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FIXATION OF DYE-TYPE RED AND BLUE INK HANDWRITING ON AGED PAPER

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ABSTRACT

In this article, red and blue ink handwriting on aged paper was fixed by fixative to prevent handwriting from fading or diffusion during aqueous deacidification. Firstly, four fixatives were selected: polydimethyl ammonium chloride (PDDA), the quaternary ammonium salt of chitosan (HACC), cationic guar gum (CGG), and benzyl triethyl ammonium chloride (TEBAC) alcohol solution, according to their fixation effects on filter paper dyes, and derive the optimal composition ratio of the above four fixatives. Experimentally derived that PDDA has an excellent fixation effect on red ink handwriting, and HACC has an excellent fixation effect on blue ink handwriting. In addition, HACC also has a positive impact on the conservation of handwriting in real archives. The mechanical properties of aged paper were improved, and the paper fibers had no obvious change after the fixation and deacidification processes.

KEYWORDS: Paper, handwriting, fixation, deacidification, archive protection.

INTRODUCTION

Recently, the preservation of aged paper materials, such as deacidification and reinforcement, has been gaining increasing attention. Previous research has pointed out that the prime causes of paper aging are the acid in the paper and the degradation of cellulose. So many researchers have taken some processes to remove acid from aged paper (Havlinova et al. 2002, Potthast and Ahn 2016, Sala et al. 2006, Tan et al. 2017). The treatment of deacidification and strengthening (Dupont et al. 2013, Piovesan et al. 2018, Souguir et al. 2012, Zhang et al. 2015) of paper files with water-soluble materials. Some essential paper files contain many different kinds of dye-type ink handwriting. Because aged paper needs to be exposed to aqueous (Li et al. 2019) or non-aqueous deacidification materials (Hubbe et al. 2017, Sequeira et al. 2006) during deacidification, if in the aqueous solution, ink handwriting would completely dissipate, which will seriously affect the value of the archive. According to a sample survey of archives from the Guangdong Provincial Archives, there is 33.83% of red and blue ink handwriting can diffuse rapidly in water. Therefore, the handwriting should be fixed first before deacidification.

The fixation mechanism of different fixatives is different due to the structures of dyes and different binding modes of fixative and fiber (Akbari et al. 2002, Rippon and Evans 2012). There are four types of the fixation mechanism: *(1)* Utilization of the film-forming property of fixative: high polymer and poly-amino compounds could be used as a fixative to form a layer of thin film on the surface of the dye molecules, which could isolate the dye molecules from the environment and improve the fastness of wet processes. These fixatives have neither reactive nor cationic groups (Carter 1996, He et al. 2009, Munoz 2007), they work universally on anionic and cationic dyes. One of the fixatives that use this binding mode is cyclododecane. It has a slow volatility and sublimate within a few days to protect the dye without changes. *(2)* Utilization of the cross-linking of dye fixative and pigment and fibers (Mughal et al. 2013, Tang et al. 2004). Reactive hydroxyl and epoxy groups can be cross-linked with reactive groups in dye molecules or hydroxyl groups on cellulose molecules to reduce the shedding of dyes. *(3)* With the utilization of the hydrogen bond attraction between fixative and fiber, some groups of the molecular structure of fixative and fiber can be combined with dye and fiber to produce a hydrogen bond or van der Waals force (Luo et al. 2020). This method is commercially available for protecting fibers on an industrial scale. *(4)* Utilization of the reaction of fixative and the dye-containing sulfonate of the acid dye of the groups or carboxylic acid salt. Insoluble compounds would be generated on the fiber with the reaction of cationic fixative and anionic dye, which can isolate dye molecules of water-soluble groups from the water solvent deacidification processes (Bredereck and Grabenstein 1988).

Initially, the fixative on the market was based on formaldehyde, which can improve the durability of the fixative on textiles by combining hydroxymethyl with the hydroxyl group on cellulose, making the color bright and shiny. However, this also increases the possibility of free formaldehyde, the release of formaldehyde in textiles exceeds the standard. Therefore the development of an efficient formaldehyde-free color fixative is the current development trend (Adair and Moore 1988).

