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Study of amur cork tree bark extract-dyed paper under artificial UVA irradiation

Yanbing Luo[1*], Xiujuan Zhang[2], Mengjia Ren[1] and Yanfei Wei[3]

Abstract

Some ancient Chinese paper artifacts dyed with amur cork tree bark extracts are currently preserved to different extents in museums and libraries worldwide. As traditional natural plant dyes, the long-term performance of the amur cork tree bark extract-dyed handmade paper has been questioned under a preserved environment. The photodegradation process of the extract-dyed papers was examined under accelerating artificial UVA irradiation conditions according to China national standard-paper and board- accelerated aging (exposure to light). Changes during aging were analyzed by optical observation and scanning electron microscopy (SEM), X-ray diffraction (XRD), thermogravimetric analysis (TGA), Fourier transform infrared spectrometry (FTIR), pH, and tensile strength and folding endurance tests. The results showed that the main components from extracted dyes played an important role in affecting the photodegradation properties of dyed paper. The changes in surface color, pH, morphology and mechanical properties after the artificial UVA accelerated degradation tests revealed that a suitable concentration of the extract dyes for maintaining the long-term irradiation stability of the paper is important.

Keywords UVA irradiation, Artificial ageing, Amur cork tree bark extract, *Daqian* paper

Introduction

This paper is part of a series of research studies on dyeing handmade papers with amur cork tree bark extracts, the reason that amur cork tree bark was chosen as the main yellow paper dyes and the properties and the most suitable concentration of dyes for maintaining the long-term thermal stability of the paper [1, 2]. Historical and more recent documentations have reported that amur cork tree bark extracts have been widely used since ancient times in China due to religious reasons; some characteristics of amur cork tree bark extracts include a bright fluorescent yellow color (C.I. Natural Yellow 18), environmentally friendly finishes, insecticidal properties of yellow alkaloid dyes, and conservation applications. As the most preferred natural basic dyes, amur bark tree extracts are also commonly used to dye matching papers for the restoration of cultural paper relics and maintenance of old features [3–6]. Berberine, the major color component of amur cork tree bark extracts, is a cationic alkaloid that can be bonded to paper fibers by hydrogen bonds and electrostatic attractive forces [2, 7]. Nevertheless, as a natural dye, colorant dyes from amur cork tree barks have some disadvantages; specifically, these dyes can fade and easily deteriorate due to heat, acid gases, light, etc. [1, 8, 9]. In our previous paper, amur cork tree bark extract-dyed handmade paper was examined under dry-heat accelerated aging experiments, and the results showed that the main components from the extracted dyes played important roles in affecting the properties of the amur cork tree bark extract-dyed paper. The most suitable concentration of extract dyes for maintaining the long-term thermal stability of the paper was an extract/water ratio of 1:2 (v/v) [1].

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Empirically, light causes berberine to change color under UV light irradiation. The fading of natural berberine and its derivative by light has been studied [10–13]. All studies have shown apparent instability in light exposure tests. As valuable as these studies have been in determining the relative merits of the tested products, the qualitative nature of the analyses and the acknowledged limitations of these standard accelerated aging protocols provide many future research directions. For the amur cork tree bark extract-dyed paper, the determination of the sensitivity of amur cork tree bark extract-dyed paper to light and the effect of different concentrations of the bark extract-dyed paper on the paper properties need to be addressed to understand the potential of light effects on the amur cork tree bark extracts on paper.

Consequently, research was carried out to explore the aging of amur cork tree extract-dyed paper under accelerating UVA irradiation conditions. Rather than predicting the aging behavior of the extracted dyed paper through an exhaustive analysis of their composition, we simply monitored the changes in appearance and in chemical and mechanical properties of the dyed papers during the aging process.

Changes in dyed paper samples to UVA light exposure enabled the examination of the photochemical stability of the extract-dyed papers and the changes in their physical properties due to photodegradation. As a consequence, the study of the paper dyed with amur cork tree bark extracts could be important to go deep into understanding their compositions and mechanisms that cause changes and enable the development of possible solutions to avoid degradation phenomena.

Experimental section

Materials

Daqian handmade paper (P), with a thickness of 94 μm and a grammage of 25.46 g/m^2 and made by the traditional Chinese handmaking paper process without fillers or sizing, was supplied by the *Daqian* Paper Shop in Sichuan Province; it exhibited slight alkalinity (pH = 7.8) due to the use of plant ash and/or lime during paper fiber preparation. The Herzberg staining test showed that the main plants were Sinocalamus bamboo (*Sinocalamus affinis* (Rendle) McClure) and mulberry bark (*Morus alba* L.). Dried amur cork tree bark was purchased from a local traditional herbal market in Chengdu, Sichuan Province, and used as received.

Preparation of colorant dyes and dyed paper

The preparation of amur cork tree bark extract-dyed paper has been reported in our previous papers [1, 2]. The dyeing process was based on traditional Chinese recipes used for laboratory procedures. Extractions were conducted in beakers with the bark to distilled water at a ratio of 1:10 (w/w) for 12 h before heating from room temperature, and the mixture then was boiled and simmered for 30 min. The extracted solution was obtained by filtering the boiled dregs using a cloth sack (the mass ratio of dissolved matter was ca. 6–8 wt%). The liquors at water/extract liquor ratios of 0:1, 1:1, 2:1 5:1, and 10:1 (*v/v*) were labeled A0, A1, A2 and A5, respectively. The amur cork tree bark extract dyed paper was prepared by completely immerging the original undyed handmade papers in different concentrations of colorant liquors at 80–90 ℃. The dyed paper was hung on glass rods at 23 ± 1 ℃ for 48 h and labeled P-A0, P-A1, P-A2, and P-A5 with color dye solutions of A0, A1, A2 and A5, respectively. The undyed paper was completely immerging the distilled water at 80–90 ℃ to have the same heat history.

