Bio-based adhesive from wheat gluten

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Abstract

Nowadays, most wood adhesives are petroleum-based derived from the limited non-renewable resource such as formaldehyde-based resins. In manufacturing and end-use of these synthetic adhesives, low molar mass toxic compounds are largely emitted. Negative effects on the human health and environment have renewed an interest in more environmental friendly adhesives. Wheat gluten (WG), an industrial by-product from wheat starch production, was of interest to develop as a bio-based adhesive because of its capacity to form a cohesive and viscoelastic mass. In this research, WG powder was dispersed in an alkaline solution to prepare the WG-based adhesive. Various adhesive formulas with different WG contents (20, 25 and 30 wt%) and concentrations of the NaOH solution used (0.05, 0.10 and 0.20 M) were formulated. The viscosity of the resulting adhesives was evaluated using Brookfield viscometer. The adhesives were applied on wood panels and then heated at 130°C for 15 minutes under pressure of 0.7 MPa to fix the adhesive bond. Tensile shear strength of the bonded wood panels was determined according to the British Standard EN 205:2003. It was found that both WG content and concentration of NaOH solution used to prepare the adhesives significantly affected their viscosity and bond strength of the wood joints. The best adhesive formula was prepared with 25 wt% WG and 0.20 M NaOH which provided the optimum bond line thickness and the highest bond strength of approximately 7.07 MPa. The adhesive bond line between the wood panels could be indicated by both optical and scanning electron microscopy techniques.

Keywords: wheat gluten, bio-based adhesive, viscosity, tensile shear strength, microscopy

Introduction

Wood bonding is considered to be one of the largest markets for adhesives (Nordqvist 2012). Currently, most wood adhesives are petroleum-based such as formaldehyde-based resin (urea-formaldehyde, phenol-formaldehyde, resorcinol-formaldehyde, and melamine-formaldehyde), polyurethane and epoxy (Imam et al. 2001). The advantages of these synthetic adhesives are their wide availability in different types, good performance, excellent durability, and moderate resistance to heat and/or moisture. However, the production and use of petroleum-based adhesives have presented a negative impact on the human health and environment. They are mainly derived from the limited non-renewable fossil source and emit low molar mass toxic compounds during product manufacturing, distribution, and end-use. The increasing environmental concern has initiated an interest in more environmental

friendly adhesives based on biopolymers derived from renewable resources (Nordqvist et al., 2013). Wheat gluten (WG) is an inexpensive industrial by-product from wheat starch processing. It is mainly composed of two protein types: gliadins and glutenins. Gliadins have globular shape and contribute to its extensibility and viscosity, while glutenins have linear structure and conduce to its elasticity and strength (Nordqvist 2012). Since WG has a cohesive behavior to form the viscoelastic mass, it has a high potential to be developed and used as a wood adhesive. The previous studies have shown that several preparing methods e.g. enzymatic hydrolysis, heat treatment, and denaturation using alkaline resulted in the high-performance WG adhesives (El-Wakil et al. 2007; D'Amico et al. 2010; Leia et al. 2010; Khosravi et al. 2011; Nordqvist et al. 2012; Ding et al. 2013). It seemed that WG content and degree of protein denaturation by alkaline strongly influenced the adhesive properties. However, only limited data on the effects of these two factors has been reported. Previously, the WG adhesives were prepared with only 0.10 M alkaline solution and the maximum WG content attempted was 24 wt% (Nordqvist et al. 2010; Nordqvist et al. 2012; Nordqvist et al. 2013). The purpose of this research was, therefore, to prepare the bio-based adhesives from WG in alkaline condition and the two factors, i.e., WG content (20, 25 and 30 wt%) and NaOH concentration (0.05, 0.10 and 0.20 M) were varied. Their effects on viscosity of the adhesives as well as bond strength of the wood jointed by the adhesives were studied.

Methodology

Preparation of wheat gluten-based adhesive

To determine the optimum formula for bio-based adhesive, wheat gluten-based adhesive with various formulas were prepared as summarized in Table 1.

Firstly, NaOH solutions with concentrations according to Table 1 were prepared. Wheat gluten powder (Zhangjiagang Hengfeng Starch Products Co. Ltd., China) was then gradually added into the alkali solution while stirred at 300 rpm by a homogenizer (IKA RW20 digital) at room temperature for approximately 90 min. After that, the wheat gluten-based adhesive was obtained.