In this study, aged newspapers and rice paper were used as ink writing carriers to simulate fixation and deacidification experiments. Dye molecules of red and blue ink were used to screen fixation materials and used to prepare fixatives and further adjust the composition of fixatives. The main component pigment of red ink azophloxine (AP), eosin Y (Y), and methyl orange (MO), and the main component pigment of blue ink direct blue (DB) and methyl blue (MB) were used. The effects of fixation and deacidification experiments on whiteness, mechanical properties and paper fibers were investigated. The optimized fixatives were applied to practical applications.

MATERIAL AND METHODS

Rice paper was produced by Anhui arching Rice paper art co. LTD; Aged newspaper (The Chinese Journal, 1980) was provided by Guangdong University of Technology Library; Filter paper (φ = 7 cm) was produced by Hangzhou WoHua filter paper co. LTD. Real archive was provided by the Finance Department of the Guangdong University of Technology. Blue ink (B-ink) and red ink (R-ink) were produced by Shanghai fine culture products co. LTD; direct blue, methyl blue, azophloxine, eosin Y, and methyl orange were produced by Shanghai McLean biochemical technology co. LTD. Polydiene dimethyl ammonium chloride (PDDA, 20 wt.%) and benzyl triethyl ammonium chloride (TEBAC, 98%) were produced by Aladdin. Chitosan quaternary ammonium salt (HACC) was produced by Guangdong haojiang chemical reagent co. LTD and cation guar gum (CGG) was produced by Qingdao Youso Chemical Technology co. LTD. Polycationic waterborne polyurethane, quaternary ammonium salt-7, quaternary ammonium salt-11, and cationic polyacrylamide were produced by Qingdao runsheng powder materials co. LTD. Other Aladdin products chemicals, with a purity higher than 99 wt. were also used.

Paper dyeing

Y aqueous solution, AP aqueous solution, and MO aqueous solution were prepared with the same mass fraction of 1.0%. MB aqueous and DB solutions were prepared with the same mass fraction of 0.2%. All these solutions were prepared with a specific temperature of 25□. Seven kinds of dye, including R-ink, B-ink were diluted with distilled water by 50:50(V/V) for sample preparation. Filter papers were immersed in the dye solutions for 1 min, then taken out and placed on dry glass dishes to dry naturally at room temperature for 24 h.

Preliminary screening of the fixation preparation

Polycationic waterborne polyurethane, quaternary ammonium salt-7, quaternary ammonium salt-11, cationic polyacrylamide, and non-aldehyde fixative was prepared as an aqueous solution and the mass ratio of 1:1 with water. PDDA was added into the solvent consisting of water and ethanol (the mass ratio of water to ethanol to polydiene dimethyl ammonium chloride was 1:1:1), and cationic cellulose was treated in the same way as polydiene dimethyl ammonium chloride. HACC and CGG were prepared as an aqueous solution, and the mass fraction was 1%and 0.1%, respectively. TEBAC alcohol solutions were prepared with a mass fraction of 20%.

Preparation of fixatives

PDDA aqueous solution was prepared in accordance with the mass ratio (PDDA/ethanol/water) 1:1:0.6, 1:3:1.2, 1:5:2.0, 1:7:3.0 and 1:9:3.6. HACC aqueous solution was prepared by mass fractions of 0.5%, 1.0%, 1.5%, 2.0% and 2.5%, respectively, then mixed with ethanol (100%) at mass ratio of 1:1. CGG was prepared into aqueous solutions of 0.1%, 0.2%, 0.3%, 0.4% and 0.5%, respectively, and a small amount of dodecyldimethyl betaine was added to help the dissolution of CGG. TEBAC was prepared into ethanol solutions of 10%, 20%, 30%, 40% and 50%, respectively. The prepared fixatives were brushed on the surface of the filter paper samples, and then samples were placed in an environment with a temperature of 23□ and relative humidity of 50% for more than 72 h.

Fixation handwriting treatment

Aged newspaper and rice paper were written “红墨水” and “蓝墨水” on the blank of the paper pattern, respectively, with R-ink and pure B-ink, and placed in an environment with a temperature of 23□ and relative humidity of 50% for more than 72 h.

Deacidification treatment

Paper samples were immersed in the calcium propionate aqueous (5%, pH = 8.79) for 10 min, then placed in an environment where the temperature was 23□, and the relative humidity was 50% for 24 h.

