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Full Length Research Paper

Utilization of bio-waste cotton (*Gossypium hirsutum* L.) stalks and underutilized paulownia (*paulownia fortunie*) in wood-based composite particleboard

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The objective of this study was to investigate some mechanical (modulus of rupture, modulus of elasticity and internal bond) and physical (thickness swelling and water absorption) properties of wood-based three-layer particleboard containing different cotton (*Gossypium hirsutum* L.) stalks and underutilized paulownia (*paulownia fortunie*) wood particle ratios (30, 50 and 70%) using urea formaldehyde resin. Addition of cotton stalk and paulownia wood in particleboard improved mechanical properties of resulting composites boards significantly. However, the water resistance decreased with increasing cotton stalk and paulownia wood particle contents. The highest mechanical properties were obtained at cotton stalk and paulownia wood particle loading of 50 and 70%, respectively. Conclusively, valuable underutilized natural resources, cotton stalk and paulownia wood can be used with the mixture of industrial wood particles in the production of particleboards with high mechanical properties.

Key words: Wood, resin, urea-formaldehyde, mechanical properties, physical properties.

INTRODUCTION

Particleboard is an engineered wood composite manufactured from wood particles, such as saw mill shavings, chips, or even sawdust, and a synthetic binder or other suitable resin, which is pressed (Akyüz et al., 2010). It used widely as component of furniture, doors and cabinets. Serious shortage of wood resources in developing countries, including Iran, have increased the importance of particleboard manufacturing from agricultural fibers and wood residues as an alternative for solid wood. Alternative raw materials such as agricultural residues, underutilized species and fast-growing species can play an important role in the particleboard industry in the future.

Recently, some researchers have focused on the use of various agricultural residues and wastes for particleboard manufacturing. Some low-cost lignocellulosic materials (LCM) used as raw material to produce composite panels from agricultural wastes had been as follows: Eggplant stalk (Guntekin and Karakus, 2008), grass clipping (Nemli et al., 2009), kiwi pruning (Nemli et al., 2003), pepper stalk (Guntekin et al., 2008), almond shell (Pirayesh and Khazaeian, 2011) and sunflower stalk (Bektas et al., 2005).

Cotton (*Gossypium hirsutum L.*) is a major crop popularly known as 'White Gold' grown primarily for fiber and oil seed all over the world (Dong et al., 2010). The cultivation of cotton generates plant residue equivalent to three to five times the weight of the fiber produced (Reddy and Yang, 2009). After harvesting the cotton balls, the entire plant, consisting stalk and leaves, is a residue which remains in the field and the farmers usually destroy it by burning (Binod et al., 2011). Burning agricultural residues causes environmental problems such as air pollution, soil erosion and decreases soil biological activity (Copur et al., 2007).

Paulownia is a fast-growing shade tree indigenous to China and South-East Asia (Bergmann, 1998). It has been highly valued for more than 2000 years in East Asia (Kalaycioglu et al., 2005). Paulownia trees have been used for different purposes such as to produce leaves useful as fodder or fertilizer, flower for honey production

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Table 1. Properties of the UF adhesive.

Property	UF[a]
Solid content (%)	60
Density (g/cm^3)	1.27
pH	7
Viscosity (cps)	63
Gel time (s)	45

[a], Urea-formaldehyde.

Table 2. Experimental design.

Panel type[a]	Particleboard composition (by weight %)		
	Industrial wood (%)	Paulownia wood (%)	Cotton stalk (%)
A	100	0	0
B	70	0	30
C	50	0	50
D	30	0	70
E	70	30	0
F	50	50	0
G	30	70	0

[a], The density of all panels was 0.70 g/cm^3.

(Yorgun et al., 2009), and wood for the production of hardwood timber (Bergmann, 1998) over the years.

Paulownia wood has also been investigated as a promising raw material for the production of wood based composites (that is, particleboard) (Kalaycioglu et al., 2005) and chemical pulp (Jiménez et al., 2005). It is soft, lightweight, ring porous, straight-grained, and mostly knot-free with a satiny luster. Its average density is about 0.35 g/cm^3 (Kalaycioglu et al., 2005). In recent years, Paulownia is receiving increasing attention as a genus suitable resource for use as a short-rotation woody species in Iran. Under the appropriate conditions, a 5- to 7-year-old tree can reach about 15 to 20 m high and the annual production is as much as 150 tons/ha (Jiménez et al., 2005). Bio-based composites will become materials to replace polymer based composites and wood in terms of their attractive specific properties, lower cost, simple process technologies, eco-friendliness, and recyclability (Ndazi et al., 2006). Guler and Ozen (2004) studied properties of particleboards made from neat cotton stalk. There is no information on using cotton stalk and paulownia wood with the presence of industrial wood particles in particleboard production.The main objective of this study was to use cotton stalk and paulownia wood as raw materials in the production of three-layer particleboard, and to evaluate physical and mechanical properties of the boards to determine if they have the required levels of properties for general uses.

