New Renewable and Biodegradable Fiberboards from a Coriander Press Cake

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ABSTRACT: New fiberboards were manufactured from a coriander cake through thermo-pressing, and the influence of thermo-pressing conditions (temperature, pressure and time) on the boards' mechanical properties, their thickness swelling and their water absorption was evaluated. Because the protein glass transition systematically occurred during molding, this resulted in effective wetting of the fibers. Consequently, all boards were cohesive, with proteins and fibers acting as binder and reinforcing fillers, respectively. Flexural properties were influenced by all tested conditions, and the optimal board was molded at 200 °C temperature, 36.8 MPa pressure and 180 s time. Its flexural strength at break and its elastic modulus were 11.3 MPa and 2.6 GPa, respectively, with the highest surface hardness of the entire study. Simultaneously, thickness swelling and water absorption were low: 51% and 33%, respectively. This board would be applicable as pallet interlayer sheeting for the manufacture of containers or furniture or in the building trade.

KEYWORDS: Coriander press cake, fiberboard, lignocellulosic fibers, proteins, thermo-pressing

1 INTRODUCTION

Coriander (*Coriandrum sativum* L.) is an annual herb native to the eastern Mediterranean. It is commonly used as a condiment or a spice, while the fruit has been used as a traditional medicine since antiquity. Indeed, coriander has been shown to exhibit a wide range of biological activities [1]. Because the coriander fruits contain both a vegetable oil and an essential oil fraction, they are particularly interesting. The main fatty acid in *Coriandrum sativum* vegetable oil is petroselinic (6Z-octadecenoic) acid, representing up to 75% of the fatty acid profile. Petroselinic acid is a rather rare positional isomer of oleic acid. It portrays some interesting properties, giving it potential for food, cosmetic and pharmaceutical industries [2, 3]. Moreover, vegetable oil from coriander fruits has recently been labeled as a novel food ingredient [4]. It is now considered safe to use as a food supplement for healthy adults, at a maximum level of 600 mg per day (i.e., 8.6 mg/kg bw per day for a 70 kg person). Therefore, the development of a sustainable new process for the extraction of vegetable oil from coriander fruits with the valorization of byproducts will be a major challenge for the years to come.

Industrial oil extraction from oilseeds is usually carried out by mechanical pressing, followed by solvent extraction with *n*-hexane. Over the last twenty years, there has been much research focusing on continuous oil extraction by mechanical pressing using extrusion technology [5–22]. In the case of coriander fruits, both single- [18, 21] and twin-screw extruders [19, 22] were used to successfully conduct the extraction of vegetable oil.

The most recent study identified the screw configuration, the device's filling coefficient, and the pressing temperature as the main factors influencing the oil extraction efficiency [22]. The highest oil recovery of the study (i.e., 47%) with concurrent low energy consumption was obtained from a pressing zone positioned immediately after the filter, and consisting of 50 mm long, reverse screws with a –33 mm pitch, with an extruder filling coefficient of 39.4 g/h rpm and a pressing temperature of 120 °C. The pressed oil

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obtained was of acceptable quality (<1.5% acidity) and pleasantly scented, leading to the conclusion that part of the essential oil was co-extracted with the vegetable oil.

After vegetable oil extraction from coriander fruits by mechanical pressing using extrusion technology, the residual oil content in the press cakes is at least 15% of the dry matter [18, 19, 22]. This could be a disadvantage for some applications, although the residual oil in the cakes might favor their conversion into usable energy through combustion, gasification or pyrolysis [23, 24].

The press cakes also contain part of the essential oil from the fruits. However, its content is never more than 0.3% of the dry matter [21, 22]. The residual essential oil in the press cakes could be extracted by means of hydrodistillation. Due to its fresh and flowery scent, it could be used in cosmetics, perfumes, shampoos, soaps and even household detergents [25], or as a mosquito repellent [26]. However, because of the low quantities of volatile oil that could be extracted, it might be more interesting to leave the volatile oil within other end products, such as agromaterials, providing them with an added value.

Another possible use of the press cakes could be the isolation of some natural antioxidants through methanolic extraction, these being potentially interesting due to their beneficial impact on health [22]. The press cakes could further be incorporated into biodegradable polymers, such as polycaprolactone and poly(lactic acid), thus acting as reinforcing fillers for the biocomposite industry [27, 28]. Finally, as renewable resources containing both proteins and fibers, the press cakes could be considered as natural composites, thus being usable for the production of biodegradable and value-added agromaterials through thermo-pressing [29–36].

This study aimed to manufacture novel renewable and biodegradable fiberboards through thermo-pressing of a coriander cake generated during vegetable oil extraction from the fruits by mechanical pressing, using a single-screw extruder. Further, it presents an evaluation of the influence of the thermo-pressing conditions (temperature, pressure and time) on the boards' mechanical properties (flexural properties, Charpy impact strength and Shore D surface hardness), their thickness swelling and their water absorption.

2 EXPERIMENTAL

2.1 Materials

Molding of the fiberboards was conducted using a press cake originating from the mechanical pressing of coriander fruits of French origin using an OMEGA 20 (France) single-screw extruder. Its residual oil content was 17.2 ± 0.1% of the dry matter (Table 1) instead of 27.7 ± 0.6% for the coriander fruits (French standard NF V 03-908). This results in an oil recovery (R_{L1}), based on the residual oil content of the press cake, of 47.6 ± 0.3%, which was calculated from the following formula:

$$R_{L1} = \frac{(Q_S \times L_S) - (Q_C \times L_C)}{Q_S \times L_S} \times 100 \quad (1)$$

where Q_S is the inlet flow rate of coriander fruits (kg/h), Q_C the flow rate of the press cake (kg/h), L_S the oil content of the fruits (%), and L_C the residual oil content of the press cake (%).

Before thermo-pressing, the press cake was crushed using an Electra F3 (France) hammer mill fitted with a 15 mm screen. One thermo-pressing condition was also used to produce a composite fiberboard made from a mixture of the press cake and coriander straw harvested at the same time as the fruits, i.e., at plant maturity. Before being manually mixed with the press cake, the coriander straw was crushed using an Electra F3 (France) hammer mill fitted with a 7.5 mm screen.

2.2 Analytical Methods

Moisture contents, mineral contents, oil contents and protein contents were determined according to the French standards NF V 03-903, NF V 03-322, NF V 03-908 and NF V 18-100, respectively. An estimation of the three parietal constituents (cellulose, hemicelluloses and lignins) contained in the solids was made using the ADF-NDF method of Van Soest and Wine [37, 38]. An estimation of the water-soluble components contained in the solids was made by measuring the mass loss of the test sample after 1 h in boiling water. All determinations were carried out in duplicate.

