

DURABILITY EVALUATION OF CEMENTITIOUS BOARDS WITH PEANUT HUSKS

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ABSTRACT

This paper presents the results of an experimental study using peanut husks as potential application in natural fiber composites for ceiling and wall covering panels. Natural fiber-reinforced cementitious composites are produced from the residue of the harvesting and processing of peanuts. Peanut husks were cut and particles higher than 0.3 mm were used as 5% (by mass) aggregates in the cement matrix. The peanut husks were treated with sodium silicate and aluminium sulphate. The composites were prepared using the slurry dewatering and pressing method with treated and untreated peanut husks. The physical and mechanical properties were evaluated after 28 days, after 365 days of natural ageing and after accelerated ageing (200 soak and dry cycles). Statistical analyses indicate a significant difference ($p < 0.05$) between the modulus of rupture of the composites with treated and untreated peanut husks. The results of the physical-mechanical properties from different ages indicated a significant difference ($p < 0.05$) for modulus of rupture, elasticity modulus, apparent porosity and water absorption between composites at 28 days and after 365 days of natural ageing or after 200 accelerated ageing cycles.

KEYWORDS:

Peanut husk, cement composites, ageing, construction material, fiber composites

INTRODUCTION

Tropical countries present significant opportunities for the production of non-wood lignocellulosic fibres (Guimarães, 1990; Tonoli et al., 2007; Tonoli et al., 2011), especially if they are available from by-products of the major commercial agricultural activities (e.g., peanut). Due to the production scale and the capacity of consuming materials, the building sector is a valid alternative destination for wastes that cannot return to the environment in a sustainable way. Agroindustry by-products generally have little application, and most of them have been used in products of low added value such as cattle food, animal litter or fuel for boilers.

Fibrous residues are suitable reinforcement materials for brittle matrices even though they present relatively poor durability performance (Joaquim et al., 2009; Tonoli et al., 2011). Accounting for the mechanical properties of the fibres as well as their broad variation range, one may develop building materials with suitable properties by means of the adequate mix design (Agopyan et al., 1989; Agopyan and John, 1989; Joaquim et al., 2009; Rabi et al., 2009). A major advantage concerning fibre reinforcement of a brittle material (e.g., cement paste, mortar or concrete) is the composite behaviour after cracking. Post-cracking toughness provided by the fibres in the material may allow large-scale construction use of such composites (Agopyan et al., 2005).

There are two approaches for the development of new composites in fibre-cement. The first one is based on the production of thin sheets and other asbestos-free components. The later components are similar to asbestos-cement ones and they are produced by well-known industrial-scale processes such as Hatschek and

Magnani methods commercially used with high acceptance for building purposes (Ikai et al., 2010; Dias et al., 2010). The second approach consists of producing composites for different types of building components like load-bearing hollowed wall, roofing tiles, and ceiling plates, which are not similar to components commercially produced with asbestos-cement (Rabi et al., 2009). This is because such materials may be used to produce lightweight building components with good mechanical performance (mainly impact energy absorption) and suitable thermal-acoustic insulation, while being economically attractive (Agopyan et al., 2005).

Since 2008, Argentina has been the first supplier for the worldwide market of peanut, followed by India and the U.S.A. 95% of the national production (almost 900,000 ton/year) of peanut is harvested and processed in the province of Cordoba - Argentina. In Brazil, the peanut production currently accounts for 296,000 ton/year and 80% of Brazilian production is in the state of São Paulo.

The peanut husk protects the peanut grain in storage where strict humidity control exists. One-third by mass of the total harvest corresponds to the peanut husk or box, which is separated during selection and processing of the peanuts. Their quantity and hardness restricts market for their use and has led to intense generation of residues with high pollution potential.

The main use for peanut husks has been incineration. Open air burning of peanuts husks generates smoke of varied composition, ashes and carbon dioxide and the main current use is as fuel in boilers. In some cases, economic impact either exists, due to the costs of transportation, storing and incineration. As a result at peanut processing plants, the availability of peanut husks is abundant, concentrated, and constitutes a potential resource to be used in building materials.

Due to the high demands on the capabilities and operative life of materials, coating technology or composite material is used to enhance the lifetime of materials. However, the stress singularities induced by the discontinuous character between the different layers may generate cracks and the composite medium thus becomes very susceptible to spallation. Thus, the concept of the so-called functionally graded material (FGM) has been introduced to eliminate singular stresses, relax residual stresses, and enhance bonding strength. The most distinct features of an FGM are the non-uniform micro-structures in materials with continuously graded macro-properties, cited by Jin and Batra (1996). Consequently, in an FGM, the interfaces between two materials disappear but the characteristics of two or more different materials of the composite are preserved. Chung Cheng (2007).

