

Alkali-treated Wheat Gluten Cross-linked with Sodium Alginate as a Bio-based Wood Adhesive for Interior Grade Particleboard

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A bio-based wood adhesive formulation free of formaldehyde and made from alkali-treated wheat gluten (WG) and sodium alginate (SA) was developed. Its formulation was optimised, and it was characterised by Fourier Transform Infrared (FT-IR) spectroscopy. The bio-adhesive was utilized to make particleboards both with virgin wood particles and recycled wood particles. A dry bio-adhesive content of 35% (w/w) was used to make samples with both type of particles. Single-layer samples of 10 mm thickness were obtained using wood particles of 1 mm (both virgin and recycled). These samples then were subjected to 3-point bending tests. Whereas the bending strength of samples made with recycled wood particles was 18.09 N/m² and therefore satisfied Type 18 of the Japanese industrial standards (JIS A 5908:2015), the bending strength of the samples made with virgin wood particles was 8.08 N/m² and satisfied 'Type 8 Base particleboard Decorative particleboard' of the Japanese standards. The density of particleboard samples made from recycled wood particles was 916 kg/m³, while that of samples made from virgin wood particles was 732 kg/m³. The alkali-treated WG and SA bio-adhesive has the potential to be used to re-manufacture particleboards, which can then be recycled and not disposed in landfills.

Keywords: Sodium alginate; Wheat gluten; Bentonite; Particleboard

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INTRODUCTION

Formaldehyde emissions released by furniture made using synthetic wood adhesives are toxic for human beings due to their carcinogenic nature (WHO 2004). Although the wood itself emits formaldehyde, its contribution is minor in comparison to the emissions due to the presence of synthetic adhesives (Salem and Böhm 2013). Formaldehyde emissions from wood furniture can be minimized by replacing the synthetic adhesives with biobased adhesive formulations during the manufacturing process. Doing so would ultimately optimize indoor air quality (Zhang *et al.* 2018). In this study, sodium alginate, wheat gluten, and bentonite were selected as raw materials to develop a new wood bioadhesive formulation. The bio-based binder of the alkali-treated wheat gluten and sodium alginate was then combined with virgin wood particles and recycled wood particles to make particleboards. The resulting wood products were then characterized to evaluate whether they satisfied industrial standards.

The basic raw material for the production of sodium alginate (SA) is brown algae (Javed *et al.* 2015). Alginates are unbranched polysaccharides and complex carbohydrate polymers derived from brown seaweeds (Pawar and Edgar 2012). Tens of millions of tons (according to frozen weight) of seaweed is harvested both offshore and onshore (Nayar and Bott 2014). Alginates are used in industrial applications due to their gelling, viscosifying, and stabilizing properties (Draget *et al.* 2004). Sodium alginate has been used as a biodegradable additive in the papermaking industry in conjugation with polyamideamine-epichlorohydrin to increase the mechanical properties of the recycled fibers (Bai *et al.* 2017). When used as a structural scaffold, SA is usually blended, modified, or copolymerized with other biopolymers (Foroughi *et al.* 2018).

The chemical structure of SA, depicted in Fig. 1, shows that when SA is dissolved in water, it is able to react with the hydroxyl groups in the wood cell walls.

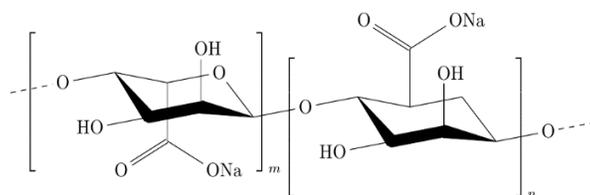


Fig. 1. Chemical structure of a sodium alginate molecule (Homayouni *et al.* 2007; Foroughi *et al.* 2018)

Alkaline bentonite binder has been used in roasted-pellet production (Gasik *et al.* 2009). The addition of bentonite has been shown to enhance the falling strength and compression strength of green pellets (Liu *et al.* 2017). By considering these factors, bentonite was selected for this study as an additive for the bio-based wood adhesive formulation. Wheat gluten (WG) is the rubbery mass that remains after the wheat dough is water washed to remove starch granules (Wieser 2007). Wheat gluten is an abundant protein source that can be obtained as a by-product from wheat starch processing (Nordqvist *et al.* 2013; Ferdosian *et al.* 2017). It consists of acid-dispersible glutenins (which have elastic properties) and alcohol soluble gliadins (which have viscous properties) (Hemmilä *et al.* 2017). Wheat gluten complex mixtures comprise of 80% wheat proteins, with the rest being lipids, polysaccharides, and minerals (Ferdosian *et al.* 2017; Hemmilä *et al.* 2017). It has been shown that rigid bio-based materials can be made using WG *via* high-temperature compression (Jansens *et al.* 2013b). In addition, WG can be blended with other polymers at high temperatures in order to manufacture bioplastics (Langstraat *et al.* 2015).

Bio-based adhesives can be made using WG due to its film-forming ability and its thermoplastic properties. Wheat gluten adhesives have been used to produce pressure-sensitive medical bandages and adhesive tapes (Day *et al.* 2006). As an elastomeric protein source, wheat gluten possesses cohesive and unique viscoelastic properties showing the suitability for material applications such as wood adhesives (Nordqvist *et al.* 2013; Mathias *et al.* 2016). The proteins derived from gluten can be divided into two groups (or subunits) according to their molecular weight, *i.e.*, high-molecular-weight (HMW) and low-molecular-weight (LMW) glutenin subunits (Khelifi and Branlard 1992; Schalk *et al.* 2017).

