Synthesis and Properties of Collagen Surface-sizing Agent Modified by Epoxy Compound

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Three modified surface-sizing agents (SA) for paper were synthesized by the cross-linking of collagen with either glycol diglycidyl ether (GDE), tetrahydrophthalate diglycidyl ester (TDE), or triglycidyl p-aminophenol (TP) and then grafting with butyl acrylate (BA). The performance and application of the sizing agent emulsions were characterized and tested. The GDE-SA had the highest viscosity. The GDE-SA coated corrugated paper had the best water resistance, smoothness, and physical and mechanical properties.

Keywords: Collagen; Sizing agent; Epoxy compound; Cross-linking agent; Corrugated paper; Water resistance

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INTRODUCTION

Paper, an important consumable product, plays an indispensable role in the daily lives of many people. China is a country characterized by large paper production. The demand for various types of paper is constantly expanding. In 2017 there were about 3,900 paper-making enterprises in China, and the national output of paper and paperboard reached 108.6 million tons, an increase of 1.35% over the previous year (Manda *et al.* 2012). With the rapid development of the paper industry, finding a favorable balance between the resources, energy, and the environment has become a key to the sustainable development of the paper industry. The development of chemical auxiliaries is no doubt a shortcut to modernize the papermaking industry, which can effectively reduce the cost of paper making and greatly improve the quality of paper (Kong *et al.* 2016). Paper sizing agents are very important processing reagents in the papermaking industry. Most paper and paperboard need to be glued to improve their physical and mechanical properties and water resistance (Di *et al.* 2018).

The traditional surface-sizing agents mainly have included animal glue, starch, alkylketene dimer, styrene acrylic polymer, and so on. However, the traditional surface-sizing agents often have the problems of high cost of raw materials, waste of grain, polluting of the environment, and so on. Furthermore, low physical and mechanical properties and poor water resistance can be obtained when a water-based formulation is being applied to the paper surface. In view of the above problems, many physical and chemical methods have been used to improve the properties of sizing agents, such as cross-linking, grafting, compounding, emulsion polymerization, and so on. Of course, the synthesis of new types of paper sizing agent is also an important pathway to solve aforementioned problems. (Yue and Fang 2015).

In 2016, China's annual output of chrome-tanned leather was approximately 63 million pieces. In the production of chrome-tanned leather, more than 80% of the tanned solid waste from the process can be categorized as hazardous waste (Sun *et al.* 2018b). If these wastes are burned or buried, a wastage of resources and environmental pollution will result (Kanagaraj *et al.* 2015). Making full use of the precious biomass resources of waste leather chips in tannery production by effective methods represents a significant opportunity to save resources. Tannery waste is rich in collagen, which has good biocompatibility, biodegradability, filling and film-forming properties. These structural and functional characteristics of collagen determine their good application prospects. At present there are fields of application of leather collagen such as preparing medicine, cosmetics, fertilizer, surface active agents, pulp and paper making, and so on (Dang *et al.* 2018).

Simple collagen has very poor mechanical properties, as it is easily degraded and absorbed in the environment. Thus, the modification of collagen—generally by chemical cross-linking—is critically important. Cross-linking agents include bifunctional materials such as diisocyanates, aldehydes, and epoxides (Zhu *et al.* 2017). Epoxy compounds are common cross-linking agents in organic synthesis (Sun *et al.* 2018a). Due to the charge polarization and ring tension in the epoxy group, the epoxy group has high reactivity and can react with $-NH_2$, phenolic hydroxyl, $-COOH$, $-SH$, $-OH$, $-C(=O)-NH_2$, *etc*., and can form cross-linked network structures to enhance the strength of the collagen molecular structure and the water resistance of collagen. In addition, the epoxy compound is less toxic compared with aldehyde cross-linking agents (Liu *et al.* 2017a). Therefore, if the collagen is extracted from waste leather and modified by epoxy compounds, new types of paper sizing agents can be prepared. This approach not only solves the environmental pollution problem, but it also opens up a new way for the development of the paper industry.

