Modified Wheat Gluten as a Binder in Particleboard Made from Reed

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ABSTRACT: To evaluate the utilization of reed in environmental friendly high-density particleboard, modified wheat gluten was used with urea-formaldehyde as a binder in different ratios, applying the optimum conditions obtained on using UF resin alone. The scanning electron microscopy of the reed fibers showed that the fibers are cylindrical in shape, which helps in enhancing the adhesion between the binder and the inner and outer surfaces of the fibers. The dependence of the mechanical properties (modulus of rupture, modulus of elasticity and the internal bond) and the physical properties (water absorption and thickness swelling) on the urea-formaldehyde/modified gluten ratios was studied. Addition of 1% and 2% boric acid as a fungicide to the binder mixture resulted in no significant change in the mechanical properties and slight improvement in the physical properties of the produced particleboard. Thermogravimetric analyses of selected samples were done to study the thermal stability of the particleboard bonded with the modified binder with and without boric acid. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 106: 3592–3599, 2007

Key words: reed; wheat gluten; urea-formaldehyde; mechanical properties; physical properties; thermogravimetric analysis

INTRODUCTION

Particleboard is a panel product manufactured by pressing wood particles or other nonwood fibers and a binder. Some of the typical applications for particleboard are packing, building, and furniture manufacture. Nonwood fibers are renewable resources that can be utilized as raw materials for particleboard production. Considerable amount of work has been carried out on the use of a wide variety of nonwood fibers in the particleboard manufacture such as wheat-cereal straws,[1–3] rice husks,[4] groundnut shells,[5] bamboo,[6] waste of tea leaves,[7,8] bagasse,[9,10] cotton, hemp and jute stalks,[11] sunflower stalks,[12] reed,[13] and reed and typha.[14] One of the nonwood fibers that can replace wood, as a raw material for particleboard manufacture, is reed. Reed is a grass-type plant that generally grows in swamplands, river bottom land, and delta areas that are found in Russia, Romania, Egypt, China, North Korea, Iraq, and Turkey. The stalks of reed may grow up to 15 feet, and the fiber length ranges from about 1.5–3.0 mm.[15]

Urea-formaldehyde (UF) is one of the most commonly used resins for particleboard industry due to a number of advantages including low cost, ease of use under a wide variety of curing conditions, low cure temperature, water solubility, resistance to microorganisms and to abrasion hardness, excellent thermal properties, and lack of color, especially of the cured resin. The major disadvantage associated with UF adhesives is the lack of resistance to moist conditions, especially in combination with heat. These conditions lead to a reversal of the bond forming reactions and consequently emission of formaldehyde.[16,17] The reaction leading to the UF products formed during UF resin synthesis and cure are reversible. In the forward direction, water is eliminated; therefore, the reverse reactions can be viewed as hydrolysis, which leads to the release of formaldehyde.[18] Because most, if not all, of these reactions are catalyzed by acid, the use of an acid catalyst, to hasten bond cure, unfortunately also increases the rate of hydrolysis and formaldehyde liberation. The reduction of formaldehyde emission levels from products bonded with UF resins has been achieved by employing one or more of several methods.[19]

There is much interest in developing more environmental friendly adhesives as natural resin. Researchers have investigated the use of natural resins derived from local plants so as to replace synthetic resins in board manufacture. Soy protein, wheat gluten, and milk casein are examples of these natural adhesives.[20] Wheat gluten (WG) has the potential to be a good binder because of its film-forming properties and its ability to reduce the release of formaldehyde.[21] WG is defined as water-insoluble protein of wheat flour that

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remains after flour dough is kneaded. It is composed of two water-insoluble proteins found in wheat flour, glutenin and gliadin. Glutenin molecules have linear configurations and the potential to form disulfide and other crosslinks. When hydrated, glutenin is firm and elastic, while Gliadin, which consists of small globular molecules, is soft and has good adhesive properties.[22] Six amino acids are contained in WG, namely, glutamic acid (2%), glutamine (37%), praline (14%), leucine (7%), lysine (1.2%), and cystine (2.1%).[23]

Generally, all of the above natural adhesives have to be modified to be used in particleboard formation. Different methods have been applied to improve the adhesion properties of, e.g., soy protein. From these methods are using urea, citric acid, boric acid, and sodium hydroxide in wheat straw-soy protein-bonded particleboard,[24] denaturing and disulphide bond cleavage,[25,26] crosslinking, acylation, and oxidation.[27–29] Modification of soy protein isolate to affect hygroscopicity, solubility, and emulsifying properties using papain and trypsin was also carried out.[30,31] Scheyer and Polsani[21] modified gluten to be used as a binder in print paste and also as a formaldehyde scavenger where melamine formaldehyde resin was used in the print paste formulation.

