Effects of adding nano-wollastonite, date palm prunings and two types of resins on the physical and mechanical properties of medium-density fibreboard (MDF) made from wood fibres

Figure 1.
Palm orchard in Bushehr province in Iran (A); a palm grove with some dried leaved to be pruned (B); the furnish after former to be hot-pressed (C).

Hamid R. Taghiyari¹
Farzad Arbabi Ghamsari¹
Ehsan Salimifard²

¹ Shahid Rajaee Teacher Training University
Faculty of Materials Engineering and New Technologies
Wood Science and Technology Department
Tehran
Iran

Auteur correspondant / Corresponding author:
Hamid R. Taghiyari – htaghiyari@srttu.edu

Doi: 10.19182/bft2018.335.a31517
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Date de publication : 12 mars 2018

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The effects of adding nano-wollastonite (NW) and date palm prunings on the physical and mechanical properties of medium-density fibreboard (MDF) made from wood fibres were studied here. Urea-formaldehyde (UF) and isocyanate (IC) resins were used to produce panels at contents of 10% and 5% respectively, based on the dry weight of the composite material. NW was used at the 5% and 10% concentrations based on the dry weight of the resins, and the results were compared with those for panels containing no NW. NW with a <100 nm particle size was used. NW was mixed and sprayed onto the material prior to formation of the mat. The results indicated distinctly lower water absorption and thickness swelling of panels produced with IC resin. Adding palm residues significantly increased the IB values in panels with both UF and IC resins. Adding NW had the effect of decreasing the mechanical properties of panels produced with UF resin, but had an increasing effect on panels made with IC resin. It was concluded that palm leaf residues can be considered as a potential raw material for producing MDF panels using both UF and IC resins. Moreover, NW is recommended as an additive in composite panels with 10% of palm residues to compensate in part for the property losses.

Keywords: minerals, nanomaterials, natural fibres, palm leaves, particleboard, thermal conductivity coefficient, wollastonite, wood-composite.
**Introduction**

Trees produce valuable constructional materials with unique properties; they help humankind keep sustainable development; and they provide beauty and clean air for both human and wildlife (Daly-Hassen et al., 2014; Arce and Moya, 2015; Gbètoho et al., 2017). Moreover, they provide raw materials for many other products and composites (Altuntas et al., 2017; Fernandes et al., 2017; Hubbe et al., 2017). However, it has also some drawbacks, like dimensional instability, vulnerability to biological deteriorating agents, and fire (Adamopoulos et al., 2012; Chan-Hom et al., 2017; Esmailpour et al., 2017; He et al., 2016; Hill, 2006; de Medeiros et al., 2016; Schmidt, 2006; Schmidt et al., 2016), and therefore, many modification methods were studied to overcome the drawbacks (Bastani et al., 2016; Hosseinpouria et al., 2016, 2017; Behr et al., 2017). One other drawback is that boards with large dimensions are not readily available at large quantities due to limitation in time and space for cultivation and harvesting of large trees (Fernandez-Puratich and Oliver-Villanueva, 2014; Andrade et al., 2016). A suitable substitute for wood boards is wood composite panels. Therefore, composite panels were also elaborated from different aspects as a substitution (Valenzuela et al., 2012; Candan and Akbulut, 2014; Tajvidi et al., 2016; Lu et al., 2017). In this connection, different resins and modified resins were also studied to improve the qualities of wood composite panels (Sheikhholeslami et al., 2016; Mantanis et al., 2017).

The constant need of composite manufacturing factories for raw materials made way to the utilization of a variety of lignocellulosic materials as well as different natural and synthetic fibers. In this connection, camel-thorn, different nuts, pruned branches, straws, kenaf, and even chicken feathers were reported to be successfully used in composite production (Parkinson, 1998; Koch, 2006). However, the main portion of composite panels consists of wood chips or fibers from fast-growing trees and trees with lower wood quality (Mendes et al., 2013; Behling et al., 2017).

