists of mulberry tree leaves [1]. Before spinning their cocoons, silkworms go through 5 instars, which takes roughly 28 days [1]. Hot air is used to dry cocoons, removing the pupae and preventing the moth from enclosing. Importantly, proper drying boosts both silk output and quality and is a recommended practice even in tropical places with year-round availability [1]. Reeling requires immersing the cocoons in hot water to weaken the sericin protein, which links the fibers together to create the thick cocoon shell. Softening allows brushes to locate and pull the end of the silk strand [1]. The process of degumming involves the complete removal of sericin, which is the silk gum that bonds the fibroin filaments together, in order to achieve a characteristic shiny, soft texture and other desirable properties. There are various sorts of silk that are commercially known and manufactured all over the world. However, most silk contains basic colors such as white (Mulberry silk, Eri silk), green (Tasar silk), and golden-yellow (Muga silk) [3]. As a result, there is a considerable demand for dyeing silk, particularly for environmentally friendly approaches.

The use of dyes to color silk also harms the environment. Water resources have been jeopardized and polluted as a result of the amount of water utilized, effluent, and untreated dye compounds. Many environmentally friendly coloring methods have been developed and used, including dyes derived from flowers, leaves, roots, barks, and so on [1, 4 - 6]. Nonetheless, ongoing research and improvement efforts are addressing drawbacks such as color instability, poor color fastness, and the utilization of highly toxic metal mordants that pose environmental hazards upon release [7, 8].

To address the labor-health and environmental impacts of the textile industry, global efforts are advancing in dyeing, functionalization, and bio-based material innovations, including fungal, bacterial cellulose, and protein-derived alternatives for applications in fashion, medicine, and technology [4, 5, 9]. However, limited research targets water consumption reduction and wastewater concerns in textile dyeing. The industry consumes significant water and generates large volumes of wastewater, with dyeing processes contributing approximately 16 % of the total wastewater [10 - 12]. Effluents often contain reactive dye residues, aerosols, and chemicals with high COD and BOD concentrations, creating hard-to-degrade compounds [11]. Each year, the textile sector discharges around 70 billion tons of wastewater, posing severe health and environmental risks and driving demand for sustainable alternatives [11]. A promising waterless coloration method is self-dyeing, achieved by feeding silkworms specially prepared mulberry leaves containing dye solutions [13, 14]. This technique reduces wastewater from conventional dyeing processes. During the fifth instar stage, silkworms consume mulberry leaves treated with dye solutions through immersion, spraying, or mixing with leaf powder feed [14].

Numerous studies on the silkworm self-dyeing method address challenges related to conventional dyeing and wastewater management. By feeding silkworms leaves infused with dyestuff, this method achieves nearly 100 % water savings compared to traditional techniques, particularly for dyes like Rhodamine B, Congo Red, Acid Orange G, and others [13 - 16]. The dye's structure significantly influences color strength values [15]. Beyond dyeing, self-modifying methods have been applied to enhance silk performance and functionality. Research focuses on altering silkworm diets with substances such as single-walled carbon nanotubes, graphene, graphene oxide, carbon-based nanomaterials, metal/metal oxide nanoparticles, and quantum dots [17 - 19]. These efforts aim to create self-modified silk fibers with superior mechanical properties, expanding their use in applications like reinforcement and surgical sutures. This innovative approach supports sustainable silk treatment and functionalization, promising an eco-friendly future for dyeing and finishing.

A study on the self-dyeing process in Bombyx mori silkworms highlighted the critical balance between water repellency and affinity in azo dyes for effective diffusion from the gut to the silk glands. The partition coefficient of dyes determines their selective binding to sericin or fibroin proteins, influencing dye distribution in cocoons and silk [13]. This eco-friendly method reduces dye-laden wastewater and pollution by integrating the dyeing process into the silkworm's biochemical pathway. The color intensity in cocoons can be regulated by adjusting dye concentrations in their diet, with no adverse effects on silkworm development [16, 20]. Structural derivatives of azo dyes served as model compounds, establishing key molecular parameters for producing internally colored silk economically [13]. Unlike previous studies, this research evaluated dyeing properties, color production, and the microstructural impacts of dyes on yarn before and after degumming, advancing new textile material development.