The goal of the present work is to investigate the addition of modified wheat gluten into particleboard made from reed, aiming to reduce the amount of UF resin used. Modification of WG adhesive is to be carried out using NaOH and urea. Also, the proper proportion of urea-formaldehyde/modified wheat gluten will be examined to achieve the best mechanical and physical properties of the produced particleboard.

EXPERIMENTAL

Raw materials used

Reed was collected and cut into small pieces and then dried to reduce moisture content to about 4–6%. The pieces were ground and screened through a mesh of 0.2 mm to remove larger particle size. Chemical composition of reed was analyzed to be 47.95% cellulose, 24.85% lignin, 22.3% pentosan, 2.9% ash, and 2.0% silica.

Urea-formaldehyde (UF) adhesive was delivered from Tanta Flax and Oil Company. It is of 60% solid content, specific gravity 1.25 g/cm^3, viscosity 400 cps, and pH 7.5–8.5 at 20°C. The formaldehyde/urea molar ratio is 1 : 1.13.

Wheat gluten (WG) was delivered from Cerestar company (France). It has the following composition: protein 79%, ash 1.0–2.0%, moisture 5.0–8.0%, fat 1.0–2.0%, fiber 1.0%, and carbohydrate 9–12%.

Boric acid was delivered from Laboratory Rasayan.

Scanning electron microscopy

The cross section of reed fibers was examined with electron microscope (JXA-840A Electron Probe Micro Analyzer Scanning Electron Microscope).

Thermal analysis

Thermal analysis was carried out using PerkinElmer thermogravimetric analyzer. Heating rate set at 10°C/min over temperature range between 50 and 750°C. Measurements were carried out in nitrogen atmosphere; the rate of flow of nitrogen was 50 cm^3/min.

Preparation of WG adhesive

Wheat gluten was added into distilled water and stirred mechanically for 20 min to prepare 15% slurry. The WG adhesive thus prepared was modified using urea and sodium hydroxide. Two series of adhesive where prepared in which urea was added 0.5 and 1.5*M* respectively, and mixed with slurry for 20 min. To each series, sodium hydroxide of different proportions (1%, 1.5%, 2%, 3%, and 4%, based on the amount of gluten) was dissolved in the least amount of distilled water, and added to the slurry and stirred for 30 min.

