International Journal of Biological Macromolecules 104 (2017) 846–853

Contents lists available at ScienceDirect

International Journal of Biological Macromolecules

journal homepage: www.elsevier.com/locate/ijbiomac

Effects of sucrose fatty acid esters on the stability and bonding performance of high amylose starch-based wood adhesive

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ARTICLE INFO

Article history:
Received 11 April 2017
Received in revised form 1 June 2017
Accepted 21 June 2017
Available online 23 June 2017

Keywords:
Starch
Wood adhesive
Bonding strength
Viscosity
Sucrose fatty acid ester

ABSTRACT

A facile approach was developed to dramatically boost the performance of high amylose starch-based wood adhesive (HASWA) by addition of sucrose fatty acid esters (SEs). The results showed that the addition of 6% (w/w, dry starch basis) SEs significantly improved the bonding strength in both dry and wet states as well as the mobility and storage stability of the adhesive. The formation of amylose-SEs complexes were proved by blue value or differential scanning calorimetry (DSC) analysis. The results also indicated that the incorporation of SEs into the HASWA hindered the aggregation of latex particles and enhanced the thermal stability of starch molecules. Furthermore, SEs addition in the adhesive system resulted in the inhibition of starch retrogradation as observed by time sweep and pulsed nuclear magnetic resonance (PNMR) analysis. These findings provide important information to prepare bio-based high performance wood adhesive by adding SEs to replace the traditional petro-chemical based stocks adhesives.

1. Introduction

The concept of wood adhesive has led to the utilization of wood composite products which are promising source for construction, furniture and building industries [1]. However, most adhesives are produced from petroleum base stocks that contain urea formaldehyde and/or phenol formaldehyde and volatile organic compounds [2]. These adhesives can create serious risks to human health and plummeting the indoor air quality by emitting these carcinogenic compounds into the environment during the production of wood composites. Moreover, these petrochemical based adhesives cannot be sustained in long term due to exhausting oil and natural gas reserves [3]. Increasing environmental and economical apprehension has induced various changes in adhesive industry in the last decades, giving more consideration to prepare bio-based adhesive systems. Therefore, it is inevitable to reinstate the petroleum based constituents in adhesive formulations. The application of renewable green natural material in adhesive system will help to produce formaldehyde-free wood adhesive in order to triumph over the above mentioned human health and environmental problems [4].

Starch, which is inexpensive and renewable natural polymer, is considered as one of the most promising candidate material to replace the synthetic polymers [5]. It is widely accepted that starches can be used in adhesive industry [6], but their bonding capacities are not sufficient to glue the constructional wood material [7]. Many strategies have been employed to improve the properties of starch-based wood adhesives by strengthening the molecular structure [8,9]. There is a considerable amount of literature in this field already [10,11] and these works normally described the preparation of starch-based wood adhesives by grafting vinyl acetate onto starch, using ammonium persulfate as an initiator. In this works, high amylose corn starch was used for the first time to prepare wood adhesive as previous reports showed that properties of corn starch are different with different amylose contents [12]. It is also reported that high amylose corn starch possess high water resistance and superior mechanical properties [13] which are essential characteristics for practical applications of wood adhesive.

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http://dx.doi.org/10.1016/j.ijbiomac.2017.06.090
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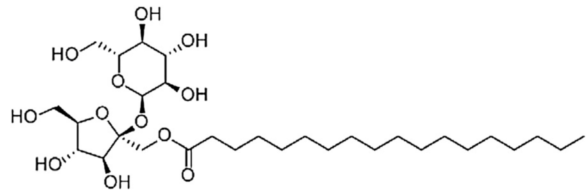


Fig. 1. Chemical structure of sucrose monostearate.

SEs is a non-ionic surfactant composed of hydroxyl groups which are hydrophilic and hydrophobic fatty alkalyl chains. This surfactant can be derived from renewable sources such as vegetable oils and carbohydrates. SEs is widely renowned for its excellent emulsifying, wetting, foaming, stabilizing, detergency and hard water resistance properties [14]. This surfactant is capturing research attention due to its characteristics such as non-toxic, biocompatible, odourless, non-cumulative and biodegradable [15]. SEs with high monoester content such as the one used in this study are more hydrophilic. The fatty acid bonded to sucrose molecule is stearic acid ($C_{18:0}$). Sucrose monoesters of stearic acid mainly consist of sucrose monoester of individual fatty acid namely stearic acid with smaller amounts of di- or tri-esters. The chemical structure of sucrose monostearate used in this study is shown in Fig. 1. Some studies have shown that surfactant could efficiently impede starch retrogradation by forming starch-surfactant complexes [16]. In this study, SEs was added before graft copolymerization of vinyl acetate onto starch to prepare HASWA. To the best of our knowledge, this is the first report in which SEs is used to improve the bonding performance of high amylose starch-based wood adhesive.