MATERIALS AND METHODS

The raw material of this study included Cotton (*G. hirsutum* L.) stalk, paulownia (*Paulownia fortunie*) wood and industrial wood particles (consisting of mixed hardwood species such as beech, oak and hornbeam). Cotton stalks were harvested in an experimental field of the Iranian Cotton Research Institute located at Hashem Abad, Gorgan (Iran) and paulownia wood was harvested from a Shastcolateh experimental forest, Gorgan (Iran) and industrial wood particles were obtained from a commercial particleboard plant in Gorgan (Iran).

The cotton stalks and paulownia wood were chipped using industrial-scale drum-chipper. The chips were then reduced into smaller particles by a knife-ring flaker. After that the particles were dried to 3% moisture content and then classified in a laboratory shaker.

Industrial wood particles and particles produced from the cotton stalks and paulownia wood remained between 3 and 1.5 and 1.5 and 0.8 mm sieve were utilized in the core and outer layers, respectively.Urea-formaldehyde (UF) resin at 8 and 12% adhesive level was used for the core and outer layers based on oven-dry weight. The properties of UF resin is given in Table 1. One percent of ammonium chloride (NH_4Cl) was added to the resin as a hardener. The particles were introduced in a drum blender and sprayed with UF and NH_4Cl for 5 min to obtain a homogenized mixture. Panels were designed consisting 35% face and 65% core layers. The target density of boards was 0.70 g/cm^3. Three panels were produced for each design. The experimental design is given in Table 2. Panel production parameters are also displayed in Table 3. The panels after conditioning (at 65% relative humidity and 20°C) to reach moisture content of about 12% were tested for modulus of rapture (MOR) and modulus of elasticity (MOE) (EN 310, 1993), internal bond strength (IB) (EN 319, 1993), thickness swelling (TS) and water absorption (WA) after 2 and 24-h immersion (EN 317, 1993).

The average of 10 and 20 measurements were reported for mechanical and physical properties respectively.Data for each test were statistically analyzed using analysis of variance (ANOVA). When the ANOVA indicated a significant difference among factors and levels, a comparison of the means was done employing Duncan test to identify which groups were significantly different

Table 3. Production parameters of particleboards.

Parameter	Value
Press temperature (°C)	170
Pressing time (min)	8
Peak pressure (kg/cm^2)	30
Thickness (mm)	16
33% NH_4Cl (%)	1
Outer layer (whole of board, %)	35
Core layer (whole of board, %)	65
Number of board for each type	3

Table 4. Comparison of chemical composition of paulownia, cotton stalk and hardwoods.

Lignocellulosic material	Hemicellulose (%)	Cellulose (%)	Lignin (%)	Reference
Cotton stalk	77.50	47.80	21.20	Ertas et al., 2010
Paulownia	78.80	48.30	22.10	Kalaycioglu et al., 2005
Hardwoods	70-78	45-50	30-35	Pirayesh and khazaeian, 2011

from other groups at 99% confidence level.

RESULTS AND DISCUSSION

Mechanical properties

Table 5 shows the results of mechanical properties of the experimental panels. As seen, particleboards containing 70% paulownia wood, had the highest MOR (26.78 N/mm^2) and MOE (2976 N/mm^2) values. Besides, the lowest MOR (11.47 N/mm^2) and MOE (1576 N/mm^2) values were measured for panel type G, containing 70% cotton stalk in the mixture. The result indicates that addition of cotton stalk up to 50% and paulownia wood up to 70% improves MOR and MOE values of the boards in comparison to the reference ones, panel type A (particleboard produced using 100% industrial wood particles). Statistical analysis found some significant difference ($p<0.01$) 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. Based on EN Standards, the minimum requirements for MOR and MOE of particleboards for interior grade type (including furniture) are 14 and 1800 MPa (EN 312-3). All the panels produced in this study provide MOR and MOE values with outstanding margin exceeding EN standard requirements for interior grade type (including furniture) except for panel type G, containing 70% cotton stalk.

