J Wood Sci (2012) 58:513–524
DOI 10.1007/s10086-012-1278-2

ORIGINAL ARTICLE

Effect of nanoclay on some applied properties of oriented strand board (OSB) made from underutilized low quality paulownia (*Paulownia fortunei*) wood

Ayoub Salari · Taghi Tabarsa · Abolghasem Khazaeian · Ahmadreza Saraeian

Received: 30 March 2012 / Accepted: 13 June 2012 / Published online: 11 July 2012

Abstract In this study, the effect of nanoclay on some applied properties of oriented strand board (OSB) made from underutilized low quality paulownia wood was investigated. Organo-modified montmorillonite (MMT) at four levels (0, 1, 3 and 5 %) was added to urea formaldehyde (UF) resin. Some chemical properties of paulownia wood (holocellulose, cellulose, lignin and ash contents, pH value and hot and cold water solubility), mechanical [modulus of rupture (MOR), modulus of elasticity (MOE), internal bond strength, screw and nail withdrawal strengths], physical (water absorption and thickness swelling) properties and formaldehyde emission of the strand boards were evaluated. Mechanical properties of all panels complied with the general-purpose OSB minimum property requirements of European Norm. With increasing 5 % nanoclay to UF resin, mechanical and physical properties of the resulting panels improved and formaldehyde emission decreased. However, none of the panels satisfied the thickness swelling and water absorption requirement. The results of X-ray diffraction and transmission electron microscope analysis confirmed the good dispersion of nanoclay in the resulting OSBs. Using paulownia as a fast-growing underutilized species not only can sustain the forests but also can supply raw material to countries facing shortage of wood.

Keywords Paulownia wood · Applied properties · Nanoclay · Formaldehyde emission · Transmission electron microscopy (TEM)

Introduction

Environmental pressure managed to prohibit forest harvesting; consequently, wood shortage-shut down of wood industries, unemployment etc. can be seen in some countries including Iran. For instance, after closure of the largest plywood plants in Germany and France [1], Iran's plywood plants discontinued their production too. The demand for wood composite products such as oriented strand board (OSB), plywood, medium density fiberboard, hardboard and veneer products has recently increased distinguishably throughout the world [2]. Natural fiber composites are also claimed to offer environmental advantages such as reduced dependence on non-renewable energy/material sources, low density, low cost, non-abrasive nature, easiness of processing, lower pollutant emissions, lower greenhouse gas emissions, enhanced energy recovery, and end of life biodegradability of components [2–5]. Alternative raw materials such as fast-growing species, underutilized species, and agricultural residues can play an important role in the particleboard industry in the future [2].

Paulownia is an extremely fast-growing deciduous tree species with vegetative propagation and tolerance to different soil and climate conditions and is original of China and its natural distribution ranges from tropical through to cool temperate climates [6]. Paulownia is an appropriate tree for intensive management in short rotation hardwood plantations because of its rapid growth, its ability to stump sprout, in exact words, paulownia does not require replanting after harvest because it regenerates from stump sprouts [7]. Besides, the use of short rotation forestry plantations is a promising tool for reducing atmospheric carbon dioxide concentration through fossil fuel substitution [8]. It is soft, lightweight, ring porous, straight-grained, and

---

A. Salari (✉) · T. Tabarsa · A. Khazaeian · A. Saraeian
Department of Wood and Paper Technology, Gorgan University of Agricultural Sciences and Natural Resources (GUASNR), Gorgan, Iran
e-mail: salariayoub@yahoo.com

mostly knot-free with a satiny luster. Under appropriate conditions, a 5- to 7-year-old tree can reach a height of about 15–20 m and the annual production is as much as 150 t/ha [6].

Oriented strand board is a reconstituted flat pressed wood-based panel composed of oriented wood strands bonded by hot-pressing using thermosetting adhesive resins. OSB panels are a relatively new kind of composites that EN-300; 1997 categorizes them in particleboard group and distinguishes among four types of OSB panels; OSB/1 is intended for general indoor use in dry conditions [9]. It is today the main substitute panel for the rather more expensive plywood, but presenting the same advantages [10]. OSB is very commonly used in light wood frame construction, such as sheathing materials of shear walls and ceiling coverings [9, 11].

Wood panel industries rely on polycondensed thermosetting resins the one of the most important is urea formaldehyde resin [12, 13]. Low price, good technological properties, absence of colors in cured polymer and easiness of application for a variety of curing conditions are of advantages of urea formaldehyde, while, formaldehyde emission and poor water resistance are its main disadvantages [14–16]. Formaldehyde is considered as a dangerous substance, and its concentration in indoor environments is restricted in many countries because of its reactivity, toxicity, and pungent odor [17, 18]. Formaldehyde has also been found to produce nasal carcinomas in mice and rats after exposure to 14.1 and 5.6 ppm of formaldehyde, respectively, over a long period of time [19]. A drastic reduction in the formaldehyde emission with a significant improvement in the durability–stability of UF bonded wood products could extend the applications and markets for these products. One of the ways to decrease the formaldehyde emission of the UF boned panels is to use nanofillers such as nanoclay and nano-SiO_2 because these materials have strong absorbability as well as high barrier property [15, 16, 20]. Fillers have modifying effects on urea formaldehyde resin not only by decreasing resin viscosity and controlling its penetration into the wood tissue and improving bonding but also economically [13]. Using organo-modified montmorillonite (MMT) (commonly called ‘‘Nanoclay’’) to reinforce polymer-based composites have raised great attention in the academic and industrial sectors, since the addition of small amount of nanoclay could substantially enhance the mechanical properties of pristine polymers [13, 20–23]. The important characteristics pertinent to application of clay minerals in polymer nanocomposites are their richest intercalation chemistry, high strength and stiffness and high aspect ratio of individual platelets, abundance in nature, high gas barrier quality and very strong absorbability [15, 24–26].

Lei et al. [20] found that slight percentages of Na^+ montmorillonite (NaMMT) nanoclay could improve the performance of thermosetting UF resin and it had an accelerating effect on the curing of UF resin that could result in physical and mechanical improvement of wood-based composites. The effects of nanoclay as reinforcement filler on the properties of MDF were investigated by Ashori and Nourbakhsh [27]. The MOR and IB strengths of the medium density fiberboard increased with increasing the weight percentage of nanoclay. In addition, when the amount of nanoclay increased to 8 %, the thickness swelling was reduced. An optimum or synergistic usage of nanoclay in the resins is essential to achieve the highest mechanical, physical, and thermal properties as well as other tailored feature that they offer [21].

In recent years, paulownia is receiving increasing attention as a genus suitable for use as a short-rotation woody species in Iran. Evaluation of this wood as a raw material in OSB production is not common. Value-added wood-based panels made from underutilize low quality wood species employing nanotechnology can be considered as an alternative solution to increased demand for raw materials and superior products in the forest industry. Employing nanotechnology on wood and wood-based composites can lead to the next generation of wood products having hyper-performance and desirable serviceability. The initial objective of this study was to investigate the suitability of paulownia wood for OSB manufacturing. In addition, the influence of nanoclay and press cycle time on the mechanical, physical and formaldehyde emission of the resulting panels and determine if such fiber resource employing nanoclay could be used to produce OSB with acceptable properties.

