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A novel porous adhesion material with ink absorbency for digital inkjet printing

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Lei Zhang,[a] Yingjie Cao,[b] Lei Wang,[a] Lu Shao,[a] and Yongping Bai*[a]

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A novel porous adhesion material for digital inkjet printing with absorbency of eco-solvent ink and removable adhesion has been successfully prepared by a simple, robust and cost-effective method. There was no traditional emulsifier with low molecular weight adopted in the poly(n-butyl acrylates) emulsion for the preparation of the porous material. Due to the property of this reactive emulsifier, there was no free emulsifier left after the polymerization, thereby reducing the drawbacks associated with traditional emulsifier. The porous material possesses high shear strength, moderate peel strength and low tack properties suitable for adhesion and could be removable many times, which offered a new method for recycling. Since the novel porous material can be utilized many times and have the potential of recycling, we believe that this material would be environmental friendly. Additionally, the absorbency of eco-solvent ink created a suitable platform for digital inkjet printing based on the novel porous material, with resistance of different atmospheres, like acid, base, oxidant, and many organic solvents.

Introduction

Water-based polymerization processes are preferred because of the strict regulations regarding the removal of volatile organic compounds (VOCs).[1-3] Emulsion polymerization is a candidate for preparing the polymers due to the emphasis on environmental-friendly.[4-6] To stabilize the emulsion, emulsifiers are used to increase the kinetic stability.[7-9] Emulsifiers with low molecular weight are used in traditional emulsion polymerization, which has negative effect on the physical properties of the polymers.[10-12] The migration of the low-molecular-weight emulsifiers to the interfaces or polymer surfaces may cause the decrease of the properties of final products.[13] Consequently, in order to reduce the detrimental effect associated with traditional emulsifiers, reactive emulsifiers are needed.[14-16] In this work, SR-10 was used to obtain the emulsion with satisfied adhesion properties and stabilities. There was no free emulsifier left after the polymerization, since SR-10 could easily react with acrylic monomers, which were widely used on account of no harm to the environment, low cost, high optical clarity and aging resistance.[17-20]

Foaming emulsion has been widely used in daily life and industrial applications.[21-23] Generally, air bubbles were generated under a strong mechanical force in the emulsion, and could be stable with foaming agent in the system.[24] The foaming system was thermodynamically unstable due to three main reasons: foam drainage, film rupture, and bubble coarsening. Thus, to obtain such a stable foaming system, foaming agents are required.[25-27] There are several kinds of foaming agents commonly used in previous reports, such as rosin, protein and sodium dodecyl sulphate. However, there are some drawbacks of these foaming agents. For example, the foaming system with rosin has low foam expansion and stability.[28] While, the viscosity of the foaming system using protein is too high, which is not suitable for practical coating.[29] Similarly, the foaming system using sodium dodecyl sulphate possesses a high foam expansion, but low foam stability.[30] Hence, a foaming agent with less disadvantages needs to be explored. Therefore, ammonium stearate was a good choice, which had more advantages, such as high stability, suitable viscosity for coating, and low cost.

Digital inkjet printing (DIP), a breakthrough in the traditional printing technology, develops quickly under the promotion of information technology and advertising industry, creating a digital image by propelling droplets of ink onto substrates.[31-35] There are lots of advantages of DIP, such as high level of efficiency, flexibility, cleanliness, creativity and competitiveness. Hence, it has been implemented in industrial markets to meet the needs for increasing productivity and lower operating cost.[36-38] With its rapid development, the requirements of different consumables, like pressure sensitive adhesives (PSAs) and ink absorption layer, are growing year by year.[39-42] The adhesion properties of the conventional PSAs are contributed by the chemical groups of the polymers. However, the shear strength is not high enough for strong bonding without any physical force. And PSAs cannot be easily removed or recycled.[43-48] Additionally, it is not suitable for DIP on the surface of the material directly, since the adhesion surface is easy to rub the nozzle of the printing machine. Furthermore, PSAs and ink absorption layer are generally coated on the two sides of one substrate, which is unfit in some special situations.[49-51] Due to its eco-friendly property, water resistance, outdoor durableness, ultraviolet (UV) resistance and low toxicity, eco-solvent ink is gradually replacing other inks. Therefore, an adhesion material with ink absorbency is needed, such as exhibition sheets, large posters for tall buildings, protective films for inside vision, etc. Furthermore, emulsion has been widely applied in the area of DIP, however, there is rare report about foamed emulsion used for DIP. Hence, foaming emulsion system was adopted for the fabrication of substrate for DIP, which can fulfil the requirements of removable adhesion properties and ink absorbency in DIP area.

