UTILIZATION OF OLIVE MILL SLUDGE IN THE MANUFACTURE OF FIBERBOARD

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The objective of this research was to investigate the utilization of olive mill sludge (OMS) as an alternative to wood in the manufacture of the medium density fiberboard (MDF). The MDF panels were manufactured using standardized procedures that simulated industrial production at the laboratory. Six panel types were made from various mixtures of hardwood fiber/dried OMS flour, 100/0, 90/10, 80/20, 70/30, 60/40, and 50/50 (by weight) percents, respectively. With increasing OMS flour content, the flexural properties of the panels, modulus of rupture and modulus of elasticity, decreased by 31.0% and 29.2% as compared to panels without OMS flour, respectively. However, the water resistance was improved by the addition of the OMS flour up to 20 wt % content. For example, the thickness swelling and water absorption values of the panels containing 20% OMS flour were 17.3% and 59.5%, while they were found for the panels without OMS flour as 21.5% and 75.6%, respectively. The findings obtained in the study showed that the OMS was capable of serving as lignocellulosic raw material in the manufacture of the MDF.

Key words: Medium density fiberboard; Mechanical properties; Olive mill sludge; Physical properties; Recycling; Wood

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INTRODUCTION

The growing demand for wood-based panels has led to continuous efforts to find new resources as an alternative to wood. The use of other renewable resources such as agricultural residues in the production of composite panels (i.e., fiberboards and particleboards) has recently been considered attractive both from the economical and environmental point of view. Value-added lignocellulosic composites made from industrial waste can be considered as an alternative solution to this problem. One of these residuals is the olive mill sludge (OMS), which is the industrial waste produced in high quantities in the olive oil industry.

The OMS, wet olive pomace, is the solid and wet lignocellulosic by-product generated in massive quantities by olive-oil extraction industries that use the two-phase centrifugation method (Fig. 1). The OMS is obtained by pressing the fruit, leaving a residue of seed husks (fragmented olive stones), seed, pulp, and peel. The olive fruit consists of pulp (70 to 90%), stone (9 to 27%), and seed (2 to 3%) on a total weight basis (Alburquerque et al. 2004). The refined OMS differs from the crude OMS mainly by lower oil content and smaller water content because it has been dehydrated during the oil extraction process. The OMS oil is extracted from the crude OMS using chemical solvents, mostly hexane, and by heat. After the extraction process, the OMS has 0.1-0.3%

olive oil based on the oven-dry weight of the OMS. Typical chemical composition of OMS before the extraction process is presented in Table 1 (Vlyssides et al. 1998).

Fig. 1. Wet olive mill sludge (OMS) generated in massive quantities by olive-oil extraction in the olive oil mill

Table 1. The Chemical Composition of Olive Mill Sludge (OMS) (Two-Phase System)

The chemical composition	Value
Moisture content (%)	56.80 ± 2.20
Fat and oils (%)	4.65 ± 1.74
Proteins (%)	2.87 ± 0.01
Total sugars (%)	0.83 ± 0.01
Cellulose (%)	14.54 ± 0.17
Hemicellulose (%)	6.63 ± 0.37
Lignin (%)	8.54 ± 0.18
Ash (%)	1.42 ± 0.09
Kjeldahl nitrogen (%)	0.43 ± 0.006
Phosphorous as P_2O_5 (%)	0.04 ± 0.003
Phenolic compounds (%)	2.43 ± 0.15
Potassium as K_2O (%)	0.32 ± 0.027
Calcium as CaO (%)	0.37 ± 0.036
Total carbon (%)	25.37 ± 2.03
C/N ratio	59.68 ±5.25

