



## Gum dispersions as environmentally friendly wood adhesives



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### ABSTRACT

Today, wood adhesives are mainly prepared from petroleum-based polymers. There is an ambition to decrease the utilization of petroleum-based raw materials and introduce bio-based polymers instead. However, the utilization of bio-based polymers is often limited due to insufficient properties in terms of water resistance or heat resistance. In this study bio-based dispersions have been prepared of locust bean gum, guar gum, xanthan gum and tamarind gum and evaluated as wood adhesives. Due to the high viscosity of the dispersions, a low dry solids content of 6 wt% was used. The film forming properties have been investigated and contact-angle measurement have been performed to obtain an indication of water resistance. Wood substrates have been bonded together and the bonding performance has been evaluated with different techniques. The gum dispersions have been compared with a commercial poly(vinyl acetate)-based wood adhesive and the results demonstrate that gums can be used as binders for wood adhesives. Locust bean gum dispersions show remarkable results – comparable to the commercial wood adhesive – even though the dry solids content is very low. The locust bean gum dispersion fulfills the D2 and WATT 91 requirements for wood adhesives according to the [European Standard EN 204](#) and [European Standard EN 14257](#).

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### 1. Introduction

Wood adhesives are today mainly produced from petroleum-based polymers; however, the fossil resource is both limited and non-renewable. Moreover, some of the adhesives have a questionable environmental impact. It is therefore desirable to introduce bio-based polymers in wood adhesives. Petroleum-based polymers are often superior in terms of bonding performance, water- and heat-resistance. To compete, the bio-based polymers therefore need to match the properties of conventional adhesives and/or introduce new valuable properties, preferably without any cost increase.

Protein, tannin, lignin, and polysaccharides are examples of interesting bio-based polymers that have been suggested for wood adhesives (Pizzi, 2006). Proteins from milk, blood and soybean have been used as wood adhesives for thousands of years. However, bio-based polymers were replaced by polymers derived from fossil resources due to enhanced properties and favorable cost development (Bye, 1990; Lambuth, 2003). Extensive research is now being

conducted to improve bonding performance and water resistance of soy protein with the intention of utilizing this raw material again (Hettiarachchy et al., 1995; Kalapathy et al., 1995; Kumar et al., 2002; Li et al., 2004; Sun, 2005). Wheat gluten has also been investigated as binder for wood adhesives (D'Amico et al., 2013; El-Wakil et al., 2007; Khosravi et al., 2010, 2011; Lei et al., 2010; Nordqvist et al., 2010, 2012a,b, 2013).

Polysaccharides are another interesting group of polymers with potential as wood adhesives. Polysaccharides are a large group of bio-based polymers and are built up by monosaccharides, joined together by glycosidic bonds, with mainly hydroxyl functional groups. A high molar mass of the polysaccharide will allow cohesive strength to the adhesive. The polar hydroxyl groups contribute with hydrogen bonding, allowing for a strong adhesion to wood. However, the polar hydroxyl groups will also make the polysaccharide hydrophilic, which has a negative impact on the water resistance.

Starch is a polysaccharide which has been investigated as a potential binder for wood adhesives. However, native starch does not have sufficient properties, and modifications are necessary to obtain the required properties. For example, starch and polyvinyl alcohol have been crosslinked with hexa(methoxymethyl)melamine, yielding an adhesive with promising results (Imam et al., 2001, 1999). Starch combined with tannins has resulted in a wood adhesive with properties

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comparable to commercial wood adhesives, interesting for ply wood applications (Moubarik et al., 2010a,b). Starch has also been evaluated as an adhesive for particleboards (Tondi et al., 2012). Furthermore, starch has been grafted with vinyl acetate, using ammonium persulfate as initiator (Wang et al., 2012; Wu et al., 2009) and further blended with silica nanoparticles (Wang et al., 2011) to improve the adhesive properties. Wheat flour, containing starch and proteins, has also been studied as a wood adhesive (D'Amico et al., 2010). Konjak glucomannan and chitin are other polysaccharides that have been evaluated as wood adhesives with successful results. Konjak glucomannan showed good dry bond strength and chitosan displayed good water resistance (Umehura et al., 2003).

Natural gums also belong to the group of polysaccharides. They can be obtained from several sources; in this study seed gums (locust bean gum, guar gum and tamarind gum) and an extracellular microbial polysaccharide (xanthan gum) have been used. Natural gums are used in the food industry and in the pharmaceutical industry, mainly as thickeners, colloidal stabilizers, and flow controllers. Furthermore, they are used in several types of adhesives such as pressure-sensitive tape, denture and medicinal adhesives, paper adhesives, pharmaceutical tablet binders, and label pastes (Conner, 1989). The research concerning wood adhesives based on seed gums are minor.

Extracellular microbial polysaccharides that are derived from bacteria have earlier been investigated as wood adhesives with successful results with dry bond strengths comparable to a poly(vinyl acetate)-based adhesive (Haag et al., 2004, 2005).

