



# Development of green waterborne UV-curable vegetable oil-based urethane acrylate pigment prints adhesive: Preparation and application



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## ABSTRACT

The energy consumption and environment pollution play an important role to the sustainable development in the dyeing and finishing industry. The aim of this study is to synthesize a series of acrylated epoxidized soybean oil based UV-curable polyurethane pigment adhesives and to reduce the consumption of energy and environment pollution in pigment printing process. The chemical composition was evaluated using fourier transform infrared spectra and nuclear magnetic resonance hydrogen spectroscopy. The thermal properties of UV-cured films were discussed by thermalgravimetric analysis (TGA) and differential scanning calorimetry (DSC). In addition, the effects of AESO content on particle size, water swelling properties and contact angles of the emulsions and films were investigated. Furthermore, the color yield, the rubbing fastness and washing fastness of the printed fabrics with acrylated epoxidized soybean oil based UV-curable polyurethane as adhesive were evaluated. The application of vegetable oil-based urethane acrylate pigments of cotton fabrics is a cleaner method that can reduce the consumption of energy and environment pollution. Compared to conventional thermally curable coatings, the UV-curable vegetable oil-based urethane acrylate pigment prints adhesive does not require a large amount of water and energy, indicating that vegetable oil-based urethane acrylate pigments have potential application in the cleaner production of textile industry.

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## 1. Introduction

In recent decades, polyurethane acrylate (PUA) is widely used as oligomer for UV adhesives due to its outstanding properties including excellent abrasion resistance and adhesion on the fabrics, superior flexibility, hardness, chemical resistance and weather ability (Sow et al., 2010). However, as more and more environmental concern around the world have promoted the efforts in the search and use of alternative raw materials in the polymer industry (Hormaiztegui et al., 2016). Waterborne polyurethane acrylate (WPUA) material with environmental protection has been paid more and more attention in recent years (Shin et al., 2013). Compared with the conventional polyurethane acrylate, waterborne polyurethane acrylate(WPUA) exhibits the attractive characteristics of environment friendly, low toxicity, competitive price,

etc. (Xu et al., 2012).

For the textile industry in China, which is the most important industries in China (Chen et al., 2017), energy consumption and environment pollution become increasingly serious. Pigment printing is the mainly method of printing application for more than about 60 percent of the printing products (Neral et al., 2006). However, thermally curable coatings printed textile must be dried and then cured with heat to convert the binders to a tough polymer, leading to consuming large amounts of energy. Compared with thermally curable coatings, UV curing technology is considered to be environmentally friendly due to the formulations release very small amounts of volatile organic compounds (VOCs) and use less energy (Mishra et al., 2015). In textile printing application of UV-curing technology has been mentioned in some articles (El-Molla, 2007) or some patents. M.M. El-Molla (2007) has prepared a series of the aqueous UV-curable binder of polyurethane acrylate oligomer based on the mixture of PEG1000 and PEG2000.

The UV-curable technology is being introduced to textile printing and dyeing. however, several problems still exist in its

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application (Elmolla and Schneider, 2006). The commercial UV-cured WPU are typically terminated with mono acrylate units, which has a low crosslink density and a slow curing rate, resulting in lower water resistance, poor mechanical properties (Jena et al., 2013). Therefore, the use of multi-double key functional group of polyurethane to improve the curing efficiency and crosslink density (Mao et al., 2017). Currently, a large of researches for WPUs are based on designing the molecular structure to improve the density of cross-link (Chen et al., 2015).

In order to modify the properties of UV-WPUAs film, silane coupling agent (Xu et al., 2015), modified acrylic resin, vegetable oil (Shahid ul et al., 2013), chitosan, and other methods such as graphene (Xu et al., 2016) was used. Luo et al. (2016) reported that the incorporation of epoxidized castor oil (ECO) into the WPUs improved the antiwater, thermal and mechanical properties of WPU films. Vikash Mishra et al. (2015) observed improved mechanical properties and water- and chemical-resistance when UV-WPU was synthesized in the presence of castor oil. Based on these studies, it has been found that chemical modification using certain compounds with a good hydrophobic or water-repellent property will be an effective way to solve the problem of poor water resistance. Therefore, the castor oil with good resistance against chemicals and solvents are one of the most suitable candidates (Li et al., 2015). However, castor oil is not as widely available as soy oil in China, and processing the castor beans to produce the oil results in a waste product containing ricin, a highly toxic protein. Acrylated epoxidized soybean oil (AESO) (Mauck et al., 2016) as one of the major soybean oil derivatives, which contains two types of functional groups (Fig. 1): one is –OH groups that can covalently bond using some reactive chemical groups, and the other is C=C bonds that are able to free-radical UV-cured. And it can be cured in the presence of oxygen and drier to form a more compact network structure by partially replacing polyester diol or polyether glycol (de Haro et al., 2016) as soft segment. In addition, the long chain non-polar fatty acid chain makes the film have excellent hydrophobicity, toughness and cold resistance, which has attracted widespread attention in the field of polyurethane (Ni et al., 2009). However, much less has been reported on the preparation and application properties of acrylated epoxidized soybean oil (AESO) based UV-curable polyurethane on printing fabrics.

