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A NOVEL GLUE REMOVER FOR PRESSURE SENSITIVE TAPES ON AGED PAPER

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ABSTRACT

Pressure-sensitive adhesives consisting of polymer (*t*-butyl acrylate, PtBA) was widely used to repair aging paper. Over time, whiteness, mechanical properties and durability of paper degrade with the aging of adhesive. In this study, novel paper-friendly glue-remover (GR), consisting of organic solvents, was designed for removal of PtBA from aging paper. Dichloromethane and hexane were screened to prepare GR according the swelling degree of aged adhesive. For detail, the component of GR was adjusted to optimize removal effect and minimize the damage of GR on aged paper. Treated with GR consisting of 40% (v/v) dichloromethane/ normal hexane, the removal rate of PtBA was high to 56%. Results of mechanical and chromatism test indicates the GR will not damage the mechanical strength or handwriting on the paper. Moreover, the pH of paper increased from 3.55 to 5.75 after removal of glue, which suggested that GR help remove acid from aging paper.

KEYWORDS: Glue remover, aging adhesives, aging paper.

INTRODUCTION

Archived materials, such as books, manuscripts, prints and paintings of the mid-19th to 20th centuries, were widely repaired by pressure-sensitive adhesives (Carter 1996), (Henniges et al. 2006). Over time, these adhesives turn yellow and become brittle, which make historical documents readily to be damaged. There is a risk of tearing aging paper, if directly peeling off the pasted pressure-sensitive adhesives from the surface of the paper. The mainly components of pressure-sensitive adhesive is polymer, such as natural rubber, synthetic rubber and acrylic polymer (Gorassini et al. 2016). To remove the pressure-sensitive adhesives, some researchers have investigated different kinds of mechanical, physical, biological and chemical methods. Liang et al. have done research on removing adhesives in mixed office waste paper according to the agglomeration-magnetic principle (Hu et al. 2004). Chen et al. introduced a flotation column method applied in OCC paperboard workshop, which achieved a remarkable performance with adhesive removal, which showed a remarkable performance with adhesive removal (Jia-xiang 2008). Additionally, some kinds of enzyme additives were investigated to decompose the grease in adhesives, improving the operating efficiency of removal equipment. (Martin 1978, Schwartz 1994). However, all these methods would destroy the fiber and handwriting of aging paper, and the solution used in some methods above contains water in, which may result in removal or damage to handwriting (Ruixiang 2012).

Although some research progress has been made in removal of adhesives from aged paper by traditional methods, but the limitations still remain, such as low removal efficiency, damaging of fibers and writing on aged paper during the removal process, etc. Research on removing pressure sensitive adhesive with a negligible effect on the properties of the paper is still in its infancy. Therefore, methods for removing adhesives from aged paper still require further improvement to protect old documents. Complementary aims include:

Investigating a water-free glue-remover to protect aged paper from deterioration caused by aging of the adhesive.

Screening low toxicity organic solvents, which are harmless to writing, mechanical strength and pH of aged paper but can aid in removing the aged adhesive.

Adjusting the composition of the glue-remover and the removal condition to achieve the best removal effect.

MATERIAL AND METHODS

Materials

The following notations were conventionally used to denote the removal of glue treatments and paper samples: China Daily News newspaper (CDN, 4/30/1981-5/25/1981), China Daily News newspaper is donated to by Yu Ping ancient book restoration center (http://www.ypbz.com.cn/gjxf/index.html) which were preserved at 26°C with a relative moisture content of 50%.

Chinese art paper (rice paper) - Polymer t-butyl acrylate (PtBA, Dongguan yikelon trading co. LTD), Biaxially-oriented Polypropylene (BOPP, Dongguan yikelon trading co. LTD.), Direct black (BN, Shanghai McLean biochemical technology co. LTD., Methyl blue (MB, Shanghai McLean biochemical technology co. LTD., Azophloxine (AP, Shanghai McLean biochemical technology co. LTD.) , Eosin Y (Y, Shanghai McLean biochemical technology co. LTD, Metanil yellow (MY, Shanghai McLean biochemical technology co. LTD.). Acid violet (CB, Shanghai McLean biochemical technology co. LTD.) . Seven organic solvents were bought from Tianjin Daomao Chemical Reagent co. LTD.

