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Article

Study of the Utilisation of Almond Residues for Low-Cost Panels

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Abstract: A large amount of research is being carried out to increase the use of renewable and eco-friendly resources like plant fibres for manufacturing new products in order to reduce energy consumption and improve their environmental impact. The almond is a deciduous tree that is native to Mediterranean areas, although nowadays the United States is the world's main almond producer. The almond fruit has three distinct parts: the inner core or flesh, the hard middle part or shell, and the outer covering of the shell, called the hull (exocarp and mesocarp). This work analyses the use of almond residues for producing eco-friendly particleboards. The ground hull of the almond has been used as a raw material, obtaining 4 different particle sizes. Eight type of board has been manufactured without using any kind of adhesive. The particle size influences some physical and mechanical properties. With particle sizes <0.25 mm it is possible to achieve greater strength in terms of modulus of rupture (MOR): 14.01 N/mm², modulus of elasticity (MOE): 2295.32 N/mm² and internal bonding strength (IB): 0.57 N/mm². This study shows that it is technically possible to manufacture boards with this material without using adhesives.

Keywords: particleboards; exocarp; *Prunus dulcis* (Mill) D.A. Webb; binderless

1. Introduction

The almond, whose scientific name is *Prunus dulcis* (Mill) D.A. Webb, belongs to the Rosaceae family and is also related to stone fruits such as peaches, plums and cherries. The almond tree is a species native to Mediterranean areas. It was spread in ancient times along the shores of the Mediterranean to North Africa and Southern Europe and, more recently, it was transported to other parts of the world, especially California (USA). The United States is the world's main producer of almonds, followed by Spain. The almond fruit has three distinct parts: the inner core or flesh, the hard middle part or shell, and the green outer covering of the shell, called the hull. The almond hull (exocarp and mesocarp) is obtained when the portion of the almond fruit that surrounds the hard shell dries and it is normally left in the field, constituting a waste product that can lead to environmental problems such as the spread of fires or pests.

Almond production generates millions of tons of waste, including the shells, hulls, pruning waste, leaves, skin and inedible part of the kernel. These residues are good raw materials for producing bioenergy and other valuable composites, but more research and development needs to be carried out in this area [1].

Numerous uses have been proposed for the green almond hull: extraction of polysaccharides [2,3], bioactive substances [4–6], antioxidants [7–11], bioenergy [12–14], activated carbon [15] and absorbent filters [16–18]. Several studies have highlighted its potential use as animal feed [19–24].

Agricultural waste also includes lignocellulosic materials that can replace natural wood, but it is necessary to demonstrate that their fibres are suitable as a raw material with which to manufacture boards for furniture, packaging and building purposes. The use of plant materials to replace wood as a raw material will play an important role in the future. The manufacture of particleboards (agglomerated panels) is continually growing due to the scarcity of wood. These are basically composite materials that are traditionally made from wood chips and a binder.

Other proposals include using the almond shell to manufacture different composites [25–29] and also the hull [30].

Wood adhesives have been developed by the petrochemical industry and offer excellent performance and good working properties, in addition to being affordable. However, it is believed that the adhesives currently used for wood composites will inevitably be restricted in the future due to a decline in the reserves of fossil resources and to harmful formaldehyde emissions.

Research has been carried out to develop particleboards using various plant species as a raw material and the latest research has focused on producing agglomerated particleboards without using a binder product: date palm [31–33], oil palm [34–39], Canary Islands palm [40], rice straw [41,42], kenaf [43,44], sugarcane [45], coconut husk [46], bamboo [47], sorghum [48], cotton stalk [49] and almond residues [30].

In this work, we propose using the almond hull (exocarp and mesocarp) as a raw material for manufacturing binderless particleboards in an attempt to develop an added value application for this waste. The objective is to obtain an entirely eco-friendly product by using almond residues to manufacture binderless agglomerated particleboards and to assess the self-binding mechanism.

2. Materials and Methods

2.1. Materials

The materials used were particles of green almond hulls from the Higher Technical College of Orihuela at Universidad Miguel Hernández in Elche.

