Industrial Crops and Products 51 (2013) 267–273

Contents lists available at ScienceDirect

Industrial Crops and Products

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

Optimization of preparation conditions of soy flour adhesive for plywood by response surface methodology

Nairong Chen, Qiaojia Lin*, Qinzhi Zeng, Jiuping Rao

College of Material Engineering, Fujian Agriculture and Forestry University, Fuzhou 35002, Fujian, China

ARTICLE INFO

Article history:
Received 30 April 2013
Received in revised form 12 August 2013
Accepted 14 September 2013

Keywords:
Soy flour adhesive
Boiling water resistance
Preparation conditions
Optimum
Response surface methodology

ABSTRACT

Response surface methodology was applied to optimize the preparation conditions of soy flour adhesive (SFA). A boiling water test was used to assess the wet strength of plywood bound with this adhesive. The effects and interactions of treatment time (X_1), treatment temperature (X_2) and additive amount of acid–salt solution (X_3) on wet strength were investigated. The regression model for the SFA preparation was significant ($p < 0.0001$). X_2 and X_3 had a significant effect on wet strength, whereas the effect of X_1 was insignificant. X_2 and X_3 showed interactions with wet strength. The optimal preparation conditions were: acid–salt solution 3.0 g, treatment time 33 min and treatment temperature 29 °C. Under these conditions, the wet strength was 1.18 ± 0.08 MPa, which is in agreement with the predicted value (1.11 MPa). An analysis of the Fourier transform infra-red spectroscopy spectra of the SFAs further confirmed the validity of the optimal preparation conditions.

1. Introduction

Adhesives prepared from biomass have been used widely in history (Frihart, 2005). In the 1970s, this type of adhesive was substituted by petro-based adhesives (e.g. phenolic resins) because of its poor mechanical properties (e.g. wet strength). Petro-based adhesives have good mechanical properties, but their raw materials such as phenol and formaldehyde are derived from nonrenewable fossil oil. Besides, toxic raw materials released from these petro-based adhesives are harmful to producers and customers. Hence, in recent years, adhesives based on biomass have attracted considerable attention as a promising alternative to petro-based adhesives.

Biomass such as starch (Karabulut et al., 2012; Wang et al., 2012), vegetable protein (Lambuth, 2003), vegetable oil (Ratna and Banthia, 2000), pectin (Wattanakorn et al., 2010), lignin (Navarrete et al., 2012) and tannins (Ping et al., 2012) could be used as adhesive raw materials. Amongst these, vegetable protein is desirable as it contains a large number of functional groups such as primary amines and carboxyl and hydroxyl groups. The main reason for soybean being the preferred adhesive raw material is not only its higher soy protein content but also because it is affordable and readily available. However, adhesives prepared from unmodified soy protein have restricted use because of their poor water resistance (Chen et al., 2012; Li et al., 2009). Therefore, interest has increased in modifying soy proteins with chemical agents, resins, enzymes, etc., to prepare a water resistant soy-based adhesive (Amaral-Labat et al., 2008; Gu and Li, 2011; Liu et al., 2010; Sun and Bian, 1999). Plywood bonded with soy-based adhesives such as soy flour treated with a polyethylenimine epichlorohydrin resin (Li et al., 2004), melamine urea formaldehyde resin (Fan et al., 2011) and polyacrylic acid solution (MPA) and sodium dodecyl sulfate (Gao et al., 2011) have improved the wet strength as well as the warm water resistance significantly. However, little information is available regarding the resistance of soy-based adhesives to boiling water. Moreover, as in most preparation processes, the properties of the soy-based adhesive involve balancing the preparation conditions to obtain the desired output characteristics. Studies have confirmed that the treatment conditions of soy protein including the additive amount of modifier, treatment time and treatment temperature affect the soy-based adhesive properties significantly (Liu et al., 2010; Xu et al., 2012; Zhang et al., 2013). Our previous work indicated that the treatment of defatted soy flour with a combination of acid, salt and alkali improved the water-resistance of the defatted soy flour-based adhesive (Lin et al., 2012). However, the preparation conditions of this adhesive have not been studied further.

Response surface methodology (RSM), which is an effective experimental design methodology, explores the relationship between several explanatory variable parameters and one or more response variable parameters by means of a mathematical model capable of predicting the values of the response variables (Sahu et al., 2010; Zhong and Wang, 2010). The main advantage of this methodology is the reduced number of experimental trials required to evaluate multiple parameters and their interactions.

* Corresponding author. Tel.: +86 0591 83715175; fax: +86 0591 83715175.
E-mail addresses: linqj208@163.com, commensurate@126.com (Q. Lin).

0926-6690/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.indcrop.2013.09.031

Table 1
Levels of parameter variables used in RSM design.