Table 1: Various formulas of the wheat gluten-based adhesives

Amount of wheat gluten (wt%)	NaOH concentration (M)	Adhesive Formula
20	0.10	20WG-0.10M NaOH
25	0.05	25WG-0.05M NaOH
25	0.10	25WG-0.10M NaOH
25	0.20	25WG-0.20M NaOH
30	0.10	30WG-0.10M NaOH

Viscosity measurement of wheat gluten-based adhesive

Viscosity of the wheat gluten-based adhesives was measured by using Brookfield Viscometer (model DV III). Spindle no. LV61, LV62, and LV63 were used. After starting the measurement, the viscosity was recorded when a constant value was reached.

Preparation of test pieces

The obtained wheat gluten-based adhesive was applied (180 g/m^2) on two teak wood panels with dimension of 120×160 mm^2. The teak wood panels were then assembled and pressed together by using a hot-press molding machine (Labtech LP-S-80) at temperature of 130°C and applied pressure of 0.7 MPa for 15 min.

The teak wood panels were bonded together according to the British Standard (BS) EN 204 (2001) and classified as a thin bond-line (adhesive layer of ≈ 0.1 mm thick). After pressing and before cutting, the panels were conditioned in a standard atmosphere (23 ± 2°C and 50 ± 5 % relative humidity for 7 days). After that, the panels were cut into four strips of width ***b*** = 20 ± 2 mm and length $\boldsymbol{l_1}$ = 150± 5 mm. The bonded sections on each test piece with width $\boldsymbol{l_2}$ = 10 ± 2 mm were then created in the middle section as shown in Figure 1. After that, the test pieces were again subjected to the conditioning in a standard atmosphere. At this step, the test pieces had to be kept in a horizontal plane and were not touched each other so that no stress was applied on them.

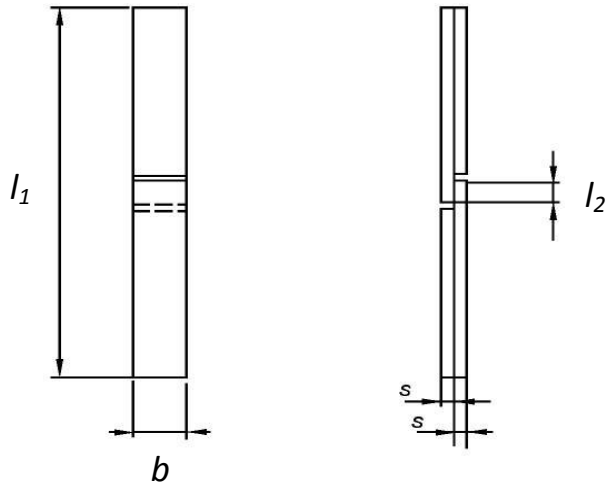

Figure 1: Lab joint bond-line image of the test piece (according to BS EN 204, 2001).

Tensile shear strength measurement

Tensile shear strength of the bonded panels was evaluated according to the BS EN 205:2003 (Wood adhesives for non-structural applications; determination of tensile shear strength of lap joints). The length and width of the test pieces were 150 mm and 20 mm respectively. It had bonded sections overlap in the middle section of 10 mm width (Figure 1). Eight test pieces were evaluated for each panel with the different wheat gluten-based adhesive formula.

The universal testing machine (UTM, Instron 5560) equipped with a 1 kN load cell was used for all such measurements. During the tests, it had to ensure that the force was applied centrally and in the plane of the bond. The test speed was set at 2 mm/min until rupture. The applied maximum force F_{max} in Newton (N) was recorded and used to calculate for the tensile shear strength (τ) in Newton per square millimeters (N/mm^2) using the following equation:

$$\tau = \frac{F_{\max}}{A} = \frac{F_{\max}}{l_2 \cdot b} \quad (1)$$

where F_{max} = the applied maximum force in Newton (N)
A = the bonded test surface in square millimeters (mm^2)
l_2 = the length of the bonded test surface in millimeters (mm)
b = the width of the bonded test surface in millimeters (mm).