For DIP application, the tack should be no more than 7 #, the 180 ° peel strength should be between 80 and 160 N/m, and the shear holding power should be more than 72 h. All the above parameters are the requirements in DIP area. Additionally, ink absorbency is evaluated with the effect of inkjet printing. The printing performance should be gorgeous, with no vortex point or white point. And the boundaries between different colors should be clear, with no halo ink phenomenon. PSA can be divided into five kinds shown in the Table 1.[52]

Table 1 Classification of PSA according the adhesion properties

Kind of PSA	180° peel strength (N/m)
Excellent permanent	>560
Permanent	400~560
Semi-removable	240~320
Removable	80~160
Excellent removable	<40

In this work, a novel porous material for DIP with removable adhesion properties and absorbency of eco-solvent ink has been successfully prepared by a simple, robust and cost-effective method. First, the poly (n-butyl acrylates) (PBA) emulsion was synthesized in the presence of reactive emulsifier, which could supply fantastic properties for the final products. Second, the adhesion properties of this porous material were determined by two components, i.e., chemical bonding between functional groups of the adhesive and the adherend, as well as the physical effect from the porous structure. Hence, the shear strength was very high, which avoided the adhesive coming off when shear forces were applied. Third, due to the PSA's absorbency of eco-solvent ink, printing on the adhesion side was performed, and could be attached to various adherends. Fourth, our final product has the tack of less than 3 #, the 180 ° peel strength of 95.8 N/m, and the shear holding time is larger than 100 h, all these properties can fulfill the requirements of DIP area very well. Last but not least, the printed PSAs had resistance to many different solvents and solutions. Hence, the porous adhesion material is suitable for various practical applications.

Experimental

Materials

Reactive emulsifier SR-10 was obtained from Adeka Co., Ltd. The structure of SR-10 is:

$$\begin{array}{l} CH_2{=}CH{-}CH_2{-}O{-}CH_2 \\ \qquad\qquad\quad | \\ R{-}O{-}H_2C{-}CHO(CH_2CH_2O)_{10}SO_3NH_4 \end{array}$$

The monomers of n-butyl acrylate (BA) and acrylic acid (AA) were provided by Wuxi Jiani Chemical Co., Ltd. Ammonium persulphate (APS) was purchased from Shanghai Lingfeng Chemical Reagent Co., Ltd. $NaHCO_3$ and NaOH were obtained from Shanghai Hongguang Chemical Co., Ltd. Foam stabilizer 9288 (ammonium stearate emulsion, with 30 % solids content), corona polypropylene (PP), methyl alcohol, ethyl alcohol, ethylene glycol, diethylene glycol, glycerol, n-decane and gasoline were provided by Shanghai Zeafee Digital Inkjet Composite Material Co., Ltd. Hydrochloric acid, potassium dichromate, zinc chloride and copper chloride were obtained from Sigma-Aldrich (China). Eco-solvent ink CMYK for digital inkjet printing was obtained from Tianjin Huaxin-Micolor ink Co., Ltd. Deionized water (DW) was used throughout the experiment.

Synthesis of PBA emulsion

First, the pre-emulsion was prepared with a stirring speed of 1500 rpm for 0.5 h using a mixture of DW (360 g), SR-10 (13.2 g), BA (640.2 g) and AA (6.6 g). Second, DW (240 g), APS (1.1 g) and $NaHCO_3$ (1.32 g) were added into the flask which was dipped in a heating batch at 82 ^{0}C and kept at the temperature constant for 15 min. The stirring speed was 200 rpm. Third, the polymerization was conducted by drop-wise addition of the pre-emulsion and the initiator solution containing DW (120 g) and APS (2.2 g) into the system respectively, which lasted about 4 h. Finally, the flask was heated to 85 ^{0}C for 1 h for the fully polymerization.

Preparation of porous adhesion materials

PBA emulsion (50 g) and ammonium stearate emulsion were mixed by a high-speed dispersion machine (dispersible plate of unchanged denticle, Jiangyin Finemachinery Co., Ltd., China) with a speed of 1000 rpm for 30 min to obtain a foaming emulsion system with large amount of air bubbles. After the dispersion process, the foamed emulsion was coated onto corona PP film using a coater QXG (Shanghai Jiehu Instrument Co., Ltd) with a coating speed of 50 mm/s. Then the coated film was dried in an oven at 95 ^{0}C for 5 min. The dry-thickness of the coating was 100 μm.