Sample parameters

Tab. 1 shows the parameters of CDN specimens used in the PtBA adhesive artificial aging experiment and the other types of paper used in the experiment. CDN were used to prepare the naturally aged paper samples in this study.

Tab. 1. The parameters of the CDN specimens.

Parameters	CDN	Measure standard
Grammage ($g \cdot m^{-2}$)	45-50	ISO 536:1995
pH of the paper surface	3.35-3.56	GB/T 13528–9
Alkali reserve ($mol \cdot kg^{-1}$)	-	ISO 10716:199
Tensile strength ($kN \cdot m^{-1}$)	1.72-1.82	ISO 1924-2:2008
Stretch (%)	0.81-0.93	ISO 1924-2:2008
Folding endurance (times)	4-6	ISO 5626:1993
Tear (mN) 1974:1990	100.2-120.2	ISO 1974:1990
Colorimetric measurements		
L	76.80-77.80	ISO 11476
a	2.87-3.87	
b	8.90-9.90	

Paper specimen dyeing

Inks diluted with DI water in 1:1 volume ratio to prepare dyeing solution and dyed Chinese art paper and printing paper as specimens to do chromatism test. Immersed both rice paper and printing paper in the dyeing solutions for 1 min, then taken out and dried at 50% RH and 23°C (ISO 187-1990).

Artificial aging of paper and adhesive

Artificial aging of paper specimens was carried out to make sure that the specimens were in a similar condition as naturally aging papers (Area and Cheradame 2011). The specimens were aged at 105°C for 72 h, which theoretically equates to 25 years natural aging (ISO 5630-1:1991, (Porck 2000). Adhesive was aged in an artificial aging process according to the processing of paper. PtBA adhesive specimens with a size of 6 x 6 cm and thickness of 100 µm coated on printing paper were aged at 105°C for different time to find the optimal time that can simulate the natural aging state of pressure sensitive adhesive.

The swelling of PtBA adhesive test

Test the swelling degree of PtBA adhesive according to the standard GB/T1690-2006 (Wang et al. 2015). After the artificial aging treatment, 0.1 g PtBA adhesive specimens were immersed in different organic solvents in bottles for 60 min to swell fully, then the samples were removed from the bottles and any surface liquid was totally dry. The weights of PtBA adhesive before and after swelling were marked as m_1 and m_2, respectively. Due to the toxicity of organic solvents, this operation needs to be carried out in the fume hood.

MATERIAL AND METHODS

Determination of chromatism

The chromatism of the paper specimens before and after immersing in the organic solvents were tested with a Color reader (CR-10, from KONICA MINOLTA SENSING, INC, JAPAN). 8 specimens of each color paper were tested at the same conditions and the average value was calculated. According to the standard chromatism formula recommended by the international lighting association (CIE), the total color change ΔE was used to indicate the chromatism of paper specimens based on the (L, a, b) * values before and after treatment for a paper specimens (Jiang, et al., 2016). ΔE was calculated as Eq. 1:

$$\Delta E = [((\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*) 2) /3]^{0.5} \quad (1)$$

The corresponding standard for chromatism and color change is showed in Tab. 2.

Tab. 2: The standard for chromatism and color change.

Grade	Discoloration	Value of chromatism
0	No color change	<=1.5
1	Very slight color change	1.6~3.0
2	Slight color change	3.1~6.0
3	Obvious color change	6.1~9.0
4	Large color change	9.1~12.0
5	Badly color change	>12.0

Determination of mechanical strength of paper

According to the standarts GB/T 12914-2008, GB/T 457-2008, GB/T 455-2002, the mechanical properties of the paper were evaluated mainly by tensile strength, bending resistance and tearing strength. Parameters for each of the 8 specimens were measured to calculate the average value to indicate the properties of mechanical strength.