This paper proposes the use of one of the agro industrial wastes for the manufacture of innovative building materials. The main objective is to evaluate some of the key properties of peanut husk cement composite in order to demonstrate a technically viable application for this waste as a particleboard material. The properties evaluated are visual aspect and physical and mechanical performance.

MATERIALS AND METHODS

Raw Materials

Cement based composites were reinforced with peanut husks. The main characteristics of the raw materials used for the preparation of the cement based composites are presented in this section.

Peanut Husks

Peanut husks (*Arachis hypogea*) were used as aggregate materials due to their varied possibilities to be cut in particles with different sizes. They were provided by AMENDOBRAS Imp. Amendoim Ltda., Bauru, estate of São Paulo, Brazil.

According to the characterization that was carried out in the Laboratory of Agricultural Microbiology, Faculty of Agricultural Sciences, National University of Cordoba, the chemical composition of the peanut husks is (percentage by mass): 54.5% of cellulose, 2.1% of hemicelluloses, 27.8% of lignin and 4.5% of extractives. The experimental analysis of the fibres composition for insoluble fibres (cellulose + lignin) and soluble fibres (hemicellulose) was determined by the gravimetric enzymatic method (Asp et al., 1983). Lignin was determined by cellulose separation from insoluble fibres by sulphuric acid washing (72%) (Van Soest and

Robertson, 1980); and carbon (C) soluble concentration was determined by the humid digestion of Walkley and Black method (Nelson and Sommers, 1982).

Thin particles of peanut husks were cut and air dried at 32°C for 5 h. Subsequently, the particles were oven dried (60°C) for 24 h. Then, the particles were classified through a sieve classifier during 5 min and four ranges of particle sizes were obtained.

The particle size distribution used in this work was based on the sizes between the # 50 and #16 sieves (M4). Small and big peanut husks particles were discarded. (Table 1)

Table 1 - Particle sizing of peanut husks used.

	Nominal sieve opening (mm)	ABNT/ASTM #	TYLER/MESH #
M4	0.300 - 1.18	50 -16	48 - 14

Ordinary Portland Cement and Carbonate Filler

High initial strength ordinary Portland cement (OPC) CPV-ARI (Brazilian Standards NBR 5733, 1983) and ground carbonate material were used in the matrix of the cement based composites. Ground carbonate material was used in order to reduce the production costs of the fibre-cement and also to mitigate volume changes of the final composite. Oxide composition of the OPC is (percentage by mass): 19.4% SiO₂, 63.5% CaO, 4.1% Al₂O₃, 2.3% Fe₂O₃, 3.1% MgO, 1.1% K₂O and 3.0% SO₃, 0.2% Na₂O. Oxide composition of the ground carbonate material is (percentage by mass): 9.0% SiO₂, 39.1% CaO, 2.2% Al₂O₃, 1.2% Fe₂O₃, 8.9% MgO, 0.2% P₂O₅, 0.4% K₂O, 0.1% TiO₂, 0.1% MnO and 0.1% Na₂O. According to particle size distribution, 50% of the particles are smaller than 11.0 μm and 16.2 μm for OPC and ground carbonate material respectively. Most of particles (90%) are smaller than 27.3 μm and 64.4 μm for OPC and ground carbonate material respectively.

Composites Formulation

Formulations were prepared using the following constituents (percentage by dry mass): 1% of cellulose pulp, 4% of peanut husks, 80% of OPC, 15% of ground carbonate material, and distillate water. In the case of formulations without cellulose pulp, the formulation was: 5% of peanut husks, 80% of OPC, 15% of ground carbonate material, and distillate water. In both cases, the water/cement (w/c) ratio obtained was 0.25.

Chemical Treatment

According to the literature, fibre treatment improves the interface between fibres and the matrix (Gram, 1983; Guimarães, 1990; Delvasto et al., 2004; Tonoli et al., 2009; Tonoli et al., 2010). Generally, chemical treatments are used to clean the fibre surface, and/or seal the fibre pores.