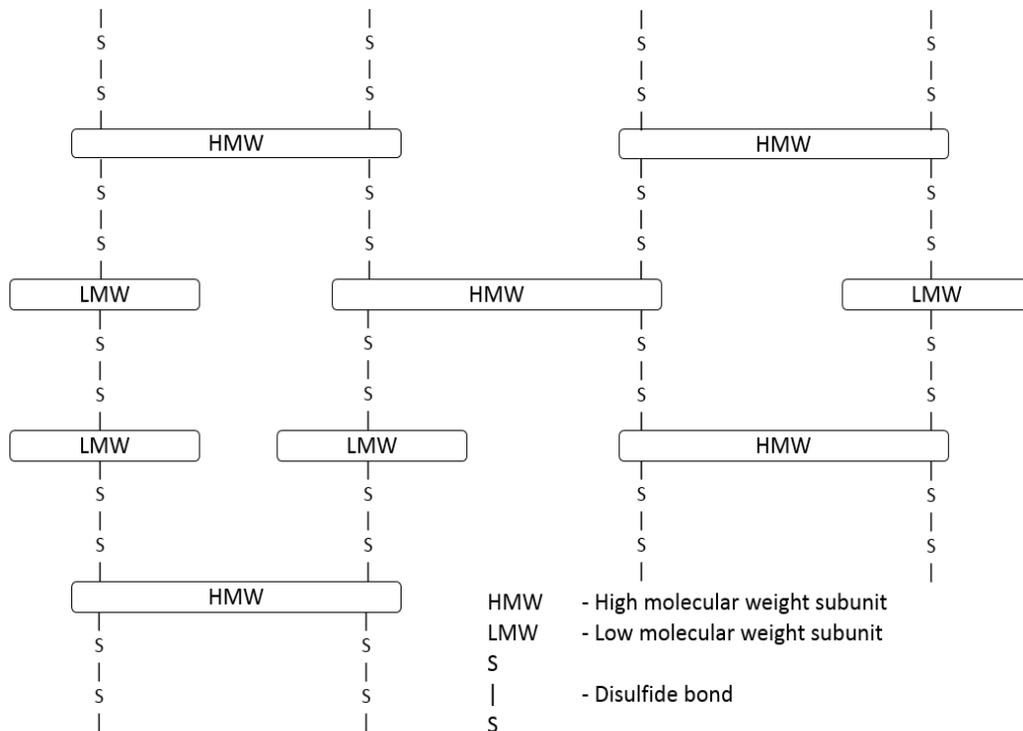


Fig. 2. A structural model for wheat gluten (Shewry *et al.* 2000; Lamacchia *et al.* 2014)

In the WG structure, the HMW subunits and LMW subunits are connected with disulfide bonds (Fig. 2). Wheat gluten consists of hundreds of proteins, and these proteins are present as monomers, oligo, or polymers linked *via* inter-chain disulfide bonds (Shewry *et al.* 2000; Wieser 2007; Lamacchia *et al.* 2014). The solubility of these proteins progressively increases with alkaline pH values (Du *et al.* 1994). In fact, the degradation of disulfide bonds can be performed by adding 0.2 M of NaOH at a temperature of 25 °C (Florence 1980). Whereas disulfide bonds seem to dominate the crosslinking at a temperature of 130 °C, the non-disulfide bonds contribute to the protein network at a temperature of 150 °C (Jansens *et al.* 2017). While WG is not dispersible in water due to its high amount of non-polar amino acids, it is dispersible in the presence of either alkalis or acids, as its isoelectric point is 7.3 (Khosravi *et al.* 2011; Hemmilä *et al.* 2017). It has been reported in literature that a mixture of WG and 0.2 M of NaOH as a wood adhesive applied on wood panels and heated to a temperature of 130 °C for 15 min under a pressure of 0.7 MPa, provided an optimum bond line thickness and a high bond strength of 7.07 MPa (Somord *et al.* 2014).

Wheat gluten mixed with starch (1:1 ratio), grafted with methylmethacrylate, crosslinked with citric acid, and nanofilled with TiO₂ has exhibited excellent thermal stability, water-resistance, and mechanical properties (Baishya *et al.* 2018). In addition, WG can be blended with synthetic wood adhesives. In order to improve the physical properties, *e.g.* water absorption and thickness swelling, of high-density particleboard using reeds instead of wood particles, 80% of the WG was modified with urea-formaldehyde resin alone as well as with the addition of 1% and 2% boric acid as a fungicide. The results showed that the mechanical properties, *e.g.*, the modulus of rupture, modulus of elasticity, and internal bond strength, did not change with the addition of boric acid (El-Wakil *et al.* 2007). On the contrary, the addition of a small proportion of an

isocyanate to hydroxymethylated or glyoxalated hydrolyzed gluten proteins yielded an improvement in the mechanical properties of the adhesive (Lei *et al.* 2010).

The addition of triacetin into a phenol-formaldehyde blended WG resin increased the internal bond strength by more than 30% (Lagel *et al.* 2015). However, when several particle sizes of alkali-denatured WG were used to prepare WG-protein wood adhesives, the size did not seem to affect the tensile strength (Nordqvist *et al.* 2010).

A two-steps method for the dispersion of WG in wood, which consisted of drying the glued chips in-between the first and the second addition, has shown to be more beneficial compared to a one-step process (Khosravi *et al.* 2011). The dispersion concentration, viscosity, and application method were the primary factors that affected the dispersion of wheat gluten into the wood particles. However, the drying temperature, presence of crosslinkers, dispersing agent, and type of wood species seemed to minimally affect the dispersion of wheat gluten into the wood particles (Khosravi *et al.* 2015). Improved bond strength and water resistance were achieved by using wood adhesives with hydrolyzed WG treated at a temperature of 90 °C and finally dispersed in 0.1 M of a NaOH water solution (aq) (Nordqvist *et al.* 2013). Bioproducts, *e.g.*, cups, plates, and boxes, were made using banana fiber-wheat gluten (Nataraj *et al.* 2018). NaOH solutions had the ability to effectively activate the wood surfaces, yielding strong adhesive bonds (Young *et al.* 1985).

Today, most of the bio-based wood adhesives that satisfy industrial standards are expensive due to the small-scale production and the high production cost. Wheat gluten as a by-product as well as a co-product in the wheat starch industry is readily available and comparatively less expensive than other bio-based wood adhesives, *e.g.* tannins (Wang *et al.* 2009; Jansens *et al.* 2013a; Ibarra *et al.* 2016). Compared to WG, smaller amounts of sodium alginate and NaOH are required to prepare an adhesive mixture. These are readily available and relatively inexpensive. Therefore, their use to prepare adhesive formulations to make interior grade particleboard that satisfies industrial standards is cost effective.