Determination of glue removal efficiency

Paper specimens were cut along the coating with PtBA adhesive area and weighed after artificial aging as described in chapter "Artificial aging of paper and adhesive", then the aged paper specimens coated with PtBA adhesive were immersed in GR for 60 min. Three parallel experiments were performed for each GR with different proportions. The total mass of adhesives and paper before immersing in GR was marked as M_1. After immersed in GR and scraped the softened PtBA adhesive, the total mass of the rest adhesive and paper was marked as M_2, the total mass of empty paper was marked as M_3. The glue removal efficiency was introduced to evaluate the rate of glue removal as shown in Eq. 2.

$$S = \frac{M_1 - M_2}{M_1 - M_3} \quad (\%) \quad (2)$$

where: S - glue removal efficiency (%),
M_2-M_1 - total removal of glue,
M_1-M_3 - total amount of adhesive before removal.

Due to the toxicity of organic solvents, this operation needs to be carried out in the fume hood.

Determination of pH

The surface pH of specimens was tested by a pH meter with flat electrode (China, DJS-0.1C/(F)). Since the mass of acid at different point of paper specimens was different, the pH of each specimen was measured for 8 times on different point and its average value was calculated.

Characterization

Scanning electron microscop SEM, Japanese Hitachi s-3400) was used to observe the changes of fiber morphology.

RESULTS AND DISCUSSION

Artificial aging of PtBA adhesive

Since the adhesive pasted on aging paper were aged, the artificial aging of adhesive was carried out to make sure that the state of adhesives specimens was close to that of naturally aging adhesives. There is no standard for the artificial aging of adhesives on paper, so it was investigated according to the Chinese GB/T 464-2008, which is for the artificial aging conditions of the paper (Jin et al., 2018). Changes of specimens, including color and properties were observed during artificial aging. As shown in Fig. 1a), b) and c), the glue coated on CDN was not aged completely when the artificial aging time was less than 72 h. The color of CDN turned to yellow and the CDN became brittle and fragile after aging 96 h (Fig. 1e). Glue films were separated from the cellulose fibers within paper, and the surfaces of glue bubbled severely.

Fig. 1: The aging of PtBA adhesive coated on CDN.(a) Unaged; (b)Aged for 24 h; (c) Aged for 48 h; (d) Aged for 72 h; (e) Aged for 96 h.

These artificial aging conditions were worse than 25 years of natural aging and different from the condition of naturally aged CDN coated adhesive. Fig. 1c) indicates that the condition of CDN after accelerated dry aging at 72 h was very close to the condition of CDN aged 25 years naturally. Since GR is investigated for aging adhesive, 72 h and 105°C were selected as the artificial aging conditions for the PtBA adhesive.

Swelling degree of PtBA adhesive in different solvents

The polymer chain of PtBA was cross-linked after artificial aging process described in chapter "Artificial aging of paper and adhesive" which prevented the PtBA from dissolving in regular solvents. The organic solvent molecules enter the interstitial polymer clearance of the cross-linked polymer, which makes the aging adhesive soft, swollen, transparent and gelatinous. The quality change rate of adhesive before and after immersed in organic solvent for 1h indicated the effect of organic solvent on the swelling of PtBA adhesive. Several different organic solvents with close polarity were selected as listed in Fig. 2. Fig. 3 shows that both the swelling degrees of aging PtBA adhesive after immersed in normal hexane and methanol were under 2. The swelling degree of aged PtBA was more than 10 in carbon tetrachloride, toluene, ethyl acetate and acetone, and the maximum swelling degree observed was about 30 in dichloromethane. Therefore, dichloromethane is the optimal swelling agent for aging cross-linked PtBA adhesive.

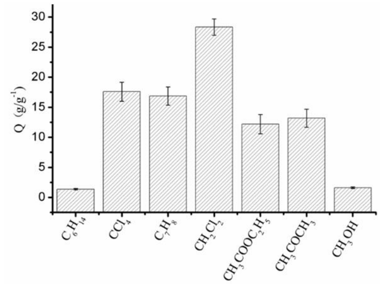

Fig. 2: Swelling degree of aged PtBA adhesive in organic solvents.