The olive oil industry is very important in Mediterranean countries, both in terms of wealth and tradition. Spain is the main world producer, followed by Italy, Greece, Turkey, Syria, and Tunisia. Large amounts of the OMS are produced in the Mediterranean region, an area that accounts for 95% of the total olive oil production worldwide. Countries of the European Union (EU) such as Italy, Spain, and France produce about 70% of olive in the world. Turkey, Tunisia, Syrian Arab Republic, and Morocco are the other important producers. Spain, the leading olive-oil producer, produces about 4 million tones of the OMS per year (Fornes et al. 2009). According to the data of Institute of Olive Researches in Turkey, 15-22 kg of olive oil and 35-45 kg of the OMS are obtained for a 100 kg of olive (IOR 2009). This given amount of the OMS is on a wet basis. Although the majority of the OMS produced in evaporation ponds is disposed of in landfill sites (Hytiris et al. 2004), the remainder is used as a heat source because of its oil content in the Aegean and the Southeastern Anatolian region of Turkey (Varol and Atimtay 2007).

The total OMS production averages 200-250 thousand/year in Turkey (Varol 2006). With the continued generation of these residues, the need for proper utilization of viable management strategies is imperative. Ayrilmis and Buyuksari (2010) reported that OMS flour could be utilized as a reinforcing filler in manufacture of wood plastic composite. They concluded that a 40/60 (by weight) formulation of the OMS flour and polypropylene was a practical choice for applications requiring a higher water resistance such as outdoor deck flooring and window frames. However, an extensive literature search did not reveal any information about the utilization of the OMS as a raw material in the manufacture of wood-based composite. The objective of this study was to investigate physical and mechanical properties of MDF panels made from various mixtures of wood fiber and the OMS. Our secondary objective was to find a solution increasing raw material requirement for the manufacture of the MDF, which is commonly used in the furniture industry in Turkey. Some physical (density, thickness swelling (TS), and water absorption (WA)) and mechanical properties (modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond (IB) strength) of the experimental panels were determined according to European standards. The MDF panels were made from various mixtures of hardwood fiber/OMS flour, 100/0, 90/10, 80/20, 70/30, 60/40, and 50/50 percents, using a method currently used in the MDF industry.

EXPERIMENTAL

Materials

Olive mill sludge having a value of 0.1-0.3% olive oil based on the oven-dry weight of the OMS was supplied from a commercial olive mill manufacturer located in Edremit, Western Turkey. First the OMS samples were dried in an oven at 60°C for 10 h to moisture content of 20 to 30% based on the oven-dry OMS solid weight. Following the drying, the OMS was then processed by a rotary grinder without adding additional water. Finally, the OMS flour passing through a U.S. 35-mesh screen and was retained by a U.S. 80-mesh screen. The OMS flour was then dried in a laboratory oven at 100°C for 24-h to moisture content of 1 to 2%. Hardwood fibers, beech wood (*Fagus orientalis* Lipsky), were obtained from a commercial MDF plant, Kastamonu Integrated Wood Company, in Kastamonu, Turkey. The wood fibers were produced using a thermo-mechanical refining process without any chemical or resin. The moisture content of the fibers, as determined by oven-dry weight, was 2 to 3% prior to treatment. A commercial liquid urea-formaldehyde (UF) resin with 65% solid content (formaldehyde/urea: 1.25) was used as an adhesive in the manufacture of the experimental MDF panels. Ammonium chloride

(NH_4Cl) solution with 20% solid content was used as a hardener for the UF resin. The resin and ammonium chloride were supplied from Kastamonu Integrated Wood Company.

MDF Manufacture

MDF panels were manufactured at the laboratory using standardized procedures that simulated industrial production. After mixing wood fibers and the OMS flour and placing the mixture into a rotary drum blender, commercial liquid urea-formaldehyde (UF) resin containing 1% ammonium chloride was applied to the mixture, with an air-atomized metered spray system at 10% (based on the weight of the oven dried raw material) in a rotating blender. The mixture was weighed and then formed into a mat on an aluminum caul plate, using a 400 mm x 400 mm forming box. When all the wood fiber/OMS flour mixture had been put into the forming box, the mats were manually precompressed. To reduce the mat height and to densify the mats, they were subjected to cold pressing. This procedure allowed for easy insertion of the mats into the hot-pressing. The fiber mats at average 10% moisture content based on the oven dry weight of fiber were subjected to hot-pressing. The press temperature, maximum panel pressure, and total press cycle were 180°C, 3.5 N/mm^2, and 8 min, respectively. The MDF panels were then trimmed to a final size of 380 mm x 380 mm x 10 mm following the cooling process. Table 2 shows the experimental design.

