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Preparation and Properties of Waterborne Varnish for On-Demand Inkjet Printing

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Abstract. *A kind of waterborne varnish for inkjet printing was synthesized, and properties of the waterborne varnish were characterized to make it suitable for the glazing requirements of inkjet printing and other printability requirements. The waterborne varnish was synthesized from epoxy resin (E-51), epoxy diluent (ED), acrylic acid, trimellitic anhydride, maleic anhydride and organic amine by three steps of ring opening reaction, esterification reaction, neutralization reaction. The viscosity, film-forming property, water absorption of waterborne varnish and the water resistance, lightness, wear resistance and bonding strength of coated paper were tested. The effects of the ratio of E-51 and ED in the polymerization system on the properties of waterborne varnish were studied. In the test of printability of self-made waterborne varnish, the absorption of digital inkjet paper to varnish and the influence of varnish on color reproduction of printed image were discussed and studied. The results show that when the molar ratio of epoxy group in E-51 and ED is about 1:1, the prepared varnish is suitable for inkjet printing, and its film-forming property, such as water resistance, adhesion and friction resistance are better. At the same time, its printability is also better.*
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1. INTRODUCTION

Traditional packaging and printing productions have some problems, such as uncertain printing quantity, inventory pressure and so on. With the continuous development of digital printing technology, graphic processing technology and network technology, significant changes have taken place in the production mode of packaging and printing [1–3]. On demand inkjet printing is a process, in which the colorant is sprayed from the nozzle to the substrate under the control of the computer, that the information will be copied in the form of graphics and text [4, 5]. With the advantages of on-demand, personalization, energy saving and environmental protection, it is a non-plate, non-pressure and non-contact printing technology. The on-demand inkjet printing technology has a wide range of markets and has become a research hotspot in recent years [6–12].

Varnish is a kind of functional coating, which is mainly used for paper packaging and printing [13–17]. The packaging printed matters, which are coated with varnish, would have a better appearance and higher durability. Furthermore, at the same time, it would be conducive to recycling [18–20]. As people pay more and more attention to environmental protection, environment-friendly varnishes have gradually become the focus of research. Among the types of varnishes, waterborne varnish has the characteristics of non-toxic, low stimulation, low volatile organic compounds (VOCs) and low cost, which has attracted increasing attention from manufacturers, consumers and scholars [21–27].

In this study, the influences of E-51 and ED on the preparation and performance of waterborne varnish in the polymerization system were explored, and the best proportion of E-51 and ED would be obtained, which would provide a reference for the preparation and synthesis of varnish. At the same time, the printability of the self-made waterborne varnish was tested, the absorption of the varnish on the digital inkjet paper and the influence of the varnish

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on the color reproduction of the printed image were also discussed and studied, which would further verify that the varnish is suitable for inkjet packaging and printing.

2. EXPERIMENTAL

2.1 *Equipments Used*

The particle sizes of the polymer were tested by a Malvern, UK particle size analyzer of model number Nano-ZS90. The viscosity of the reactant system was tested by a viscometer, the model of which was DVS and the brand was Brookfield. The coating samples were made by xb-1 bar coater, which was produced by Shandong Yuanwei Instrument Equipment Co., Ltd. Adhesion and friction resistance were tested by QFZ-II paint film adhesion tester and MCJ-01A wear tester respectively. The digital inkjet proofs were printed by Epson SureColor P9080 inkjet printer. The absorption test of digital inkjet paper to varnish was completed by Kruss dsa100s automatic contact angle tester. The color density of the inkjet color sample was measured by using X-rite standard edition spectrodensitometer.

2.2 *Materials Used*

The varnishes in the experiment were synthesized by E-51 epoxy resin, polyethylene glycol diglycidyl ether, polypropylene glycol diglycidyl ether (ED), acrylic acid, triethylamine, trimellitic anhydride and dimethylaminoethyl methacrylate. Digital inkjet paper used three kinds of inkjet digital paper, the gram weight was 70 g/m^2, 80 g/m^2 and 100 g/m^2 respectively.