The goal of this work was to evaluate the effects of SEs on the performance of HASWA. The bonding strength and viscosity variations were evaluated. Dynamic viscoelastic measurement was studied to evaluate the gelation behavior of amylose. Furthermore, the characteristics as well as the thermal stability and distribution and mobility of water molecules in wood adhesive system were analyzed to confirm the quality improvements of wood adhesive with addition of SEs.

2. Experimental

2.1. Materials

Ordinary corn starch was provided by Wuhan Jinruicheng Co. (China). Citric acid monohydrate, tri-sodium citrate dihydrate, ammonium persulfate, urea and vinyl acetate (VAc) were of analytical grade and produced by Sinopharm Chemical Reagent Co., Ltd. Enzyme pullulanase (activity: 1000U/mL) was obtained from Yuan Ye Biological Co., Ltd., Shanghai, China. High amylose starch was obtained by enzymolysis (pullulanase) approach (detailed procedure for high amylose starch preparation is given in Section 2.2) having amylose content of 44.46% as determined by iodine calorimetric method of Garg and Jana [17] using absorbance at 620 nm. The content of amylose (44.46%) was in agreement with the previous reports [18]. SEs was obtained from Hangzhou Riling Co., Ltd (Table S1). SEs in this study is the admixture of different fatty acid esters consisting of 57% monoester, 28% di-ester and 10% tri-ester of a mixture of stearic and palmitic acid ester in a 70:30 proportion. The weight percent of monoester, diesters and polyesters was 56.7, 37.5 and 5.8, respectively [19]. Distill water was used all along preparation.

2.2. Synthesis of high amylose starch-based wood adhesive (HASWA) with SEs

About 15 g of normal corn starch was taken and dissolved in 100 mL of buffer solution (32 mL citric acid monohydrate, 68 mL tri-sodium citrate dehydrate) in three necked round-bottomed flask and stirred in boiling water for 5 min to gelatinize the starch slurry. After gelatinization, the reaction mixture was placed in a water bath and 45 μL of pullulanase enzyme (1000 ASPU/mL) was added to carry out the process of enzymolysis by constantly stirring the reaction mixture for 2 h at 50 °C. After enzymolysis, high amylose starch was obtained followed by the deactivation of enzyme by placing the reaction mixture in the boiling water for 5 min and the pH of the reaction mixture was adjusted to 4.5 with 0.5 mol/L hydrochloric acid. SEs (3–12% dry starch basis) was added to the reaction mixture and allowed to react for 15 min at 70 °C. Subsequently, 1 g of ammonium persulfate was added followed by 18 mL of vinly acetate over a period of 2 h under nitrogen protection. Afterward, the temperature of the reaction mixture was increased to 85 °C and kept for 30 min. Following this, the temperature was decreased to 50 °C, 10 mL of urea solution was added and the pH was adjusted to 4.8 using 0.5 mol/L NaOH. In the end, the graft-copolymerized high amylose starch-based wood adhesive was cooled to room temperature and stored in a plastic bottle.

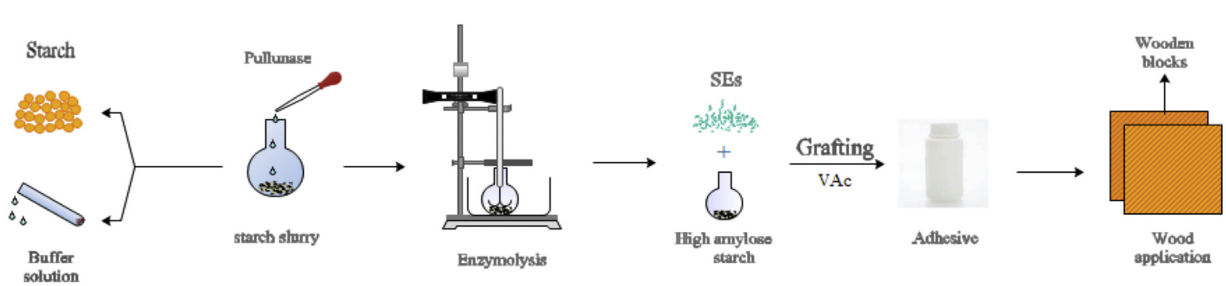
A conventional HASWA was synthesized following the aforementioned procedure but without adding SEs for comparison purpose. A schematic procedure is illustrated below in Scheme 1 to prepare HASWA with addition of SEs.

2.3. Intrinsic viscosity measurement

A rotational viscometer NDJ-8S (Ping Xuan Shanghai Scientific Instrument Co., China) was used to measure the intrinsic viscosities of the adhesive samples at 30 °C and 30 rpm using rotor No. 4. The samples were replicated three times and results were presented as averages.

2.4. Shear adhesive strength test

The strength test of the adhesive samples was performed following the procedure of Wang et al. [10] All the tests were performed in triplicate and the results were reproducible within deviation of ± 2.5 MPa.



Scheme 1. Schematic illustration of preparation of high amylose starch-based wood adhesive.