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Physicochemical properties of modified soybean-flour adhesives enhanced by carboxylated styrene-butadiene rubber latex

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ABSTRACT

As an abundant and renewable resource with high biodegradability, soybean-flour (SF) adhesive has aroused further interest by many researchers. Modified soybean-flour (MSF) adhesives with high solid content offer great potential as alternatives to petroleum-based adhesives. The objective of this study was to investigate the physicochemical properties of MSF adhesives modified by carboxylated styrene-butadiene rubber latex (XSBRL). The solid content, viscosity, adhesion, structural change, and morphological properties of the MSF/XSBRL adhesives were characterized in detail. Viscosity of the MSF adhesives was reduced remarkably and the dry and wet shear strengths of the MSF/XSBRL adhesives were enhanced with the increased addition of XSBRL, which acted as both a viscosity reducer and enhancer. X-ray diffraction patterns and morphological images indicated that the cured adhesives became more compact and homogeneous (following XSBRL modification), which prevented damage by moisture or water. Attenuated total reflection-Fourier transformation infrared spectroscopy showed that no apparent chemical reaction occurred between XSBRL and the MSF adhesives. The introduction of polar carboxyl groups contributed to high adhesion, superior emulsibility and good flowability of XSBRL, which improved the adhesion performance and reduced the viscosity of the MSF adhesives.

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1. Introduction

The various forms of wood utilization represent an extremely large and diverse market for adhesives [1]. Most adhesives used in wood composites are formaldehyde-based adhesives, such as phenol formaldehyde (PF), urea formaldehyde (UF), melamine urea formaldehyde (MUF), etc [2,3]. However, the problem is the formaldehyde emission during production and in the first periods of the lifetime of the thermosetting resins panels [2–4]. In recent years, due to the pollution caused by petroleum-based adhesives and the increasing environmental concern, Soy-based adhesives with good adhesion properties have been considered as alternatives for petroleum-based polymers in the manufacture of adhesives since they are derived from renewable resources. On the contrary, the resources of petroleum-based adhesives are non-renewable and limited [5–8]. Polyurethane (PUR) adhesives are used as formaldehyde free wood adhesives, but their short pot life, high cost and low durability has limited their extensive use in the wood-based panel industry [9].

Soy-based adhesives, developed initially in 1923, are inexpensive, abundant, and environmental-friendly. This abundant protein resource can be obtained as a by-product from the processing of soybean oil industry. Industrial utilization of soy proteins for the production of biodegradable resins has led to increased interest in these materials for further potential applications as well as enhancing the values of such systems [10–12]. Soy proteins as wood adhesives have many unique properties such as renewability, abundance, operability, low press temperature, and high wood-binding ability with relatively high moisture content [13]. However, soy-based adhesives cannot be used extensively, and their development is limited because of poor water resistance and bonding strength.

Substantial research has been conducted in the last few decades to improve the adhesion properties of soy proteins for manufacture of wood products [10,14–16]. Soy proteins can be used in the industry because of the functional properties. The modification of soy proteins aims to improve the functional properties by altering their molecular structure or conformation at

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the secondary, tertiary, and quaternary levels using physical, chemical, or enzymatic agents [5,17,18]. The adhesion properties of soy protein adhesives can be improved via the use of denaturants, reducing agents, or cross-linking agents as well as via enzyme hydrolysis [19–23]. However, their overall performance is still inferior to the petroleum-based adhesives, especially in terms of wet adhesion strength.

Blending is an important method in composites manufacture and has gained intensive attention because of the strong economic incentives arising from the use of polymer blends [24]. These blends may combine the advantages of both components or possess better properties than either component. Therefore, the blending of soy proteins with other polymers is obviously of practical significance and, potentially, improved water resistance for wood applications [25–27,6]. Soy proteins can be modified into functional copolymers that interact and react with commercial synthetic adhesives to enhance adhesion performance [8]. However, the properties of polymer blends greatly depend on their morphology, compatibility, and interaction between components [14].

Carboxylated styrene-butadiene rubber latex (XSBRL) is a water dispersed copolymer obtained via the polymerization of butadiene, styrene and a small amount of unsaturated carboxylic acid. XSBRL is widely used in the carpet, paper, non-woven fabric, construction, paint, water-based adhesives and other industries [27]. Some panel manufacturers blend XSBRL with UF resins for the production of man-made board covers as substitutes for the polyvinyl acetate emulsion adhesives. The shear strength and formaldehyde emission of plywood bonded with XSBRL modified UF resins could meet the requirements of China National Standard for type II plywood. The reason is that the emulsion bonding performance improved by the carboxyl groups will enhance the polarity of the adhesives. The butadiene and unsaturated carboxylic acid play important roles when used to modify UF resins [28].