Artificial UVA acceleration aging

Samples were preconditioned according to ISO 187–1990 [14] (23 ± 1 ℃ and 50 ± 2% RH) for 24 h and cut into suitable sizes for testing. Artificial UVA irradiation degradation tests were based on GB/T 40278-2021 (Paper and Board-Accelerated Ageing Exposure to Light) using an accelerating UVA aging chamber [15]. To study the photodegradation of paper samples, four 40 W fluorescent ultraviolet (UVA) sunlamps (UVA-340) with a maximum wavelength of 340 nm were installed on a bracket in an accelerated artificial chamber (ATLAS UV test, American) without opening water-spray system. The distance between the lamp and samples was kept constant at 20 cm. The light intensity at the sample surface was approximately 0.68 W/m^2 measured by a UVA radiometer. The temperature inside the chamber was fixed at approximately 50 ± 0.5 ℃. Photodegradation was conducted in a room environment without humidity rate control. The samples were collected at predetermined times for a total of 30 d to evaluate the extent of photodegradation. At predetermined periods, specimens for each test were removed from the chamber and conditioned for 24 h.

Characterization

Colorimetric measurement

The CIE-Lab values of the color changes were measured by a solid CM-700D reflection spectrophotometer (from Minolta Co., Japan, D_{65} light source and 10° observer) according to the standard ISO 11475:2004 [16]. The CIE 1976 L*a*b* color space was used to determine the color difference (ΔE) by the equation $\Delta E = \Delta E = \sqrt{(\Delta L)^2 + (\Delta a)^2 + (\Delta b)^2}$, where changes in red–green (Δa), yellow–blue (Δb) and lightness (ΔL)

for each sample were plotted against exposure time. At least 10 measurements were carried out for each sample, and the results were averaged.

pH tests

The pH measurement of the paper samples was in accordance with GB/T 13528-2015 [Paper and Board-Determination of Surface pH] [17] with little modification. The pH value of the extract was analyzed using a Mettler Toledo In Lab Surface pH meter (Mettler Toledo, Switzerland). A drop of distilled water was added to the paper surface, and the flat-surface combined pH electrode was pressed against the wetted paper surface until the measured value was stable, which took approximately 5 min. The reported results were an average of five determinations. Before the pH measurement, the sensor was calibrated using the provided standard buffer solutions of 4.0, 7.0 and 9.2.

Thermal analysis

Thermogravimetric analysis (TGA) was used to evaluate the thermal stability of paper samples via a Thermal TA550 instrument. Approximately 4–5 mg of the paper samples was heated at a heating rate of 10/min from room temperature to 800 ℃ under an ultrahigh-purity nitrogen flow.

μ-ATR mid-FTIR analysis

The structural changes occurring in the samples upon accelerated aging treatment were monitored by transmission and micro-attenuated total reflection (μ-ATR) mid-FTIR. A Thermo Nicolet iZ10 FTIR instrument equipped with a μ-ATR crystal was used to collect FTIR spectra in the wavenumber range of 650–4000 cm^{-1} with a 16 cm^{-1} resolution and 128 scans. The spectra of paper samples were collected and compared by correlating absorbance values corresponding to the same wavenumbers.

X-ray diffraction analysis

XRD (X-ray diffraction) analyses were performed using a PANalytical diffractometer (Netherlands, radiation at Cu Kα1 = 1.54 Å, 40 kV × 40 mA) with a scan step size of 0.026°. The samples were scanned in fixed time mode under a diffraction angle 2θ range of 4–60°. A tube of copper was used for the measurement. The mA and kV were adjusted.

Mechanical property measurements

Tensile strength tests were performed according to the ISO-1924-2 standards and the Technical Association of the Pulp and Paper Industry (TAPPI) T-494 [18, 19] with a TMI 84-56 tensile tester (horizontal) (Testing Machines, Inc., Holland) at a test speed of 25 mm/min. The paper samples for the tensile strength tests were ca. 250 mm long and 15 ± 0.1 mm wide in the horizontal and transverse directions, respectively. The tensile strength tests were carried out in a standard atmosphere (RH of 50±2% and temperature of 23±1 ℃).

The folding endurance experiments were performed on a TMI 31–23 double folding endurance tester (Testing Machines, Inc., USA) according to ISO 5626:1993 [20], TAPPI/T511 [21] and GB/T 457-2008 [22]. The double-fold force was 175 per minute, and the applied force was 0.5 kg. The tested paper samples were ca. 140 mm long and 15 ± 0.1 mm wide in the horizontal and transverse directions, respectively. The folding endurance tests were carried out in a standard atmosphere (RH of 50 ± 2% and temperature of 23 ± 1 ℃). The reported values were the average of ten determinations.

The prepared paper strips were free of abnormalities, wrinkles and creases. The reported values were the average of ten determinations.

Microscope examination

The morphology of the surface of the samples before and after irradiation was investigated using a Hitachi S-4800 instrument. Scanning electron microscopy (SEM) images were recorded and operated at a 5 kV working voltage. All specimens were sputter coated with gold prior to examination.