The almond hulls were left to dry outdoors for 6 months. They were then shredded in a blade mill. The particles were collected in a vibrating sieve and 4 particle sizes were selected: particles that passed through the 4 mm sieve but were retained in the 2 mm one (2 to 4 mm), particles that passed through the 2 mm sieve but were retained in the 1 mm one (1 to 2 mm), particles that passed through the 1 mm sieve but were retained in the 0.25 mm one (0.25 to 1 mm) and particles that passed through the 0.25 mm sieve (<0.25 mm). The approximate humidity of the particles was 11%.

2.2. Methods

Binderless particleboards were manufactured, their properties were evaluated experimentally and the results were analysed using statistical methods.

The manufacturing process consisted of forming the mat of the board using almond residues with four different particle sizes. The mat was formed in a mould of dimensions 600 mm × 400 mm and was subjected to pressure and heat in a hot plate press at a pressure of 2.5 MPa and a temperature of 120 °C for 30 min. Once cooled, half of the boards were placed in the press again for a further 30 min at the same temperature. The panels were then left to cool in a vertical position. The approximate dimensions of the particleboards were 600 × 400 × 5 mm. The production characteristics of the 8 types of panels are shown in Table 1 and several of the panels are shown in Figure 1. Six panels of each type were manufactured.

Table 1. Characteristics of the type of panels manufactured.

Type of Board	No. of Boards	Particle Size (mm)	Time (min)	Pressure (Mpa)	Temperature (°C)
A1	6	<0.25	30	2.5	120
A2	6	<0.25	30 + 30 = 60	2.5	120
B1	6	0.25 to 1	30	2.5	120
B2	6	0.25 to 1	30 + 30 = 60	2.5	120

C1	6	1 to 2	30	2.5	120
C2	6	1 to 2	30 + 30 = 60	2.5	120
D1	6	2 to 4	30	2.5	120
D2	6	2 to 4	30 + 30 = 60	2.5	120

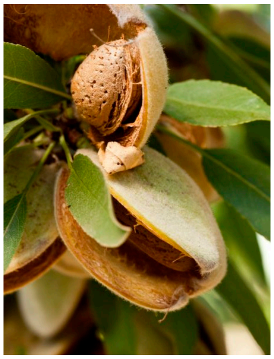

Figure 1. Almond hulls (exocarp and mesocarp) and the binderless particleboards obtained.

The properties were determined according to the European standards established for wood particleboards [50]. Before performing the tests, samples of the dimensions specified in the European standards were cut from each board. Six samples were cut for the bending test, 6 for the density test, 3 for the immersion in water test and 3 for the internal bonding strength test. Before performing the tests, the samples were placed in a normal climate chamber at a temperature of 20 °C and relative humidity of 65%.

The properties of the boards were measured according to the European standards: density [51], thickness swelling (TS) and water absorption (WA) after 2 and 24 h immersed in water [52], internal bonding strength (IB) [53], modulus of rupture (MOR) and modulus of elasticity (MOE) [54]. The boards were evaluated according to the European standard [55]. The mechanical tests were performed with the IMAL testing machine (Italy), which complies with the required velocity for each test, as specified in the European standards applicable to the tests.

A chemical analysis of the material was performed to study the concentration of polymeric sugars in the sample, which was quantified using high-performance liquid chromatography (HPLC) according to NREL/TP-510-42618 [56]. The material was ground to obtain a particle size of <0.25 mm and 0.5 grams of material were used, diluted in 100 mL of distilled water at room temperature. The samples were left in distilled water and stirred for 48 h. The process used was HPLC and the chromatography system comprised an Agilent column with HP processor.

Error bars and standard deviation were obtained for the mean values of the tests and analysis of variance (ANOVA) was performed. The statistical analysis was performed using SPSS v. 25.0 software from IBM.

3. Results and Discussion

3.1. Density

The mean density values of each type of board are shown in Figure 2. No significant differences can be observed between the different types of boards. They are high-density boards: 1343 ± 74 kg/m^3. The statistical analysis shows that the density does not depend on the particle size or the pressing time.