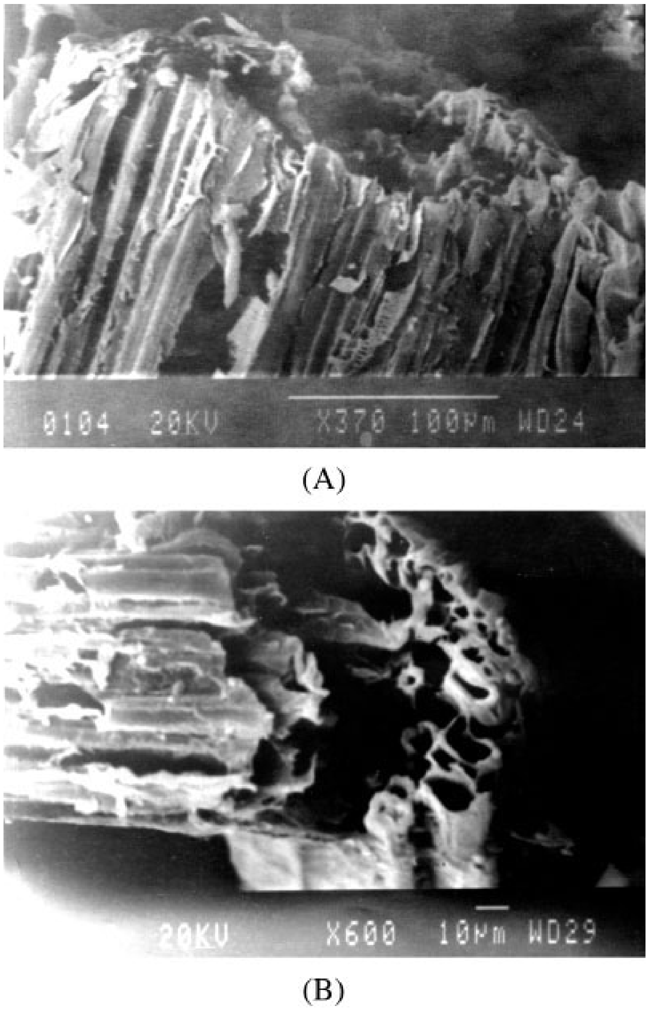
Board formation

To achieve the best conditions for board formation, UF resin was first used alone as the adhesive. The amount of resin added, based on the resin solid content and the oven dry weight of the reed, were 8%, 10%, and 12%. Ammonium chloride solution was used as a hardener and paraffin wax emulsion as water-repelling agent. The resin was sprayed onto the particles; the hand-formed particle mats were then pressed at platen temperatures of 130, 145, and 160°C, pressing time of 8, 12, and 16 min, and a pressure of 3.9 N/mm^2.

Different ratios of urea-formaldehyde/modified wheat gluten (UF/G) were used to partially substitute UF resin. These ratios were 80/20, 60/40, 40/6, 20/80, and 0/100 UF/G, respectively. The conditions used for the thus prepared board formation were resin concentration 12%, pressing temperature 160°C, pressing time 12 min, and pressing pressure 3.9 N/mm^2.

Specimens for testing

After hot pressing, boards were conditioned at 20°C and 65% relative humidity for 72 h and then trimmed and cut into specimens of specific dimensions suitable to various tests. The specimens were conditioned again at temperature of 20°C and 65% relative humidity for 72 h. Testing of specimens was carried out



(A)

(B)

Figure 1 SEM images of reed fibers.

according to EN standards. For mechanical properties, modulus of rupture (MOR) and modulus of elasticity (MOE) were measured according to EN 310, 1993,[32] and internal bond (IB) was according to EN 319, 1993,[33] by using LLOYD INSTRUMENTS LR 10K universal testing machine. Physical properties, water absorption (WA) and thickness swelling (TS), were determined according to EN (317–1993).[34] Five specimens from each sample were tested, and results were averaged.

RESULTS AND DISCUSSION

Scanning electron microscopy

Figure 1 shows the SEM pictures of the reed fibers. The pictures are magnified 370,000 and 600,000 times, as shown in Figure 1(A, B), respectively. The pictures show that the fibers are cylindrical, which permits greater surface area exposed to the resin used; consequently more bonding between the fibers and the resin is expected.

Urea formaldehyde resin-bonded reed particleboard

Since the bond quality is mainly affected by the pressing conditions, and since the curing of the resin is disturbed on using severe conditions, the pressing conditions must be adjusted to obtain boards with optimum mechanical and physical properties. After adjustment of these conditions, substitution of UF resin with the modified natural adhesive is carried out to obtain boards with comparable properties with the conventional UF-bonded reed particleboard and with standard requirements.

The effects of different pressing parameters, i.e., UF resin concentration (8%, 10%, and 12%), pressing time (8, 12, and 16 min.) and pressing temperature (130, 145, and 160°C) were studied to furnish samples with maximum mechanical and physical properties

Mechanical properties

Figure 2 represents the dependence of mechanical properties (MOR, MOE, and IB) on the UF concentra-

