

Effect of the Particle Geometry and Adhesive Mass Percentage on the Physical and Mechanical Properties of Particleboard made from Peanut Hull

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Peanut hull residues were considered for the manufacturing of particleboards. Various concentrations of two types of adhesive—polymeric diphenylmethane diisocyanate (MDI) and urea-formaldehyde (UF)—were separately combined with four types of peanut hull particles (fine, mixed, coarse particles, and peanut hull powder) to manufacture particleboards with a certain target density. The confidence level of the effect of the selected production parameters on the physical and mechanical properties of the panels was evaluated. The results showed that increasing the adhesive mass percentage significantly improved the dimensional stability of the boards. A better mechanical performance was achieved for the MDI-bonded boards compared with the UF-bonded boards. Superior bonding between the MDI adhesive and the peanut hulls with different particle geometries was also observed; the peanut hull powder and coarse particles were unsuitable for the manufacturing of panels, due to the risk of an internal blowout. The water resistance of the panels was poor, whereas the mechanical strength of the peanut hull particleboard met the class M-1 requirement of the ANSI A208.1 (2009) standard for wood particleboard.

Keywords: Peanut hull particleboard; Urea-formaldehyde; Polymeric diphenylmethane diisocyanate; Physical and mechanical properties; Microstructure; Chemical composition

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INTRODUCTION

The peanut is a popular crop that is cultivated all over the world, mostly in the tropics and subtropics. About 46 million tons of peanuts are harvested throughout the world every year, resulting in 7 million tons of peanut hull obtained as by-product (Guler and Buyuksari 2011). The peanut hulls are often discarded as waste material and immediately burnt or buried after harvest (Dias *et al.* 2016; Miranda *et al.* 2016). Due to the shortage of wood resources and the growing problem of environmental pollution, there is growing interest in using various agricultural residues to produce functional materials, including composite panels (Rowell 1995; Nemli *et al.* 2008; Mohammad *et al.* 2015). In previous studies, residues such as cotton stalk, rice straw, and other fiber-based materials have been successfully used as raw materials for the production of particleboard (Guler and Ozen 2004; Li *et al.* 2010; Uliana *et al.* 2016; Yan *et al.* 2016; Zhao *et al.* 2016; Guler *et al.* 2016).

In general, the raw materials used to fabricate particleboard must have a chemical composition similar to that of wood, which usually contains cellulose,

hemicellulose, and lignin (Copur *et al.* 2007; Barros Filho 2011; Fiorelli *et al.* 2012; Dias *et al.* 2016). Peanut hull consists of 68.8% holocellulose, which includes 42.5% α -cellulose and 28% lignin (Guler *et al.* 2008). The similar lignocellulose content indicates that peanut hull is a good candidate material to replace wood in the manufacturing of particleboard.

Guler *et al.* (2008) tried using a mixture of European black pine (*Pinus nigra* (Arnold)) wood chips and peanut hull (*Arachis hypoqaea* (L.)) with different hull contents (0%, 25%, 50%, 75%, and 100%) and reported an optimal peanut hull to wood chip ratio of 1:4. The panels produced at the optimized ratio showed good physical and mechanical properties.

In addition to mixed with wood as some parts of alternative material in exploring particleboard, peanut hull has been employed as the sole raw material bonded with urea-formaldehyde (UF) adhesive for the fabrication of particleboard with a density of 0.8 g/cm³, meeting the requirements of the TS EN 312 (2005) (Guler and Buyuksari 2011). However, there was a problem in UF bonded board with non-wood particles. The performance of particleboard made from wheat straw with UF was poor (Boquillon *et al.* 2004). A poor quality was also observed by Li *et al.* (2010) when using a UF adhesive to bond rice straw. To overcome the poor distribution of the UF adhesive in the agriculture residues, polymeric diphenylmethane diisocyanate (MDI) has been introduced as a novel, formaldehyde-free adhesive in the particleboard manufacturing industry (Mo *et al.* 2003). Grigorious (1998) found that mixing an UF adhesive with a MDI adhesive improves the quality of the straw particleboard. This resulted from a higher bond-forming ability in MDI adhesive which improved the distribution of adhesive within particles (Li *et al.* 2013). With the higher public sensitivity to the release of formaldehyde in traditional furniture particleboard, the production of healthier and more environmentally friendly particleboard without formaldehyde has become highly desirable (Wang *et al.* 2006).