Artificial aging experiment

Papers were aged at 105□ for 72 h in an electric thermostat (ISO5630-1: 1991). After being placed on dry glass dishes for 24 h with a temperature of 25□ and a humidity of 50%, tested the properties of paper accelerated aging at 105□ for 72 h (Wójciak 2016), which theoretically equates to 25 years of natural aging (Barrow 1963, Zervos et al. 2014).

Colorimetric measurement

A color reader (CR-10) was used to test the whiteness of the samples. The whiteness of each paper was the average value of 8 times the measurements performed at the same conditions. The CIE1994 (L, a, b)* system was the chromatic coordinates standard according to ISO 11467: 2000. The ΔE was used to indicate the total color difference, which was based on the (L, a, b)* values measured before and after treatment or before and after artificial aging for targeting paper samples (Pan 2019). ΔE was determined as:

$$\Delta E=\left[\left(\Delta L^*\right)^2+\left(\Delta a^*\right)^2+\left(\Delta b^*\right)^2\right]^{0.5} \qquad (1)$$

The value of L* indicates the brightness coefficient, reflecting the luminance of the measured position: The larger the value, the closer to white. Conversely, the closer to black (100 white, 0 black). The larger the value of a*, the closer to red. Conversely, the closer to green. The larger the value of b*, the closer to yellow. Conversely, the closer to blue (Dotson 1991).

SEM

The morphological fiber analysis of the untreated and treated paper samples was performed with an SEM (S-3400N-, Hitachi Ltd., Japan). The paper sample is cut to a suitable size and attached to the sample table with carbon conductive tape. The fiber morphology is observed after gold spraying with an ion-sputtering instrument.

RESULTS AND DISCUSSION

Selection of fixed materials

As shown in Tab. 1, the preliminary screening of a variety of fixed materials was conducted, among which blank experiment and no aldehyde fixative were used as screening references. For the R-ink, PDDA and HACC had a better fixation effect, and the values of the total color difference were 7.1 and 10.5, respectively. For the B-ink, HACC, CGG, and TEBAC had a better fixation effect, and the values of the total color difference were 9.5, 10.8, and 7.7, resp. Therefore, PDDA, HACC, CGG, and TEBAC were selected as fixed materials, which were used for the proportioning optimization and subsequent deacidification experiments.

Tab. 1: Values of the total color difference ΔE (-) of fixed materials for R-ink and B-ink.

Materials	R-ink	B-ink
Blank samples	31.6	26.9
PDDA	7.1	13.7
Polycationic waterborne polyurethane	22.2	19.3
Quaternary ammonium salt -7	13.6	18.9
Quaternary ammonium salt -11	15.6	21.3
HACC	10.5	9.5
Cationic polyacrylamide	21.3	20.7
CGG	11.3	10.8
Cationic cellulose	14.5	12.7
TEBAC	12.4	7.7
No aldehyde fixative	7.8	15.1

Optimization of fixative

In order to better dissolve PDDA in ethanol aqueous solution, the ternary liquid phase diagram of PDDA, ethanol, and water was drawn to control the amount of water and find the critical line to minimize the effect of the water in fixatives on the color or handwriting of aged paper, as shown in Fig. 1. Point A represented the soluble point when the PDDA ethanol solution was added to water. Point B represented the three-phase miscibility points. Five different proportions of PDDA fixatives were prepared according to the phase diagram, and the mass ratios (PDDA/ethanol/water) were 1:1:0.6, 1:3:1.2, 1.2:5:2.0, 1:7:3.0, and 1:9:3.6.

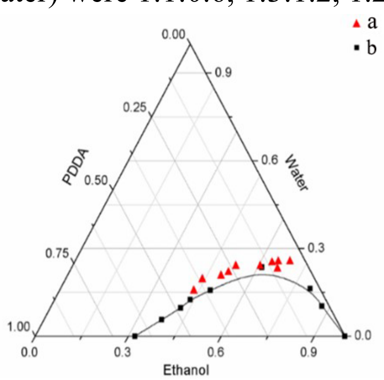

Fig.1: Three-phase diagram of PDDA, ethanol, and water (a-experiment preparation concentration; b-critical path point).