Hence, this research study was carried out to explore the possibility of using alkali-treated wheat gluten crosslinked with sodium alginate as a bio-based wood adhesive to make interior-grade particleboard. The evaluation of the quality of the bio-adhesives was performed by assessing the bending strength and the internal bond strength of the resulting particleboards.

EXPERIMENTAL

Materials

Wheat gluten was purchased from Buy Whole Foods Online Ltd (Kent, United Kingdom). Sodium alginate was bought from Sigma Aldrich (St. Louis, MO). Bentonite and NaOH were bought from Fisher Scientific (Waltham, MA). Kastamonu Entegre (Adana, Turkey) provided 1 mm virgin wood particles, and 1 mm recycled wood particles were provided by the French Institute of Technology for Forest-based and Furniture Sectors (Champs-sur-Marne, France).

Preparation of the Samples

The wood particles of 1 mm size were dried in a vacuum oven at 130 °C until the moisture became less than 5%. A mixture of wheat gluten, sodium alginate, and bentonite were mixed with the wood particles (virgin and recycled particles) while changing various

parameters such as weight ratios. A NaOH solution (1 M) was sprayed on the mixture until the moisture content was 18%. The mixture was then hot-pressed for 15 min at a temperature of 150 °C at a pressure of 1.4 MPa in a hydraulic hot press machine (50 ton press acting on 5" Hydraulic Ram) to make single-layer particleboard sample. The dimensions of the samples prepared were 210 mm × 210 mm × 10 mm. The samples were cut as in Fig. 3 and underwent a 3-point-bend-test and an internal bond (IB) test using the 5/100 kN Instron 5500R Electro-mechanical machine. Samples from three particleboard specimens prepared with the same formulation and conditions were used for the testing. An environmental scanning electron microscope (E-SEM) was used to analyze the composition of the samples.

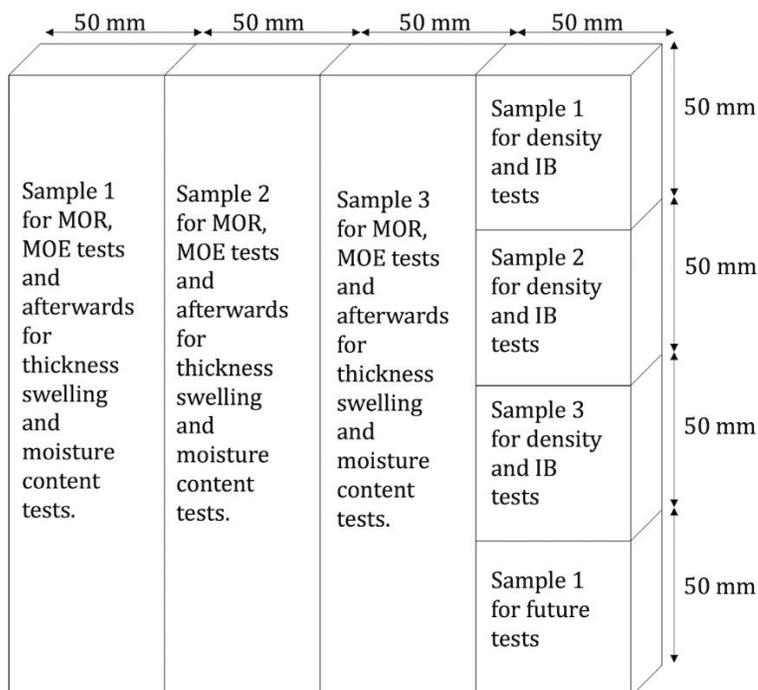


Fig. 3. Cutting to the size of the particleboard for physical and mechanical tests.

Taguchi Design of Experiments

In order to speed up the optimization of the experimental work, based on the number of variable parameters, *e.g.* the amount of wood particles, bentonite, sodium alginate, and wheat gluten, a relevant Taguchi orthogonal array was carried out. MINITAB 16 software (Penn State University, University Park, PA) was used to analyze the experimental results. This was helpful in reducing the number of experimental trials necessary for the optimization of the bio-adhesive formulation.

Environmental scanning electron microscopy (E-SEM) analysis

Samples for the environmental-scanning electron microscope (E-SEM) were prepared by crushing both types of particleboard samples (from virgin and recycled particles using hammer blows) and then gold coating them. A FEI XL30 (Thermo Fisher Scientific, Waltham, MA) machine was used for the analysis.

X-ray diffraction analysis

Prior to the x-ray diffraction analysis, the wheat gluten, sodium alginate, and the appropriate mixture of wheat gluten and sodium alginate samples were mixed with a NaOH solution (1 M). The amount of the solution used was measured afterwards. The measurements were then performed at room temperature. Oxford Diffraction Xcalibur 2 was used for the X-ray diffraction.

Fourier transform infrared (FTIR) analysis

Fourier transform infrared spectroscopy (FTIR) was employed to investigate the absorption characteristics of the uncured (alkali-treated WG) and cured samples following synthesis with the aim of identifying the chemical bonds of the compounds and the transformation of their functional groups subjected to curing. The FTIR analysis was performed using an attenuated total reflection (ATR) accessory that allowed the determination of the change in chemical functional groups under different process conditions.

In this experiment, FTIR equipment (Jasco 6200, Easton, MD) was used in conjunction with a PIKE MIRacle ATR accessory. An AZnSe crystal plate was fitted as the ATR sampling interface. Initially, the set up was calibrated in air to reduce the effect of environmental conditions before the sample assessments occurred over a wide wavenumber range (4000 to 550 cm^{-1}). The ATR-FTIR absorbance spectra of the uncured and cured resins of the alkali-treated wheat gluten crosslinked with sodium alginate were compared, and the results are shown in Fig. 9.

RESULTS AND DISCUSSION

Statistical Analysis

The Taguchi experimental design method was used to evaluate the influence of the compositions of the resin mixture on the modulus of rupture (bending strength), as shown in Table 1. It was found that an increase in the bentonite quantity in the resin reduced the modulus of rupture of the sample, while an increase in the wheat gluten quantity increased it (Fig. 4).