2.3 Particle Size Distribution

The extrusion press cake was examined with a Nachet France Z 45 P (France) × 15 binocular magnifier. Six different photographs were taken and analyzed using the Archimed 4.0 (France) software. The particle size distribution was determined through manual measurements of the diameter of all particles on the photographs, using the ImageJ (USA) software. The tapped densities of both press cake and coriander straw were measured in a Granuloshop Densitap ETD-20 (France) volumenometer with simultaneous measurements of the corresponding apparent densities, i.e., before compaction.

2.4 TGA Measurements

Thermogravimetric analyses (TGA) of the press cake and neat pressed coriander oil were performed with a

Shimadzu TGA-50 (Japan) analyzer. Dynamic analysis was conducted under air at a heating rate of 5 °C/min, from 20 to 750 °C. Before analysis, the samples of press cake were equilibrated in a climatic chamber (60% RH, 25 °C) for three weeks. For all measurements, the mass of the test sample was around 10 mg for the press cake and around 8 mg for the coriander oil. The sample weights were measured as a function of temperature and stored. These data were later used to plot the percentage of undegraded sample (1–*D*) (%) as a function of temperature, where

$$D = \frac{W_0 - W}{W_0}, \quad (2)$$

with W_0 and W the weights at the starting point and during scanning, respectively. All measurements were carried out in duplicate.

2.5 DSC Measurements

Differential scanning calorimetry (DSC) of the press cake was performed from a deoiled material with a Mettler Toledo DSC 1 STARe System (Switzerland) power compensation calorimeter. The purge gas used was nitrogen of analytical quality at a flow rate of 50 mL/min. Temperature and energy calibration was carried out with zinc (T_f = 419.5 °C), indium (T_f = 156.6 °C) and distilled water (T_f = 0 °C) before the beginning of the tests. The analyses were performed with hermetic 120 µL stainless steel capsules (plus an empty reference capsule) fitted with O-rings resistant to an internal pressure of 20 bar (Mettler Toledo). They were carried out at a heating speed of 5 °C/min from 25 to 250 °C. Before analysis, the deoiled press cake was dried in a ventilated oven (60 °C, 12 h). Sample mass was around 10 mg and measurements were made in triplicate. The treatment of the obtained data was carried out using the STARe software (Mettler Toledo).

2.6 DMTA Measurements

Dynamic mechanical thermal analysis (DMTA) of the press cake was used to evaluate its thermo-mechanical behavior. It was performed from the deoiled material with a Triton Technology Tritec 2000 (UK) DMTA analyzer. The pre-dried powder was placed in a small metallic container (30.0 mm × 7.0 mm × 1.3 mm), inert from the point of view of relaxations. DMTA measurements were realized according to the two points bending technique with a 1 Hz frequency, a 50 µm displacement, and a 3 °C/min heating rate on a −50 to 180 °C range. Distance between the two points was 10 mm.

2.7 Thermo-pressing

The coriander press cake was molded through thermo-pressing inside an aluminum mold, using a 400 ton capacity Pinette Emidecau Industries (France) heated hydraulic press and producing 150 mm square fiberboards. The mold was equipped with vents to allow the expression of residual oil from the press cake during molding. The press cake was dried in a ventilated oven (60 °C, 12 h) before molding, in order to minimize vapor generation during thermo-pressing and so to restrict the risk of defects such as blisters inside the fiberboards [36]. On molding, the moisture content of the press cake was 2.1 ± 0.1%. The mass of the press cake was 200 g for all tested thermo-pressing conditions. These comprised 160–200 °C mold temperature, 24.5–49.0 MPa applied pressure and 60–300 s molding time (Table 2). Moreover, two additional boards (board number 12 and board number 13) were manufactured using the molding conditions of board number 7, i.e., 200 °C mold temperature, 36.8 MPa applied pressure and 180 s molding time. On the one hand, board number 12 was produced using 400 g press cake instead of 200 g. On the other hand, board number 13 was obtained using a manual mixture of 200 g press cake and 36.8 g coriander straw (i.e., 15% mass content). The latter was also dried in a ventilated oven (60 °C, 12 h) before molding, and its moisture content was then 3.1 ± 0.2%. Two fiberboards were manufactured for each of the tested thermo-pressing conditions. The first one was used to assess the mechanical properties for bending, while the second one was used for measuring Shore D surface hardness, Charpy impact strength, thickness swelling, and water absorption.

The oil recovery during molding was calculated from the following formulae:

$$R_{L2} = \frac{\left(m_C \times L_C\right) - \left(m_{FB} \times L_{FB}\right)}{m_C \times L_C} \times 100 \quad (3)$$

where R_{L2} is the oil recovery during molding relative to the residual oil contained in the press cake (%), m_C the mass of press cake used for thermo-pressing (g), m_{FB} the mass of equilibrated fiberboard (g), and L_{FB} the oil content of the fiberboard (%).

$$R'_{L2} = R_{L2} \times \frac{100 - R_{L1}}{100} \quad (4)$$

where R'_{L2} is the oil recovery during molding relative to the total amount of oil in the coriander fruits (%), and R_{L1} is the oil recovery in the single-screw extruder based on the residual oil content of the press cake (%).

The total oil recovery (extraction of the oil from coriander fruits in the single-screw extruder and expression

of the residual oil from the press cake during molding) was calculated from the following formula:

$$R_{LT} = R_{L1} + R'_{L2} = R_{L1} + \left(R_{L2} \times \frac{100 - R_{L1}}{100} \right) = \left(R_{L1} \times \frac{100 - R_{L2}}{100} \right) + R_{L2} \quad (5)$$

where R_{LT} is the total oil recovery through single-screw extrusion and press cake molding, relative to the total amount of oil that the coriander fruits contain (%).

2.8 Mechanical Properties for Bending

An Instron 33R4204 (USA) universal testing machine fitted with a 500 N load cell was used to assess the flexural properties of the test specimens according to the French standard NF EN 310, including breaking load (F), flexural strength at break (σ_f) and elastic modulus (E_f). The test specimens were 150 mm long and 30 mm wide. Their thickness was measured at three points with a digital sliding caliper with a 0.01 mm resolution, and the mean value (t) was recorded to calculate their volume and section. All specimens were weighed to calculate their mean apparent density (d). The test speed was 2 mm/min and the grip separation was 100 mm. Test specimens were cut, and equilibrated in a climatic chamber (60% RH, 25 °C) for three weeks before being tested. All determinations were carried out through four repetitions.

2.9 Charpy Impact Strength

A 0-40 daN cm Testwell Wolpert (France) Charpy machine was used to assess the impact strength of the unnotched test specimens according to the French standard NF EN ISO 179, including the absorbed energy (W) and the resilience (K). The test specimens were 60 mm long and 10 mm wide. Their thickness was measured at three points with an electronic digital sliding caliper with a 0.01 mm resolution, and the mean value (t) was recorded to calculate their section. Impact strength measurements were made at 23 °C using the three-point bending technique, with a grip separation of 25 mm. Test specimens were cut, and equilibrated in a climatic chamber (60% RH, 25 °C) for three weeks before being tested. All determinations were carried out through sixteen repetitions.

