# DEVELOPMENT OF WASHED COTTONSEED MEAL-BASED WOOD ADHESIVES FOR INDUSTRIAL APPLICATION Zhongqi He USDA-ARS, Southern Regional Research Center New Orleans, LA

### Abstract

Plant seed meal-based products can be used as green wood adhesives. We have tested the bonding performance of defatted cottonseed meal (CSM), water-washed cottonseed meal (WCSM), and cottonseed protein isolate (CSPI). CSM was cheap but showed poor water resistance. CSPI showed better adhesive performance, but it was expensive. WCSM, a new product we developed, possessed the adhesive strength comparable to CSPI, but it is more cost-efficient and environment-friendly than CSPI. To promote it as an industrial wood adhesives, WCSM was produced in a pilot-scale. The product was characterized in regards to chemical composition, physical properties and bonding strength in order to establish the quality standards in industrial production. Practical application of the pilot-produced WCSM product was explored. Per the experimental data, WCSM-based adhesives could be used as low-temperature glues for the domestic small utensils bonding (such as in pencil making), high-temperature adhesives for interior non-structural plywood making, and greener blending adhesive with urea formaldehyde resin for wood based composites production.

# **Introduction**

Defatted cottonseed meal-based adhesives have great potential as sustainable eco-friendly adhesives (He, 2017) as both the meal and its protein isolate both showed wood bonding capability (Cheng et al., 2013; He and Cheng, 2017a). However, the water resistance of cottonseed meal-based adhesives is poor, and the protein isolate is expensive in preparation. Thus, we (He et al., 2014a) developed a novel meal product by water washing of the defatted cottonseed meal. The new product, washed cottonseed meal (WCSM), showed the adhesive performance comparable to cottonseed protein isolate (He et al., 2016a; He et al., 2014b). For promoting the practical application, WCSM has been characterized and further produced in a pilot scale (He et al., 2015; He et al., 2016b), and the effects of several operational parameters on the bonding performance have been evaluated (He et al., 2017; Li et al., 2017). This work reported the adhesive performance of WCSM alone, or its mixtures with synthetic resins. The information presented in this work would be helpful in formulating industrially applicable WCSM-based wood adhesives.

#### **Materials and Methods**

WCSM was prepared from mill-produced cottonseed meal in a pilot scale, and its basic properties were reproted in an earlier paper as described previously (He et al., 2016b). The pilot-produced WCSM was ground by a hammer mill (Model W-6-H, Schutte Buffalo Hammermill, Buffalo, NY, US), passed through a 0.5-mm screen. Wood bonding and testing was performed in reference to relevant industrial standards. WSCM was first mixed with water and applied to wood substrate by a brush. Each thin board was divided in two parts and, after spreading the adhesive on both parts, they were overlapped and put under the press. If needed, synthetic poly(vinyl acetate) (Vinavil 2259 L) or urea formaldehyde (UF) resins were mixed with WCSM to produce hybrid adhesives to examine the blending effects of WCSM. The lap-shear tensile strength of these preconditioned wood specimens was measured with a Materials Tester. The tensile shear strength at break (MPa) was reported as the adhesive strength of the tested WCSM products.

# **Results and Discussion**

# WCSM as non-structural interior bonding glues

For exploration of the practical application of the WCSM as wood adhesive, we have collaborated with an industrial partner in Italy to test WCSM for non-structural interior bonding (such as in furniture making) in comparison with their synthetic glue poly(vinyl acetate) (Vinavil 2259 L) (He and Chiozza, 2017). In this experiment, we conducted four types of adhesive strength measurements per European Union standards (Fig. 1). These data indicated that the application rate (adhesive content) in the testing range was not critical as press temperature in determining the bonding strength of WCSM. The dry shear strength of WCSM was 10.7 and 9.3 MPa, respectively, with the application rated of 150 g m<sup>-2</sup> (WCSM 1) and 250 g m<sup>-2</sup> (WCSM 2). The average of 10 MPa was lower than that of Vinavil 2259 L, but met the requirement of EU class D1 adhesives. However, both values of the wet (0.4 MPa) and soaked (1.2 MPa) strength of WCSM were well below the EU requirements of D2 and D3 adhesives. The hot

strength of WCSM was around 9.0 MPa, just about 10% decrease in average, compared to the dry adhesive strength. Indeed, the hot strength of WCSM was greater than synthetic Vinavil 2259 L and met the heat resistance (8 MPa) of EU standard. In summary, these data suggested that WCSM meets the criteria of class D1 wood adhesives used for interior non-structural purpose. The heat resistance data also meet the criteria of class D3, but the water resistance data failed as D3 adhesive used for protected outside non-structural purpose.

Testing	Testing	Adhesive strength (MPa)		
type	conditions	Vinavil	WCSM1	WCSM 2
D1,10 MPa	Dry Strength	15.8	10.7 (good)	9.3 (marginal)
D2, 8 MPa	Soaked Strength	10.8	1.2 (poor)	_ a
D3, 2 MPa	Wet Strength	4.8	0.5 (poor)	0.3 (poor)
Heat resis-	Hot strength	7.0	8.9 (excellent)	9.1 (excellent)
tant 8 MPa				

Fig. 1. Adhesive strength of WCSM adhesives with the application rates of 150 g m<sup>-2</sup> (WCSM 1) and 250 g m-2 (WCSM 2) per European Union standards EN204, EN205 and EN14257.



