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Synthesis of Carboxymethyl Cellulose-Acrylate-OvPOSS Graft Copolymer and its Application in Paper Protection

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As an important part of cultural heritage, paper cultural relics have important historical, scientific and humanistic values. However, a large number of paper cultural relics are suffering from aging, mildew, yellowing, cracks, embrittlement and other threats, or are even completely damaged. To enhance the strength of paper cultural relics and prolong the life of paper cultural relics, a novel CMC-acrylate-OVPOSS graft copolymer was synthesized with carboxymethyl cellulose (CMC), methyl methacrylate (MMA), butyl acrylate (BA), hydroxyethyl methacrylate (HEMA), glycidyl methacrylate (GMA), octavinyl polyhedral oligomeric silsesquioxane (OVPOSS). The synthesized materials were analyzed by infrared spectroscopy, X-ray diffractometer, thermal analyzer and X-ray photoelectron spectroscopy. In addition, the reinforcement solution was coated on the surface of the paper, and the protective effect of the reinforcement solution on the paper was evaluated by testing the mechanical and optical properties of the paper. When the reinforcement solution with a mass concentration of 10 % was coated on the paper, the mechanical properties of the paper were greatly improved, and the gloss and whiteness were basically unchanged.
Keywords: paper cultural relics, paper protection, carboxymethyl cellulose, acrylate.

1. INTRODUCTION

Papermaking technology is one of the four great inventions of ancient China. Paper is the crystallization of the long-term experience and wisdom of the working people in ancient China, and it has promoted the cultural development of China, Arabia, Europe and even the whole world [1]. As one of the four ancient civilizations, China's long history has left us a wealth of cultural heritage, among which paper cultural relics, as the main body of cultural inheritance, are rich in variety and number [2, 3]. However, with the influence of time, preservation conditions, historical reasons and other objective factors, a large number of paper cultural relics appeared mildew, yellowing, cracks, embrittlement and other problems, and even completely powdered, making many precious paper materials destroyed in the long history [4 – 6]. Therefore, it is a very important and urgent task to carry out the protection work of paper cultural relics and the research on the reinforcement and protection materials and techniques of paper cultural relics for a country with thousands of years of history and a large number of paper-based cultural relics.

Cellulose is the most abundant natural renewable resource on earth, which comes from plant photosynthesis and microbial metabolism. Green plants grown through photosynthesis are the main source of cellulose in nature, and the annual regeneration amount reaches billions of tons, which is the most usable renewable resource in nature [7]. The main component of paper is cellulose, and a small amount of hemicellulose and lignin, the higher the degree of polymerization of cellulose, the better the mechanical properties of paper. As a natural polymer material, cellulose has good compatibility with paper, and its molecules are linear polymers, which have enough adhesion strength to fibers and can "bridge" between fibers [8, 9]. Carboxymethyl cellulose (CMC), as a derivative of cellulose, retains the advantages of cellulose, such as low price, abundant material, non-toxic, biodegradable, and has the similar chemical structure to paper fiber [10]. The abundant hydroxyl groups on the chain can form hydrogen bonds with the hydroxyl groups on the paper fiber, and can also be endowed with excellent characteristics through chemical modification, to obtain paper heritage protection materials with excellent performance. Graft copolymerization is one of the most commonly used methods. The grafted polymers prepared by this method not only retain the excellent properties of CMC itself, but also give it new functions [11]. The acrylic monomer is often used as graft copolymer, and the synthesized material has the advantages of good film formation, low toxicity, water resistance, weather resistance and thermal stability. Suitable polyacrylate material coated on the paper, not only can make the paper fiber thickening, fiber binding force increase, and enhance the strength of the paper, but also can play a toughening, anti-aging and other effects. At the same time, the organic-inorganic composite material OVPOSS with a three-dimensional nanostructure was added to the synthesis process. The organic substituent of OVPOSS is -CH=CH_2, which can be homopolymerized and copolymerized with other monomers, making the synthesized material possess the excellent properties of wear resistance and weather resistance of OVPOSS [12, 13].

In this study, a paper protector was synthesized by soap-free emulsion polymerization using CMC, MMA, BA, GMA, HEMA and OVPOSS as raw materials. After

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the protectant was applied to the surface of the paper, the tensile strength, folding resistance, tearing degree, gloss, whiteness, quantity and thickness of the paper were studied and analyzed. The results showed that the mechanical properties of the coated paper increased greatly, and the glossiness and whiteness of the coated paper had no obvious effect, which accorded with the principle of "repairing the old as the old, keeping the original appearance".