Chromatism of paper

The chromatism (ΔE) of paper specimens before and after immersion were determined to investigate the solubility of writing dyes in organic solvents. The results were shown in Tabs. 3-7.

Tab. 3: The chromatism of dyed paper specimens before and after immersed in dichloromethane.

Dyes	CB	MB	Y	BN	MY	AP
1	1.23	12.55	2.75	3.57	1.47	18.67
2	1.12	12.54	2.46	3.05	1.06	18.55
3	1.35	12.94	2.95	3.65	1.47	19.04
average value	1.24	12.68	2.72	3.42	1.33	18.75
standard deviations	0.12	0.23	0.25	0.33	0.24	0.25

Tab. 4: The chromatism of dyed paper specimens immersed in methanol.

Dyes	CB	MB	Y	BN	MY	AP
1	4.66	6.41	2.05	4.07	17.60	16.82
2	4.15	6.05	1.84	3.84	17.01	17.06
3	4.91	6.79	2.16	4.36	17.87	16.21
average value	4.58	6.42	2.01	4.09	17.49	16.70
standard deviations	0.39	0.37	0.16	0.26	0.44	0.44

Tab. 5: The chromatism of dyed paper specimens immersed in carbon tetrachloride.

Dyes	CB	MB	Y	BN	MY	AP
1	2.79	8.78	5.43	2.73	1.39	0.92
2	2.57	8.75	5.17	2.64	1.22	0.73
3	2.93	9.18	5.27	2.96	1.41	0.80
average value	2.76	8.91	5.29	2.78	1.34	0.82
standard deviations	0.19	0.24	0.13	0.17	0.11	0.09

Tab. 6: The chromatism of dyed paper specimens immersed in ethyl acetate.

Dyes	CB	MB	Y	BN	MY	AP
1	4.51	2.65	2.21	0.89	1.50	2.28
2	4.17	2.59	2.02	0.81	1.37	2.17
3	4.67	2.87	2.42	0.95	1.65	2.47
average value	4.45	2.70	2.22	0.88	1.50	2.30
standard deviations	0.26	0.14	0.20	0.07	0.14	0.15

Tab. 7: The chromatism of dyed specimens immersed in acetone.

Dyes	CB	MB	Y	BN	MY	AP
1	1.48	10.39	7.99	3.31	4.96	4.79
2	1.55	10.65	7.55	3.65	4.65	4.45
3	1.32	10.31	7.65	3.73	4.72	4.84
average value	1.45	10.45	7.73	3.57	4.78	4.70
standard deviations	0.12	0.18	0.23	0.22	0.16	0.21

The ΔE of AP-dyeing specimens before and after immersed in dichloromethane and methanol were beyond 15, which indicated that dyes and pigment molecules had strong solubility in both dichloromethane and methanol. The ΔE values of papers immersed in n-hexane were less than 2.0. The solubility of dyes in hexane was the smallest. Dichloromethane will damage handwriting on paper while n-hexane is one solvent without damage on handwriting of paper. Therefore, dichloromethane and n-hexane were mixed to prepare a paper-friendly GR.

Optimization of the composition of GR

Considering both the effects of organic solvents on swelling of adhesive and handwriting on paper, GR was prepared by mixing n-hexane and dichloromethane at different volume ratios. As shown in Fig. 3, the chromatism of paper increased with the dichloromethane content in mixed solvent.

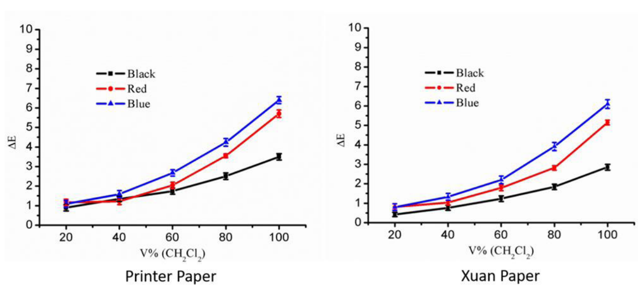

Fig. 3: Effect of GR composition on chromatism.