The total color difference values of samples dyed with MO, Y, AP, MB, and DB before and after being soaked in water were 37.4, 36.6, 39.7, 51.2, and 11.1, respectively. The total color difference values of samples dyeing with R-ink and B-ink soaked in water directly were 31.6 and 26.9, resp. By increasing the ratio of anhydrous ethanol, the fixation effect of PDDA was enhanced first and then reduced. The best quality ratio of PDDA, ethanol, and water was 1:5:2. After being treated with PDDA fixative at the best ratio, the total color difference values of samples dyed with MO, EY, AP, MB, and DB before and after being soaked in water were 12.7, 9.8, 12.7, 38.6, and 8.2, resp. The samples dyed with red and blue ink were 2.1 and 7.5, resp.

The fixation effect of HACC on samples dyed with DB was not obvious. However, when the content of the HACC stock solution was 2.5%, the total color difference values of the sample were about 10 less than that of samples treated with HACC fixative at the other rations. That was probably because the viscosity of HACC increased at this ratio, so the shedding of MB would have been prevented. The effect of different concentrations of HACC on other pigment dyes was not much different. For red and blue ink, the higher the concentration of HACC, the better the fixation effect. However, HACC with high viscosity would change the appearance of aged paper. Therefore, the best content of the HACC stock solution was 1.5%.

The chemical structure of CGG is similar to the chemical structure of cellulose so that it can be well combined with paper cellulose and fix a part of the pigment on fibers. When the concentration of the cationic guar gum ranged from 0.1% to 0.5%, the samples' total color difference values were about 10. Considering the effects of viscosity, the best concentration of CGG in GCC fixative was 0.3%. After being treated with the GCC fixative with that concentration, the total color difference values of samples dyed with red ink and pure blue ink before and after soaking in water were 9.2 and 10.2, respectively.

The higher the concentration of TEBAC, the more obvious the fixation effect. However, excessive TEBAC concentration will affect the appearance of the paper, so the concentration of 30% TEBAC ethanol solution was selected. The total color difference values of samples dyed with R-ink and B-ink were 11.7 and 7.8, respectively.

The results demonstrated that the preliminarily screened PDDA, HACC, CGG and TEBAC have better color fixing effect. Due to the poor color fastness of most dyes, there are certain bleaching and dyeing problems, after controlling the concentration of fixative, the paper appearance is greatly guaranteed (Sun and Dong 2008, Zhou and Tang 2016). Fixing agent on MO, Y, AP, DB, red and blue ink fixing effect is very obvious. Because of the chemical instability of MB, the fixation effect is poor. PDDA overall color fixing effect is good, especially effective for red ink.

Fixation effect of fixative on R-ink and B-ink

The fixation effect of PDDA, HACC, CGG, and TEBAC on R-ink and B-ink were investigated in calcium propionate aqueous (5%, pH = 8.79). The results are shown in Fig. 2. PDDA has the best fixation effect on R-ink in the four kinds of fixative, and HACC on B-ink, respectively. The total color difference values of filter paper with R-ink are 2.8 after deacidification and 2.3 after artificial aging. So PDDA is an excellent fixative for R-ink because of its shallow effect. Similarly, HACC is a significantly more excellent fixative for B-ink.

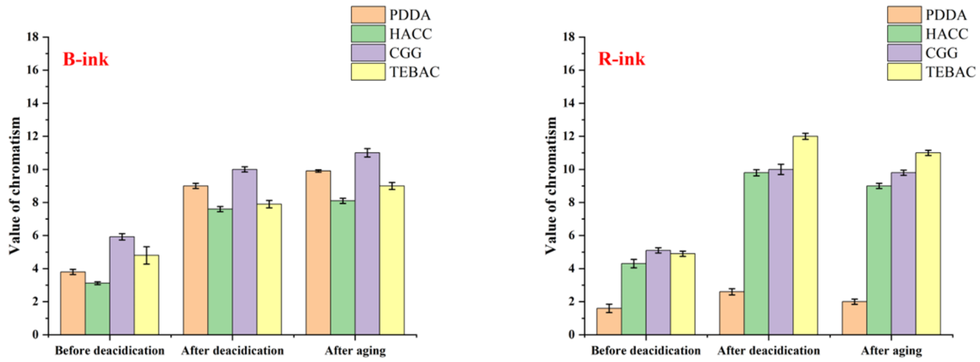

Fig.2: Fixation effect of fixatives on red ink and blue ink.

