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Aging time improves adhesive performance of handmade starch paste for restoration of ancient Chinese books and its mechanism of action

Changwei Wang[1†], Yuqi Yao[2†], Yue Zhang[3] and Xiao Yao[1*]

Abstract

In the restoration of ancient Chinese books, handmade starch paste serves as a paper adhesive, distinguished from traditional starch paste preparation methods. It involves special processes such as starch washing and aging, relying entirely on the artisanal expertise throughout the entire process. The study recreates the process of making handmade starch paste for the restoration of traditional ancient books and investigates the effects of aging time on the apparent viscosity, rheological properties, and adhesive performance of the paste. The results indicate that during aging, the pH of the starch paste decreases significantly, but it has a minimal impact on its apparent viscosity, rheological properties, and paper softness. However, it notably enhances the adhesive performance, with the optimal results observed after 3 days of aging. This is attributed to the decrease in residual protein content in the starch, as well as the significant improvement in swelling power and solubility of the starch. The results of infrared spectroscopy and XRD testing reveal that there are no significant changes in the molecular and crystalline structures of starch during the aging process. The acidic environment produced by starch fermentation promotes protein hydrolysis, emerging as the primary reason for the improved adhesive performance of the paste.

Keywords Ancient book restoration, Handmade starch paste, Aging time, Adhesive performance, Action mechanism

Introduction

The extensive ancient books are not only crucial components of valuable cultural heritage, bearing rich historical and cultural connotations, but also serve as important mediums for documenting the material civilization and spiritual civilization of great nations [1]. However, due to various natural and societal factors, ancient books have suffered a degree of damage, and the responsibility for restoring these ancient books bears the crucial task of prolonging their longevity and safeguarding their invaluable contents. As the adhesive used in the restoration of ancient books, the performance of handmade starch paste directly affects the quality of the restoration work [2].

The principle of ancient book restoration work is "restore it as it was," meaning that when restoring ancient books, the goal is to preserve their original state as much as possible [3]. Before and after restoration, efforts are made to maintain the original appearance. Handmade starch paste plays a crucial role in bonding pages and supporting painting cores. Therefore, there are strict

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performance requirements for the paste used in ancient book restoration.

First and foremost, it must be reversible, meaning that pages repaired with the paste can be unmounted with almost no negative effects [4]. Although industrial paper adhesives such as CMC (carboxymethyl cellulose) and PVA (polyvinyl alcohol) are reversible under appropriate conditions, they may still cause slight damage to the paper during detachment in practical operations, which is unacceptable when dealing with ancient books. Secondly, the paste should exhibit a clean, white texture, possess suitable adhesive properties, be adjustable in thickness, and maintain its adhesive properties over time [5].

In practical conservation practices, handmade starch paste is gently removed by fully moistening the bonding area, leveraging its inherent reversibility. This is related to the hydrogen bonding strength within their molecular structure. The polar hydroxyl groups in the chain-like starch molecules readily form hydrogen bonds with water molecules, making the paste highly hydrophilic, resulting in a "swelling" phenomenon. Therefore, under high humidity (above 90% RH) or when fully moistened with water, the adhesive bond between the paste and the binding material will undergo desorption [6], as depicted in Fig. 1. Due to the intrusion of water molecules, the distance between the paper bonding interfaces increases from L (before swelling) to L+2ΔL (after swelling), disrupting the hydrogen bonds formed between the paste and the bonded material. This results in the creation of free hydroxyl groups that become new water molecule attraction points, leading to the infiltration of more water molecules. As a result, the hydrogen bonds within the adhesive structure gradually break, leading to a reduction in adhesive strength.

Handmade starch paste used for ancient book restoration nowadays is either prepared by staff according to ancient formulas. The basic ingredient for making the paste is flour that has been processed by ancient methods to remove gluten and impurities, as this type of flour has been proven through practical experience and scientific experimentation to yield excellent results [7, 8]. Fan [5] provides a detailed introduction to the preparation, storage, and usage of handmade starch paste, emphasizing the importance of proper paste application.

In previous related studies, the focus has typically been on the appropriate usage of handmade starch paste, with little attention given to the preparation process and its underlying reasons. Moreover, the production of the paste heavily relies on artisanal expertise, as there are currently no standardized references or comprehensive systematic studies on its production process. Therefore, the primary aim of this study is to recreate the production process of handmade starch paste, drawing from existing literature and practical knowledge. The main focus is to examine how the aging process affects different properties of the paste and delve into its mechanism of action. By doing so, this research seeks to advance the scientific and standardized production and application of the paste, ultimately improving the quality of ancient book restoration.