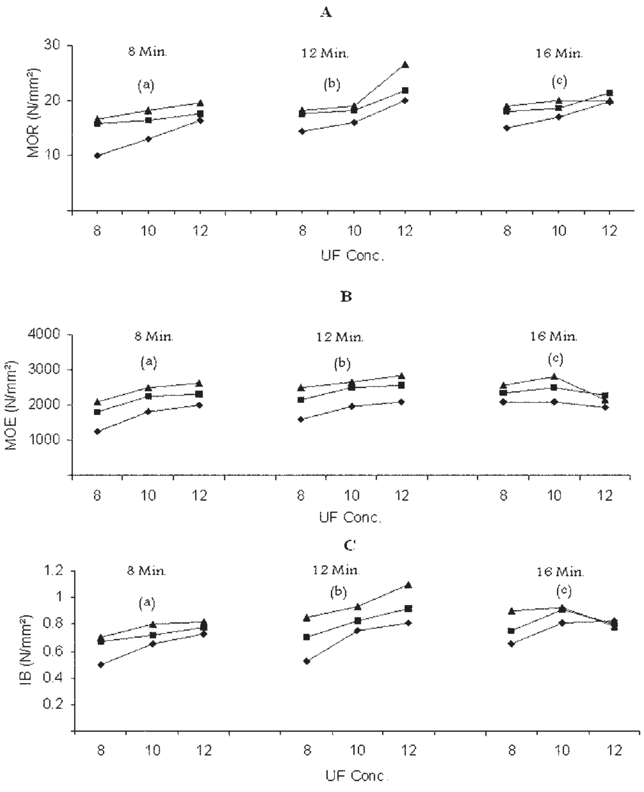


Figure 2 UF concentration dependence of MOR (A), MOE (B), and IB (C) of particleboard prepared under various pressing conditions for 8 (a), 12 (b), and 16 (c) min at temperatures 130°C (◆), 140°C (■), and 160°C (▲).

tion of boards prepared under various pressing conditions. From Figure 2, it is clear that increasing UF content, pressing time, or pressing temperature caused significant improvement in the mechanical properties.

MOR values range from 10.1 to 26.6 N/mm^2 [Fig. 2(a)]. MOE values range from 1324 to 2830 N/mm^2 [Fig. 2(b)]. The IB values range from 0.498 to 1.1 N/mm^2 [Fig. 2(c)]. All the particleboard samples showed to have mechanical properties higher than the EN standards. These previous results show that UF-bonded reed particleboard samples possessing UF concentration of 12%, prepared under pressing time of 12 min and pressing temperature of 160°C, possessed maximum mechanical properties. This improvement may be ascribed to the spreading of the adhesive within the particles over wide particle surface as well as in the curing of the adhesive.[35–37] On the other hand, upon increasing pressing time to 16 min at 160°C using 12% resin concentration, the MOE and IB properties were deteriorated. This deterioration in the board properties can be attributed to the degradative effect and disintegration of resin under such conditions.

Physical properties

Figure 3 represents the dependence of the physical properties (WA and TS) of UF-bonded reed particleboard using different UF concentration, prepared under different pressing temperatures and pressing times. Conditions that furnished high mechanical board properties were suitable for improving the physical properties, i.e., low water absorption and low thickness swelling. This expected coincidence is based on the previously mentioned reasons in addition to the presence of 1%, wax which is a water repellent agent.

Modified adhesive binder (UF/G)-bonded reed particleboard

The adhesive strength of protein glue depends on the ability to disperse in water and on the interaction of nonpolar and polar groups of the protein with wood material. In a native protein, the majority of polar and nonpolar groups are not accessible because of the internal bonds resulting from Van der Waals forces, hydrogen bonds and hydrophobic interactions. For this reason protein glue is a poor adhesive, and chemical changes are required to break the internal bonds and uncoil or disperse the polar protein molecules so as to possess superior properties to the unmodified protein.[38] The alkali was used primarily to create strong polar groups, such as amide, hydroxyl, and carboxyl groups resulted from the hydrolysis of protein molecules producing peptide chains with suitable molecular weight. Thus, NaOH was expected to unfold protein molecules and expose them to available polar groups for stronger adhesion.[39] Urea is an important denaturation agent that can unfold the secondary helical structure, as well as tertiary and quaternary structures of a protein. Its oxygen and hydrogen atoms can actively interact with hydroxyl groups in WG. This reaction breaks down the hydrogen bonding in the protein body and unfolds the protein complex, resulting in enhanced adhesion bonding strength.[40]