Peanut husks were chemically treated with Sodium Silicate and Aluminium Sulphate. The process is described by Ramirez Sarmiento (1996). Cut peanut husks were oven dried at 60°C for 24 h. Then, they were immersed in a solution of Sodium Silicate and Aluminate Sulphate and mechanically stirred. Each 25 g of peanut husks were immersed in 500 g of Sodium Silicate solution at 5 wt. % for 1 h. The husks were filtered in a vacuum system for 5 min. After the same amount of the husks was impregnated with 500 g of Aluminates Sulphate solution at 30% wt for 1 h, and they were filtered in a vacuum system for 5 min. They were dried at environment air condition for 1 h. Later, peanut husks were oven dried for 24 h, cooled and stored in sealed container for additional 24 h. After this treatment, the peanut husks were ready to be used in the cementitious mixtures.

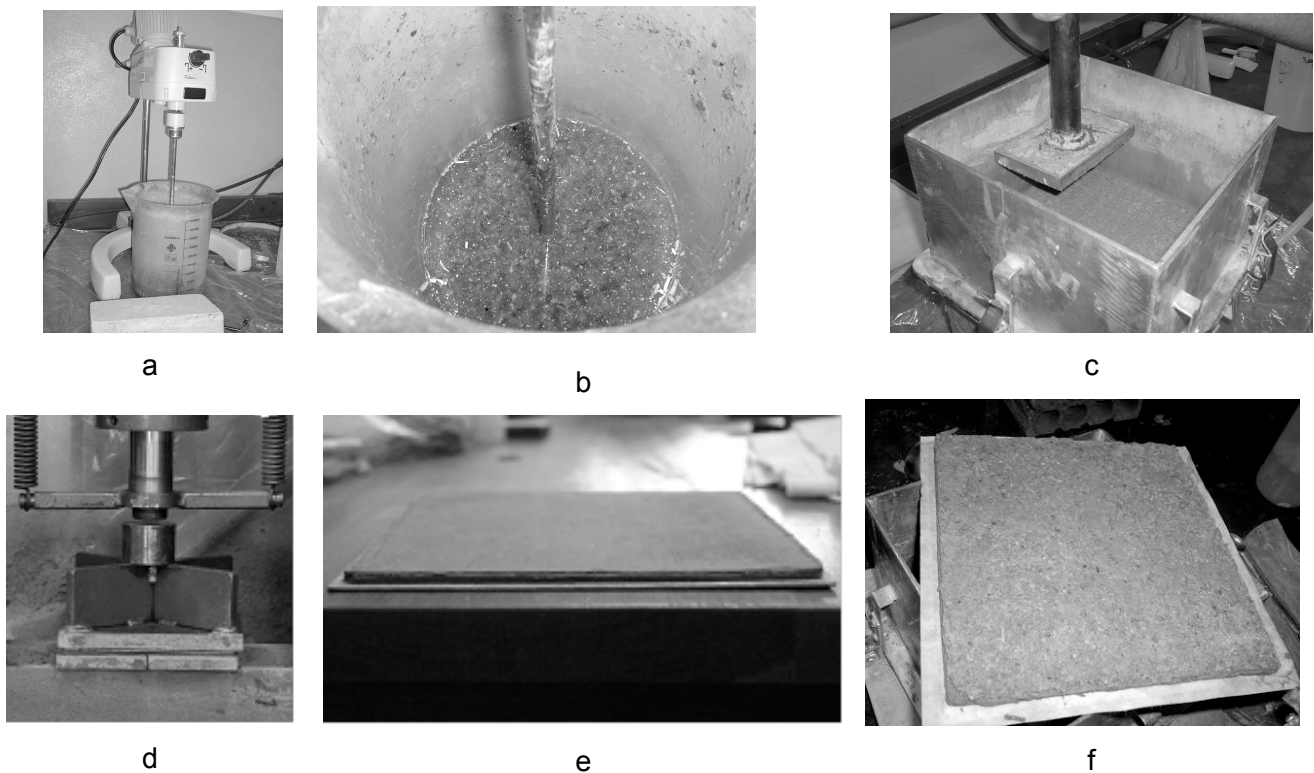


Figure 1 - Composite preparation: (a) mechanic mixer; (b) floating husks; (c) molding at the evacuable casting chamber; (d) pressing; (e) pad composite after pressing; and (f) composites after curing in plastic bags.

Composite Preparation, Physical and Mechanical Characterization

The cement based composites were moulded in pads measuring 200 mm x 200 mm. They were prepared in laboratory using the slurry vacuum de-watering followed by pressing technique as described by Santos et al. (2011).