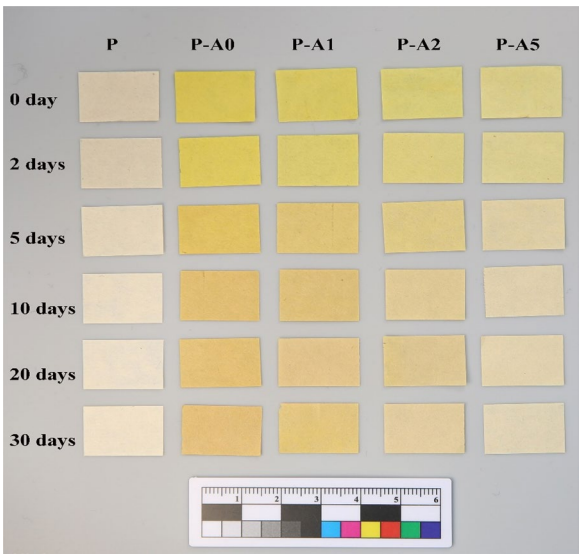


Fig. 1 Changes in color shades for different samples with ageing time

Results and discussion

Evaluation of color changes

Figure 1 shows a good range of color shades for the paper samples before and after artificial UVA accelerated degradation treatment with different concentrations for different irradiation times. The corresponding color difference (ΔE) and color coordinates are shown in Fig. 2 and listed in Table 1, respectively. From Fig. 1, the color of the extract-dyed paper was bright yellow according to the extract concentrations. Since light-induced color fading was a well-known characteristics of natural dyes, color fading was observed for the papers dyed with extracts in Fig. 1 at different irradiation times. According to the △E graph (Fig. 2), the progressions of ΔE were evident. Moreover, all samples showed a slow initial color change with △E slowly increasing in the first 2 days; this was followed by a rapid increase where △E rapidly increased, and then a slowdown in the color change occurred. The undyed and dyed papers had similar responses to UVA irradiation at the same time, despite the difference in intensity. Due to the fast color fading, the color difference of extract-dyed paper changed more than that of undyed paper during aging. A higher extract concentration correlated to a greater color change. The color changed to a great extent, showing a color difference of 4.73 for undyed paper (P) and 13.42 for P-A0 after irradiation for 5 days.

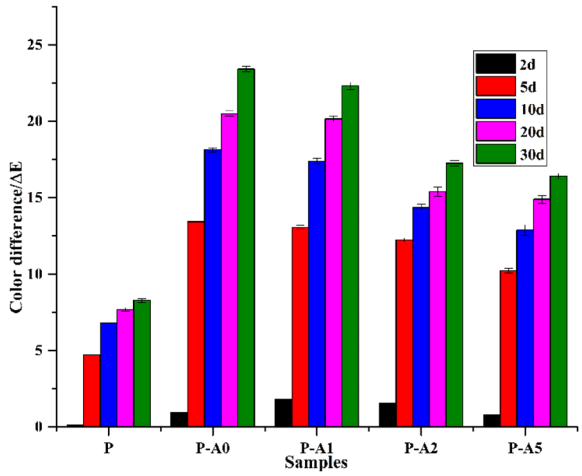


Fig. 2 Changes in the color difference with ageing time

To determine the difference in paper and amur cork tree bark extracts in response to UVA irradiation, the changes in L*, a* and b* of all samples were measured and are listed in Table 1. In this experiment for undyed paper, the increase in ΔL* meant that the paper became brighter; additionally, the slight decrease in a* and b* color coordinates with increasing irradiation aging time meant the loss of red to a more green tint and the loss of yellow to more blue tint, respectively. For the extract-dyed paper samples, the significant decrease in the L* and b* values and increase in the a* values with increasing irradiation aging times were evident and caused by the introduction of natural yellow amur cork tree bark colorant dyes. A higher colorant dye concentration correlated to a greater change in the color coordinates. The color coordinate change trend under the UVA accelerated degradation test was different from the dry-heating aging process and led to the different color results under UVA and dry-heating aging conditions.

Table 1 Changes of color coordinates with ageing time

Ageing time	Colour parameters	P	P-A0	P-A1	P-A2	P-A5
2d	ΔL	0	− 0.47	− 0.35	− 0.39	− 0.04
	Δa	− 0.08	0.8	0.68	0.48	0.27
	Δb	− 0.1	− 1.63	− 1.32	− 0.47	− 0.29
5d	ΔL	4.66	− 4.58	− 3.66	-— 1.91	0.5
	Δa	− 0.36	6.75	6.36	5.62	4.43
	Δb	− 0.58	− 10.6	− 10.78	− 10.57	-9.2
10d	ΔL	6.58	− 6.06	− 4.39	− 1.78	0.88
	Δa	− 0.67	7.79	7.25	6.42	4.61
	Δb	− 1.61	− 15.21	− 15.02	− 14.49	− 11.92
20d	ΔL	7.35	-— 6.11	− 3.6	− 1.85	1.95
	Δa	-0.8	8.08	7.32	6.55	4.85
	Δb	− 2.14	− 18.4	− 17.1	− 16.0	− 13.84
30d	ΔL	7.83	− 6.3	− 3.37	− 0.57	2.94
	Δa	− 0.92	8.0	7.26	6.32	4.64
	Δb	− 2.52	− 21.06	− 20.84	− 18.19	− 15.46