Morphological observation

The wood panels were carefully cut and prepared for the bond line observation. Prior to the analysis, the cross section of the specimens was polished using Prius PRESI Power machine (MECAPOL P230) at speed of 300 rpm. Surface of the specimens was then cleaned with ethanol before the observation. Optical microscopy (OM) and scanning electron microscopy (SEM) were used to examine the bonded joint of the specimens. The observations were carried out using the optical microscope (OM) Zeiss/Axiotech and scanning electron microscope (SEM) LEO 1450 VP (with operating voltage of 10 kV). The specimens for SEM were mounted on an aluminum stub and coated with a thin layer of gold under vacuum before examining.

Results and Discussion

After the WG-based adhesives with different formulas were prepared, their viscosities were measured and summarized in Table 2. For the adhesives prepared with 0.10 M NaOH solution, with increasing the content of WG protein, the viscosity of the adhesive dispersions considerably increased. The higher amounts of added WG directly resulted in a larger number of the protein molecules presented in the adhesive dispersions. Therefore, a higher degree of molecular entanglement and a decrease in free volume for protein molecules to move were presumed as illustrated in Figure 2. This led to the higher viscosity of the adhesive dispersions with the higher contents of WG.

Table 2: Viscosity of wheat gluten-based adhesives with different formulas

Adhesive Formula	Spindle [a]	Speed (rpm)[b]	Viscosity(cP)
20WG-0.10M NaOH	LV1	60	97
25WG-0.05M NaOH	LV2	70	265

25WG-0.10M NaOH	LV2	70	392
25WG-0.20M NaOH	LV2	33	874
30WG-0.10M NaOH	LV3	19	6270

[a,b] Different spindles and speeds were used due to a large difference in the viscosity of the adhesives with different formulas.

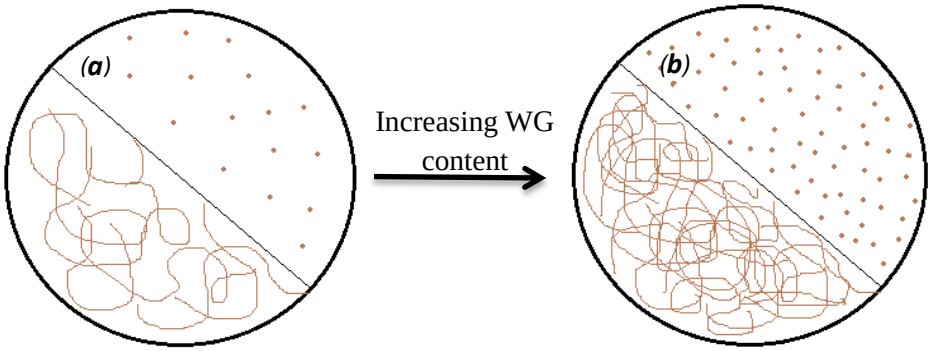

Figure 2: Molecular chain entanglement and free volume in WG-based adhesive dispersions with a) low and b) high WG contents.

For wood adhesive application, the viscosity of adhesive should not be too high or too low when applied on wood panel. This is because the viscosity of adhesive has influence on the thickness of bond line and ability of adhesive to penetrate into the wood surface. Both factors are subsequently determined the bond strength of jointed panels (Kumar et al. 2002; Nordqvist et al. 2013).