2.10 Shore D Surface Hardness

A Bareiss (Germany) durometer was used to assess the Shore D surface hardness of the fiberboards according to the French standard NF EN ISO 868. Fiberboards were equilibrated in a climatic chamber (60% RH, 25 °C) for three weeks before being tested. All determinations were carried out through 48 repetitions (24 repetitions for each side of the fiberboard).

2.11 Thickness Swelling and Water Absorption

Four 50 mm × 50 mm samples were used to determine thickness swelling (TS) and water absorption (WA) of the fiberboards. For this, samples were submerged in distilled water at 25 °C for 24 h. TS was determined according to the French Standard NF EN 317 and the thickness of each sample was measured at four points, midway along each side 10 mm from the edge before and after soaking in distilled water. Each sample was also weighed to an accuracy of 0.01 g to determine WA values.

3 RESULTS AND DISCUSSION

3.1 Physicochemical Characterization of the Press Cake

The press cake used in this study originated from the extraction of vegetable oil from coriander fruits of French origin through mechanical pressing, using an OMEGA 20 single-screw extruder. Its residual oil content was 17.2% of the dry matter (Table 1). This led to an oil recovery in the single-screw extruder (R_{L1}), based on the residual oil content in the press cake, of 47.6%. In addition to lipids, the press cake consisted of a mixture of proteins (18% of the dry matter) and lignocellulosic fibers (54% of the dry matter), including cellulose, hemicelluloses and lignins (Table 1). Therefore, the press cake could be considered as a natural composite. It further contained minerals and water-soluble components, representing 6% and 16% of the dry matter, respectively. Before

Table 1 Chemical composition (% of the dry matter) and apparent and tapped densities (g/cm^3) of the press cake and the coriander straw.

Raw material	Press cake	Coriander straw
Chemical composition (% of the dry matter)		
Minerals	6.2 ± 0.0	4.7 ± 0.2
Lipids	17.2 ± 0.1	1.6 ± 0.0
Proteins	18.2 ± 0.3	2.8 ± 0.0
Cellulose	26.6 ± 0.5	51.5 ± 0.3
Hemicelluloses	22.2 ± 0.4	15.0 ± 0.4
Lignins	5.0 ± 0.1	13.6 ± 0.3
Water-soluble components	16.0 ± 0.1	10.7 ± 0.3
Densities (g/cm^3)		
Apparent density	0.416 ± 0.004	0.072 ± 0.002
Tapped density	0.482 ± 0.003	0.081 ± 0.001

Results in the table correspond to the mean values ± standard deviations.

thermo-pressing, the press cake was crushed using an Electra F3 hammer mill fitted with a 15 mm screen. After milling, it consisted of almost spherical particles with a mean diameter of around 170 µm (Figure 1). Its apparent and tapped densities were 0.416 and 0.482 g/cm^3, respectively (Table 1).

A thermogravimetric analysis of the press cake was conducted and the TGA degradation curve under air appears in Figure 2. An initial mass loss was observed at 100 °C corresponding to water evaporation. The moisture content of the equilibrated press cake was 7.8 ± 0.1%, and the mass loss observed in the TGA curve corresponded to approximately the same mass percentage. The thermal degradation of organic compounds occurred in two main stages. The first degradation phenomenon (between 225 and 350 °C) led to a loss of approximately 50% of the dry matter in the sample. Moreover, two different subgroups of organic compounds were degraded in this temperature range. Indeed, the dTGA curve revealed two different peaks, associated with thermal degradation midpoints of 294 °C and 330 °C, respectively. The second degradation phenomenon was observed at much higher temperatures, i.e., between 450 and 525 °C. However, it was associated with a lower mass loss (around 30% of the dry matter in the sample).

Literature data provided some information on the thermal degradation of fibers. Hemicelluloses degrade around 270–330 °C, and then cellulose, around 320–380 °C. Finally, lignins have a wide peak in the range from 200 to 450 °C [39–42]. Moreover, thermal degradation of vegetable proteins has also been observed from an industrial sunflower ca

ke, above 250 °C and below 350 °C [43]. Further, a TGA analysis was conducted from neat pressed coriander oil and revealed one main degradation phenomenon that led to a loss of nearly 90% of the test sample mass. The corresponding onset and midpoint were

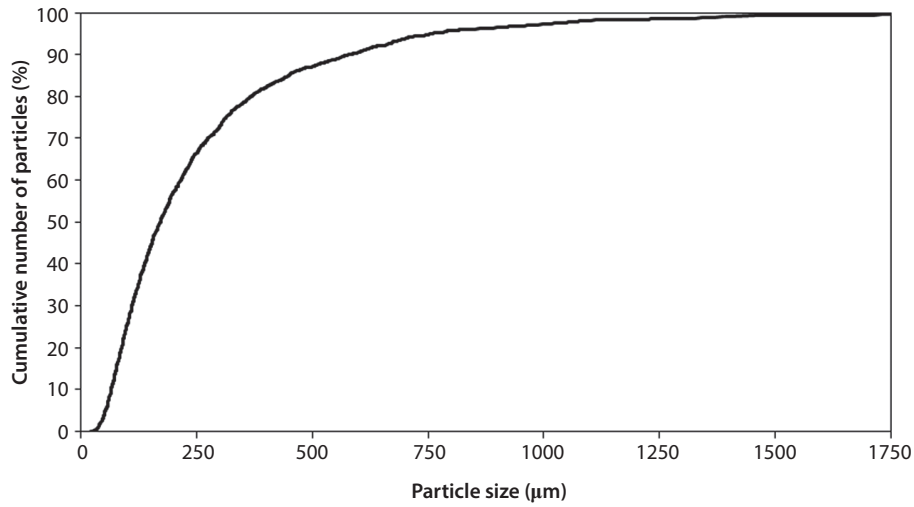

Figure 1 Particle size distribution in the press cake.

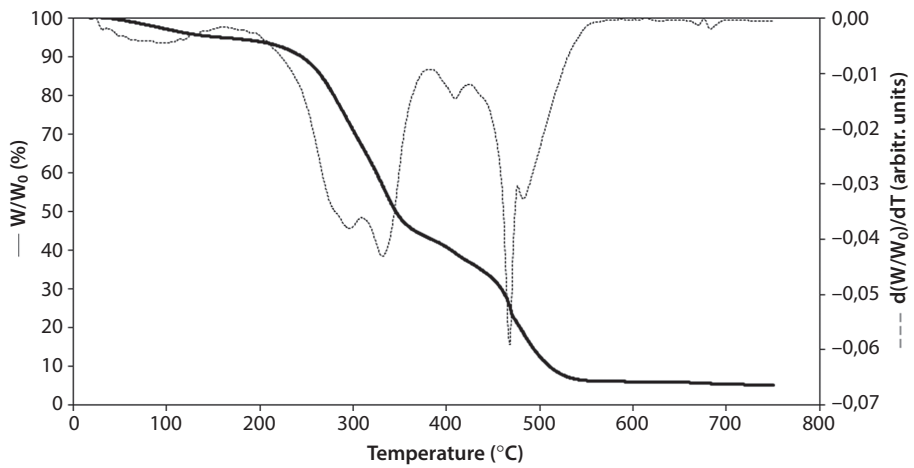

Figure 2 TGA degradation curve under air and at 5 °C/min of the press cake.