Mechanical characterizations

Mechanical properties are an important direct indication of polymer properties. Colorant fading is usually associated with changes in characteristics, such as mechanical properties. The average mechanical values of tensile strength and folding endurance with different extract concentrations at different aging times are shown in Figs. 3 and 4, respectively. The mechanical properties of handmade paper are different in terms of the orientation along the transverse direction (TD) and longitudinal direction (LD). The mechanical results showed that the addition of amur cork tree bark extracts greatly increased the mechanical properties of the paper, especially along the longitudinal direction (LD) of the paper samples, which was ascribed to the electrostatic interaction due to the chemical reaction between the colorants and paper fibres and contained sugar materials that covered the fibre surface. Artificial UVA accelerated degradation decreased mechanical properties. The higher the concentration was, the faster the decrease. As time went on, the mechanical properties of samples with different amur cork tree bark extract concentrations tended to approach; moreover, the mechanical properties for all dyed samples were larger than those for undyed paper at the same irradiation time. According to the mechanical results, the fastest tensile strength decrease rate occurred at 10 d. However, the fastest folding endurance change rate occurred after irradiation for 2 days. These results indicated that the folding endurance was easily affected under UVA accelerated degradation conditions. Compared with the dry-heat aging results, artificial UVA accelerated degradation induced a greater mechanical decrease.

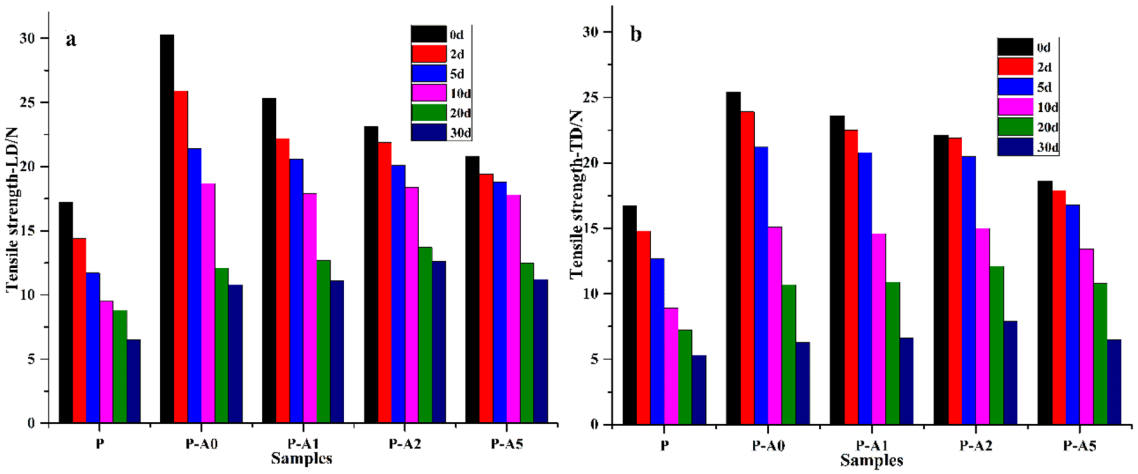


Fig. 3 Changes in tensile strength for samples with ageing time

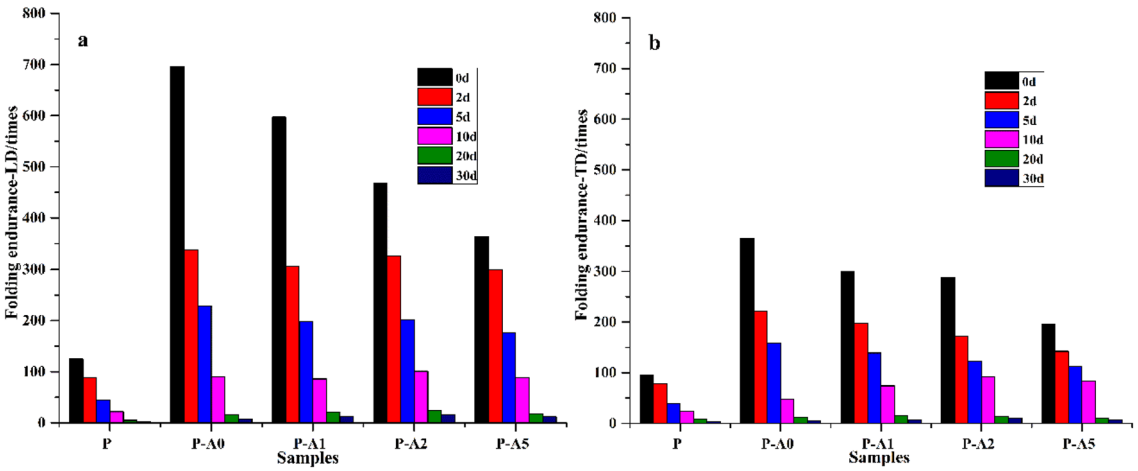


Fig. 4 Changes in folding endurance for samples with ageing time

SEM observations

Mechanical properties are usually accompanied by some physical changes, such as changes in morphological structure. SEM was used to observe the surface morphology of the samples before and after accelerated aging. Representative typical SEM images with different concentrations of amur cork tree bark extracts before and after being subjected to UVA irradiation for 30 days are shown in Fig. 5. SEM images showed that all paper fibers were destroyed to various degrees after UVA accelerated degradation. Seriously broken fibers were clearly observed for the undyed paper (P-30) and the amur cork tree bark extract dyed paper P-A0-30 and P-A1-30. Broken fibers and holes in P-30 and P0-30 indicated that fibers of both undyed and undiluted extract-dyed paper were more seriously destroyed after 30 days of aging than other samples. Fewer broken fibers were observed on the surface of P-A2-30; thus, P-A2 was not damaged as much as the other samples. We have reported that P-A0 and P-A1 led to the agglomeration of color dyes on the paper surface, however, the colorant-dyed paper surface exhibited better dispersibility than that of higher extract concentrations when the concentration of extracts were at A2 [1]. For P-A5-30, due to the fewer extracts, the paper fibers were evidently destroyed compared with P-A2-30. Similar to the results of dry-heat acceleration aging, SEM results showed that a suitable concentration of amur cork tree bark extracts, which could maintain the best dispersibility of colorant dyes on the paper surface, could improve the stability of paper samples.