2.3 *Sample Preparation and Performance Tests*

2.3.1 *Sample Preparation*

The waterborne varnishes were synthesized by the following steps. Firstly, a certain proportion of E-51 and ED were esterified with acrylic acid. Secondly, when the acid value of the system decreased to a certain value, trimellitic anhydride and maleic anhydride were added to further esterification. Finally, dimethylaminoethyl methacrylate and triethylamine were introduced to neutralize the system, and deionized water was added to adjust the solid content [28, 29]. Seven groups of E-51 and ED with epoxy group molar ratio of 3:1, 2:1, 1.5:1, 1:1, 1:1.5, 1:2, 1:3 were selected in the experiment, and the varnishes, prepared during the experiment, were marked as sample ①–⑦.

The grammage of the paper, used in this experiment, is 70 g/m^2, 80 g/m^2 and 100 g/m^2 respectively. And these three kinds of inkjet digital paper were recorded as DP70, DP80 and DP100 respectively. The samples were printed by Epson inkjet printer. The substrate was placed in a room with a temperature of 21 ± 1°C and a relative humidity of $50 \pm 5\%$ for 24 h before printing, which simulated the printing environment [30]. The post-press glazing process was completed by coating with bar coater.

2.3.2 *Performance Tests*

Viscosity. DVS digital viscometer was used to measure the viscosity of the system at a temperature of 23 ± 0.5°C. The rotor and speed would be selected, and each sample would be measured at least twice at the same temperature. The average value was calculated as the final viscosity of the system.

Solid Content. The solid content of the sample was tested according to ISO 3251:2003.

Particle Size. The sample was diluted with deionized water to a solution with a mass fraction of 0.5%, and the particle size was tested by a particle size analyzer at normal temperature.

Water Absorption. The dried curing film would be cut into square pieces, which have a mass of about 0.2000 g. After weighing accurately (m_0), these square pieces would be immersed in distilled water and taken out after 24 h. The filter paper was used to wipe off the water on the surface of the membrane quickly, and the mass of the membrane (m_1) would be weighed accurately. The water absorption (a) would be calculated according to formula (1).

$$A = \frac{m_1 - m_0}{m_0} * 100\%. \quad (1)$$

In Formula (1), m_1 is the mass of the membrane after immersion, and m_0 is the mass of the membrane before immersion.

Water Resistance. The varnishes were dried to form a film and soaked in water for 50 min. Pulverization, cracking, light loss and other phenomena would be observed to see whether they will appear on the samples. If there were no phenomena as described above, it would be judged as qualified.

Adhesion and Friction Resistance. The adhesion of the coating was tested according to the Chinese national standard GB 9286-98 tape paper method. The abrasion resistance of the coating was tested according to the Chinese national standard GB/T 1768-2006.

Absorption of Varnish on Digital Inkjet Paper. The absorption of self-made waterborne varnish on digital inkjet paper with different grammage was tested. The absorption process of varnish on paper was observed by contact angle tester, and the corresponding time was recorded.

Color Measurements of Paper, Prints and Varnished Samples. In order to evaluate the influence of varnish on the color reproduction performance of printed image, the CIE $L*a*b*$ values of prints and varnished samples were measured by spectrophotometer according to ISO 5-4:2009 standard. ΔE_{00} equation [30] was used to calculate the corresponding color difference. Each sample would be measured ten times, and the average value was taken as the experimental result.

3. RESULTS AND DISCUSSION

3.1 *Viscosity of Waterborne Varnish*

In this experiment, the combination of ED and E-51 was used to reduce the viscosity of epoxy acrylic resin system on the premise of ensuring the mechanical properties of prepolymer

Table I. The initial viscosity of the reaction system under different ratios of E-51 and ED.

