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# Eco-friendly dyeing of silk fabric by using *Artemisia argyi* as natural dye and *Phyllanthus emblica* as natural mordant

Lili Feng, \* Anyuan Ye, xuemei Yu and Zhihua Chen

Natural dyes commonly exhibit low dye uptake and inferior color fastness in textile applications, making the utilization of mordants necessary for enhancing dyeing performance. However, ecological toxicity and low biocompatibility are the drawbacks of traditional metal mordants. This study proposed an eco-friendly alternative by employing *Phyllanthus emblica* (PE) as natural mordant, and systematically explored its enhancement mechanism and mordanting effects on silk dyed with *Artemisia argyi* dye. The results demonstrated that the addition of PE extract under the dyeing conditions (pH 4, 90 °C, 30 min) significantly enhanced the color depth of silk fabric, as indicated by an increase in the *K/S* value from 2.82 to 5.05. The color fastness was also improved, with both washing and rubbing fastness increasing by approximately 0.5 grades. Furthermore, the dyed fabric exhibited excellent antibacterial activity, achieving a 93.03% bacteriostatic rate against *Staphylococcus aureus*. This study provides new insights for developing eco-functional textiles through sustainable dyeing processes.

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## 1. Introduction

In the modern textile industry, environmental preservation and sustainability have emerged as critical issues. Synthetic dyes, while providing bright colors and good color fastness, often contain hazardous chemicals that can harm both human health and the environment.<sup>1,2</sup> Natural dyes derived from plants, animals, and microorganisms present an environmentally sustainable alternative,<sup>3–5</sup> offering biodegradability, non-toxicity, and multifunctional properties including antioxidant, antibacterial, and anti-inflammatory effects.<sup>6–8</sup>

*Artemisia argyi* (*A. argyi*), a perennial herbaceous plant belonging to the Asteraceae family, represents one of the most promising natural dye sources among various plant-derived colorants. It is widely distributed across northern temperate regions, especially in Asia, Europe and North America.<sup>9,10</sup> This plant holds significant importance in traditional Chinese medicine, where its dried leaves and the whole plant are extensively utilized for clinical treatment and daily health care.<sup>11–13</sup> Moreover, *A. argyi* has demonstrated considerable potential in the field of textile since it is capable of simultaneous imparting coloration and biofunctionalization to textiles. Textiles treated with *A. argyi* exhibit significant antibacterial properties and UV resistance, facilitating their broad application in medical textiles and sportswear.<sup>14</sup>

However, natural dyes are organic macromolecules that primarily adhere to natural fibers through non-covalent interactions such as van der Waals forces and hydrogen bonds. This

weak binding mechanism result in dyeing disadvantages including low dye uptake and unsatisfactory color fastness.<sup>15,16</sup> To address these challenges, metal mordants have been conventionally employed in natural dyeing processes. However, the utilization of metal mordants raise substantial environmental concerns due to their ecological toxicity.<sup>17,18</sup> There is a growing need to search for alternative natural mordants with low toxicity for eco-friendly dyeing application. In recent years, various types of natural mordants have been explored as sustainable alternatives to conventional metal mordants.<sup>19</sup> These include plant-based mordants derived from tannin-rich sources like oak galls, pomegranate rinds, and sumac; non-vegetable mordants from animal or mineral sources such as chitosan and clay; and oil mordants like sulfonated castor oil.<sup>20</sup> These natural mordants function through mechanisms including chelation, cross-linking, and complex formation with both dye molecules and textile fibers, significantly improving dye uptake and color fastness while reducing environmental impact.

Among these alternatives, tannic acid, a polyphenol extracted from various plant tissues (roots, stems, leaves, and fruits), has shown particular promise as an eco-friendly alternative.<sup>21,22</sup> Its effectiveness can be attributed to a dual mechanism: the abundant phenolic hydroxyl groups form strong hydrogen bonds with textile fibers, anchoring the mordant firmly to the substrate, while its polyphenolic structure enables chelation and complex formation with dye molecules. As a result, tannic acid not only enhances dye adsorption and color fixation but also exhibits broad compatibility with a variety of natural dyes and textile materials.<sup>23</sup> Its renewable nature and biodegradability make it an ideal candidate for sustainable textile

College of Textile and Apparel, Quanzhou Normal University, Quanzhou, Fujian, China. E-mail: jll827@qztc.edu.cn



production, aligning with the growing demand for environmentally dyeing process in the textile industry. Moreover, tannic acid demonstrates remarkable biological activities including potent antioxidant, antiviral, and antibacterial properties, which can impart functional value to treated textiles.<sup>24–26</sup>

*Phyllanthus emblica* (*P. emblica*), a significant medicinal horticultural plant, is notably rich in tannic acid, particularly in its bark and fruits.<sup>27,28</sup> In this study, tannic acid extracted from *Phyllanthus emblica* fruits was employed as natural mordant for silk fabric dyed with *A. argyi*. The dyeing process was systematically investigated, and the mordanting mechanism was further elucidated. Additionally, antibacterial activity of the dyed silk fabrics was characterized to evaluate their potential for developing sustainable functional textiles.