Materials and methods

Materials

Strands for OSB panels were manufactured from paulownia (*Paulownia fortunei*) logs by producing 1 mm thick rotary-cut veneers from them and converting the resulting veneers to pieces with 15 mm width and 100 mm length by circular saw. Paulownia logs were harvested from Shastcolateh Educational Forest, Gorgan (Iran). The strands were dried to 3 % moisture content in an oven. The UF resin was produced by a local plant with characteristics given in Table 1. Ammonium chloride (NH_4Cl) was added as a hardener to UF resin at a level of 1 % based on oven-dry weight of resin. The commercial NaMMT nanoclay used with a cation exchange capacity (CEC) value of 92.6 mequiv/100 g was purchased from Southern Clay Products Co., USA. It was added to the resin at four levels of 0, 1, 3 and 5 % based on oven-dry weight of resin. The characteristics of NaMMT are given in Table 2. Experimental design is given in Table 3.

Table 1 Properties of the UF adhesive

Properties	UF[a]
Solid content (%)	60
Density (g/cm^3)	1.27
pH	7
Viscosity (cps)	200
Gel time (s, 100 °C)	45

[a] Urea–formaldehyde

Table 2 The characteristics of Na + montmorillonite nanoclay

Supplier designation	Closite® Na$^+$
Organic modifier	None
Density (kg/m^3)	2860
Moisture (%)	4–9
Weight loss ignition (%)	7

OSB manufacturing

The OSB panels were manufactured using standardized procedures that simulated industrial production at the laboratory. Urea formaldehyde (UF) resin was used at a level of 10 % based on oven-dry weight of wood strands to produce the panels. The UF resin was mixed with the NaMMT nanoclay by mechanical stirring for 360 s at the room temperature to get a good dispersion and partly exfoliation of NaMMT in the UF resin [20]. It seems the UF resin can also wet NaMMT and partly penetrates into the silicate layers during this time. Then, the UF resin containing well-dispersed nanoclay was sprayed into the strands using a rotary blender. The resin viscosity increased once NaMMT was added to the mixture; however, there was no significant effect on the resin spraying. The resinated strands (10 × 15 × 1 mm) 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/cm^3. Three panels were produced for each group. The experimental design is shown in Table 4. The dimensions of the produced panels were 420 × 420 × 10 mm. Ammonium chloride (NH_4Cl) was added as a hardener in UF resin at a level of 1 % based on the oven-dry weight of resin. Mats were hand formed in a forming frame and then the strand mats were manually pre-pressed. The strands mats at 10 % moisture content were exposed to the hot press. All composites were conditioned at 65 ± 5 % relative humidity and 20 ± 1 °C for about 2 weeks.

Testing methods

Chemical properties of the paulownia were determined and specimens were sampled and prepared according to Tappi T 257 cm [28] Standard. Holocellulose and cellulose contents were determined according to the chloride method [29]. The lignin T 222 cm [30] and ash T 211 om [31] contents and hot and cold water solubility T 207 om [32] were also measured. The pH value was determined according to Browning [33].

Table 3 The experimental design

Board type	Press time (min)	Strand (wt%)	UF (%)	Nanoclay (%)
A_1B_1	7	90	10	0
A_1B_2	7	90	10	1
A_1B_3	7	90	10	3
A_1B_4	7	90	10	5
A_2B_1	10	90	10	0
A_2B_2	10	90	10	1
A_2B_3	10	90	10	3
A_2B_4	10	90	10	5

Table 4 Production parameters of oriented strand boards

Parameter	Value
Press temperature (°C)	170
Peak pressure (kg/cm^2)	30
Thickness (mm)	10
Dimensions (mm)	420 × 420
Number of boards for each type	3

Mechanical tests were performed on a Universal Instron testing machine with a load capacity of 20000 N. Physical and mechanical tests on the experimental OSBs were conducted in accordance with appropriate European Norm (EN) and ASTM Standards.

The flexural properties for the OSBs, MOR and MOE, were determined on specimens of nominal 250 × 50 × 10 mm dimensions cut from each experimental panel in accordance with EN 310 [34] standard. Ten internal bond (IB) specimens of a nominal 50 × 50 × 10 mm dimensions were cut from each experimental panel in accordance with EN 319 [35] standard. The average of 10 and 20 measurements were reported for mechanical and physical properties, respectively. A tensile load was applied at a rate of 1 mm/min and the stress was determined using the load at which the specimen failed. In accordance with ASTM D1037 [36] guidelines for screw and nail withdrawals, screws and nails were driven into the OSB specimens. The screws were driven through the full thickness of the panel. Lead holes with the diameter of 70/100 of screw root diameter and 10 mm depth were opened to the screw points of screw withdrawal specimens. There were no lead holes opened to the nail withdrawal specimens. The nailing set-up was prepared similar to the one used for screws, and a pulling force with a constant speed of 2.5 mm/min was

Table 5 Comparison of chemical composition and density of paulownia (current study) and hardwoods and softwoods (3)

Raw material	Holocellulose (%)	α-Cellulose (%)	Lignin (%)	pH	Ash (%)	Solubility (%)			
						Alcohol–benzene (2/1)	1 % NaOH	Hot water	Cold water
Paulownia	69	45.1	29.3	5.7	3.6	–	–	6.8	5.3
Hardwoods	70–78	45–50	30–35	–	0.35	2–6	14–20	2–7	4–6
Softwoods	63–70	45–50	25–35	–	0.35	2–8	9–16	3–6	2–3

applied until the nails were completely separated from the specimens. Maximum applied force at the point of separation was recorded from the indicator of the test machine. A Universal Testing Machine (Schenk 40 kN) was used to obtain the withdrawal strength of nail and screw fasteners in the specimens with dimension 50 × 50 × 10 mm.

Measuring and weighing of test specimens were done according to EN 325 [37]. Thickness and length of specimens were measured using digital calipers accurate to 0.01 mm. Weight measurements were made using a balance accurate to 0.01 g. Fifteen water absorption (WA) and thickness swelling (TS) specimens of a nominal 50 × 50 × 10 mm were cut from each experimental board for use in determining WA and TS after 2 and 24 h submersions in accordance with EN 317 [38] standard. The average thickness was determined by taking several measurements at specific locations on the specimen. The specimens were then submerged in a horizontal position under 25 mm of distilled water maintained at room temperature (20 °C). After 2 and 24 h of submersions, the specimens were drip dried for 10 min, wiped of any surface water, and the thickness was determined as before. Density of the specimens (50 × 50 × 10 mm) was measured according to EN 323 [39].

The influence of nanoclay on the formaldehyde emission was evaluated by the desiccator method by randomly taking 20 samples from each type of panel following Standard AS/NZS 1859.1 [40].

Characterization

X-ray diffraction (XRD) was applied to study the exfoliation of NaMMT nanoclay in the structure of produced panels. The specimens were OSB containing 1 % NaMMT, OSB containing 3 % NaMMT, OSB containing 5 % NaMMT and pure NaMMT. The XRD was performed in a Bruker X-ray diffractometer (D8 Advanced, Germany) using Cu Kα radiation ($\lambda = 1.54$ nm). The samples were scanned in 2θ ranges 2–10° at a rate of 0.3°/min. The generator was operated at 40 kV and 30 mA. The interlayer spacing (d_{001}) of nanoclay was calculated in accordance with Bragg equation: $2d \sin\theta = \lambda$.