With large amounts of agricultural waste of peanut hulls without reasonable utilization in China, this study was conducted to evaluate the feasibility of manufacturing composite panels from peanut hulls with different particle geometries and different amounts of UF and MDI adhesives. The manufacturing of particleboard using agricultural residues is well known, but the comparison with two different adhesive bonded boards, and the deeper exploration from a micro perspective is extremely rare when considering the bonding mechanism of particleboards. Above all, such healthy utilization of peanut hulls in the industrial manufacturing of green, environmentally friendly particleboard a potentially effective method to solve problems with waste residues, which result in value-added products.

EXPERIMENTAL

Preparation of Peanut Hull Samples with Different Particle Geometries

Whole raw peanut hulls were obtained from a farm in the Henan Province, China. They were harvested by a local farmer and put through a stripping machine (DX-HSBK20, Qufu Dexin Machinery Equipment Co. Ltd., Qufu, China) to separate the peanut seeds from the hulls. The average moisture content of the obtained peanut hulls was between 10% and 13%. After delivery, the peanut hulls were divided into four types of samples using a sieve with an opening size of 12.7 mm. The peanut hulls were classified into four different particle geometries: the original particle mixture, the coarse particles remaining on top of the sieve, the fine particles which passed through

the sieve, and a powder with a particle size below 1 mm that were milled using a knife ring flaker (BX 4612/5, NJ-Yunqing Mechanical and Electrical Equipment Co. Ltd., Nanjing, China).

Adhesive Preparation

The commercial urea-formaldehyde (UF) and polymeric diphenylmethane diisocyanate (MDI) adhesives used in this study were procured from Sanlian Adhesive (Shenzhen, China) and Guanhai Chemical Products Corporation (Zhengzhou, China), respectively. The UF was dispersed in water to a solids content of 46.05%, and 1% (compared with the dried quantity of peanut hull) ammonium chloride (NH_4Cl , CAS 12125-02-9, Linhang Chemical Co. Ltd., Yangzhou, China) was added to the UF as hardener.

Manufacturing of the Peanut Hull Particleboard

Five different amounts of adhesive were chosen to evaluate the effect of the adhesive mass percentage on the performance of the peanut hull particleboard. For the UF, either 2 wt.%, 4 wt.%, 6 wt.%, 8 wt.%, or 10 wt.% were added. For the MDI, either 8%, 10%, 12%, 14%, or 16% were added. All prepared materials were placed into the hot-press machine (YBV-1S, Junlong Machinery Co. Ltd., Shiyang, China) and pressed at 170 °C by applying a pressure of 3.5 to 4.5 N/mm² for 5 min to prepare boards with a target density of 0.8 g/cm³. The dimensions of the particleboard fabricated in this study were 420 mm (length) x 420 mm (width) x 15 mm (thickness) after pressing. The boards were edge-trimmed to dimensions of 350 mm (length) x 350 mm (width) x 15 mm (thickness). First, the optimum amount of adhesive was determined according to the physical and mechanical results. Particleboards were fabricated from the four different types of particle samples to determine the optimum particle geometry. An orthogonal experimental design was employed to optimize the performance of the MDI-bonded board and to determine the effect and the order of the three interacting factors, *i.e.*, the particle geometry, the adhesive mass percentage, and the target density (Table 1).

Table 1. Orthogonal Experimental Scheme Employed for the Peanut Hull Particleboard Manufacturing Experiments

MDI-bonded Board Test Sample Designation	Particle Geometry	MDI Adhesive Mass Percentage (wt.%)	Target Density (g/cm ³)
A	fine	6	0.7
B	fine	8	0.8
C	fine	10	0.9
D	mixture	6	0.9
E	mixture	8	0.7
F	mixture	10	0.8
G	coarse	6	0.8
H	coarse	8	0.9
I	coarse	10	0.7

Physical and Mechanical Testing

The samples for measuring the water absorption (WA), thickness swelling (TS), internal bond strength (IB), modulus of elasticity (MOE), and modulus of rupture (MOR) were prepared, and the properties were tested in accordance with the ASTM D 1037-06a (2006) standard for evaluating the properties of wood-based fiber and particle panel material. All mechanical properties were determined using a universal

testing machine (LD23.502, Labsans Corporation, Shenzhen, China). The TS and WA values were measured after 24 h of immersion in distilled water at 20 °C. Each given value corresponds to the average, which was determined by studying six samples cut from two particleboards fabricated under the same conditions.