2. EXPERIMENTAL

2.1. Materials and instruments

Xuan paper was purchased from Jingxian county, Anhui province. Sodium carboxymethyl cellulose (CMC), methyl methacrylate (MMA), butyl acrylate (BA), glycidyl methacrylate (GMA) Potassium persulfate (KPS), sodium bicarbonate ($NaHCO_3$), sodium hydroxide (NaOH), acetone and hydroxyethyl methacrylate (HEMA) were purchased from Aladdin company. Eight vinyl polyhedral oligomeric silsesquioxane (OVPOSS) was obtained from Hangzhou Tuomu Technology Co., Ltd. Aqueous ammonia ($NH_3 \cdot H_2O$) and ethanol were purchased from chemical reagents of Luoyang City.

2.2. Preparation of CMC-acrylate-OVPOSS emulsion

The experiment accurately weighed 1 g CMC in a four-mouth flask, added an appropriate amount of deionized water, raised the temperature to 70 ℃ and stirred to dissolve. After the solution was completely dissolved, the temperature was raised to 80 ℃, and the air was drained with nitrogen for 30 min. Half of the KPS aqueous solution was added, which was in contact with CMC solution for 10 min, and then the mixed monomers (MMA, BA, GMA, HEMA) accounting for 1/4 of the total monomer mass were added by drops. When blue light appeared in the solution, the remaining monomer and OVPOSS dispersed in the monomer were dripping, and the remaining initiator was dripping at the same time. The dripping acceleration of the monomer was controlled to be 6 – 7 seconds and a drop was added. The finishing time of the monomer dripping was slightly earlier than that of the initiator aqueous solution. After the reaction was over, the emulsion was cooled to room temperature, and then the pH was adjusted to slightly alkaline with 20 % ammonia water, and the emulsion was discharged for use. The synthesis of CMC-acrylate-OVPOSS is shown in Fig. 1.

Purification of the product: the emulsion was poured into the ethanol solution and stirred while inverted to demulsify and precipitate. After centrifugal separation, the emulsion was washed with anhydrous ethanol and centrifuged several times. The precipitate was put into a vacuum drying oven and dried to constant weight at 60 ℃ to obtain the coarse graft product. An appropriate amount of crude graft product was taken into the Soxhlet extractor and extracted with acetone as a solvent for 24 h. The homopolymer was removed to obtain the purified graft products. And, the grafting rate of the product can reach 514 %, and the grafting efficiency can reach 78.7 %.

2.3. Preparation of simulated paper samples

To evaluate the protective effect of the prepared materials on paper, Xuan paper was selected as the simulated paper sample. The paper samples were dipped in the reinforcement solution for 5-10 min, then taken out and dried at room temperature.

2.4. Analysis and testing methods

FT-IR test of the samples: The purified samples were scanned by the Nicolet IS5 Fourier Transform Infrared spectrometer.

XRD test of the samples: The purified samples were determined by D8 Advance X-ray diffractometer.

Thermal analysis test of samples: The samples were determined by STA 2500 Regulus synchronous thermal analyzer.

XPS test of samples: The samples were determined by Thermo Fisher Scientific K-alpha X-ray photoelectron spectroscopy.

The tensile strength of paper: According to the Chinese standard GB/T 12914-2008, the tensile strength of paper samples (15 mm in width and 150 mm in length) was tested by using a PN-TT300 computerized tensile testing machine.