The morphology structure of the composites was investigated by a Philips (Model EM 208, Netherland) transmission electron microscope (TEM) with an acceleration voltage of 100 kV. The ultra-thin slides were provided by a Leica Ultracut UCT (Germany).

Statistical analysis

OSB panels were produced according to a factorial design (Table 3). Factors were nanoclay loading (4 levels, 0, 1, 3 and 5 %) and press time (2 levels, 7 and 10 min). This experimental design allows an evaluation of the effects of nanoclay loading and press cycle time on the physical and mechanical properties and formaldehyde emission of OSB panels. Analysis of variance (ANOVA) and Duncan multiple comparison tests (at 95 and 99 % confidence level) were conducted using SPSS software.

Results and discussion

Chemical properties

Results of the chemical analyses of paulownia wood are presented in Table 5. The average holocellulose content of the samples was found to be 69 %, which is slightly lower than that of hardwoods stated in the literature [3]. Based on the findings in this study, average cellulose and lignin content were also determined as 45.1 and 29.3 %, respectively. The cellulose value is comparable, but the amount of lignin is lower than the value of typical hardwoods. One of the most important chemical properties of wood is its pH level. It has an important role in developing good bonding between resin and particle, which results in enhanced panel properties [4]. The average pH value of the samples was 5.7. Urea–formaldehyde used as binder in panel manufacture has a pH value ranging from 5.5 to 6.0 [10, 41]. Comparable pH values of wood and resin is critical to have a good glue line between particles that influences both the physical and mechanical properties of the panels [4, 41]. Hot water procedure removes a part of extraneous components, such as inorganic compounds, tannins, gums, sugars, starches, and fatty derivates [42]. Hot and cold water solubility values were higher than both softwoods and hardwoods investigated in previous studies (Table 5).

Mechanical properties

Applied mechanical properties important for the application wood-based composites include not only flexural property but also the internal bonding and nail and screw withdrawal strength. The flexural modulus is a measure of resistance to panel deflection. It is one of the most important mechanical properties of wood composite panels since it affects the serviceability and the structural performance for both exterior and interior applications. The most important facing properties for structural design are the flexural properties because these properties are needed for determining panel deflections, in-plane deformations, and buckling loads. Besides, screw withdrawal resistance, an important engineering property for the potential applications of these materials, was also evaluated. Mechanical properties of all panels were found to comply with general-purpose OSB minimum property requirements of EN 300 [43] values for use in dry conditions. In general, all panels made with 5 % nanoclay and pressed for 7 min had the highest values of mechanical properties among the other types of specimens.

Modulus of rupture (MOR)

According to the analysis of variance (ANOVA), the MOR was significantly influenced by the amount of nanoclay and press time at a confidence level of 99 % but their interaction was not significant (Table 6). In addition, all mechanical properties of the boards were improved with incorporation of nanoclay to 5 % (Fig. 1). In general, concerning mechanical properties, optimum condition was using 5 % nanoclay and press time of 7 min. Results showed a significant ($P < 0.01$) decrease in the MOR value by increasing press time from 7 to 10 min. However, the interaction of above-mentioned variables was not significant. As shown in Fig. 1, the highest MOR value was observed for panel type A1B4.

Modulus of elasticity (MOE)

Similar to MOR, MOE was influenced by the addition of nanoclay and increasing press time at confidence level of 99 % (Table 6).

Results indicated also that the maximum value of MOE was attained at nanoclay concentration of 5 % (Fig. 1). The data in Fig. 1 show that the property of the boards was improved when the nanoclay content increased from 1 to 3 % and 5 %. The boards pressed for 10 min showed significantly ($P < 0.01$) lower MOE values in comparison with those pressed for 7 min. The interaction of above-mentioned variables was not significant.

Table 6 Analysis of variance (ANOVA) for applied treatments

Treatment	Nanoclay	Press time	Interaction (nanoclay × press time)
Modulus of rupture (MOR)	**	*	ns
Modulus of elasticity (MOE)	**	**	ns
Internal bonding (IB)	**	**	*
Screw holding strength (SH)	*	**	ns
Nail holding strength (NH)	**	**	ns
Water absorption (h)			
2	**	**	ns
24	*	*	ns
Thickness swelling (h)			
2	**	**	*
24	**	*	ns
Formaldehyde emission	**	**	**

ns not significant

* Significant difference at the 5 % level ($P \leq 0.05$ %)

** Significant difference at the 1 % level ($P \leq 0.01$ %)

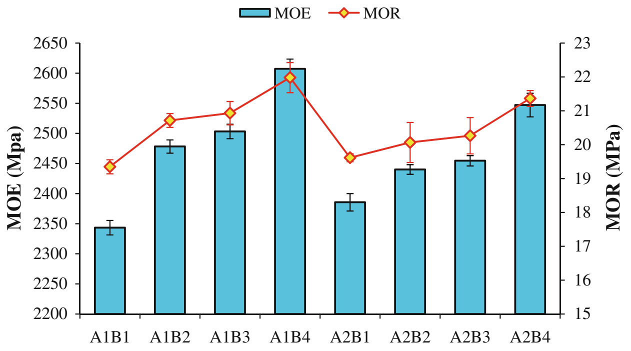

Fig. 1 MOR and MOE of various board types. Standard deviations are shown as *error bars*

Internal bonding strength (IB)

IB values of the experimental panels ranged from 0.45 to 0.60 N/mm^2 (Fig. 2), in other word, all panels met minimum IB requirement (0.30 N/mm^2) of EN 300 Type 1 panel for use in dry conditions. The highest IB value was observed for panel type A1B4, while the lowest was observed for A1B1 type panel. The internal bond strength (IB) of the boards was significantly affected by nanoclay at a confidence level of 99 %. Results showed a significant ($P < 0.01$) decrease in the IB value by increasing press time from 7 to 10 min. The interaction between the nanoparticle loading and press time was significant. As

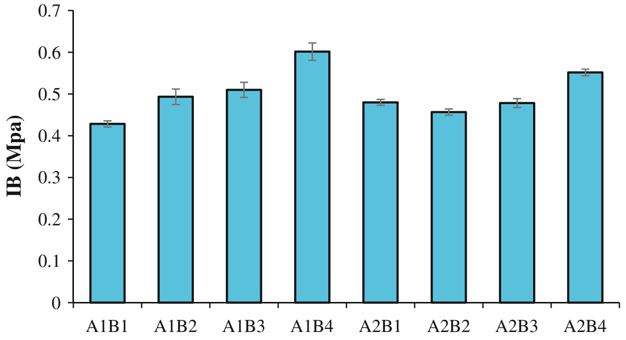

Fig. 2 IB of various board types. Standard deviations are shown as *error bars*

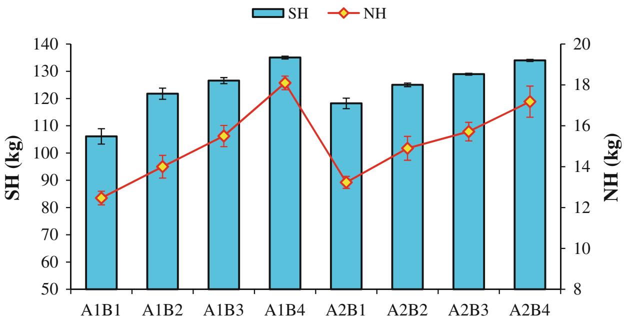

Fig. 3 Screw and nail holding strengths of various board types. Standard deviations are shown as *error bars*

shown in Fig. 2, maximum IB values were attained at the 5 % nanoclay concentration.