## 2. Materials and methods

### 2.1 Materials

*A. argyi* power and fresh *P. emblica* were purchased from a local agricultural trader. Silk fabric weighting  $33\text{ g m}^{-2}$  was purchased from Hangzhou Furan Silk Co., Ltd. *Escherichia coli* ATCC 29522 and *Staphylococcus aureus* ATCC 6538 were provided by American Type Culture Collection. Yeast powder and peptone were purchased from Sangon Biotech (Shanghai) Co., Ltd. The chemical reagents were obtained from Aladdin Reagent Co., Ltd.

### 2.2 Methods

**2.2.1 Preparation of *A. argyi* dye and *P. emblica* extract.** The *A. argyi* dye was naturally extracted by employing ethanol as the

solvent. With a solid-to-liquid ratio of  $1\text{ g } 10\text{ ml}^{-1}$ , the extraction procedure maintained at a temperature of  $50\text{ }^{\circ}\text{C}$  for 50 min. Following filtering, the *A. argyi* dye solution was named as AA dye.

The preparation of the *P. emblica* extract involved the following procedure. The *P. emblica* fruits were first subjected to thorough washing and cut into small pieces, which were then mixed with distilled water at a solid-to-liquid ratio of  $1\text{ g } 10\text{ ml}^{-1}$ . The mixture underwent mechanical disruption using a high-speed blender to facilitate the release of intracellular components. The resulting mixture was filtered and centrifuged at 7000 rpm for 10 min to remove insoluble impurities. After centrifugation, the suspension named PE extract was carefully collected and preserved for utilization in the subsequent experiments (Fig. 1).

**2.2.2 Dyeing silk with AA dye and PE extract.** The PE extract was employed as natural mordant in the dyeing of silk fabrics with AA dye and the simultaneous mordanting method was employed to evaluate the mordanting effect. Additionally, related dyeing parameters including the amount of AA dye (2.5–10 ml) and PE extract (0–30 ml), pH (2–6), dyeing temperature ( $45\text{--}90\text{ }^{\circ}\text{C}$ ) were optimized to explore the suitable dyeing process. For subsequent experiments, the following representative treatments were selected: undyed silk fabric (WS), silk fabric dyed with only AA dye (AS), silk fabric dyed with AA dye and PE extract (APS), silk fabric dyed with AA dye and PE extract under pH 2–6 (APS2–APS6).

### 2.3 Characterization

**2.3.1 UV-vis analysis.** The spectral characteristics of the obtained PE extract was analyzed using a Lambda 365 UV-vis

