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PROPERTIES OF SUNFLOWER HUSK PARTICLEBOARD GLUED WITH POLYMERIC DIPHENYLMETHANE DIISOCYANATE ADHESIVE

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ABSTRACT

This study investigates the suitability of sunflower husks as an alternative raw material for the production of particleboards. For this purpose, shredded sunflower husks bonded with 5% polymeric diphenylmethane diisocyanate (pMDI) were used. Single-layer particleboards were manufactured at three density levels: 800, 850, and 900 kg·m^{-3}. The physical and mechanical properties of the boards were evaluated, including modulus of rupture, modulus of elasticity, internal bond strength, screw withdrawal resistance, water absorption, and thickness swelling after 24 h of water immersion. The results indicate that increasing board density leads to improved mechanical properties. The manufactured boards meet the requirements of EN 312 for board types P2 and P3. In addition, the use of pMDI adhesive contributes to reduced water absorption and low thickness swelling.

Keywords: *particleboards; sunflower husks; polymeric diphenylmethane diisocyanate (pMDI); high-density; properties*

INTRODUCTION

The origins of particleboard manufacturing technology are associated with the utilization of wood residues from roundwood processing [1]. However, modern production increasingly relies on high-quality wood resources, including veneer logs. The growing demand for wood raw materials in construction, wood-based panel manufacturing, and energy production has led to increased consumption and rising prices. This situation necessitates the exploration of alternative, renewable, and cost-effective raw material sources. The use of agricultural lignocellulosic residues in particleboard production offers a viable approach to reducing wood consumption and lowering production costs.

Numerous studies have demonstrated the feasibility of using agricultural lignocellulosic residues as partial or complete substitutes for wood in particleboard manufacturing. Various raw materials have been investigated, including wheat straw [2–4], white mustard straw [5, 6], rapeseed straw [7–9], corn stalks [10], rice straw [11], flax [12], cotton stalks [13], grapevine prunings [14–16], miscanthus stalks [17], bagasse, canola, and hemp [18,47], hemp fibers [19],

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and sunflower stalks [20–22]. Despite their significant potential, the large-scale industrial application of these materials depends on their availability in sufficient quantities, their concentration in specific locations, and year-round supply.

Sunflower husks represent a promising raw material that meets these requirements, as they are generated in large quantities at vegetable oil production facilities. Sunflower (Helianthus annuus L.) is one of the most economically important crops in the European Union. Sunflower seeds are widely used for the production of vegetable oil and related products, ranking among the major global oilseed crops after soybean, oil palm, and rapeseed [23]. In Europe, sunflower seeds are among the most important oilseeds, with significant production, import, and processing activities [24].

Bulgaria plays a key role in the European sunflower sector. It has one of the largest sunflower seed processing capacities in the EU, reaching nearly 3 million tons annually, while domestic production is approximately 2 million tons [24]. The country is also a leading exporter of sunflower products, accounting for a substantial share of EU exports to non-EU markets. In recent years, sunflower seed imports have increased significantly, further contributing to the availability of processing residues such as sunflower husks [27].

The annual production of sunflower seeds in Bulgaria ranged between approximately 1.78 and 2.14 million tons in the period 2021–2023 [25]. On a global scale, Bulgaria ranks among the top ten producers of sunflower seeds [26]. Considering both domestic production and imports, and assuming that sunflower husks account for approximately 25% of the seed mass [28], the potential annual generation of sunflower husks in Bulgaria is estimated to be between 0.55 and 0.76 million tons.

The chemical composition of sunflower husks is comparable to that of wood, consisting of cellulose (27–34%), hemicellulose (27–29%), lignin (24–25%), and extractives (around 9%) [29,30]. This similarity highlights their strong potential as an alternative lignocellulosic raw material for particleboard production.

A review of the literature indicates that while numerous studies have focused on sunflower stalks, research on sunflower husks remains relatively limited. Borysiuk et al. [31] investigated three-layer particleboards with densities of 650 $kg \cdot m^{-3}$ using urea-formaldehyde (UF) adhesive, incorporating 10–30% sunflower husks in the core layer. Although increasing the husk content reduced mechanical properties, the boards still met the requirements of EN 312 for P2-type boards. However, effective bonding was identified as a key challenge due to the presence of oily substances on husk surfaces.

Cosereanu et al. [32] studied the influence of particle size and geometry on the properties of particleboards made from sunflower husks. Their results showed that finer particles improved board compaction, resulting in lower water absorption (WA) and thickness swelling (TS). In contrast, coarser particles led to increased porosity, higher WA and TS values, and reduced mechanical performance, with some boards failing to meet EN 312 requirements.