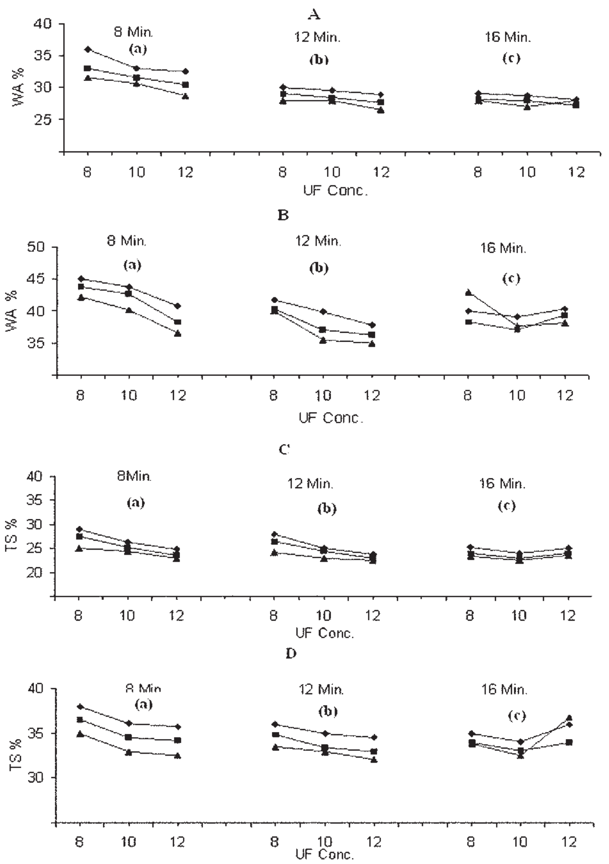


Figure 3 UF concentration dependence of WA (A and B), and TS (C and D) after 2 and 24 h, respectively of particle board prepared under various pressing conditions for 8 (a), 12 (b), and 16 (c) min at temperatures 130 (◆), 145 (■), and 160°C (▲).

Urea-formaldehyde was added to gluten in different proportions namely 80/20, 60/40, 40/6, 20/80, and 0/100, keeping the resin content 12%, based on the dry weight of reed, pressing temperature 160°C, and pressing time 12 min. Gluten was modified before mixing with UF using urea of 0.5 and 1.5*M* concentration. For each urea concentration, 1%, 1.5%, 2%, 3%, and 4% NaOH was used, based on the amount of gluten. The mechanical properties of the produced boards are shown in Figure 4, and the physical properties in Figure 5.

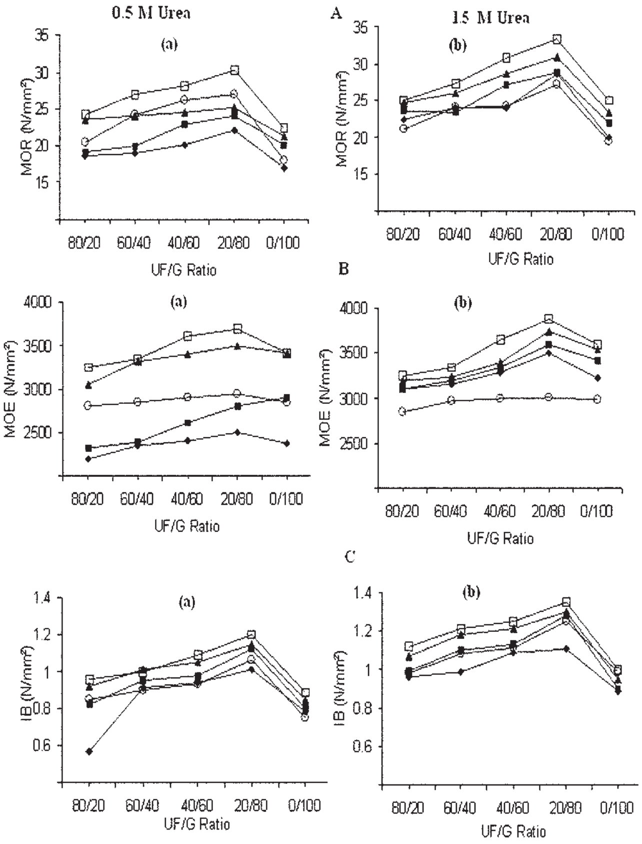


Figure 4 Dependence of MOR (A), MOE (B), and IB (C) on the UF/G ratio of particleboard prepared using different concentrations of urea 0.5*M* (a) and 1.5*M* (b), and NaOH 1% (♦), 1.5% (■), 2% (▲), 3% (□), and 4% (○).