225 °C and 350 °C, respectively. Associated with the onset of its thermal degradation, the smoke point of coriander oil is thus similar to many other vegetable oils (e.g., 232 °C in the case of refined sunflower oil).

Consequently, the first thermal degradation stage (225–350 °C) could be associated with the simultaneous degradation of water-soluble components, proteins, hemicelluloses and lignins, followed by the degradation of lipids and cellulose. Because TGA analysis of the press cake was conducted under air atmosphere, the second stage, situated around 475 °C, would then correspond to the oxidation of the degradation products from the previous stage. At the end of the measurement, the undegraded sample represented 5.2% of the test sample mass. The undegraded compounds logically corresponded to minerals, whose content in the press cake was 6.2% of the dry matter (Table 1). To conclude, no thermal degradation occurred inside the press cake before 225 °C. Thus, it is reasonable to suppose that the three mold temperatures chosen for thermo-pressing, i.e., 160, 180 and 200 °C, were relevant. Indeed, no organic compounds inside the press cake, especially proteins, were likely to thermally degrade during molding, even at 200 °C, which contributed a priori to the production of fiberboards with preserved mechanical properties.

DSC analysis of the press cake was conducted on a deoiled material that was dried in a ventilated oven (60 °C, 12 h) before being tested. Thus, it was conducted from a material with approximately the same moisture content as that of the press cake used for thermo-pressing, i.e., 2.2% instead of 2.1%. The DSC curve revealed a glass transition phenomenon, situated around 140–145 °C (Figure 3). And, this glass transition was still observed using the DMTA technique, the loss factor (tan δ) curve revealing a significant peak in the same temperature range simultaneously with a rapid decrease in the storage modulus (E′) (Figure 4). This glass transition was attributed to that of proteins. Indeed, even if other materials inside the press cake such as hemicelluloses could reveal the same phenomenon [44–47], this glass transition temperature

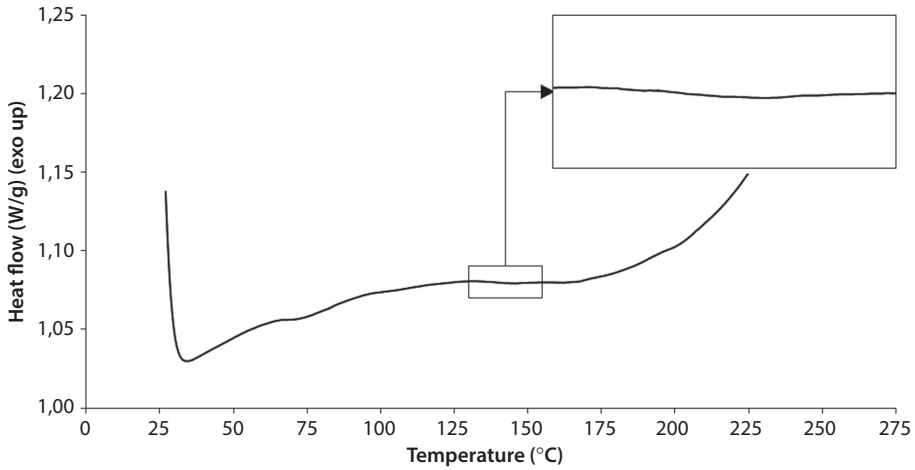

Figure 3 DSC curve in a pressure-resistant capsule of the deoiled press cake after drying in a ventilated oven (60 °C, 12 h).

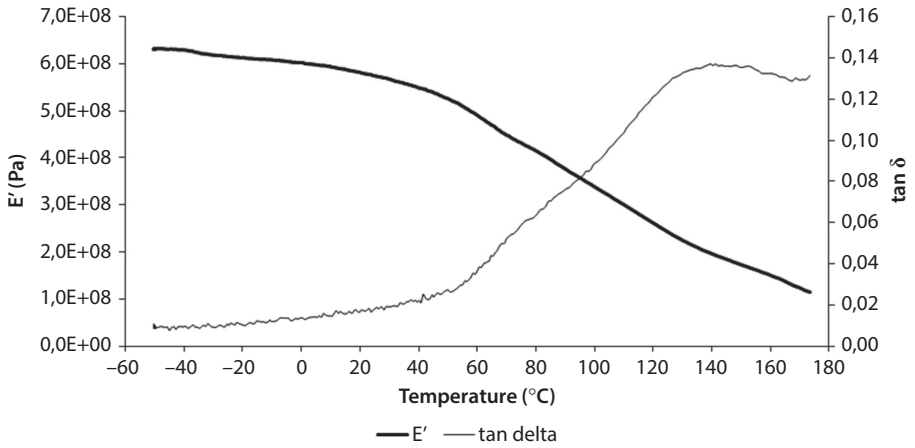

Figure 4 DMTA curve in a small metallic container of the deoiled press cake after drying in a ventilated oven (60 °C, 12 h).

perfectly agreed with that of sunflower proteins showing the same amount of water [36, 48].

3.2 Influence of Thermo-Pressing Conditions on the Characteristics of Fiberboards

The press cake was dried in a ventilated oven (60 °C, 12 h) before thermo-pressing, and its moisture content was 2.1% at molding. For boards 1 to 11, the press cake quantity was 200 g (i.e., 889 mg/cm^2) and the applied thermo-pressing conditions were 160–200 °C mold temperature (three levels), 24.5–49.0 MPa applied pressure (three levels), and 60–300 s molding time (three levels) (Table 2). These values were similar to those used in a previous study conducted from a deoiled sunflower cake originating from the whole plant [36]. The eleven fiberboards that were manufactured using these thermo-pressing conditions were all cohesive, with proteins and fibers acting respectively as a natural binder and reinforcing fillers.

The influence of the mold temperature (i.e., 160, 180, and 200 °C) on the mechanical properties of the fiberboards was studied through boards 1, 2 and 7. These were manufactured from medium values for both applied pressure and molding time, i.e., 36.8 MPa and 180 s, respectively. The mold temperature always being higher than the protein glass transition temperature (140–145 °C), this change systematically occurred during molding. Thus, the proteins were always in a rubbery state. However, the protein-based resin became consistently less viscous with increasing mold temperature, and this contributed undoubtedly to a progressive improvement in fiber wetting. This is probably the reason why the fiberboards became increasingly mechanically resistant as the mold temperature increased, the 200 °C mold temperature logically leading to the most resistant board (Table 2). As an example, as the mold temperature increased from 160 to 200 °C, the flexural strength at break increased from 4.5 to 11.3 MPa. Similarly, the elastic modulus and the Shore D surface hardness increased from 0.9 to 2.6 GPa and from 61 to 71°, respectively. At the same time, because the board density also tended to increase (until 1.30 for board 7), this resulted in a reduction of its internal porosity, thus contributing to a strongly reduced water sensitivity (51% thickness swelling instead of 157% at 160 °C mold temperature, and 33% water absorption instead of 167%).