This study provides a scientific premise on the self-dyeing method of silk for various new natural that had not been tested before (Nanocurcumin from Turmeric powder, Caesalpinia sappan dyestuff) with the expectation of their compatibility with natural dyestuffs. Silk self-dyeing added value and significance to an eco-sustainable method of color creation. Furthermore, synthetic dyes (Acid Red 88, Basic Red 13) were also tested to expand the list of colorants applicable to this self-dyeing method. Of particular note, unlike the report on the molecular weight of dyes below 400 g/mol, which demonstrated a high color-producing capability for silk [13], the results in this study contributed to a clearer understanding of the role of molecular weight in the coloration effectiveness of the self-dyeing method. Besides, colorimetric, morphological, and thermal of these self-dyed silk fibers have been thoroughly evaluated via Commission on Illumination Lab (CIELab) color space, visible light scattering, and reflection, Scanning Electron Microscope (SEM), Thermal Gravimetric Analysis (TGA). In addition, the influences of silk degumming on these properties of self-dyed silk were also investigated and discussed.

2. MATERIALS AND METHODS

2.1. Materials

Chemicals: *Caesalpinia sappan* dyestuff was provided by Maiwa (Canada). Nanocurcumin derived from Turmeric powder was obtained from Faculty of Chemistry Engineering – Ho Chi Minh City University of Technology (Viet Nam). Acid Red 88 was purchased from Sigma-Aldrich (USA). Basic Red 13 was provided by Rifa (Korea).

Bombyx mori (Mulberry species) and mulberry leaves were supplied by Bao Loc's mulberry plantations (Lam Dong, Viet Nam).

2.2. Coloring mulberry leaves

Using the spraying technique, mulberry leaves were dyed with dyeing solutions such as *Caesalpinia sappan* dyestuff (286.28 g/mol), nanocurcumin (368.4 g/mol), Acid Red 88 (400.38 g/mol), and Basic Red 13 (389.36 g/mol) at a concentration of 2000 ppm. The liquor ratio was 1.5:1 (the weight in grams of mulberry leaves to the volume in milliliters of dyeing solution). These colorful leaves were line-dried until their moisture content was in equilibrium with the surrounding air.

2.3. Self-dyeing of silkworm

Since the fourth day of the fifth instar, colorful mulberry leaves have been fed to the Bombyx mori silkworms. There was a three-day feeding period. The collected cocoons were boiled for five minutes at 90 ºC before being reeled using Silk reeling M/C-KTR1 (Korea).

2.4. Characterization

2.4.1. Color analysis

Based on the Kubelka-Munk theory as shown in Equation 1 [21], the color strength (K/S) of self-dyed silk fibers was measured and estimated using an X-Rite Color i5 Spectrophotometer and Color iControl software (X-Rite, USA) in the wavelength range of 400 nm to 700 nm. Color difference ΔE represents the geometrical distance between the corresponding points (of the two samples) in the color space (the distance from the test sample and the analysis sample) [22, 23] which was computed as Equation 2. Where K, S, and R, respectively, stand for absorbance, scattering, and reflectance; ΔL indicates any difference in brightness and is denoted by (+) if the sample that reproduces is lighter (brighter) than the witness specimen, and by (–) if it is darker than this; ΔL determines the lightness/darkness difference between a sample and a standard; Δa determines the red/green color distinction between the standard and the sample; Δb determines the yellow/blue color difference between a standard and a sample [22].

$$\mathrm{K/S} = \frac{(1-\mathrm{R})^2}{2\mathrm{R}} \tag{1}$$

$$\Delta\mathrm{E} = \sqrt{\Delta\mathrm{L}^2 + \Delta\mathrm{a}^2 + \Delta\mathrm{b}^2} \tag{2}$$

2.4.2. Scanning electron microscope

Using a JSM-IT200 InTouchScope™ Scanning Electron Microscope (Japan), the morphology of self-dyed silk fiber was examined.

