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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).

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RÉSUMÉ

Effets de l'ajout de nano-wollastonite, de résidus de palmier-dattier et de deux types de résine sur les propriétés physiques et mécaniques de panneaux de fibres de bois à densité moyenne

L'étude porte sur les effets de l'ajout de nano-wollastonite (NW) et de résidus de palmier-dattier sur les propriétés physiques et mécaniques de panneaux de fibres à densité moyenne. Des résines d'urée-formaldéhyde (UF) et d'isocyanate (IC) ont été utilisées à 10 % et 5 %, respectivement, du poids sec du matériau pour produire les panneaux. NW à granulométrie ‹ 100 nm a été utilisé à 5 et 10 % du poids sec des résines, mélangé et pulvérisé sur le matériau avant formation du tapis de fibres. Les résultats ont été ensuite comparés à ceux pour les panneaux sans NW. Ils indiquent clairement une moindre absorption d'humidité et un moindre gonflement en épaisseur pour les panneaux produits avec ajout de résine IC. L'ajout de résidus de palmier-dattier a augmenté significativement les valeurs IB des panneaux produits avec les deux résines, UF et IC. L'ajout de NW a réduit les propriétés mécaniques des panneaux produits avec la résine UF, et amélioré celles des panneaux produits avec la résine IC. En conclusion, les résidus de palmier-dattier peuvent être considérés comme matière première potentielle pour la production de panneaux de fibres à moyenne densité en utilisant les deux résines, UF et IC. D'autre part, l'ajout de NW est recommandé pour la production de panneaux composites avec 10 % de résidus de palmier pour compenser en partie leur moindre résistance mécanique.

Mots-clés : minéraux, nanomatériaux, fibres naturelles, feuilles de palmier, panneaux de particules, coefficient de conductivité thermique, wollastonite, matériau de bois composite.

ABSTRACT

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

The effects of adding nano-wollastonite (NW) and date palm prunings on the physical and mechanical properties of medium-density fibreboard (MDF) 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 were 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, nano-materials, natural fibres, palm leaves, particleboard, thermal conductivity coefficient, wollastonite, wood-composite.

RESUMEN

Efectos de agregar nanowollastonita, residuos de palma datilera y dos tipos de resina en las propiedades fisicomecánicas de tableros de fibras de madera de densidad media

Se estudiaron los efectos de la adición de nanowollastonita (NW) y residuos de palma datilera en las propiedades físicas y mecánicas de tableros de fibras de densidad media. Para producir los tableros, se utilizaron resinas de urea formaldehído (UF) e isocianato (IC) al 10% y 5%, respectivamente, del peso seco del material. Se utilizó la NW, con un tamaño de partícula de ‹100 nm, al 5 y 10% del peso seco de las resinas, mezclándola y pulverizándola sobre el material antes de la formación de la manta de fibras. Posteriormente se compararon los resultados con los de tableros sin NW y éstos indican claramente una menor absorción de humedad y un menor hinchamiento en grosor de los tableros producidos con resina IC. La adición de residuos de palma datilera incrementó significativamente los valores IB de los tableros con ambas resinas, UF e IC. La adición de NW disminuyó las propiedades mecánicas de los tableros producidos con la resina UF y mejoró las de los tableros producidos con la resina IC. En conclusión, los residuos de palma datilera pueden considerarse como potencial materia prima para la producción de tableros de fibras de densidad media empleando ambas resinas, UF e IC. Además, se recomienda añadir NW para la producción de tableros compuestos con 10% de residuos de palma para compensar, en parte, su menor resistencia mecánica.

Palabras clave: minerales, nanomateriales, fibras naturales, hojas de palma, tableros de partículas, coeficiente de conductividad térmica, wollastonita, material de madera compuesta.

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; Hosseinpourpia *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 (Sheikholeslami *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 hyrcanum*), 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/m^3. Four replicate boards were made for each treatment and the panels were conditioned at 25±2° C and 42±3% relative humidity for three 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 leave preparation

Date palm pruned leaves from *Phoenix dactylifera* L. were cut and dried from Borazjan city (Dashtestan) located in Bushehr province in Iran (latitude: 29º 16' 11» N, longitude: 51º 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.

Table I.
Composition of the Wollastonite Gel (Taghiyari *et al.*, 2013bc) evaluated in the study.

Component	Proportion (% w/w)
CaO	39.77
SiO_2	46.96
Al_2O_3	3.95
Fe_2O_3	2.79
TiO_2	0.22
K_2O	0.04
MgO	1.39
Na_2O	0.16
SO_3	0.05
Water	4.67

Wollastonite and resin application

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/cm^3.

Table II.
Dimensions of specimens prepared for testing according to specifications of ISIRI Standard 9044 PB Type 2 (compatible to ASTM Standard D-1037).