The results suggested that the ratio from 0% to 40% was expected to achieve the aim on protecting handwriting on aged paper. Treated with the GR consisting of less than 40% (v/v) dichloromethane, the chromatism of paper specimens is less than 3. Beyond that limit, the chromatism of paper specimens increases rapidly with the increase of content of dichloromethane.

Effect of composition of glue-remover and exposure period on removal rate

The proportion of dichloromethane and n-hexane will affect the swelling of aging adhesive, and the aging adhesive cannot be fully removed from paper after a single removal treatment, which means that repeated treatments are needed. Thus, the optimum ratio of adhesive remover and the best lengths of time for degumming were explored. The effect of GR composition on removal rate is shown in Fig. 4a, the glue removal efficiency increased with the increase of the dichloromethane proportion, which suggested that of dichloromethane, had good effect on swelling of the aged cross-linked PtBA adhesive. The removal rate of glue increases rapidly when the volume fraction of dichloromethane is in the range of 20%~40%. Since dichloromethane has a strong effect on the swelling degree of aged PtBA adhesive, the removal ration of PtBA would not increased so much until the volume content of dichloromethane was more than 40%. In addition, the chromatism of paper specimens will increase rapidly beyond that limit. Therefore, the optimal volume content of dichloromethane is 40%.

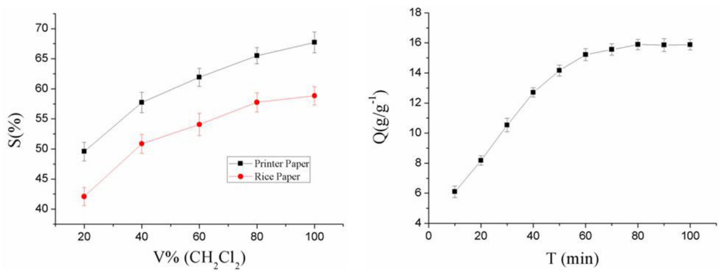

Fig. 4: Effect of GR composition and exposure period on glue removal efficiency.

Times ranging from 0 to 100 min were explored to find out the optimal time to immerse the adhesive during removal process. From Fig. 4b, it can be seen that the cross-linking structure expands over time, with a linear increase in swelling degree from 0 to 40 minutes, eventually leveling out between 80 and 100 minutes. The increase of swelling degree was relatively slow and the swelling equilibrium after more than 100 min. Compared to Fig. 5a, the yellowing degree of the adhesive in Fig. 5b was weaker, which indicated that while some surface adhesive had been removed in Fig. 5 b), there was still some adhesive remaining on the paper.

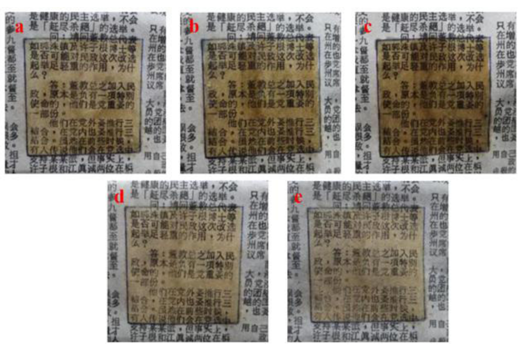

Fig. 5: The effect of moving adhesives on aging newsprint with different processing times: (a)Untreated aging newsprint; (b)Newsprint after 1 degumming treatment; (c) Newsprint after 2 degumming treatment; (d) Newsprint after 2 degumming treatment; (e)Newsprint after 4 degumming treatment.

Fig. 5c was newsprint coated with glue after twice removal processes, it can be seen that the adhesive in the middle position of newsprint was removed. Fig. 5d was newsprint coated with glue after three times removal processes.

Most of the aged adhesive had been removed. The handwriting on the newsprint was retained better, and the remaining part of the glue is tightly bonded with the cellulose fibers in the paper and is difficult to remove. Fig. 5e was newsprint coated with glue after four times removal processes. After the previous three times removal process, almost all fiber within paper and ink was exposed, and after the fourth removal process, the fiber and ink within paper was damaged, causing damage to the paper. Therefore, three times removal is enough to remove the aged adhesive efficiently without damaging the fibers within the paper.