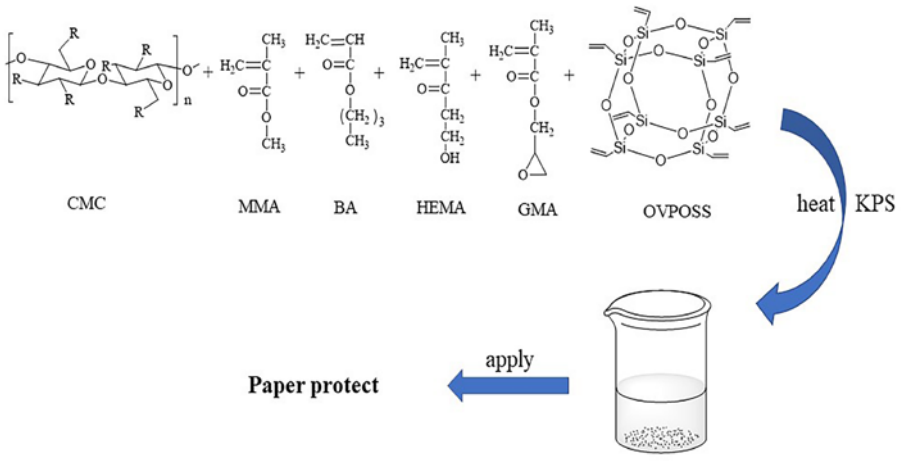


Fig. 1. Synthesis and application of CMC-acrylate-OVPOSS graft copolymer

The folding endurance of paper: According to the Chinese standard GB/T 457-2008, the folding endurance of paper samples (15 mm in width and 140 mm in length) was tested by PN-NZ135 double fold instrument.

The tearing strength of paper: According to the Chinese standard GB/T 455-2002, the tearing strength of the paper samples (63 mm in width and 75 mm in length) was tested by using the computerized PN-TT1000 paper tearing instrument. The gloss of paper: according to the Chinese standard GB/T 8941-2013, the gloss of the paper samples (100 mm in width and 100 mm in length) was tested with the PN-GM gloss instrument. The brightness of paper: according to the Chinese standard GB/T 7974-2002, the brightness of the paper samples (100 mm in width and 100 mm in length) was tested using the PN-48B whiteness instrument.

3. RESULTS AND DISCUSSION

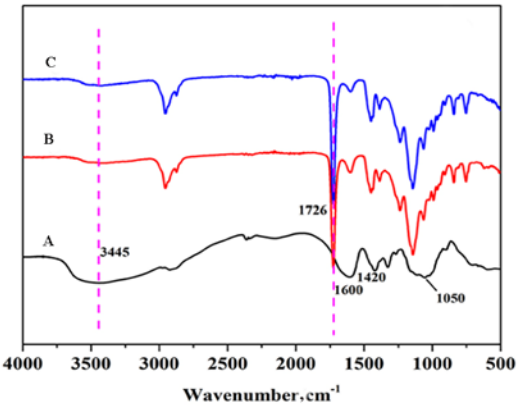
3.1. Infrared characterization of samples

To observe and compare the chemical structure of the samples, CMC, CMC-acrylate and CMC-acrylate-OVPOSS were analyzed by FT-IR spectroscopy and the results were shown in Fig. 2.

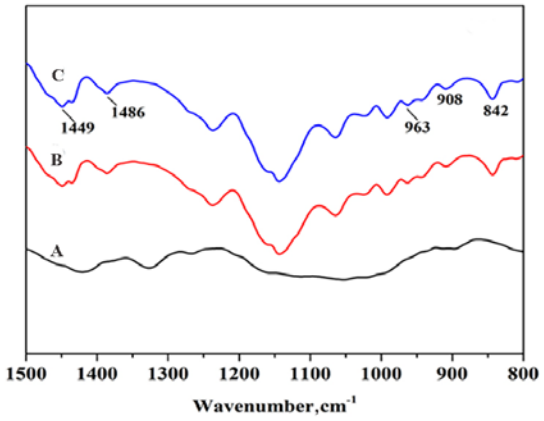
It can be seen from Fig. 2 that curve a was the infrared spectrum of CMC, and the broad peak near 3445 cm^{-1} was the stretching vibration absorption peak of -OH. At 1420 cm^{-1} and 1600 cm^{-1}, there were symmetric and asymmetric stretching vibration absorption peaks of the -COO group, respectively [14]. The stretching vibration absorption peak of C-O-C in the CMC chain was around 1050 cm^{-1} [15]. As can be seen from Fig. 2 b and c, compared with the FT-IR of CMC, the spectra of CMC-acrylate and CMC-acrylate-OVPOSS had all the characteristic absorption peaks of CMC, the -OH absorption peak near 3445 cm^{-1} was significantly weakened, and the absorption peak of C=O in acrylate monomer appeared at 1726 cm^{-1}. In addition, in the partially amplified spectrum, 2955 cm^{-1} and 1386 cm^{-1} were the absorption peaks of the stretching vibration and symmetrical deformation vibration of the methyl group in the acrylate, respectively. At 2874 cm^{-1} and 1449 cm^{-1} were the absorption peaks of stretching vibration and bending vibration of the methylene group respectively [16]. The stretching vibration absorption peak of C-O in acrylate was at 1143 cm^{-1}; 908 cm^{-1} was the characteristic peak of the epoxy group, indicating the successful introduction of the GMA monomer. At the same time, no obvious C=C absorption peak was found in the range of 1670 – 1620 cm^{-1}, which indicated that all acrylate monomers participated in the grafting reaction. Due to the small amount of OVPOSS addition, the infrared characteristic peak was weak, and another characterization of OVPOSS was made.