Statistical Analysis

Duncan's multiple range test and analysis of variance were performed on duplicate sets of experimental data to determine if there were significant differences among the production parameters ($p < 0.05$). SPSS 19.0 software (IBM, Armonk, NY, USA) was used to analyze the gathered data.

Microstructure and Chemical Analysis

Scanning electron microscopy (Quanta450, FEI, Hillsboro, OR, USA) and energy dispersive X-ray spectroscopy (INCA X-ACT250, Oxford Instruments, London, UK) were employed to analyze the microstructure and chemical composition of the fabricated peanut hull particleboards.

RESULTS AND DISCUSSION

Particleboard Properties

Thickness swelling and water absorption are the key parameters for describing the dimensional stability of wood composites. Li *et al.* (2010) observed that the TS, WA, and LE values of rice straw particleboard increased when the UF adhesive mass percentage was increased from 12 wt.% to 16 wt.%. A similar relationship was observed between the adhesive mass percentage and the TS and WA values (Fig. 1). By increasing the MDI adhesive mass percentage, the TS was reduced from 60.08% to 4.03%, and the WA decreased from 88.20% to 40.88%. For the UF adhesive-bonded boards, the TS declined from 48.53% to 14.27%, and the WA decreased from 97.19% to 47.24%. Similarly, high TS values of agricultural residue board have been reported near 60.70% for particleboards made from tobacco leafs (Kalaycioglu 1992). The poor dimensional stability of the particleboards can be improved by increasing the adhesive mass percentage or coating the board with a hydrophobic substance (Nemli *et al.* 2005). Furthermore, a heat treatment, the use of a water repellent chemical (such as paraffin), and the acetylating of the particles also effectively improve the water repellency (Guler and Ozen 2004; Guler *et al.* 2006).

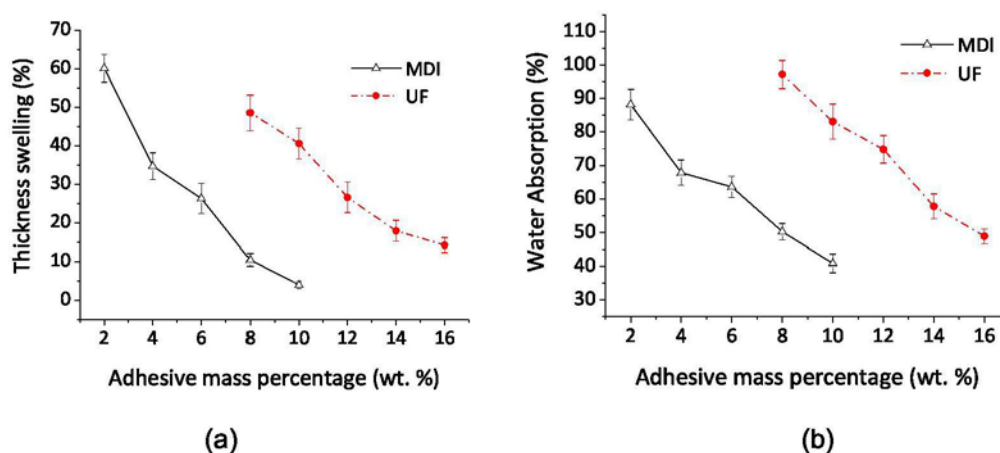


Fig. 1. Effect of the adhesive mass percentage on the physical properties of the prepared peanut hull particleboards. (a) Thickness swelling (TS) and (b) water absorption (WA)