Table 2. Experimental Design

MDF panel type	Wood fiber and OMS formulations for the MDF (by weight %)	
	Wood fiber (%)	OMS (%)
A	100	0
B	90	10
C	80	20
D	70	30
E	60	40
F	50	50

A total of 12 experimental panels, two for each type of panel, were made in the laboratory. The density values of the MDF panels were between 0.66 g/cm^3 and 0.69 g/cm^3. Prior to physical and mechanical tests, all the samples were conditioned for three weeks at 20°C and 65% relative humidity until no changes in the weights were detected by regular weighing of the samples. The MDF panels made from 100% beech wood fibers were considered as control panels.

Determination of Water Resistance

Water resistance of the panels, TS and WA, was evaluated according to the EN standard. Fifteen samples, 50 mm × 50 mm × 10 mm, from each type of panel type were used for the TS and WA properties. The samples were immediately weighed. The sample thickness was determined by taking a measurement at a specific location, the diagonal crosspoint, on the sample. After 24-h of submersion, the samples were drip-dried for 10 min, wiped clean of any surface water, and weighed. The density and water resistance of the samples were evaluated according to the test methods and requirements of EN 323 (1993) and EN 317 (1993), respectively.

Determination of Mechanical Properties

Tests of the flexural properties, the MOR and MOE, were conducted according to EN 310 (1993). A total of 12 samples with dimensions of 250 mm x 50 mm x 10 mm, six parallel and six perpendicular to the panel surface, were tested for each panel type to determine MOR and MOE. The flexural samples were tested on a Losenhausen Universal testing system equipped with a load cell with a capacity of 10 kN. The MOR test was conducted in accordance with the third point loading method at a span-to-depth ratio of 20:1. The crosshead speed was adjusted so that failure would occur within an average of 60 s. Load-deflection data for the calculation of the sample's MOE were recorded at the 10% and 40% values of failure load (P_{max}). The IB tests were conducted on the samples according to EN 319 (1993). Ten samples, 50 mm x 50 mm x 10 mm, from each type of panel were used for the IB strength. The load was continuously applied to the samples throughout the tests at a uniform rate of motion of the movable cross-head of the testing machine of 1.2 mm/min until failure occurs.

Statistical Analysis

For the physical and mechanical tests, all multiple comparisons were first subjected to an analysis of variance (ANOVA) at $p<0.01$ and significant differences between the mean values of the MDF panels samples were determined using Duncan's multiple range test.

RESULTS AND DISCUSSION

Physical Properties

Table 3 shows the average density, TS, and, WA values of the MDF panels containing OMS flour. The differences between the treatment groups and the test results are shown in Table 3 as letters. All panel types showed statistically significant differences ($p < 0.01$) in the TS and WA values from each other. Panel type C had the lowest TS and WA values, while the highest values were found for panel type F.

Table 3. The Average Values of the Physical Properties of the MDF panels

MDF Panel Type	Physical Properties		
	Density (g/cm^3)	Thickness swelling (TS) (24-h) (%)	Water absorption (WA) (24-h) (%)
A	0.66 (0.04)	21.5 (1.6) a[1]	75.6 (5.3) a
B	0.68 (0.07)	19.1 (2.2) b	67.3 (4.5) b
C	0.67 (0.03)	17.3 (1.5) c	59.5 (3.8) c
D	0.67 (0.03)	22.6 (2.0) d	71.4 (6.5) d
E	0.69 (0.05)	23.7 (2.3) e	80.9 (4.4) e
F	0.67 (0.04)	24.8 (2.8) f	88.5 (6.8) f
Quality requirements for MDF	> 0.60[2]	Max. 15[3]	-

[1] Groups with same letters in column indicate that there was no statistical difference ($p < 0.01$) between the samples according to Duncan's multiply range test. Values in parentheses are standard deviations.