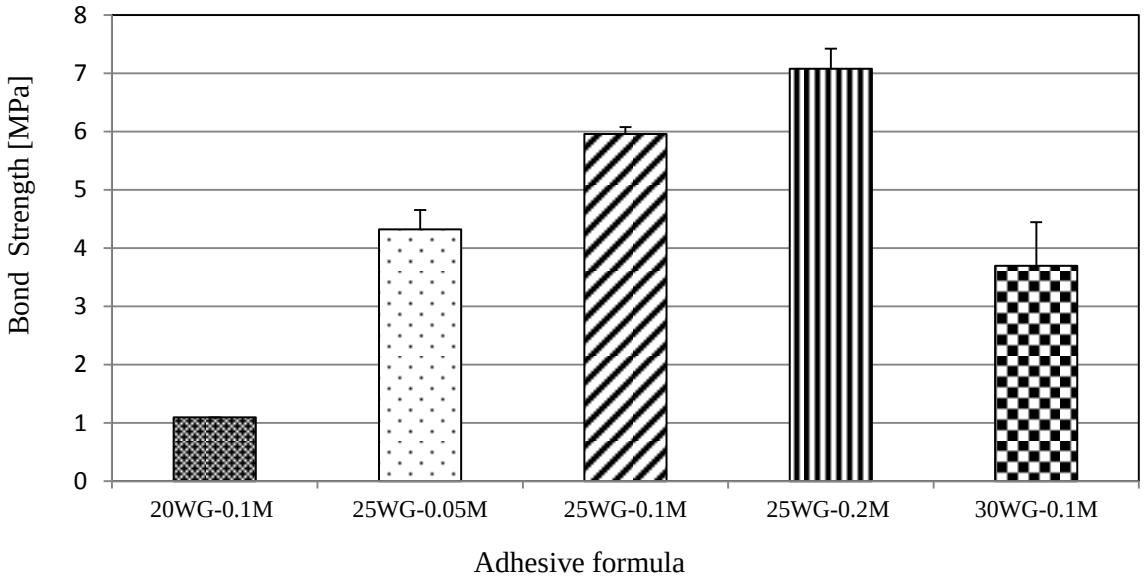

Figure 3: Tensile bond strength of the wood joints bonded with different formulas of wheat gluten-based adhesive.

The tensile bond strength of the joints bonded with different adhesive formulas is summarized in Figure 3. With increasing WG content in the adhesives from 20 wt% to 25 wt%, the bond strength of the wood joints markedly increased. At WG content of 20 wt%, the viscosity of the adhesive was low (Table 2) which possibly caused a very thin bond line in the wood joints and resulted in a weak bond. At higher WG content of 25 wt%, with the increased viscosity of the adhesive, this should allow a thicker bond line in the wood joints as well as a sufficient penetration of the adhesive into the wood surfaces. As a result, the bond strength of this wood joint was observed to considerably increase.

On the other hand, when WG content was increased from 25 wt% to 30 wt%, the bond strength of the wood joints was noticeably decreased. From the experimental observation, the thick bond line in this wood joint can be obtained. However, the very high viscosity of this formula led to a difficulty in adhesive penetration into the wood surfaces. Thus, a decrease in degree of mechanical bonding (interlocking) supposedly reduced the bond strength in this wood joint. The balance between the amount of adhesive in the bond line and in the woods is required for superior bond strength of the joints (Nordqvist et al. 2010).

In this work, the viscosity of the adhesive prepared with 25 wt% WG was considered to be the optimum since the highest bond strength of the wood joints was obtained in this adhesive formula as shown in Figure 3. The bond line of the wood joints bonded with the 25 wt% WG-based adhesive was examined by the two techniques; scanning electron microscopy and optical microscopy as shown in Figure 4a and b. The bond line between the wood panels can be noticed in the two photographs. However, the bond line thickness could not be estimated. A degree of the adhesive penetration into the wood panels also could not be observed. In the future work, some modifications such as staining the adhesive with indicative dyes or

particles prior to the microscopy analysis should be applied for a better indication of the bond line thickness as well as a degree of the adhesive penetration (Nordqvist et al. 2013).