In this study, epoxides were used as cross-linking agents to explore the cross-linking effects of glycol diglycidyl ether (GDE), tetrahydrophthalate diglycidyl ester (TDE), and triglycidyl p-aminophenol (TP) on collagen. Using butyl acrylate (BA) polymer as hydrophobic chain and ammonium persulfate as initiator, the cross-linked collagen was grafted and modified to prepare a new environmentally friendly papermaking sizing agent. Three kinds of sizing agents used in the sizing of corrugated paper. The properties of the three types of sizing agents were compared with respect to the paper ring crush index, tensile strength index, and water resistance.

EXPERIMENTAL

Materials and Instruments

Collagen powder (The average molecular mass is approximately 3000 Da) was provided from Hebei Zhongpi Dongming Environmental Technology (Shijiazhuang, China). Sodium hydroxide, potassium persulfate, sodium bisulfite, and hydrochloric acid were obtained from Kermel (Tianjin, China). Alkylketene dimer (AKD), sodium chloride, and butyl acrylate were purchased from Aladdin (Shanghai, China). Glycol diglycidyl ether was obtained from Evonik Degussa AG (Shanghai, China). Tetrahydrophthalate diglycidyl ester was obtained from Aldrich (Shanghai, China). Triglycidyl p-aminophenol was purchased from Hongshun Biotechnology (Shanghai, China). The corrugated paper was received from Canfor (Shenzhen, China).

Synthesis of Sizing Agent

The GDE-SA, TDE-SA, and TP-SA were obtained by GDE, TDE, TP cross-linking modified and BA graft modified collagen. First, 3 g of industrial collagen powder was dispersed into 30 mL of water. After swelling, it was heated to 45 °C and stirred until it was completely dissolved. The collagen solution was obtained by gauze filtration. The 30 mL as-prepared collagen solution with the concentration of 0.1 g/mL was added into a three-necked flask with the capacity of 250 mL. Under stirring and refluxing, a small amount of NaCl solution was added, and the pH value was adjusted to 10 with 10% NaOH aqueous solution. Then, 6% of GDE (TDE, TP) in the amount of collagen was added dropwise as a cross-linking agent, and the reaction system was kept for 3 h at 50 °C to obtain cross-linked collagen. Subsequently, the system was heated to 80 °C with stirring, and 4.5 g of BA was pre-emulsified for 30 min. The mixture was kept for 3 h after 0.6 g ammonium persulfate was added. Finally, the product was filtered with gauze.

Characterization

The pH, solid content, and viscosity of collagen and the three sizing agent emulsions were tested by pH meter, electronic moisture meter, and viscometer, respectively. Fourier transform-infrared (FT-IR) spectroscopy was performed on a Bruker Vector-22 spectrometer (Karlsruhe, Germany) in the wavenumber range from 500 cm^{-1} to 4500 cm^{-1}. The molecular weight of the samples was tested by gel permeation chromatography (GPC). The mobile phase was 0.1 mol/L $NaNO_3$. All samples were diluted with distilled water to 0.01 wt.%. The particle size distribution of the samples was measured by Zetasizer (Nano ZS, Malvern, UK). Thermogravimetric analysis (TGA; TA Instruments, Q500, Shanghai, China) was used to analyze the thermal stability of the collagen and three sizing agents. The heating rate was 5 °C/min, with a temperature range from room temperature to 600 °C. The flow of nitrogen gas was 60 mL/min. The hydrophilicity of the coated paper was studied using a contact angle measuring device (Attension Theta, Shanghai, China). The transverse section of the paper fibers was characterized by scanning election microscopy (SEM; Hitachi Limited, S4800, Tokyo, Japan).

The thickness of paper was measured by paper thickness tester (CHY-C2A, Jina, China). The tear strength was tested using paper tearing tester (60-2600_PROTEAR, Jina, China). The tensile index was detected by tensile index tester (HH-KZ30/300/500, Hanzhou, China). The ring crush strength was measured using a computer-controlled compression tester (DC-KY3000A, Sichuan, China). And the stiffness was performed by paper and cardboard tester (YQ-Z-1, Sichuan, China). According to the requirements of test instruments, the paper was cut into corresponding size. Each sample was taken nine parallel samples, and each of them was tested three times. The average value was taken as the test result. The corrugated paper physical and mechanical properties were detected according to GB/T 6544-2008 (2008), GB/T 22364-2008 (2008), GB/T 12914-2008 (2008), and GB/T 2679.8-1995 (1995).