Thermogravimetric analyses

Polymer thermal stability could be affected by changes in mechanical properties. A TG experiment was conducted to ensure the thermal stability of paper samples before and after artificial UVA accelerated degradation, and T_i(℃) (the initial decomposition temperature) and T_{max}(℃) (the maximum decomposition temperature) before and after UVA accelerated degradation treatment at different times with different extract concentrations are listed in Table 2. From Table 2, the thermal stability of all samples decreased with aging time and with increasing extract concentration. When the concentration of extract was lower than A2, a higher concentration of the amur cork tree bark extracts correlated to a lower initial decomposition temperature (T_i) and the maximum decomposition temperature (T_{max}). The T_i and T_{max} values for P-A5 were slightly higher than those for undyed paper during its entire aging process. According to our previous study, when the concentration of amur cork tree bark extract was larger than A2, agglomerations could be observed, which decreased the interaction between the main components and paper fibers [1]. The A2 concentration of amur cork tree bark extract exhibited relatively good dispersibility when dyeing with paper samples and improved the thermal stability of the paper samples by the stronger hydroxyl-nitrogen bond and hydroxyl groups and viscous substances.

Spectrum characterizations

From the X-ray diffraction patterns of paper samples before and after UVA aging tests (Fig. 6), the signals at $2\theta = 15.23°$ and 22.23°, which corresponded to the (101) and (002) planes of cellulose, shifted to approximately 15.68° and 22.91°, indicating chemical bonding between the paper fibers and colorant dyes. Under artificial UVA accelerated degradation conditions with different extract concentrations at different aging times, the 2θ for aged samples was separated into two groups: signals at $2\theta = 15.23°$ and 22.23°, which were ascribed to P-30 and P-A5-30, and signals at $2\theta = 15.38°$ and 22.39°, which were ascribed to P-A0-30, P-A1-30 and P-A2-30 paper samples. The difference between the two groups was attributed to the concentration of the amur cork tree bark extracts. Therefore, the XRD results showed the degradation led to the transformation and generation of compounds under artificial UVA accelerated degradation tests.

The FTIR spectra of paper samples with different extract concentrations at different aging times are presented in Fig. 7. Molecular changes could be observed clearly. The shoulder bands at 1641 cm^{-1} and 1600 cm^{-1} formed one peak at 1638 cm^{-1}; the band at 1513 cm^{-1} disappeared after aging for 5 days. Moreover, a new signal at 1728 cm^{-1} was observed for aged samples according to the extract concentrations and aging times. Specifically, a higher concentration and longer aging time correlated to a stronger peak at 1728 cm^{-1}. The peak at 1728 cm^{-1} was identified as the C=O stretching (asymmetric) of a carboxylate group in the samples; thus, some acidic products were produced during the UVA aging process.

pH tests

The amur cork tree bark extract is weakly acidic (pH = 5.6), and the original undyed paper is slightly alkaline (pH 7.8). The extract-dyed papers demonstrated weak acidity to weak alkalinity due to the residual alkaline materials in the paper neutralizing the weak acidic compounds in the extracts [1]. The progressions of the pH change before and after artificial UVA accelerated degradation treatment with different concentrations for different times are shown in Fig. 8; the results showed that the acidic compounds were produced during the irradiation aging process. According to Fig. 8, the decrease in pH for P-A0 was greater than that for