Number	Sample ①	Sample ②	Sample ③	Sample ④	Sample ⑤	Sample ⑥	Sample ⑦
Viscosity (mPa s)	1350.08	692.07	512.00	272.05	226.50	189.10	133.22

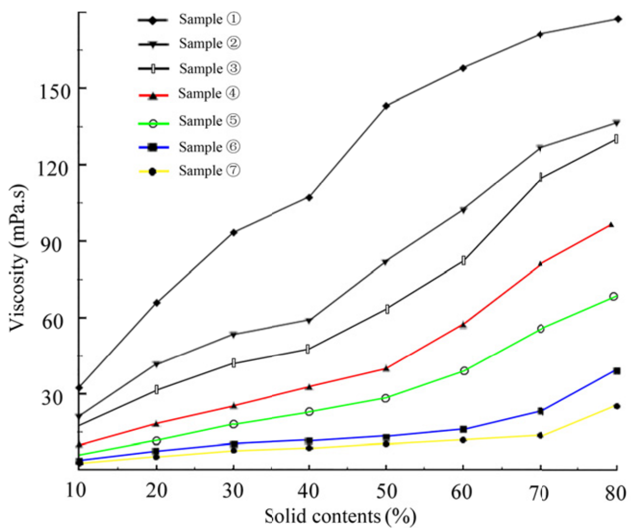

Figure 1. Effect of the ratio of E-51 and ED on the viscosity of varnish with different solid contents.

system. It is difficult to use E-51 in inkjet varnish system because of its high viscosity. The results of the experiments are shown in Table I.

It can be seen from Table I that with the increase of ED addition, the viscosity of the initial resin system decreases. When the epoxy molar ratio of E-51 to ED is 1:1, the viscosity of the initial system decreases greatly from 8325.80 mPa s to 272.05 mPa s, and the viscosity reduction rate would be 96.73%.When the proportion of ED is increased, the average molecular weight in the polymer system becomes smaller and the distribution of prepolymer with large molecular weight will be smaller, which may reduce the viscosity of the whole polymer system.

In this experiment, the viscosities of the samples ①–⑦ were tested, and the viscosities of the varnish system were tested with different ratios of E-51 and ED and different solid contents. The experimental results are shown in Figure 1.

As shown in Fig. 1, the viscosity will increase when the solid content of the prepared varnish increases from 10% to 80%. The viscosities of sample ①–③ system will increase rapidly with the solid content more than 40%, and the viscosities of most varnish are more than 50 mPa s, which cannot meet the requirements of inkjet printing. The viscosities of sample ④–⑦ are relatively small, which can also meet the requirement of low viscosity even when the solid contents become high. In the varnish polymer system, the reduction of relative molecular weight and solid content will reduce the opportunity proportion of intermolecular instantaneous entanglement and the possibility of hindering the orientation of molecular chain along the shear force direction, which will eventually reduce the viscosity of the varnish polymer system.

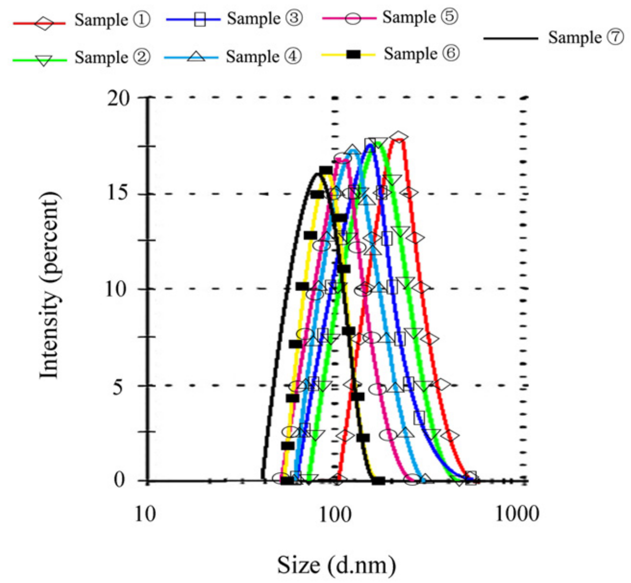

Figure 2. The particle size of the varnish system with different molar ratios of epoxy groups (E-51:ED).

