



Preparation and properties of a starch-based wood adhesive with high bonding strength and water resistance



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ARTICLE INFO

Article history:

Received 5 February 2014

Received in revised form 15 August 2014

Accepted 17 August 2014

Available online 2 September 2014

Keywords:

Wood adhesive

Starch

Silane coupling agent

Bonding strength

Water resistance

ABSTRACT

A Highly efficient method was developed for preparing starch-based wood adhesives with high performance, using H_2O_2 , a silane coupling agent and an olefin monomer as an oxidant, cross-linking agent and comonomer, respectively. The effects of various parameters on the shear adhesive strength were investigated in the dry state (DS) and wet state (WS). The results indicated that the bonding strength of starch-based wood adhesives could reach 7.88 MPa in dry state and 4.09 MPa in wet state. The oxidation could reduce the content of the hydroxyl transforming into carboxyl and aldehyde groups, and the graft copolymerization enhanced the thermal stability, which improved the bonding strength and water resistance. The starch-based adhesive and the fractures in the bonded joints were analyzed via Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM) and thermogravimetric analysis (TGA). The improved properties were attributed to the modified of microstructure of the graft-copolymerized starch-based adhesive.

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1. Introduction

Starch has many diverse applications in food production and other industries (Fishman, Coffin, Konstance, & Onwulata, 2000; Le Corre, Bras, & Dufresne, 2010). Chemically modified starches extend the range of physical properties available for various uses, because they exhibit excellent physicochemical properties that are markedly altered from those of their parent starches (Zdanowicz, Schmidt, & Szychaj, 2010; Zhang et al., 2007). Starches have been widely used in the adhesives industry (Kennedy, 1989), but their bonding capacity is insufficient for gluing wood. The graft copolymerization of synthetic polymers onto a starch backbone is among the best ways to improve the bonding properties of starch (Lei, Du, Wu, Xi, & Dong, 2014). Many reports describe the synthesis, characterization, and properties of starch graft copolymers (Bruyn, Sprong, Gaborieau, Rober, & Gilbert, 2007; Gong, Wang, & Tu, 2006; Lai, Don, Liu, & Chiu, 2006; Tanrattanakul & Chumeka, 2010). Nevertheless, these starch-based wood adhesives still lack the high bonding strength and water resistance. Furthermore, compared with the conventional adhesive materials, starch-based adhesives are typically too weak for practical use (Tanrattanakul & Chumeka, 2010).

The molecular structure of starch adhesives must be strengthened to develop high-performance wood adhesive.

Starch oxidation is an alternative method for improving starch properties that is widely used in industry. Oxidation is a chemical modification in which carboxyl and carbonyl functional groups can be introduced into the starch chains. At a suitable temperature and pH, starch can react with several oxidizing reagents (Veelaert, de Wit, Gotlieb, & Verhé, 1997; Kuakpetoon & Wang, 2001; Wing & Willett, 1997) to form oxidized starch. Oxidized starch prepared from hydrogen peroxide has been of particular research interest (El-Sheikh, Ramadan, & El-Shafie, 2010; Zhang, Zhang, Wang, & Wang, 2009). Hydroxyl groups, primarily at the C-2, C-3, and C-6 positions, are transformed into carbonyl or carboxyl groups via oxidation (Kuakpetoon & Wang, 2006; Kurakake, Akiyama, Hagiwara, & Komaki, 2009). However, the carbonyl and carboxyl groups exhibit high-activity, easily participating in various reactions, such as cross-linking (Yanbo, Chengfei, & Meina, 2009). Hydrogen peroxide decomposes into hydrogen ions and water, making it an environmental friendly oxidizing reagent. Therefore, in this research, the hydrogen peroxide was used as the oxidizing reagent.

The oxidized starch adhesive exhibited low bonding strength and water resistance preventing its use in practical applications. Therefore, in order to improve the performance of starch adhesives, sodium dodecyl sulfate (Li et al., 2014), urea (Wang, Gu, Li, Hong, & Cheng, 2013), silane coupling agent and olefin monomer

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were added to the oxidized starch to produce a graft-copolymerized wood adhesive (Bruyn et al., 2007; Gong et al., 2006; Lai et al., 2006; Tanrattanakul & Chumeka, 2010). The bonding strength and water resistance of the adhesive have improved significantly, confirming the positive effect of adding the silane coupling agent and olefin monomer to the adhesive system. The structure of adhesive was analyzed to determine the interaction between the grafted starch and the silane coupling agent. The thermal properties of adhesive and its viscoelasticity have confirmed that the quality of the graft copolymerized starch-based wood adhesive was improved.

At present, most studies showed that the bonding strength and water resistance of starch-based wood adhesive were usually too weak to practical use (Wang, Gu, Hong, Cheng, & Li, 2011; Bordes, Pollet, & Avérous, 2009). The purpose of the study was to investigate in the bonding strength and water resistance performance of starch-based wood adhesive prepared by grafting copolymerization and adding the silane coupling agent.