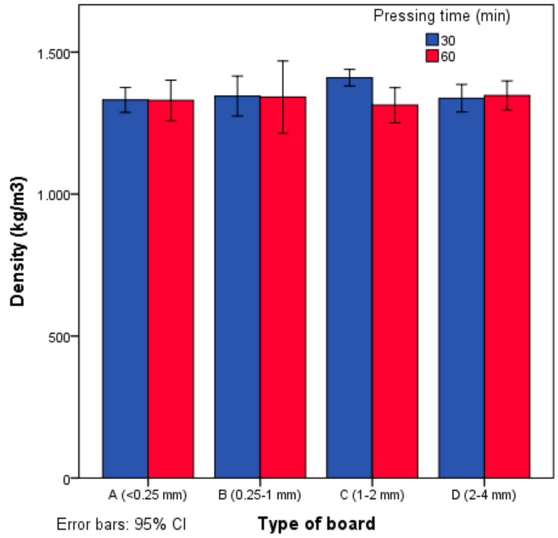

Figure 2. Mean density according to type of board.

3.2. Thickness Swelling (TS) and Water Absorption (WA) after 2 h and 24 h Water Immersion

The test was performed on three samples of each board. The mean thickness swelling (TS) results after 2 and 24 h are shown in Table 2. In the test after 2 h, the boards with the lowest TS were type C1 with 8.77%, and the boards with the highest TS were type D2 with 17.51%. After 24 h, the type A2 boards had a TS of 18.24% (minimum value), and for the type D2 boards it was 33.43% (maximum value). Statistically (Table 3), it can be seen that it depends on the particle size and the pressing time.

Table 2. Thickness swelling (TS) and water absorption (WA) after 2 and 24 h according to type of board and pressing time.

Type of Board	Pressing Time	TS 2 h (%)	TS 24 h (%)	WA 2 h (%)	WA 24 h
A1	30	11.11 (5.26)	22.11 (7.74)	11.66 (6.21)	26.28 (8.18)
A2	30 + 30	9.10 (1.05)	18.24 (3.94)	12.11 (5.68)	21.17 (7.37)
B1	30	13.54 (3.12)	26.06 (4.02)	13.09 (7.36)	30.88 (8.21)
B2	30 + 30	16.35 (6.06)	24.04 (1.84)	20.42 (7.79)	34.51 (9.68)
C1	30	8.77 (4.27)	18.62 (7.29)	6.83 (3.73)	16.65 (6.22)
C2	30 + 30	10.24 (5.76)	20.24 (9.48)	20.16 (10.55)	28.34 (7.93)
D1	30	9.62 (6.25)	22.58 (9.12)	8.24 (5.18)	22.10 (10.79)
D2	30 + 30	17.51 (6.37)	33.43 (7.06)	18.18 (8.88)	39.73 (10.04)

(…) standard deviation.

Table 3. Analysis of variance (ANOVA) of the results of the tests.

Factor	Properties	Sum of Squares	d.f.	Half Quadratic	F	Sig.
Particle size	TS 2 h (%)	336.553	3	112.184	3.692	0.019
	TS 24 h (%)	869.605	3	289.868	5.340	0.003
	WA 2 h (%)	238.837	3	79.612	1.031	0.388
	WA 24 h (%)	1403.742	3	467.914	3.616	0.020
	MOR (N/mm^2)	220.893	3	73.661	19.123	0.000
	MOE (N/mm^2)	6,164,529.410	3	2,054,843.137	12.550	0.000
	IB (N/mm^2)	1.079	3	0.360	2.926	0.027
Pressing time	TS 2 h (%)	109.232	1	109.232	3.210	0.080
	TS 24 h (%)	76.860	1	76.860	1.111	0.297
	WA 2 h (%)	708.820	1	708.820	11.134	0.002
	WA 24 h (%)	699.873	1	699.873	5.032	0.030