Screw and nail withdrawal strength

According to the results, the screw and nail withdrawal strength was affected by nanoclay and press time at 99 and 95 % confidence level, respectively (Table 6). In case of screw and nail withdrawal strength, similar to the other mechanical properties and the highest screw and nail withdrawal strength values of 135 and 18 N/mm was obtained for the OSBs made with UF resin containing 5 % nanoclay pressed for 7 min (Fig. 3). The panels pressed for 10 min showed significantly ($P < 0.05$) lower screw and nail withdrawal strength values in comparison with those pressed for 7 min. In all samples, the withdrawal strength of screw was found higher than that of nail.

The strength properties of wood-based composites are depend on many factors such as physical and mechanical properties of individual wood particles, their interfacial adhesion, the orientation and aspect ratio of particles [3, 44, 45]. Cellulose, lignin, and hemicelluloses contents of the lignocellulosics had strong influences on the mechanical properties [46]. Aspect ratio, rather than particle size, has the greatest effect on strength and stiffness [42]. The fiber length of paulownia (*Paulownia fortunei*) wood is around 0.8–1 mm and its aspect ratio is about 25 [47], which are less than those of some hardwoods and softwoods [42] and even less than those of fibers from agro-materials like oilseed stalk, corn stalk, and reed stalk [48]. Density of paulownia (*Paulownia fortunei*) wood is quite low (0.26 g/cm^3) in comparison to many common hardwoods like beech, oak, hornbeam, and ironwood [49]. Low density strands disturb the interface bond strength and require high compaction to reduce porosity and combination of locked-in internal stresses with an evaporating steam from the furnish leads to catastrophic delamination of interface bonds specially for UF bonded panels [4]. In hardwoods, vessels, morphological structural elements in longitudinal series exposed in transverse section constitute about 10–46 % of the stem and are cells of relatively large diameters (50–300 nm) which appear as the open vertical tubes within the wood structure because their end walls have partially dissolved [10]. It was also reported that resins with low molecular weights easily penetrated into the wood cell walls; however, the resins with much higher molecular weights tended only to reach the wood cell lumen instead of the cell walls [50]. Too high flow ability leads to an overpenetration of the resin into the wood and to a starved glue line [10]. In such a case, no glue line and hence no bonding strength can be formed. Resin penetration is a function of resin viscosity, chemical compatibility of the resin with the strands surface and physical condition of the strands surface [4, 10]. The quality of bonding and hence the properties and the performance of the wood-based panels and beams are determined by three main parameters: (1) the wood, especially the wood surface, including the interface between the wood surface and the bond line; (2) the adhesive; (3) the working conditions and process parameters [10]. Nanoparticles by decreasing the UF resins viscosity could prevent too much of adhesive from filtering into the wood to create the phenomena of permeation or inadequate bonding [15]. Penetration of glue in the sense of filling of cell cavities—not cell walls—has received considerable attention [51]. At low weight OMMT loadings, the enhancement of properties of composites is attributed to the lower percolation points created by the high aspect ratio nanoclays. The increase in properties may also be attributed to the formation of intercalated and exfoliated nanocomposite structures formed at these clay loadings [20, 26, 51, 52]. The properties of panels pressed for lower time (7 min) were better than those pressed for more time (10 min). By taking into account that nanoclay has an accelerating effect on UF curing time [20], the reason could be because of partial degradation of cured UF polymer and damages of paulownia wood cell wall as it is thinner than common woods [42, 47]. When wood-based composites are hot-pressed under conditions of high temperature and low moisture content, compressive stresses are imparted to the individual wood cells resulting in damaged wood cell walls and strength losses [10, 53].

NaMMT exfoliates in UF resin and urea formaldehyde molecules could penetrate the galleries between clay layers [20]. Furthermore, when wood-based composites are hot-pressed under conditions of high temperature and low moisture content, compressive stresses are imparted to the individual wood cells resulting in damaged wood cell walls and strength losses [10]. Hot-water extractives can diffuse to the surface, thus blocking cells and reducing contact of the matrix with the hydroxyl groups (–OH) of cellulosic material resulting in inferior interfacial bonding strength [48]. Besides, water-soluble extractives, which their amount in paulownia woods is high, have significant effect on the UF resin gel time, which is important in the determination of the adhesion [54]. Extractives can have adverse effects on the curing of adhesives, leading to poor particle–particle bond strength and may cause blows and severely deteriorating the internal bond strength [46]. The negative effect of extractives on the mechanical properties of particleboard has been stated by many authors [46, 55]. Besides, paulownia wood contains a high amount of ash, which can result in weak bonding between particles as well as very low internal bonding strength within panels [46]. As a result, OSB from paulownia requires additional treatments; chemical modification of strands [4, 14]. Biological treatments of anaerobic digestion (AD) [56] using MDI resin instead of [57] and surface coating of the end product [58]. Similar mechanical results have been reported for panels made using underutilized biomaterials [2, 3, 41, 44, 46, 59].

Physical properties

Water absorption and thickness swelling

Reconstituted wood-based products have a well-documented problem of water sorption and lack of dimensional stability. Figures 4 and 5 show the values of the water absorption and thickness swelling for the composites, which vary depending upon the nanoparticle loading and press time. The results of ANOVA for TS and WA of the OSBs after 2 and 24-h water immersion times are given in Table 6. According to the results, water absorption (both after 2 and 24 h water immersion) significantly influenced by the amount of nanoclay and press time at confidence levels of 95 and 99 %, respectively. On the other hand, thickness swelling (both after 2 and 24 h water immersion) significantly influenced by the amount of nanoclay and press time at a confidence level of 99 % and again the interaction was not significant. Both mentioned properties of the panels were improved with incorporation of nanoclay to 5 % and press time decreased from 10 to 7 min. In general, concerning physical properties, optimum condition was using 5 % nanoclay and press time of 7 min. Nevertheless, none of the panels did satisfy the TS and WA requirement of EN Standard for general-purpose usage. Results indicate that with incorporation of nanoclay to 5 %, the WA and TS of the panels decrease significantly and after that increases. The data in Figs. 4 and 5 show that the panel type A1B4 had the lowest average WA and TS values. The maximum water absorption was recorded for controls panels (A1B1).