Table 1. Changes of Quantities of Adhesive Components and the Resulted Bending Strength for Taguchi Analysis

| Sample number | Wood particles (g) | Bentonite (g) | Sodium alginate (g) | Wheat gluten (g) | Bending strength (Nm^{-2}) |
|---------------|--------------------|---------------|---------------------|------------------|---------------------------------------|
| 1 | 200 | 45 | 30 | 0 | 4.21 |
| 2 | 200 | 40 | 35 | 0 | 4.43 |
| 3 | 200 | 29 | 57 | 0 | 4.80 |
| 4 | 200 | 43 | 57 | 0 | 5.20 |
| 5 | 200 | 29 | 71 | 0 | 5.88 |
| 6 | 200 | 43 | 71 | 0 | 6.20 |
| 7 | 200 | 31 | 46 | 31 | 6.07 |
| 8 | 200 | 31 | 31 | 46 | 6.65 |
| 9 | 200 | 15 | 62 | 31 | 6.22 |
| 10 | 200 | 15 | 31 | 62 | 7.03 |

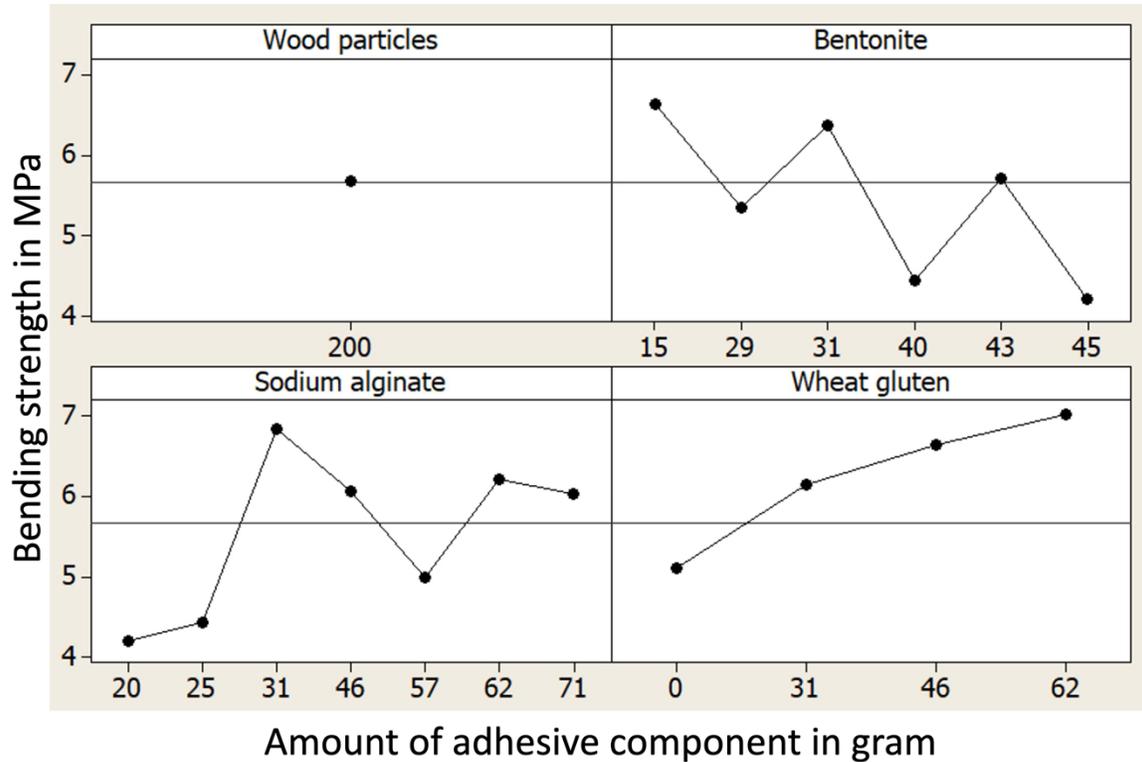


Fig. 4. Taguchi experiment design analysis for the inputs

By considering the Taguchi analysis results, the weight proportions of the wood particles, wheat gluten, and sodium alginate were chosen as 62%, 31%, and 7% (w/w), respectively, in order to achieve optimal results for the bending strength. A lower amount of sodium alginate was chosen to have better physical properties of the final samples. The necessary concentration of NaOH was 1M in order to minimize the agglomeration of the resin mixture.

Environmental scanning electron microscopy (E-SEM) analysis

The optimized adhesive resin was used to prepare particleboards using virgin wood particles and recycled wood-based panel particles. Both particleboards were made from particles of the same size. Increased amounts of N and O were observed as shown in Table 2 in the recycled wood particleboard, which was due to the presence of urea-formaldehyde, as observed by the composition analysis of EDX via E-SEM.

Table 2. Elemental Analysis of Samples Made with Virgin Wood Particles and Recycled Wood Particles

| Sample type | Sample number | Element percentages | | |
|-------------------------|---------------|---------------------|-------|-------|
| | | C | N | O |
| Virgin wood particles | 1 | 54.20 | 8.45 | 34.12 |
| | 2 | 56.26 | 5.10 | 37.71 |
| | 3 | 38.20 | N/A | 33.70 |
| Recycled wood particles | 1 | 44.33 | 13.81 | 39.84 |
| | 2 | 45.28 | 12.99 | 40.85 |
| | 3 | 46.77 | 14.32 | 38.36 |

The wood cell lumens were also observed *via* E-SEM images of the virgin wood particleboard samples (Fig. 5). However, the lumens were not observed in the particleboards made using recycled wood-based panel particles, which were comparatively higher in density and had different mechanical properties.