On the basis of these initial results, the influence of both pressure and time was studied through boards 3 to 11, using for all of them the mold temperature considered as the optimal one, i.e., 200 °C. Comparing the two minimal applied pressures, i.e., 24.5 and 36.8 MPa, the pressure increase resulted in the fiberboard densification and so in the increase in both flexural properties and Shore D surface hardness, and this occurred for the three tested molding times (Table 2 and Figure 5). Moreover, such a tendency was already observed in a previous study [36]. However, for these two minimal pressures, even if the increase in molding time from 60 to 180 s led to an increase in the board density and so in its mechanical properties (bending properties plus surface hardness), a reduction in these characteristics occurred for an additional increase in molding time. Thus, the 300 s molding time was considered as excessive, leading to a partial degradation of biopolymers inside the boards, in particular proteins whose binding ability probably diminished. The best mechanical properties were thus obtained under 36.8 MPa applied pressure and 180 s molding time, corresponding to board number 7.

For the maximal applied pressure, i.e., 49.0 MPa, the board density and bending properties slightly increased as the molding time increased from 60 to 300 s (Table 2 and Figure 5), and this tendency was already observed from a deoiled sunflower cake [36]. At the same time, the Shore D surface hardness was high, varying from 66 to 70°. However, all fiberboards molded at 49.0 MPa pressure remained less mechanically resistant than board 7, even at 300 s molding time. This could be the consequence of excessive applied shear (49.0 MPa) in combination with the maximal temperature (200 °C), which in turn could lead to the beginning of degradation phenomena inside boards 5, 8 and 11.

The water sensitivity of boards 3 to 11 was studied through measurements of their thickness swelling and water absorption after 24 h soaking in distilled water. Because the increase in fiberboard density resulted in a reduction of the internal porosity of the material, a correlation could be made between the density and the water sensitivity of the fiberboards (Figure 6). Indeed, the more the density, the less the thickness swelling and the less the water absorption. Thus, the densest fiberboard (1.30 density), i.e., board 7, which also corresponded to the best bending properties (11.3 MPa flexural strength at break and 2.6 GPa elastic modulus) and the highest value for Shore D surface hardness (71°), was the most water-resistant board as well, thickness swelling and water absorption being only 51% and 33%, respectively. Molded under 200 °C temperature, 36.8 MPa pressure and 180 s time, board number 7 was considered as the optimal board of the entire study. It complied with the French standard NF EN 312 (standard dedicated to the specifications for particleboards), type P2 (i.e., boards for interior layouts, including furniture, used in dry conditions) for flexural properties (recommendations of 11 MPa and

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Table 2 Thermo-pressing conditions, mechanical properties, thickness swelling and water absorption of the thirteen manufactured fiberboards.

Fiberboard	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**
Thermo-pressing conditions													
Temperature (°C)	160	180	200	200	200	200	200	200	200	200	200	200	200
Pressure (MPa)	36.8	36.8	24.5	36.8	49.0	24.5	36.8	49.0	24.5	36.8	49.0	36.8	36.8
Time (s)	180	180	60	60	60	180	180	180	300	300	300	180	180
Press cake (g)	200.0	200.0	200.0	200.0	200.0	200.0	200.0	200.0	200.0	200.0	200.0	400.0	200.0
Coriander straw (g)	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	35.3
Flexural properties													
H_{FB} (%) [a]	6.14 ± 0.05	6.41 ± 0.11	7.17 ± 0.02	8.50 ± 0.10	8.46 ± 0.13	6.93 ± 0.14	7.16 ± 0.08	7.60 ± 0.05	8.15 ± 0.18	6.91 ± 0.11	3.17 ± 0.23	7.86 ± 0.10	7.90 ± 0.07
t (mm)	6.36 ± 0.05	6.20 ± 0.06	6.52 ± 0.04	6.24 ± 0.10	6.11 ± 0.14	6.15 ± 0.08	5.93 ± 0.06	6.00 ± 0.16	6.29 ± 0.07	6.11 ± 0.04	5.91 ± 0.08	11.96 ± 0.17	8.00 ± 0.10
d	1.27 ± 0.01	1.28 ± 0.00	1.24 ± 0.01	1.27 ± 0.01	1.28 ± 0.02	1.29 ± 0.01	1.30 ± 0.01	1.29 ± 0.02	1.26 ± 0.02	1.28 ± 0.01	1.29 ± 0.01	1.24 ± 0.01	1.22 ± 0.02
F (N)	36.6 ± 0.5	52.0 ± 1.9	30.3 ± 2.0	49.7 ± 2.8	66.2 ± 2.5	70.6 ± 3.1	79.5 ± 2.3	68.4 ± 4.2	68.1 ± 2.7	69.4 ± 2.9	74.7 ± 2.0	101.1 ± 6.6	70.3 ± 3.4
s_f (MPa)	4.5 ± 0.1	6.8 ± 0.2	3.6 ± 0.2	6.4 ± 0.4	8.9 ± 0.3	9.3 ± 0.4	11.3 ± 0.3	9.5 ± 0.6	8.6 ± 0.3	9.3 ± 0.4	10.7 ± 0.3	3.5 ± 0.2	5.5 ± 0.3
E_f (MPa)	929 ± 7	1457 ± 56	785 ± 43	1418 ± 49	1895 ± 32	2141 ± 121	2625 ± 156	2074 ± 170	1797 ± 55	1968 ± 99	2305 ± 91	520 ± 40	1070 ± 58
Charpy impact strength													
W (mJ)	70 ± 7	67 ± 8	73 ± 7	82 ± 7	86 ± 5	80 ± 5	75 ± 6	82 ± 6	73 ± 4	77 ± 5	66 ± 4	198 ± 69	500 ± 68
K (kJ/m^2)	1.09 ± 0.11	1.08 ± 0.12	1.12 ± 0.10	1.32 ± 0.12	1.40 ± 0.08	1.30 ± 0.09	1.26 ± 0.10	1.36 ± 0.10	1.15 ± 0.06	1.25 ± 0.09	1.12 ± 0.07	1.66 ± 0.58	6.25 ± 0.85
Surface hardness													
Shore D (°)	60.8 ± 4.7	63.6 ± 3.6	59.5 ± 5.9	65.0 ± 3.7	66.2 ± 4.2	68.8 ± 3.8	70.8 ± 3.1	69.7 ± 3.5	68.6 ± 3.1	68.0 ± 3.1	68.0 ± 2.8	66.3 ± 3.3	58.9 ± 4.4
Thickness swelling and water absorption													
TS (%)	157 ± 18	160 ± 6	148 ± 13	137 ± 13	124 ± 11	69 ± 10	51 ± 4	42 ± 6	78 ± 9	67 ± 4	55 ± 4	141 ± 11	190 ± 5
WA (%)	167 ± 7	112 ± 8	173 ± 13	71 ± 3	67 ± 7	39 ± 3	33 ± 1	42 ± 5	65 ± 7	52 ± 4	40 ± 1	131 ± 12	191 ± 4

Results in the table correspond to the mean values ± standard deviations.