Paper Surface Sizing

The process of surface sizing for papers used in this work was basically the same as that used in industry. Corrugated base paper was used as the base-stock, and it was cut from pristine papers to a size of approximately 16 cm × 20 cm (Lin *et al.* 2016). The coating method was used with the horizontal roller type surface sizing using the K303 MULTI coater (Dongguan Haida Equipment Co., Ltd., Dongguan, China).

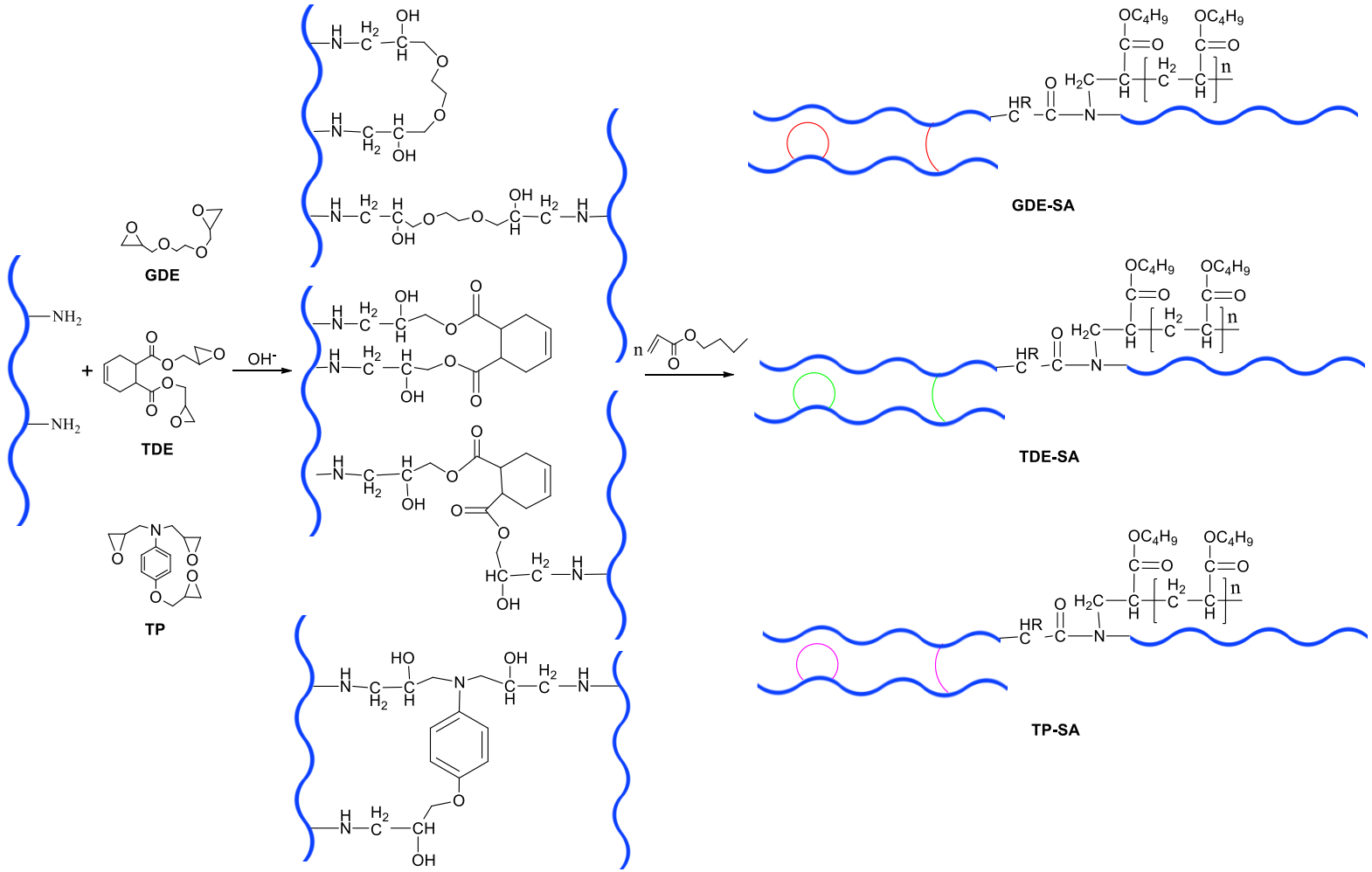

Fig. 1. Schematic diagram of preparation of hydrophobic sizing agents (GDE-SA, TDE-SA, and TP-SA)

The amount of sizing was controlled by choosing appropriate coating bars and coating speed. The sized papers were dried at 105 °C and equilibrated for 24 h at a temperature of 25 °C and 65 ± 20% relative humidity (Dong *et al.* 2014). The amount of sizing agent was adjusted to end up with the same sizing amount on the corrugated paper, which was 8 g/m^2 (Yang *et al.* 2016).