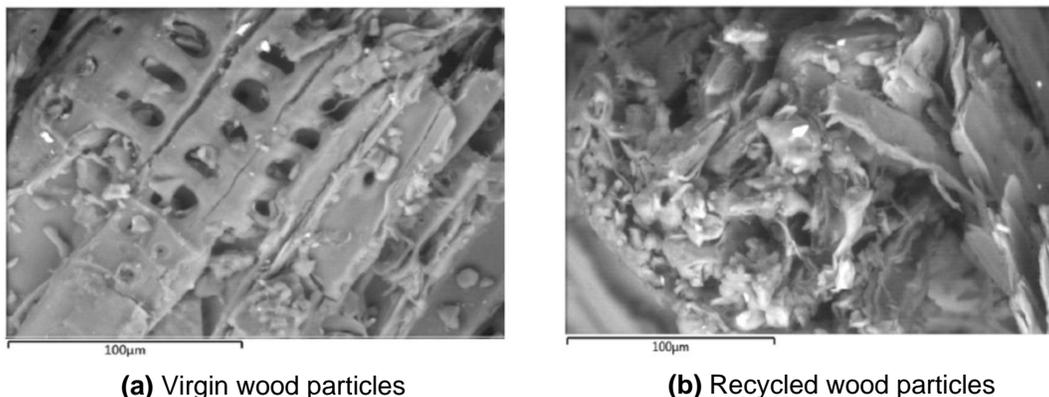


Fig. 5. E-SEM images of the particleboard samples prepared using virgin wood particles and recycled wood-based panel particles

X-ray diffraction analysis (XRD)

The resin was subjected to XRD analysis to evaluate its crystallinity and compare it to WG and sodium alginate separately. Figure 6 shows their diffraction patterns. The crystallinity indexes (CI) were calculated according to Eq. 1,

$$\text{Crystallinity Index} = \left[\frac{I_{200} - I_{am}}{I_{200}} \right] \times 100\% \quad (1)$$

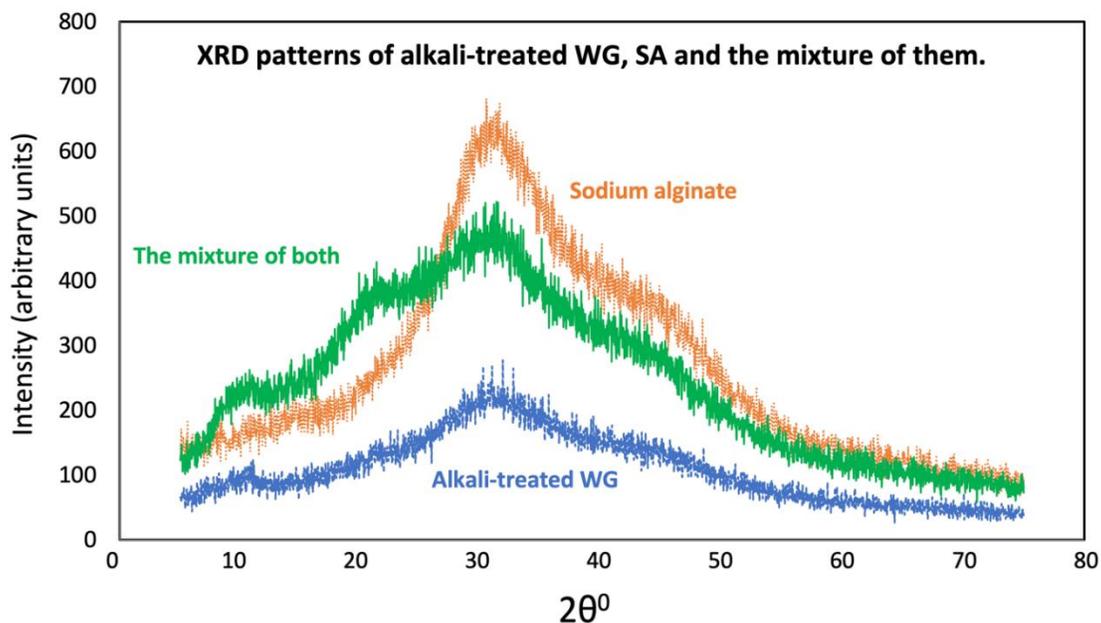


Fig. 6. XRD analysis of the alkali-treated wheat gluten, sodium alginate, and a mixture of both

where I_{200} is the peak intensity corresponding to the crystalline fraction and I_{am} is the intensity of the amorphous fraction. Then, the CI were compared by using the diffraction patterns (as shown in Fig. 7).

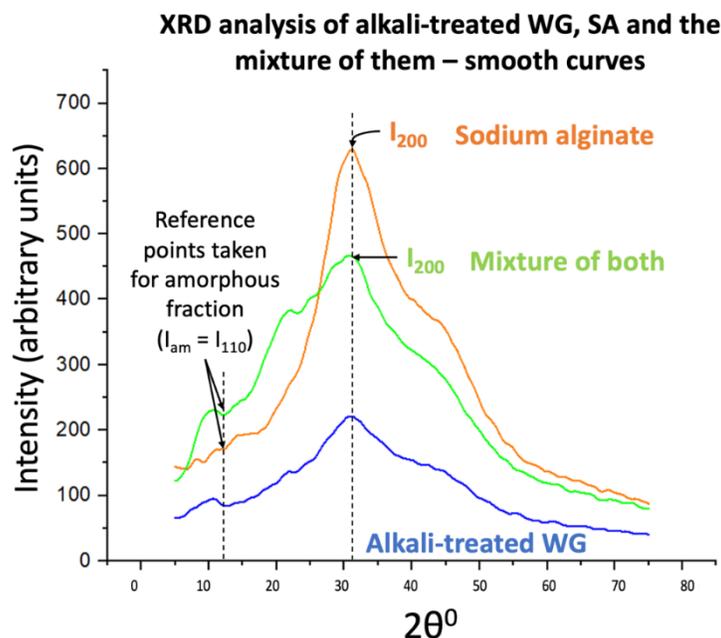


Fig. 7. XRD analysis of alkali-treated wheat gluten, sodium alginate and their mixture using smooth curves taken by using OriginPro2021 software

The crystallinity index of the alkali-treated mixture (WG+SA) was lower than the other tested samples (Table 3). A lower crystallinity index implies a higher bond strength. This shows that the modification of wheat gluten and sodium alginate can increase the strength of the final wood composite (Amini *et al.* 2019).