Zeleniuc et al. [33] examined the effect of adhesive type and content on particleboards made from sunflower husks. Their findings demonstrated that pMDI and phenol-formaldehyde (PF) adhesives significantly improve both physical and mechanical properties compared to urea-formaldehyde adhesives. In particular, pMDI adhesive provided high internal bond strength and reduced water absorption and thickness swelling, even at lower adhesive contents.

The literature clearly indicates that achieving high-performance particleboards from sunflower husks requires adhesives with strong bonding capabilities and high water resistance. Polymeric diphenylmethane diisocyanate (pMDI) adhesives are particularly suitable due to their high reactivity, strong adhesion, and excellent resistance to moisture [34]. The bonding mechanism involves reactions between isocyanate groups (–N=C=O) and hydroxyl groups (–OH) in lignocellulosic materials, forming urethane bonds that enhance strength and durability [35]. Additionally, pMDI adhesives can achieve comparable or superior performance at lower dosages compared to conventional adhesives such as UF, MUF, and PF [36].

For particleboard production, polymeric 4,4′-methylenediphenyl diisocyanate (pMDI), a liquid adhesive with 100% solids content, is commonly used [37].

The aim of this study is to evaluate the suitability of sunflower husks as a raw material for the production of high-density particleboards bonded with pMDI adhesive. The novelty of the study lies in the investigation of high-density single-layer particleboards made entirely from sunflower husks and bonded with a relatively low adhesive content (5% pMDI), with a focus on their physical and mechanical performance in relation to EN 312 requirements.

2 MATERIALS AND METHODS

2.1 Materials

The sunflower husks (*Helianthus annuus* L.) used in the experiment were obtained as a by-product of industrial vegetable oil production. The initial moisture content of the raw material was 9.9%.

The husks were processed into particles using a FCH-500 hammer mill (SKSM “POWER”, Yambol, Bulgaria), equipped with a sieve with round openings of 10 mm in diameter.

The resulting particles had a bulk density of 205 kg·m^{-3}. A visual representation of the particles is shown in Fig. 1.

Figure 1. Particles of sunflower husks

The particle size distribution was determined using a laboratory sieving machine with sieve openings ranging from 4.0 mm to 0.2 mm. Particles smaller than 0.2 mm were removed from the mixture. The particle size distribution is presented in Fig. 2.

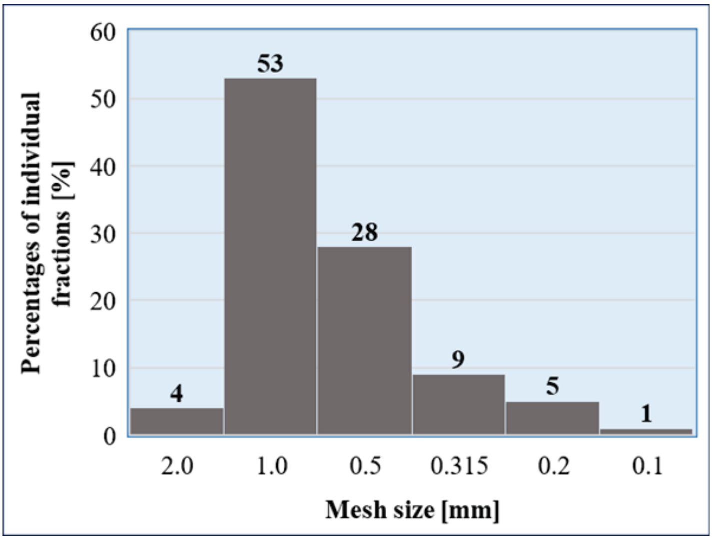

Figure 2. Particle-size distribution of sunflower husk particles: percentage of each fraction

2.2 Adhesives

A solvent-free polymeric diphenylmethane diisocyanate (pMDI) adhesive, trade name ONGRONAT WO 2750 (BorsodChem Zrt., Hungary), was used as a binder. The main properties of the adhesive are presented in Table 1.

Table 1. Properties of polymeric diphenylmethane diisocyanate (pMDI)

NCO % as wt, %	Viscosity 25°C as mPa·s	Acidity as ppm HCl	Functionality
min 30 max 32	min 170 max 230	max 300	2.7

2.3 Production of panels

Particleboards (PB) with dimensions of 500 × 500 × 18 mm were manufactured at target densities of 800, 850, and 900 kg·m^{-3}. The pMDI content in all boards was 5%.