RESULTS AND DISCUSSION

The structural differences between collagen and the three sizing agents are illustrated in the schematic diagram of cross-linked grafting modifications in Fig. 1. Under alkaline conditions, the epoxy groups of the cross-linking agents (GDE, TDE, and TP) were attacked by amino side chains of collagen, leading to the fracture of C−O and the formation of N−C bonds. When the amino side chains of the same collagen attacked the epoxy groups of cross-linking agents, the intramolecular cross-linking bond formed (Liu *et al.* 2017b). When the amino side chains of different collagen attacked the epoxy groups of cross-linking agents, the intermolecular cross-linking bond formed (Liu *et al.* 2016a). Subsequently, the cross-linked collagens were further modified by BA, and the hydrophobic sizing agents (GDE-SA, TDE-SA, and TP-SA) were successfully prepared.

The physical properties of GDE-SA, TDE-SA, and TP-SA are shown in Table 1. The solid content of the three sizing agents was 27.21 wt.%, 23.94 wt.%, and 6.25 wt.%, respectively. Their pH values were 3.8, 3.6, and 3.3, respectively, and the viscosity was 10.04 mPa·s, 9.25 mPa·s, and 6.24 mPa·s, respectively. Obviously, all three modified sizing agent emulsions exhibited an acidic pH value, and the GDE-SA exhibited the greatest viscosity.

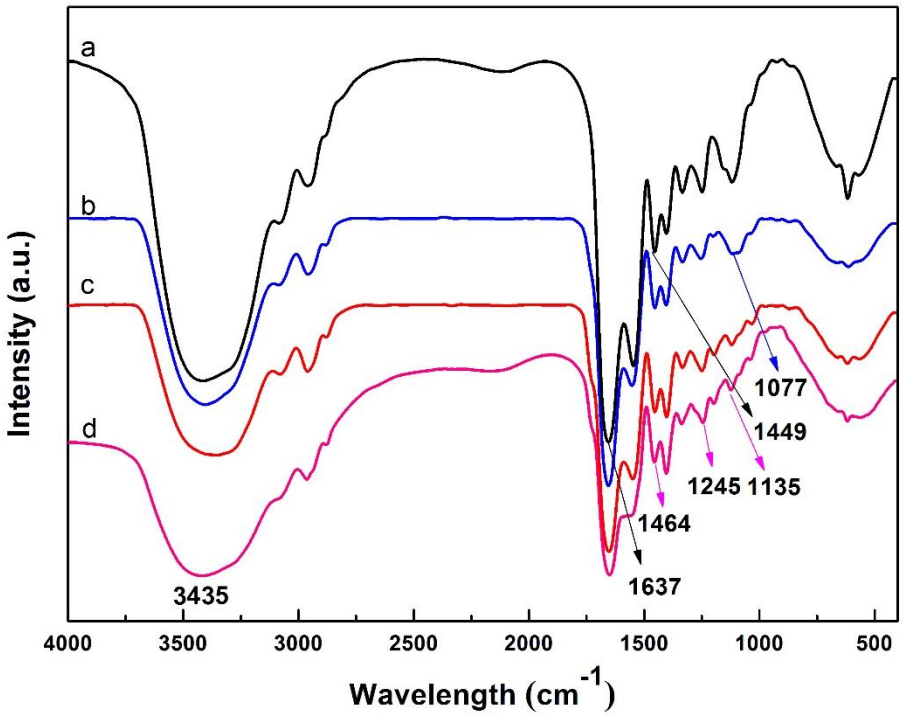

Fig. 2. FT-IR spectra of collagen (a), GDE-SA (b), TDE-SA (c), and TP-SA (d)