2.4.3. Thermogravimetric analysis

Thermogravimetric analysis (TGA) was conducted using a Mettler Toledo TG209 F1 thermometer (Netzch, Germany). The thermograms were produced at temperatures ranging from 25 to 1000 ºC in a nitrogen environment with a constant gas flow rate of 10 mL/min and a heating rate of 10 K/min.

3. RESULTS AND DISCUSSION

3.1. Coloration effectiveness of silk self-dyeing for artificial and natural colorants

Figure 1 provides images of silk samples through the self-dyeing process using natural and artificial colorants. Generally, as per visual observation (Figure 1), it is clear that natural colorants (Caesalpinia sappan, nanocurcumin) didn’t yield significant coloration effectiveness for cocoons of Bombyx mori via the self-dyeing process. On the contrary, in terms of artificial colorants, Acid Red 88 has prevailed over its counterparts in coloration capacity, followed by Basic Red 13. To elucidate the dyeing capabilities of silk for different dyes reports from prior studies highlighted molecular weight (around 400 g/mol) as a determining factor in the dye’s permeation through the silk gut into the glands within the silkworm’s body [16]. Concerning molecular weight, all the dyes used hardly exceeded but rather fluctuated around the 400 g/mol threshold, such as nanocurcumin (368.38 g/mol) [24], Acid Red 88 (400.38 g/mol) [25], and Basic Red 13 (389.36 g/mol) [26]. It can be inferred that the molecular weight of the dye is not an absolute determinant, as previously suggested, but a factor facilitating efficient absorption through the silkworm's silk gut. Studies highlight key factors in effective silk self-dyeing, including molecular weight (≤ 400 g/mol) and the balance of hydrophobicity (aromatic groups) and hydrophilicity (polar groups) of the silk cocoon.

Figure 1. Self-dyed cocoons with various artificial (Acid Red 88, Basic Red 13) and natural (*Caesalpinia sappan*, nanocurcumin) colorants.

Figure 2 illustrates the K/S curves of self-dyed silk samples before and after degumming, as well as in comparison with pristine silk. K/S was calculated based on the amount of light absorbed (K) and scattered (S) by the sample at a particular wavelength. The findings in this study were once again validated through the K/S color values of silks self-dyed with pre and post-curing dyes (Figure 2). The Acid Red 88 has demonstrated a good coloring effect on the self-dyeing process compared with most of the studied dyes of both natural and artificial sources. In addition, it can be seen that after degumming with MS and Na_2CO_3, the K/S values of all dyed silk samples were significantly reduced, which proves that the surveyed natural and artificial colorants are adsorbed and distributed mainly in the sericin layers.

Before degumming, the K/S curves of self-dyed silk samples Acid Red 88 (Figure 2c) showed a peak in the range of 500 - 550 nm, suggesting that this wavelength range was more effective in producing better color than other wavelengths outside the peak range. These peaks indicated the maximum absorption of light by the dye molecules, resulting in a more intense and evenly distributed color on the surface. However, for certain types of self-dyeing drugs such as *Caesalpinia sappan* (Figure 2a), nanocurcumin (Figure 2b), and Basic Red 13 (Figure 2d), no peak waves were observed, so it can be concluded that these samples did not produce any color during the self-dyeing process in the wavelength range of 400 - 700 nm. The K/S curves also showed a gradual decrease in K/S values as the wavelength increased from 400 to 700 nm. This decrease in K/S values indicated a decrease in the amount of color strength by the dye molecules at longer wavelengths. Prior to degumming, a notable peak was observed in Acid Red 88, with peak wavelengths ranging from 525 to 535 nm, respectively. This peak exhibited higher K/S values compared to the other self-dyed silk samples, indicating that the Acid Red 88 (with a peak of 0.73 ± 0.42 K/S at a wavelength of 530 nm) was more effective in coloring the silk prior to the removal of the sericin layer.