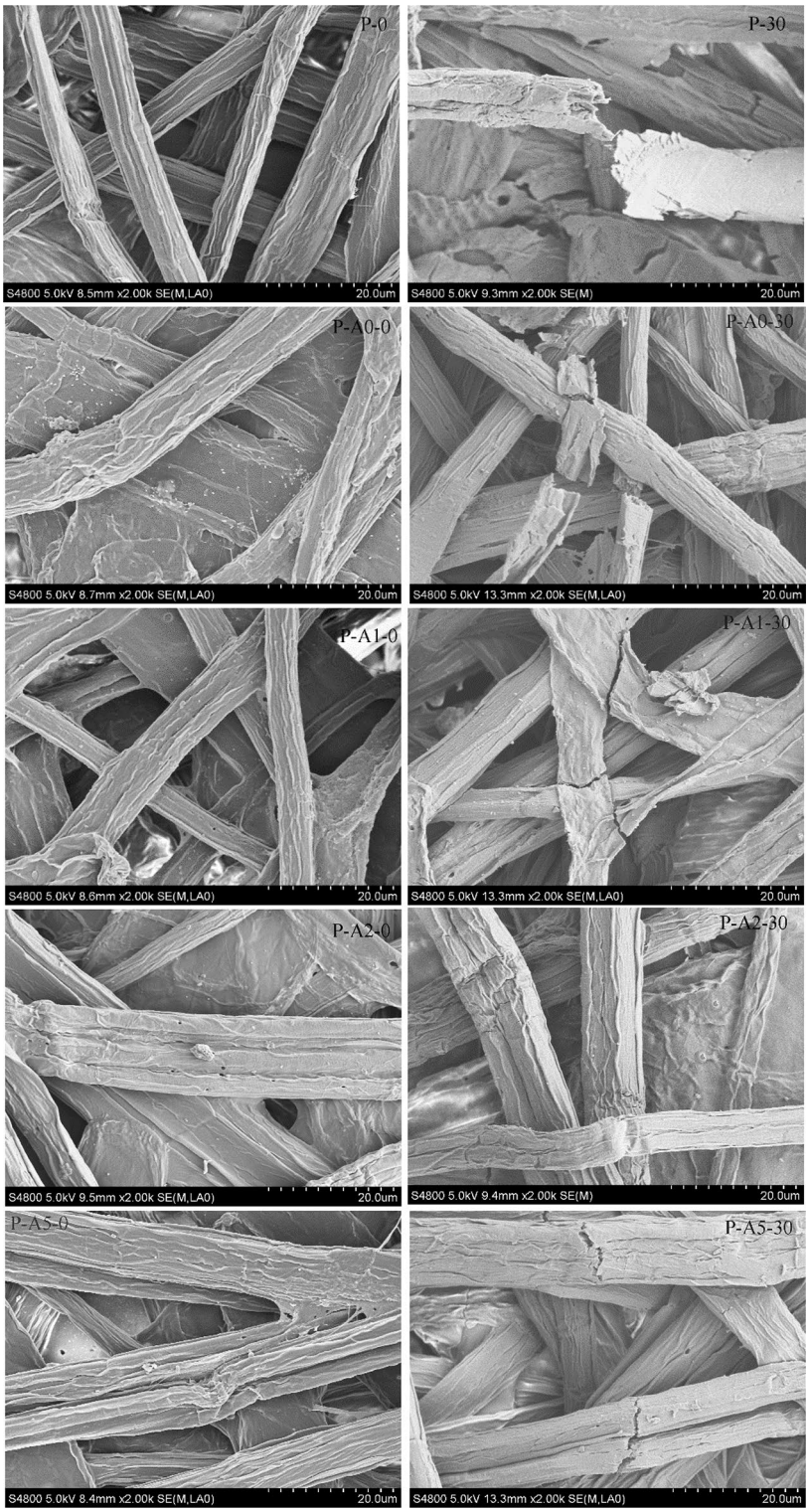


Fig. 5 SEM photographs for the surface of P, P-A0, P-A1, P-A2 and A5 with ageing time. (P-0, P-A0-0, P-A1-0, P-A2-0, P-A5-0: 0day; P-30, P-A0-30, P-A1-30, P-A2-30, P-A5-30: 30days)

Table 2 TG data of samples with ageing time

Ageing time	0d(℃)		5d(℃)		10d(℃)		20d(℃)		30d(℃)	
	T_i	T_{max}	T_i	T_{max}	T_i	T_{max}	T_i	T_{max}	T_i	T_{max}
P	328.4	348.1	326.2	346.9	322.3	342.2	319.4	338.8	312.1	334.1
P-A0	323.7	348.9	322.1	346.1	320.8	343.2	318.9	340.3	310.5	335.9
P-A1	325.4	350.4	323.5	347.7	321.5	344.1	320.1	341.2	312.3	336.4
P-A2	327.9	352.1	325.4	348.9	322.9	346.8	321.6	342.6	314.7	338.1
P-A5	327.8	350.8	325.8	347.3	322.1	343.4	321.1	339.3	313.4	335.6

T_i: the initial decomposition temperature; T_{max}: the maximum decomposition temperature

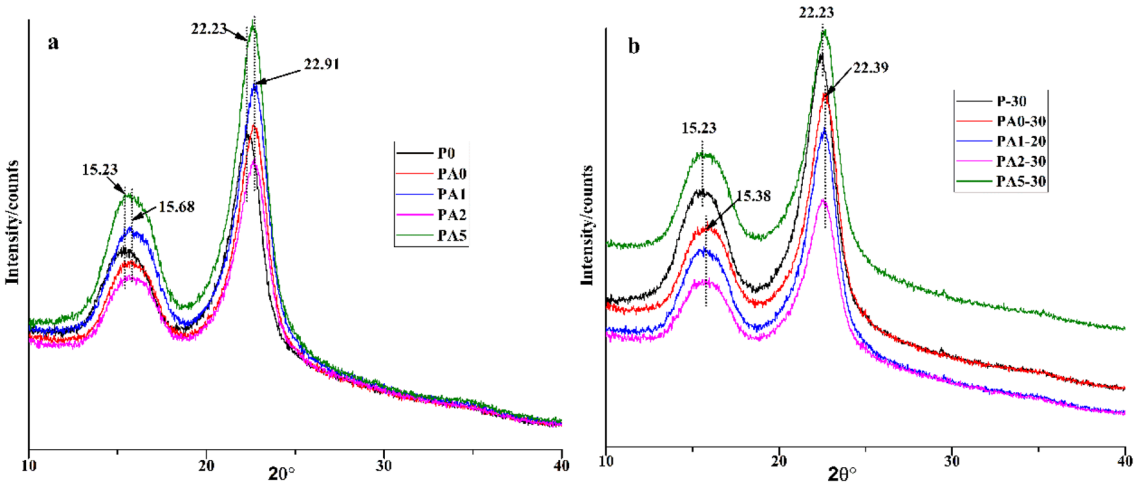
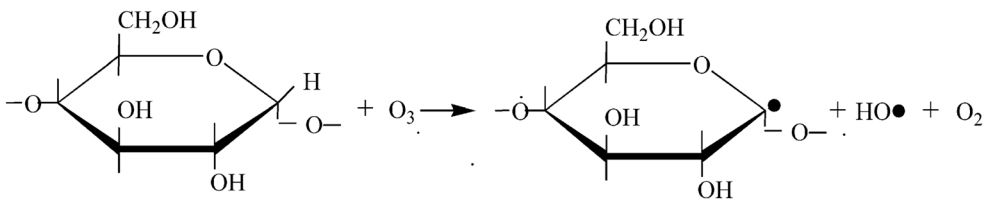