3.2. XRD characterization of samples

Fig.3 showed the XRD spectrum of CMC, CMC-acrylate and CMC-acrylate-OVPOSS. Fig. 3 showed that CMC at 2θ = 20°had an obvious diffraction peak, it was a chain of polysaccharides in a semicrystalline state of characteristic absorption peak [17]. CMC-acrylate and CMC-acrylate-OVPOSS in 2θ = 20° also had the peak, and their peak were almost the same, and the intensities of both peaks were weaker than CMC. The reason is that the grafted acrylate polymer on the CMC chain enlarged the proportion of the amorphous region, destroyed the original space structure of CMC, and the regularity and symmetry of the molecular chain of the introduced polymer were poor, which led to the decrease of the crystallinity of CMC, which further indicated that the occurrence of graft copolymerization.



a



b

Fig. 2. a – FT-IR spectra; b – amplified spectra of three samples: A – CMC; B – CMC-acrylate; C – CMC-acrylate-OVPOSS

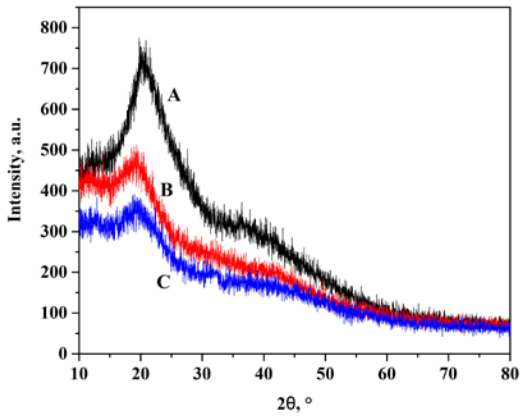


Fig. 3. X-ray diffraction patterns of three samples: A – CMC; B – CMC-acrylate; C – CMC-acrylate-OVPOSS

3.3. Thermal analysis of samples

To study the change of thermal properties of CMC grafted with acrylate and OVPOSS, the thermal gravimetric analysis of CMC, CMC-acrylate and CMC-acrylate-OVPOSS were carried out. As shown in Fig. 4, they were the TG curve and the DTG curve respectively. It can be seen from the figure that the thermal decomposition of the sample CMC was mainly divided into three stages, the first stage was from 23 ℃ to 194 ℃, and the thermal weight loss reached 8.6 %, which was mainly due to the evaporation of water adsorbed on the CMC surface. The second stage started at 194 ℃ and ended at 320 ℃. The weight loss of the sample was relatively rapid, and the thermal weight loss reached 37.8 %. This may be due to the oxidation of C=O and C–H in the main chain of CMC. Combined with DTG, CMC had two peaks at 67.5 ℃ and 284.6 ℃, corresponding to the maximum decomposition rate of CMC in these two stages. The third stage was from 320 ℃ to 500 ℃. The weight loss of CMC was slow, the thermal weight loss was about 9.8 %, and the carbon residue was 43.8 %, which was mainly due to the aromatization of carbon residue in CMC [18].

According to the TG and DTG curves of CMC-acrylate, the decomposition stage was mainly divided into four stages, and the thermal weight loss rate of the sample was 2 %, 10.8 %, 70.4 % and 4 %, respectively. The first stage was from 22 ℃ to 190 ℃, which was mainly the loss of adsorbed water in the grafted polymer. The second stage started from about 190 ℃ to 300 ℃, and the sample weight loss was relatively rapid, mainly due to the decomposition of the CMC chain in the graft of CMC-acrylate. The maximum decomposition temperature was 278.3 ℃, which was consistent with the thermal decomposition temperature range of CMC. In the third stage, from about 300 ℃ to 420 ℃, the graft copolymer lost weight rapidly, which was probably due to the decomposition of part of the CMC chain and acrylate branch chain in the copolymer, and the maximum decomposition temperature was 368.2 ℃. The fourth stage was 420 – 500 ℃, and the weight loss was relatively slow. It was mainly the pyrolysis of some graft copolymers that was difficult to decompose. Meanwhile, the carbon residue reached 12.8 %, which was greatly reduced compared with CMC, mainly because a large number of acrylate monomers were grafted onto the CMC chain.