Characterizations of porous adhesion materials

Viscosity was measured by a rotational viscometer (Brookfield Engineering Laboratories Inc, USA) at 25 ^{0}C. The surfaces of porous materials were measured by a digital microscope (Gaosuo Digital Technology Co. Ltd., China). Contact angle was measured using a JC2000D contact angle meter (Zhongyikexin Sci. & Tech. Co. Ltd., China). The picture of color piece was printed on the materials by digital printing equipment Mutoh VJ-1604 (Colormade Digital Technology Co. Ltd., China). Stabilities of the material after digital inkjet printing were tested under conditions of water, inorganic ions, organic solvents, acids and bases.

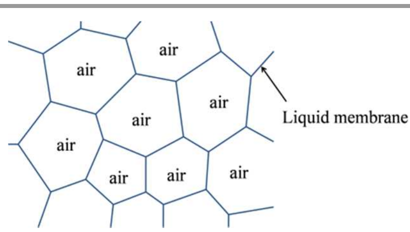
The adhesion properties were tested by ball tack tester CZY-J, shear strength tester CZY-S and electron tensile testing machine XWL (Jinan Labthink Mechanical & Electrical Technology Co. Ltd., China). A 100 mm×100 mm porous material was cut down for tack test. Steel balls of different size were released from the top of an incline with an angle of 30 °. The tack was obtained by the measurement of the balls rolled onto the tape after a distance of 100 mm for slipping, and the serial number of the biggest ball which stopped on it was recorded. A 25 mm×70 mm porous material was cut down for test of shear holding time. A tape distance of 20 mm was applied on one of the steel sheet, and the rest for another. After a dwelling time of 1h, the sample was clamped with a 2 kg load. The shear holding power was measured by the time that elapsed between the application of load and the completed exfoliation of the tape from steel sheet. Porous material of 25mm in width was cut down for 180° peel test. It was applied to the steel sheet (or other materials) as an adherend with a roller moving back and forth three times with a speed of 300 mm/min to make a contact. After a dwelling time of 20 min, the sample was tested with a drawing speed of 300 mm/min. The peel force at a distance of 75 mm was recorded. Porous material of 20 mm in width was cut down for test of shear strength. A distance of 20 mm was applied to the adherend with a roller moving back and forth three times. And it was tested with a drawing speed of 300 mm/min.

Results and discussion

Effect of the foam stabilizer on the foaming system

In the foaming system, the surface area grows with the formation of bubbles, as well as the energy of the whole system. According to the Gibbs theory, all the systems tend to stay in a low energy state. Hence, when the foam stabilizer was added into the system, the surface tension decreased, and the pressure difference also decreased, thus leading to the slow speed of foam drainage, which benefited the stability of bubbles. In the meanwhile, the addition of foam stabilizer enhanced the surface intensity and viscosity of the liquid membrane (shown in Scheme 1), which made the drainage harder. So the decrease of the thickness of the liquid membrane became slower, and the air permeability reduced, which were also beneficial to the stability of bubbles.

Fig. 1 shows the images of the PBA emulsion with foam stabilizer under a dispersion speed of 1000 rpm for 30 min with the amount of foam stabilizer of 5 wt %. The volume obviously increased during the dispersion process. After the high-speed dispersion, the viscosity increased from 50 to 2000 mPa·s. The volume became four times larger than that of the mixture before stirring. The formation of a large number of air bubbles was contributed by the high-speed dispersion, while the stability of air bubbles in the foamed emulsion were due to the existence of ammonium stearate emulsion, which was used as a foam stabilizer. That was because ammonium stearate could effectively reduce the surface tension of the emulsion, thus leading to the stability of air bubbles, as discussed in the paragraph above. The half-life time ($t_{1/2}$) of the foam was 192 h, which was better than other foaming system of previous result.[53]



Scheme 1 Illustration of liquid membrane

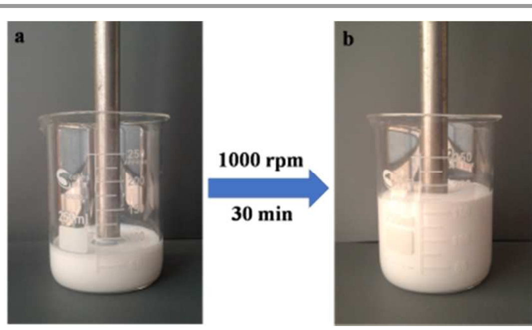


Fig. 1 Images of PBA emulsion with foam stabilizer before (a) and after (b) stirred. (The amount of ammonium stearate emulsion was 5 wt %)