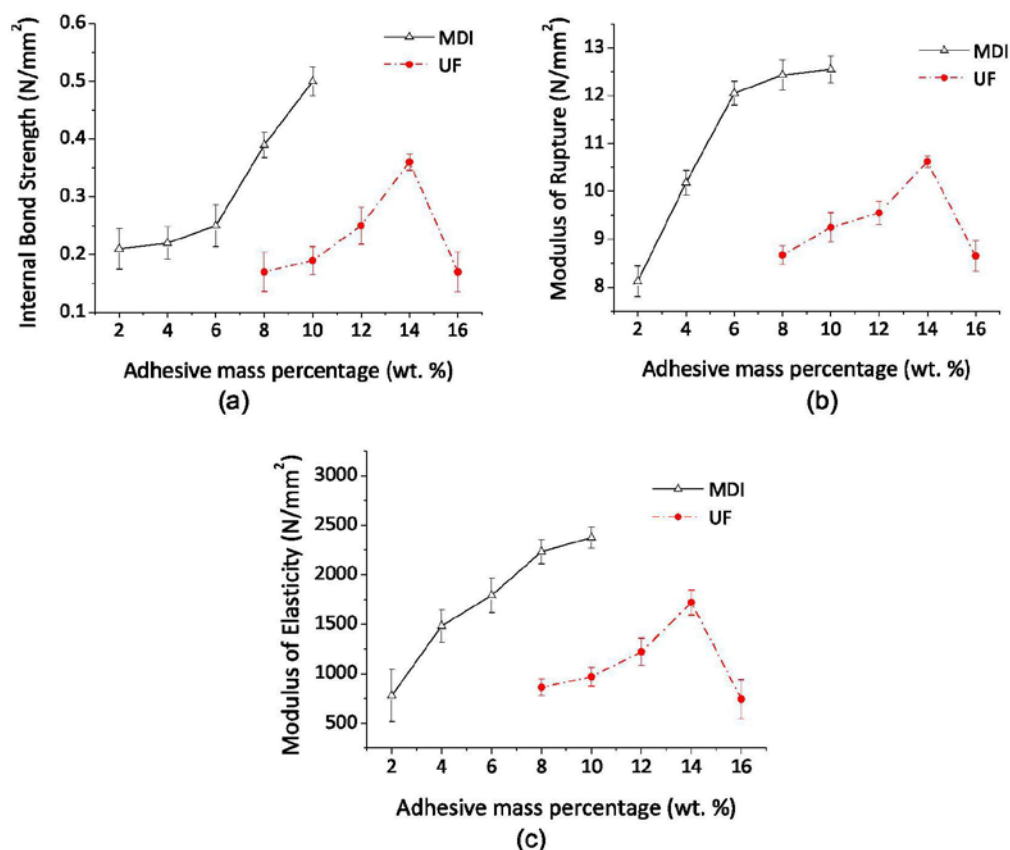


Fig. 2. Effect of the adhesive mass percentage on the mechanical properties of the prepared peanut hull particleboards. (a) Internal bond strength, (b) modulus of rupture, and (c) modulus of elasticity

The effect of the adhesive mass percentage on the IB strength and static bending properties (MOR and MOE) of boards is illustrated in Fig. 2. All mechanical properties of the MDI-bonded boards were enhanced by increasing the adhesive mass percentage. There was a visible increment in the IB and the MOR strength when more than 6 wt.% MDI was added to the peanut hull particles.

For the UF bonded-boards, the mechanical properties were first enhanced, but then dropped rapidly when the UF adhesive mass percentage was higher than 14 wt.%. The IB, MOR, and MOE values first increased from 0.17 to 0.39 N/mm², 8.67 to 10.62 N/mm², and 864 to 1720 N/mm², respectively, when the UF adhesive mass percentage was increased from 8 to 14 wt.%.

The IB, MOR, and MOE values decreased to a minimum of 0.17 N/mm², 0.12 N/mm², and 743 N/mm², respectively, for the samples prepared with 16 wt.% UF. This result can be traced back to an internal blowout due to a high vapor emission as part of the pressure release process, which is a common failure in wood and particleboard panels.

The evaporation of more than half of the water contained in the UF adhesive easily and quickly generates a high vapor pressure, whereas using the anhydrous MDI adhesive avoids the risk of an internal blowout caused by water vapor. The increase of the bending properties with increasing adhesive mass percentage is probably due to the increase of the internal bonding strength.