Effect of removal agent on pH of paper surface

Moldy CDN were used to investigate the effect of GR on the pH of the paper. From Tab. 8, data shows that the pH value of moldy CDN was 4.15 on average, and the average pH values of aged paper was 3.66.

Tab. 8: The chromatism of dyed paper specimens immersed in n-hexane.

Dyes	CB	MB	Y	BN	MY	AP
1	1.15	0.71	0.23	0.94	0.45	0.79
2	1.23	0.59	0.26	0.89	0.42	0.70
3	1.33	0.64	0.25	1.05	0.47	0.74
average value	1.24	0.65	0.25	0.96	0.45	0.75
standard deviations	0.09	0.06	0.02	0.08	0.02	0.05

After the immersion, the average pH value of aged CDN was 5.38. Because the organic acid was dissolved by GR, the pH values of the paper increased. There are many factors that can cause paper acidification, one example is that some acidic substances are not soluble in organic solvents, so the acid in CDN is not fully neutralized after the treatment (Win and Okayama 2012, Zhang et al.). In Fig. 6 are SEM images of these samples.

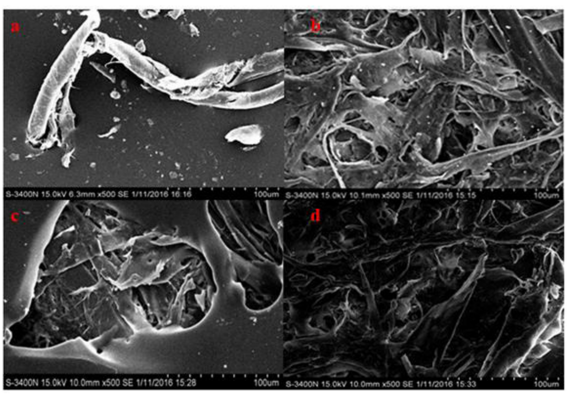

Fig. 6: SEM of CDN before and after removal: (a)Paper coated glue before removal;(b)Paper without glue before removal;(c) Paper coated glue after removal;(d)Paper without glue after removal.

In Fig. 6a, only some of the cellulose fibers were exposed on the surface of the paper, while the rest was covered, because the paper was coated well with PtBA adhesive. However, in Fig. 6b, the fibers of CDN without PtBA adhesive were clearly visible. It was evident that fibers within CDN paper were damaged and messy after natural aging and were covered with impurities. From Fig. 6c, there was a few cellulose fiber of CDN coated with adhesive could be seen. Comparison between Fig. 6b and d) showed that almost no changes occurred in the morphology of the cellulose fibers before and after immersed in GR. After immersed in GR, the fiber were still broken and messy, but the impurities on the surface of the fiber were cleaned. Obviously, the GR also contributed to the removal of plaque.

Effect of removal agent on mechanical strength of paper

The mechanical strength of paper is mainly evaluated through parameters such as tensile strength, elongation, folding strength, and tearing strength (Lin et al. 2008). The mechanical strength parameters of the Rice paper and printing paper were determined before and after immersed in the GR with different volume ratios of n-hexane and dichloromethane.

Fig. 7 shows that the tensile strength of printing paper and rice papers remains mostly unchanged; the folding strength of printing paper only changes slightly, and the folding strength of rice paper also remains mostly unchanged; the change of tearing degree of printing paper fluctuates from 0.5~2.5mN.