IB values of the experimental panels ranged from 0.36 to 0.70 N/mm^2. The highest IB value was observed for panel type D, while the lowest was observed for G type panel. IB value of the panels also decreased with increasing cotton stalk percentage in the mixture but increased with increasing paulownia wood particle ratio. The minimal requirement of internal bond strength for general purposes (EN 312-2) interior usage (EN 312-3) and load-bearing board (EN 312-4) are 0.24, 0.35 and 0.50 MPa, respectively. In general, all the boards except for panel type G met the IB requirements of EN standard for different usage. The strength properties of bio-based composites depend on many factors such as physical and mechanical properties of individual particles, their interfacial adhesion, the orientation, density (or compact ratio) and aspect ratio of particles (Ayrilmis et al., 2009; Nourbakhsh and Ashori, 2010). Besides, cellulose, lignin and hemicelluloses contents that were different for all three raw material of this study (Table 4) had strong influences on the mechanical properties (Habibi et al., 2008). Cotton stalk density (0.28 to 0.31 g/cm^3) is lower than paulownia wood (0.35 g/cm^3) and woody raw material density (0.40 to 0.75 g/cm^3) (Guler and Ozen, 2004; Kalaycioglu et al., 2005). Lower density or higher compact ratio and bulky characteristics of cotton stalk and paulownia wood particles in comparison to industrial wood particles can be one of the main reasons of the mechanical properties improvements. However, regarding cotton stalk at high loading (70%), in the gluing stage they did not glue well, relatively forming lumps resulting in an interrupted glue line between the particles and consequently lower mechanical properties.

Physical properties

The results of ANOVA and Duncan's mean separation test for TS and WA of particleboards containing cotton

Table 5. The mechanical properties of particleboards made from cotton stalk, paulownia wood and industrial wood particles and the test results of ANOVA and Duncan's mean separation tests.

Mechanical property	Board type	Mean[a]	Standard deviation	Standard error	X_{Min}[b]	X_{Max}[c]	p[d]
	A	16.02[s]	0.049	0.015	15.98	16.12	*
	B	19.50[u]	2.714	0.858	14.89	23.79	*
	C	20.03[u]	1.581	0.499	16.99	22.00	*
MOR (N/mm^2)	D	26.78[t]	1.235	0.390	24.73	28.39	*
	E	16.92[s]	1.296	0.409	14.97	18.45	*
	F	17.19[s]	0.698	0.221	16.26	18.10	*
	G	11.47[p]	0.737	0.233	10.19	12.37	*
	A	2254.30s	36.435	11.522	2300.00	2399.00	*
	B	2345.20[su]	215.886	68.269	1971.00	2571.00	*
	C	2536.30[ut]	185.677	58.716	2284.00	2890.00	*
MOE (N/mm^2)	D	2976.60[v]	195.191	61.725	2637.00	3296.00	*
	E	2357.30[su]	166.146	52.540	2128.00	2610.00	*
	F	2608.00[t]	80.209	25.364	2505.00	2733.00	*
	G	1576.00[p]	153.511	48.544	1360.00	1890.00	
	A	0.57[su]	0.032	0.010	0.53	0.62	*
	B	0.65[su]	0.108	0.034	0.53	0.85	*
	C	0.67[u]	0.064	0.020	0.55	0.75	*
IB (N/mm^2)	D	0.70[u]	0.101	0.032	0.54	0.87	*
	E	0.61[su]	0.123	0.038	0.44	0.78	*
	F	0.62[su]	0.128	0.040	0.35	0.70	*
	G	0.36[p]	0.139	0.043	0.10	0.55	*

[a], Mean values are the average of 10 specimens; [b], minimum value; [c], maximum value; [d], significance level of 0.01 (for ANOVA); [p,s,u,t,v], values having the same letter are not significantly different (Duncan test). MOR, Modulus of rapture; MOE, modulus of elasticity; IB, internal bond strength.

stalk and paulownia wood particles for 2 and 24-h water immersion times are given in Table 6. With the addition of cotton stalk and paulownia wood particle to the particleboard TS and WA values increased from 11.81 to 29.42% and 59.87 to 104.34% for 2-h water immersion time in comparison to the panel type A (particleboard produced using 100% industrial wood particles). Likewise, for 24-h water immersion time, these figures were 18.03 to 32.35% and 67.82 to 116.88%. Contrary to the mechanical properties, TS and WA values deteriorated with increasing cotton stalk and paulownia wood loading in the panels that can be related to lower density, higher porosity, of these materials in comparison to industrial wood particles than mean more voids and pores for water uptake. Besides, variation in the TS and WA values of the experimental boards can be attributed to differences in chemical composition of the raw material (Table 4) and differences in their pH value with the binders' one. Difference in hemicelluloses, cellulose and lignin ratio of raw material can result in different dimensional stability of the resulting panels (Pirayesh and Khazaeian, 2011). One of the most important chemical properties of bio-based fibers that have an important role in developing good bonding between resin and particles as well as hardening of resin is pH (Nemli et al., 2008). Inequity of pH value of raw material and binder can lead to different water absorption properties of resulting boards (Nemli et al., 2008).