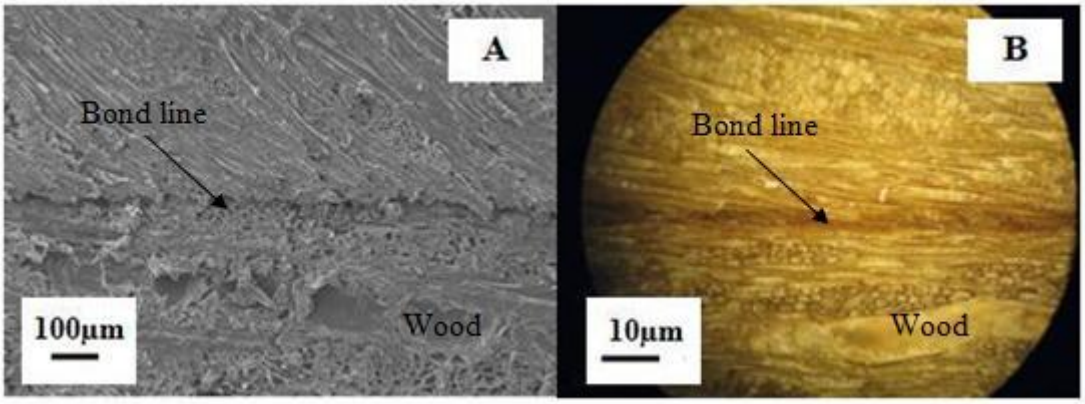

Figure 4: a) Scanning electron micrograph and b) optical micrograph of cross-section of the wood joints bonded by 25%wt WG-based adhesive.

At the constant WG content of 25 wt%, the concentration of NaOH solution used to prepare the adhesives was varied at 0.05 M, 0.10 M and 0.20 M. It was found that when the concentration of NaOH was increased, the viscosity of the adhesive dispersions was gradually increased (Table 2). With increasing the concentration of NaOH, it was observed that the pH of the adhesive dispersions was increased from 7 to 12. The isoelectric point of WG proteins was reported to be around pH 7 to 8 (Nordqvist et al. 2012). This pH shift would result in a larger negative charge on the protein molecules and an unfolding of the three-dimensional protein structure by cleaving both intra- and intermolecular linkages as illustrated in Figure 5. As a consequent, the unfolded molecules could lead to more chain entanglement and an increase in the viscosity of the adhesive dispersions.

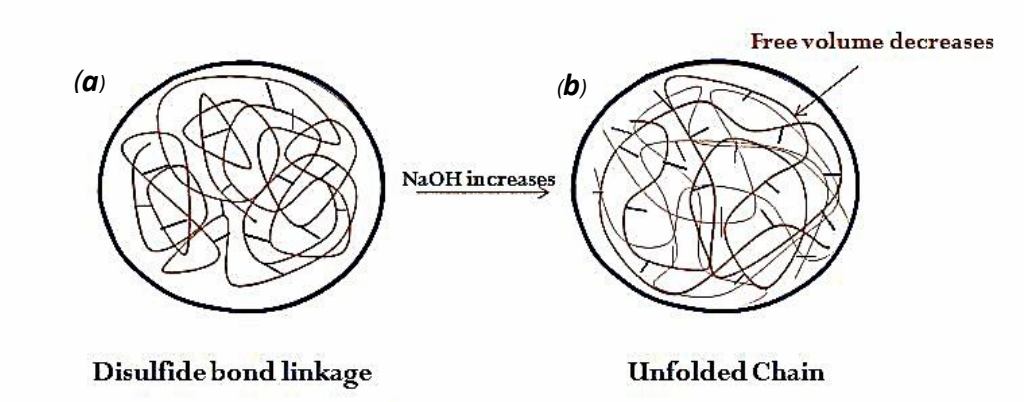

Figure 5: WG protein structure at a) isoelectric point and b) pH above isoelectric point.

In the adhesive formula of 25 wt% WG, with increasing concentration of NaOH, the bond strength of the wood joints linearly increased (Figure 3). Since NaOH acts as a denaturing agent, it disrupts the network structure of WG protein and results in the unfolded protein structure. For adhesive application, this change in protein structure should allow a larger number of functional groups on WG molecules available for further intermolecular interaction (crosslinking) when the adhesives were later applied and heated to bond the joints as illustrated in Figure 6. With a higher degree of crosslinking, the stronger adhesive bonds could be formed, leading to the higher bond strength as the NaOH concentration used in the adhesive preparation increased.

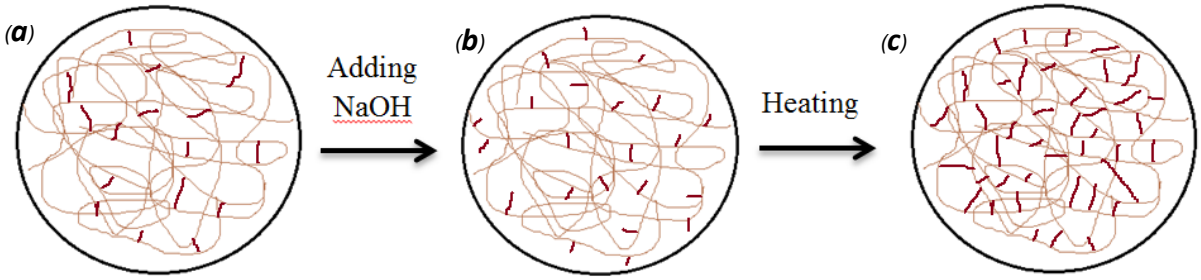

Figure 6: WG protein structure a) before and b) after addition of NaOH c) after crosslink by heating.

