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Processing of urea-formaldehyde-based particleboard from hazelnut shell and improvement of its fire and water resistance

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SUMMARY

The purpose of this study was to manufacture urea-formaldehyde-based particleboard from hazelnut shell and eliminate its disadvantages such as flammability, water absorption, swelling thickness by using fly ash and phenol-formaldehyde. Synthesized urea-formaldehyde and grained hazelnut shells were blended at different ratios ranging from 0.8 to 3.2 hazelnut shell/urea-formaldehyde and dried at 70°C in an oven until constant weight was reached. In addition, other parameters affecting polymer composite particleboard from hazelnut shell and urea-formaldehyde were investigated to be the amount of fly ash, amount of phenol formaldehyde and the effects of these parameters on bending stress, limit oxygen index, water absorption capacity and swelling in the thickness. The optimization results showed that the maximum bending strength was $4.1\,\mathrm{N/mm^2}$, at urea-formaldehyde ratio of 1.0, reaction temperature of 70°C, reaction time of 25 min, hazelnut shell/urea-formaldehyde resin of 2.4 and mean particle size of 0.1 mm. Although the limited oxygen index and smoke density of composite particleboard without fly ash has 22.3 and 1.62, with fly ash of 16% (w/w) according to the filler has 38.2 and 1.47, respectively. Water absorption and increase in the swelling thickness exponentially decreased with increasing phenol formaldehyde. Copyright © 2009 John Wiley & Sons, Ltd.

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KEY WORDS: particleboard; flame retarder; hazelnut shell; fly ash; swelling

1. INTRODUCTION

Generally, particleboards, hardboard and fiberboard are among the most popular materials used in interior and exterior applications such as floor, wall and ceiling panels, office dividers, bulletin boards, cabinets, furniture, insulator, coater, counter tops and desk tops [1]. They are conceived and

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developed in research and development centers and in pilot plants. The producing of particleboards can be related to the decided economic advantage of low-cost wood raw material and inexpensive agent and simple processing. In addition, hazelnut is abundantly available in Turkey with their shells having no economical value other than being used as very low grade fuel. Thus, agricultural residues instead of wooden particles are used extensively for producing of boards. Among the raw materials are almond shell [2], pomace [3, 4], apricot shell [5], kiwi prunings [6], wheat straws [7], bamboo [8], cotton seed hulls [9], flax shiv [10], rice straw-wood [11], vine prunings [12], coir pith [13] and wood flour [14]. Besides agricultural wastes, waste cardboards containing aluminium have been used in order to manufacture low density boards [15]. On the other hand, polymers such as urea formaldehyde, phenol formaldehyde, melamine formaldehyde, polyethylene and polyvinylidene are commonly used as a binder. Urea formaldehyde is the most economic and useful adhesive among these binders. These developments are also ecologically sound, as is previously wasted material now and can be used for manufacturing useful products, thereby conserving our natural resources.

Although particleboards have economical and ecological advantages, they have three important disadvantages such as flammability, water absorption and wood-attacking insects. The disadvantages limit the extensive usage of boards and wooden materials. In order to decrease their flammability, either fire retarders can be dropped during the manufacturing process, or mineral agents can be added into the finish. Alternatively, non-flammable binders can be used or non-flammable coating compounds can be applied to the surface. These methods can be used individually or jointly. Boric acid, ammonium phosphates and borates, ammonium sulfate and chloride, zinc chloride and borate, phosphoric acid, dicyanodiamide, sodium borate, antimony oxide, magnesium hydroxide and halogenide compounds are used as flame resistant agents. Some of them, have been forbidden by EPA due to their toxicity. In addition, vermiculite, perlite, gypsum and cement are used as flame retarders [16]. Other alternative binders, optional to synthetic resins, used for production of fire-resistant boards are calcium silicate, magnesite or other mineral bonding agents [17]. Three-layer boards with mineral particles in the face layers resistant to flame are manufactured [18].

The purpose of this study was to determine the effects of hazelnut shell/urea-formaldehyde ratio on the hardness and bending strength of composite particleboard produced from hazelnut shell. In addition, coal powerplant fly ash was added into the particleboard composition as a flame retarder instead of chemicals or natural minerals. Also, synthesized phenol-formaldehyde resin was mixed into the urea formaldehyde and hazelnut shell blends, in order to prevent water absorption and swelling thickness.