The water uptake of wood can be mainly ascribed to hydrogen bonding of water molecules to the free hydroxyl groups presented in cellulose and hemicelluloses [60, 61]. Additionally, large numbers of porous tubular structures present in wood accelerate the penetration of water by the capillary action [22, 42]. OSB compared to other wood-based composites like plywood has poor water resistance in contact with water that is direct result of higher pressure needed to consolidate the OSB mat [1]. Within this framework, the consolidation strain induced during the pressing process was defined as a function of the targeted

Fig. 4 WA (after 2 and 24 h) of various board types. Standard deviations are shown as *error bars*

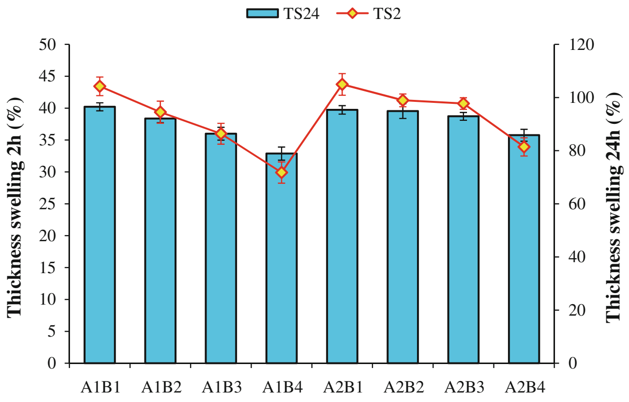

Fig. 5 TS (after 2 and 24 h) of various board types. Standard deviations are shown as *error bars*

thickness for the panel product, the flake thickness, and, finally, the number of deposited strands [62]. Wood-based panels such as fiberboard, plywood, or OSB are also subjected to reversible and/or irreversible swelling under wet and dry conditions. The amount of the reversible swelling is directly related to hygroscopicity of wood [63]. It is known that the springback in the wood-based composites occurs usually after manufacture of the panels. The springback is an irreversible thickness swelling, which occurs after wetting the composites, is attributed to the release of applied stresses accompanied by some loss of the glue bonds [64]. The springback indicates debonding of the adhesion between the wood elements and the adhesives. Upon gluing, it was apparent that the strands due to bulk volume of the material did not glue well, resulting in interrupted glue line between the particles. Urea formaldehyde is not a waterproof binder and absorbs moisture when exposed to humid conditions. When the panels were immersed in water, high swelling took place because of high porosity of the strands and their hygroscopicity, spring back (the release of built in compressive forces brought about during panel manufacturing) and water absorption affinity of urea formaldehyde [65]. Thickness swelling is also affected by bond quality and adhesive property [14]. The wood porosity affects also the type and form of the adhesive as it affects the ability of the substrate to absorb water and other solvents from the adhesive, as well as allowing some of the adhesive to be absorbed over larger surface areas [10]. All the treated specimens strongly resisted water absorption and as a result, thickness swelling decreased. Nanoclay increases the tortuous path for water transport and as a result water diffusivity decreases [22, 52]. Decreasing available space for water absorption due to occupation of voice spaces in the wood by the nanoclay can be another mechanism for the lower water uptake of nanocomposites [66]. The presence of nanoclay in the composite hinders the permeation of water through the composite [22]. Another reason can be obstruction of capillaries in wood by the modifier so that water lost its main channel [22, 50]. Under the combined influence of pressure and heat, cell lumens in hardwoods collapse, and a certain amount of fractures in cell walls develops and such densification also is responsible for its dimensional instability in the form of the hygroscopic swelling and wood with the highest degree of densification had the highest thickness swelling [53]. Values of thickness swelling of the panels decreased with increasing press time. Similar results were observed by several researchers [2, 67, 68]. Thickness of wood samples decreases with increasing densification period this phenomenon can be explained as a consequence of softening of solid wood with better distribution of temperature as a result of longer press time [2, 53, 69]. Nevertheless, none of the boards did satisfy the TS and WA requirement of EN Standard for general-purpose usage. Thickness swelling is also affected by bond quality and adhesive property [14]. There are many other methods to improve the water absorption as well as thickness swelling of wood based panels; adding wax (0.5–1 %) to the mixture of adhesive and particles during manufacturing process, decreasing the "springback" affect by reducing the density of panels and chemical modification of particles/strands [2, 14, 69]. Besides, using nano-emulsion of paraffin can lead to better water resistance [70]. Similarly, lower physical properties have been reported for particleboards made using biomaterials [41, 44, 59, 69, 70].

Formaldehyde emission

The addition of nanoclay into OSB significantly (at confidence level of 99 %) decreased formaldehyde emission value (Table 6). The maximum decrease in the formaldehyde emission of the panels was recorded for those made with UF containing 5 % nanoclay. Besides, increasing press time from 7 to 10 min significantly ($P < 0.01$) decreased the formaldehyde emission. Depending on addition of the nanoclay in the panels, the formaldehyde emission values ranged from 14.5 to 43 % lower than the panels made with untreated UF adhesive and pressed for 7 min (Fig. 6). The minimum value of formaldehyde gas emission was measured at nanoclay concentration of 5 %. In general, it was revealed that the formaldehyde emission was affected by nanoclay incorporation and press time. Panel type A2B4 had the lowest formaldehyde emission value with 0.645 mg/L followed by panel type A1B4 (0.690 mg/L), panel type A2B3 (0.740 mg/L), panel type A1B3 (0.790 mg/L), panel type A2B2 (0.900 mg/L), panel type panel type A1B2 (0.967 mg/L), panel type A2B1 (1.090 mg/L) and panel type A1B1 (1.130 mg/L). The decrease in formaldehyde emission of the OSB panels with nanoclay was attributed to barrier property (shielding effect) of the nanoparticles [20, 26, 52, 71]. In fact, the organoclay could improve the barrier performance of composites based on the simple "tortuous path" model [52]. NaMMT is exfoliated in UF resin and urea formaldehyde molecules could penetrate the galleries between clay layers and exfoliate the clay [20]. The similar results were reported in previous studies [12, 20, 52]. Due to extremely high aspect ratio, specific surface area, small size effects for its insufficient surface atoms and high reactivity, nanoclay could easily cross with the active groups of pure resins and improve bonding strength of the modified resin [20, 25, 26, 71]. Nanoclays consist of stacks of sheet like silica platelets with the thickness of about 1 nm and extremely large surface area and aspect ratios [21]. Since clay layers constitute a barrier to gases and water, forcing them to follow a tortuous path, the

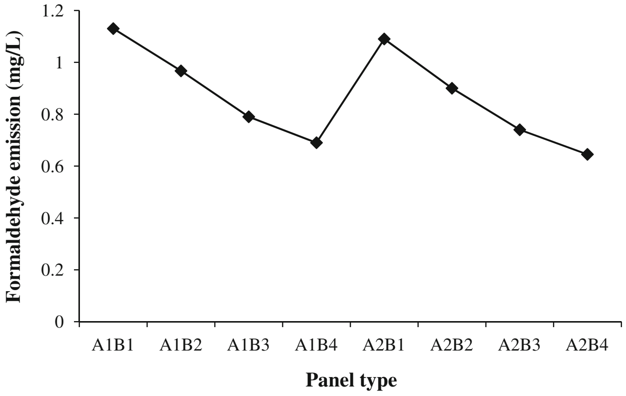

Fig. 6 Formaldehyde emission of various board types

introduction of nanoclays into polymer bio-structures has been shown to greatly improve barrier properties [72]. Post-treatment methods to decrease and minimize formaldehyde release are based on compounds like ammonia, ammonium salts, or urea [73]. Another effective way to reduce formaldehyde release is the addition of formaldehyde-binding substances (‘‘scavengers’’) to the resin or to the wood particles [45]. Urea also acts as a buffer to control the pH and improve the stability of UF resins. Ammonium chloride acts as an acid catalyst of the curing reaction and as a scavenger [73]. Coating panel's surfaces with decorative overlays are to eliminate the release of formaldehyde [74]. Besides, processing condition of wood based panels have important role on respective formaldehyde emission so that the higher the press cycle time the lower the formaldehyde emission at service [12].