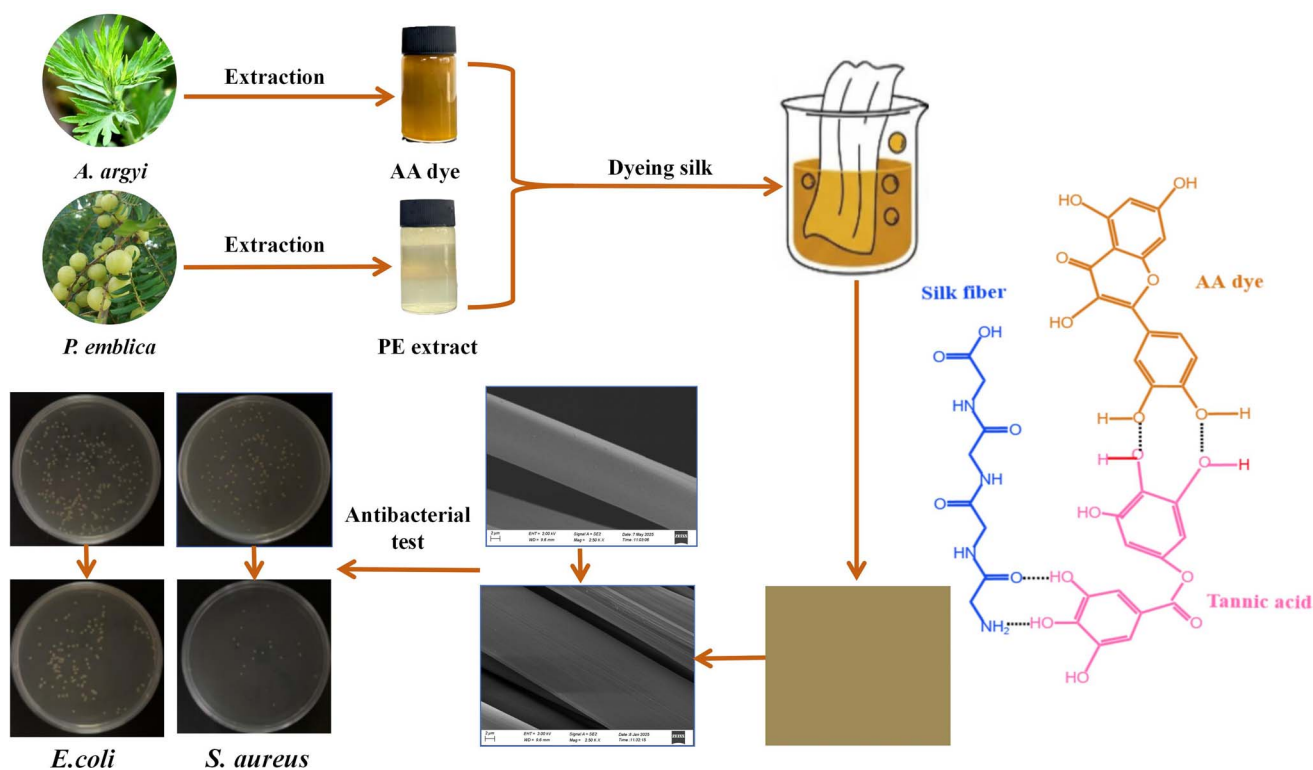


Fig. 1 The experimental process of dyeing silk fabric with AA dye and PE extract.

spectrophotometer (PerkinElmer, USA) within the visible light spectrum ranging from 200 to 700 nm.

**2.3.2 Scanning electron microscopy (SEM).** The morphology of silk samples was analyzed using a scanning electron microscope (Phenom Pharos, USA). Before SEM analysis, all samples were sputter-coated with gold and the pictures were observed under 2500 magnification.

**2.3.3 Fourier transform infrared (FTIR) spectra.** The infrared spectra of samples were tested using the Nicolet iS10 Infrared Raman spectrometer (Thermo Fisher, USA) with the wavelength range of 4000–800  $\text{cm}^{-1}$ .

**2.3.4 X-ray photoelectron (XPS) spectra.** The XPS spectra of fabrics was measured using a PHI 5000 VersaProbe III X-ray photoelectron spectrometer (ULVAC-PHI, Japan) with a monochromatized Al K $\alpha$  source operated at 15 kV and 50 W.

**2.3.5 Color characteristics.** The color characteristics of dyed silk fabrics were tested using a Datacolor 800 spectrophotometer (Datacolor company, USA) under illuminant D<sub>65</sub>. The test result was the average of observed values from five different positions.

**2.3.6 Color fastness.** The rubbing and washing fastness of dyed silk samples were evaluated according to ISO 105-X12 and ISO105-C10, respectively.

**2.3.7 Antibacterial property.** Samples were tested for antibacterial properties against *S. aureus* and *E. coli* in accordance with the textile antibacterial standard (GB/T 20944.3-2008). The detailed experimental protocol followed the published literature.<sup>29</sup>

## 3. Results and discussion

### 3.1 Characterization of PE extract and AA extract

As shown in Fig. 2a, the PE extract exhibited absorption peaks around 212 nm and 276 nm, which corresponded to the absorption peaks of tannic acid.<sup>30</sup> This indicated that YE extract contained a rich amount of tannic acid. Meanwhile, the absorption spectra showed a highly pH sensitivity. A gradually decrease in the absorption peak at 212 nm was observed as the pH increased from 1 to 7. Additionally, in alkaline circumstances, an absorption peak appeared at about 320 nm. This

might be due to the presence of hydroxyl groups on the tannic acid molecules, which deprotonated and produced oxoanions ( $\text{O}^-$ ) in alkaline conditions. It thus enhanced the conjugation of the molecule and the delocalization range of the  $\pi$ -electrons, causing the redshift from 280 nm to 320 nm.

The infrared absorption spectra of PE power and AA power were characterized to further elucidate their molecular structures and functional groups. As shown in Fig. 2b, a broad peak around 3408  $\text{cm}^{-1}$  corresponds to the O–H stretching vibrations of phenolic hydroxyl groups, indicating the presence of numerous hydroxyl groups in the structure of PE power. A peak around 1726  $\text{cm}^{-1}$  can be attributed to carbonyl bond ( $\text{C}=\text{O}$ ) in the ester group.<sup>31</sup> The aromatic  $\text{C}=\text{C}$  stretching vibrations are responsible for additional peaks at about 1631  $\text{cm}^{-1}$ , which validate the existence of the benzene ring.<sup>32–34</sup> A peak around 1057  $\text{cm}^{-1}$  corresponds to the C–O stretching vibrations of the ester groups, further characterizing the functional groups within tannic acid.<sup>35</sup> In contrast, the spectrum of AA power also exhibits a broad O–H stretching band around 3405  $\text{cm}^{-1}$  and aromatic  $\text{C}=\text{C}$  vibrations at 1631  $\text{cm}^{-1}$ . However, the absence of a carbonyl peak at 1726  $\text{cm}^{-1}$  distinguishes it from tannic acid, indicating a lack of typical ester groups. Instead, the structure of AA dye is further evidenced by a peak at 1404  $\text{cm}^{-1}$ , associated with O–H in-plane bending or aromatic ring vibrations, and a characteristic aromatic C–H out-of-plane bending vibration at 618  $\text{cm}^{-1}$ ,<sup>36</sup> which suggests a specific flavonoid-like aromatic substitution pattern.