Table 3. Crystallinity Indexes of the Samples

| Sample | I_{200} | I_{am} | CI (%) |
|---------|-----------|----------|--------|
| WG | 225 | 85 | 62 |
| SA | 632 | 165 | 74 |
| WG + SA | 467 | 225 | 52 |

Fourier transform infrared (FTIR) analysis

The results of the FTIR analysis of wheat gluten and the cured and uncured alkali-treated wheat gluten crosslinked with sodium alginate resins are shown in Fig. 8 (full range) and Fig. 9 (in detail). The assignment of the peaks is presented in Tables 4 and 5.

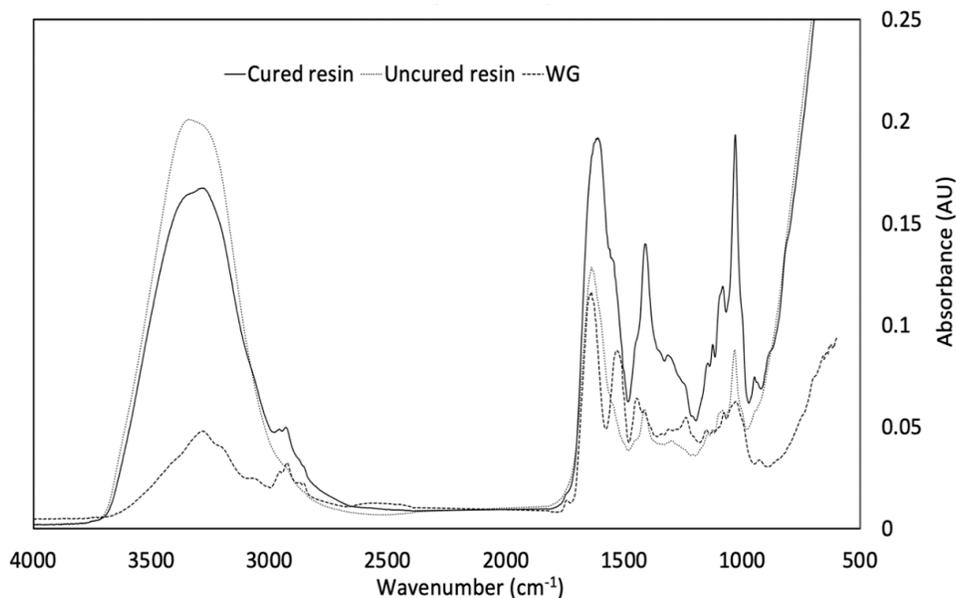


Fig. 8. FTIR analysis of wheat gluten and the alkali-treated cured and uncured resins (full range)

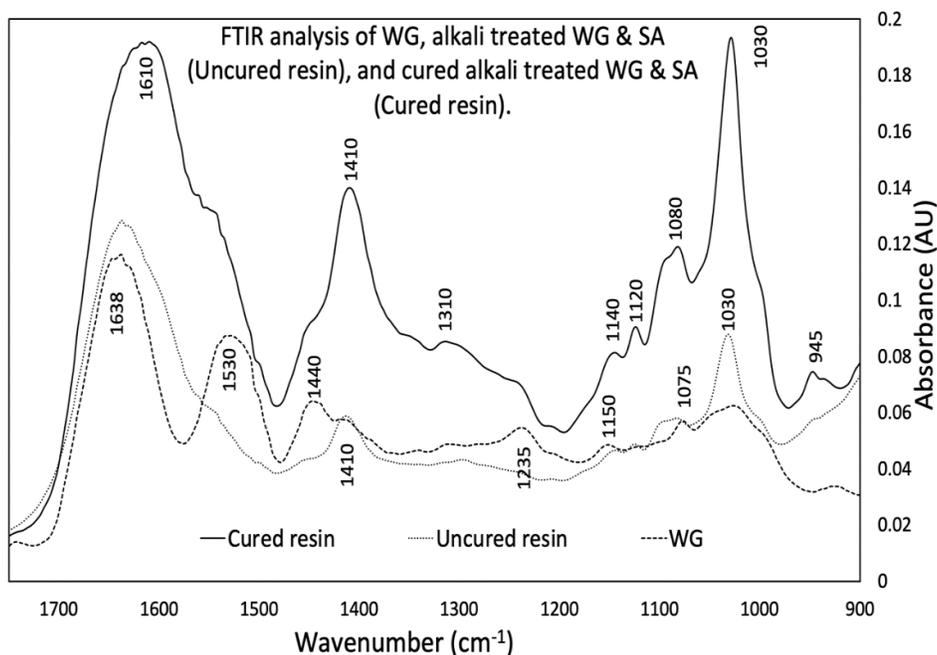


Fig. 9. FTIR analysis of wheat gluten and the alkali-treated cured and uncured resins

The peak shown in Table 4 at 1638 cm^{-1} of wheat gluten can be assigned to the stretching frequency of the C=O in the amide groups. The stretching C=N, C=C, C-N, and N-H bending can be seen at 1530 cm^{-1} . The peak disappeared in the uncured resin mixture with sodium alginate and in wheat gluten treated with NaOH probably due to a partial crosslinking reaction. In addition, the peak at 1440 cm^{-1} , which can be assigned to the C-O-H bending of wheat gluten, was shifted to 1410 cm^{-1} when WG and SA were alkali-treated, probably as a result of the formation of S=O or sulfone groups. A dramatic increase in the intensity of the peak at 1410 cm^{-1} can be seen after the samples underwent heat treatment (curing), which produced more sulfone groups. Phosphorus and potassium are

contained in the fertilizers that are used in winter to enhance the wheat yield and quality. The peaks at 1235, 1150, and 1075 cm^{-1} exemplify the presence of phosphorus and potassium in wheat gluten (Gaj *et al.* 2012). The C-O-C stretching of sodium alginate and S=O sulfoxide stretchings of the partially bonded sulfoxide, which arise from the crosslinking of the LMW/HMW with sodium alginate, were observed at 1030 cm^{-1} of the uncured resin curve. Followed by the heat treatment, the intensity of the peak located at 1030 cm^{-1} of the cured sample increased and the peak also broadened (Fig. 9). This might represent the formation of more C-O-C and S=O sulfoxide stretching during the heat treatment *via* the crosslinking of sodium alginate and sub-units of wheat gluten (as shown by the reaction in Fig. 10).