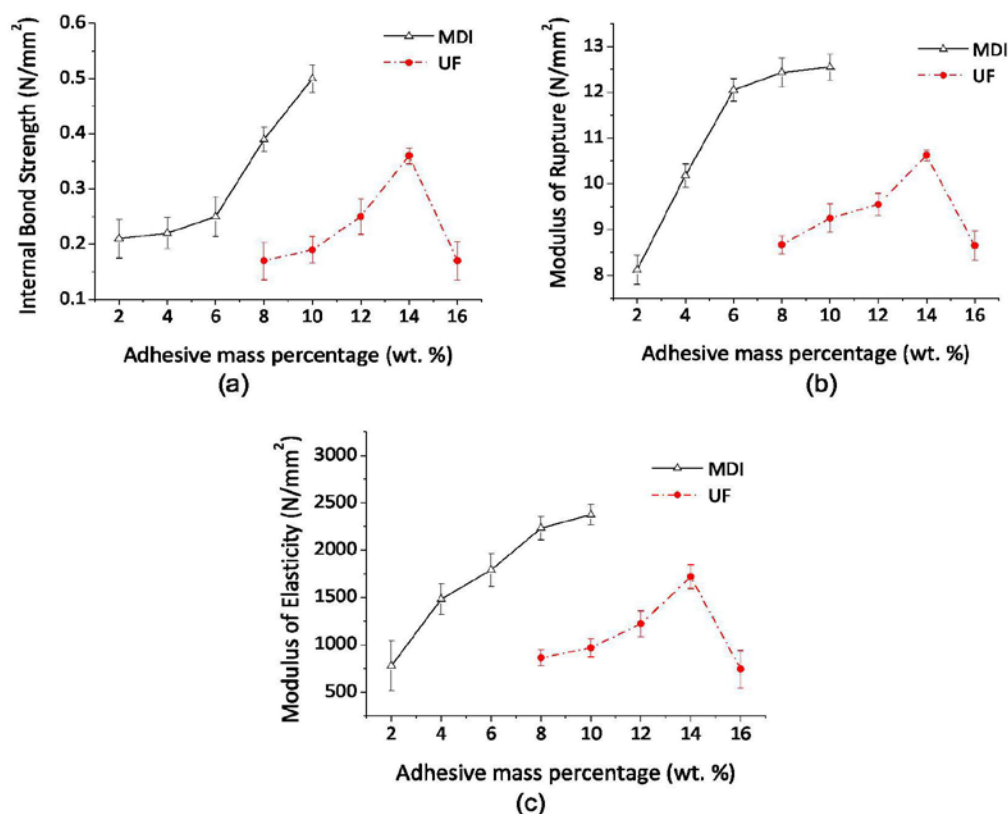


Fig. 3. Effect of the particle geometry on the mechanical properties of the fabricated peanut hull particleboards. (a) Internal bond strength, (b) modulus of rupture, and (c) modulus of elasticity.

With adhesive mass percentages of 8 wt.% and 14 wt.% determined to be the optimal contents for the MDI- and the UF-bonded boards, respectively, the effect of the particle geometry on the mechanical properties (IB, MOR, and MOE) of the prepared particleboards was studied (Fig. 3). Compared with the UF-bonded boards, a better mechanical performance was obtained for the MDI-bonded boards. In general, particleboard made with larger particles shows a higher mechanical strength (Yemele *et al.* 2008). However, the best mechanical performance in the experiments was achieved by the samples made from the mixture particles, rather than for the largest, coarse particles. The results showed that increasing the particle size obviously improved all mechanical properties, and the maximum IB, MOR, and MOE values of 0.39 N/mm², 12.65 N/mm², and 2292 N/mm², respectively, were observed for the particle mixture for both the MDI- and the UF-bonded specimens. Furthermore, increasing the particle size by using the large, coarse hulls, however, resulted in a decrease of the IB, MOR, and MOE values by about 7.69%, 8.77%, and 16.32%, respectively, for the MDI-bonded specimens, and they were significantly decreased by about 66.67%, 64.88%, and 78.39%, respectively, for the UF-bonded specimens. In the UF-bonded specimens, combining a lower adhesive mass percentage of 14 wt.% with larger, coarse particles caused an internal blowout. In this study, the best IB, MOR, and MOE values observed for the MDI-bonded specimens exceeded the requirements of the ANSI A208.1 (2009) standard (0.36 N/mm², 10.0 N/mm², and 1550 N/mm², respectively) in the M-1 class for medium-density wood particleboard.