### 3.2 Dyeing silk fabric with PE extract

Fig. 3a illustrates the variations in  $K/S$  values and apparent color of silk fabrics dyed with AA dye and PE extract. It is evident that the dyed fabrics exhibited a relatively low  $K/S$  value of 2.38 when PE extract was not present. The  $K/S$  of the dyed fabrics enhanced corresponding to the increased addition of PE extract, reaching a maximum value of 3.01 at 20 mL PE extract addition. This may be attributed to the presence of tannic acid in the PE extract, which formed a complex with the AA dye, facilitating the fixation of the dye onto the fiber and thereby deepening the color.

Given the critical role of pH value in the dyeing process of protein fibers, the influence of pH was also investigated in this

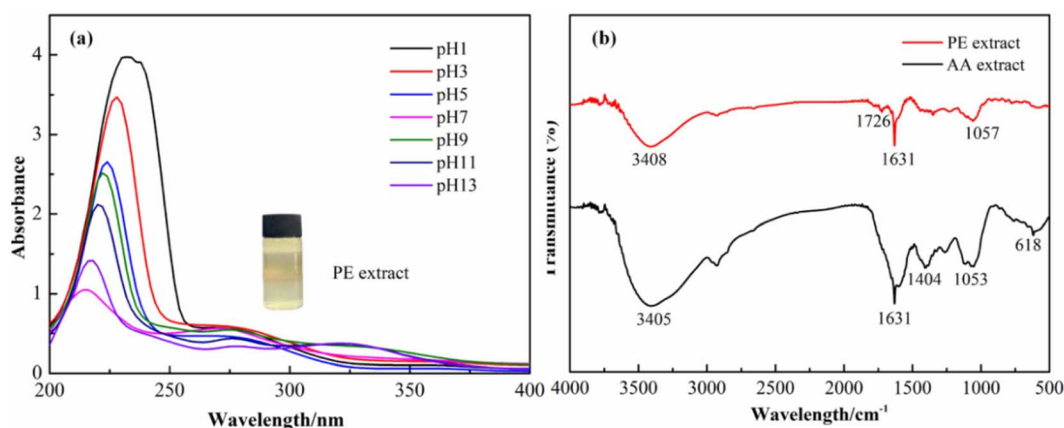


Fig. 2 UV-vis spectra of PE extract (a), FTIR spectra of PE power and AA power (b).