Figure 2 displays the FT-IR spectra of collagen, GDE-SA, TDE-SA, and TP-SA. As shown as Fig. 2a, the characteristic absorption peaks at 1637 cm^{-1} (amide I, C=O stretching) and 1449 cm^{-1} (amide III, C=O stretching) were assigned to the stretching of C=O in the peptide bond (Li *et al.* 2015; Pietrucha and Safandowska 2015). The absorption peaks of the amide I band and the amide III band of the three kinds of sizing emulsions move to high wave numbers, which is due to the rearrangement of the groups on the molecular chains of the modified collagen (Wang *et al.* 2014). As displayed in Fig. 2b, the absorption peak at 1077 cm^{-1} was assigned to the C−O−C bond of ether group, indicating that the collagen had been successfully modified (Du *et al.* 2018). Similarly, as displayed in Fig. 2c and 2d, the absorption peaks of C−C(C=O)−O bond at 1135 cm^{-1} also indicate that BA was successfully grafted on the surface of collagen (Wen *et al.* 2017a).

Table 1. Properties Comparison of Three Kinds of Modified Emulsions

Physical Properties	GDE-SA	TDE-SA	TP-SA
Appearance	milky white	milky white	dark brown
Solid Content (wt.%)	27.21	23.94	6.25
pH	3.8	3.6	3.3
Viscosity (mPa·s)	10.04	9.25	6.24

The GPC results of collagen and three sizing agents are shown in Table 2. TDE-SA possessed the appropriate average molecular weight, so it exhibited the most superior polydispersity. In detail, the M_n, M_w, M_p, and M_z of TDE-SA were 2026, 3347, 3381, and 5338, respectively, and its polydispersity coefficient was 1.65. These results indicated that the collagen emulsion is easy to reunite, and the polydispersity is poor. Furthermore, in the sizing agents obtained after modification, the characteristics of collagen emulsion are changed, and the polydispersity is better (Zhang *et al.* 2016). To further study the polydispersity of collagen and three sizing agents, the particle size was tested. Figure 3 shows the size distributions of the collagen, GDE-SA, TDE-SA, and TP-SA emulsions. The average particle size of collagen, GDE-SA, TDE-SA, and TP-SA was 122 nm, 90 nm, 93 nm, and 92 nm, respectively. The particle sizes of the modified emulsions were smaller than the collagen emulsion, indicating that the modified emulsions are more stable than collagen emulsion.

Table 2. GPC Characteristics of Collagen, GDE-SA, TDE-SA, and TP-SA

	Mn[a]	Mw[b]	Mp[c]	Mz[d]	Mz+1	Polydispersity
Collagen	2298	5984	3281	13186	22495	2.6033
GDE-SA	1766	3546	3435	6696	11252	2.0076
TDE-SA	2026	3347	3381	5338	8138	1.6520
TP-SA	2444	4631	3622	8425	13508	1.8951

[a] M_n, The number average molecular weight; [b] M_w, the weight average molecular weight; [c] M_p, the mass average molecular weight; [d] M_z, the viscosity average molecular weight

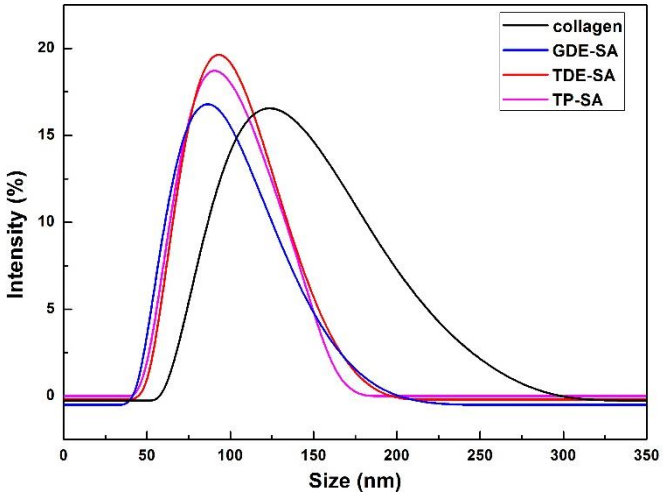

Fig. 3. Particle size distributions of collagen, GDE-SA, TDE-SA, and TP-SA

Prior to TG characterization (Fig. 4), the four emulsions were oven-dried for 12 h at 60 °C. The small weight loss at 0 to 100 °C can be attributed to the residual moisture in the dried samples (Yuan *et al.* 2018). At 150 to 280 °C, collagen displayed a small weight loss because the chain of collagen molecules is more stable, and the structure is not destroyed in this temperature range. The three sizing agents lost more weight than collagen, which can be attributed to the loss of small molecular chain segments. This result indicated that GDE, TDE, and TP were successfully cross-linked with collagen, and small molecular chain segments were formed on the surface of collagen (Striugas *et al.* 2017). In addition, the obvious weight loss observed in four dried samples at 215 to 400 °C was attributed to the destruction of the collagen framework (Si *et al.* 2018). Among them, unmodified collagen had the greatest weight loss, indicating that the structure of modified collagen was more stable.