After degumming, the K/S curves of all self-dyed silk samples showed a significant decrease, indicating a decrease in the amount of dye molecules present in the silk samples. The K/S values of the self-dyed silk samples after degumming were similar to those of Pristine silk, indicating that the dye molecules were primarily located in the sericin layer of silk. The peaks of *Caesalpinia sappan* (Figure 2a), Acid Red 88 (Figure 2c), Basic Red 13 (Figure 2d), and nanocurcumin (Figure 2b) observed in the K/S curves before degumming gradually disappeared after degumming, indicating that the dye molecules were removed from the silk samples during the degumming process. The K/S values of *Caesalpinia sappan* (Figure 2a), Acid Red 88 (Figure 2c), Basic Red 13 (Figure 2d), and nanocurcumin (Figure 2b) were decreased to only about 0.2 to 0.4, much lower than before degumming.

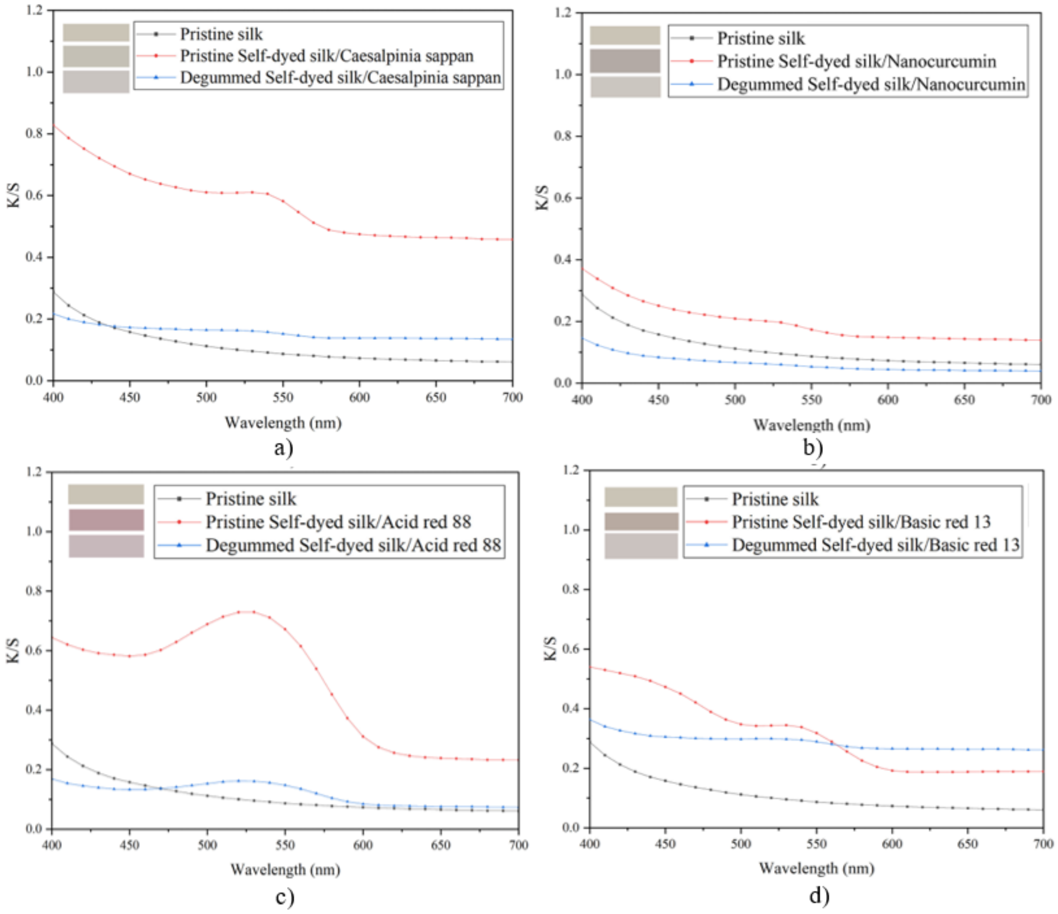

Figure 2. K/S spectra of self-dyed silk with *Caesalpinia sappan* (a), nanocurcumin (b), Acid Red 88 (c), Basic Red 13 (d) before and after degumming.