Dispersion behavior

Figure 7a shows the XRD profile of pure NaMMT. It can be seen that nanoclay shows peaks at 7.4, 14.5, 20.5, 21.1, 26.79, 28.1, 35.23, 43.79, 54.33, and 62.30°. The peak at $2\theta = 7.2°$ is the strongest one that corresponds to a *d*-spacing of 1.21 nm according to Bragg's law [20]. Figure 7b shows the diffraction patterns for pure NaMMT, OSB containing 1 % NaMMT, OSB containing 3 % NaMMT and OSB containing 5 % NaMMT in the range of $2\theta = 2$–10°. It can be seen that the strong peak of NaMMT at $2\theta = 7.2°$ disappears completely in all OSB containing NaMMT, which demonstrates that nanoclay was quite exfoliated and delaminated in UF resin. This shift of the XRD peak to lower value of 2θ indicates that the exfoliated structure was generated during the hot press. In fact, the sharp narrow diffraction peaks show crystalline structure of nanoclay, while the broad peak is an amorphous structure after exfoliation. This finding is compatible with previous studies [20, 52]. They reported that NaMMT is exfoliated in UF resin. Urea formaldehyde molecules could penetrate the galleries between clay layers, and the clay was exfoliated.

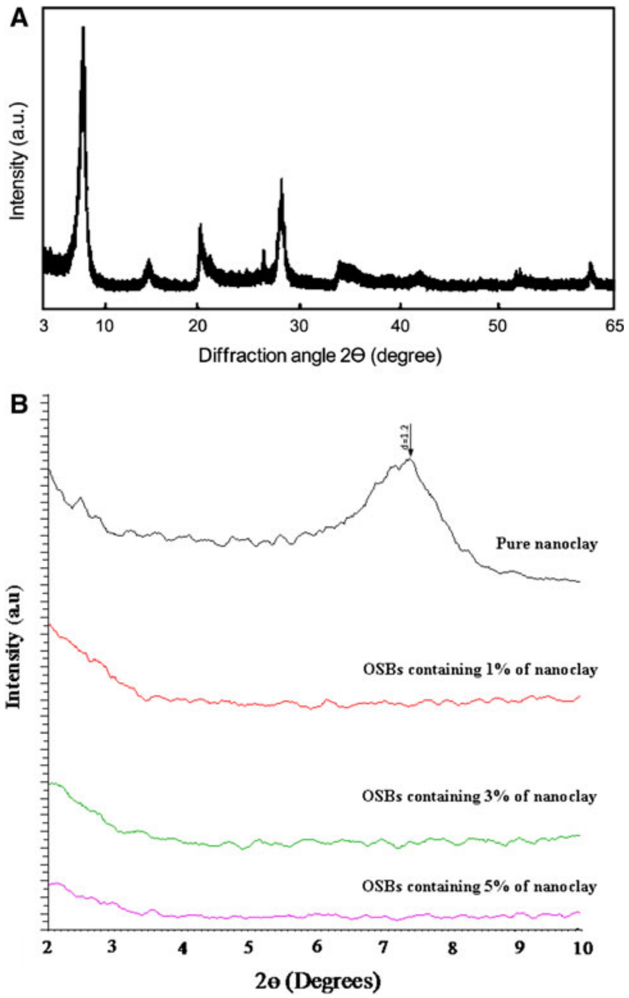

Fig. 7 **a** XRD pattern of pure nanoclay, **b** XRD pattern of OSBs containing different nanoclays

Figure 8 shows the dispersion state of the composites as observed by TEM. The dark line represents the intersection of the silicate layers, while the white background corresponds to UF matrix. When the loading level of NaMMT into the UF is 5 % (Fig. 8c) NaMMT exhibited better dispersion and distribution of the nanoclay within the polymer matrix than the 1 and 3 % of nanoclay content (Fig. 8a, b). By increasing the level of NaMMT to 5 %, the dispersion of nanoclay became more homogenous and well dispersed.

Conclusions

In this study, the potential of underutilized low quality paulownia wood as a bio-resource in OSB manufacturing was studied. Based on initial findings of this study, it can

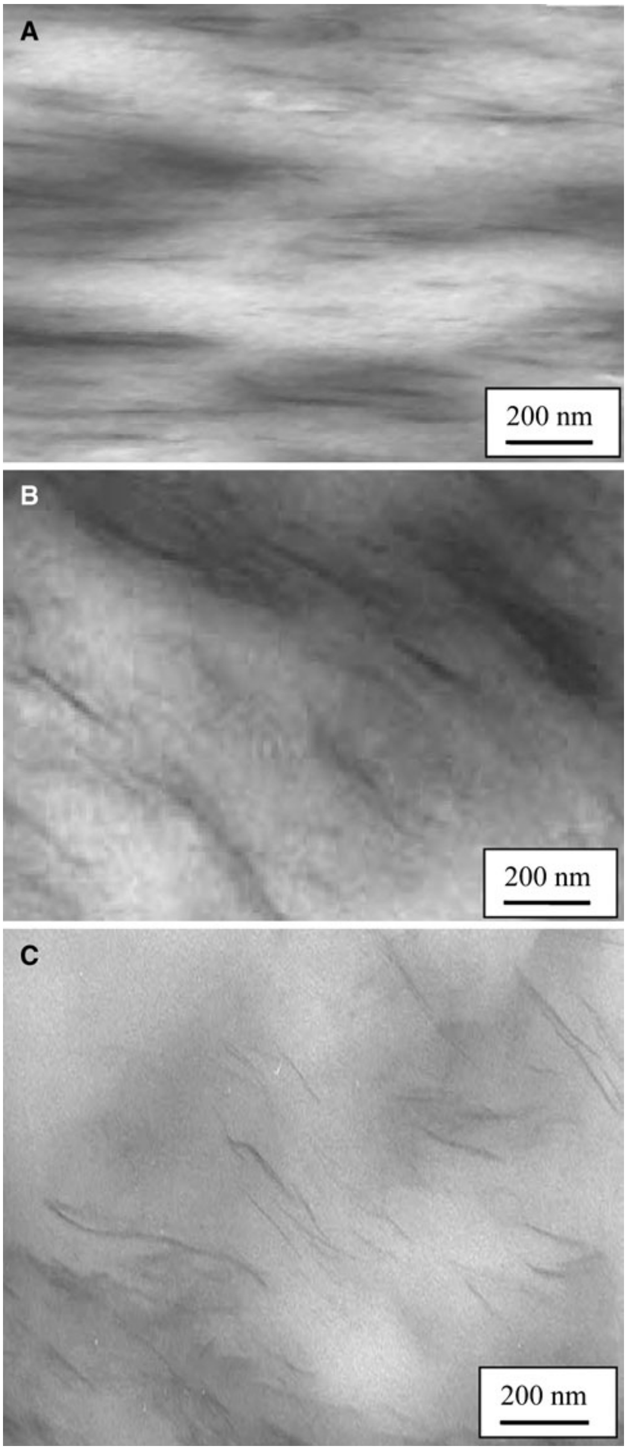

Fig. 8 **a** The TEM pattern of UF resin specimen containing 1 % nanoclay, **b** the TEM pattern of UF resin specimen containing 3 % nanoclay, **c** the TEM pattern of UF resin specimen containing 5 % nanoclay

be concluded that underutilized paulownia wood could be used in manufacturing OSB. The addition of small percentage (5 %) of nanoclay into UF resins appeared to improve considerably the applied properties of the resulting panels. XRD characterization indicated that nanoclay completely exfoliated when mixed with UF and TEM images approved the results. The influence of nanoclay addition was particularly noted in OSB by the increase in mechanical property, water resistance, and reduction of formaldehyde emission. Valuable wood-based panels made from underutilize low quality paulownia wood employing nanotechnology can be considered as an alternative solution to increased demand for raw materials. Finding this type of new application area for underutilized paulownia wood can lead to decreasing the pressure on the forests and alleviating raw material shortage in wood industry in developing countries.