Materials and methods

Materials

Wheat flour, a commercial product with 11%wt gluten (Yihai Kerry Arawana Holdings Co., Ltd., China). Xuan paper, made of ~80% fibers of sandalwood and ~20% straw fibers (Jing County, Anhui province), has an average thickness of 0.068 mm and an average paper

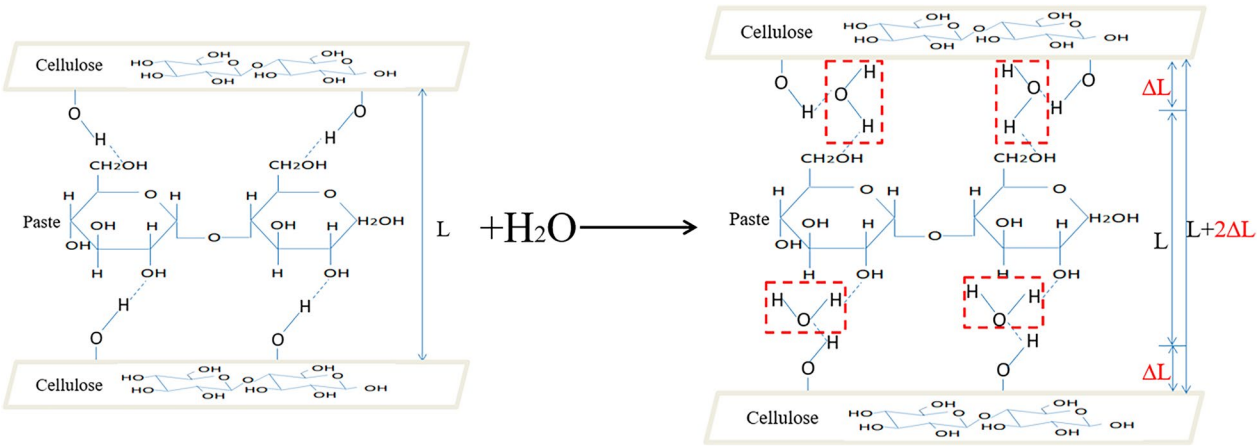

Fig. 1 The desorption process of water molecules in paste adhesive structure

grammage of 38.7 g/m^2. Silk fabric, made by Huzhou Yunhe Shuanglin Silk fabric Co., Ltd., China.

Sample preparation

Handmade starch paste preparation

The preparation of handmade paste was shown in Fig. 2, which mainly included the following four steps:

Step I. Gluten removal

The traditional extraction method was employed to extract wheat starch. Wheat flour and water were mixed in a 2:1 ratio to form a dough, which was then allowed to stand for 20 min. The dough was added with water and continuously kneaded. Insoluble proteins in flour aggregated into gluten, while starch separated out with the water flow [9]. A small amount of gluten particles may mix into the starch slurry. Using an 80-mesh filter effectively removed gluten particles from the starch slurry. The main purpose of removing gluten was to eliminate the insoluble proteins in flour, as the proteins can affect the gelatinization of starch and make the paste prone to deterioration.

Step II. Aging

The cleaned starch slurry from step I was placed in a constant temperature and humidity curing box (model HSP, China) at 20 ℃ and stood for aging. The starch naturally settled, while the upper liquid turned yellow with a slight acidic taste [10]. The aging period ranged from 0 to 7 days, with 0 days representing unaged starch. Separating the liquid phase from the solid phase, the precipitated solid was dried to obtain starch with different aging periods, while the supernatant was dried to obtain yellow colloidal substances containing different hydrolysis products of starch. The average moisture content of starch was about 11.8%.

Step III. Paste preparation

The starch obtained from step II was mixed with water in a 1:4 ratio and heated in an 80 ℃ water bath. The slurry was continuously stirred until it thickened from a thin consistency to a semi-transparent one (cooked pulp), indicating the completion of starch gelatinization [11].

Step IV. Paste application

The cooked pulp would cool into a solid and needed to be diluted with water before use. A suitable amount of it was mixed with water in a 1:3 ratio to obtain a starch paste with moderate viscosity for ancient book restoration.

Gelatinization process of starch

The preparation of starch paste essentially involves the process of starch gelatinization at high temperatures [12]. With the increase in temperature, the order of starch granule molecules is disrupted, leading to a rapid increase in the viscosity of the starch suspension, forming a semi-transparent viscous colloidal substance after stirring.

As depicted in Fig. 3, starch granules (I) are typically enveloped by amylopectin and are insoluble in cold water, forming only a suspension. When the starch suspension is heated, the starch granules undergo a process of water absorption and swelling in a wet and hot environment (II). Once the gelatinization temperature is reached, hydrogen bonds between amylose molecules break, gradually disrupting the crystalline structure, while the outer layer of amylopectin around the granules also ruptures (III). The starch volume increases to several tens or hundreds of times its original size, transforming the suspension into a translucent, viscous colloidal substance, indicating gelatinization completion. During cooling, the colloid undergoes regeneration of amylose and forms ordered or crystalline amylopectin molecules (IV) [13].