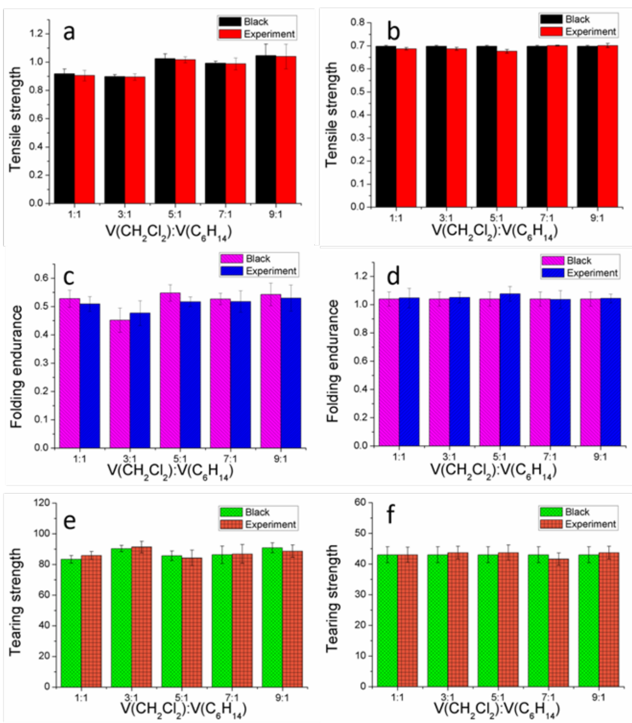

Fig. 7: Effect of glue-remover on mechanical properties of paper: (a) printer paper; (b) rice paper; (c) printer paper; (d) rice paper; (e) printer paper; (f) rice paper.

Exploration on the removal mechanism

Natural aging or artificial aging will lead to the crosslinking reaction of the PtBA adhesive polymer chains, since it was polymerized by tertiary acrylate as a monomer. Regular solvents are unable to dissolve the PtBA adhesive easily when it becomes hard, crisp and yellow after the crosslinking reaction, but it will be separated with good permeation of an organic solvent. Cross-linking polymers will expand in organic solvents spontaneously, which is also known as "swelling". Limited swelling refers to cross-linking polymer swelling in solvent which can reach the equilibrium state and remain in two-phase state when cross-linking polymer contact solvent and no longer inhalation solvent after a certain time (Youcef et al. 2017). As shown in Fig. 8, organic solvent molecules enter the crosslinking polymer gap between the polymer and the paper and expand the gap through infiltration. Seven kinds of organic solvents of different polarity were selected as degumming agents to swell the adhesive and test the swelling degree of the aged adhesive after immersing in solvent. The results were shown in Tab. 9.

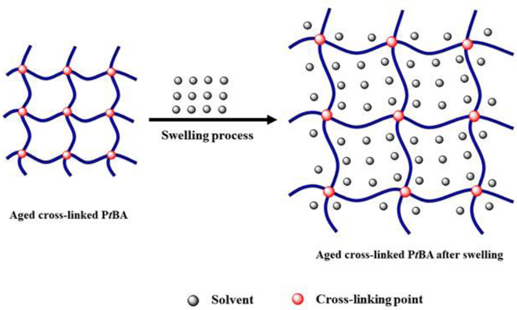

Fig. 8: The model of swelling of PtAB.

Tab. 9: The pH value of molded CDN before and after immersed in GR.

Sample	pH before aging	pH after aging	pH after soaking
1	4.12	3.39	5.55
2	4.25	3.68	5.49
3	4.09	3.65	5.33
4	4.11	3.85	5.32
5	4.21	3.73	5.22
average value	4.15	3.66	5.38

We can see from Tab. 10, dichloromethane as an organic polar solvent has a good swelling and softening effect on the aged cross-linked PtBA adhesive.

Tab. 10: The swelling degree of PtBA in organic solvents.