Conclusion

Both WG content and concentration of NaOH used to prepared bio-based adhesives significantly influenced their viscosities and adhesive properties. The optimum WG content was found to be 25 wt%. At this WG content, the adhesive had not too low or too high viscosity, resulting in a sufficient bond line thickness and degree of adhesive penetration into the wood panels. Thus, the highest bond strength in the wood joints boned by the 25 wt% WG adhesive was obtained. The bond line between the wood panels can be observed by both scanning electron microscopy and optical microscopy. In the adhesive preparation, with increasing concentration of NaOH solution, the viscosity of the adhesive was gradually increased. When pH of the WG adhesive dispersion shifted away from the isoelectric point, the WG molecules would change into unfolded structures. This caused more chain entanglement and, hence, an increase in the viscosity of the adhesives. The bond strength of the wood joints was linearly increased with concentration of NaOH solution that was used to prepare the adhesives. When the adhesives were applied on the wood panels and heated to set, the unfolded structures with a larger number of available functional groups on WG molecules could form higher density crosslinks, leading to the stronger adhesive bonds. In this work, the best adhesive formula was found to be prepared by 25 wt% WG content in 0.20 M NaOH solution which provided the highest bond strength of the wood joints approximately 7.07 MPa. To fulfill today's requirements for wood adhesives, the adhesive performance needs to be further improved, practically in the water resistance.

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References

D'Amicoa S., Hrabalovab M., Müllera U., Berghoferd E. (2010) Bonding of spruce wood with wheat flour glue—Effect of press temperature on the adhesive bond strength. Industrial Crops and Products 31: 255–260.

Ding R., Su C., Yang Y., Li C., Liu J. (2013) Effect of Wheat Flour on the Viscosity of Urea-formaldehyde Adhesive. International Journal of Adhesion & Adhesives 41: 1–5.

El-Wakil N.A., Abou-Zeid R.E., Fahmy Y., Mohamed A.Y. (2007) Modified wheat gluten as a binder in particleboard made from reed. Applied Polymer Science 106: 3592-3599.

Imam S.H., Gordon S.H., Mao L., Chen L. (2001) Environmentally friendly wood adhesive from a renewable plant polymer: characteristics and optimization. Polymer Degradation and Stability 73: 529-533.

Khosravia S., Nordqvist P., Khabbaz F., Johanssona M. (2011) Protein-based adhesives for particleboards—Effect of application process. Industrial Crops and Products 34: 1509–1515.

Kumar R., Choudhary V., Mishra B., Varma I.K. (2002) Adhesives and plastics based on soy protein product. Industrial Crops and Products 16: 155-172.

Leia H., Pizzi A., Navarrete P., Rigolet S., Redl A., Wagner A. (2010) Gluten Protein Adhesives for Wood Panels. Journal of Adhesion Science and Technology 24:1583-1596

Nordqvist P. (2012) Exploring the wood adhesive performance of wheat gluten. Ph.D. Thesis. KTH chemical science and engineering: Sweden.

Nordqvist P., Khabbaz F., Malmstr¨oma E.(2010) Comparing bond strength and water resistance of alkali-modified soy protein isolate and wheat gluten adhesives. International Journal o Adhesion & Adhesives 30: 72–79.

Nordqvist P., Lawther M., Malmström E., Khabbaz F. (2012) Adhesive properties of wheat gluten after enzymatic hydrolysis or heat treatment – A comparative study. Industrial Crops and Products 38: 139– 145.

Nordqvist P., Nordgren N., Khabbaz F., Malmström E. (2013) Plant proteins as wood adhesives: Bonding performance at the macro- and nanoscale. Industrial Crops and Products 44: 246– 252.