Test	Sample Dimensions (mm)
Center Point Loading (MOR/MOE)	400 × 75 (loading span: 370)
Internal Bond Strength (IB)	75 × 75
Water absorption/Thickness Swelling	75 × 75

Physical and mechanical testing

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

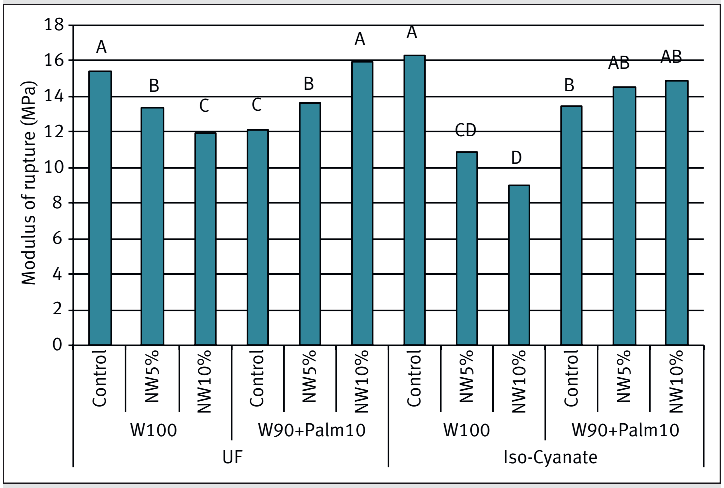

Figure 2.
Effects of addition of date palm pruning residues (Palm) and/or nano-wollastonite (NW) on modulus of rupture of MDF panels (values represent averages of eight replicates) (Letters on each column represent Duncan Multiple Range Groupings at 95% level of confidence).

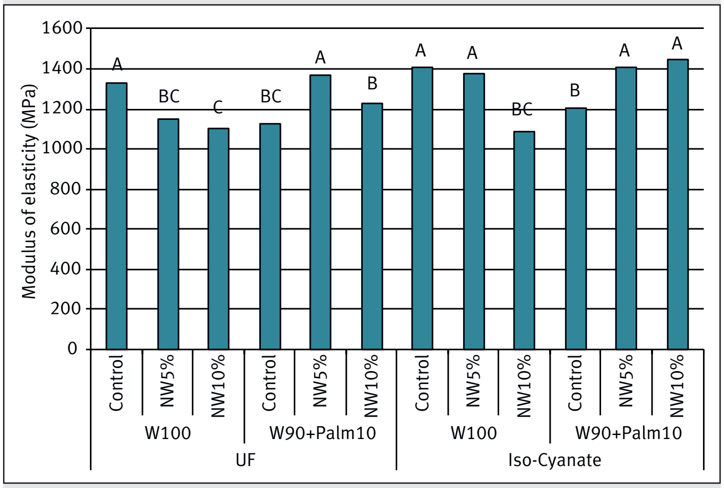

Figure 3.
Effects of addition of date palm pruning residues (Palm) and/or nano-wollastonite (NW) on modulus of elasticity of MDF panels (values represent averages of eight replicates) (Letters on each column represent Duncan Multiple Range Groupings at 95% level of confidence).

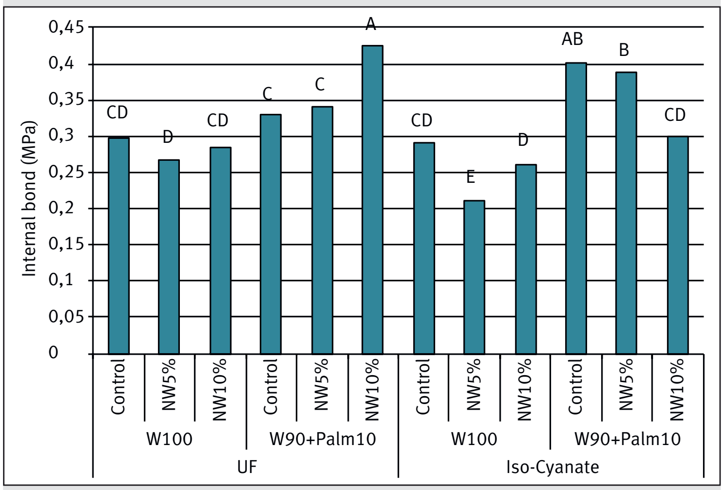

Figure 4.
Effects of addition of date palm pruning residues (Palm) and/or nano-wollastonite (NW) on internal bond of MDF panels (values represent averages of eight replicates) (Letters on each column represent Duncan Multiple Range Groupings at 95% level of confidence).