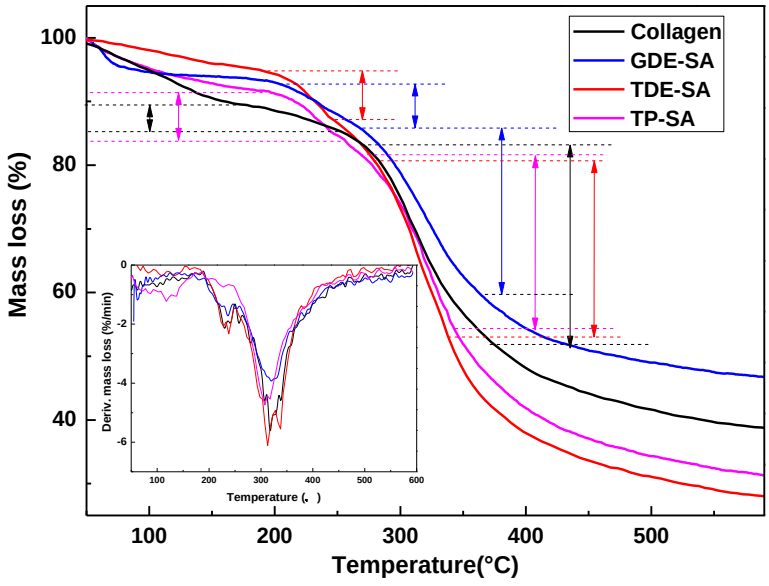

Fig. 4. TG curves of collagen, GDE-SA, TDE-SA, and TP-SA, and the inset is the corresponding

DTG curve

To further study the water resistance of the corrugated paper before and after being coating with the three sizing agents, the corresponding static contact angles were detected. As displayed in Fig. 5, the water drop was completely adsorbed by the surface of uncoated corrugated paper in a very short time of 15 s (Fig. 5a), but the large static contact angles were maintained for a long time after the corrugated paper was coated by the sizing agents (Fig. 5b to 5d). The largest static contact angle (117.2°) was obtained on the surface of corrugated paper coated by GDE-SA (Fig. 5b), indicating that the GDE-SA exhibited the most superior water resistance effect. The water resistance was attributed to the hydrophobic groups of GDE, TDE, and TP. In addition, the ether bonds of GDE exhibited stronger hydrophobicity than the ester bond of TDE, but the water resistance effect TP was weaker than GDE and TDE for the existence of amino groups in the structure (Wang *et al.* 2016).

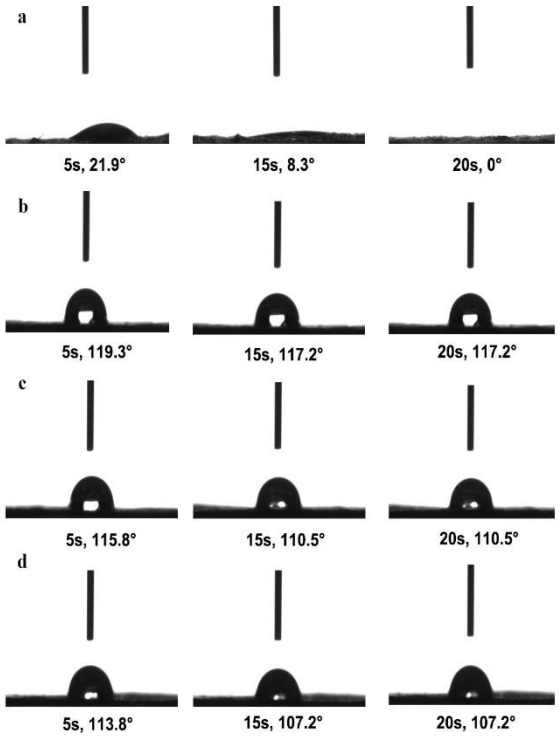

Fig. 5. Static contact angles of water drop with corrugated paper before (a) and after being coated by GDE-SA (b), TDE-SA (c), and TP-SA (d) in different standing time