[2] Quality requirement for dry-process fiberboards (MDF) according to EN 316 (1999).

[3] Quality requirements for general-purpose MDF panels for use in dry conditions and nominal thickness > 9 mm to 12 mm) according to EN 622-5 (2006).

The TS values of all the MDF types did not meet maximum requirement of TS (max. 15%) after 24-h of submersion for general purpose MDF panels of EN 622-5 (2006) Standard. High swelling could be mainly attributed to the fact that no wax or other hydrophobic substance was used during the MDF manufacture.

Water resistance of the MDF panels was improved by the addition of the OMS flour up to 20 wt %. However, further additions decreased the water resistance. The results of the present study were in agreement with the literature. Shi et al. (1999) reported that different sizes of polymeric fluff to MDF panels improved their water resistance. Nemli (2003) found that the addition of 10 wt % wood dust into particleboard improved its thickness swelling. In another study, Ayrilmis et al. (2009) stated that water resistance of MDF panels was improved by addition of 10 wt % cone flour but further additions adversely affected the water resistance of the panels. These studies reported that small particles could fill the spaces between fiber or particles and act as matrix material to improve adhesion properties. The similar results were also observed in this study. With the addition of 20 wt % OMS flour in the MDF, the average TS and WA values decreased by 19.5% and 21.3% as compared to the values of the MDF panels made from 100 wt % wood fibers, respectively.

Mechanical Properties

The MOR and MOE of the MDF panels decreased with increasing OMS flour content (Table 4). The MDF panels made from 100 wt % wood fibers (panel type A) had the highest MOE and MOR values while the lowest values were found for the panels containing 50 wt % OMS flour (panel type F) (Table 4). All the panel types did not met MDF minumum requirements (22 N/mm^2 and 2500 N/mm^2) for use in dry conditions of EN 622-5 (2006). The MOR and IB values of the panels containing 10 wt % OMS flour were slightly lower than the minimum values stated in EN 622-5.

Table 4. The Average Values of the Mechanical Properties of the MDF Panels

MDF Panel Type	Mechanical Properties		
	Modulus of rupture (MOR) (N/mm^2)	Modulus of elasticity (MOE) (N/mm^2)	Internal bond strength (IB) (N/mm^2)
A	21.6 (0.88) a[1]	2370 (125) a	0.49 (0.15) ab
B	21.3 (1.05) a	2243 (132) b	0.57 (0.14) c
C	18.7 (0.91) b	2205 (164) b	0.52 (0.10) b
D	17.4 (0.85) c	1985 (146) c	0.47 (0.18) a
E	16.3 (1.16) d	1827 (152) d	0.41 (0.15) d
F	14.9 (0.94) e	1678 (161) e	0.37 (0.12) e
Quality requirements for MDF	Min. 22.0[2]	Min. 2500[2]	Min. 0.60[2]

[1] Groups with same letters in column indicate that there was no statistical difference ($p < 0.01$) between the samples according to Duncan's multiply range test. Values in parentheses are standard deviations.

[2] Quality requirements for general-purpose MDF panels for use in dry conditions and nominal thickness > 9 mm to 12 mm) according to EN 622-5 (2006).