Compared with the thermogravimetric profiles of CMC-acrylate, the thermogravimetric analysis curves of CMC-acrylate-OVPOSS showed similar decomposition temperatures, which were also mainly divided into four stages. The first stage started from 15 ℃ to 190 ℃, and the thermogravimetric loss was about 2.1 %. The second stage was 190 – 298 ℃, and the thermogravimetric loss was about 10.3 %, which may be due to the decomposition of the CMC chain in the graft copolymer. The third stage was 298 ℃ to 425 ℃, and the thermogravimetric loss was about 65.9 %. The maximum decomposition temperature was 370.8 ℃, which was 2.6 ℃ higher than that of CMC-acrylate copolymer. The fourth stage was 425 – 500 ℃, the weight loss of the sample was relatively slow, and the thermal weight loss was about 3 %.

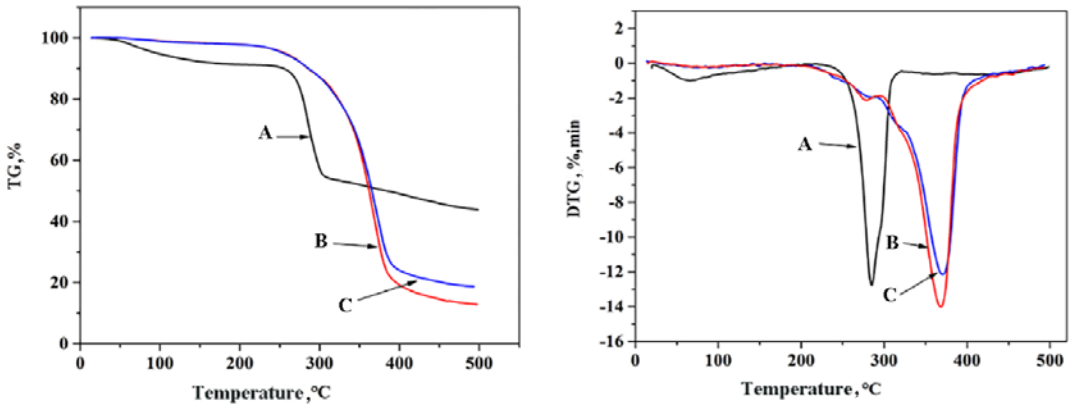


Fig. 4. TG and DTG curves of three samples: A – CMC; B – CMC-acrylate; C – CMC-acrylate-OVPOSS

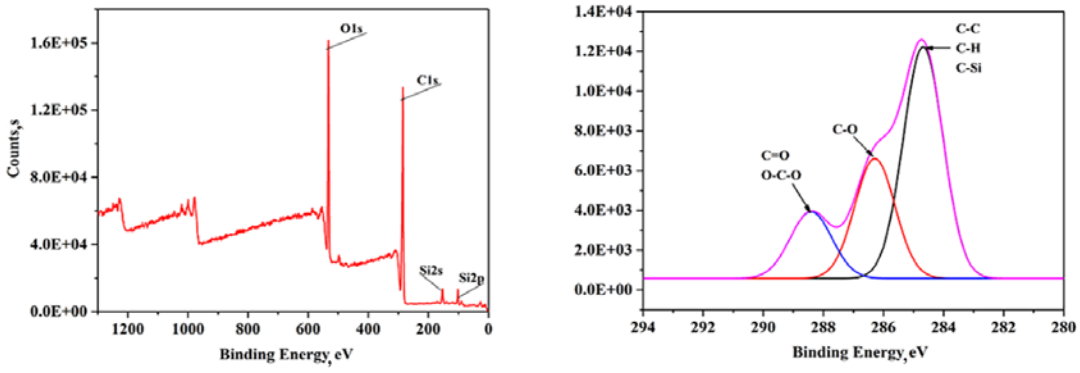


Fig. 5. XPS spectrum of CMC-acrylate-OVPOSS and high-resolution spectrum of C 1s

The carbon residue reaches 18.7 %, which was 5.9 % higher than the 12.8 % of CMC-acrylate. It showed that OVPOSS reacted with CMC-acrylate, thus improving the thermal stability and ablation resistance of the material.