Effect of modified adhesive binder (UF/G) on the mechanical and physical properties

Figure 4 represents the general pattern of the effect of the UF/G ratio on the mechanical properties (MOR, MOE, and IB) using various concentrations of NaOH and urea. Increasing the NaOH concentration increases the MOR, MOE, and IB of the particleboard samples; this increase is pronounced till 3% NaOH. The improved properties are due to the presence of the strong polar groups, resulted from the effect of NaOH. For urea-modified gluten, the mechanical properties of particleboard increase as the concentration of urea increases. Significant improvement of mechanical properties was observed using 1.5*M* urea. On using different UF/G ratios, an improvement of mechanical properties was obtained till UF/G adhesive binder ratio 20/80. This high gluten ratio shows that modified gluten is responsible for the adhesion character of the used adhesive binder because of the above-mentioned reason. The results also showed that using a natural adhesive alone, i.e., zero UF concentration is not recommended for bonded reed particleboard. MOR, MOE, and IB range from 18.6 to 30.34, 2200 to 3690, and 0.567 to 1.2 N/mm^2, respectively, for 0.5*M* urea at different concentrations of NaOH and adhesive binder proportion. The values of the same mechanical properties on using 1.5*M* urea were 21.5–33.37, 3097–3880, and 0.962–1.325 N/mm^2, respectively, at different concentrations of NaOH and adhesive binder proportions. MOR standard is 14 N/mm^2 for general purpose (EN, 312–2, 1996)[41] and 15 N/mm^2 for interior fitments (including furniture) (EN, 312–3, 1996).[42] MOE standard is 1950 N/mm^2 for interior fitments (including furniture) (EN, 312–3, 1996)[42] for use in dry conditions, and the IB standards are 0.31 N/mm^2 for general-purpose boards, 0.45 N/mm^2 for interior grade type according to EN, 312–2 (1996)[41] and EN, 312–3 (1996),[42] respectively. All the produced samples showed better values when compared with EN standard values.

The same conditions for gluten modification and using the same UF/G ratio, which produced samples possessing high mechanical properties, results in samples with improved WA and TS after 2 and 24 h. The results are shown in Figure 5. This may be because WA and TS were affected by bond quality[43] and adhesive properties, as well as the presence of water repellent agent (wax). Gluten contains relatively small

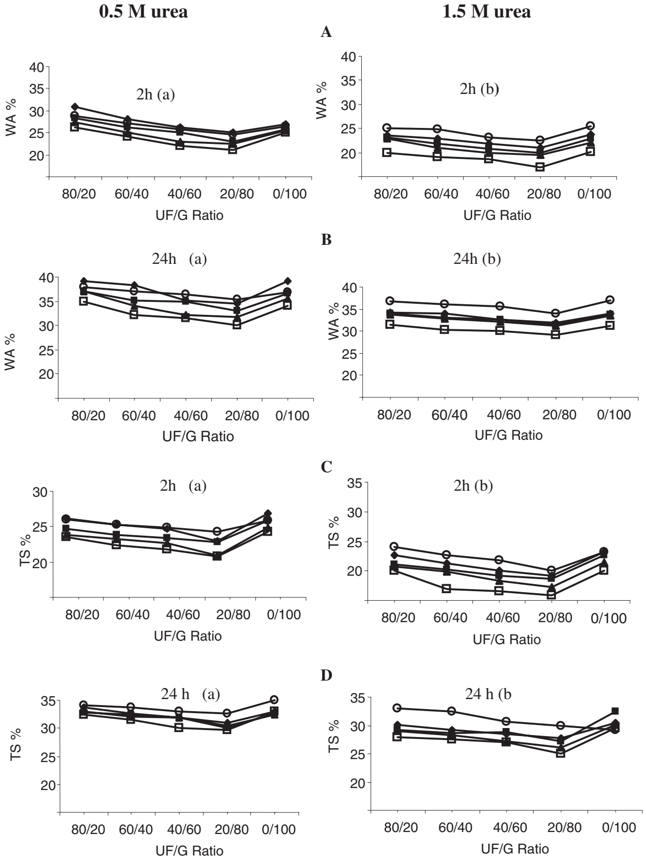


Figure 5 Dependence of WA (A and B), and TS (C and D) after 2 and 24 h, respectively, on the UF/G ratio using different concentrations of urea 0.5*M* (a) and 1.5*M* (b), and NaOH 1% (♦), 1.5% (■), 2% (▲), 3% (□), and 4% (○).