MOR (N/mm²)	31.065	1	31.065	3.732	0.045
MOE (N/mm²)	877,907.085	1	877,907.085	4.021	0.039
IB (N/mm²)	0.273	1	0.273	2.703	0.107

d.f.: degrees of freedom. F: Fisher–Snedecor distribution. Sig.: significance. MOR: modulus of rupture. MOE: modulus of elasticity. IB: internal bonding strength

Table 2 also shows the mean WA values and the error bars after 2 and 24 h. The water absorption percentage values after 2 h range between 6.83% for type C1 and 20.42% for type B2; there were large differences between some of the types of boards. After 24 h the water absorption increases, giving a value of 16.65% for type C1 and 39.73% for type D2. From the data in Table 3, it can be concluded that the WA depends on the particle size and the pressing time, except for WA after 2 h.

Some kind of water-repellent product could be used to minimise swelling and water absorption, such as those used in the wood industry.

3.3. Modulus of Rupture (MOR) and Modulus of Elasticity (MOE)

Six samples of each board were cut to determine the bending properties, three longitudinally and three transversally. The results of the bending tests are shown in Figures 3 and 4, where the mean modulus of rupture (MOR) and modulus of elasticity (MOE) values can be observed for each type of board.

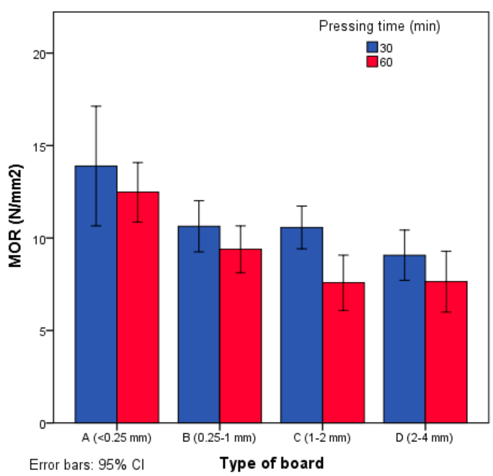

Figure 3. MOR according to type of board.

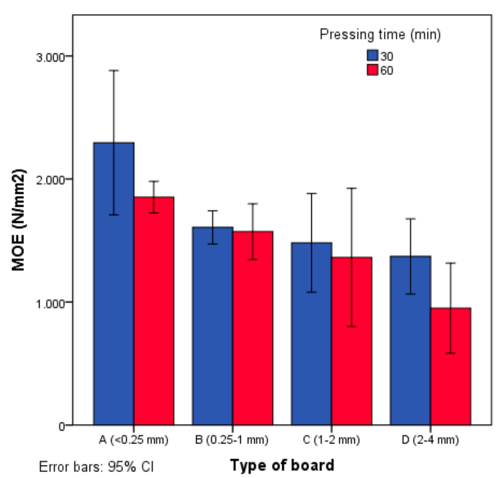

Figure 4. MOE according to type of board.

The values obtained for the modulus of rupture and modulus of elasticity depend on the particle size used, decreasing as the particle size increases. The pressing time has a negative influence, since the longer the time, the worse the performance obtained. Greater strength is achieved with particle sizes <0.25 mm, obtaining a MOR value of 14.01 N/mm^2 and a MOE of 2295.32 N/mm^2. The statistical analysis performed (Table 3) shows that the MOR and MOE depend on the particle size and the pressing time. The smaller the particle size and the shorter the pressing time, the better the results obtained in the bending test.

3.4. Internal Bonding Strength (IB)

The results of the internal bonding strength (IB) test are shown in Figure 5, obtaining high values that range between 0.57 and 1.27 N/mm^2.

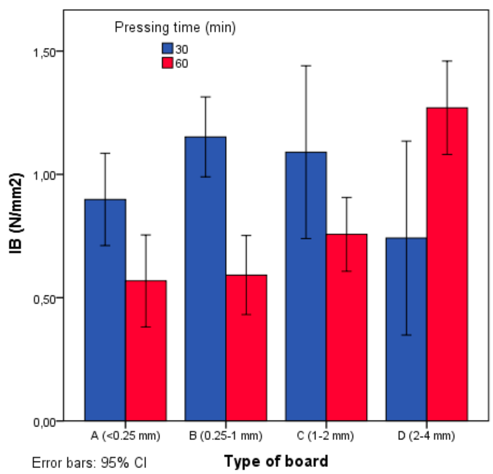

Figure 5. Internal bonding strength according to type of board.