To study the sizing effect of three different sizing agents on corrugated paper, the transverse section of the corrugated paper before and after being coated by the sizing agents was characterized by SEM. As shown as Fig. 6, uncoated corrugated paper fibers exhibited loose and disorganized arrangement. The corrugated papers treated with GDE-SA, TDE-SA, and TP-SA exhibited improved compact fiber arrangement, which indicated that the three sizing agents successfully penetrated into the interspace of the fibers by physically or chemically binding mode with paper fibers (Chen *et al.* 2017). The corrugated paper treated by GDE exhibited the most compact and smooth fiber arrangement, which explains why the corrugated paper coated by GDE-SA exhibited the largest static water contact angles. This is because GDE showed the best cross-linking effect, which significantly enhanced the strength of the collagen molecular structure. The

combination between sizing agent and corrugated fiber was stronger, making the corrugated paper fiber more compact. Therefore, the combination of GDE-SA and paper fiber was stronger when the GDE-SA was applied to coating corrugated paper, leading to the most compact and smooth fiber arrangement (Liu *et al.* 2016b).

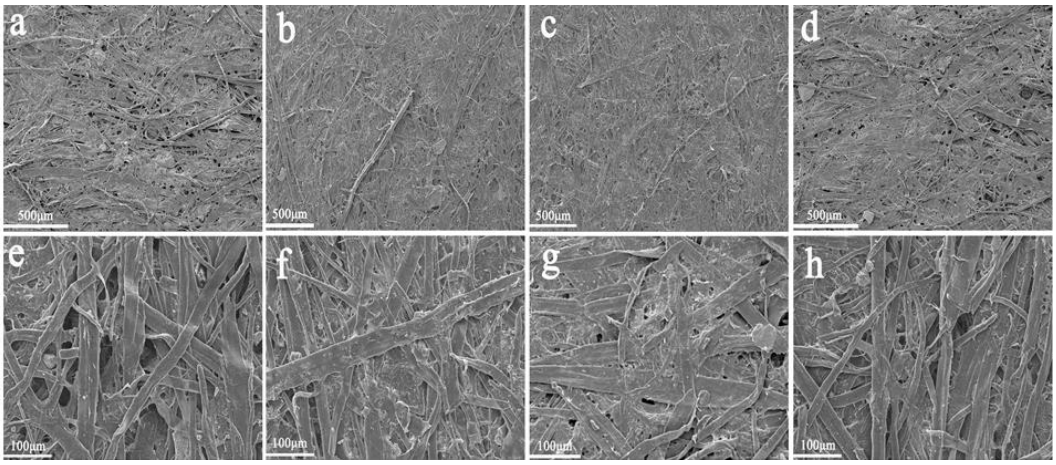

Fig. 6. SEM images of corrugated paper before (a, e) and after being coated by GDE-SA (b, f), TDE-SA (c, g), and TP-SA (d, h) in different magnification

To study the stability of the physical and mechanical properties, the thickness, stiffness, tear strength, tensile strength index, and ring crush index were determined, as shown in Table 3. The uncoated corrugated paper thickness, stiffness, tear strength, tensile strength index, and ring crush index values were smaller. The corrugated paper treated with the three sizing agents exhibited improved physical and mechanical properties. The corrugated paper coated by GDE-SA exhibited better physical and mechanical properties than the other two sizing agents. The thickness, stiffness, tear strength, tensile strength index, and ring crush index were 1.2 times, 2.1 times, 1.6 times, 1.5 times, and 2 times as high as that of corrugated paper, respectively. GDE exhibited a better cross-linking effect on collagen, so the corrugated paper coated by GDE-SA had better physical and mechanical properties (Du *et al.* 2014).