[a] *H_{FB} is the moisture content of the fiberboard. Fiberboards were equilibrated in a climatic chamber (60% RH, 25 °C) for three weeks before moisture measurements.*

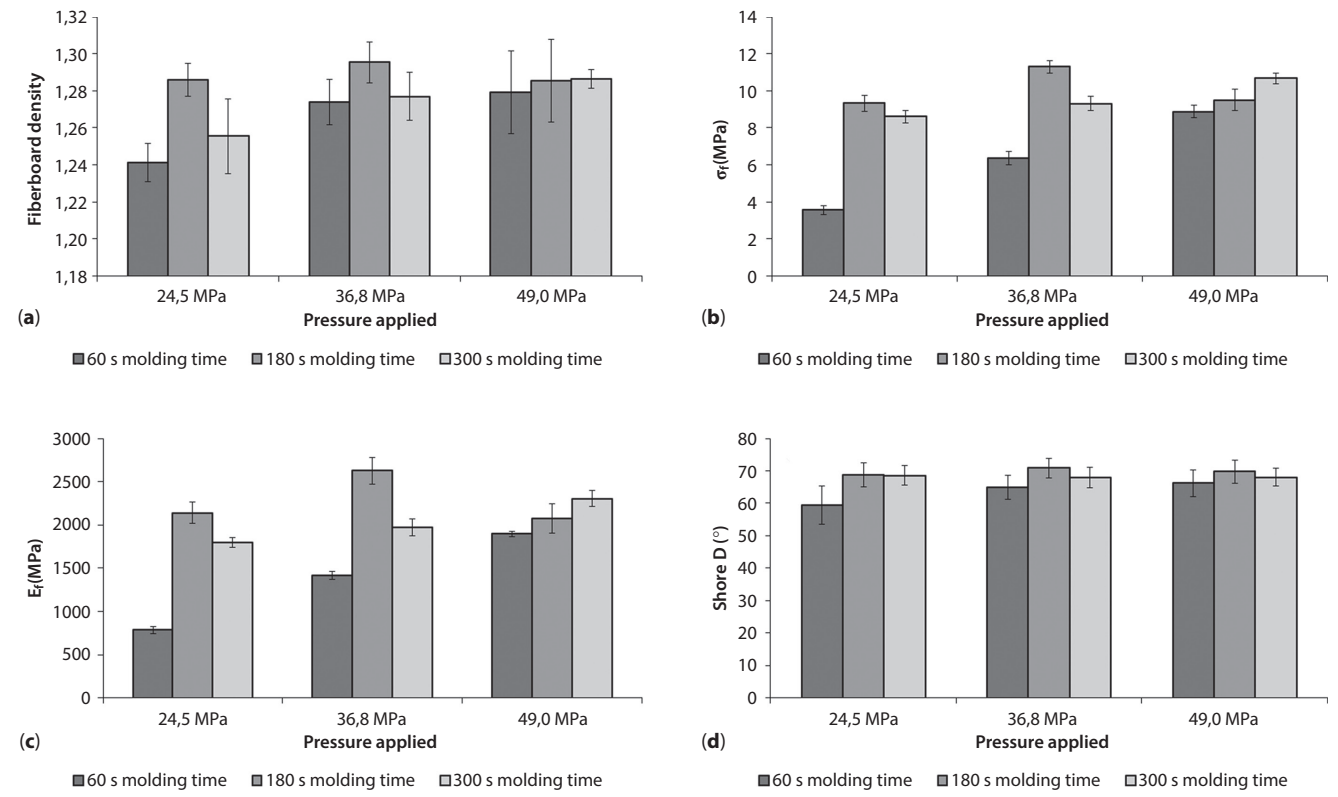

Figure 5 Density (**a**), flexural strength at break (**b**), elastic modulus (**c**), and Shore D surface hardness (**d**) of the fiberboards as functions of the applied pressure and the molding time (200 °C mold temperature).

1.8 GPa for flexural strength at break and elastic modulus, respectively, for boards with a 6 mm thickness).

Concerning the Charpy impact strength, the latter was always low for boards 1 to 11 (Table 2) and the maximal resilience was only 1.4 kJ/m^2 (case of board 5). This was due to the global brittle behavior of fiberboards, which therefore showed a high stiffness. Two solutions were considered in order to improve the impact resistance of the optimal board, i.e., board 7. On the one hand, increasing the board thickness by doubling the mass used in its manufacture (case of board 12) allowed a slight increase (+ 32%) in its resilience. On the other hand, manually adding 15% of coriander straw to the press cake before thermopressing (case of board 13) resulted in a very important increase in resilience (+ 396%). The straw constituted a byproduct of coriander culture. It revealed very low apparent and tapped densities compared to the press cake, and high fiber content: 51% cellulose, 15% hemicelluloses and 14% lignins (Table 1). Conversely, contents for minerals, lipids and proteins were much lower, representing only 5%, 2% and 3% of the dry matter, respectively. In conclusion, the addition of coriander straw to the press cake was a promising solution to significantly improve the impact resistance of the optimal board. Moreover, this fiber is not commercially available for the moment, as the coriander harvest only concerns the fruits. Therefore, this could be seen as an additional source of income for farmers, possibly justifying, in the future, their harvest in the field. However, it should be noted that the improvement in Charpy impact strength by coriander straw addition also resulted in a reduction of all other boards' characteristics, i.e., the flexural properties, the Shore D surface hardness, the thickness swelling and the water absorption (Table 2).

3.3 Oil Expression during Molding

For all fiberboards, part of the residual oil in the press cake was expressed through the sidewall vents of the mold during molding, which was due to the applied pressure. Concurrent oil expression and molding had been previously observed starting from a cake produced after the biorefinery of sunflower whole plant in a twin-screw extruder [31–33] or from a jatropha press cake [35]. Thus, a decrease in the residual oil content inside the fiberboards was systematically observed: up to 7.0% of the dry matter for board 10 (Table 3) instead of 17.2% in the press cake (Table 1). This further contributed to the increase in the total oil recovery (R_{LT}), reaching 81% for board 10 (Table 3).