2. MATERIALS AND METHOD

The chemicals employed for the production of urea formaldehyde and phenol formaldehyde binder were 37% formaline solution, 99.99% sodium hydroxide, urea and phenol (Merck). Concentrated NaOH solution was used as a catalyst in the reaction. The experimental set-up for the urea formaldehyde and the phenol formaldehyde reactions consisted of a spherical flask reactor, a reflux condenser, a thermometer and a heating mantle with a magnetic stirrer. A 1000 ml three-necked volumetric pyrex flask was used as a reactor. Hazelnut shell was used throughout the study as a reinforcement material. These shells were ground, sieved and dried in an oven at 105°C for about 1 h. Twenty five percent (w/w) of ammonium sulfate according to the binder was added into the

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mixture of resin and shell as a hardener. The mixture of binder and filler was pressed in the cast at 80°C temperature and at 100 atu pressure for 15 min by means of hydrolic press.

Optimum conditions of urea formaldehyde and phenol formaldehyde reactions were obtained from earlier studies [2, 3]. Reaction temperature was controlled within ± 2°C by means of thermostat. After the polymerization reactions had been completed at specified temperature and time, hazelnut shell was added into the polymer until it was saturated and the mixture was then cast into a mould and dried for one day at atmospheric conditions. The cast material was removed from the mould, turned upside down and dried again for one day. Later, it was dried at 70 ± 2°C temperature in an oven until constant weight was reached. In order to see how the manufacturing conditions affected the particleboard features, hazelnut shell/urea-formaldehyde ratio, fly ash/urea-formaldehyde ratio, phenol formaldehyde/urea-formaldehyde ratio were chosen as parameters. The non-flammability of the material in which coal fly ash was added instead of chemicals was investigated. Fly ash was supplied from Yatağan-Muğla coal power plant in Turkey. Briefly, non-flammability tests were carried out according to ASTM E 160-50 standard. In addition, water absorption capacity and swelling specifications of the plates produced were determined according to EN 317 and ASTM-D 1037 standards.

The mechanical properties were specified by means of bending test. Shore A hardness test was performed by means of Durotech M202 hardness tester. Bending strength from three points was done according to EN 310 standard by using Shimadzu AG-I apparatus. For three point bending test, the span length was 50 mm and the cross head speed was 10 mm/min. The dimension of the rectangular specimen was 12 mm high, 30 mm wide and 60 mm long. A minimum of three specimens for bending tests on each specimen were done in order to establish the average values. The burning characteristics of particleboards contained various amounts of fly ash determined by Dynisco limited oxygen index instrument and smoke density equipment.

3. RESULTS AND DISCUSSION

The influence of hazelnut shell/urea/fomaldehyde ratio on the mechanical properties of particleboard was researched in this study, such as urea/formaldehyde ratio of 1.0, reaction time of 20 min, mean particle size of 0.4 mm at the reaction temperature of 70 ± 2°C. In addition, it was seen that at the end of the above conditions, the transition was from transparent to cloudy stage, which is the indication of the completion of reaction [2]. It can be seen in Figure 1 that hazelnut shell/urea-formaldehyde ratio highly affects the mechanical properties of particleboard. The increase in the hazelnut shell/urea-formaldehyde ratio from 3 to 7 led to a decrease in the three points bending strength of material as a result of the decreasing binder. The increase in the hazelnut shell/urea-formaldehyde ratio from 0.8 to 2.4 led to a rise in three points bending strength of composite material as a result of increasing reinforcement material. It was recorded that the bending strength of the particleboard was 4.1 N/mm^2 above the optimum conditions. The rising trend reversed at higher ratios because of insufficient binder. The decreasing trend resulted from a domination of poor adhesion between particle and urea formaldehyde resin. Trial-and-error experiments to find out the appropriate hazelnut/urea-formaldehyde ratio showed that, hazelnut/urea-formaldehyde ratio of 2.4 corresponding to the highest strength value was a reasonable choice and it was recorded as a constant parameter in further experiments. In addition, hardness of this optimum material was determined as 98.2 Shore A. It is believed that the recording of the higher hazelnut shell/urea