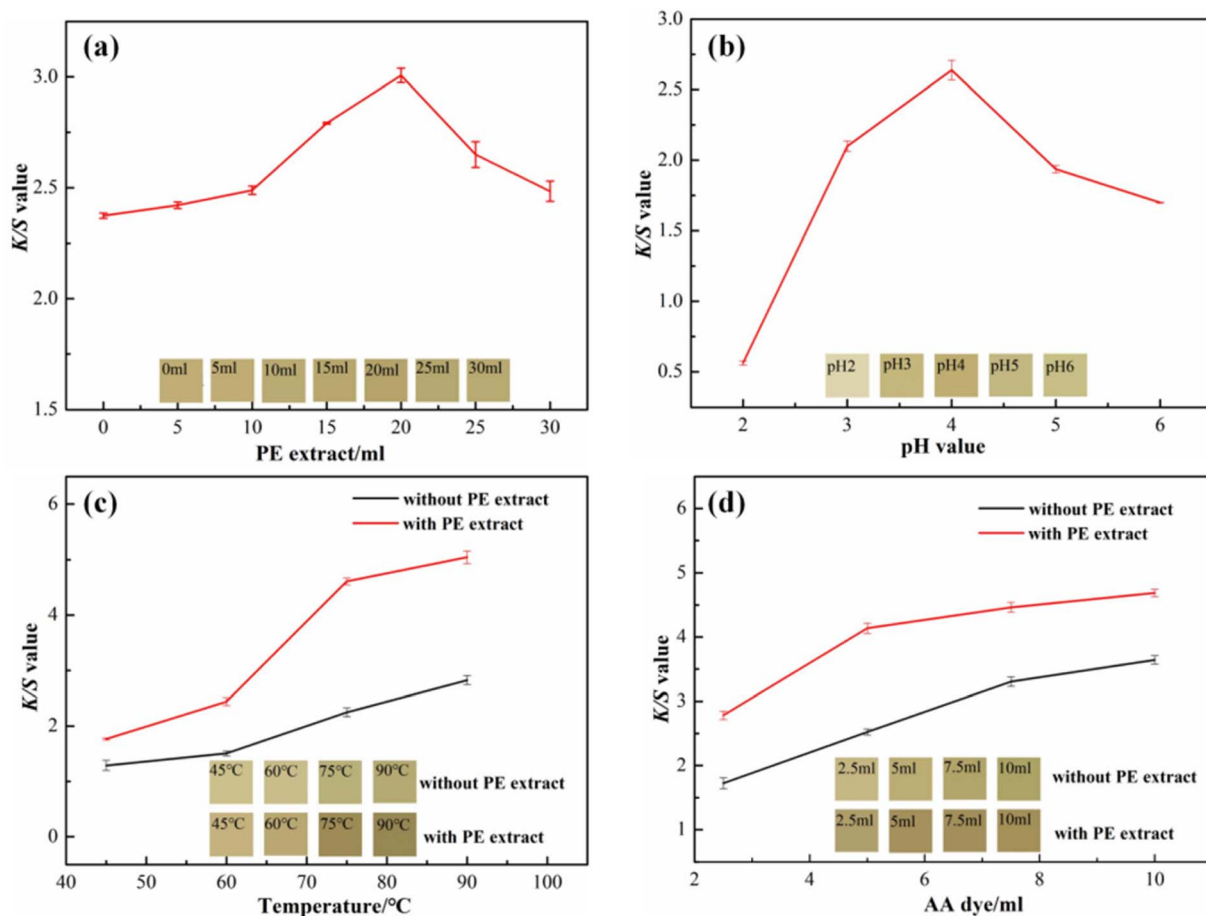


Fig. 3 The influence of PE extract concentration (a), pH value (b), dyeing temperature (c), AA dye concentration (d) on  $K/S$  value of dyed silk fabric.

study. An acidic range of pH 2–6 was chosen for the studies in order to avoid the reduction in silk strength caused by an excessively acidic environment. As shown in Fig. 3b, the  $K/S$  was lowest at pH 2 with a value of 0.56. Further increase in pH value led to a significant enhancement in the  $K/S$  value, reaching a maximum at pH 4. Conversely, a gradual decline in  $K/S$  values was observed when the pH exceeded 4. The observed color depth variation can be attributed to pH-dependent changes in the surface charge of both tannic acid and silk, which directly modulate their electrostatic interactions. Under acidic conditions ( $\text{pH} < 4$ ), the carboxyl groups of tannic acid remain largely protonated and electrically neutral,<sup>37</sup> resulting in diminished electrostatic attraction toward the protonated amino groups of silk. This leads to weak binding affinity and poor mordanting efficacy. In contrast, at pH 4, tannic acid undergoes partial deprotonation and acquires a net negative charge, while silk maintains a positive surface charge. The resulting strong electrostatic attraction promotes effective adsorption of tannic acid onto the silk fiber, thereby improving the subsequent binding of natural dye and color depth. At pH values above 5, however, both tannic acid and silk exhibit negative surface charges. The resulting electrostatic repulsion hinders adsorption, leading to reduced mordanting efficiency and a corresponding decline in the  $K/S$  value of the dyed silk fabric.

Since the stability of AA dye and strength of silk fibers will be impacted by high temperatures, the dyeing temperature was set within the range of 45–90 °C. As depicted in Fig. 3c, the addition of PE extract exerted a minimal impact on color depth at lower dyeing temperatures below 60 °C. Nevertheless, a significant rise in the  $K/S$  value and a marked enhancement in the mordanting effect were observed as the dyeing temperature increased above 60 °C. These results indicated that the dyeing of silk with AA dye was greatly improved by the addition of PE extract. As seen in Fig. 3d, this enhancement was further demonstrated in dyeing procedure with varying addition amounts of AA dye.

As bio-mordant, tannic acid can concurrently establish hydrogen-bonding interactions with the  $-\text{NH}_2$  groups of silk fiber and the  $-\text{OH}$  groups of the AA dye, thus yielding a ternary

Table 1 Color fastness of dyed silk fabric

Sample	Rubbing fastness		Washing fastness			
	Dry	Wet	Color change	Cotton	Nylon	Wool
AS	3–4	3	3–4	3–4	4	3–4
APS	4	3–4	4	4	4	4