Fig. 6 Changes in XRD for samples with ageing time

the other samples and resulted in a pH of 4.7 when aged for 30 d. Cheun reported that some materials, including acidic products, could be produced during the aging process for the extracts from the amur cork tree bark [23]. Therefore, the accumulated components from degraded samples led to a further decrease in pH. The lower pH of dyed paper before aging led to a lower pH after UVA accelerated degradation, which demonstrated that the residual weakly acidic compounds in dyed paper potentially accelerated the degradation of the paper samples.

The aging of the paper under UVA irradiation conditions involved several main processes: UVA absorption, chemical bond cleavage and oligomer fragment formation. It was reported that the general mechanism of cellulose under UVA irradiation was a free radical process based on its polysaccharide structure [24–26]. Ying et al. demonstrated that traditional handmade paper could be photosensitized to H_2O_2 and $O_2^{\bullet-}$ by photoinducing electron transfer under UVA irradiation [27–29]. Moreover, UV light could lead to the production of various reactive species by UV photons interacting with molecular oxygen to promote formation of ozone molecules [30]. Ozone could electrophilically attack the anomeric carbon atom (C_1) on cellulose, either by interacting with the hydrogen atom of the glucose ring or breaking the glycosidic bond; this would detach C_1 and form a macroradical and carbon-centered radical by a radical chain reaction [24, 29, 31], as follows:



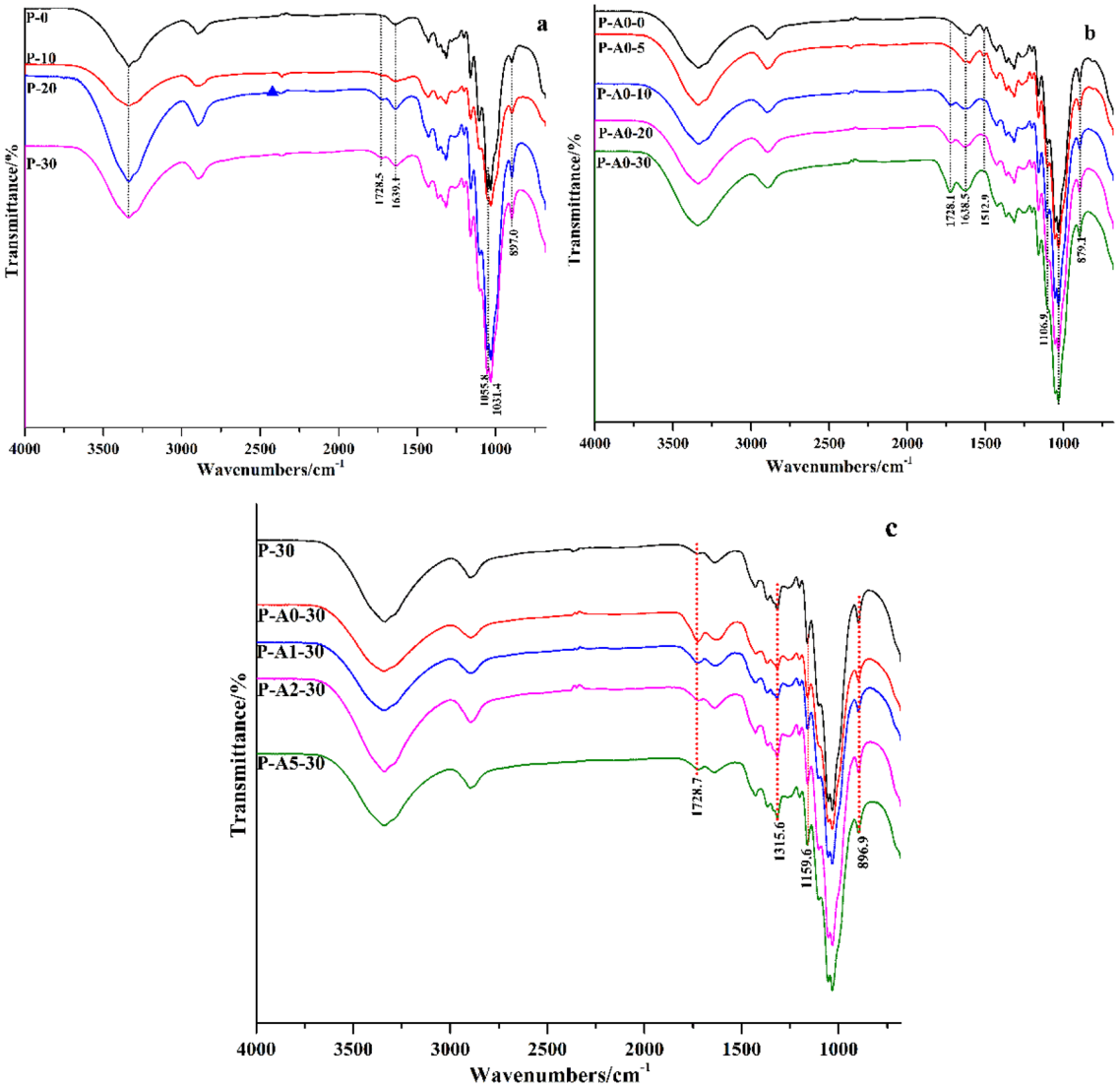


Fig. 7 FTIR results for samples with ageing time

Reactive oxygen species, such as OH•, $O_2^{\bullet -}$, and H_2O_2, initiate the degradation reaction by attacking the paper fiber polymers to accelerate chain cleavage and generate carboxylic acids, carbon dioxide, water, and oligomers.