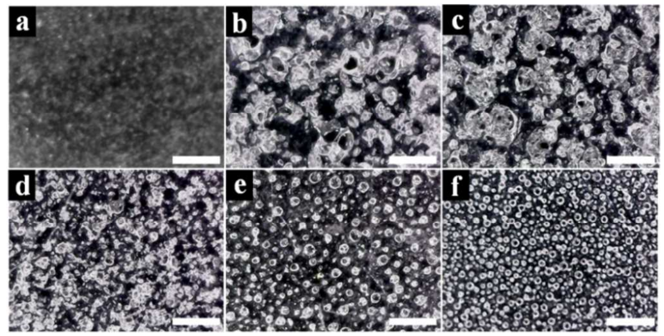


Fig. 2 Microscope photographs of obtained adhesion materials with pores using foamed emulsion with different amounts of foam stabilizer. (a. 0 %, b. 1 wt %, c. 2 wt %, d. 3 wt %, e. 4 wt %, f. 5 wt %). The scale bars equal 300 μm.

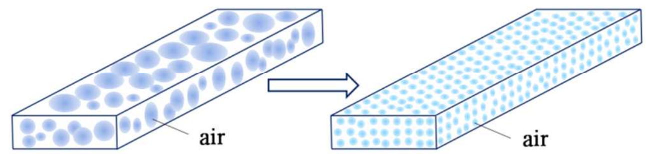


Fig. 3 Schematic images of the microstructure's change of the adhesion material with the growth of the amount of the foam stabilizer. The circles represent air bubbles.

Effect of the foam stabilizer on the microstructure of porous adhesion materials

Porous materials were prepared by the foamed emulsion. Fig. 2a-f shows the microstructure of the dry final materials with pores using different amounts of foam stabilizer ammonium stearate emulsion. The size of pores decreased and the amount of the pores increased with the growth of the amount of ammonium stearate emulsion. The pores were not well-distributed when the amount of ammonium stearate emulsion was less than 3 wt % (Fig. 2b-d). When the amount of ammonium stearate emulsion reached 4 wt %, there was obvious appearance of uniform pores (Fig. 2e). The reason was probably that when there was enough foam stabilizer, the pores could be easily stabilized without merging with each other. Hence, we can see lots of pores in Fig. 2f with the amount of 5 wt %, which was illustrated by Fig. 3.

Effect of foam stabilizer on the adhesion properties of porous materials

The properties of the adhesion material can be judged by several aspects, such as the 180° peel strength, shear strength and the tack properties.[46] The following section will discuss the effect of foam stabilizer on the properties of adhesion material.

As is shown in Fig. 4a, the 180° peel strength of the porous materials decreased with the growth of the amount of the ammonium stearate emulsion. The strength was 137.9 N/m when there was no foam stabilizer, while the strength decreased gradually to 94.5 N/m with the amount of 6 wt %. Few pores with different sizes formed with a small amount of ammonium stearate emulsion, while more well-distributed pores formed when the amount of ammonium stearate emulsion increased, which could be obviously observed from Fig. 2. Therefore, the porous material was easy to be peeled off from the adherend, since the adhesion area between the adhesive layer and adherend decreased. The 180° peel strength of the porous material can fulfill the requirements of removable adhesion properties.[52]

Fig. 4b shows the effect of the amount of the ammonium stearate emulsion on the shear strength of the porous materials. The shear strength went up with the increase of the amount of the ammonium stearate emulsion. The strength was only 0.13 MPa, while the strength increased to 0.70 MPa with the amount of 5 wt %, since more well-distributed pores formed with the growth of the amount of ammonium stearate emulsion. When the addition of ammonium stearate emulsion was too much, the viscosity of the system would be too high, which made the liquid membrane much brittle, thus making the bubbles easy to merge. So the stability of the whole system decreased with a high ratio of ammonium stearate emulsion. From Fig.4b, when the addition was 6 wt %, the shear strength decreased to 0.69 MPa.