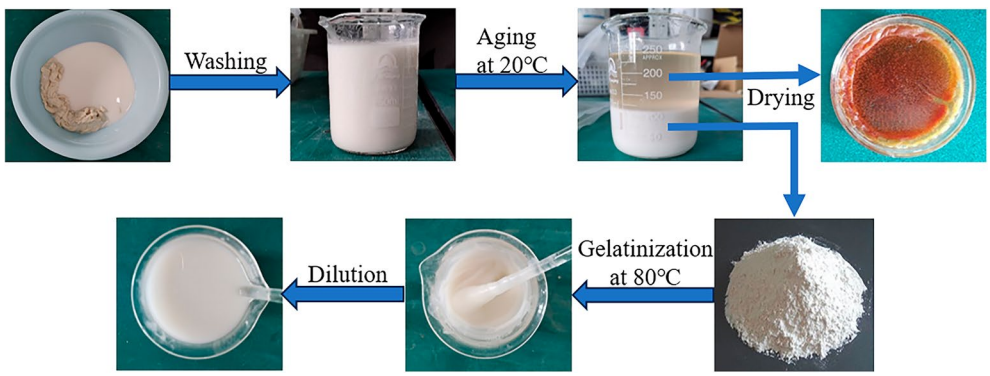

Fig. 2 Handmade starch paste preparation process

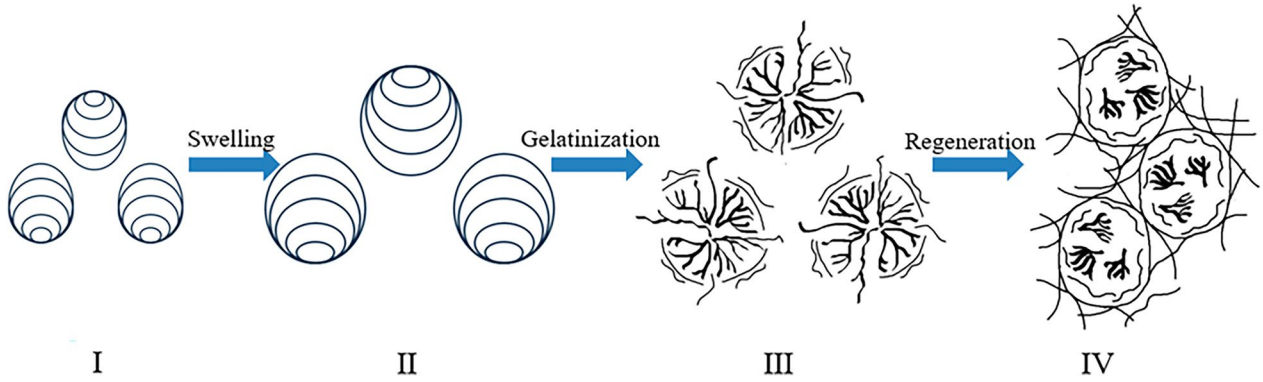

Fig. 3 High temperature gelatinization process of starch

Xuan paper sample

Xuan paper was selected from the middle section and cut into dimensions of 40 cm horizontally and 30 cm vertically. The Xuan paper that had not been coated with paste was used as the blank control group. 18 g of the paste was evenly brushed onto the surface of the Xuan paper sample using a brush. After natural drying at 25 ℃, tensile strength and softness tests could be conducted on the paper samples.

Silk fabric sample

For artworks composed of non-rigid materials such as paper and silk fabric, peel strength can be used to evaluate the adhesive bonding strength [14]. The fiber strength in paper material is lower compared to the silk protein in silk material. During the peeling process, the paper sample often does not separate at the bonding interface, but rather the paper itself undergoes delamination and damage, making it difficult to analyze the peel strength of the paper sample. Therefore, silk fabric is chosen as the base material to analyze the peel strength.

The traditional wet mounting method was employed for mounting as illustrated in Fig. 4, where the bonding section is 150×150 mm. 10 g of the paste was evenly brushed onto the bonding section of the silk fabric using a brush, ensuring no air bubbles were present in the mounted fabric. Weight was applied to ensure adequate contact between the upper and lower layers of silk fabric and the paste. After natural drying at 25 ℃, tests for the peel strength of the silk fabric could be conducted.

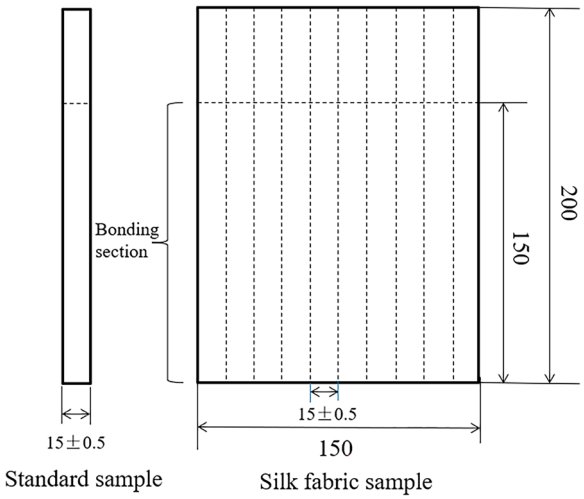

Fig. 4 Preparation of silk fabric sample

pH determination

During the aging process of starch slurry, the increase in the acidity of the slurry is mainly caused by the substantial proliferation of microorganisms resulting from starch fermentation. A pH meter (model PHS-25, China) was used to measure the pH of the supernatant and the paste made from aged starch at 25 ℃.

An acidic environment could accelerate cellulose degradation [15], so the acidity of the adhesive may also promote the acidification of paper. According to ISO 6588-1:2021 [16], the pH of paper sample was measured using the cold extraction method, with uncoated Xuan paper as the blank sample. The air-dry sample was shredded to approximately 5–10 mm^2, and 2 g of the sample was weighed and added to 100 ml of distilled water. Cold extraction was conducted at 25 ℃, followed by thorough agitation, and then the pH was measured.