It was reported [23] that the amur cork tree bark extracts under UV irradiation treatment produced isobenzofuran-1,3-dione 4,5-dimethoxy, 1,3-dioxolo[4,5-g] lisoquinolin-5(6H)-one,7,8-dihydro, canthine-6-one and dihydroberberine accompanied by the disappearance of quinic acid and other products. The factors that affect the UVA accelerated degradation tendency of the paper should affect the degradation of paper. In the case of papers that are dyed with the amur cork tree bark extracts, the uniform dispersed extract on the paper surface improved the paper's stability by forming electrovalent bonds, as indicated by the TG and mechanical results. Moreover, as a flavonoid compound, berberine could directly remove some hydroxyl free radicals and superoxide free radicals via single electron transfer and could partly inhibit the paper aging with a certain concentration of color dyes. However, existing weakly acidic residues could not be neutralized when the concentration of the extract liquor was high and led to agglomeration on the paper surface, such as P-A0 and P-A1, which accelerated the degradation of paper fibers. Additionally, agglomerated color extracts could reduce the opportunity to form hydroxyl free radicals with paper cellulose, which led to the further degradation of paper fibers.

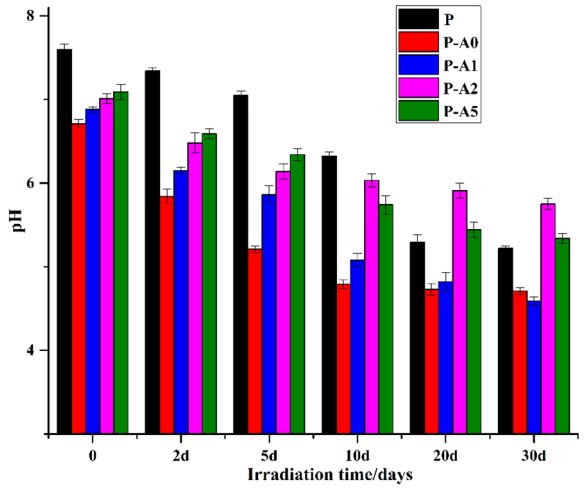


Fig. 8 Changes in the pH values for samples with ageing time

Conclusions

This study investigated the effect of amur cork tree bark color extract on handmade paper under artificial UVA accelerated degradation conditions. The handmade Daqian paper was dyed with different concentrations of amur cork tree bark extracts by diluting the original color dyes (A0) to different concentrations using deionized water. The prepared samples were subjected to a UVA accelerated degradation test under four 40 W fluorescent ultraviolet sunlamps (UVA-340) at 50 ℃ without water spraying. Structural and compositional characteristics were determined by color, pH, SEM, TG, XRD, μ-FTIR, and mechanical property changes. As an alkaloid compound, berberine could remove the free radicals formed during the paper UVA irradiation process via single electron transfer, form chemical bonds between the paper fibers and could potentially inhibit paper photodegradation to a certain extent. High concentrations of extract liquors led to the agglomeration of colorant dyes and decreased the opportunity to form interactions between paper fibers. Moreover, the weakly acidic extracts decreased the pH of the paper samples and further led to weakly acidic conditions for high extract concentration dyes. The present photodegradation artificial study showed that a water/extract liquor ratio of 2:1 (*v/v*) was a suitable concentration for the preservation of amur cork tree bark extract-dyed paper.

In conclusion, our results confirmed that traditional amur cork tree bark extract dyes affected the irradiation stability of dyed handmade paper. Combined with previous dry-heating artificial research, for color fading natural dyes, the concentration of colorant dyes need to be considered when amur cork tree bark extracts are used to restore cultural paper relics.

Abbreviations

ISO	International Organization for Standardization
RH	Relative humidity
TAPPI	Technical association of the pulp and paper industry
LD	Longitudinal direction
TD	Transverse direction
SEM	Scanning electron microscopy
XRD	X-ray diffraction
TGA	Thermogravimetric analysis
FTIR	Fourier transform infrared spectrometer
μ-ATR	Micro-attenuated total reflection
CIE	International commission on illumination
P	Undyed paper samples
A	Amur cork tree barks
P-A0, P-A1, P-A2, P-A5	Indicated the water/extract liquor ratios of 0:1. 1:1, 2:1 and 5:1, respectively

Acknowledgements

The authors grtefully acknowledge Lingzhu Yu for her help with SEM measurements and Liang Zhang for his assistants with mechanical tests.

Author contributions

Data of the research work were collected by YBL, MJR and YFW. XJZ and YBL prepared the manuscript. YBL revised it. All authors read and approved the final manuscript.

Funding

The Science and Technology Support Programme of Sichuan Province (Grant No.:2023YFS0494) and the Joint Funds of the National Natural Science Foundation of China (Grant No.: U19A2045).

Availability of data and materials

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Competing interests

The authors declare no competing interests.

Received: 5 August 2023 Accepted: 27 November 2023
Published online: 06 December 2023

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