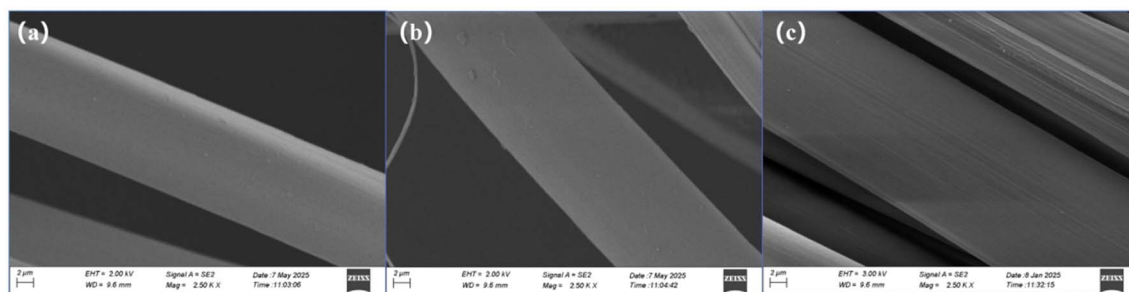


Fig. 4 SEM images of the fabric samples, WS (a), AS (b), APS (c).

“dye-tannic acid-silk” complex.<sup>38–40</sup> The color fastness of the dyed silk fabrics was moderately improved by this supramolecular network,<sup>41</sup> as seen by the 0.5 grade rise in rubbing fastness and washing fastness shown in Table 1.

### 3.3 Characterization of dyed silk fabric

**3.3.1 SEM analysis.** The SEM images reveal that the surface morphologies of WS and AS were essentially identical (Fig. 4a and b), indicating that the AA dyeing process did not substantially alter the surface morphology of the silk fabrics. However, consistent with previous reports on the effect of tannic acid on silk,<sup>42</sup> the PE-dyed silk (APS) exhibited groove-like patterns on its surface (Fig. 4c). This change in surface morphology, which may be attributed to the acidic constituents in the PE extract, resulted in an increased surface area of the silk fiber. The greater surface area contributed to enhanced adsorption of the AA dye, thereby improving the overall dyeing efficiency.

**3.3.2 FTIR analysis.** Fig. 5a shows that the FTIR spectra of WS, AS, and APS are substantially comparable, indicating that the dyeing process with AA and the mordanting with PE extract did not alter the overall chemical structure of the silk fabric. For the silk fibers, the region around 3286  $\text{cm}^{-1}$  in the infrared spectrum is indicative of the coupling between the  $\text{-NH}$  stretching vibrations and the amide stretching vibrations.<sup>43</sup> The distinctive peak observed near 1625  $\text{cm}^{-1}$  is associated with the carbonyl ( $\text{-C=O}$ ) stretching vibrations that constitute the amide I band.<sup>44</sup> Additionally, the peak located at 1518  $\text{cm}^{-1}$  is attributed to the  $\text{N-H}$  in-plane vibrations that constitute the amide II band.<sup>45</sup> Notably, a slight decrease in transmittance is observed for the APS sample at these specific wavelengths. This change can be attributed to the mordanting mechanism of tannic acid from the PE extract. The abundant phenolic hydroxyl groups ( $\text{-OH}$ ) of tannic acid interact with the functional groups on the silk fabric, such as amino ( $\text{-NH}_2$ ) and amide ( $\text{-CO-NH-}$ ) groups, primarily through hydrogen