Sunflower husk particles were blended with the adhesive in a mechanical mixer equipped with a rotating blade shaft to ensure uniform distribution. The adhesive-coated particles were then manually formed into a mat inside a wooden forming box placed on a steel plate. The mat was pre-pressed manually to improve compaction before hot pressing.

The formed mat was subsequently transferred to a hot press using the steel plate. To ensure the target thickness, two steel spacers (18 mm thickness) were placed on both sides of the mat. Hot pressing was carried out in a hydraulic laboratory press with electric heating (PMC ST 100, Manni S.p.A., Italy), shown in Fig. 3.

Figure 3. Production of a sunflower husk particleboard in a laboratory press

The pressing temperature was 175°C. The specific pressure ranged from 2.8 MPa to 0.6 MPa during the pressing cycle. The press factor was 15 s/mm. The hot pressing schedule is presented in Table 2.

Table 2. Hot pressing schedule for sunflower husk particleboards

Stage	Pressure,MPa	Temperature, °C	Time, s
Closing	0 - 2.8	175	10
1	2.8	175	30
2	1.2	175	120
3	0.6	175	90
Opening	0.6 - 0	175	20

Table 3. Cross-section of the produced sunflower husk particleboards

Types of manufactured particleboard		
Panel A	Panel B	Panel C
		

After production, the boards were cooled and conditioned for 24 h under laboratory conditions to reach equilibrium conditions. Subsequently, the particleboards were cut into test specimens in accordance with EN 326-1 [38].

The physical and mechanical properties of the particleboards were determined according to EN 310 [39], EN 317 [40], EN 319 [41], EN 320 [42], EN 322 [43], and EN 323 [44]. Each property was evaluated using eight test specimens.

For the screw withdrawal resistance test, standard particleboard screws (4.5 × 50 mm) were used.

Mechanical testing was performed using a universal testing machine HST-50E (Jinan Hensgrand Instrument Co. Ltd., China). The classification of the boards was carried out in accordance with EN 312 [45], considering requirements for board types P2 to P7 with thicknesses between 13 and 20 mm.

Statistical analysis of the experimental data was performed using Microsoft Excel (Data Analysis ToolPak, Microsoft Corporation, Redmond, WA, USA).

RESULTS AND DISCUSSION

The obtained values of the physical and mechanical properties of the produced sunflower husk particleboards are presented in Table 4.

Table 4. Physical and mechanical properties of manufactured sunflower husk particleboards

Property	Unit	Panel A	Panel B	Panel C	REF
Density	$kg{\cdot}m^{-3}$	798 (37.5)	853 (34.16)	896 (39.1)	645 (9.4)
Moisture content	%	4.8 (0.35)	4.5 (0.26)	4.7 (0.33)	8.7 (1.3)
Thickness swelling after 24 h	%	11.6 (1.59)	14.2 (1.6)	14.9 (1.15)	-
Water absorption after 24 h	%	32.0 (3.56)	22.6 (2.8)	19.4 (1.91)	-
Modulus of rupture	$N{\cdot}mm^{-2}$	15.2 (0.536)	21.2 (1.36)	22.7 (1.74)	-
Modulus of elasticity	$N{\cdot}mm^{-2}$	1955 (85.92)	2248 (244.9)	2262 (193.88)	-
Internal bond	$N{\cdot}mm^{-2}$	0.62 (0.08)	0.79 (0.07)	1.05 (0.12)	-
Resistance to axial withdrawal of screws	$N{\cdot}mm^{-1}$	65.5 (5.2)	73.2 (8.3)	97.6 (5.4)	60.4 (4.7)

Note: Mean values are based on 8 specimens. Standard deviations are given in parentheses. The reference board (REF) is an industrial three-layer particleboard bonded with UF resin.

The statistical analysis of the experimental data was performed using one-way analysis of variance (ANOVA). All tests were evaluated at a significance level of $\alpha = 0.05$.

The results confirm statistically significant differences ($p < 0.05$) between the tested boards for all investigated properties, indicating that board density has a strong influence on their physical and mechanical performance.

Table 5. Analysis of variance (ANOVA) for manufactured sunflower husk particleboards

Water absorption after 24 h						
Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	691.245	2	345.624	47.76	1.532E-08	3.46
Thickness swelling after 24 h						
Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	48.373	2	24.186	13.89	0.000143	3.46
Modulus of rupture						
Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	255.94	2	127.97	74.0	3.08E-10	3.46
Modulus of elasticity						
Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	480570.1	2	240285	6.86	0,00507	3.46

Internal bond						
Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	0.726	2	0.363	38.84	8.7771E-08	3.46
Resistance to axial withdrawal of screws						
Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	4489.053	2	2244.526	53.35	5.86191E-09	3.46

The ANOVA results (Table 5) show statistically significant differences among the produced particleboards for all investigated properties, including water absorption, thickness swelling, bending strength, modulus of elasticity, internal bond strength, and screw withdrawal resistance ($p < 0.05$ in all cases).