The results from Table 1 indicated that the L*, a*, b* values and the color difference (ΔE) of the self-dyed silk samples before and after degumming, compared to the undyed silk, demonstrated color variations. The findings also revealed differences in color between the silk samples before and after degumming. This indicates that the degumming process affects the color system of the samples, although the extent of change varied for each dye.

Prior to degumming, the nanocurcumin, *Caesalpinia sappan*, and Basic Red samples had similar average L* values, indicating that their brightness was approximately the same. Among them, *Caesalpinia sappan* had the highest brightness with a value of 80.22 ± 3.86, so the brightness of this sample stood out more compared to the other four samples. Acid Red 88 had a lower brightness (67.57 ± 2.23), this sample had the lowest brightness among the 4 dye samples. After degumming, the L* values of all samples increased significantly, indicating that the sericin coating had darkened the colors of the samples. Among them, the nanocurcumin sample had the highest (88.37 ± 0.66) and Acid Red 88 had the greatest increase in average L* value after degumming (83.22 ± 0.63). The nanocurcumin had the least and lowest increase in average L* value (76.79 ± 0.43), and the difference in L* between measurements of this sample was very large.

Table 1. L*, a*, b*, and ΔE measurement results with 4 dye samples before and after degumming.

	Before degumming			
	Caesalpinia sappan	Nanocurcumin	Acid Red 88	Basic Red 13
L*	80.22 ± 3.86	76.79 ± 0.43	67.57 ± 2.23	74.94 ± 0.98
a*	0.61 ± 0.22	2.49 ± 0.37	13.71 ± 0.77	4.02 ± 0.50
b*	6.45 ± 1.96	7.26 ± 2.15	2.33 ± 2.00	6.48 ± 0.43
ΔE	6.24 ± 3.60	9.60 ± 0.58	21.92 ± 1.74	14.27 ± 5.53
	After degumming			
L*	85.57 ± 1.40	88.37 ± 0.66	83.08 ± 0.88	82.10 ± 1.87
a*	1.19 ± 0.17	0.70 ± 0.16	5.58 ± 0.75	1.66 ± 0.70
b*	2.68 ± 0.38	4.78 ± 0.72	1.49 ± 0.62	3.33 ± 2.04
ΔE	5.16 ± 0.45	3.86 ± 0.48	8.59 ± 0.94	6.37 ± 1.25

Before degumming, the ΔE values of the samples were relatively large, ranging from 6.24 ± 3.60 to 21.92 ± 1.74, with Acid Red 88 being the most effective with a ΔE of 21.92 ± 1.74 and *Caesalpinia sappan* being the least effective with a value of 6.24 ± 3.60. However, after degumming, the ΔE values decreased significantly, ranging only from 3.86 ± 0.48 to 8.59 ± 0.94. Before degumming, Acid Red 88 was evaluated as having the best color efficiency, but after the degumming process, its color efficiency decreased significantly. This indicates that the color efficiency of the samples is very high when the sericin layer is not removed. The dyes mainly bond with the sericin layer, which is difficult to bond and penetrate deep into the fibroin.

Prior to degumming, the average a* color parameter of the 4 dye samples was located on coordinates of red colors with values ranging from 0.61 ± 0.22 to 13.71 ± 0.77. The a* parameter with high positive values reflected the predominance of red over green [27]. So, Acid Red 88 had the highest red color with an a* value of 13.71 ± 0.77, and the lowest was *Caesalpinia sappan* with a value of 0.61 ± 0.22. After degumming, the average a* values of the 4 samples tended to decrease but remained positive, indicating that the red color of the samples would become lighter.