The objectives of this study were to investigate the physico-chemical properties of the modified soybean-flour (MSF) adhesives with commercial XSBRL, such as adhesion, structural change, viscosity, and morphological properties.

2. Materials and methods

2.1. Materials

Soybean-flour (SF) with an average protein content of 45.2 wt% and moisture content of 5.0 wt% was purchased from Sanhe Hopefull Group Oil Grain Food Co. Ltd (Hebei, China). XSBRL was purchased from Xinxiang Helue lida power materials Co., Ltd. (Henan, China). Other chemical reactants were analytical grade and obtained from Beijing Chemical Reagents Co., Ltd (China). The Poplar veneer (*Populus tomentosa Carr*) was obtained from Wen'an County (Hebei, China) with a moisture content of 8.0 wt% and the bulk density was about 0.35–0.40 g/cm^3. Polyamideamine-epichlorohydrin (Solid content, approximately 50 wt%) was used as the cross-linking agent. This was synthesised in the laboratory.

2.2. Preparation of MSF/XSBRL adhesives

The following is a representative procedure for the preparation of the MSF adhesives and XSBRL modified MSF adhesives.

MSF adhesives: SF (28 g), tap water (72 g), and crossing-linking agent (12 g) were sequentially added to a three-neck flask and stirred for 30 min at room temperature.

MSF/XSBRL adhesives: Various proportions (0 wt%, 5 wt%, 10 wt%, 15 wt%, 20 wt%, 25 wt%) of XSBRL based on MSF adhesives content were added to the MSF adhesives and further mixed until uniform at room temperature.

2.3. Solid content of the MSF/XSBRL adhesives

The solid content of the adhesives were determined using the oven-drying method. Approximately 3 g (weight α) of the adhesive was placed into an oven with the temperature set to 120 ± 2 °C for drying until a constant weight (weight β) was obtained. The drying process took approximately 120 min. The value of the solid content was calculated using the following equation. The average value of the solid content was calculated from three parallel samples.

$$\text{Solid content } (\%) = \frac{\beta(g)}{\alpha(g)} \times 100\%$$

2.4. Dynamic viscoelastic measurement

The apparent viscosity of the fresh MSF/XSBRL adhesives were determined using a rheometer with a parallel plate fixture (20 mm diameter). The distance was set to 1 mm for all of the measurements. The experiments were conducted under a steady shear flow at 23 °C. The shear rates ranged from 0.1 to 240 s^{-1} in 10 s^{-1} increments. All of the measurements were conducted in triplicate, and the average value was reported.

2.5. X-ray diffraction (XRD)

The adhesives were cured in an oven at 120 ± 2 °C until a constant weight was obtained and ground into a powder. XRD patterns were recorded on an XRD diffractometer (XRD-6000, Shimadzu, Kyoto, Japan) using a cobalt source and 0.2 theta scan ranging from 5° to 80° at 45 kV and 30 mA.

2.6. Attenuated total reflection-Fourier transformation infrared spectroscopy (ATR-FTIR)

The adhesives were cured in an oven at 120 ± 2 °C until a constant weight was obtained and ground into powder. The ATR-FTIR spectra of the cured adhesives were recorded on a Nicolet 7600 spectrometer (Nicolet Instrument Corporation, Madison, WI) from 500 to 4000 cm^{-1} with a 4 cm^{-1} resolution using 32 scans.

2.7. Scanning electron microscopy (SEM)

The samples were poured into a piece of aluminium foil and dried in an oven at 120 ± 2 °C until a constant weight was achieved. A Hitachi S-3400N (Hitachi Science System, Ibaraki, Japan) scanning electron microscope was used to observe the fractured surfaces and side surfaces of the MSF/XSBRL adhesives. The surface was sputter coated with gold prior to examination.

2.8. Preparation of three-layer plywood samples

Poplar veneers with dimensions of 400 × 400 × 1.6 mm^3 (width × length × thickness, moisture content: 8.0 wt%) were used. The MSF/XSBRL adhesives were coated on both sides of the core veneer using a brush. The dosage of adhesive was maintained at approximately 188 g/m^2. The adhesive-coated veneer was stacked between two uncoated veneers with the grain direction of the two adjacent veneers perpendicular to each other. Before hot pressing, the veneers with adhesives were allowed to stand at room temperature for 10 min and then assembled. The stacked veneers were hot-pressed at 1.0 MPa at 120 °C for 6 min. After hot-pressing, the panels were stored at ambient temperature for at least 24 h prior to cutting it into specimens for evaluation of shear strength and water resistance.

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