3.2 *Particle Size of Waterborne Varnish*

The proper particle size has great influence on the range of use and performance of the varnish. In this experiment, the particle sizes of varnish sample ①–⑦ were tested to explore the effect of the compound ratio of E-51 and ED on the particle size of the system. The synthetic varnish would be checked for whether it would be suitable for inkjet printing. The experimental results are shown in Figure 2.

It could be seen from Fig. 2 that as the amount of ED increases, the particle sizes of the varnish system, which were prepared in the experiment, become smaller and smaller. The particle sizes of the varnish system in sample ④–⑦ are about 100–200 nm, with high and narrow peaks. The results show that the particle sizes of the varnishes are uniform and the distribution is concentrated, which would be suitable for on-demand inkjet printing.

3.3 *Water Absorption of Waterborne Varnish*

The prepared varnish samples ①–⑦ were diluted to different solid contents for drying and curing, and the water absorption of the varnishes was measured as shown in Figure 3.

It can be seen from Fig. 3 that when the solid content of samples ①–⑦ is less than 40%, with the increasing of solid content, the water absorption rate of the varnish-cured film layer decreases gradually. The varnish with higher solid content has relatively high internal cross-linking density, which is not conducive to the penetration of water into the film.

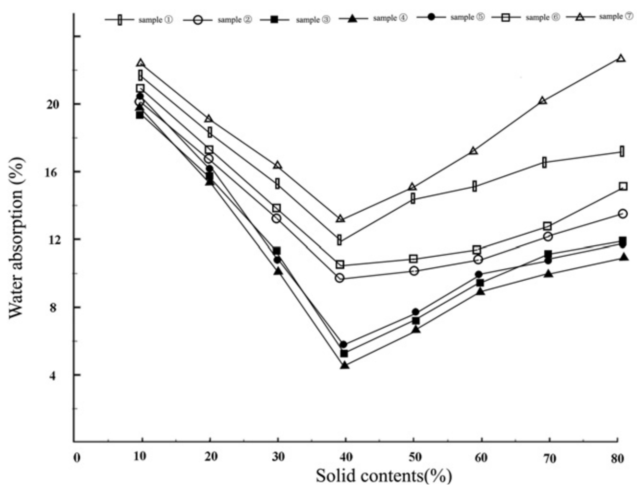

Figure 3. Effect of the ratio of E-51 and ED on the water absorption of varnish.

Table II. Water resistance of waterborne varnish under different ratios of E-51 and ED.

Sample	Performance		
	Pulverization	Crack	Glossy
①	Qualified	Qualified	Qualified
②	Qualified	Qualified	Qualified
③	Qualified	Qualified	Qualified
④	Qualified	Qualified	Qualified
⑤	Qualified	Qualified	Unqualified
⑥	Unqualified	Qualified	Unqualified
⑦	Unqualified	Unqualified	Unqualified

However, the number of hydrophilic groups will also increase with the increase in solid content. Hence, when the solid content of varnish becomes greater than 40%, the water absorption of the cured film will increase. The experimental results show that the water absorption of sample ④ is 4.6%, and the performance of the sample is the best.

3.4 *Water Resistance of Waterborne Varnish*

The waterborne varnish prepared in this experiment contains hydrophilic groups such as hydroxyl group, ether group, carboxyl group and amino group, which have certain water solubility. But it is required to have certain water resistance after being dried in the process of using varnish. Therefore, in this experiment, samples ①–⑦ with a solid content of 40% were tested for water resistance, and the results are shown in Table II.