Comparing boards 1, 2 and 7, which were all molded using medium values for pressure (36.8 MPa) and time

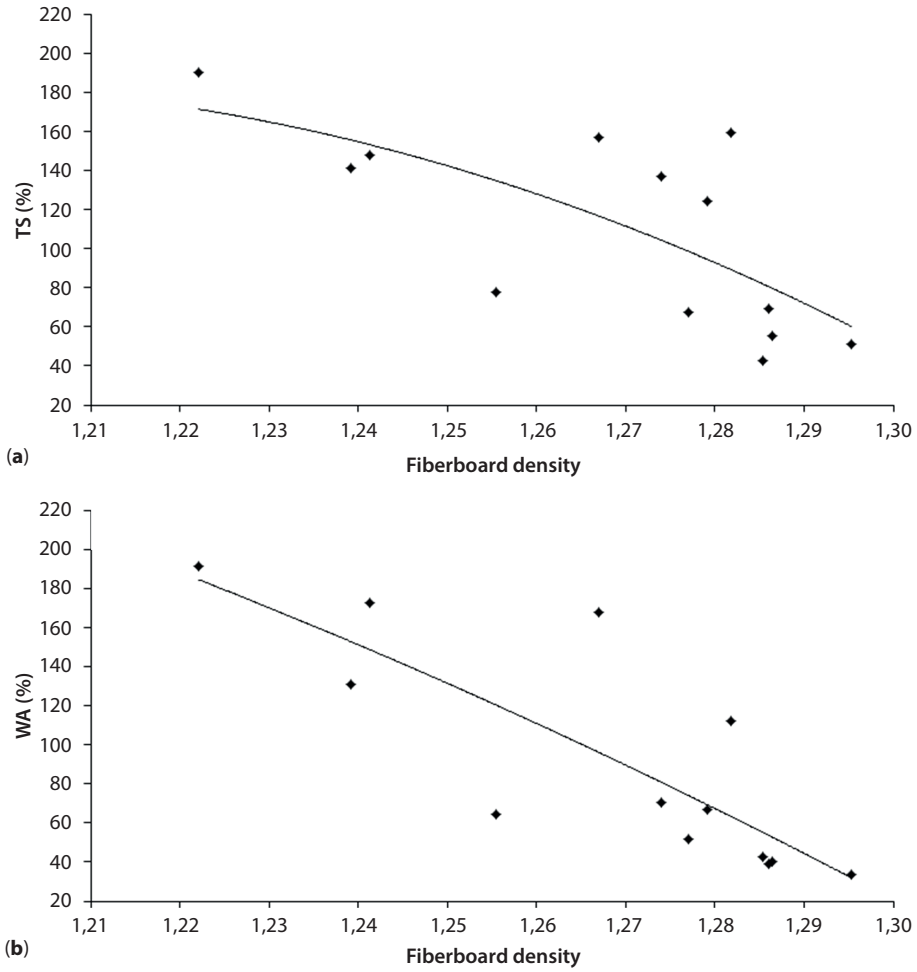

Figure 6 Thickness swelling (**a**), and water absorption (**b**) of the fiberboards as functions of their density.

(180 s), the increase in temperature from 160 to 180 °C had no influence upon the quantity of oil expressed during molding. Indeed, residual oil contents inside boards 1 and 2 were not significantly different, leading to similar oil recoveries (R_{L2}) (52–53%) and to a 75% total oil recovery (R_{LT}) in both cases (Table 3). On the contrary, further increase in mold temperature (until 200 °C) largely favored oil expression during molding. This was due to its viscosity decrease, facilitating the migration of oil through the mold. This resulted in a better impoverishment in lipids of board 7 and a significant increase in both oil recoveries: 61% oil recovery during molding (R_{L2}) and 80% total oil recovery (R_{LT}). The same tendency was already observed for sunflower oil [31–33] and jatropha oil [35]. In conclusion, the 200 °C mold temperature, which already contributed to the production of more mechanically resistant and less water-sensitive fiberboards, also favored oil expression on molding, thus leading to an increase in total oil recovery (R_{LT}).

Boards 3 to 11 were all molded at the 200 °C optimal mold temperature, making it possible to analyze the influence of both applied pressure (24.5–49.0 MPa) and molding time (60–300 s) on oil expression during molding (Table 3 and Figure 7). For boards 3 to 5, which involved the minimal molding time, i.e., 60 s, the pressure increase contributed to a progressive improvement in oil expression, as illustrated by the decrease in residual oil inside the fiberboards. This further led to the increase in the oil recovery during molding (R_{L2}) and the total oil recovery (R_{LT}): from 44 to 55%, and from 71 to 76%, respectively. For the two maximal molding times, i.e., 180 and 300 s, the quantity of expressed oil was much more important, leading to a better impoverishment of the fiberboards in lipids. In addition, as already observed for sunflower oil [33], the boards' residual oil content tended to decrease with the increase in both pressure and time.

The two lowest values for the residual oil content of the fiberboards were obtained with a maximal molding time, i.e., 300 s, using 36.8 MPa and 49.0 MPa of applied pressure. They were 7.0% and 7.2% of the dry matter, respectively, corresponding to boards 10 and 11. The oil recovery during molding (R_{L2}) was then

Table 3 Quantification of the oil expressed during molding of the fiberboards.

Fiberboard	1	2	3	4	5	6	7	8	9	10	11	12	13
m_{FB} (g)	188.4	189.8	191.7	188.8	188.6	185.3	182.3	183.5	186.3	185.9	182.7	368.1	234.0
L_{FB} (% of the dry matter)	8.98 ± 0.05	9.05 ± 0.01	10.64 ± 0.00	10.11 ± 0.00	8.81 ± 0.01	8.44 ± 0.01	7.74 ± 0.05	8.24 ± 0.01	8.64 ± 0.03	7.02 ± 0.07	7.17 ± 0.09	9.01 ± 0.05	10.14 ± 0.03
R_{L2} (%)	53.0 ± 0.3	52.4 ± 0.1	43.9 ± 0.0	48.3 ± 0.0	55.0 ± 0.1	56.9 ± 0.1	61.2 ± 0.2	58.7 ± 0.0	56.2 ± 0.1	64.1 ± 0.4	62.4 ± 0.4	54.8 ± 0.3	36.9 ± 0.2
R'_{L2} (%)	27.8 ± 0.5	27.4 ± 0.1	23.0 ± 0.0	25.3 ± 0.0	28.8 ± 0.1	29.8 ± 0.1	32.0 ± 0.4	30.7 ± 0.1	29.4 ± 0.3	33.5 ± 0.6	32.7 ± 0.7	28.7 ± 0.5	19.3 ± 0.5
R_{LT} (%)	75.4 ± 0.8	75.1 ± 0.4	70.6 ± 0.3	72.9 ± 0.3	76.4 ± 0.4	77.4 ± 0.4	79.7 ± 0.7	78.4 ± 0.4	77.1 ± 0.6	81.2 ± 0.9	80.3 ± 1.0	76.3 ± 0.8	66.9 ± 0.8

Results in the table correspond to the mean values ± standard deviations.
The oil recovery in the single-screw press based on the residual oil content in the press cake (R_{L1}) was 47.6 ± 0.3%.