Coded and un-coded variables	Levels				
Coded-variables (Z_i)	−1.682	−1	0	1	+1.682
Treatment time (X_1, min)	4.77	15.00	30.00	45.00	55.23
Treatment temperature (X_2, °C)	18.2	25.0	35.0	45.0	51.8
Additive amount of acid–salt solution (X_3, g)	0.48	1.50	3.00	4.50	5.52

RSM has been used widely in scientific research, such as on cellulose nanocrystals (Tang et al., 2011; Wang et al., 2010), activated carbons (Ahmad and Alrozi, 2010; Sahu et al., 2010) and wood-based panels (Islam et al., 2012; Li et al., 2009). To the authors' knowledge, no study has been conducted on the optimization of the preparation conditions of soy flour adhesive (SFA) using the RSM approach.

In this study, a SFA treated by a combination of acid and salt was prepared and this adhesive was then used to make plywood. A standard RSM design called a central composite design (CCD) was used to optimize the preparation conditions, including additive amount of acid–salt solution, treatment time and treatment temperature, with the purpose of obtaining optimal preparation conditions for preparing a boiling-water resistant SFA.

2. Materials and methods

2.1. Materials

All chemicals were of analytical grade and were purchased from Sinopharm Chemical Reagent Beijing Co., Ltd. (China). A solution (acid–salt solution) containing both salt and acid was prepared by adding 1% of calcium chloride into hydrochloric acid solution (37%). Defatted soy flour (DSF) containing 53.4% (dry basis) protein was purchased from Shandong Wonderful Industrial Group Co., Ltd. (China). The 98% DSF had been screened through a 200 mesh screen, according to the supplier. *Poplar* veneer 300 mm × 300 mm in size, 1.2–1.3 mm thick and with a moisture content of 10–12 (wt.%) was supplied by Vicwood Industry Co., Ltd. (Suzhou, China). The diglycidyl ether of bisphenol-A (DGEBA) epoxy resin (E-51) was purchased from the China Petroleum and Chemical Corporation.

2.2. SFA preparation

SFA preparation was according to our previous work (Lin et al., 2012), with minor modifications. DSF (20 g) and distilled water (80 g) were added to a three-necked flask and stirred for 40 min at a 35 °C water bath. After the different masses of acid–salt solutions had been added into the DSF slurry, it was stirred for varying times at different temperatures (Tables 1 and 2), to obtain a modified DSF slurry. The modified DSF slurry was adjusted to pH 11 with sodium hydroxide solution (30%) at room temperature and finally 10% (w/w) of the DGEBA was blended with the slurry and stirred for 10 min. The obtained SFA was stored in a refrigerator.

2.3. Preparation of plywood

All obtained SFA was used to prepare duplicate samples of 3-ply wood by coating 140 g/m^2 of the adhesive on each veneer layer. The assembly time, pressing temperature, pressure and time were set at 10 min, 160 °C, 1.0 MPa and 3.6 min, respectively. Average results are presented from the duplicates.

2.4. Mechanical plywood properties

The plywood wet strength was determined by following exactly the conditions and methods described by the Chinese National Standards GB/T 9846-2004. A piece of plywood was cut into ten 100 mm × 25 mm specimens. The boiling water resistance and bonding strength of the developed SFA was evaluated through the glue line wet strength of the plywood samples subjected to boiling-water soaking pretreatment. Under these conditions, the plywood specimens were soaked in boiling water for 4 h and oven-dried at 63 ± 3 °C for 20 h, boiled again for 4 h and cooled at room temperature for 10 min. The wet strength was conducted by a tensile testing machine (MTS, USA) at a crosshead speed of 10 mm/min.

Table 2
Experimental scheme and results.

Run number	Treatment time (X_1, min)	Treatment temperature (X_2, °C)	Additive amount of acid–salt solution (X_3, g)	Wet strength (Y, MPa)
1	−1 (15.00)	−1 (25.0)	−1 (1.50)	0.57
2	1 (45.00)	−1	−1	0.73
3	−1	1 (45.0)	−1	0.82
4	1	1	−1	0.80
5	−1	−1	1 (4.50)	0.89
6	1	−1	1	0.90
7	−1	1	1	0.00
8	1	1	1	0.00
9	−1.682 (4.77)	0 (35.0)	0 (3.00)	0.94
10	1.682 (55.23)	0	0	0.86
11	0 (35.00)	−1.682 (18.2)	0	0.99
12	0	1.682 (51.8)	0	0.00
13	0	0	−1.682 (0.48)	0.72
14	0	0	1.682 (5.52)	0.00
15	0	0	0	1.06
16	0	0	0	1.09
17	0	0	0	0.97
18	0	0	0	1.11
19	0	0	0	0.98
20	0	0	0	1.00