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 with cellulosic components of wood and palm fibers (Taghiyari *et al.*, 2016b), resulting in an increase in IB values. The second mechanism was the absorption of resin by NW particles, resulting in a decrease in IB 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

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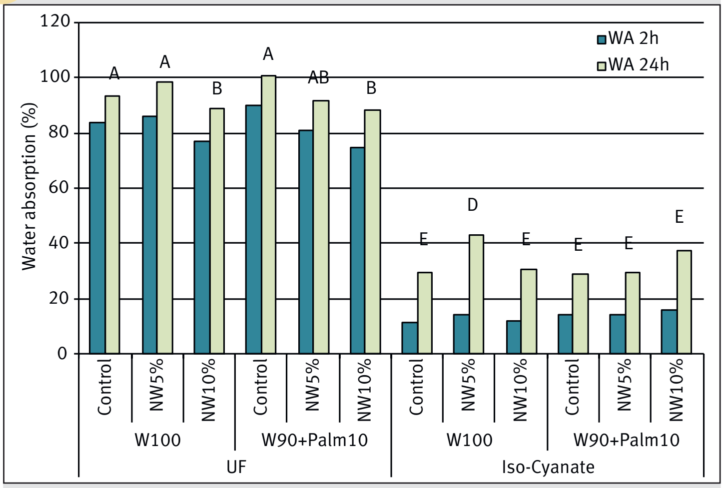

Figure 5.
Effects of addition of date palm pruning residues (Palm) and/or nano-wollastonite (NW) on water abruption (WA) of MDF panels (values represent averages of eight replicates) (Letters on each column represent Duncan Multiple Range Groupings at 95% level of confidence).

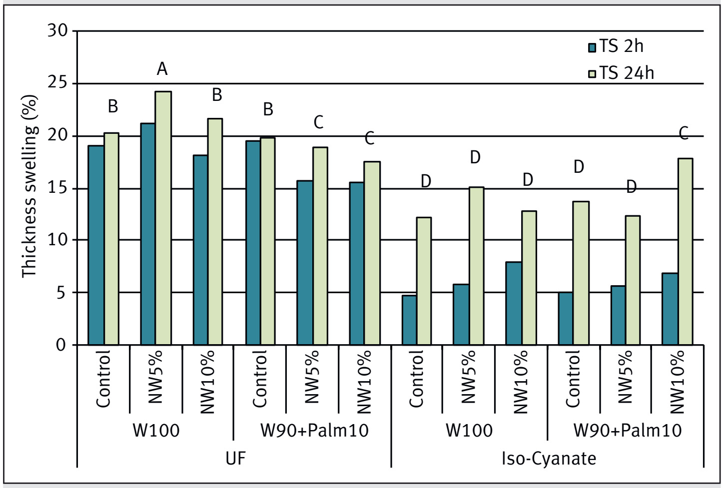

Figure 6.
Effects of addition of date palm pruning residues (Palm) and/or nano-wollastonite (NW) on thickness swelling (TS) of MDF panels (values represent averages of eight replicates) (Letters on each column represent Duncan Multiple Range Groupings at 95% level of confidence).

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

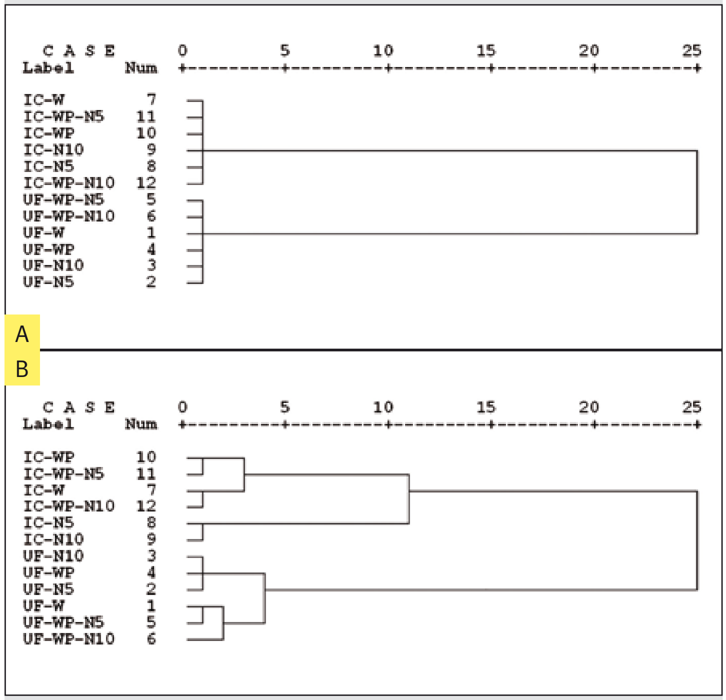

Figure 7.
Cluster analysis of the twelve treatments based on only physical (A) and all physical and mechanical properties (B) (IC=isocyanate resin; UF=urea-formaldehyde resin; W=wood; P=palm residues; N=nanowollastonite).