From Table II, when the amount of ED is too large, it will affect the water resistance of the varnish film. The water resistance of samples ①–④ film is good, and there are no cracking, pulverization, light loss and other phenomena. Sample ⑤ have a slight loss of gloss. The light loss of sample ⑥ is more serious than that of sample ⑤. The water resistance of the film of sample ⑦ is the worst, and there are some phenomena such as cracking, pulverization and losing gloss. With the increase of ED dosage, the number of hydrophilic groups in the varnish prepolymer system will also increase under the same solid content. After the varnish is dried, the probability of hydrophilic groups on the surface of the paint film will also increase, which will make its water resistance worse.

Table III. The adhesion and friction resistance of waterborne varnish.

Performance	Sample						
	①	②	③	④	⑤	⑥	⑦
Adhesion	Yes	Yes	Yes	Yes	Yes	No	No
Friction resistance (g)	0.0011	0.0024	0.0031	0.0036	0.0048	0.0072	0.0108

3.5 *Adhesion and Friction Resistance of Waterborne Varnish*

Stick the tape paper on the film and tear it off after pressing, after the coating film is cured. If the film layer is torn off by the tape paper, the adhesion would be deemed to be unqualified. If the film is intact, it would be regarded as qualified. The friction resistance of the film is tested according to the test standard of the wear resistance of the film.

Under the condition of friction pressure of 20 ± 0.3 N, the sample will be rubbed 50 times with a friction testing machine. The sample will be weighed when the chip is removed, and the weight loss will be calculated simultaneously. The same sample will be tested many times, and the arithmetic mean value will be taken as the result. The above test results, in this study, are shown in Table III. If the adhesion test is qualified, it would be recorded as “Yes”, whereas the unqualified one would be recorded as “No”.

As can be seen from Table III, when the amount of ED is too large, the adhesion of the film formed by varnish will be relatively poor. Especially when the epoxy molar ratio of E-51 to ED exceeds 1:2, more films will be stuck. The friction resistance of the coating also decreases with increasing ED content, and especially when the ratio of E-51 to ED exceeds 1:1.5, the weight loss weight will increase significantly. For the varnish polymer system, the molecular weight will directly affect the mechanical properties. Different molecular weights have different mechanical properties. The higher the molecular weight, the better the hardness and strength, and the better the adhesion and friction resistance.

After the experimental tests (3.1–3.5) and analysis of the results, the comprehensive performance of waterborne varnish of sample ④ is found to be better, and will be used in the following experiments and tests.

3.6 *Absorption of Waterborne Varnish on Digital Inkjet Paper*

The waterborne varnish prepared in this experiment is intended to be used in inkjet printing. Therefore, it is necessary to test whether the absorption on commonly used inkjet digital paper is good.