Apparent viscosity and rheological properties

The apparent viscosity of the paste was directly measured using a digital viscometer (model NDJ-8S, China) at 25 ℃, while the rheological properties were obtained using a six-speed rotational viscometer (model ZNN-D6,

China). Experimental data of the flow curve were fitted using the power-law model [17]:

$$\tau = K \cdot r^n$$

where τ is the shear stress (Pa), γ is the shear rate(s^{-1}), K is the consistency coefficient (Pa•s^n) and n is the flow behavior index.

Tensile strength

Tensile strength refers to the maximum tension that a paper sample can withstand per unit width before breaking. The paper samples were cut into test strips measuring 150 mm in length and 15 mm in width, with 10 strips each for horizontal and vertical directions testing according to GB/T 12914-2018 (Chinese standard) [18]. The average maximum tensile strength and elongation were measured using a horizontal computerized tensile tester (model WZL-B, China) with a pulling rate of 20 mm/min.

Softness

Softness refers to the maximum vector sum of the sample's resistance to bending and the frictional force between the sample and the testing gap. The smaller the softness, the softer the sample. The softness of the paper samples was measured using a Tissue Softness Analyzer (model TSA, Germany), following the method described by Xu et al. [19]. The samples were cut into test strips of 100 mm by 100 mm, with 10 strips tested in both horizontal and vertical directions. The results are the average values for the horizontal and vertical directions.

Peel strength

The silk fabric samples were cut into strips measuring 200 mm in length and 15 mm in width for peeling tests by a horizontal computerized tensile tester (model WZL-B, China) according to ASTM D903 [20], with a stretching rate of 20 mm/min.

FT-IR spectroscopy

The chemical characteristics of wheat flour, aged starch, and starch slurry supernatant were investigated using Fourier-transform infrared spectroscopy (FT-IR) [21]. All spectra were collected in the spectral range of 4000–400 cm^{-1} using a Bruker Tensor-27 spectrometer (Germany) at room temperature, with an average of 16 scans and a spectral resolution of 4 cm^{-1}. All spectra were baseline-corrected and normalized.

Protein content

Wheat flour contains about 11% protein. In Sect. "Sample preparation", the process of gluten removal could only remove the insoluble proteins in the flour, mainly including glutenin and gliadin. A portion of the water-soluble proteins, however, remained in the aged starch after drying. The determination of protein content used the Kjeldahl method for nitrogen determination [22].

Swelling power and solubility

Swelling power and solubility of the starch were determined according to the method described by Shang et al. [23] with modification. 2% (w/v) starch suspension (50 ml) was maintained at 80 ℃ for 30 min, cooled, and then centrifuged at 3000 rpm for 20 min. The supernatant was placed in glass dishes and dried. The swelling power was evaluated as the wet sediment weight divided by initial dry matter weight excluding water-soluble starch. The solubility was expressed as the percentage of dried solid weight based on the weight of dry sample.

X-ray diffraction

X-ray diffraction patterns of the starch with different aging periods were obtained using an X-ray diffractometer (model Ragaku Ultima IV, Japan). The diffractometer was operated using Cu-Kα radiation at 25 mA and 35 kV with the scanning speed of 5°/min from 5 to 40°, and step intervals was 0.02°. Relative crystallinity (%) was calculated as the percentage ratio of the diffraction peak area to the total diffraction area [24].

Result and discussion

pH

During the aging periods of starch slurry, starch fermentation leads to the proliferation of microorganisms, which produce organic acids. The organic acids cause the pH of the slurry to decrease continuously, as shown in Fig. 5. At aging 0 days, the pH of the starch slurry is 6.41, close to neutral. But during days 1–4 of aging, the acidity

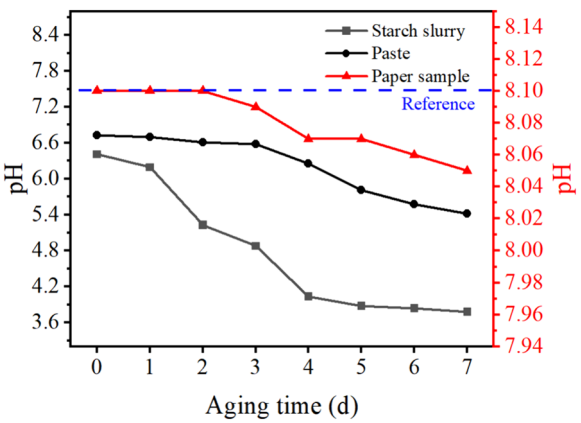

Fig. 5 Effect of aging time on the pH of starch slurry, paste and paper sample (25 ℃)

of starch slurry noticeably increases. By aging 4 days, its pH drops to 4.04, with a slight further decrease observed thereafter. Similarly, the pH of the paste also decreases with prolonged aging time. Before aging 3 days, there is little change in pH, remaining close to neutral. However, after 3 days, the pH of the paste decreases significantly from 6.58 to 5.42.