Solvent	polarity	Viscosity (Pa•s)	Boiling point (^{0}C)	Absorption wavelength (nm)	Swelling degree
n-hexane	0.06	0.33	69	210	1.3
tetrachloromethane	1.6	0.97	77	265	17.6
methylbenzene	2.4	0.59	111	285	16.9
dichloromethane	3.4	0.44	240	245	28.3
ethyl acetate	4.3	0.45	77	260	12.2

acetone	5.4	0.32	57	330	13.2
methanol	6.6	0.6	65	210	1.6

The swelling degree increased with the solvent polarity up until the polarity was as large as that of dichloromethane, following the principle of similar phase solution. Comparing the swelling degrees of aging PtBA adhesive immersed in tetrachloromethane and methylbenzene, it was found that they did not comply with the principles mentioned the above. This indicated that solvent molecules need to penetrate the aged cross-linked polymer, so that the organic molecular could reduce the bonding force between the polymer molecules to swell it. The size of tetrach-loromethane molecular is smaller than that of methylbenzene, so the tetrach-loromethane molecule could more easily enter gaps of the aged PtBA adhesive. The swelling result of the aged PtBA adhesive immersed in tetrach-loromethane and methylbenzene also can supported this conclusion. This also explains why the degrees of swelling of the aged PtBA immersed in ethyl acetate and acetone.

When dichloromethane molecules swell the aged PtBA adhesive, small molecules enter into the gaps of the PtBA adhesive to reduce the bonding force between the aged cross-linked polymer molecules. The aged PtBA adhesive becomes soft, expanded, and transparent after immersion in dichloromethane; this made the adhesive to be easily stripped from the fiber within paper.

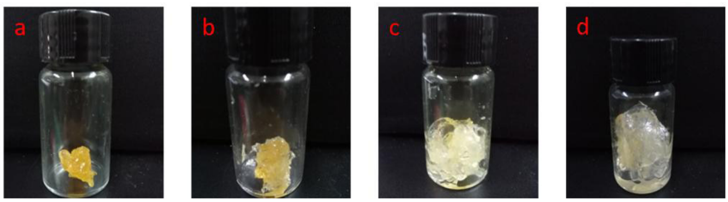

Fig. 9: Aged adhesive swelled for different times: (a) Without swelling; (b) Swelling for 20 min; (c) Swelling for 40 min; (d) Swelling for 60 min.

It could be seen from Fig. 9 that the solvent molecules enter the grid of the polymer molecules to expand the cross-linking structures, and the swelling degree increased up until 40 min. During 40~60 min, an elastic contraction force could be produced by the polymer due to the stretch of the net chain structure, slowing the entrance of solvent molecules into the polymer after immersion for 40 min, so the degree of swelling increased relatively slow. The balance between the tension and contraction forces could be achieved at 60 min, resulting in swelling equilibrium. Therefore, the optimal time for swelling of aged PtBA adhesive was 60 min.

The GR molecules could enter into the network of the aged cross-linked polymer to swell the aged PtBA adhesive and effectively soften it. When the volume content of dichloromethane is less than 60%, the glue removal rate increases with dichloromethane content. Additionally, the solubility of ink in the glue-remover is less than 3 when the volume content of dichloromethane is less than 40%. In order to achieve the purpose of "Repair it without changing its original appearance" and protect handwriting on paper, the V percentage (dichloromethane) should be controlled at 40%. It is the maximum limit of dichloromethane, below which the handwriting can keep as its original appearance. At this optimum proportion, GR is an efficient remover of the cross-linked adhesive on aged paper with a removal rate of 56%. The pH value of moldy CDN also increased due to the removal of mildew and impurities on CDN. This method is easy to operate, effective and environmentally friendly, and has great prospects and scope of application.

One avenue for future research is to investigate different types of pigments used in painting, calligraphy, or other techniques found in aged documents, as these pigments may have different solubility conditions and may require different solvents or compositions in order to produce an effective GR. The research on the mechanism of this glue-remover is still not complete, and more research needs to be done to fully understand this system.

CONCLUSIONS

After 72 h aging, the artificial aging state of pressure sensitive adhesive coated on the paper is closest to state of the pressure sensitive adhesive aged in the natural environment. When the V% (CH_2Cl_2) is 40%, the effect of removing glue agent on handwriting of papers is the least. At this ration of CH_2Cl_2 and normal hexane, the removal efficiency can reach 56%, so this ratio is the optimum proportion of the glue agent. The glue removal agents have good removal efficiency on commercially available adhesive or coated adhesive. At the same time, pH value of newsprint increased. In a word, this formula of glue removal agents has great application prospect for paper document protection.

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