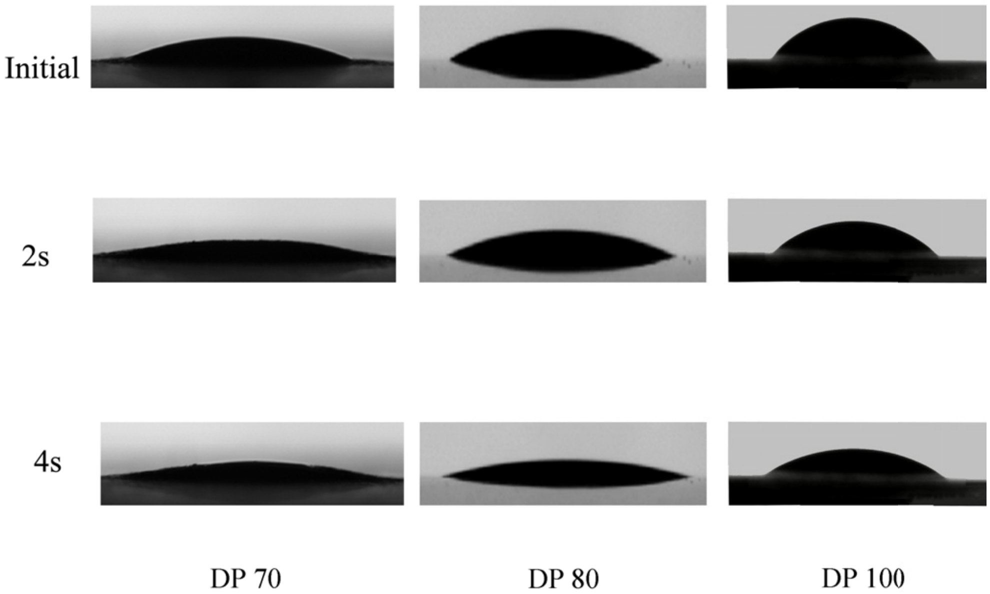

Figure 4. Absorption of waterborne varnish on different papers.

In this experiment, the absorption processes of waterborne varnish on different papers were tested, while the absorption of the varnish on different papers was recorded by contact angle tester. The absorption processes of the varnish on DP 70 (70 g/m^2), DP 80 (80 g/m^2) and DP 100 (100 g/m^2) digital inkjet paper are shown in Figure 4.

As can be seen from Fig. 4, the varnish gradually spreads completely on the paper within 4 s. Among them, the evaporation and penetration time of varnish on the surface of 70 g/m^2 digital inkjet paper is less than that on 80 g/m^2 and 100 g/m^2 digital inkjet paper. With the increase in gram weight of paper, the spreading and absorption process of varnish will take more time to complete.

The digital inkjet papers used in this experiment are all two-side offset papers. The coating on the surface of the base paper and the structure of the paper sheet may affect the absorption capacity of the paper and the penetration capacity of the varnish on the paper at the same time. As the paper weight increases, the paper structure would become firmer. If the amount of coating on the surface of the such a paper became larger, both of them would inhibit the absorption of varnish on the paper.

3.7 *Effect of Varnish on Color Reproduction of Inkjet Printing Image*

Varnish can improve the relative performance of printed matter, while at the same time trying not to reduce the image color reproduction performance of the original printed matter.

In this experiment, the square field color blocks of inkjet printing pattern are designed, which have yellow (Y), magenta (M), cyan (C) and black (K), as can be seen in Figure 5.

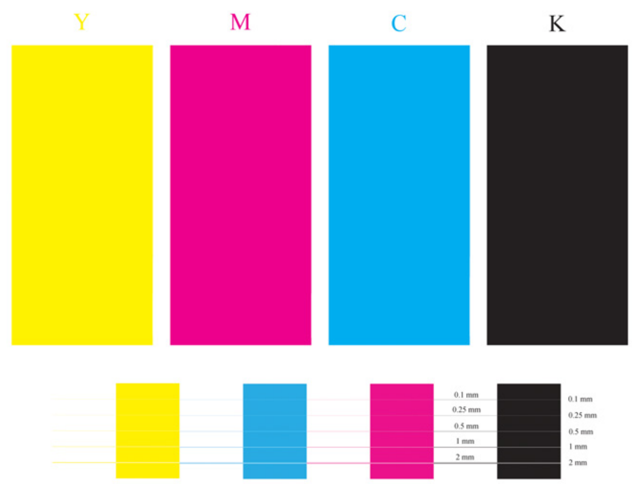

Figure 5. Digital inkjet printing sample.

Varnish will be applied to the inkjet printing sample, and with its absorption and drying on the sample, a thin film will be formed. With the increase in the amount of varnish coating, the thickness of the film will also show a certain increase. In order to verify the effect of varnish on the color restoration of digital inkjet printing, five kinds of coating rods (4 μm, 6 μm, 8 μm, 10 μm and 12 μm) were used for coating. The coating experiment was carried out on digital inkjet paper with different gram weight. The L ∗ a ∗ b∗ values of each color of the printed matter after the varnish drying were tested and compared with the uncoated printed matter, and then the color difference ΔE_{00} was obtained.