Iran is not rich in forest land but 220,000 hectares of date palm cultivation out of 770,000 hectares around the world are located in Iran (Hosseinkhani, 2015). Date palm leaves should be pruned regularly to keep them healthy and fertile; in fact, each date palm tree produces 10-20 kg pruned residues annually (Hosseinkhani, 2015), making an abundant source of lignocellulosic raw material to be used for industrial purposes. Date palm pruned residues were reported to be successful in production of MDF at pilot plant scale and using 100% pruned leaves of date palm (Hosseinkhani, 2015). However, Iran’s MDF manufacturing plants are designed for production of MDF with wood fibers; therefore, a percentage of date palm pruned leaves to be mixed with wood fibers would be a practical method to provide a portion of the raw material in each production batch, and to use the same machinery for MDF production at the same time. Similar procedure was reported to have promising results for Camel-thorn (Alhagi maurorum) fibers in MDF (Taghiyari et al., 2016a).

Nanotechnology and nanomaterials have been effective to improve properties of many materials and overcome some of their drawbacks (Majidi, 2016; Matinise et al., 2016; Harsini et al., 2017; Pethig, 2017; Suganya et al., 2017). In this regard, nano-wollastonite (NW) was reported to improve thermal conductivity coefficient of composite mats (Taghiyari et al., 2013ab), physical and mechanical properties, biological resistance against wood-deteriorating fungi, and fire resistance of solid wood species and composites (Karimi et al., 2013; Taghiyari et al., 2014ab). NW was also reported to make bonds with wood chemical components (Taghiyari et al., 2016b). Moreover, it was reported that wollastonite has no health hazards for human or wildlife (Huuskonen et al., 1983ab; Maxim and McConnell, 2005; Taghiyari and Sarvari Samadi, 2016). Therefore, separate sets of panels were produced with NW mixed in the resin before being sprayed on the composite furnish.

**Experimental methods**

**Composite panel production**

A dry process was used to produce the MDF panels. In order to do this, industrial defibrated fibers were purchased from Sanaye Choobe Khazar Company, Iran. The fibers were composed of beech (Fagus orientalis), alder (Alnus glutinosa), maple (Acer hyporcanum), hornbeam (Carpinus betulus), and three poplar species (Populus nigra, Populus deltoides, and Populus euroamericana). The resin and NW had solid contents of 60% and 40%, respectively. The resin/NW mixture was sprayed on the wood fibers and thoroughly mixed in a rotating drum before hot pressing. Average moisture content at the time of pressing was 7.5%. The blended fibers were manually poured into a 45 × 45 cm frame to form the mat which was then pressed at 170°C and 160 bar for 4 minutes to produce 16 mm thick panels with a target density of 700 kg/m³. Four replicate boards were made for each treatment and the panels were conditioned at 25±2°C and 60% relative humidity for 3 weeks after pressing. The outer 40 mm around each panel was cut away before the final specimens were cut to eliminate source of bias as a result of lower consolidated panel material that might not be representative of each treatment panel.

**Palm leaf preparation**

Date palm pruned leaves from Phoenix dactylifera L. were cut and dried from Borazjan city (Dashtestan) located in Bushehr province in Iran (latitude: 29° 16’ 11” N, longitude: 51° 13’ 7” E, elevation above sea level: 70 m), and they were taken to Shahid Rajaee Teacher Training University for defibration (figure 1). They were first washed and then cooked in boiling water for two hours before being defibrated in refiner in two runs. The defibrated palm fibers were added to wood fibers at 10% w/w basis. Separate sets of MDF panels were produced with 0% palm content as control specimens.
Nano-Wollastonite (NW) emulsion with solid content of 40% and particle size of < 100 nm was purchased from Mehrabadi Machinery (Mehrabadi Machinery Mfg. Co., Iran); the chemical composition of NW is explained in table I. NW was blended with urea formaldehyde (UF) and isocyanate (IC) resins at three levels (0, 5, and 10 % w/w) based on the dry weight of resins. These two NW-content levels were chosen based on previous projects (Taghiyari et al., 2014ab; Taghiyari and Sarvari Samadi, 2016). The resin contents were 10% and 5% for UF and IC resins, respectively, based on the dry weight bases of the fiber content of composite panels. The UF resin was purchased from Sari Resin Manufacturing Company, Sari, Iran. It contained 10% of UF with a viscosity of 200-400 cP, 47 seconds of gel time, and a density of 1.277 g/cm³.