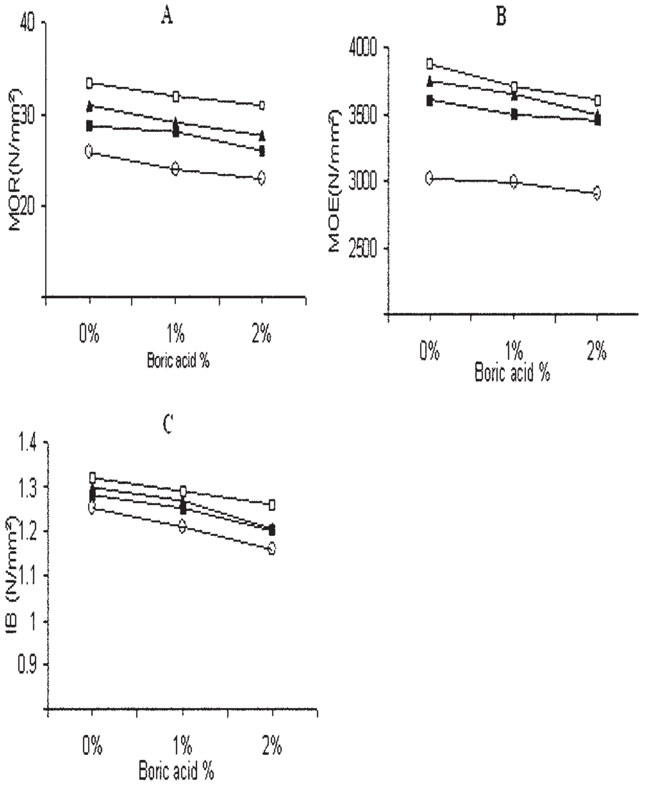


Figure 6 Dependence of MOR (A), MOE (B), and IB (C) on the boric acid concentrations of particleboard prepared using 1.5*M* urea and different concentrations of NaOH 1.5% (■), 2% (▲), 3% (□) and 4% (○).

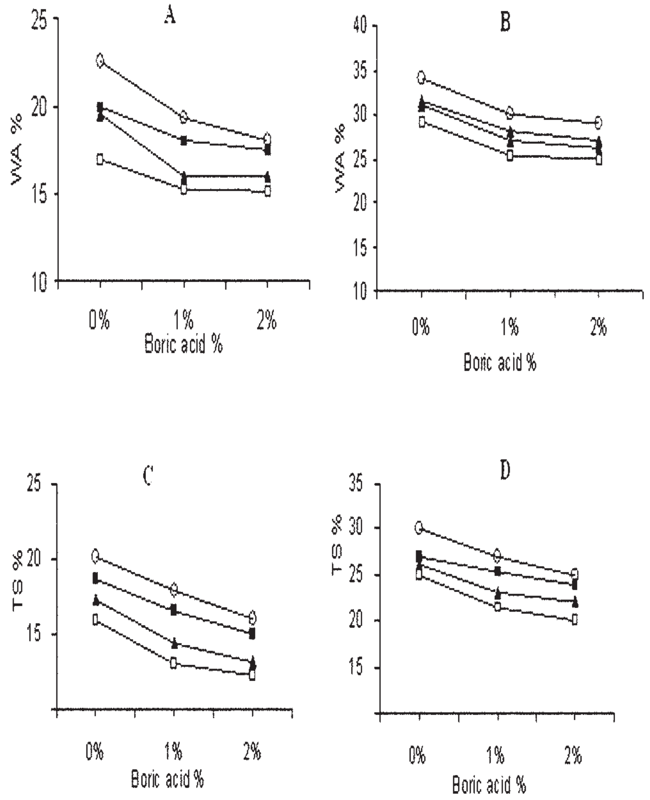


Figure 7 Dependence of WA after 2 and 24 h (A and B) respectively, and TS after 2 and 24 h (C and D) respectively, on the boric acid concentrations of particleboard prepared using 1.5*M* urea and different concentrations of NaOH 1.5% (■), 2% (▲), 3% (□), and 4% (○).

ratio of carbohydrate, compared with other proteins, e.g., soy protein (35% carbohydrate),[24] which is responsible for the water absorption. Results show that WA and TS values after 2 and 24 h ranged from 30.1% to 25% and from 22.7% to 15.9%, respectively, using 1.5*M* urea and different concentrations of NaOH and adhesive binder proportions. It is clear from the results that all the boards have comparable TS to the standard values. This is due to contribution of enhanced adhesion bonding of gluten, bonding of the cured UF resin, and the water repellent agent (wax).