Before degumming, the average b* value of the other samples was located on coordinates of yellow-blue colors with values ranging from 2.33 ± 2.00 to 7.26 ± 2.15. Since all b* values were positive, it could be concluded that, in the tested samples, yellow hues were dominant over blue hues (as mentioned in Figure 1). After degumming, the average b* values decreased to around 1.49 ± 0.62 to 4.78 ± 0.72. Combining L*, a*, and b* values, it can be seen that all 4 samples had a strong yellowish-red color before degumming, which became lighter after 4 rounds of degumming, with an increase in brightness. The degree of change varied for each sample, with Acid Red 88 being the most noticeably altered.

3.2. Surface morphological properties of self-dyed silk with natural and artificial colorants

Figure 3 shows the surface morphology of self-dyed silk samples before and after degumming at 1000 magnification.

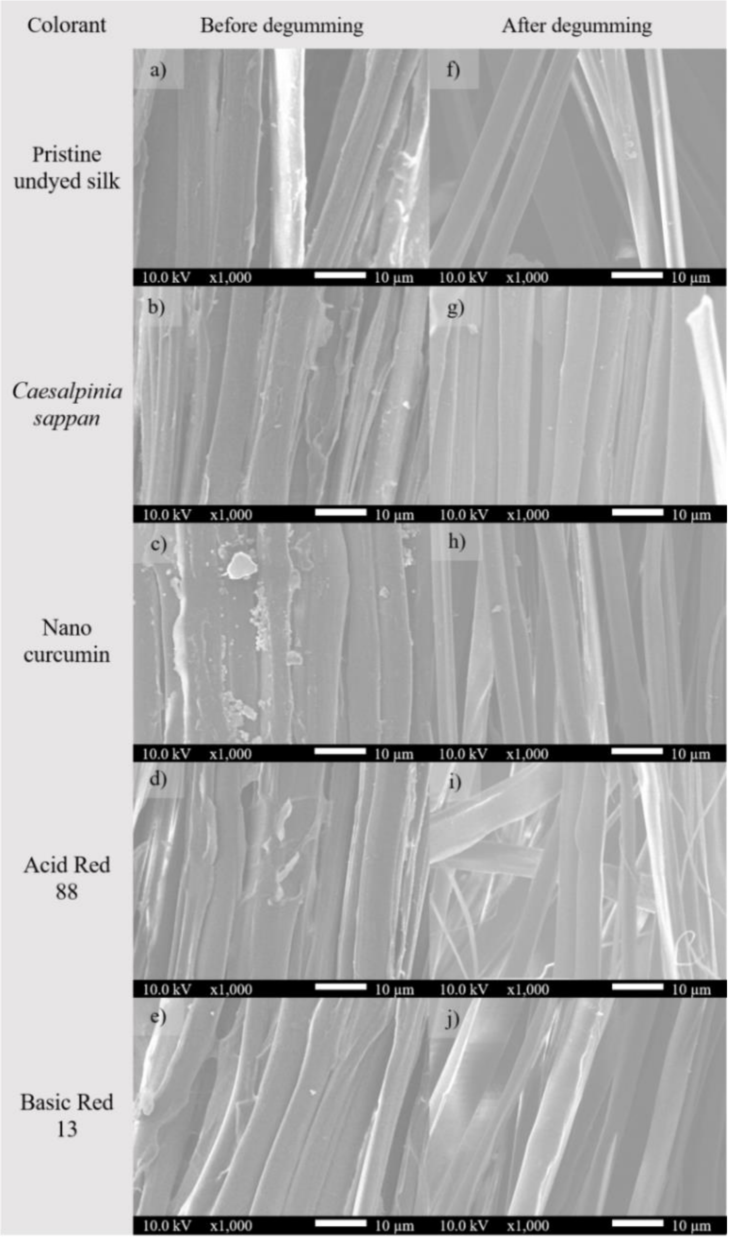

Figure 3. SEM images of silk samples before and after degumming.