Acknowledgments The authors gratefully acknowledge financial assistance of Iran Nanotechnology Initiative Council and the Research Deputy of Gorgan University of Agricultural Sciences and Natural Resources.

References

1. Papadopoulos AN, Traboulay E (2002) Dimensional stability of OSB made from acetylated fir strands. HolzalsRoh-und Werkstoff 60(2):84–87
2. Ashori A, Nourbakhsh A (2008) Effect of press cycle time and resin contents on physical and mechanical properties of particleboard panels made from the underutilized low-quality raw materials. Ind Crops Prod 28(2):225–230
3. Pirayesh H, KhazaeianA(2012) Using almond (*Prunusamygdalus* L.) shell as a bio-waste resource in wood based composite. Composites Part B 43: 1475–1479
4. Ndazi B, Tesha JV, Nisanda ETN (2006) Some opportunities and challenges of producing bio-composites from non-wood residues. J Mater Sci 41:6984–6990
5. Joshi SV, Drza LV, Mohanty AK, Arora S (2004) Are natural fiber composites environmentally superior to glass fiber reinforced composites? Composites Part A 35:371–376
6. Jiménez L, Rodriguez A, Ferrer JL, Pérez A, Angulo V (2005) Paulownia, a fast growing plant, as a raw material for paper manufacturing. Afinidad 62:100–105
7. Bergmann BA (1998) Propagation method influences first year field survival and growth of Paulownia. New Forest 16:251–264
8. Martinez E, Lucas-Borja ME, Andres Abellan M, Lopez Serrano FR, Garcia Morote A, del Cerro Barja A (2010) Aprovechamientoenergetico de *Paulownia* spp. en el ambitomediterraneo. Revista Montes 102:5–12
9. Rebollar M, Pérez R, Vidal R (2007) Comparison between oriented strand boards and other wood-based panels for the manufacture of furniture. Mater Des 28:882–888
10. Dunky M, Pizzi A (2002) Wood adhesives. In: Chaudhury M and Pocius AV (eds) Adhesive science and engineering—2: surfaces, chemistry and applications, chap 23. Elsevier, Amsterdam, pp 1039–1103
11. Wang Z, Li L, Gong M (2012) Measurement of dynamic modulus of elasticity and damping ratio of wood-based composites using the cantilever beam vibration technique. Constr Build Mater 28:831–834
12. Aydin I, Gursel C, Semra C, Cenk D (2006) Effects of moisture content on formaldehyde emission and mechanical properties of plywood. Build Environ 41:1311–1316

13. Doosthoseini K, Zarea-Hosseinabadi H (2010) Using Na + MMT nanoclay as a secondary filler in plywood manufacturing. J Indian Acad Wood Sci 7(1–2):58–64
14. Abdolzadeh H, Doosthoseini K, Karimi AN, Enayati AA (2011) The effect of acetylated particle distribution and type of resin on physical and mechanical properties of poplar particleboard. Eur J Wood Prod 69(1):3–10
15. Lin Q, Yang G, Liu J, Rao J (2006) Property of nano-SiO_2/urea formaldehyde resin. Front For China 2:230–237
16. Roumeli E, Pavlidou E, Papadopoulou E, Vourlias G, Bikiaris D, Paraskevopoulos KM, Chrissafis K (2012) Synthesis, characterization and thermal analysis of urea formaldehyde/nanoSiO_2 resins. Thermochimica Acta 527:33–39
17. Wang S, Yang T, Lin L, Lin C, Tsai M (2008) Fire-retardant-treated low formaldehyde-emission particleboard made from recycled wood-waste. Bioresour Technol 99:2072–2077
18. Nemli G, Ozturk I (2006) Influences of some factors on the formaldehyde content of particleboard. Build Environ 41:770–774
19. Kim S, Kim HJ (2005) Effect of addition of polyvinyl acetate to melamine–formaldehyde resin on the adhesion and formaldehyde emission in engineered flooring. Int J Adhes Adhes 25:456–461
20. Lei H, Du G, Pizzi A, Celzard A (2008) Influence of nanoclay on urea-formaldehyde resins for wood adhesives and its model. J Appl Polym Sci 109:2442–2451
21. Haq M, Burgueno R, Mohanty AK, Misra M (2008) Hybrid bio-based composites from blends of unsaturated polyester and soybean oil reinforced with nanoclay and natural fibers. Compos Sci Technol 68:3344–3351
22. Khanjanzadeh H, Tabarsa T, Shakeri A (2012) Morphology, dimensional stability and mechanical properties of polypropylene—wood flour composites with and without nanoclay. J Reinf Plast Compos 31(5):341–350
23. Reddy N, Yang Y (2009) Properties and potential applications of natural cellulose fibers from the bark of cotton stalks. Bioresour Technol 100:3563–3569
24. Zeng QH, Yu AB, Lu GQ, Paul DR (2005) Clay-based polymer nanocomposites: research and commercial development. J Nanosci Nanotechnol 5:1574–1592
25. ArafaIM FaresMM, Barham AS (2004) Sol-gel preparation and properties of interpenetrating, encapsulating and blend silica-based urea-formaldehyde hybrid materials. Eur Polym J 40:1477–1487
26. Khanjanzadeh H, Tabarsa T, Shakeri A, Omidvar A (2011) Effect of organoclay platelets on the mechanical properties of wood plastic composites formulated with virgin and recycled polypropylene. Wood Mat Sci Eng 6(4):207–212
27. Ashori A, Nourbakhsh A (2009) Effects of nanoclay as a reinforcement filler on the physical and mechanical properties of wood-based composite. J Compos Mater 43:1869–1875
28. T 257 cm. Sampling and preparing wood for analysis, TAPPI Standards; 1985
29. Wise LE, Karl HL (1962) In: Earl Libby C (ed) Cellulose and hemicelluloses in pulp and paper science and technology, vol 1. McGraw Hill Book Co, New York
30. T 222 om (1998) Acid-insoluble lignin in wood and pulp. TAPPI Standards
31. T 211 om (1993) Ash in wood and pulp. TAPPI Standards
32. T 207 om (1999) Water solubility of wood and pulp. TAPPI Standards
33. Browning BL (1967) In: Holocellulose content in wood, methods of wood chemistry, vol. I–II. Interscience Publishers, New York
34. EN 310 (1993) Determination of bending strength and modulus of elasticity. EN 312. 2003. Particleboards—specifications
35. EN 319 (1993) Particleboards and fiberboards; determination of tensile strength perpendicular to the plane of the board
36. ASTM D 1037-96 (1999) Standard methods for evaluating properties of wood-base fiber and particle panel materials. American Society for Testing and Materials, Philadelphia
37. EN 325 (1993) Determination of dimensions of test pieces. EN 326-1 (1993) Sampling, cutting, and inspection. Part 1
38. EN 317 (1993) Particleboards and fiberboards—determination of swelling in thickness after immersion in water
39. EN 323 (1993) Determination of density
40. Australian/New Zealand standard AS/NZS 1859.1 (2004) Reconstituted wood-based panels Specifications. Part 1: particleboard
41. Nemli G, Demirel S, Gümüokaya E, Aslan M, Acar C (2009) Feasibility of incorporating waste grass clippings (*Loliumperenne* L.) in particleboard composites. Waste Manag 29:1129–1131
42. Ashori A, Nourbakhsh A (2010) Reinforced polypropylene composites: effects of chemical compositions and particle size. Bioresour Technol 101:2515–2519
43. European Committee for Standardization. Wood-based panels, Brussel, Belgium. EN 300. 1997: Oriented strand boards (OSB)—definitions, classification specifications
44. Copur Y, Guler C, Akgul M, Tascioglu C (2007) Some chemical properties of hazelnut husk and its suitability for particleboard production. Build Environ 42:2568–2572
45. Ayrilmis N, Buyuksari U, Avci E, Koc E (2009) Utilization of pine (*Pinuspinea* L.) cone in manufacture of wood-based composite. For Ecol Manag 259:65–70
46. Pirayesh H, Khazaeian A, Tabarsa T (2012) The potential for using Walnut (*Juglansregia* L.) shell as a raw material for wood-based particleboard manufacturing. Compos B. doi: 10.1016/j.compositesb.2012.02.016
47. Ziaeddin Hosseini S, Afra E (2006) A study of fiber characterization and chemical composition of paulownia (*Paulownia fortunei*) in Gorgan region Iranian (in Persian). J Nat Resour 58(4):871–878
48. Nourbakhsh A, Ashori A (2010) Wood plastic composites from agro-waste materials: analysis of mechanical properties. Bioresour Technol 101:2525–2528
49. Salari A (2012) Influence of nanoclay and nanosilica on properties of oriented strand board (OSB) Product from Paulownia. M.S. Thesis 2012. Department of Wood and Paper Technology, Gorgan University of Agricultural Sciences and Natural Resources (GUASNR), Gorgan
50. Shi J, LI J, Zhou W, Zhang D (2007) Improvement of wood properties by urea formaldehyde resin and nano-SiO_2. Front For China 2(1):104–109
51. Gindl W, Schoberl T, Jeronimidis G (2004) The interphase in phenol–formaldehyde and polymeric methylene diphenyl-di-isocyanate glue lines in wood. Int J Adhes Adhes 24:279–286
52. Zahedsheijani R, Faezipour M, Tarmian A, Layeghi M, Yousefi H (2011) The effect of Na + montmorillonite (NaMMT) nanoclay on thermal properties of medium density fiberboard (MDF). Eur J Wood Prod. doi:10.1007/s00107-011-0583-6
53. Buyuksarı U, Hiziroglu S, Akkılıc H, Ayrılmıs N (2012) Mechanical and physical properties of medium density fiberboard panels laminated with thermally compressed veneer. Composites Part B 43:110–114
54. Buyuksari U, Ayrilmis N, Avci E, Koc E (2010) Evaluation of the physical, mechanical properties and formaldehyde emission of particleboard manufactured from waste stone pine (*Pinuspinea* L.) cones. Bioresour Technol 101:255–259
55. Nemli G, Aydin A (2007) Evaluation of the physical and mechanical properties of particleboard made from the needle litter of Pinus pinaster Ait. Ind Crops Prod 26(3):252–258
56. Zheng Y, Pan Z, Zhang R, EI-Mashad HM, Pan J, Jenkins BM (2009) Anaerobic digestion of saline creeping wild ryegrass for biogas production and pretreatment of particleboard material. Bioresour Technol 100:1582–1588