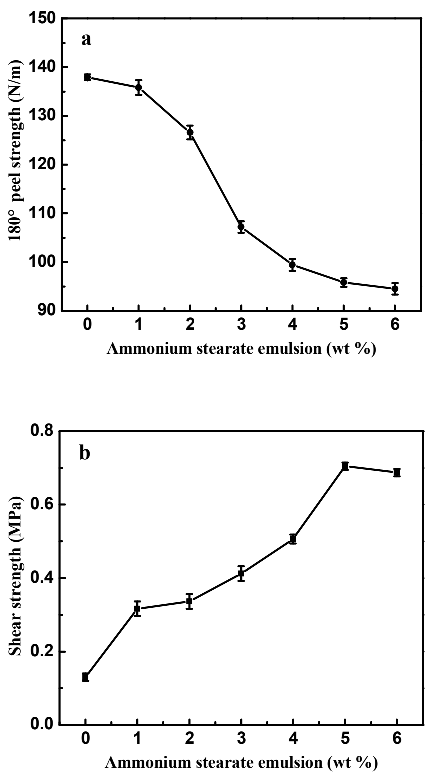


Fig. 4 Effect of the amount of ammonium stearate emulsion on 180° peel strength (a) and shear strength (b)

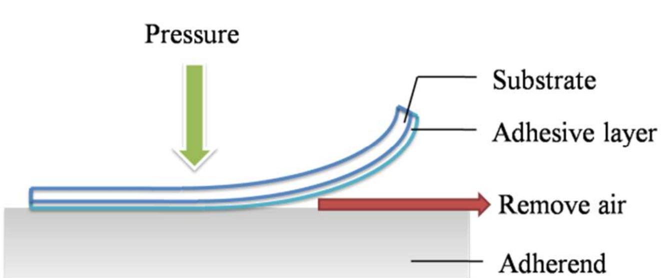


Fig. 5 Schematic image of adhesion process of porous material (The substrate was PP, the adhesive layer was porous polymer, and the adherend could be stainless steel, glass and aluminium)

Fig. 5 is the schematic image of the adhesion process between porous material and adherend. The adhesion properties are mainly determined by two parts: one is the chemical properties of the polymer adhesives; the other is the physical properties of pores, which eliminated the air inside the pores under a certain pressure, thus leading to the pressure difference between the inside and outside of pores. The low T_g of the polymer, had benefits for removing air when the material was adhesion to the adherend for its flexibility, which can fulfill the requirements of adhesives. And the high shear strength appeared under a pressure when adhesion, due to removing air from the pores.

The shear holding time of the porous materials were more than 100 h with the application of a high shear force, which also confirmed the data in Fig. 4b. Based on the above results, an ideal porous material with well-distributed pores could be obtained when the amount of ammonium stearate emulsion was 5 wt %, which is better than the reported result of 500 min based on poly(n-butyl acrylate-co-acrylic acid) latexes.[42]

Table 2 shows the 180° peel strength and shear strength of porous material bonded on six kinds of surfaces. The porous materials can be adhesive to many kinds of surfaces. Generally, traditional adhesives used for adhesion on polar surfaces cannot be adhesive to glass, since the composition of glass consisted of inorganic compounds. The adhesion properties of porous material bonded on glass showed that the bonding was enhanced.

The tack properties of porous materials were less than 3 #. The steel balls did not stop on the surface of the porous adhesion layer due to the low tack, since there were a large number of pores, which increased the roughness of the surface. Yet, it gave a nice bond with the adherend under a light pressure. And the materials could be easily removed from the adherend without any residue due to their light tack properties.

Performance of digital inkjet printing

Digital inkjet printing can be performed on the porous material, which has absorbency of eco-solvent ink. The amount of the ammonium stearate is 5 wt%.

Fig. 6a shows the porous material printed with seven color pieces. The ink output of seven colors from left one to right one were 100, 100, 100, 100, 400, 200 and 200 %. There was no vortex point or white point on the surface of the material. The printing performance was gorgeous with various kinds of colors. And there were clear boundaries between different colors, with no halo ink phenomenon. The contact angle of the porous material before digital inkjet printing (Fig. 6b) was 102.3±2.4°, while it decreased to 97.5±1.7° after digital inkjet printing (Fig. 6c). That was because there were additives in the ink, which increased the hydrophility.