In general, the surface morphology of silks before and after degumming did not change significantly displayed in Figure 3. However, for self-dyed silk before degumming (Figures 3.a,b,c,d,e), because most of the colorants are eliminated during biosynthesis in silkworms, the self-dyeing process with various colorants is said to be less efficient such as *Caesalpinia sappan*, nanocurcumin, and Basic Red 13 (Figures 3.b,c,e). Self-dyeing with these ineffective colorants did not produce any difference in the surface morphology of silk compared with pristine undyed silks (Figure 3a). However, for the effective colorant in self-dyeing as Acid Red 88, the surface morphology of silk changed remarkably, especially the discontinuity in the sericin layer (Figure 3d). Corresponding to the conclusion about the coloring efficiency of self-dyeing with various dyes, the higher the colorant is mainly distributed in the sericin layer, the higher the amount of colorant will cause the sericin layer to become disrupted.

After degumming (Figures 3.f,g,h,i,j), most of the sericin layer was removed, therefore silk surface is smoother with no obvious difference between self-dyed silk and conventional undyed silk. However, it can be observed that for the effective colorants in self-dyeing and Acid Red 88, many fibrils appear in the silk structure (Figures 3i), which could not be observed in either undyed silk or self-dyed silks with colorants with poor coloring efficiency (Figures 3.f,g,h,j). It can be seen that, although effective colorants such as Acid Red 88 are mainly distributed on the sericin layer, they still have a significant effect on the fibroin structure of silk during biosynthesis. The results of this morphological surface are analogous to a previous study [14]. Based on the findings, it can be concluded that the self-dyed process does not have a significant impact on the morphology of silk. This finding can pave the way for future studies on modifying the fibroin structure of silk for various applications.

3.3. Thermal stability of self-dyed silk with natural and artificial dyes

Figure 4 presents TGA diagrams of self-dyed silk samples before and after degumming that provided information on the thermal stability of self-dyed silks with various synthetic and natural colorants.

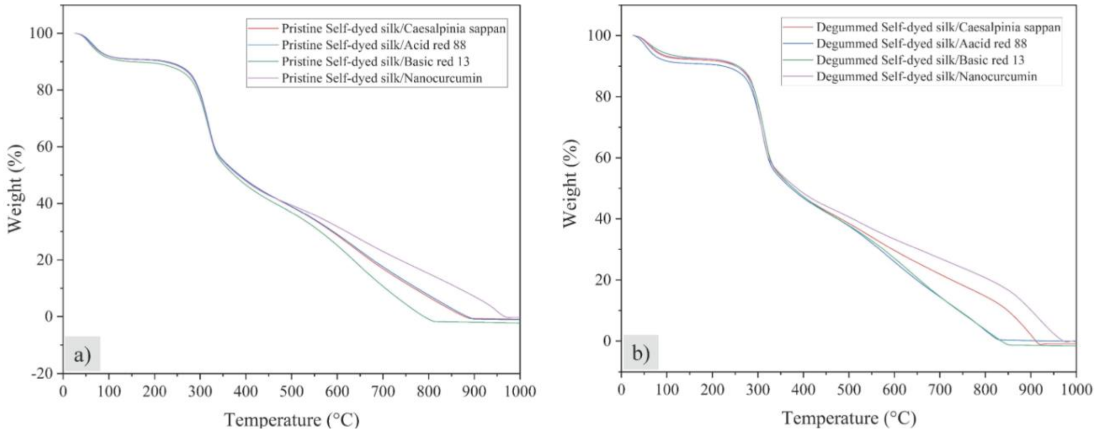

Figure 4. TGA diagrams of self-dyed silks before (a) and after (b) degumming.