**Wollastonite and resin application**

Specimens were cut from the panels for physical and mechanical tests in accordance with the dimensions and procedures described in ISIRI Standard 9044 PB Type 2; this standard is compatible to procedures described in ASTM Standard D-1037 (table II). Two specimens for each physical and mechanical tests were cut from each panel replicate.

Center point loading tests were carried out on an Instron 4486 Universal Testing machine over a 380 mm span. The loading speed was adjusted at a rate of 2 mm per minute; data were registered to calculate modulus of elasticity (MOE) and modulus of rupture (MOR).

Internal bond strength was determined by gluing aluminum blocks to each face of the test sample using hot-melt glue; pulling force was then applied to the aluminum blocks. Internal bond strength (IB) was calculated as the maximum load required to fail the sample over the surface area.

Water absorption was measured by weighing samples before and after 2 or 24 hours of soaking in distilled water. Thickness swelling was determined on the same samples by measuring thickness at five pre-determined points on each sample before and after the water immersion. The five points comprised of four points on each corner, and one in the center.

**Statistical analysis**

A two-way analysis of variance (ANOVA) was carried out using SAS software, version 9.2 (2010) at a 95% level of confidence. Duncan multiple range test was then performed to determine groupings for each properties. Hierarchical cluster analysis, including dendrograms and Ward methods with squared Euclidean distance intervals, was carried out using SPSS/18 (2010). Fitted-line, contour, and surface plots were made using Minitab software, version 16.2.2 (2010) (Taghiyari et al., 2017).

**Results and discussion**

**Flexural properties**

The highest and lowest modulus of rupture (MOR) values were found in the W100-IC (16.3 MPa) and NW10%-IC (9 MPa) treatments, respectively (figure 2). Addition of palm fibers to panels also resulted in a decrease in MOR values of panels without NW content and in panels produced both with UF and IC resins. This was as a result of weaker strength of palm fibers. Addition of NW resulted in a decrease in MOR values in panels without date palm pruning residues, produced with both UF and IC resins. This was because NW particles absorbed part of the resin around themselves (Taghiyari et al., 2013a), preventing this part of resin to be actively involved in the process of sticking fibers together. However, addition of NW had an improving effect on MOR values in panels with 10% palm fiber content. It is hypothesized that NW could
better be integrated with palm fibers; however, chemical studies should be carried out to finalize the reasons for this improvement.

Results of the modulus of elasticity (MOE) tests demonstrated that the highest (1,411 MPa) and lowest (1,090 MPa) MOE values were found in the same treatments as the highest and lowest MOR values (figure 3). The addition of NW resulted in a decrease in panels without palm residues, and in an increase in MOE values of panels containing palm fibers; the fluctuations were not statistically significant in all cases. The increasing and decreasing trends were similar to MOR values, though the proportions were not the same for the addition of NW5% and NW10%.

**Internal bond strength**

The highest and lowest internal bond (IB) values were found in the Palm10%-NW10% with UF resin (0.43 MPa) and W100%-NW5% with IC resin (0.21 MPa) panels, respectively (figure 4). A general trend was obvious in that the addition of palm pruning residues resulted in an increase in IB values. This was as a result of higher slender ratio of palm leaves fibers (Hosseinkhani, 2015), making the core section of panels more integrated and more strongly bonded together. Similar improvement in IB values was also reported in MDF panels containing camel-thorn chips with higher slender ratio (Taghiyari et al., 2016a). However, no clear trend was found as a result of the addition of NW or palm residues, indicating that two mechanisms were simultaneously interacting. The first mechanism was the formation of bonds between NW components with wood cell wall polymers (Taghiyari et al., 2016b). Addition of palm to the mat did not have significant effect on WA values. As to the panels produced with IC resin, addition of NW and palm did not significantly affect WA values.