Table 3. Application Performance Comparison of Corrugated Paper

	Thickness (mm)	Standard Deviation	Stiffness (mN·m)	Standard Deviation	Tearing Strength (mN)	Standard Deviation	Tensile Strength Index (N·m/g)	Standard Deviation	Ring Crush Index (N·m/g)	Standard Deviation
CP[a]	0.224	0.001	4.41	0.02	184.360	3.332	35.25	0.03	4.21	0.10
GDE-CP[b]	0.265	0.002	9.21	0.03	302.045	5.673	52.53	0.05	8.23	0.03
TDE-CP[c]	0.248	0.002	8.95	0.02	206.907	4.231	44.53	0.02	7.92	0.03
TP-CP[d]	0.257	0.001	8.00	0.04	254.973	3.267	46.78	0.06	8.02	0.04
AKD-CP[e]	0.246	0.003	8.93	0.03	205.903	4.536	45.03	0.04	7.53	0.08

[a]CP, Corrugated paper; [b]GDE-CP, The corrugated paper coated by GDE-SA; [c]TDE-CP, The corrugated paper coated by TDE-SA; [d]TP-CP, The corrugated paper coated by TP-SA; [e]AKD-CP,

The corrugated paper coated by AKD.

To further study the bending resistance of the corrugated paper before and after coating with the sizing agents, the state of water drop on the folded corrugated paper was tested. As shown in Fig. 7, the water drops were absorbed by the uncoated corrugated paper, and as the corrugated paper was folded more times, more water was absorbed (Fig. 7a). Thus, the uncoated corrugated paper had poor bending resistance (Jiao *et al.* 2018). The water drops were kept completely on corrugated paper coated by the three sizing agents (Fig. 7b to 7d), indicating that the corrugated paper after sizing had improved bending resistance (Wen *et al.* 2017b). The water drops were best kept on the corrugated paper coated by GDE-SA (Fig. 7b). The water drops remained completely on the corrugated paper that had been folded 20 times, and none was absorbed by the paper. This indicated that the corrugated paper treated by the GDE-SA had the best bending resistance.

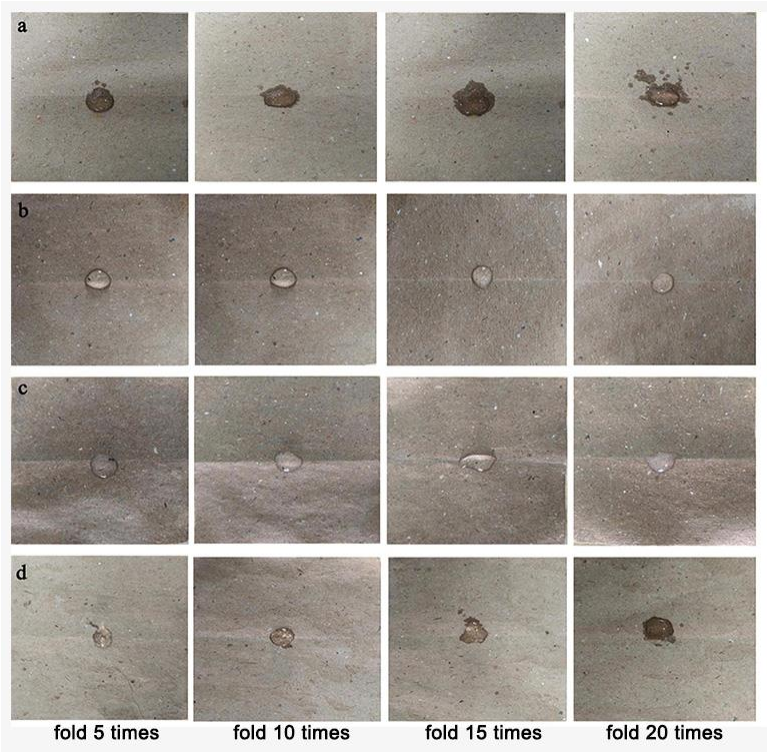

Fig. 7. The state of the water drops on folded corrugated paper after 5 s of dripping.

CONCLUSIONS

1. All three modified surface-sizing agent emulsions exhibited an acidic pH value, and the glycol diglycidyl ether surface-sizing agent (GDE-SA) exhibited the greatest viscosity.
2. The three modified sizing agent emulsions were more stable and had a narrower distribution of particle size than collagen emulsion.
3. The three modified sizing agents exhibited higher thermal stability than collagen.
4. The GDE-SA coated corrugated paper exhibited the best water resistance,

smoothness, and physical and mechanical properties.

5. The corrugated paper treated by the GDE-SA had the best bending resistance.

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