In Fig. 3a and c, the samples were dyed with R-ink and B-ink, respectively. Then PDDA and HACC were brushed on the right side surface of the red sample and blue sample, respectively. After fixation, the samples were soaked in calcium propionate aqueous (5%, pH = 8.79) for 10 min. The results are in Figs. 3b and d. The left of papers without fixation was severely faded. The R-ink on the filter paper was almost completely faded, which was more severe than the B-ink. However, the R-ink on the right side of the paper with fixation held up more perfectly than B-ink (Xing et al. 2022).

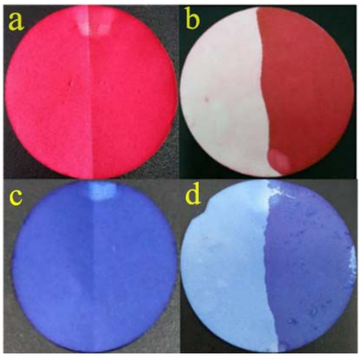

Fig. 3: Fixation effect of fixative on filter papers: (a, c) before treatment; (b, d) after fixation and deacidification.

Effect of fixative on handwriting after deacidification

Since paper files containing ink handwriting are very valuable, the deacidification process of handwriting protection was first studied through simulation experiments. In the paper archive, newspaper and rice paper are the main carriers of information, so the simulation experiment uses these two paper samples for fixed color deacidification. Paper samples of red and blue handwriting were prepared, PDDA and HACC fixatives were brushed on the aged newspaper (The Chinese Journal from 1980) and rice paper signed by orange frame in Fig. 4. PDDA fixative was used on red handwriting, and HACC fixative was used on blue handwriting. In Fig. 4a and b for the aged newspaper, the unfixed red ink handwriting faded and scattered obviously after deacidification. However, the fixed handwriting faded a little and was kept clear. Blue ink handwriting was relatively kept well than red ink handwriting, as shown in Fig. 4c and d. The results were much better for the rice paper, as shown in Figs. 4e to h. In a word, PDDA and HACC fixatives had basic ideal effects on red handwriting and blue handwriting, resp.

Fig. 4: Fixation effect on the handwriting of the aged newspaper (a-d) and the rice paper (e-h) before and after fixation. (a, c, e, g) before treated; (b, f) red handwriting after fixed and deacidification; (d, h) blue handwriting after fixed and deacidification.

From these results it is clear that since red and blue inks contain water-soluble anionic dyes (Kirby and Cates 1986), the use of cationic color fixative can be combined with anionic dyes to produce water-insoluble precipitates (Liu and Zhao 2017), so as to achieve color fixation, to a large extent, to avoid blotting, spreading and other phenomena when the handwriting meets the water. In particular, it showed a good fixation effect in the fixation and deacidification experiment of simulated paper archives of newspaper and rice paper (Fig. 4), which was conducive to the further fixation of physical paper archives.

Effect of fixative on the whiteness of paper AFTER DEACIDIFICATION

The total color difference value of the prepared samples of the aged newspaper and rice paper before and after being treated with PDAA or HACC were all lower than 0.5. According to the evaluation criteria for chromatic aberration, this color difference change belongs to the non-discoloration level. As shown in Tab. 2 and Tab. 3, after deacidification, the L value of samples increased significantly, much more than the change of the a and b values.

Tab. 2: Effect of PDDA on the total color difference value ΔE (-) of paper.

Sample		Blank	After fixation	After deacidification	After aging
Newspaper	L	74.7 ± 0.2	74.1 ± 0.1	76.3 ± 0.1	75.4 ± 0.1
	a	5.9 ± 0.1	6.1 ± 0.2	6.1 ± 0.1	5.8 ± 0.2
	b	16.2 ± 0.1	16.3 ± 0.1	16.1 ± 0.2	17.3 ± 0.2
	ΔE	-	0.64	1.61	1.14
Rice paper	L	88.6 ± 0.1	88.7 ± 0.1	89.7 ± 0.1	88.5 ± 0.1
	a	4.6 ± 0.2	4.6 ± 0.2	4.8 ± 0.1	4.7 ± 0.2
	b	6.3 ± 0.2	6.5 ± 0.1	6.5 ± 0.2	6.8 ± 0.2
	ΔE	-	0.22	1.13	0.51

The larger the L value, the greater the paper brightness. There are two reasons for the increase in the brightness of the sample. The first reason could be possibly caused by calcium ions compounds on the paper improving the whiteness of the paper. The second reason is that the yellow degradation compounds produced by aged paper dissolve in the washing water, increasing the paper's brightness. The results showed that the total color difference value of samples after fixation and deacidification was around 1.5, which the increase in L may cause. The paper will turn yellow for the paper archive that has been kept for a long time. If deacidification can improve the brightness of the paper, it is possibly beneficial to the paper.