Fig. 2. Adhesive strength of WCSM-based adhesives at 40 °C under the pressing pressure of 1.0 MPa

With an industrial partner's input, we further tested WCSM for a specific application in pencil making. In this work, eight WCSM-based adhesives were formulated to bond pine wood pencil sandwich (Fig. 2). The solid contents ranged from 27% to 49%. These data indicated all formulations possessed certain bonding ability. The general trend was that the higher the press time, the stronger of the adhesive strength. Specifically, Formulation A and D show the highest adhesive strength, and Formulation B shows weak bonding strength with 60 and 120 min pressing time. The adhesive strength values of Formulation E, F, G and H was just between those of the first four formulations. Per the result, four formulations, A, B, C and D, were applied to bond the "real" pencil sandwich slates. These bonded sandwiches were sent back to make eco-friendly pencils (Fig. 3). Those pencils were then subject to the industrial Temperature Cycle Test (Table 1). For the Temperature Cycle test, all pencil samples were submitted at the following cycle of 24 h at 25°C, 24 h at -5 °C, and 24 h at 45 °C. This cycle was repeated 4 times. Two pencils were tested for each conditions. The data in Table 2 show that all pencils made from the slates bonding with formulations A-D for 120 min passed through the industrial Temperature Cycle Test. However, only half of those pencils made with 60 min passed the test, indicating that long bonding time (120 min) is needed for making the pencils passed T Cycle test.



Fig. 3. Application of WCSM-based adhesive to bond pencil slat sandwich to make pencils.

Table 1. Pencil slat sandwich glue conditions and subsequent results of the industrial temperature cycle (T cycle)

test of the pencils made by these sandwiches						
Testing #	Adhesive	Pressure (MPa)	Time (min)	T cycle (2 pencils)		
1	A (30%)	1	120	ok, ok ‡		
2	B (39%)	1	120	ok, ok		
3	C (40%)	1	120	ok, ok		
4	D (49%)	1	120	ok, ok		
5	A (30%)	1	60	ko, ok		
6	B (39%)	1	60	ko, ok		
7	C (40%)	1	60	ko, ok		
8	D (49%)	1	60	ko, ok		

†. ok=good pencils, ko=open pencils.

# **Blending WCSM with synthetic resins**

We first tested the bonding performance of the blend of WCSM and Vinavil 2259 L (Fig. 4). With either the hot (130 °C) or cold (23 °C) press temperature, the blend of WCSM and Vinavil 2259 L showed similar adhesive performance as the Vinavil 2259 L and its water diluted solution. These data also showed that, unlike the pure WCSM adhesives with failed bonding with cold pressing (He and Cheng, 2017a; He and Cheng, 2017b), the blend did not show negative impacts on the adhesive performance of Vianvil 2259 L. These data further suggested that, whereas blending did not improve the adhesive strength of the synthetic Vianvil 2259 L, WCSM could be mixed with a poly(vinyl acetate) resin to tailor adhesive formulations for optimizing adhesive and operational conditions (such as, used in room temperature or appropriate solid content of adhesives).



Fig. 4. Effect of press temperature on the adhesive strength of synthetic glue Vinavil 2259 L (Vinavil ) and its blends with WCSM (Vinavil/WCSM) and water (Vinavil/water) in the ratio of 100:10.



Fig. 5. Dry (a) and soaked (b) strength of UF resin, WCSM, and their blends (200 psi= 1.38 MPa).

We further tested the hybrid adhesives composed of WCSM and urea formaldehyde (UF) resin with different ratios (Liu et al., 2017). The dry adhesive strength of WCSM and UF was 480 and 550 psi, respectively (Fig. 5). For the hybrid adhesives, the dry shear strength was about 450, 460, 520, 5301, and 410 psi for 10%, 20%, 30%, 40%, and 50% of WCSM, respectively. These values indicated that the adhesive strength of these hybrid adhesives was at the same level of those of WCSM and UF. On the other hand, the soaked strength of UF and WCSM and WCSM was quite low with the value around 200 psi. Blending the two together with the ratios of 10, 20, 30 and 40% of WCSM raised the soaked strength to >400 psi, indicating that blending was an effective approach to improve the water resistance of UF-alone or WCSM-alone based adhesives. In addition, replacement of UF resins with WCSM could reduce the formaldehyde emission from UF resins, leading to a more environmentally friendly feature of the final wood based composites with hybrid adhesives. In other words, the mixtures of UF and WCSM would be greener wood adhesives than UF resins.

#### <u>Summary</u>

Washed cottonseed meal may be used as the conventional D1 (interior) wood adhesives for nonstructural application. Four WCSM-based adhesive formulations were designed for bonding pencil sandwich slats. With appropriate bonding conditions, the pencil made from those bonded sandwiches passed the industrial temperature cycle test. Washed cottonseed meal can also be blended with synthetic resins to form hybrid adhesives. The blending can be used for improving either the operational conditions (such as polyvinyl acetate) or water resistance and emission reduction (such as urea formaldehyde).

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