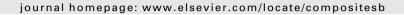
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The potential for using walnut (Juglans regia L.) shell as a raw material for wood-based particleboard manufacturing

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ABSTRACT

In this study, the suitability of walnut shell as a renewable agricultural residue for panel manufacturing was investigated. Particleboards containing different walnut shell particle ratios (0%, 10%, 20%, 30%, 40% and 100%) were made using urea-formaldehyde (UF) resin. Some chemical properties of walnut shell (holocellulose, α cellulose, lignin and ash contents, alcohol-benzene solubility, 1% NaOH solubility, hot and cold water solubility), mechanical (modulus of rupture, modulus of elasticity and internal bond strength) and physical properties (thickness swelling and water absorption) of the particleboard were determined. The addition of walnut shell particles greatly improved the water resistance of the panels. However, flexural properties and internal bond strength decreased with increasing walnut shell particle content. The results indicated that panels can be manufactured utilizing walnut shell particles up to 20% without falling below the minimum EN Standard requirements of mechanical properties for general purpose use. Conclusively, walnut shell, an annual residue, could be utilized with mixture of wood particles in the manufacture of particleboard used for outdoor environments due to lower thickness swelling and water absorption.

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1. Introduction

Increased demand for raw materials in the forest industry as a result of population growth and depletion of the natural resources, have directed researchers towards non-wood bio-based alternatives for composite manufacturing [12,13]. Agricultural residues are excellent alternative materials to wood because they are inexpensive, easily processed, plentiful, and renewable [2,27].

Some of the problems associated with industrial usage of agricultural residues in the forest industry include the high cost of collection, transportation and storage that can be overcome by establishing small-scale plants close to rural areas [10]. A selection of agricultural residues have been successfully used in particleboard manufacturing [9] and recent advances in the particleboard industry in the United States show a bright outlook for bio-based particleboards and plants [7].

Iran as a developing country does not have sufficient raw material to supply its forest industry demands. Consequently, several researchers in Iran have investigated the suitability of agro-based residues; canola straw [47], wheat straw [44], almond shell [34], and bagasse [43] in the production of wood-based composites.

Walnut (Juglans regia L.) is an important crop that is cultivated throughout the world's temperate regions for its edible nuts [35].

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Worldwide walnut production was approximately 2.2 million tons from a total of 834 thousand ha in 2009 [22]. Since walnut shell comprises 67% of the total weight of the fruit [25], around 1.5 million tons of walnut shell is left behind each year. Walnut shell, an annual agricultural waste, is the ligno-cellulosic material forming the thin endocarp or husk of the walnut tree fruit. Farmers harvest grains and burn or otherwise dispose the residues (shell, husk, stalks, etc.), although burning agricultural residues causes serious environmental problems [23]. When there is enhanced interest in industrial utilization of agricultural residues and wastes, this can mean for farmers, a second income from their plantation [10].

Iran with a walnut production of 150,000 tons ranks as the world's third biggest producer after China and the United States [16]. Walnut shell, as a bio-waste, has no economical value or industrial usage in Iran and generally is discarded or burned in the stove in the winter. Value-added wood-based panels made from agricultural residues can be considered as an alternative solution to increased demand for raw materials in the forest industry. Walnut shell could play an important role in the manufacture of value-added wood-based panels such as particleboards and may be more efficient use of the walnut shell. Guru et al. [14] studied the suitability of walnut shell for production of polymer based composite. Up to now, there is no information on using walnut shell with mixture of wood particles in production of particleboard. Therefore, the aim of this study was to investigate the suitability of walnut shell in production of three-layer particleboard as

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a supplement and to alleviate the shortage of raw material in forest industry.

2. Material and methods

The raw material of this study included industrial wood particles, and consisted of industrial wood particles (mixed hardwood species such as hornbeam, beech and oak) that was provided by a commercial particleboard plant in Gorgan, Iran and walnut (*Juglans regia* L.) shell, that was collected from the field after the walnut harvest in Sfejir, Iran. The adhesive was urea–formaldehyde (UF) which was produced by a local plant with characteristics given in Table 1.