Locust bean gum is a galactomannan with a main chain of mannose units with galactose side chains on approximately every fourth mannose unit, Fig. 1a. It has a high molar mass of approximately 310 kg/mol. Guar gum is also a galactomannan with a galactose side chain on approximately every second mannose unit, Fig. 1a. Guar gum has a lower molar mass compared to locust bean gum, approximately 220 kg/mol. Xanthan gum is an anionic polysaccharide with a very high molar mass, above 2000 kg/mol, but can be as high as 13,000–50,000 kg/mol. Its main chain consists of glucose units with alternating mannose and glucuronic acids. Most of the inner mannose residues are acetylated and approximately half of the terminal mannose units are pyruvated, Fig. 1b. Tamarind gum is a xyloglucan with a main chain based on glucose units with side chains of xylose units, some equipped with galactose units, Fig. 1c.

The aim of this study is to evaluate adhesive properties of different gums. A commercial poly(vinyl acetate)-based wood adhesive was used to benchmark the experimental results obtained for the dispersions, even though it is based on a latex system.

## 2. Experimental

### 2.1. Materials

Locust bean gum from *Ceratonia siliqua* seeds, xanthan gum from *Xanthomonas campestris* and guar gum were purchased from Sigma–Aldrich. Tamarind gum from Innovassynth containing ~85 wt% xyloglucan and deoiled tamarind gum from Premcem Gums containing ~40 wt% xyloglucan were kindly supplied by Cel-lutech AB, Sweden. Brown liquor was kindly supplied by Domsjö Fabriker AB, Sweden. The brown liquor contains mainly lignin, sugars, polymeric sugars as well as trace elements such as manganese, calcium and iron. Cascol® 3304, a poly(vinyl acetate)-based wood adhesive was kindly supplied by Akzo Nobel and used as a reference. Cascol is a thermoplastic wood adhesive classified as a D2 adhesive according to standard EN 204 (EN204, 2001). Proxel XL-2 (biocide) was obtained from Arch UK and Rocima 520S

(fungicide) from Dow. Particleboards (15 cm × 15 cm), veneers and beech wood panels were kindly supplied by Akzo Nobel, Casco Adhesives AB.

### 2.2. Preparation of dispersions of gums

Locust bean gum, guar gum, xanthan gum, tamarind gum from Innovassynth and Premcem (24 g) were dispersed in de-ionized water (380 ml) while heated at 60 °C in an oil bath for 6 h and stirred (150–200 rpm) using an overhead stirrer, resulting in 6 wt% dispersions.

Tamarind gum from Premcem was further used to prepare an 11 wt% dispersion by adding 48 g gum in de-ionized water (380 ml) using the procedure described above.

Locust bean gum (24 g) was also dispersed in brown liquor (380 ml) using the same procedure as described above.

All dispersions were allowed to cool to room temperature and then 0.15% (w/w) solutions of both Proxel XL-2 (biocide) and Rocima 520S (fungicide) were added to prevent mold growth.

### 2.3. pH measurement of the gum dispersions

pH was measured with a pH glass electrode (JENWAY) connected to a pH meter (3510 pH meter JENWAY) 24 h after the preparation.

### 2.4. Viscosity measurements of the gum dispersions

The viscosity was measured 24 h after the preparation with a strain-controlled dynamic frequency sweep. The measurements were performed in a TA AR2000 rheometer using 25 mm parallel plates. The frequency was set from 1 rad/s to 500 rad/s using 25% strain. All measurements were conducted in triplicates.

### 2.5. Film formation

The dispersions were used to prepare films on glass substrates using a 700 µm applicator 24 h after dispersion preparation. The films were allowed to dry for 10 min at 120 °C and further conditioned at 23 °C and 50% relative humidity for 24 h prior to further analysis.

### 2.6. Contact-angle measurement

To study the hydrophobicity and obtain an indication of the water resistance the films were subjected to contact-angle measurements. The contact-angle measurements were performed on a KSV instrument CAM200 equipped with a Basler A602f camera, using 5 µL droplets of deionized water. Images were recorded immediately after applying the water droplet and after 40 s. Results are averages of five measurements. The water contact angles were determined using the CAM software.

### 2.7. Specimen preparation

#### 2.7.1. Application Method 1: particleboards bonded with veneers

Particleboards and veneers were conditioned at 23 °C and 50% relative humidity for at least seven days. The 24 h old dispersion was used to bond a particleboard (15 cm × 15 cm) with veneer. The adhesive was applied on the particleboard (360 g adhesive/m<sup>2</sup>). The specimens were hot-pressed with a Fontijne Grotnes Lab Pro 400 at 120 °C and 0.9 MPa, for 5 or 2.5 min.

#### 2.7.2. Application Method 2: veneers

The beech veneers were conditioned at 20 °C and 65% relative humidity for at least seven days. Veneer strips (0.6 mm in

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