Table 2 Adhesion properties of porous material bonded on several surfaces

Adherend surface	180° peel strength (N/m)	Shear strength (MPa)
Stainless steel	95.8	0.70
Glass	66.0	0.47
Aluminium	90.6	0.68
Wood	57.7	0.32
Polyurethane paint	81.3	0.58
Card board	85.6	0.61

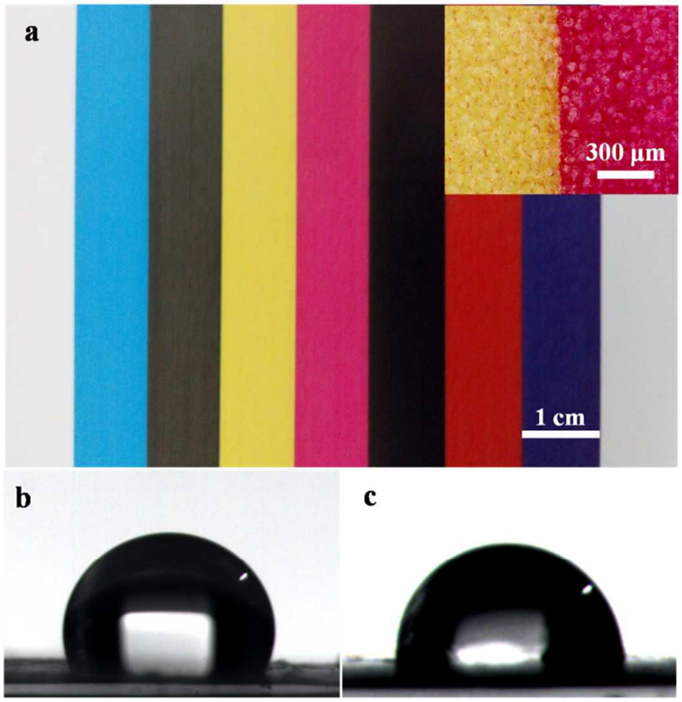


Fig. 6 The porous material (5 wt% of ammonium stearate) printed with color piece (a), and the contact angle of the porous material before (b) and after (c) digital inkjet printing

Table 3 Adhesion properties of porous material after digital inkjet printing

Adherend surface	180° peel strength (N/m)	Shear strength (MPa)
Stainless steel	86.4	0.63
Glass	50.2	0.31
Aluminium	77.6	0.61
Wood	42.8	0.28
Polyurethane paint	62.6	0.43
Card board	69.8	0.57

Table 4 Test conditions of various chemical media resistance

Chemical media	Solutions containing inorganic ions	Organic solvents	Acids	Bases
Test conditions (72 h)	Potassium dichromate (strong oxidant) Zinc chloride Copper chloride	methyl alcohol ethyl alcohol ethylene glycol diethylene glycol glycerol n-decane gasoline	pH=3 pH=5	pH=10 pH=12

The adhesion properties of the porous material after digital inkjet printing are shown in Table 3. After digital inkjet printing, the porous material can also bond to various kinds of surfaces, which indicated that the material was suitable for practical applications. To my best knowledge, our material was the first one possessing the ability of both ink absorbency, and removable adhesion after digital inkjet printing.

Stabilities of the material after digital inkjet printing

Water resistance of the material after digital inkjet printing was tested by immersing in water for 1 month, there was no fading on the printed material. And the boundaries between different colors were still clear, with no halo ink phenomenon, which indicated that the water resistance of porous material could satisfy for practical application. Similarly, chemical media resistance of the material was also tested under various conditions (Table 4). After kept in the different conditions for 72 h, the boundaries between different colors were still clear, with no halo ink phenomenon or fading.

Conclusions

Digital ink printing was performed based on the porous PSA layer. The optimal amount of ammonium stearate emulsion is 5 wt % for the preparation of the porous material, which can form a large amount of well-distributed pores. The porous material had removable adhesion properties when bonded to various kinds of surface, due to the adhesion from chemical and physical bonding. Our final product has the tack of less than 3 #, the 180 ° peel strength of 95.8 N/m, and the shear holding time is larger than 100 h, all these properties can fulfill the requirements of DIP area very well. The material has absorbency of eco-solvent ink, which can be used as inkjet coatings, with resistance of chemical media, such as inorganic ions, organic solvents, acids and bases. This porous material can also be adhesive to different surfaces after digital inkjet printing. To my best knowledge, our material was the first one possessing the ability of both ink absorbency, and removable adhesion after digital inkjet printing, which is suitable for digital inkjet printing and many other applications.

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[a] School of Chemical Engineering and Technology, Harbin Institute of Technology, Harbin 150001, P.R. China. Fax: +86-451-86418270; Tel: +86-451-86413711. Corresponding authors: baifengbai@hit.edu.cn (Y. Bai); zhanglei0807@126.com (L. Zhang); leiwangmaryland@gmail.com (L. Wang).

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