An acidic environment will accelerate paper acidification, so adhesives used for paper should ideally be as close to neutral as possible [25]. The pH of the reference paper is 8.10, indicating weak alkalinity. This is due to the addition of fillers such as calcium carbonate in the manufacturing process of Xuan paper [26]. The acidity of the paste can affect the pH of the paper sample. The paste before aging 3 days has almost no impact on the pH of the paper sample, whereas paste after 3 days will lower the pH of paper sample. Although the decrease of the pH is only 0.05 in numerical value, it has a certain negative impact on the long-term stability of the paper, making it unfavorable for prolonged preservation. This suggests that despite the increase in acidity during the aging process, the impact on the paste and paper is relatively minimal before aging 3 days.

Apparent viscosity and rheological properties

The paste used in the restoration of ancient books has stringent requirements for its apparent viscosity. If the paste is too thick, it becomes difficult to apply, leading to deformation of the adhered pages and causing secondary damage to the ancient books. On the other hand, if the paste is too thin, it may result in hollow areas and poor adhesive properties, causing the repaired pages to detach on their own after some time.

The study discusses obtaining the moderate viscosity pastes by controlling the ratio of starch to water in the gelatinization and dilution steps, and testing its apparent viscosity at 25 ℃ as shown in Fig. 6a. Overall, there is not a significant difference in apparent viscosity among them. The paste aged for 0 days has slightly higher apparent viscosity, while the apparent viscosity of pastes aged for 1–7 days fluctuates around 35 to 39 mPa s. However, for polymer fluids like pastes, which typically exhibit some degree of pseudoplasticity, there may be fluctuations in measurements, so further rheological properties are measured.

The steady flow behaviors of pastes were investigated, and the results are shown in Fig. 6b and Table 1. According to Fig. 6b, the flow behavior profile of each sample is well fitted by the power-law model ($R^2 = 0.9988–0.9991$). The consistency coefficient (K), flow behavior index (n), and coefficients of determination (R^2) for each flow curve are presented in Table 1. All pastes have a pseudoplastic flow behavior for n values < 1 (0.8534–0.8565), indicating a shear-thinning behavior, that is viscosity decreases with frequency or increasing shear rate. The n values remain almost unchanged, while the consistency coefficient K initially decreases, followed by a slight subsequent rise. This indicates that the 7-day aging duration does not appear to significantly impact the apparent viscosity and rheological properties of the paste.

Table 1 Power-law parameters of aged starch (25 ℃)

Samples	Formula	K/Pa•s^n	n	R^2
Aging 0 days	$\tau = 0.1137\dot{r}^{0.8534}$	0.1137	0.8534	0.9988
Aging 3 days	$\tau = 0.0895\dot{r}^{0.8565}$	0.0895	0.8565	0.9991
Aging 7 days	$\tau = 0.0972\dot{r}^{0.8552}$	0.0972	0.8552	0.9990

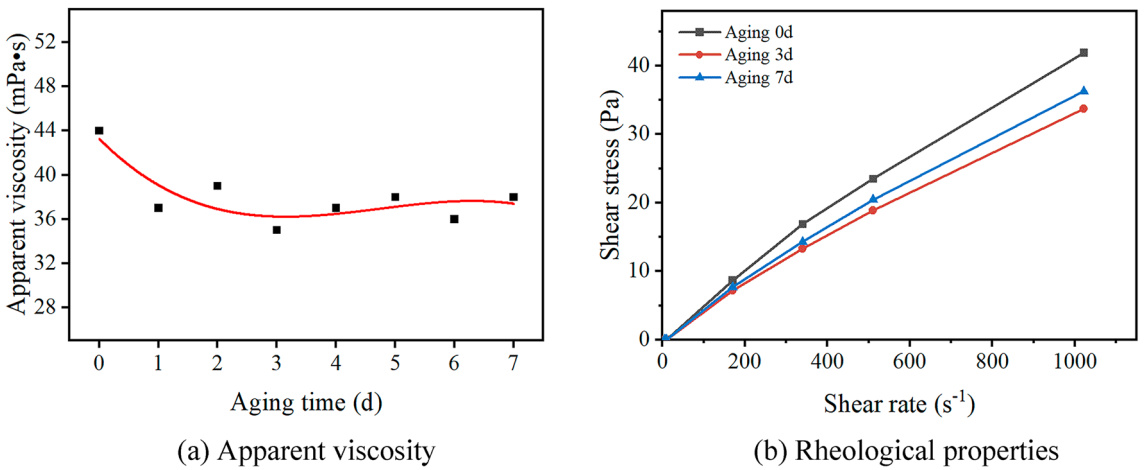

Fig. 6 Effect of aging time on apparent viscosity and rheological properties (25 ℃). **a** Apparent viscosity. **b** Rheological properties

Tensile strength

Coating pastes with different aging periods were applied on paper samples to compare the tensile strength, and the results are shown in Fig. 7. There are certain differences in the mechanical properties of the paper in the transverse and longitudinal directions. In the transverse direction, both tensile strength and elongation are higher than in the longitudinal direction, indicating that the flexibility of the paper is greater in the transverse direction. This is attributed to the unique papermaking process of Xuan paper, which determines the orientation of the paper fibers [27]. In the transverse direction, the fiber orientation is dominant, resulting in strength significantly greater than the bonding strength between fibers.