The experimental test results are shown in Figure 6. The test sample number is represented by A B, where A is the ink color and B is the paper sample code (for example, if the color

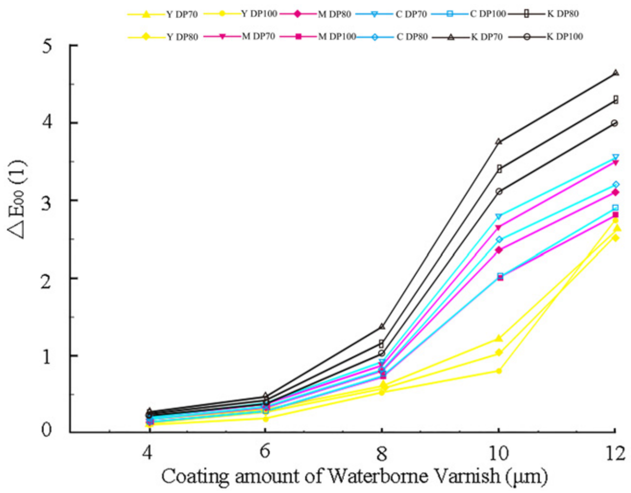

Figure 6. Influence of different amounts of varnish coating on ΔE_{00}.

difference of yellow printed on 70 g/m^2 paper is tested, the test sample number would be recorded as Y DP70).

It can be seen from Fig. 6 that when the coating amount of waterborne varnish becomes greater, the color difference of digital inkjet paper with different gram weight has the same increasing trend. Especially if the coating amount of varnish is more than 8 μm, the color difference of each color will increase significantly. In terms of color, the black color difference increases obviously with the increase in coating amount of the varnish. The color difference changes of magenta and cyan are close relatively, but when the coating amount of varnish is more than 8 μm, the color difference will increase greatly. The change of yellow color difference is small. But when the coating amount of varnish becomes more than 10 μm, the color difference of yellow will increase obviously.

With the increase of the film thickness of the varnish on the printed image information surface, the optical properties of the printed matter might be affected and then the image color reproduction ability would be affected at the same time. Nevertheless, the ΔE_{00} of all samples are within the range of less than 5, which can hardly be seen by human eyes [30], and does not affect the graphic reproduction quality of inkjet packaging.

4. CONCLUSIONS

The results of synthesis experiments show that when the molar ratio of ED and E-51 epoxy groups is 1:1 (sample ④), the reactive waterborne varnish system has better performance, such that the average particle size is less than 0.2 μm with moderate viscosity. The film performance test results show that when the molar ratio of ED and E-51 epoxy is 1:1 (sample ④), the water absorption of waterborne varnish film is low, and the water resistance, adhesion and friction resistance are good, which can be used in on-demand inkjet printing system. The physical and chemical properties of the prepolymer for waterborne varnish have a decisive influence on the performance of the glazing system, and its film-forming properties, transparency and abrasion resistance would need to be further improved. The mature and stable synthesis process would also need to be determined by refining experiments.

The test results of the printability of the waterborne varnish show that the varnish can spread well on digital inkjet paper and has good absorption. The test results of color reproduction performance of inkjet printing images also show that the varnish prepared in this experiment has little negative influence on the image reproduction performance of original digital inkjet printing products, and the color difference changes caused by the varnish are within the range of human vision, which can be ignored.

The waterborne varnish prepared in this experiment is not only suitable for digital inkjet printing online process glazing but also for off-line process glazing. However, in the course of the experiment, it is found that if the surface of unglazed printed matter is stained, wrinkled and has other pollution phenomena, the surface of the printed matter after glazing would also have corresponding pollution. Therefore, it is suggested that digital inkjet online glazing or online coating hybrid glazing technology should be used to reduce the pollution of printed matter before glazing.

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