2. Materials and methods

2.1. Materials

Cornstarch was provided by Changchun Dacheng Corn Co. (China). The silane coupling agent ($\text{CH}_2=\text{CH}-\text{Si}(\text{OC}_2\text{H}_5)_3$, A-151) was obtained from Nanjing Forward Chemical Co. (China). The hydrogen peroxide (H_2O_2 , 30%), ferrous sulfate (FeSO_4), butyl acrylate ($\text{CH}_2=\text{CHCOO}(\text{CH}_2)_3\text{CH}_3$, BA), vinyl acetate ($\text{CH}_3\text{COOCH}=\text{CH}_2$, VAc), ammonium persulfate ($(\text{NH}_4)_2\text{S}_2\text{O}_8$, APS), polyvinyl alcohol (PVA), sodium dodecyl sulfate ($\text{CH}_3(\text{CH}_2)_{11}\text{OSO}_3\text{Na}$, SDS) and sodium hydroxide were purchased from Tianjin Kemiao Chemical Reagent Co. (China). All the other reagents were analytical grade.

2.2. Synthesis of graft-copolymerized starch-based wood adhesive

A graft-copolymerized starch-based wood adhesive was prepared as follows: 10 g dried corn starch and 100 mL distilled water were placed into a four-necked round-bottom flask and stirred at 80 °C for 30 min. After the starch gelatinization, the reaction temperature was cooled to 55 °C, following by the addition 0.1 g ferrous sulfate and H_2O_2 . Oxidization after 1 h, the temperature was again raised to 80 °C, and 3 g PVA was dropped into the mixture four-necked round-bottom flask. Stirred after 0.5 h, the mixture was cooled to 60 °C, and 0.1 g of SDS, 0.05 g of APS and silane coupling agent were added. Grafting after 0.5 h, 0.05 g APS, BA and VAc were added dropwise over 2 h (addition at constant speed of 0.2 mL/min), and the reaction continued for 1 h. Finally, the temperature was raised to 80 °C and kept for 30 min and then cooled to room temperature.

2.3. Shear adhesive strength test

The shear adhesive strength of the adhesive samples was tested according to the industry standard HG/T2727-1995 (China, 1995). Freshly cut 25 mm × 25 mm × 10 mm pieces of wood (*Betula platyphylla*, 0.62 × 103 kg/m) were glued with adhesives under a static pressure of 1 MPa at 25 °C for 24 h. The shear adhesive strength of the glued samples in the dry or wet (after immersing in water at 30 °C for 3 h) state was determined using a WDT-10 shear adhesive strength analyzer (KQL Co., China). The shear adhesive strength was calculated as follows: $\sigma M = F_{\text{max}}/A$ in which σM (MPa) is the shear adhesive strength, F_{max} (N) is the observed maximum failing load, and A (m^2) is the bonding surface of the sample. The testing speed was 5 mm/min. All the tests

were replicated 10 times, and the results were presented as the averages.

2.4. Intrinsic viscosity measurement

The intrinsic viscosity was measured using an NDJ-5S rotational viscometer (Ping Xuan Shanghai Scientific Instrument Co., China) with no. 4 rotor at 30 r/min and 30 °C. An extended period was required to obtain a stable reading. All the tests were replicated 3 times, and the results presented as the averages.

2.5. Fourier transform infrared (FTIR) spectroscopy

The adhesive samples were precipitated with ethanol, washed with distilled water, and dried to obtain the adhesive solids. The solids were extracted with acetone using a Soxhlet extraction device at 70 °C for 48 h to remove the homopolymers of the VAc and BA monomers and then dried. The extracted solids were fully milled with potassium bromide and compressed for FTIR analysis on a Nexus 6700 FTIR spectrometer (Nicolet Co., USA). Each sample was scanned 32 times over the 4000–400 cm^{-1} region at a resolution of 4 cm^{-1} .

2.6. Scanning electron microscopy (SEM)

The surfaces of cast films of the adhesive samples and the fractured surface of the sample under dry conditions and the fractured surface were coated with gold under vacuum. Then, all samples were observed under a scanning electron microscope (Quanta-200, Holland).

2.7. Thermogravimetric analysis (TGA)

The solid adhesive samples were extracted with acetone using a Soxhlet extraction device at 70 °C for 48 h to remove the homopolymers of the VAc and BA monomers and then was dried. The thermal stability of the solid samples was analyzed using a NETZSCH TGA209 thermogravimeter (NETZSCH Co., Germany). The samples (5 mg in a 70 μL alumina pan) were heated from 25 to 600 °C at a rate of 10 °C/min under a 30 mL/min nitrogen gas flow.

3. Results and discussion

Given the vast hydroxyl structure of the native starch, oxidized starch is hydrophilic and therefore cannot be used as a wood adhesive without modification. Consequently, the water resistance of starch adhesives must be improved. In this paper, a silane coupling agent and an olefin monomer were added to oxidize starch to produce a starch-based wood adhesive. The synthesis process is provided in Scheme 1. All oxidized starches were prepared after gelatinization, and then, the effects of the oxidant, catalyst, and monomer contents and the mole ratio of the monomer and coupling agent on the shear adhesive strength in the dry (DS) and wet states (WS) were investigated in detail.

3.1. Effect of oxidant content on shear adhesive strength

The native starch was oxidized using hydrogen peroxide; some of the hydroxyl groups in the starch were transformed to carbonyl or carboxyl groups via oxidation (Sangseethong, Termvejsayanon, & Sriroth, 2010), significantly improved the water resistance of the starch adhesive. Fig. 1a depicted the effect of the oxidant content on the shear adhesive strength. When the oxidant content was increased from 0 to 3.0 (by weight based on the solid content of adhesive), the shear strength in the DS increased from 3.28 to 7.30 MPa, and the shear strength WS increased from 1.40

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