Aside from the adhesive mass percentage and the particle geometry, the target density of the boards was also a very important factor affecting the physical and mechanical properties of the panels (Dias *et al.* 2005). The results of the range

analysis conducted for the physical and mechanical properties of the prepared peanut hull particleboards are presented in Table 2. The physical properties (TS and WA) were mostly affected by the target density (C), and significant P values were obtained for the effect of the WA. ANOVA showed a significant effect with a confidence level of 95% (Table 3). When increasing the target density, more particles were included in the board at a higher compaction ratio for the same panel dimensions, resulting in a poor dimensional stability, which was mainly attributed to the higher proportion of the hygroscopic raw materials (Hammen *et al.* 2001). The bending properties (MOR and MOE) were highly dependent on the internal bonding strength of the boards, and all three mechanical properties were significantly impacted by the particle geometry (A) and the adhesive mass percentage (B) at a confidence level of 99% ($P < 0.01$), with the primary factor being the particle geometry. Some physical properties of the fiber-reinforced composites strongly depend on the fiber form factor (*i.e.*, the length to diameter ratio), which determines the flexibility and strength of boards through disparate fibers (Boquillon *et al.* 2004). Thus, combining a suitable and eligible fiber raw material with a bonding adhesive guarantees high quality in fabricated particleboards.

Table 2. Range Analysis of MDI-bonded Peanut Hull Particleboards

Types of Mechanical Properties	IB			MOR			MOE		
	k_1	k_2	k_3	k_1	k_2	k_3	k_1	k_2	k_3
A	0.34	0.39	0.46	10.08	12.96	11.55	1485	2226	1860
B	0.37	0.34	0.48	11.42	12.22	12.35	1656	2198	2327
C	0.36	0.37	0.39	9.70	10.25	10.63	1583	1815	2227
Order	A > B > C			A > B > C			A > B > C		

Types of Physical Properties	TS			WA		
	k_1	k_2	k_3	k_1	k_2	k_3
A	24.71	22.88	19.36	68.46	65.55	62.26
B	23.09	21.60	22.25	70.17	67.49	66.63
C	19.25	22.76	24.93	64.44	68.19	78.63
Order	C > A > B			C > A > B		

Note: A, the particle geometry; B, adhesive mass percentage; C, target density

Table 3. ANOVA Results for MDI Adhesive-bonded Particleboard Properties

Factor Types	P value For Given Property				
	TS	WA	IB	MOR	MOE
A	0.377	0.085	0.000**	0.000**	0.000**
B	0.802	0.201	0.001**	0.000**	0.001**
C	0.320	0.044*	0.012*	0.000**	0.012*

* $P < 0.05$; ** $P < 0.01$

Surface Characterization

SEM micrographs of the peanut hull surface (Fig. 4a) revealed a rough interface of stacked flakes with various vacancies. This rugged structure was not conducive for the wetting and coating of the hull by the adhesive, and the better performance of MDI-bonded boards in the results above can be explained by the better wettability and the smaller contact angle (Shen *et al.* 2011). A structure similar to the tracheid form in wood can be seen in Fig. 4b. Some gaps and holes between the particles were filled with the adhesive, and the combination of raw particles and the adhesive had a large impact on the internal bonding and bending strength of the produced boards. The poor dimensional stability of the boards may have been related

to the tubular and pore structure found in the peanut hulls, which led to the formation of a capillary system, thereby promoting the absorption of water. Therefore, filling parts of the holes and pores with water-repellent chemicals or adding more adhesive was recommended and essential for improving the water resistance and inhibiting the formation of the capillary system. The EDS (electronic differential system) spectrum obtained for the peanut hull shows the mass percentage of carbon, oxygen, and silicon at 40.86%, 31.95%, and 27.19%, respectively. The high amount of hydrophobic silicon in the peanut hull restricted the wetting and coating of the adhesive, resulting in the poor performance of the UF-bonded boards when dispersed in water. Focused on such defects, steam-based pre-treatment, optimizing adhesive by adding modifier, and seeking another better adhesives could be used to deal with the solutions (Han *et al.* 1998; Khristova *et al.* 1998; Zhang *et al.* 2003). The MDI-bonded boards with their higher bondability, smaller contact angle, and absence of formaldehyde had a much higher potential for replacing wood in residential applications and environments.