The tensile strength of paper samples coated with starch paste is significantly higher than that of the reference. The paste infiltrates the pores of the paper, and as the moisture evaporates, the paste dries to form a film, thereby significantly improving the paper's tensile strength and elongation. The tensile strength of the paper shows a trend of initially increasing and then stabilizing. Before aging 3 days, the tensile strength of the paper gradually increases. At aging 3 days, the paper exhibits the highest tensile strength. Compared to aging 0 day, the transverse tensile strength increased by 79.6%, elongation increased by 61.0%, longitudinal tensile strength increased by 42.9%, and elongation increased by 25.3%. After aging 3 days, there is little change in the tensile strength and elongation of the paper.

Softness

Softness is crucial for the restoration of ancient books using starch paste. Therefore, the effect of starch paste with different aging times on the softness of paper samples was tested, as shown in Fig. 8. The paste-coated paper samples all exhibited higher softness compared to the reference, while the differences between samples with different aging times were relatively small. This indicates that the application of starch paste increases the rigidity of the paper to some extent, with a minimal correlation to aging time, but the change in softness at aging 3 days is relatively small.

Peel strength

The peel strength characterizes the adhesive strength of the adhesive. If the paste has strong adhesive strength, the bonding is less likely to loosen, which is beneficial for the long-term preservation of ancient books. As shown in Fig. 9, peel strength shows an initial increase followed by stabilization, consistent with the trend observed in the tensile strength of the paper. Before aging 3 days, peel strength gradually increases, reaching its maximum at aging 3 days with a 23.9% improvement compared to aging 0 day.

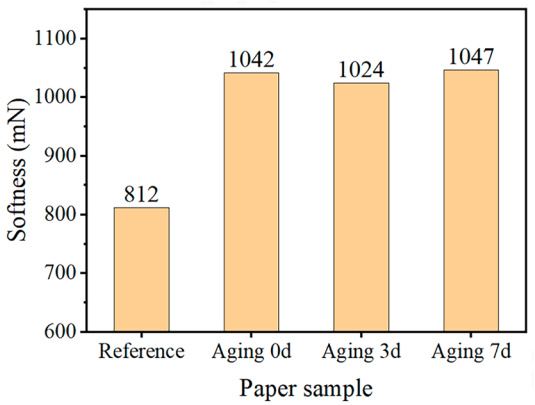

Fig. 8 Effect of starch paste with different aging times on softness

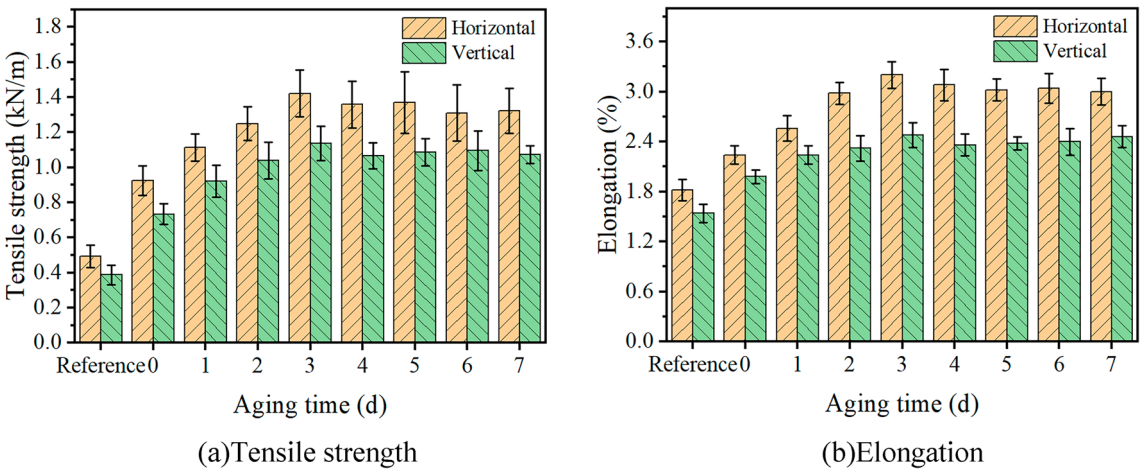

Fig. 7 Effect of aging time on tensile strength. **a** Tensile strength. **b** Elongation

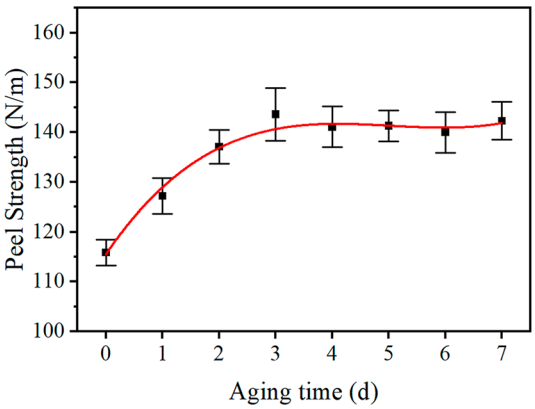

Fig. 9 Effect of aging time on peel strength

FT-IR spectroscopy

The FT-IR spectra of wheat flour, aged starch, and starch slurry supernatant are shown in Fig. 10. From the magnified view of Fig. 10b, it can be observed that only wheat flour exhibits distinct absorption peaks around 1663 cm^{-1} and 1604 cm^{-1}, which are associated with the amide I band in the secondary structure of proteins [28]. This indicates that gluten removal effectively eliminates insoluble proteins from wheat flour.