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 Re-square (77%) between the two properties (figure 10). However, neither high and nor significant R-square values were found between any other properties studied in this research project.

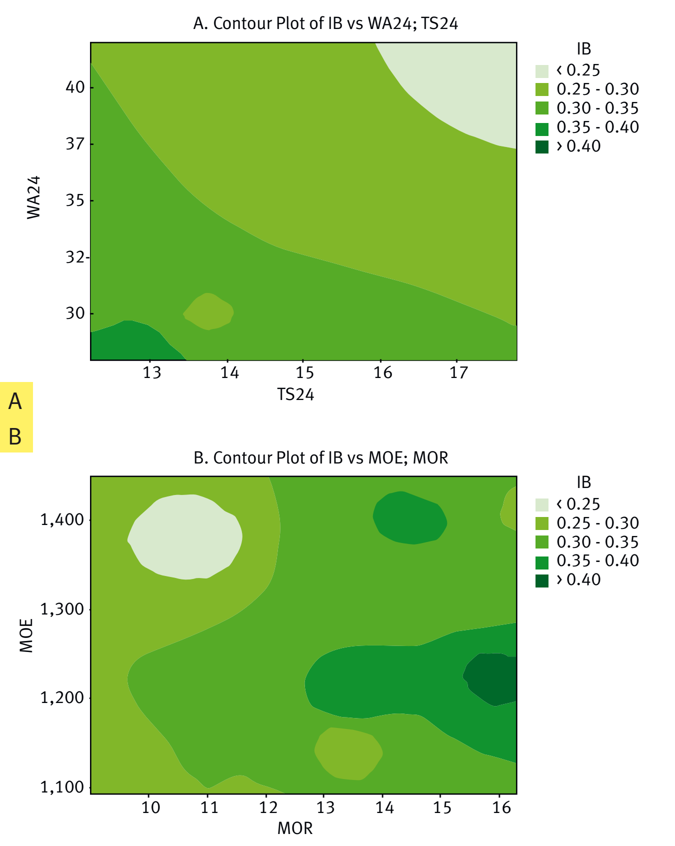

Figure 8.
Contour plots between internal bond (IB) versus water absorption (WA) and thickness swelling (TS) values after 24 hours of immersion in water (A), and between IB versus modulus of rupture (MOR) and modulus of elasticity (MOE) values (B).

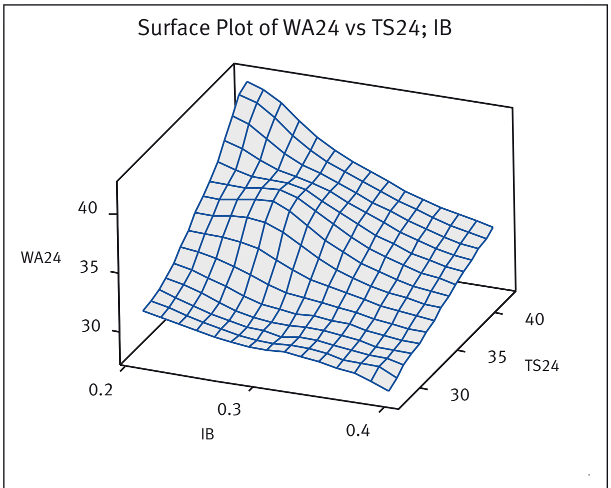

Figure 9.
Surface plot between internal bond (IB) versus water absorption (WA) and thickness swelling (TS24) values after 24 hours of immersion in water.

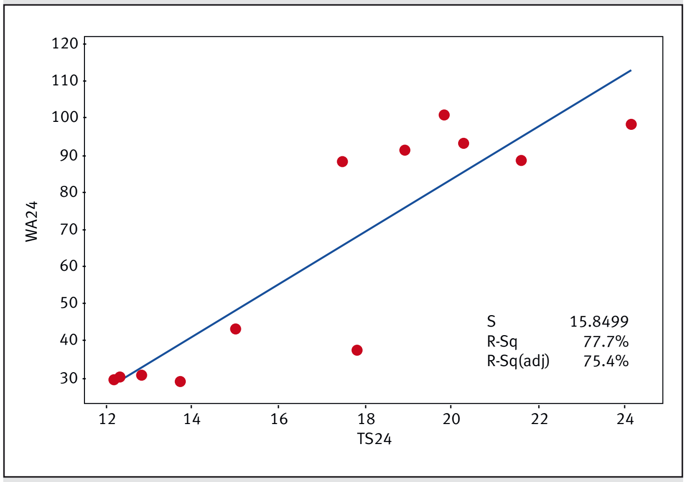

Figure 10.
Fitted-line plot between water absorption (WA) and thickness swelling (TS) values after 24 hours of immersion in water.

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 draw-backs 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.

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