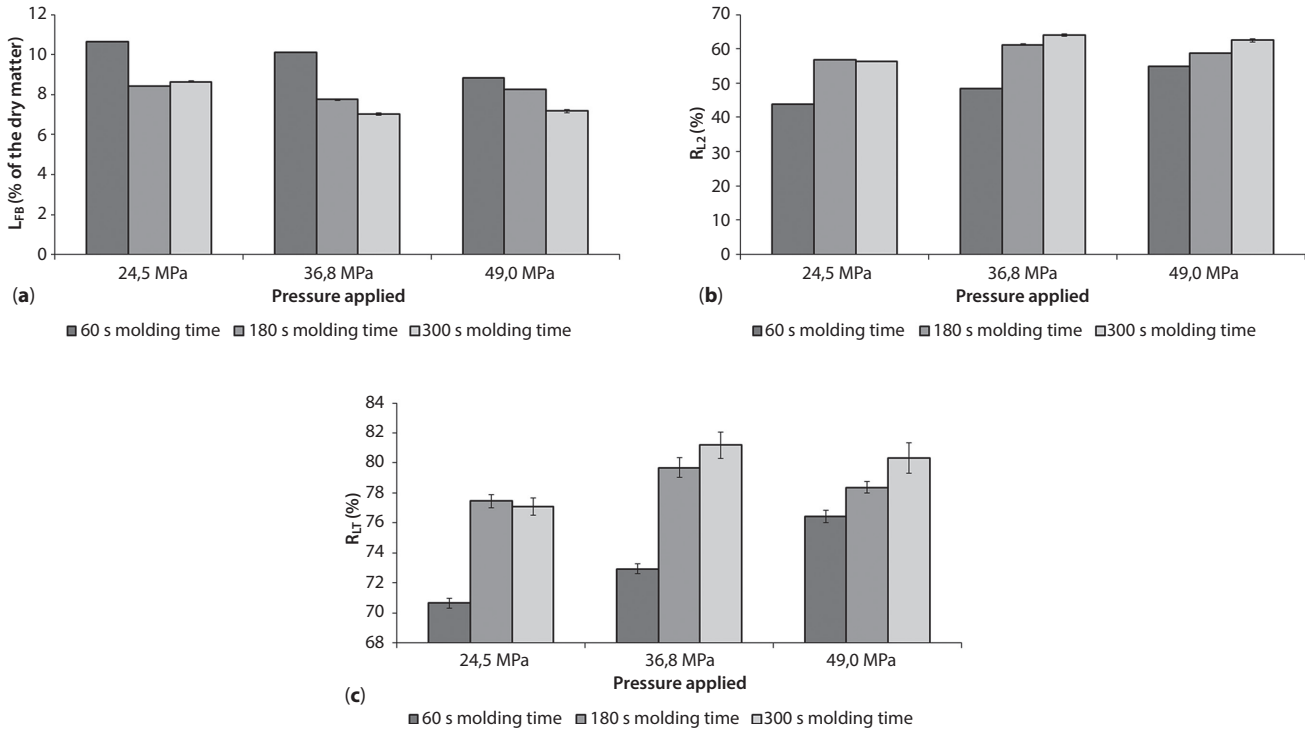

Figure 7 Oil content of the fiberboard (**a**), oil recovery during molding relative to the residual oil contained in the press cake (**b**) and total oil recovery relative to the total amount of oil that the coriander fruits contain (**c**) as functions of the applied pressure and the molding time (200 °C mold temperature).

62–64%, leading to an 80–81% total oil recovery (R_{LT}). The residual oil content of the fiberboard molded under optimal thermo-pressing conditions, i.e., board number 7, was also less than 8% of the dry matter, and this was associated with a nearly maximal total oil recovery (R_{LT}) of 80%.

As previously observed in the case of sunflower oil [33], the increase in board thickness, which was the case for boards 12 and 13, slightly unfavored oil expression. The oil recovery during molding (R_{L2}) decreased from 61% for board 7 (200 g press cake quantity) to 55% for board 12 (400 g press cake quantity). Logically, the total oil recovery (R_{LT}) slightly decreased as well (from 80% to 76%). In the case of board 13, however, this decrease in oil recovery is mostly due to the addition of coriander straw to the press cake before molding. Indeed, the vegetable oil was partly absorbed by the straw, the latter acting as a retention agent for residual oil inside the fiberboard. Both oil recoveries (R_{L2}) and (R_{LT}) were then only 37% and 67%, respectively.

Because the oil expression during molding was only partial, fiberboards still contained residual oil. As an example, for the 200 °C mold temperature that led to the most mechanically resistant boards (Table 2) and to the best oil recoveries during molding (Table 3), fiberboards still contained between 7.0 and 10.6% residual oil. It is sure that all fiberboards revealed a global hydrophilic character. However, it is also reasonable to assume that the residual oil will contribute to making them slightly less water-sensitive and more durable than deoiled thermo-pressed agromaterials.

The oil expressed during molding could be collected. Its filtration would first eliminate the small solid particles driven through the vents of the mold during thermo-pressing. Then, this could further be complemented by a refining step. The obtained refined oil could still be used for its high content in petroselinic acid, to synthesize a large number of interesting platform molecules for the food, cosmetic and pharmaceutical industries [22]. However, because it was of lower quality compared to the pressed oil produced continuously in the single-screw extruder, its use for human feeding seemed more delicate. On the contrary, it could be used for two new applications, i.e., as a biolubricant or to be transformed into biodiesel after transesterification of triglycerides with methanol to produce fatty acid methyl esters (FAME) [35, 49].

4 CONCLUSION

New renewable and biodegradable fiberboards were manufactured through thermo-pressing of a press

cake produced during the extraction of vegetable oil from coriander fruits using a single-screw extruder. Proteins acted as a binder inside the boards, leading to the production of cohesive panels inside which the fibers contributed to their mechanical reinforcement. The three thermo-pressing conditions that were investigated (i.e., temperature, pressure and time) affected the boards' mechanical properties. Even if the glass transition of proteins systematically occurred during molding, a 200 °C mold temperature was required in order to significantly improve fiber wetting inside the boards. The most mechanically resistant fiberboard (11.3 MPa flexural strength at break, 2.6 GPa elastic modulus and 70° Shore D surface hardness) was molded under 200 °C mold temperature, 36.8 MPa applied pressure and 180 s molding time. Furthermore, the water sensitivity of this optimal board was quite low (51% thickness swelling and 33% water absorption). In regard to these characteristics, the latter could find applications in various industrial fields such as the handling and storage industry (interlayer sheets for pallets), the manufacture of containers or furniture, or the building trade (floor underlayers, interior partitions or ceiling tiles). Moreover, thermo-pressing consisted not only of molding new, cohesive fiberboards, but also contributed to a significant increase in the vegetable oil recovery: from 48% after extrusion to a maximum of 81% after thermo-pressing (80% in the case of the optimal board).

REMARK FROM AUTHORS

The first two authors have both participated equally to this work. Therefore, they are both considered as principal authors.

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