3.4. XPS characterization of samples

To obtain the surface element composition and chemical bond state of the CMC-acrylate-OVPOSS graft copolymer, XPS analysis was performed on it, as shown in Fig. 5.

As can be seen from Fig. 5, the full spectrum of photoelectron spectroscopy of the polymer clearly showed the silicon spectrum in the polymer. In the C 1s spectrum, through curve fitting to get three points peaks, showed that the polymer had three states in the C: electron binding energy of 284.68 eV was C or H atoms or Si atoms connected C atom (–C–C, –C–H, –C–Si). The electron binding energy 286.28 eV was the C atom connected with only one non-carbonyl O atom (–C–O), and the electron binding energy 288.41 eV was the C atom connected with one carbonyl O atom or two non-carbonyl O atoms (C=O, –O–C–O–) [19]. XPS spectra showed that the acrylate monomer and OVPOSS were successfully grafted onto the CMC chain.

3.5. Changes in properties of paper samples before and after treatment

The changes in the machine direction (MD) and cross direction (CD) mechanical strength of the paper samples before and after treatment with 10 % reinforcement solution were respectively shown in Table 1 and Table 2.

It can be seen from Table 1 and Table 2 that the tensile strength, elongation, folding strength and tearing strength of the reinforced paper in both vertical and horizontal directions were greatly improved compared with that of the blank paper after the reinforcement solution treatment. The tensile strength of the paper increased by 111.6 % and 36.0 % in the vertical and horizontal directions, respectively. Paper or cardboard by tension to break the length of the extension and the original length of the ratio known as the paper elongation, is an important index to measure the toughness of the paper. Compared with blank paper, the elongation of reinforced paper was increased by 167.8 % and 155.1 % respectively. The folding strength of paper was increased by 736.6 % and 277.5 % respectively, and the folding resistance of paper was improved significantly. Paper tearing strength refers to the force required to tear paper to a certain length, is an important indicator of the mechanical properties of paper. The study showed that the tearing strength of paper was related to the length of the paper fibers and the binding force between the fibers. Compared with blank paper, the tearing strength of reinforced paper was increased by 76.2 % and 69.4 % respectively. The mechanical properties of reinforced paper were better than that of blank paper, this may be because the reinforcement solution was filled in the paper fiber, the paper surface formed more and more dense film, and the bonding force between the paper fibers increased. At the same time, because the CMC in the reinforcement solution could play a certain bridging role, deepening the connection between the paper fibers, there may be a small amount of grafted epoxy ring may also have a ring opening reaction with the hydroxyl group on the paper to play a connecting role, so that the paper's ability to resist external force impact was enhanced, and the paper's mechanical strength was increased. The changes in glossiness, whiteness, thickness and quantity of paper before and after the reinforcement solution treatment were shown in Table 3. It can be seen from Table 3 that the glossiness and brightness of the coated paper decreased by 2.0 % and 3.0 % respectively compared with the blank paper sample, while the thickness and weight of the paper increased by 2.2 % and 4.9 % respectively. These changes in paper properties were not obvious and could hardly be recognized by the naked eye, which conformed to the principle of "repair as old as old" for paper protection. Compared with other polymer materials (such as cellulose and cellulose derivatives, polyacrylic acid, silicon fluoride resin), carboxymethyl cellulose-acrylate-OVPOSS graft copolymer protective material is safe, has good permeability, has certain strength, has good compatibility with the simulated paper relics, and does not have the disadvantages of uneven coating of protective material, making paper thicker, affecting handwriting and so on.