57. Yang P, Zhang F (2004) Study on cure conditions of PMDI-based binder in use of wheat straw particleboard. China Adhesives 14:37–39
58. Nemli G, Kırcı H, Serdar B, Ay N (2003) Suitability of kiwi pruning for particleboard manufacturing. Ind Crops Prod 17:39–46
59. Guntekin E, Karakus B (2008) Feasibility of using eggplant stalks (*Solanum melongena*) in the production of experimental particleboard. Ind Crops Prod 27:354–358
60. Nourbakhsh A, Farhani Baghlani F, Ashori A (2011) Nano-SiO_2 filled rice husk/polypropylene composites: physico-mechanical properties. Ind Crops Prod 33:183–187
61. Gwon JG, Lee SY, Chun SJ, Doh GH, Kim JH (2010) Effects of chemical treatments of hybrid fillers on the physical and thermal properties of wood plastic composites. Part A 41:1491–1497
62. Oudjehane A, Lam F (1998) On the density profile within random and oriented wood-based composite panels: horizontal distribution. Compos B 29B:687–694
63. Hsu WE, Schwald W, Shields JA (1989) Chemical and physical changes required for producing dimensionally stable wood-based composites. Part 2: chemical and physical changes required for producing dimensionally stable wood-based composites. Wood Sci Technol 23(3):281–288
64. Mohebby B, Gorbani-Kokandeh M, Soltani M (2009) Springback in acetylated wood based composites. Constr Build Mater 23:3103–3106
65. Biswas D, Bose SK, Hossain MM (2011) Physical and mechanical properties of urea formaldehyde-bonded particleboard made from bamboo waste. Int J Adhes Adhes 31(2):84–87
66. Deka BK, Maji TK (2010) Effect of coupling agent and nanoclay on properties of HDPE, LDPE, PP, PVC blend and Phargamites karka nanocomposite. Compos Sci Technol 70(12):1755–1761
67. Tabarsa T, Chui YH (1997) Effects of hot-pressing on properties of white spruce. Forest Prod J 47:71–76
68. Welzbacher CR, Wehsener J, Rapp AO, Haller P(2008) Thermo-mechanical densification combined with thermal modification of Norway spruce (*Piceaabies* Karst) in industrial scale—dimensional stability and durability aspects. HolzRohWerkst 66:39–49
69. Tabarsa T, Jahanshahi S, Ashori A (2010) Mechanical and physical properties of wheat straw boards bonded with a tannin modified phenol-formaldehyde adhesive. Compos B 42(2):176–180
70. Bektas I, Guler C, Kalaycioglu H, Mengeloglu F, Nacar M (2005) The manufacture of particleboards using sunflower stalks (*Helianthus annuus*) and poplar wood (*Populus alba* L.). J Compos Mater 39(5):467–473
71. Kord B, Hemmasi AH, Ghasemi I (2010) Properties of PP/wood flour/organo modified montmorillonite nanocomposites. J Wood Sci Technol 45:111–119
72. Adame D, Beall GW (2009) Direct measurement of the constrained polymer region in polyamide/clay nanocomposites and the implications for gas diffusion. Appl Clay Sci 42:545–552
73. Kim S (2009) Environment-friendly adhesives for surface bonding of wood-based flooring using natural tannin to reduce formaldehyde and TVOC emission. Bioresour Technol 100:744–748
74. Nemli G, Kalaycıoglu H (2006) The resistances of several types of overlaying materials against cigarette burn, scratch, and abrasion. Build Environ 41:640–645