Effect of boric acid on the mechanical and physical properties

Some selected samples were chosen to test the effect of boric acid addition, as an insecticide and fungicide,[44] on the mechanical and physical properties. 1% and 2% boric acid, based on the weight of gluten, was added to the 20/80 UF/G-bonded reed particleboard, which possesses the optimum mechanical and physical properties at 1.5*M* urea under different concentrations of NaOH. Figures 6 and 7 reproduce the results. From the two figures, Addition of 1% and 2% boric acid to the binder mixture resulted in no significant change in the mechanical properties and slight improvement in the physical properties of the produced particleboard. Boric acid could interact with cellulose from reed, and small amount of carbohydrate which present in gluten

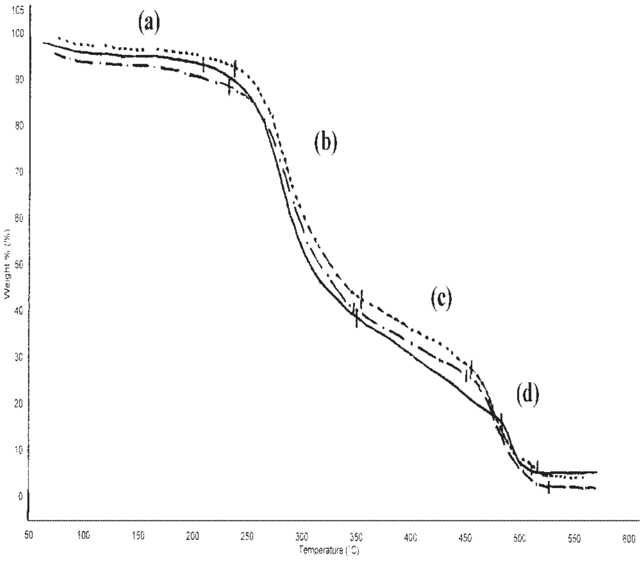


Figure 8 TGA curiives of UF-bonded reed particleboard (—), UF/G-bonded reed particleboard (. . . . . .), and UF/G-bonded reed particleboard with 1% boric acid (− −).

and crosslinks might occur among the complexes by interacting with those long-chain molecules in the amorphous region in cellulose and hold cellulose together much tighter. This inhibits the penetration of water molecules into the complex, reducing water absorption.[24]

Thermogravimetric analyses

Thermogravimetric analyses (TGA) of some selected samples A, B, and C were done to study the stability of the UF-bonded reed particleboard (A) as well as the UF/G-bonded reed particleboard (B, C), where sample B was of UF/G ratio 20/80, 3% NaOH and 1.5*M* urea, and sample C possesses UF/G ratio 20/80, 3% NaOH, 1.5*M* urea and 1% boric acid. The TGA diagram of the samples is shown in Figure 8, where the TGA curves of the investigated samples exhibit four main weight-loss stages, denoted as stages a, b, c, and d. Stage a, which is accompanied by a slight decrease in weight, is ascribed to the evaporation of water and easily volatile materials. The main degradation processes, Stages b and c, are attributed to pyrolysis and the evaporation of large amounts of the pyrolytic products. The onset temperatures of the weight loss stage b were 209.11, 237.56, and 231.66°C for samples A, B, and C, respectively. This increase in the onset degradation temperature of the UF/G-bonded reed particleboard than UF-bonded reed particleboard is again attributed to the inclusion of polar hydroxyl, amide, and carboxyl groups, which leads to an increase in the bonding strength between the binder and the fiber chains; consequently more energy is needed for degradation. Sample B, which possesses maximum mechanical and physical properties, shows significant stability toward decomposition. From the figure, we can conclude that addition of boric acid did not significantly deteriorate the thermal stability of the UF/G-bonded reed particleboard because of the mentioned reasons. On the other hand, the onset degradation temperatures of Stage c were 350.68, 352.88, and 351.94°C for samples A, B, and C, respectively. These temperatures are nearly equal. The onset degradation temperatures of Stage d were 490, 455.5, and 449.66°C for samples A, B, and C, respectively. The residual masses remaining at the end of the degradation process were 4.746%, 5.117%, and 3.32%, for samples A, B, and C, respectively. The temperature at which the degradation process ended (called ash formation temperature were 517.31, 523.96, and 531.09°C, for samples A, B, and C, respectively.

CONCLUSIONS

Modified wheat gluten can partially substitute UF resin, up to 80%, in particleboard made from reed. The mechanical and physical properties of the thus formed samples, with or without boric acid, meet the standard requirements. Using modified wheat gluten in bonded reed particleboard results in an improvement in thermal stability.

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