Table 4. FTIR Assignments for Wheat Gluten and Uncured Resin Mixture

| Wavenumber (cm^{-1}) | Remarks | Reference |
|---------------------------------|---|---|
| 1638 | Stretching frequency of C=O of amide group | Goel <i>et al.</i> (2017) |
| 1530 | Stretching C=N, C=C, C-N stretching and N-H bending | Daniel-da-Silva <i>et al.</i> (2008); Movasaghi <i>et al.</i> (2008); Kläusler <i>et al.</i> (2014) |
| 1440 | C-O-H bending | Reusch (2013) |
| 1235 | Phosphate vibration, C-O stretching | Chiriboga <i>et al.</i> (1998); Movasaghi <i>et al.</i> (2008) |
| 1150 | C=O, P=O, P-O-C (P-O-P) asymmetric stretching | Filip <i>et al.</i> (2008); Movasaghi <i>et al.</i> (2008) |
| 1075 | Symmetric stretching of phosphodiesteres | Fujioka <i>et al.</i> (2004) |
| 1030 | C-O-C stretching, S=O sulfoxide stretching | Narain (2010); Santiago-Medina <i>et al.</i> (2017) |

Table 5. FTIR Assignments for Cured Resin Mixture

| Wavenumber (cm^{-1}) | Remarks | Reference |
|---------------------------------|---|--|
| 1610 | Aromatic C=C | Emmanuel <i>et al.</i> (2015) |
| 1410 | Stretching vibration of COO- | Yuan <i>et al.</i> (2017) |
| 1610 | Stretching of C-N bonds | Jitendra <i>et al.</i> (2015); Ji and Guo (2018) |
| 1140 | C-O-C anti-symmetric stretching, S=O Sulfone | Zhang <i>et al.</i> (2013); Reusch (2013) |
| 1120 | Strong (O-CH ₂), Mannose-6-phosphate | Yoshida <i>et al.</i> (1997); Ooij (2003) |
| 1080 | Symmetric phosphate stretching (P=O stretching and P-O-C (P-O-P) bonds) | Filip <i>et al.</i> (2008); Movasaghi <i>et al.</i> (2008) |
| 1030 | C-O-C stretching, S=O sulfoxide stretching | Reusch (2013); Santiago-Medina <i>et al.</i> (2017) |
| 945 | =NOH oxime N-O | Reusch (2013) |

The shift of the peak in the uncured sample from 1638 to 1610 cm^{-1} after curing (Table 5) might represent the formation of clear aromatic C=C vibrations (Brudler *et al.* 1995; Emmanuel *et al.* 2015). During the curing process, the C=O of the amide groups from the wheat gluten proteins crosslinked with the aromatic sodium alginate

(Satheeshabadu and Mohamed 2015). The peak at 1410 cm^{-1} in the cured sample might correspond to the reaction of $-\text{COONa}$ from the sodium alginate with the amino acids from the wheat gluten protein. Stretching of C-N bonds of the amide is represented by the peak at 1310 cm^{-1} .

The C-O-C anti-symmetric stretching and S=O sulfone represented at 1140 cm^{-1} might be due to the sulfonyl groups formed (Fig. 10). The formation of sulfoxide and sulfonyl bonds revealed that the wheat gluten subunits were separated due to the alkali treatment, and they were probably interacting with ionized sodium alginate.

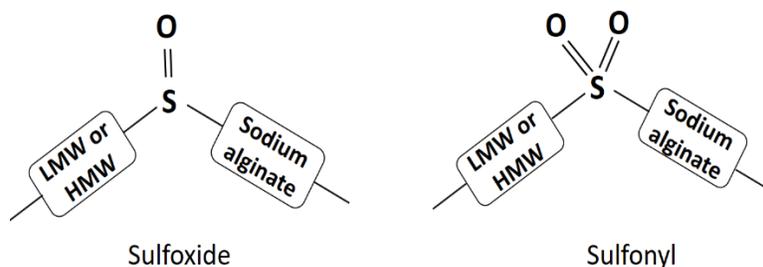


Fig. 10. Crosslink reaction between the low or high molecular weight subunits of wheat gluten with sodium alginate

As mentioned above, wheat gluten harvested during winter contains phosphorus and potassium fertilizers (Gaj *et al.* 2012). The peaks at 1120 and 1080 cm^{-1} observed in the cured resin indicated the presence of phosphorus in wheat gluten even after curing. Bonding of the hydroxyls with the $-\text{NH}$ groups from the amino acid monomers of wheat gluten can be seen at the newly formed peak at 945 cm^{-1} in the cured resin. Therefore, the FTIR analysis established that the curing promoted additional peaks that showed the reactivity after heat treatment, enabling functionalization, as expected.

The resin was used to prepare particleboard samples using virgin wood particles and recycled wood-based panel particles of the same size (1 mm).

The bend strengths and the internal bond values of the virgin and recycled wood samples obtained are presented in Table 6. The IB strength is interrelated with the material density.

Table 6. Mechanical Properties of Particleboard Samples

| Type of Wood Particle | IB Strength (Nm^{-2}) | Bending Strength (Nm^{-2}) | JIS A 5908:2015 |
|-----------------------|----------------------------------|---------------------------------------|-----------------|
| Virgin | 0.52 ± 00 | 8.08 ± 0.15 | Type 8 |
| Recycled | 1.29 ± 01 | 18.09 ± 0.07 | Type 18 |

Note: The values of the three different samples used for the testing are reported in Table S1 in the Appendix.

Particleboard samples made using virgin wood particles satisfied the requirements for “type 8 base particleboard decorative particleboard” according to JIS A 5908:2015 (2015), while recycled wood particles satisfied the requirements for type 18. The particleboards made using recycled wood particles had greater than twice the IB value due to its high density and less porous nature. The wood cells of the recycled wood particles were filled with previously used synthetic adhesives.