As shown in Fig. 10a, the infrared spectrum of flour and starch granules shows a characteristic broad stretching peak of hydroxyl groups near 3400 cm^{-1}, and the small band at around 2926 cm^{-1} is attributed to the C-H stretching of CH_2 groups [29]. The absorption peak near 1656 cm^{-1} corresponds to the amorphous region's absorption peak of water adsorbed by starch [30]. The peak around 1435 cm^{-1} is attributed to the CH_2 bending vibration, while the absorption peak near 1370 cm^{-1} corresponds to the bending vibration of the CH_2 bond. The absorption peak around 1157 cm^{-1} is assigned to the stretching vibration of C-O and C–C bonds, and the peak near 1017 cm^{-1} is associated with the stretching vibration of the C-O bond and the bending vibration of C–OH. The absorption peak near 764 cm^{-1} is attributed to the stretching vibration of the C–C bond, and the peak around 576 cm^{-1} is assigned to the skeletal mode vibration of starch [31]. The infrared spectra of starch at different aging periods show almost no variation, indicating that the aging process has minimal impact on the functional groups of starch.

On the other hand, the supernatant containing various hydrolysis products exhibits an infrared spectrum distinct from that of starch. There is a strong absorption peak around 3422 cm^{-1}, which includes N–H (amide A band) and sugar hydroxyl stretching vibrations. The absorption peak near 2930 cm^{-1} is associated with the C-N stretching vibration (amide B band), and the absorption peak near 1636 cm^{-1} is attributed to the stretching vibration of C=O stretching vibration (amide I band) [32]. The peak around 1422 cm^{-1} is assigned to both the C=O stretching vibration and the N–H bending vibration (amide III band), and the absorption peak near 1057 cm^{-1} is associated with the stretching vibration of the C-O bond. This indicates that the supernatant not only contains glucose or polysaccharides as hydrolysis products of starch but also includes amino acids or peptides from the hydrolysis of proteins.

Protein content

According to existing research reports, during the process of starch gelatinization, proteins bind to the surface of starch granules to form stable complexes, hindering both the swelling of starch granules and the leaching of amylose molecules, thereby delaying starch gelatinization [33]. Protein hydrolysis products are identified in

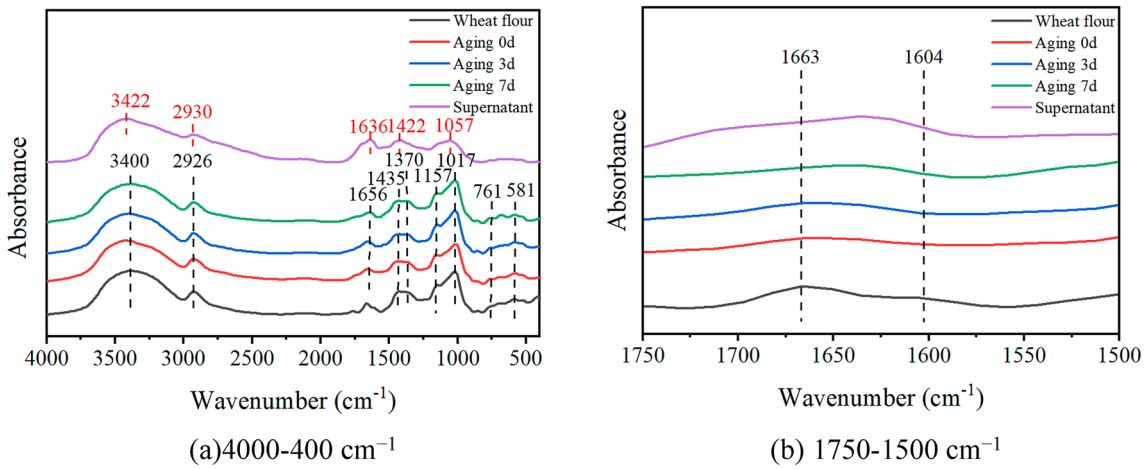

Fig. 10 FT-IR spectra of wheat flour, aged starch and supernatant. **a** 4000–400 cm^{-1}. **b** 1750–1500 cm^{-1}

the infrared spectrum of the supernatant. Therefore, further testing is conducted to determine the protein content in the starch, with the results depicted in Fig. 11. Removal of insoluble glutenin and gliadin proteins from wheat flour can reduce the protein content in starch from approximately 11% to 1.92%, leaving behind soluble proteins within the starch. During the aging process, there is a significant decrease in protein content, reaching around 1.5% after aging 3 days, indicating a decrease of 21.9% compared to aging 0 day.

Swelling power and solubility

Swelling power and solubility respectively refer to the ability of starch to absorb water and expand during heating, and the degree to which it dissolves in water. Both of these factors reflect the extent to which starch undergoes gelatinization and forms colloidal suspensions in water [34]. Swelling power and solubility of starch at different aging periods are depicted in Fig. 12. Both swelling power and solubility of starch show a gradual increase before reaching their peak at aging 3 days, after which they stabilize and remain relatively constant.