**Water absorption**

Results of WA measurement showed distinct difference between panels produced with IC resin in comparison to their UF-resin counterparts (figure 5). Addition of NW tended to decrease WA both in panels with IC and UF resins; the decreasing trend was more obvious in IC panels. This decrease was attributed to the formation of bonds between NW components with wood cell wall polymers (Taghiyari et al., 2016b). Addition of palm to the mat did not have significant effect on WA values. As to the panels produced with IC resin, addition of NW and palm did not significantly affect WA values.

**Thickness swelling**

Addition of NW and palm residues has nearly the same effects on thickness swelling (TS) values as they had on WA values (figure 6). TS values also illustrated distinctly lower values for panels produced with IC-resin in comparison to panels with UF-resin, though IC-resin
content was only half of UF-resin content in all treatments. In this connection, IC is classified as a WBP (weather and boil proof) adhesive; therefore, the significant difference between IC and UF demonstrated significant effect of resin type on the overall physical properties of composite panels.

**Cluster analysis**

Cluster analysis of the twelve treatments based on the physical properties showed clear distinct grouping of treatments with IC-resin versus treatments with UF-resin (figure 7A). Within-group clustering of either of resins showed very close grouping of the treatments (figure 7A). This implied the addition of NW and palm residues did not significantly affect dimensional stability and water absorption of panels. However, cluster analysis based on all physical and mechanical properties showed that although there was a clear distinction between the two resins, but within-group variations also occurred (figure 7B). This indicated that opposite to physical properties, mechanical properties of composite panels depended on a variety of factors involved in the production process like NW-content and palm residues. In other words, addition of NW and palm residues had significant effect on the overall mechanical properties of the panels; and they had both increasing and decreasing effects on different treatments, depending on the components of panels.

Contour plot of IB values versus WA24 and TS24 clearly showed an inverse relationship with both WA24 and TS24 properties (figure 8A). Surface plot also demonstrated decreasing trends of both WA24 and TS24 properties as the IB values increased (figure 9). Contour plots of IB versus MOR and MOE values illustrated that IB had a direct relationship with MOR values, indicating that as IB increased, so did MOR values. However, no particular trend could be observed between IB and MOE values. Fitted-line plot between WA24 and TS24 values showed rather high and statistically significant R-square (77%) between the two properties (figure 10). However, neither high nor significant R-square values were found between any other properties studied in this research project.
Conclusion

Medium-density fiberboard (MDF) panels were produced in the present research project with two different resins of urea-formaldehyde (UF) at 10% content, and isocyanate (IC) at 5% content. In order to find new sources of fibers to satisfy the increasing needs for raw material for composite industry, 10% palm leaf residues was added to the composite mat to be compared with those panels with pure wood fibers. Moreover nano-wollastonite was added to the mat at two contents of 5% and 10% to improve some of the drawbacks of composite panels. Results showed that IC-resin has a significantly lower water absorption and thickness swelling while mechanical properties varied case by case. Addition of NW had a decreasing effect on the mechanical properties of panels produced with UF resin, but it had an increasing effect on panels with IC resin. Addition of palm residues had a significant increasing effect on IB values in panels with both UF and IC resins. It was concluded that palm leaf residues can be considered a potential source of raw material for MDF panels produced with both UF and IC resins. Moreover, NW is also recommended to be added in composite panels with 10% of palm residues to compensate for part of the property losses.

Acknowledgments

The authors acknowledge constant scientific support of Prof. Olaf Schmidt from the University of Hamburg, as well as the support of Alexander von Humboldt Stiftung.
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Cirad - Campus international de Baillarguet, 34398 Montpellier Cedex 5, France - Contact : bft@cirad.fr - ISSN : L-0006-579X