The FTIR analysis performed showed the presence of crosslinking in the alkali-treated wheat gluten and sodium alginate resin, whereas the XRD analysis demonstrated the high bonding strength of the resulting panel. The mechanical tests revealed that the sample made with the novel bio-based adhesives can satisfy industrial standards, according to JIS A 5908:2015 (2015). While the type 8 particleboards did not meet the thickness swelling (TS) requirements, the type 18 particleboards did; for this type of particleboard the TS should be less than 12%. The TS achieved in this study for both types of particleboard was 22%.

The use of bentonite as a hardener in the wood adhesive reduced the bending strength and increased the thickness swelling. This corresponded to the brittle nature of bentonite clay as well as its tendency to swell. After conducting the Taguchi experimental design analysis, bentonite was not included in the resin formulation, even though it is known to penetrate through the wood cell walls and make mechanical interlocks.

A comprehensive thermal and rheological analysis, *e.g.*, TGA/DTG, DSC, and DMA, can be done to characterize the resin behavior, but this was out of the scope of this paper. The physical properties, *e.g.*, water-resistance, of the adhesive formulation can be improved during future research.

CONCLUSIONS

1. The disulfide bonds between the low molecular weight (LMW) and high molecular weight (HMW) proteins subunits break with the addition of NaOH. In an aqueous medium, the -ONa and -OH groups in sodium alginate ionize to O^- , Na^+ , and H^+ ions. Some of the ionized O^- from the sodium alginate form bonds *via* crosslinking with unbonded disulfide in the low and high molecular weight subunits of wheat gluten (Fig. 10). The rest of the ionized O^- bonds with the hydroxyl groups in the cellulose, hemicellulose, and lignins of wood cell walls. The adhesive formulation of alkali-treated wheat gluten (WG) and sodium alginate (SA) in the ratio of 6:1 wt. satisfies the Type 8 of JIS of the particleboard samples made with virgin wood particles.
2. There was improvement in the mechanical and physical properties when the resin was used with recycled wood panel particles, which contained urea-formaldehyde adhesive. This reveals that the proteinaceous wood adhesive can be used to re-manufacture wood-based panels, as proteinaceous adhesives can be blended with synthetic wood adhesives. This can divert the wood-based panel industry from a linear economy to a circular economy. The research was carried out based on the bending strength and internal bond strength as an initial step using an experimental design approach.

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APPENDIX

Table S1. Mechanical and Physical Properties of Particleboard Samples Made by Using Virgin Wood Particles and Recycled Wood Particles

| | Particleboard made with virgin wood particles | | | Particleboard made with recycled wood particles | | |
|---|---|----------------|----------------|---|----------------|----------------|
| Sample number | 1 | 2 | 3 | 1 | 2 | 3 |
| Length (mm) | 200.00 | 200.00 | 200.00 | 200.00 | 200.00 | 200.00 |
| Width (mm) | 49.03 | 49.34 | 50.06 | 49.67 | 50.31 | 49.82 |
| Thickness (mm) | 10.10 | 9.87 | 10.05 | 9.67 | 9.87 | 10.19 |
| Span (mm) | 150.00 | 150.00 | 150.00 | 150.00 | 150.00 | 150.00 |
| Failure load (N) | 180.00 | 178.00 | 175.50 | 376.45 | 392.70 | 414.00 |
| MOR (N/mm²) | 8.10 | 8.33 | 7.81 | 18.24 | 18.03 | 18.01 |
| Average MOR (N/mm²) | 8.08 | ± | 0.15 | 18.09 | ± | 0.07 |
| P1 (10% of F load) | 18.00 | 17.80 | 17.55 | 37.65 | 39.27 | 41.40 |
| P2 (40% of F load) | 72.00 | 71.20 | 70.20 | 150.58 | 157.08 | 165.60 |
| P1 value | 18.02 | 17.81 | 17.54 | 37.64 | 39.29 | 41.42 |
| P2 value | 72.01 | 71.22 | 70.21 | 150.58 | 157.09 | 165.61 |
| d1 value | 1.44 | 1.45 | 1.42 | 1.38 | 2.18 | 2.10 |
| d2 value | 1.88 | 1.90 | 1.84 | 2.03 | 2.82 | 2.73 |
| MOE (N/mm²) | 2049.50 | 2110.93 | 2082.28 | 3264.18 | 3210.50 | 3155.25 |
| Average MOE (N/mm²) | 2080.90 | ± | 17.75 | 3209.98 | ± | 31.45 |
| Length (mm) | 49.93 | 49.97 | 50.05 | 50.02 | 49.97 | 49.96 |
| Width (mm) | 49.03 | 49.34 | 50.06 | 49.67 | 50.31 | 49.82 |
| Thickness (mm) | 10.10 | 9.87 | 10.05 | 9.67 | 9.87 | 10.19 |
| weight (g) | 18.01 | 17.93 | 18.43 | 22.00 | 22.45 | 23.51 |
| Density (kg/m³) | 728.40 | 736.81 | 731.92 | 915.71 | 904.76 | 926.94 |
| Average density (kg/m³) | 732.38 | ± | 2.44 | 915.81 | ± | 6.40 |
| Failure load (N) | 1275.54 | 1284.90 | 1292.76 | 3255.00 | 3189.00 | 3212.00 |
| IB (N/mm²) | 0.52 | 0.52 | 0.52 | 1.31 | 1.27 | 1.29 |
| Average IB (N/mm²) | 0.52 | ± | 0.00 | 1.29 | ± | 0.01 |
| Mass before dry (g) | 19.78 | 19.93 | 19.35 | 21.65 | 21.00 | 20.73 |
| Mass after dry (g) | 18.23 | 18.44 | 17.73 | 19.36 | 18.88 | 18.49 |
| Moisture content (%) | 9% | 8% | 9% | 12% | 11% | 12% |
| Average MC (%) | 9% | ± | 0% | 12% | ± | 0% |
| Thickness before (mm) | 10.10 | 9.87 | 10.05 | 9.67 | 9.87 | 10.19 |
| Thickness after (mm) | 11.27 | 11.16 | 11.22 | 10.58 | 10.65 | 11.20 |
| Thickness swelling (%) | 12% | 13% | 12% | 9% | 8% | 10% |
| Average TS (%) | 12% | ± | 0% | 9% | ± | 1% |