As can be seen in Figure 5, the internal bonding strength depends on the particle size, which is corroborated by the statistical result obtained in Table 3. For a pressing time of 30 min, better values are obtained with a medium particle size and for a pressing time of 30 + 30 min the results improve as the particle size increases. The ANOVA performed (Table 3) indicates that the IB does not depend on the length of time in the hot plate press. Table 4 shows a comparison of the results obtained with the values required by the European standards [55] to determine the compatibility of uses of boards with a thickness of 4 to 6 mm.

Table 4. Characteristics of the type of panels manufactured and classification.

Type of Board	MOR	MOE	IB	TS 24 h
A1	14.01	2295.32	0.90	22.11
A2	12.48	1852.04	0.57	18.24
B1	10.64	1606.83	1.15	26.06
B2	9.39	1571.91	0.59	24.04
C1	10.57	1481.50	1.09	18.62
C2	7.58	1362.81	0.76	20.24
D1	9.07	1370.56	0.74	22.58
D2	7.64	950.65	1.27	33.43
Type P1 [56]	11.50	-	0.31	-
Type P2 [56]	12.00	1950.00	0.45	-
Type P3 [56]	14.00	1950.00	0.50	20.00

The boards manufactured with a smaller particle size and 30-min pressing time (type A1) could only be used as general purpose boards for use in dry conditions (P1) or as boards for interior fitments including furniture (P2). To achieve P3 classification (non-structural boards for use in humid conditions) it would be necessary to consider adding water-repellent substances to reduce the TS value.

In a previous study [50] with a temperature of 110 °C lower strength values were obtained. In this case, by increasing the temperature to 120 °C the strength of the boards has been increased by more than 50%, so further testing is needed to determine the influence of temperature.

Better properties were achieved with almond hulls than with date palm [30,34] or oil palm [35,39] using a lower temperature and, therefore, lower energy expenditure.

With particles of rice straw [42] and Canary Islands palm [40] better properties are achieved when the boards are subjected to pressure in cycles, but with almond hulls their properties decrease. It is necessary to carry out tests manufacturing these boards for shorter times in the hot-plate press, as this material behaves differently when subjected to heat and pressure.

Almond hulls have a large amount of polysaccharides [2,3], so it is possible that some of them may reach the glass transition temperature and contribute to self-binding.

3.5. Result of the Chemical Analysis

Table 5 shows the results obtained in the chemical analysis of sugars performed.

Table 5. Results of the chemical analysis of sugars.

Sugars	g/100 g	St. Dev.
Glucose	-	-
Xylose	3.76	0.09
Arabinose	1.80	0.06

The percentages of sugars detected are very low. Xylose is the most abundant sugar found in the almond exocarp and mesocarp, which contributes most to the formation of the boards. These sugars, although only present in a small proportion, can favour self-binding of the particles.

4. Conclusions

The boards manufactured with a particle size <0.25 mm and 30 min of pressing (type A1) achieve properties that are compatible with use for interior walls and furniture.

The boards were manufactured at a temperature of 120 °C (which can be considered very low) with a pressing time of 30 and 30 + 30 min, so it is necessary to perform further tests with new combinations to try to achieve better properties.

All the properties analysed depend on the particle size used, except for density, for which no significant differences were found between the different types of boards that were tested.

The pressing time influences the MOR, MOE and WA. The longer the time in the hot-plate press, the lower the MOR and MOE and the higher the WA, thus resulting in particleboards with worse properties.

The boards obtained have a high density, so one of the potential markets for this type of board could be as the central core of laminated flooring, for which high-density wood fibre boards are currently being used.

Sugar content, especially xylose, contributes to self-binding of the particles.

This study shows that it is technically possible to manufacture binderless particleboards with almond hulls.

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