These findings confirm that increasing the density of sunflower husk particleboards significantly improves their mechanical properties. In particular, higher-density boards (Panel C) demonstrate superior bending strength, stiffness, internal bond strength, and screw holding capacity compared to lower-density boards.

At the same time, a decrease in water absorption is observed with increasing density, which can be attributed to improved compaction and reduced void content within the board structure. However, thickness swelling shows a slightly increasing trend with density, which may be related to the swelling behavior of lignocellulosic particles and their interaction with the adhesive system.

When compared to the reference industrial board (REF), the sunflower husk boards exhibit significantly higher density and comparable or superior screw withdrawal resistance, despite using only 5% pMDI adhesive.

According to EN 312 classification, the produced boards meet the requirements for P2 and P3 type boards, while the higher-density variants approach or meet selected requirements for higher-performance board types.

3.1 Water Absorption and Thickness Swelling

The 24 h water absorption values of the produced sunflower husk particleboards are relatively low. For boards with a density of 800 $kg \cdot m^{-3}$, the water absorption is 32%, while for densities of 850 $kg \cdot m^{-3}$ and 900 $kg \cdot m^{-3}$ it decreases to 22.6% and 19.4%, respectively. This corresponds to a reduction of approximately 39.3% when comparing the highest-density boards with those of 800 $kg \cdot m^{-3}$.

The observed decrease in water absorption with increasing density can be attributed to improved compaction of the particle structure. The small dimensions of sunflower husk particles facilitate better packing and deformation under pressure, which reduces void spaces within the board. As density increases, the reduced porosity limits water penetration into the internal structure of the boards.

A similar trend was reported by Zeleniuc et al. [33], where sunflower husk particleboards with a density of 535 $kg \cdot m^{-3}$ and 6% pMDI resin exhibited significantly higher water absorption (76%), which was associated with their lower compaction level.

The thickness swelling values after 24 h ranged from 11.6% to 14.9%. In contrast to water absorption, thickness swelling shows a slight increasing trend with increasing density, with a rise of approximately 28.4% between 800 $kg \cdot m^{-3}$ and 900 $kg \cdot m^{-3}$ boards.

This behavior can be explained by the higher elastic recovery potential of more densely compacted lignocellulosic particles when exposed to water. Although higher density improves particle bonding, it may also increase internal stresses that contribute to swelling.

The relatively low thickness swelling values are also attributed to the use of pMDI adhesive, which forms strong chemical bonds with hydroxyl groups in lignocellulosic materials, resulting in a compact and moisture-resistant structure that limits water penetration.

The obtained results indicate that sunflower husk boards with densities of 800 and 850 $kg \cdot m^{-3}$ meet the EN 312 requirements for P3-type boards in terms of thickness swelling.

Comparable results were reported by Xinwu et al. [47], who obtained thickness swelling values of approximately 8% for bagasse-based particleboards with a density of 880 $kg \cdot m^{-3}$ and 5% pMDI content.

In contrast, Zeleniuc et al. [33] reported higher thickness swelling values (29.7%) for sunflower husk particleboards, which can be associated with lower density and different adhesive formulations.

3.2 Modulus of Rupture and Modulus of Elasticity

The modulus of rupture (MOR) of sunflower husk particleboards increases with increasing density. For boards with a density of 800 $kg \cdot m^{-3}$, the MOR is 15.2 $N \cdot mm^{-2}$. When the density increases to 850 $kg \cdot m^{-3}$, the MOR rises by approximately 39.5% to 21.2 $N \cdot mm^{-2}$. A further increase in density to 900 $kg \cdot m^{-3}$ results in a minor increase to 22.7 $N \cdot mm^{-2}$ (about 7% compared to 850 $kg \cdot m^{-3}$).

These results suggest that the most intensive improvement in bending strength occurs within the density range of 800–850 $kg \cdot m^{-3}$, where the most effective particle rearrangement and compaction take place. At higher densities, the effect of additional compaction becomes less pronounced.

The obtained MOR values indicate that boards with a density of 800 $kg \cdot m^{-3}$ meet the requirements for EN 312 board type P4, while boards with densities of 850 and 900 $kg \cdot m^{-3}$ satisfy the higher requirements for type P7 particleboards.