TGA analysis confirmed that the self-dyeing of the above dyes changed the thermal properties of silk both before and after degumming. The thermal stability of the self-dyed silks is markedly different over the entire temperature range from about 25 to 1000 ºC. Most of the thermal stability between the self-dyed silk samples before and after degumming exhibited similar thermal properties below 350 ºC, and the difference was significantly different when the temperature was raised above 350 ºC. It can be said that this finding is related to various factors such as the amount of carbon and other substances, as well as changes in amino acids within silk. This relationship was previously demonstrated through the analysis of amino acid content in self-dyed silk compared to regular white silk [14]. Furthermore, the presence of various dye types and their individual characteristics is also a contributing factor to the differences observed in the thermal properties of various self-dyed silk threads and so forth due to the existence of different colored compounds in the biosynthesis of sericin and fibroin. More specifically, the main cause of the mass loss of silk fibers at temperatures below 100 ºC is water evaporation, while the bulk loss above 200 ºC is related to the loss of gases that have low molecular weight (H_2O, CO_2, and NH_3) [28]. These gases are attributed to the disruption of amino acid side chain residues and the cleavage of peptide bonds in the amorphous region of the silk [29]. The fibroin of Bombyx mori silk exhibits a degradation step with a maximum decomposition temperature of 350 ºC. However, there are differences in the thermal properties between raw silk (containing sericin) and degummed silk (sericin removed), with corresponding thermal decomposition steps at 325 - 350 ºC and 340 - 360 ºC for sericin-containing and sericin-free fibers, respectively. Variations in thermal properties are also evident among different post-degumming samples, possibly linked to the type of dye ingested by the silkworms along with alterations in the silk synthesis process during fiber formation. The results demonstrate chemical and physical changes in self-dyed silk samples using different dyeing methods. However, it's observed that silk doesn't completely decompose even at temperatures as high as 1000 ºC due to its characteristic semi-crystalline structural features and the phase transition kinetics from amorphous regions within the silk to crystalline after the heating process concludes [30]. Therefore, it can be said that the difference in thermal stability between the self-dyed silks is due to the quantitative change of carbon, nitrogen, oxygen, and hydrogen in the protein composition of the silk. Degradation of fibroin molecules has been recorded which can be observed at about 350 ºC - the decomposition temperature of Bombyx mori silk [30].

4. CONCLUSION

This research provides a foundation for self-dyeing silk using novel natural and synthetic colorants like nanocurcumin, Caesalpinia sappan, Acid Red 88, and Basic Red 13. Acid Red 88 showed the highest color efficiency, with a significant enhancement at 525 - 535 nm before degumming (K/S value: 0.73 ± 0.42). After degumming, brightness increased across all samples, with nanocurcumin,achieving the highest value (88.37 ± 0.66), while redness decreased, and yellowness rose. Acid Red 88 exhibited the highest color effectiveness (ΔE: 8.59 ± 0.94).

The self-dyeing process caused noticeable changes in silk morphology, particularly in the sericin layer, though dyes like Caesalpinia sappan, nanocurcumin,, and Basic Red 13 yielded no visible coloration. The study highlights the potential of self-dyeing to create sustainable textiles, reduce water consumption, and address environmental concerns. Further research is needed to optimize formulations, explore dye absorption mechanisms, and evaluate other dye types to enhance process efficiency and commercialization potential. This work advances sustainable silk dyeing and contributes to cleaner textile technologies.

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CRediT authorship contribution statement. Uyen Nguyen Tu Tran: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft preparation, Writing – review and editing. Hung Ngoc Phan: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft preparation, Writing – review and editing. Son Minh Ngoc Nguyen: Formal analysis, Investigation, Writing – original draft preparation, Writing – review and editing. Thao Thanh Hoang: Conceptualization, Writing – review and editing, Supervision. Huong Mai Bui: Conceptualization, Methodology, Writing – review and editing, Supervision.

Declaration of competing 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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