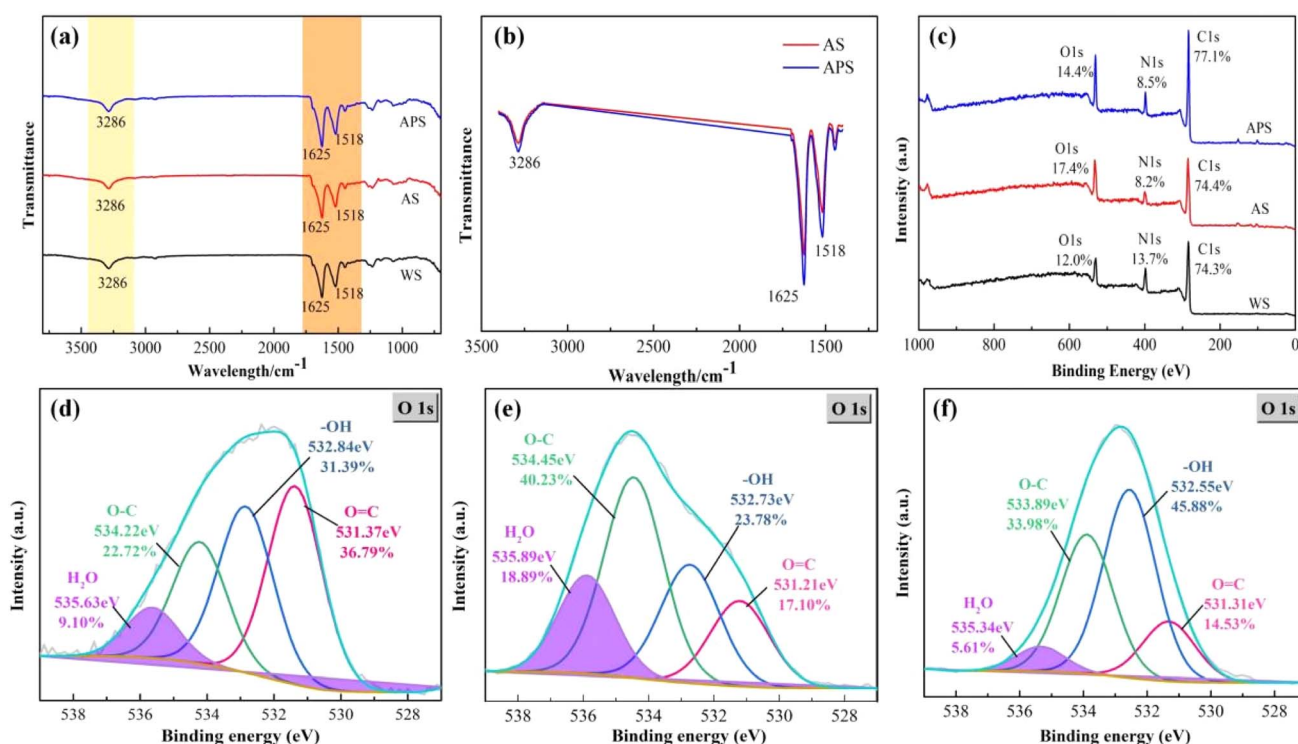


Fig. 5 FTIR spectra of the silk fabrics (a and b), full X-ray photoelectron spectroscopy of silk fabrics (c), narrow-spectrum X-ray photoelectron spectroscopy of WS (d), AS (e), APS (f).



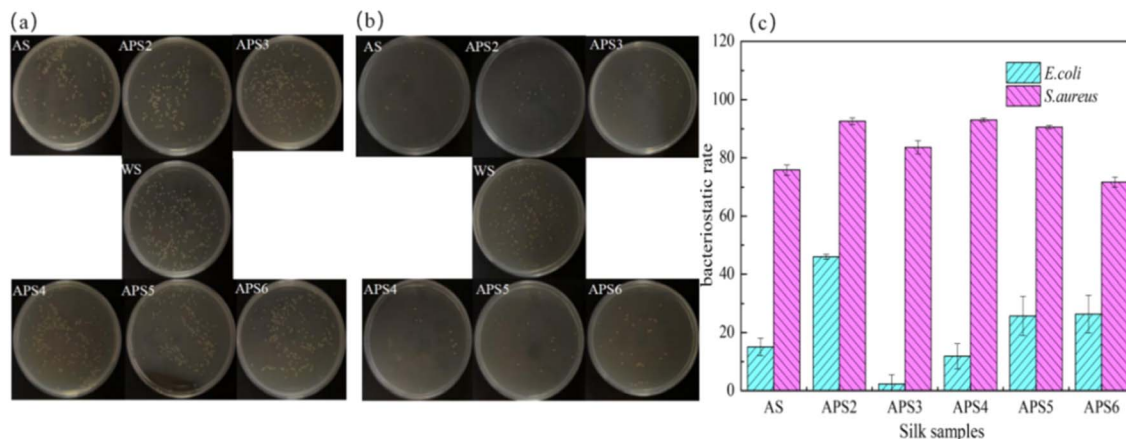


Fig. 6 Antibacterial activity of dyed silk fabrics against *E. coli* (a) and *S. aureus* (b), the bacteriostatic rate (c).

bonding. The anchored tannic acid subsequently acts as a molecular bridge, forming additional hydrogen bonds with polar groups in the natural AA dye. This cross-linking results in a stable “silk-tannic acid-dye” ternary complex, as evidenced by the observed decrease of C=O content in the XPS analysis of the mordanted fabric. The formation of this complex enhances dye binding and reduces the vibrational freedom of the involved functional groups, leading to a slight decrease in transmittance observed in the FTIR spectrum of the APS fabric.