The modulus of elasticity (MOE) shows a similar trend. For densities of 800, 850, and 900 $kg \cdot m^{-3}$, the MOE values are 1955, 2248, and 2262 $N \cdot mm^{-2}$, respectively. Increasing density from 800 to 850 $kg \cdot m^{-3}$ leads to an improvement of approximately 15%, while further densification does not result in a significant increase.

This indicates that a near-maximum structural compactness is achieved at around 850 $kg \cdot m^{-3}$, beyond which additional density increase has a limited effect on stiffness. All tested boards meet the EN 312 requirements for type P3 particleboards in terms of MOE.

3.3 Internal Bond Strength

The internal bond strength (IB) increases significantly with increasing board density. Values of 0.62, 0.79, and 1.05 $N \cdot mm^{-2}$ were obtained for boards with densities of 800, 850, and 900 $kg \cdot m^{-3}$, respectively.

The improvement in IB is attributed to increased contact area between particles, reduced void content, and enhanced adhesive performance of pMDI at higher compaction levels. The strongest bonding is achieved in the highest-density boards due to the formation of a more continuous and compact internal structure.

The obtained results indicate that boards with a density of 800 $kg \cdot m^{-3}$ meet EN 312 requirements for type P6, while boards with densities of 850 and 900 $kg \cdot m^{-3}$ satisfy the requirements for type P7 boards.

Comparable results were reported by Zeleniuc et al. [33], who obtained a relatively low IB value of 0.25 $N \cdot mm^{-2}$ for sunflower husk boards with a density of 535 $kg \cdot m^{-3}$ and 6% pMDI content, which was attributed to insufficient densification.

Rofii et al. [46] also demonstrated the strong influence of pMDI, reporting IB values between 0.8 and 2.0 $N \cdot mm^{-2}$ in wood-based composites bonded with 6% pMDI, which was associated with the excellent wetting and bonding ability of the adhesive.

3.4 Resistance to Axial Withdrawal of Screws

The screw withdrawal resistance of sunflower husk particleboards increases with density. For the board with a density of 800 $kg \cdot m^{-3}$, the value is 65.5 $N \cdot mm^{-1}$, which is higher than that of the reference industrial board (645 $kg \cdot m^{-3}$), which shows 60.4 $N \cdot mm^{-1}$.

Increasing the density to 900 $kg \cdot m^{-3}$ results in a significant improvement, reaching 97.6 $N \cdot mm^{-1}$, which corresponds to an increase of approximately 49%.

The improvement in screw holding capacity is related to the higher compaction of the board structure and increased frictional resistance between particles. A denser internal structure provides better mechanical interlocking and load transfer around the screw insertion zone.

CONCLUSIONS

The produced sunflower husk particleboards with densities ranging from 800 to 900 $kg \cdot m^{-3}$ demonstrate improved physical and mechanical performance as a result of increased compaction of the particle structure.

Low 24 h water absorption values were obtained, which are attributed to the reduced porosity and improved internal bonding within the board structure. Thickness swelling values ranged between 11.6% and 14.9%, with a slight increase observed as density increased. All tested boards meet the EN 312 requirements for type P3 particleboards in terms of thickness swelling.

The modulus of rupture increases significantly with increasing density, particularly in the range from 800 to 850 $kg \cdot m^{-3}$. Due to the effective bonding performance of pMDI adhesive, boards with densities of 800 $kg \cdot m^{-3}$ meet the requirements for EN 312 type P4, while boards with densities of 850 and 900 $kg \cdot m^{-3}$ satisfy the requirements for type P7 particleboards.

The modulus of elasticity shows an increasing trend up to a density of 850 $kg \cdot m^{-3}$, after which further densification does not result in significant improvement, indicating a near-optimal structural compactness at this density level.

Internal bond strength increases markedly with board density, confirming the beneficial effect of both increased compaction and the strong adhesive performance of pMDI. As a result, the produced boards meet the highest EN 312 requirements (P6 and P7 classes) at higher density levels.

The screw withdrawal resistance of sunflower husk particleboards (800 $kg \cdot m^{-3}$, 5% pMDI) exceeds that of conventional industrial three-layer particleboards bonded with UF resin at lower density, demonstrating the potential of sunflower husks for structural applications.

Overall, the results confirm that sunflower husks, combined with pMDI adhesive, are a promising raw material for producing high-density particleboards with competitive physical and mechanical properties.

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CONTENTS

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