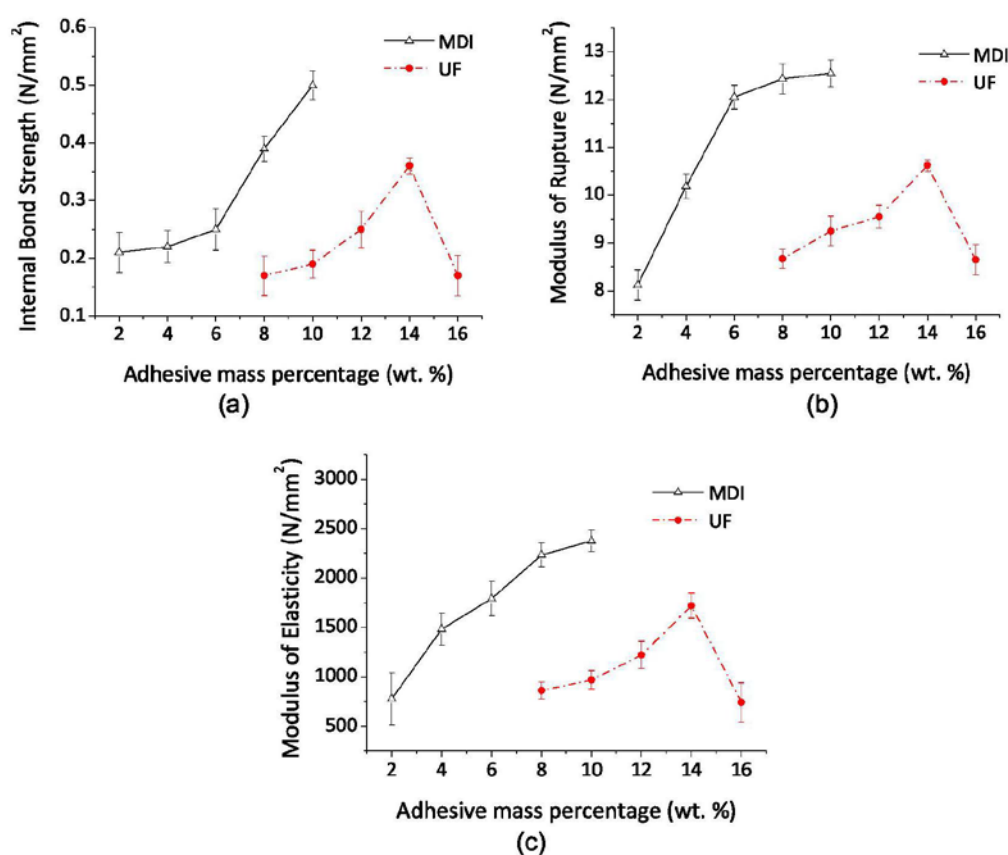


Fig. 4. SEM micrographs of (a) peanut hull surface and (b) peanut hull fracture; (c) EDS spectrum of peanut hull

CONCLUSIONS

1. Increasing the adhesive mass percentage significantly improved the dimensional stability of the peanut hull particleboards fabricated in this study.
2. Peanut hull powder and coarse particles were not suitable for the manufacturing of particleboard panels, due to risk of an internal blowout during the hot pressing process.

3. An overall better performance was achieved for the MDI-bonded boards, where the target density significantly affected the TS and WA with a confidence level of 95%, and the particle geometry and the adhesive mass percentage affected the mechanical strength (IB, MOR, and MOE) at a confidence level of 99% in the primary and secondary order.
4. The water resistance performance of the boards was poor, whereas the mechanical strengths of the boards met the class M-1 requirement of the ANSI A208.1 (2009) standard for wood particleboard.
5. The presence of tubular and porous fiber morphology, consisting of a capillary system which strongly promotes water absorption, resulted in a poor dimensional stability of the boards. In addition, the high weight percentage of silicon (27.19%), combined with the rough interface, was conducive to the coating and wetting of the peanut hull by the MDI due to a smaller contact angle. Because peanut hull bonded with an MDI adhesive exhibits a nice aroma, formaldehyde-free emissions, and a qualified strength, it has huge potential for utilization in residential environments.

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