As described in Sect. "pH", the adhesive strength of the paste is derived from the gelatinization of starch. The adhesive performance of the paste is positively correlated with its starch gelatinization capability and negatively correlated with protein content. The enhanced gelatinization capacity of aged starch may be associated with the hydrolysis of water-soluble proteins in an acidic environment.

X-ray diffraction

The X-ray diffraction patterns and relative crystallinity of starch at different aging periods are presented in Fig. 13. All starch granules show strong diffraction peaks at 15°, 17°, 18° and 23°, and there are two connected double peaks at 17° and 18°, indicated that starches are all

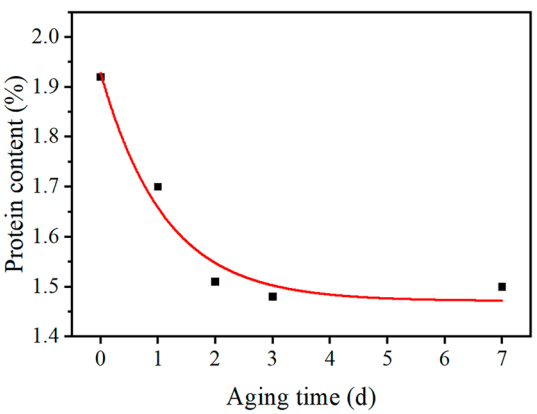

Fig. 11 Effect of aging time on protein content in starch

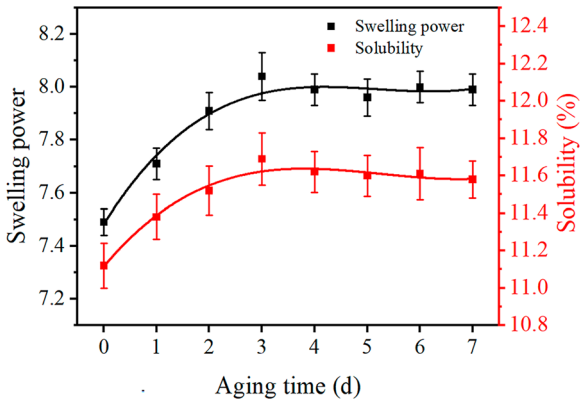

Fig. 12 Effect of aging time on Swelling power and solubility of starch

typical A-type crystal structure, as reported by previous researchers [35]. In addition, a minor diffraction peak appeared at 20°, which is the characteristic of amylose–lipid complex.

The relative crystallinity of starch is slightly reduced, and the decrease in crystal order can enhance the gelatinization ability of starch to a certain extent. However, overall, the 7-day aging duration does not have a significant impact on the X-ray diffraction pattern of starch. It implies that the crystalline structure of starch remains nearly unchanged during the aging periods.

Conclusion

Based on the results of this study, it can be concluded that the aging process has a relatively minor effect on the apparent viscosity, rheological properties, and paper softness, but it significantly improves the tensile strength and peel strength. The optimal aging time at 20 ℃ is found to be 3 days, during which the paste exhibits the highest

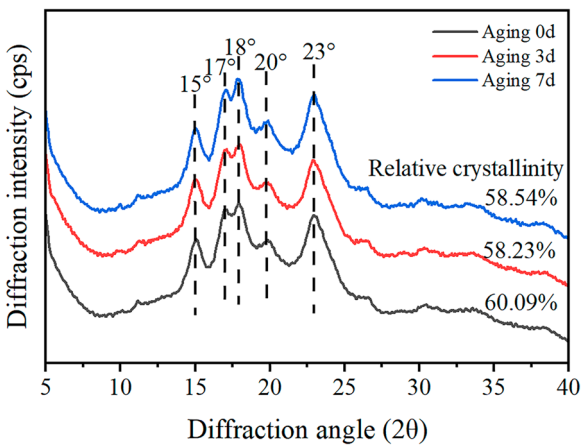

Fig. 13 X-ray diffraction patterns of aged starch

tensile and peel strengths. At this point, the paste has a relatively minor impact on the pH of paper sample and possesses a moderate viscosity, making it suitable for applying. These characteristics align perfectly with the requirements for use in the restoration of ancient books.

Analysis based on infrared spectroscopy and XRD reveals that there are almost no significant changes in the molecular and crystalline structures of starch granules. However, after aging, protein hydrolysis products are found in the supernatant, and the protein content in the starch decreases significantly. The adhesive strength of the paste originates from the gelatinization of starch, and its adhesive properties are positively correlated with swelling power and solubility of starch, while negatively correlated with the protein content. This indicates that the hydrolysis of residual proteins in the starch under acidic conditions is the fundamental reason for the significant improvement in the adhesive performance of the paste.

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Author contributions
C.W.: Conceptualization, Methodology, Data curation, Writing—review & editing. Y.Y.: Investigation, Formal analysis, Writing—original draft. Y.Z.: Investigation, Resources, Data curation. X.Y.: Conceptualization, Resources, Supervision.

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Availability of data and materials
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Declarations

Competing interests
The authors declare no competing interests.

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