With the addition of the OMS flour, the MOR and MOE values of the MDF panels decreased by 31.0% and 29.2%, respectively. This was mainly attiributed to the geometry of the OMS flour. The flexural properties of the MDF panels are strongly influenced by the properties of their constituent materials, their volume ratio, and the interaction among them. Especially, physical and mechanical properties of the individual wood fibers, and the manner in which these components combined in structure significantly affect bending properties of the MDF. The MDF panels having high bending strength can be obtained by using a homogeneous material with a high degree of slenderness (ratio of the fiber length to the diameter of the fiber) of 150 (Maloney 1977). The OMS flour consists of almost round particles, and their aspect ratios are very low as compared to beech wood fiber which is 49. The average fiber length and diameter of beech wood are 1.1 mm and 0.0224 mm, respectively (Istek et al. 2005).

It is generally accepted that longer fibers achieve an increased network system by themselves and result in increased bending properties of the fiberboard. The lower MOR and MOE of the MDF panels containing the OMS flour can be explained by the small size of the OMS flour in the structure, resulting in low fiber ratios and ultimately leading to poor physical fiber-to-fiber contact). These results were in agreement with previous studies (Hill 1978; Lee et al. 2006; Copur et al. 2008; Ayrilmis et al. 2009). For example, Lee et al. (2006) reported that static bending properties and tensile strength of bagasse-based fiberboards increased as slenderness ratio of bagasse fiber bundles and particles increased from 3 to 26.

Wood is a lignocellulosic material made up of three major constituents (cellulose: 42-44%, hemicelluloses: 27-28%, and lignin: 24-28%) with some minor constituents (extractives: 3-4%) (Walker 2006). The major portion of the wood is crystalline cellulose, which is higher than that of OMS flour. The aligned fibril structure of the cellulose along with strong hydrogen bond has high stiffness thus addition of the OMS flour can decrease the stiffness of the fiberboard. The lower MOR and MOE values of the MDF panels containing higher amounts of the OMS flour can be related to lower amounts of cellulose and lignin materials in the cell walls of the OMS flour (Table 1).

The maximum value of the IB strength was reached at 10 wt % OMS flour content and then decreased as the OMS flour content reached 50 wt %. The IB values of the panel types B and C were 16.3% and 6.1% higher than that of the control group, respectively. However, further additions decreased the IB strength. This was attributed to the fact that small particles could easily fill the space, had high surface area to bond, and provided better adhesion. As a consequence, it could be concluded that the adhesion bond between the surfaces in the MDF was improved by the addition of 10 wt % OMS as much interaction occurred between the surfaces. The reason for the lower IB strength above 10 wt % OMS flour content can be attributed to the weak adhesion between the wood fibers and OMS flour. In this case, by increasing the resin level depending on the OMS flour content in the panel, the IB strength of the MDF panels can be improved. Another reason for the low IB value of the MDF panels containing above 10 wt % OMS was that with higher OMS content, there was more specific surface area and less resin content for surface area unit. In addition, the low internal bond strength of panels at higher levels of the OMS flour can also be due to its less fibrous anatomical structure as compared to the wood fibers. Similar results were also observed in previous studies (Shi et al. 1999; Nemli 2003; Copur et al. 2008; Ayrilmis et al. 2009). Although all the panel types did not meet the minimum IB requirement (0.60 N/mm^2) for use in dry conditions of EN 622-5 (2006), the IB strength of panel type B was slightly lower than the standard value.

CONCLUSIONS

The following general conclusions were drawn from the present study:

1. With increasing OMS flour content up to 20 wt %, the TS and WA values of the MDF panels decreased by 19.5% and 21.3% as compared to the MDF panels made from 100 wt % wood fibers. However, further additions decreased the water resistance.
2. The MOR and MOE of the MDF panels decreased with increasing OMS content. Depending on the OMS flour content, the MOR and MOE values of the MDF panels decreased by 31.0% and 29.2%, respectively.
3. The IB values of the panel types B and C were 16.3% and 6.1% higher than that of the MDF panels made from 100 wt % wood fibers, respectively.
4. The findings obtained in the study showed that the OMS was capable of serving as lignocellulosic raw material in the manufacture of the MDF.

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