Tab.3: Effect of HACC on paper value of the total color difference ΔE (-).

Sample		Blank	After fixation	After deacidification	After aging
Newspaper	L	74.6 ± 0.1	74.3 ± 0.2	75.3 ± 0.1	74.4 ± 0.1
	a	7.9 ± 0.1	7.5 ± 01	7.1 ± 0.2	7.8 ± 0.3
	b	17.2 ± 0.2	17.3 ± 0.1	17.1 ± 0.2	17.8 ± 0.1
	ΔE	-	0.26	1.07	0.64
Rice paper	L	88.8 ± 0.2	88.8 ± 0.1	89.9 ± 0.2	88.5 ± 0.2
	a	4.7 ± 0.1	4.8 ± 0.1	5.1 ± 0.3	5.2 ± 0.2
	b	6.4 ± 0.1	6.5 ± 0.1	6.8 ± 0.1	7.2 ± 0.2
	ΔE	-	0.14	1.53	0.99

From the effect of deacidification fixation on paper whiteness, after the application of PDDA and HACC on newspaper and rice paper, L, a and b values have little change, but the total color difference is less than 1.5 (Tabs. 2 and 3), which does not affect the appearance of paper pattern. Conform to the principle of preserving the original form of archives and the principle of minimal intervention. Calcium propionate deacidification solution can improve the whiteness of paper to a certain extent. After artificial aging, the change value of paper color difference is small (Zhang 2017), indicating that fixed color deacidification can alleviate the aging of paper (Yong 2020), reflecting a certain anti-aging ability.

Effect of fixative on paper fibers

Having the most excellent fixation effect on handwriting, PDDA was used to observe the effect on paper fibers. As shown in Fig. 5a, the aged newspaper is without treatment, 30 years of the aged paper has been acidified and yellowing, so the fiber has more messy and small particles. SEM showed that the fiber has no noticeable difference (Figs. 5a-c). Clean and full fiber is in rice paper (Fig. 6a). On Figs. 6b and c is presented respectively the fiber of rice paper with PDDA and rice paper without fixation after artificial aging. The fiber before and after aging also has no obvious difference, but fiber became wizened after artificial aging. The paper fiber has no obvious change of aged newspaper and rice paper before and after artificial aging. It can be stated that the color-fixative will not cause a negative effect on the paper for a long time.

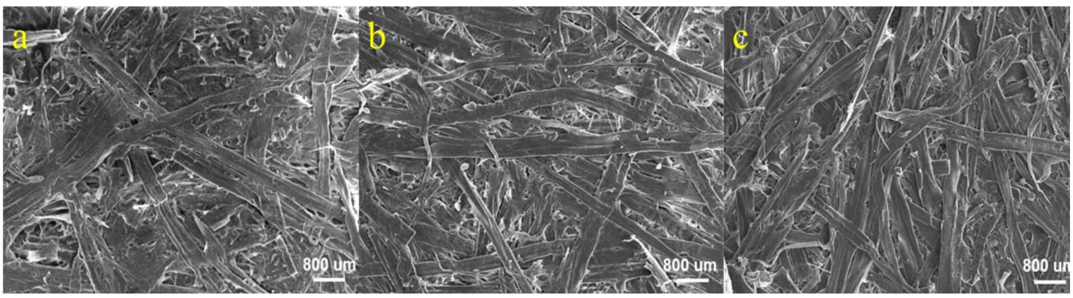

Fig. 5: SEM images of the fibers from aged newspaper: (a) untreated; (b) after fixation and aging; (c) untreated and after aging.

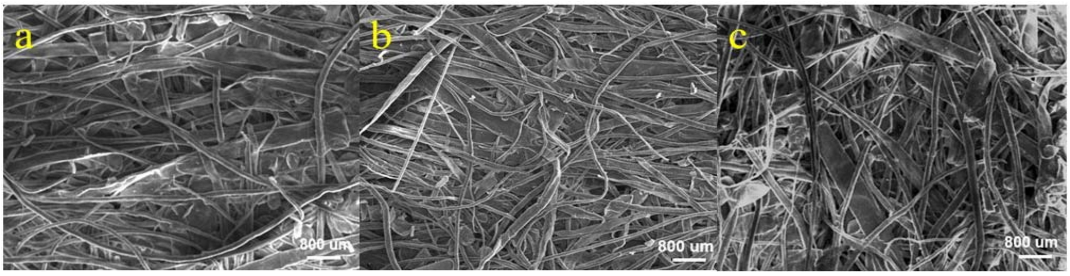

Fig. 6: SEM images of the fibers from rice paper: (a) untreated; (b) after fixation and aging;(c) untreated and after aging.