We have recently developed a series of AESO-based UV-curable polyurethane pigment prints adhesive which can reduce the consumption of energy and water in pigment printing process. The results are very interesting in the cleaner production of finishing process. Hence, in this work, the synthesis of UV-curable polyurethane adhesive from the AESO with different content bio-polyols was accomplished. The structures and thermal properties of the AESO-based UV-WPUAs were characterized. The printing performance and thermal stability of the resultant polyurethane coatings were investigated. The colorfastness of the fabrics printed with these pigment prints adhesives were also analyzed. Additionally, the influence of the vary irradiation time on the degree of curing and the water resistance was discussed.

## 2. Experimental

### 2.1. Materials

Soybean oil was obtained from the local market. Triphenyl phosphine (TPP) was purchased from Aladdin Industrial Corporation (Shanghai, China). Hydroquinone was received from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). 4-methoxyphenol was obtained from Sigma-Aldrich Co. LLC. Isophorone diisocyanate (IPDI, AR, >99.0%(GC)), 2-hydroxyethyl methacrylate (HEMA, AR, >95.0%(GC)) and 2,2-dimethylol

propionic acid (DMPA, AR, >97.0%(T)) were purchased from TCI (Shanghai) Chemical Industry Co., Ltd. Poly (caprolactone glycol) (PCL, 1000 g/mol) was obtained from Jining macro Ming chemical reagent CO., Ltd. (Jining, China). Dibutyltindilaurate (DBTDL, AR), triethylamine (TEA, AR, 101.19 g/mol) and acetone were obtained from Sinopharm Chemical Reagent Co., Ltd. Pigment Red 8111 FFG from Jiang Wei Da City Auxiliary Factory (Zhejiang, china), Darocur 1173 was obtained from Sinopharm Chemical Reagent Co., Ltd.

Isophorone diisocyanate (IPDI) was used without further purification. However, acetone was dehydrated with 4 Å molecular sieves for at least two weeks, PCL1000 was dewatered at 120 °C for about 2 h in under vacuum conditions and the 2,2-dimethylol propionic acid (DMPA) was dried at 120 °C for almost 4 h, then put them in dryer before use.

### 2.2. Synthesis of epoxidized soybean oil

Soybean oil (25.0 g) and formic acid (6.25 g) were mixed in a three-necked round-bottom flask, equipped with dropping funnel, mechanical stirrer and the thermometer. To start the reaction, hydrogen peroxide solution (28.0 g) was slowly added into the three-necked flask reaction system. The reaction was continued for 4.5 h and maintained at the temperature of 50 °C. The crude product was filtered and washed repeatedly with deionized water to obtain a pH of 7.0.

### 2.3. Synthesis of acrylated epoxidized soybean oil

Epoxidized soybean oil (21.0 g), triphenyl phosphine (TPP) (2.40 g), and 4-methoxyphenol (0.24 g) as a catalyst and inhibitor were added into a three-necked flask equipped with mechanical stirrer, a nitrogen inlet and constant pressure dropping funnel. The mechanical stirrer was maintained at 500 rpm. Acrylic acid (3 g, 1/0.95 equivalent amount to epoxy content) was added drop wise to the stirred evenly reaction mixture and maintained at 80 °C for 1 h. Then the reaction system was heated to 120 °C for 3 h. The reaction was monitored by measuring acid values per half an hour. After the reaction, the final product was analyzed by FT-IR. A representation of this reaction was illustrated in Scheme (1) Fig. 1 (Kahraman et al., 2006).

### 2.4. Preparation of epoxidized soybean oil acrylate based UV-WPU

The synthetic route of this reaction was shown in Scheme (2) Fig. 1. The detailed synthesis experimental formulations are illustrated in Table 1. The reaction was carried out in a 250 mL. Under nitrogen, IPDI was mixed with PCL1000 and different amounts of AESO was added in a four-necked round-bottom flask equipped with a mechanical stirrer, thermometer and snake-shaped reflux condenser. then 3–4 drops of DBTDL were added as a catalyst. The reaction was continued for 3 h at 60 °C, until the content of –NCO reached theoretical value. Then DMPA dissolved in acetone was added in the reaction system and was held at 75 °C for 2 h. During the reaction, acetone was added into the round-bottom to reduce the viscosity of the system. 2-Hydroxyethyl Methacrylate (stabilized with MEHQ) was injected into the system. And then the reaction was maintained at 75 °C for 3 h. After the temperature was reduced to 40 °C, TEA (equivalent to total molar content of (DMPA), as a neutralizer, was then added for 30 min while stirring. The mixture was emulsified with deionized water at 1500 rpm for 30 min. After which the acetone was distilled off by rotary vacuum evaporator, to obtain slight yellow blue emulsion.

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