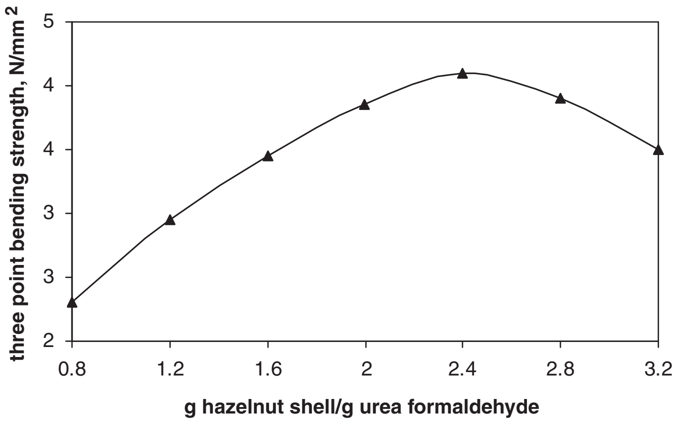

Figure 1. Effect of hazelnut shell/urea-formaldehyde ratio on the three points bending strength of particleboard.

Table I. XRD analysis of Muğla-Yatağan coal powerplant fly ash.

Compounds		(w/w) %
Silica, SiO_2		47.08
Metal oxide, R_2O_3		29.47
Titanium dioxide, TiO_2	(R_2O_3)	01.03
Ferric oxide, Fe_2O_3	(R_2O_3)	06.48
Alumina, Al_2O_3	(R_2O_3)	21.96
Calcium oxide, CaO		16.03
Magnesium oxide, MgO		02.12
Sodium, oxide, NaO		01.28
Potassium oxide, K_2O		00.93
Sulfur trioxide, SO_3		02.13
Phosphor pentoxide, P_2O_5		00.14

formaldehyde ratio than in the previous study was caused due to casting by pressure method instead of freely [2].

The addition of fly ash to the particleboard affects the burning characteristics of particleboards [18]. In order to research the influence of fly ash on the combustion characteristics of particleboards, the experiments were carried out with five particleboards containing fly ash up to 16% (w/w) as a part of filler materials (fly ash+hazelnut shell). XRD analysis results of Muğla-Yatağan coal powerplant fly ash, which was used as a flame retarder were given in Table I.

The percentages of limit oxygen were recorded for each sample, as can be seen from Figure 2. Limit oxygen values increased from 22.3 to 33.8 with increasing amount of fly ash as a part of filler in the particleboard from 0 to 16% (w/w). Smoke density was decreased from 1.62% to 1.47% with increasing fly ash percent. It can be deduced from the burning test results that fly ash can be used in the particleboard as a flame retarder instead of chemicals or mineral agents [17].

The greatest disadvantage of paticleboards is the effect of water. In order to eliminate water absorption of particleboards, synthesized phenol formaldehyde was used as a binder together

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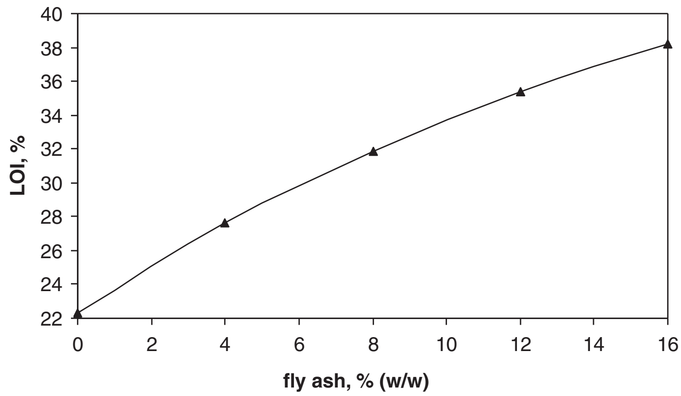

Figure 2. Effect of fly ash on the limited oxygen index of particleboard; 2.4 g filler/g urea formaldehyde.