Table 1. The tensile strength, elongation, folding endurance and tearing strength in the machine direction (MD) of paper samples and its standard deviation (*S*)

Paper sample	Tensile strength, $kN \cdot m^{-1}$		Elongation, %		Folding endurance, times		Tearing strength, mN	
	MD	*S*	MD	*S*	MD	*S*	MD	*S*
Untreated paper	0.956	0.075	1.15	0.063	43.4	4.1	251.5	16.1
Treated paper	2.023	0.108	3.08	0.11	363.1	25.3	443.2	21.5

Table 2. The tensile strength, elongation, folding endurance and tearing strength in the cross direction (MD) of paper samples and its standard deviation (*S*)

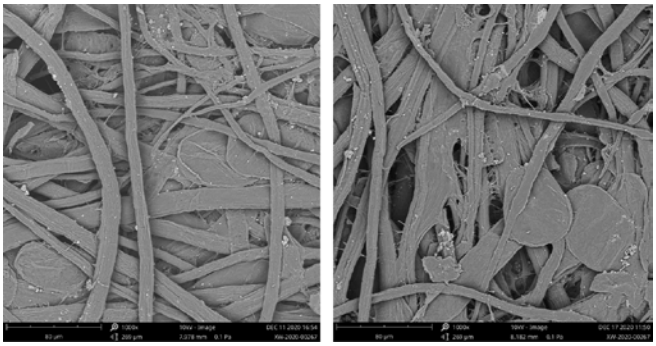
Paper sample	Tensile strength, $kN \cdot m^{-1}$		Elongation, %		Folding endurance, times		Tearing strength, mN	
	CD	*S*	CD	*S*	CD	*S*	CD	*S*
Untreated paper	0.439	0.043	1.27	0.058	4	0.81	296.1	19.5
Treated paper	0.597	0.054	3.24	0.14	15.1	1.6	501.7	26.2

Table 3. The glossiness, brightness, thickness and weight of paper samples and their standard deviation (S)

Paper sample	Glossiness, Gu	*S*, Gu	Brightness, %	*S*, %	Thickness, μm	*S*, μm	Weight, g/m^2	*S*, g/m^2
Untreated paper	4.9	0.063	83.0	0.14	48.7	0.087	32.06	0.084
Treated paper	4.8	0.063	80.5	0.13	50.8	0.095	34.52	0.077

3.6. SEM test of paper samples

The SEM spectrum of the paper sample before and after treatment with a protective solution is shown in Fig. 6. It can be clearly seen from Fig. 6 that, compared with the blank paper, the gap between fibers of the treated paper has been filled to a certain extent, and the connections between fibers have increased. This is the main reason for the increase in paper adhesion and paper strength.

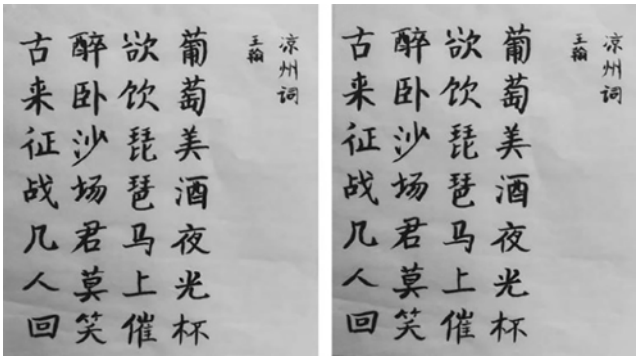


a b

Fig. 6. SEM image of the paper pattern before and after coating: a – before coating; b – after coating

3.7. Handwriting and ink test of paper samples

The handwriting and ink experiment of paper is shown in Fig. 7.



a b

Fig. 7. Appearance changes of the pattern before and after coating: a – before coating; b – after coating

It can be seen from the figure that the reinforcement material is colorless and transparent, does not cover the handwriting, and does not spread the ink. The appearance of the paper does not change in color, hue or luster, and keeps the original appearance, which is in line with the principle of "repair as old as before".

4. CONCLUSIONS

In this study, a novel CMC-acrylate-OVPOSS graft copolymer emulsion for the protection of paper cultural relics with CMC as the substrate and MMA, BA, GMA, HEMA, and OVPOSS as the graft was synthesized. The protection material has a certain strength, good thermal stability and good compatibility with the simulated paper relics. When the paper sample was coated with 10 % reinforcement solution, the mechanical strengths of the paper in the machine direction and cross direction were significantly stronger than that of the blank paper, and the optical properties of the paper were not changed significantly. The handwriting and ink test of the paper pattern showed that the reinforcement liquid hardly affected the appearance of the paper, which was conform to the principle of "repairing the old as the old, keeping the original appearance". Furthermore, CMC-acrylate-OVPOSS had a positive effect on the protection of paper cultural relics.

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