These results suggest that the old newspaper fiber after the fixative fixing aging has no obvious change, but the blank group after aging fiber fracture is serious, and after aging water loss, there is a wizened phenomenon; There is no significant difference between the fiber of rice paper after solid aging and the fiber of only aging without solid color, and there is no obvious change compared with the fiber of untreated, which indicates that the rice paper has good stability and durability. So, the fixative can protect the fiber of the paper to a certain extent (Reyden 1992).

Application of fixative in real object archives

The real object archives are precious, and the existing discarded archives are from the finance department of Guangdong University of Technology. HACC fixative to the handwriting on the discarded archives with B-ink was applied (Fig. 7). The blue handwriting has faded due to changes in time and environment (Fig. 7a). HACC fixative was brushed on the handwriting on the top right corner of the file. Then, the discarded archives were placed in an environment of 50% relative humidity at 23°C for 24 h before being placed in the acidifying agent (Fig. 7b). During the deacidification process, the unfixed handwriting (lower left corner) was obviously faded, while the fixed writing (upper right corner) almost had no change. After soaking for 10 min, dried the discarded archive naturally (Fig. 7c). Compared with Fig. 7a, the fixed handwriting was almost unchanged, and the fixative shows a good fixation effect. The handwriting without fixation faded after deacidification and could not maintain its original appearance. Therefore, HACC fixative has a significant effect on the fixation of handwriting. The pH of discarded archive before deacidification was 6.84. After deacidification, the pH increased to 7.69. The archive was alkalescent, which could prevent further acidification and alleviate acidification hydrolysis of archival fibers.

The fixative with good color fixing effect in the deacidification simulation experiment is applied in the actual archives. Because the ink in the actual archives has a history of more than 20 years, the ink and paper are firmly combined, so the fixative can well cover the handwriting and fiber, and will not fall off with the ink, so that the fixative can play a better role (Es-sabbeur et al. 2023).

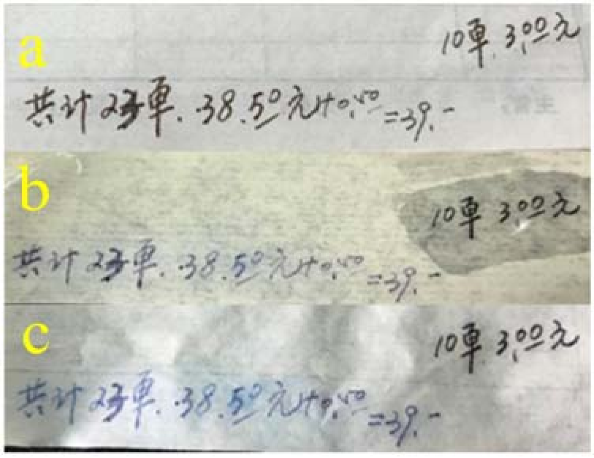

Fig. 7: Application of fixatives on the real object archives: (a) before treatment; (b) after fixation with HACC and acidification; (c) after drying.

CONSLUSIONS

In this study, PDDA and HACC were screened from 10 fixatives to protect R-ink and B-ink handwriting on aged paper. The results show that when the mass ratio of PDDA, ethanol and water is 1:5:2, the fixation effect of PDDA on R-ink handwriting is better, and the total color difference of R-ink dyed pattern decreases from 31.6 to 2.1 after being fixed. 1.5% HACC stock solution has a better fixing effect on B-ink handwriting, and the total color difference decreases from 26.9 to 7.2 after being fixed. HACC effectively protects the only remaining archives of real objects with B-ink handwriting. In addition, fixation and deacidification can alleviate the aging of paper, so that the paper has a certain anti-aging ability, the fixative can protect the paper fiber to a certain extent, the paper fiber after fixation and deacidification treatment has no obvious change by SEM observation.

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