Walnut shells were first cleaned of dirt and impurities, and then chipped by a knife ring flaker. Next, the chipped shells and wood particles were classified in laboratory shaker. Particles that remained between 3-1.5 mm sieve and 1.5-0.8 mm sieve were utilized in the core and outer layers, respectively. Particles were oven dried at 100 ± 3 °C to reach the target moisture content (3%). Urea formaldehyde (UF) resin at 9% and 11% levels (based on oven dry weight) was used for the core and outer layers respectively. One-percent ammonium chloride (NH₄Cl) was also added to the resin as a hardener. Particleboard panels were manufactured using standardized procedures that simulated industrial production at the laboratory. The particles were placed in a drum blender and sprayed with urea formaldehyde and ammonium chloride for 5 min to obtain a homogenized mixture. Panels were designed consisting of 35% particles at the face layers and 65% at the core layer. The resinated particles then were pressed into panel mat using a laboratory scale hydraulic hot press (OTT, Germany). Thickness of panels was controlled by stop bars and panels target density was 0.7 g/cm³. Three panels were produced for each group. The experimental design is shown in Table 2. The dimensions of the produced particleboards were $42 \times 42 \times 1.6$ cm. The produced particleboards conditioned at 20 °C and 65% relative humidity to reach moisture content of about 12% before trimming to final dimension of $40 \times 40 \times 1.6$ cm.

The panel production parameters were also displayed in Table 3.

Chemical properties of the walnut shell were determined and specimens were sampled and prepared according to Tappi T 257 cm [42] Standard. Holocellulose and cellulose contents were determined according to the chloride method [45]. The lignin T 222 om [41] and ash T 211 om [39] contents were also measured. Alcohol-benzene T 204 cm [37], hot and cold water T 207 om [38] and 1% NaOH T 212 om [40] solubility were determined.

Some mechanical properties; modulus of rupture (MOR) [18], modulus of elasticity (MOE) [18] and internal bond strength (IB) [20] and physical properties; thickness swelling (TS) and water absorption (WA) [19] were determined for the produced particle-boards. The average of 10 and 20 measurements were reported for mechanical and physical properties respectively. The data obtained was statistically analyzed using analysis of variance (ANO-VA) and Duncan's mean separation tests.

Table 1 Properties of the UF adhesive.

Properties	UF ^a
Solid (%) Density (g/cm³) pH Viscosity (cps) Free formaldehyde (%)	63 1.273 7.6 64 0.15
Gel point (100 °C)	55

^a Urea-formaldehyde.

Table 2Experimental design.

Board type ^a	Raw material		
	Walnut shell (%)	Wood (%)	
A	0	100	
В	10	90	
C	20	80	
D	30	70	
E	40	60	
F	100	0	

 $^{^{\}rm a}$ The density of the boards made from walnut shell and wood chips was 0.70 g/cm $^{\rm 3}.$

Table 3Production parameters of particleboards.

Parameter	Value
Press temperature (°C)	180
Pressing time (min)	5
Peak pressure (kg/mm ²)	25
Thickness (mm)	16
Dimensions (mm)	420×420
33% NH ₄ Cl content (%)	1
Outer layer (whole of board%)	35
Middle layer (whole of board%)	65
Number of boards for each type	3

3. Results and discussion

Certain chemical properties of the walnut shell and some other crop residues as well as hard/softwoods are listed in Table 4. A comparison between walnut shell and other crops; peanut hull [12], hazelnut hull [10], cereal straw [21], cotton carpel [1] and softwoods and hardwoods [36], indicated that walnut shell had the lowest holocellulose (46.6%) and α cellulose (25.4%) contents. In terms of lignin, walnut shell had the highest content (49.1%) compared to the other crop residues as well as hardwoods and softwoods. Ash content of walnut shell was similar to cereal straw but much higher than hardwoods and softwoods. Alcohol-benzene solubility was close to cereal straw but lower than peanut hull and wood species. 1% NaOH solubility was similar to peanut hull and cereal straw but higher than wood species. Hot water solubility was close to peanut hull and cereal straw and higher than hardwoods and softwoods. Finally, cold water solubility of walnut shell was close to cereal straw and higher than softwoods and hardwoods.

Table 5 shows the results of mechanical properties of produced panels. The highest MOR (16 N/mm²) and MOE (2309 N/mm²) values were measured for particleboard produced using industrial wood particles. Besides, the lowest MOR (5.8 N/mm²) and MOE (1152 N/mm²) values were determined for panels type F, including 100% walnut shell. The result indicated that the increasing the walnut shell content in the mixture significantly decreased the MOR and MOE values of the particleboards. Statistical analysis found some significant differences (P < 0.01) between some groups means for MOR and MOE values. Significant differences between groups were determined individually for these tests by Duncan's multiple comparison tests. The result of Duncan's multiple range tests are shown in Table 5 by letters. All panel types showed statistically meaningful differences (P < 0.01) in their MOR and MOE properties from each other. Depending on the amount of walnut shell particle in the particleboards, average decreases in MOR values varied from 8.1% to 63.8% as compared to average of the panel type A (Fig. 1). MOE values showed similar trends to result of the MOR. The average MOE values of the particleboard with walnut shell particles decreased from 2.8% to 50% as compared to values

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