**3.3.3 XPS analysis.** To investigate the effect of AA dye and PE extract on silk fabrics, the full spectrum of XPS was employed to study the changes in the content of carbon (C), nitrogen (N) and oxygen (O) elements on WS, AS, and APS fabric. Fig. 5c reveals that the oxygen content of the original silk fabric (WS) was 12.0%. After treatment with AA dye and PE extract, the oxygen content of the dyed fabrics increased to 17.4% (AS) and 14.4% (APS), respectively. This marked increase could be attributed to the abundant polyphenols and flavonoids present in AA dye and PE extract. The XPS narrow-spectrum analysis was applied to analyze the differences in the oxygen bond content of the dyeing silk fabric. The O 1s peak was decomposed into main three peaks, including the C=O bond at 531.37 eV, the –OH bond at 532.84 eV and the C–O bond at 534.22 eV.<sup>46</sup> As can be seen from Fig. 5d, the proportions of the C=O bond, the –OH bond and the C–O bond in the original silk fabric (WS) were 36.79%, 31.39% and 22.72%, respectively. It is evident from Fig. 5e that the C=O bond content decreased to 17.10% (Fig. 5e) for AS fabric and 14.53% for APS fabric (Fig. 5f). This decline suggests a reduction in –CONH bonds on the silk fibers, likely due to the formation of hydrogen bonds with –OH groups from the AA dye and tannic acid present in the PE extract. In contrast, the C–O bond content increased to 40.23% in AS and 33.98% in APS fabrics. This increase indicates enhanced C–O bonding after dyeing with AA dye, possibly resulting from the adsorption of polyphenols and flavonoids onto the silk fabric.

### 3.4 Antibacterial properties

The AA dye and PE extract both contain nature substances with antibacterial properties.<sup>47–49</sup> Therefore, the antibacterial properties of the dyed silk fabrics were assessed against *E. coli* and *S.*

*aureus*. As depicted in Fig. 6a, the bacteriostatic rates against *E. coli* were relatively low across all samples, with the highest bacteriostatic rate observed in APS2 at approximately 40%. Nevertheless, the dyed silk fabrics exhibited remarkable antibacterial activity against *S. aureus* (Fig. 6b). The different bacteriostatic rates observed against *E. coli* and *S. aureus* can be attributed to the differences in their cell wall structures. As a Gram-positive bacterium, *S. aureus* possesses a thick peptidoglycan layer,<sup>50,51</sup> which is susceptible to disruption by flavonoids and tannic acid present in the AA dye and PE extract. In contrast, Gram-negative bacteria such as *E. coli* have a more complex cell wall structure, containing only 5–20% peptidoglycan, which is further shielded by an outer membrane rich in lipopolysaccharides.<sup>52</sup> This outer membrane acts as a permeability barrier, significantly restricting the penetration of antibacterial compounds and consequently reducing their efficacy.

The incorporation of PE extract significantly enhanced antibacterial performance. The sample mordanted with PE extract (APS4) showed 93.03% inhibition against *S. aureus*, substantially exceeding the 75.82% of the AS sample (Fig. 6c). This enhancement is attributed to tannic acid in the PE extract, which not only promoted the uptake of flavonoid-rich AA dye but also exerted a synergistic effect with the incorporated flavonoids. The proposed synergy involves a multi-targeted mechanism, including co-disruption of the cell membrane, inhibition of key enzymes, and chelation of essential metal ions.<sup>53</sup>

Notably, sample APS2 exhibited antibacterial activity against *S. aureus* comparable to that of APS4, despite its lower *K/S* value. This phenomenon is attributed to the highly acidic microenvironment of APS2, which maintains flavonoids in a protonated state. This protonation enhances their lipophilicity, thereby facilitating integration into and disruption of the bacterial cell membrane.<sup>54</sup> The resultant membrane damage enables deeper cellular penetration and inhibition of intracellular enzymes, ultimately leading to cell death.

## 4. Conclusion

This study has demonstrated that PE extract served as an effective nature mordant for sustainable silk dyeing with AA

dye, overcoming the inherent limitations of conventional metal mordants. The tannin-rich PE extract enhanced dyeing performance through dual mechanisms including surface etching of silk fibers and formation of stable ternary complexes with AA dye and silk fabrics *via* hydrogen-bonding networks. Under the dyeing conditions (pH 4, 90 °C, 30 min), PE-mordanted silk fabrics exhibited a 79.08% increase in *K/S* value compared to the unmordanted silk fabrics. Meanwhile, the experimental results demonstrated that the bio-mordanted silk fabrics achieved 0.5-grade improvements in rubbing fastness and washing fastness relative to unmordanted fabrics. Furthermore, synergistic interactions between the AA dye and tannin-rich PE extract imparted exceptional antibacterial properties, achieving 93.03% bacteriostatic rate against *S. aureus*. This study successfully developed an eco-friendly dyeing technology using tannin-rich PE extract as an effective alternative to conventional metal mordants. The proposed method not only significantly improved the dyeing performance of silk fabrics but also endowed them with excellent antibacterial properties, while avoiding heavy metal pollution. These findings provide new insights for developing environmentally friendly functional textiles. Future studies could focus on the synergistic effects and underlying mechanisms of combined mordant systems to optimize dyeing performance. Moreover, standardized extraction protocols, assessments of textile reusability, and comprehensive life-cycle analyses should be further explored to facilitate the commercialization of natural mordants.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

The authors declare that the data supporting the findings of this study are available within the paper.

## Acknowledgements

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