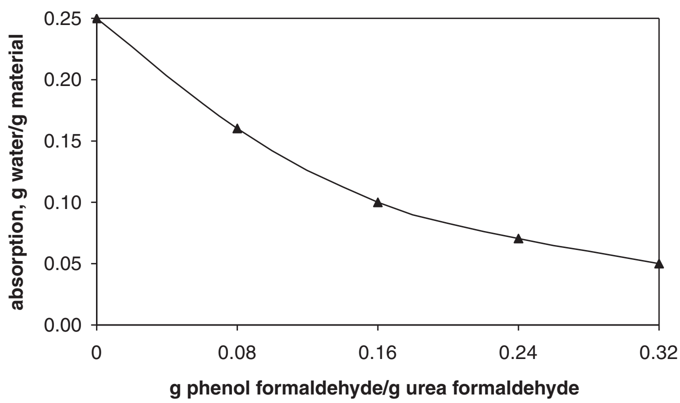

Figure 3. Effect of phenol formaldehyde/urea-formaldehyde ratio on the water absorption capacity of particleboard; 2.4 g hazelnut shell/g binder.

with urea formaldehyde at different ratios. The phenol formaldehyde/urea-formaldehyde ratio was increased up to 0.32 (w/w) to determine the water absorption capacity and increase in the swelling thickness at which the hazelnut shell/binder resin ratio of 2.4 by mass. Depending on the ratio of each trial, the amount of phenol formaldehyde is substituted with the same amount urea formaldehyde by mass. As illustrated in Figures 3 and 4, phenol formaldehyde/urea-formaldehyde ratio reversely affected the absorption capacity and swelling thickness of particleboard. The increase in the ratio up to 0.32 led to a decrease exponentially from 0.25 to 0.05 (w/w) in the water absorption capacity and from 0.210 to 0.033% in the swelling thickness of the composite material.

It can be deduced from these results that phenol formaldehyde can be mixed with urea formaldehyde to decrease water absorption capacity and swelling increase in the thickness of particleboards. It is thought that these hydrofobic properties result from cross link binding in resin and improving of resin filler adhesion [19].

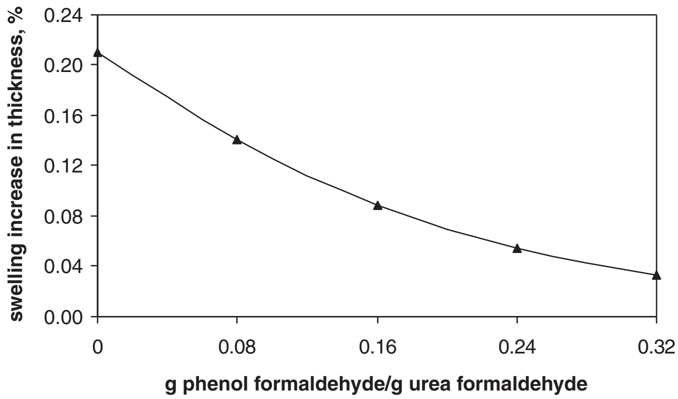

Figure 4. Effect of phenol formaldehyde/urea-formaldehyde ratio on the swelling increase in the thickness of particleboard; 2.4 g hazelnut shell/g binder.

4. CONCLUSION

It is concluded that, urea-formaldeyde-based composite particleboard can be produced from hazelnut shell, which has no economical value other than being used as very low grade fuel at the optimum conditions determined. The optimization results showed that the maximum bending strength was 4.1 N/mm^2, at urea-formaldehyde ratio of 1.0, reaction temperature of 70°C, reaction time of 25 min, hazelnut shell/urea formaldehyde resin of 2.4 and mean particle size of 0.1 mm. Fly ash, which creates air pollution can be used as a flame retarder instead of chemicals or mineral agents in the composite material. Thus, it prevents environmental pollution. Although the limited oxygen index and smoke density of composite particleboard without fly ash has 22.3 and 1.62, with fly ash of 16% (w/w) according to the filler has 38.2 and 1.47, respectively. In addition, phenol formaldehyde can be utilized as a water resistant. Water absorption and increase in the swelling thickness exponentially decreased with increasing phenol formaldehyde.

Waste raw material and low operating cost make this study a promising one for technological application. Optimum conditions recorded in this work may be applied to industrial processes economically and are environment friendly. Obtained polymer composite particleboard can be recommended for use in interior and outer applications in the fire risk and high humidity media securely and economically.

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