Level of pH of the raw material of this study, cotton stalk 4.74 (Kargarfard and Latibari, 2011), paulownia wood 5.38 (Kalaycioglu et al., 2005), hard woods 5.96 (Nemli et al., 2008) and the resin, 7, was relatively different. Also, lack of good glue line between bulky cotton stalk and paulownia wood particles can be another reason of the lower TS and WA In general, all boards did not satisfy the TS and WA requirement of EN standard for general purpose usage. Chemical modification of agricultural fibers as well as coating particleboard surfaces with melamine-resin impregnated papers or laminates can results in more dimensionally stable panels (Azizi et al., 2011 ; Ndazi et al., 2006). Adding wax to coat the lignocellulosic fibers can improve their water resistance (Ayrilmis et al., 2009). Especially, MDI resin can perfectly bond agricultural wastes to wood fibers (Yang and Zhang, 2004). Similarly, lower physical properties have been reported for particleboards made using agricultural

Table 6. Thickness swelling (TS) and water absorption (WA) test results of ANOVA and Duncan's mean separation tests of particleboards produced from cotton stalk, paulownia and industrial wood particles.

Physical property	Board type	Soaking time (h)	Mean (%)[a]	Standard deviation	Standard error	X_{Min} [b]	X_{Max} [c]	p[d]
Thickness swelling (TS)	A	2	11.81[p]	0.338	0.107	11.20	12.28	*
	B	2	15.73[s]	0.419	0.132	15.10	16.40	*
	C	2	22.14[u]	0.773	0.244	21.30	23.37	*
	D	2	23.25[ut]	0.530	0.167	22.31	23.91	*
	E	2	24.47[tv]	1.725	1.545	21.48	26.30	*
	F	2	24.67[v]	0.889	0.281	23.17	25.81	*
	G	2	29.42[w]	1.702	1.538	27.68	32.21	*
	A	24	18.03[p]	0.316	0.100	17.40	18.40	*
	B	24	21.81[s]	1.521	0.481	19.14	23.61	*
	C	24	24.36[u]	0.702	0.222	23.15	25.20	*
	D	24	28.01[t]	0.413	0.130	27.43	28.50	*
	E	24	28.57[t]	1.458	0.461	26.02	30.19	*
	F	24	30.96[v]	1.650	0.521	28.55	33.23	*
	G	24	32.35[w]	0.721	0.228	31.02	33.19	*
Water absorption (WA)	A	2	59.87[p]	3.181	1.005	57.50	68.50	*
	B	2	64.94[s]	4.933	1.560	59.57	74.52	*
	C	2	69.72[u]	3.578	1.131	63.43	73.26	*
	D	2	74.03[t]	3.454	1.092	70.31	79.76	*
	E	2	75.70[t]	1.035	0.327	74.41	77.47	*
	F	2	76.69[t]	0.979	0.309	75.22	78.06	*
	G	2	104.34[v]	1.827	0.577	102.17	107.38	*
	A	24	67.82[p]	0.470	0.148	66.90	68.40	*
	B	24	80.04[s]	4.920	1.556	70.41	87.73	*
	C	24	86.26[u]	5.259	1.663	77.93	92.39	*
	D	24	94.19[t]	4.043	1.278	86.55	98.66	*
	E	24	92.30[t]	1.427	0.451	90.09	93.84	*
	F	24	93.83[t]	2.676	0.846	90.70	97.31	*
	G	24	116.88[v]	3.532	1.116	110.95	121.05	*

[a], Mean values are the average of 20 specimens; [b], minimum value;[c], maximum value; [d], significance level of 0.01 (for ANOVA); [p,s,u,t,v,w], values having the same letter are not significantly different (Duncan test).

wastes and residues (Copur et al., 2007; Bektas et al., 2005; Nemli et al., 2009; Guntekin and Karakus, 2008).

Conclusion

In this study, the potential of two underutilized fibrous materials for particleboard manufacturing was studied. Kinds of fibrous material and their ratios significantly influenced the mechanical properties of the panels. Addition of cotton stalk up to 50% and paulownia wood particles up to 70% improved the mechanical properties of the resulting boards; however, physical properties decreased with increasing cotton stalk and paulownia wood particles ratios in the mixture. This problem may be easily solved by the addition of paraffin to the boards. Based on initial findings of this study, it can be concluded that cotton stalk and underutilized paulownia wood could be utilized in particleboard production. Finding this type of new application area for cotton stalk and underutilized paulownia wood can lead to decreasing pressure on the forests, alleviation of raw material shortage of wood industry in developing countries and providing second income from the plantation along with environmental benefits.

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