Eucalyptus pulp was dispersed in water for 5 min. The peanut husks and the matrix components were then added to form a slurry material. The final mixture formed with approximately 25% of solids was stirred at 1000 rpm for 4 min. The slurry was transferred to the evacuable casting box and vacuum was applied until a solid surface formed. The pads of each formulation were pressed individually at 3.2 N/mm^2 for 5 min. Pads were then sealed wet in a plastic bag to cure at room temperature for two days and then cured in a controlled climate chamber for additional five days. The moulding technique and curing procedures adopted for the composites manufacture are depicted in Figure 1.

Physical and Mechanical Tests

Pads were cut wet into four 165 mm x 40 mm flexural test specimens using a diamond saw cooled with water. Specimen thickness was approximately 6 mm. Prior to mechanical test, specimens were oven dried at $60 \text{ }^\circ\text{C}$ for 24 h, after completing twenty eight days of cure.

Physical properties (water absorption - WA, bulk density - BD and apparent porosity - AP) were obtained from the average of four specimens for each formulation, following the procedures specified by the ASTM C 948-81 (1981) Standards.

Mechanical tests were performed in a universal testing machine Emic DL- 30000 equipped with 1 kN load cell. A four-point bending configuration was employed for the determination of the values of modulus of rupture (MOR), limit of proportionality (LOP), modulus of elasticity (MOE) and toughness (T). A span of 135 mm and a deflection rate of 1.5 mm/min were adopted in the bending test.

Wet/Dry Accelerated Cycles and Weathering Conditions Aging

The wet/dry accelerated cycles test aims to simulate natural ageing with exposure to alternated cycles of wetting and drying. Specimens were successively immersed in water at 20 ± 5 °C during 18 h and exposed to the temperature of 60 ± 5 °C for 6 h in a ventilated oven as following standard EN 494 [23]. Each wet/dry procedure represents one cycle and it was performed for 200 times (200 cycles).

Specimens were exposed during 1 year under natural weathering conditions, in Pirassununga, Brazil (latitude $21^{\circ}59'46''$ S, longitude $47^{\circ}25'33''$ W, and mean elevation of around 627 m). During the period under consideration the maximum and minimum temperatures were 21.4 °C and 21.1 °C, respectively, average wind speed was 1.9 km/h, rainfall was approximately 1300 mm/year, maximum and minimum relative humidity were 84.2% and 62.5%, respectively, and solar radiation was around 376 cal/cm²/day.

RESULTS AND DISCUSSION

Visual Aspect

During the mixing procedure, the fibrous husks float. Consequently, most of them stay on the upper surface of the plates during the moulding process. The results are attractive and with textural surfaces leading to a natural aspect to the plates.

The aspect of the pads suggests an interesting use as textured wall-covering panels (Figure 2). The addition of the peanut husk particles combines good distribution of particles in the cement mass and attractive surface appearance.

Having intentionally obtained FGM we found it resulted in better visual appearance and functionality. One of the surfaces had an attractive appearance, texture and colour while the other side was smooth cement and had probably better hygroscopic properties

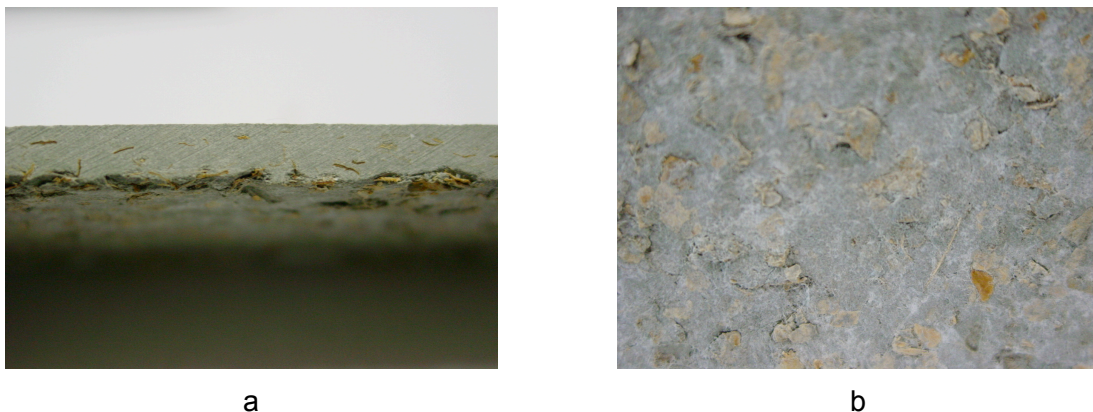


Figure 2 - Composite after curing: (a) cross-section of the composite; (b) surface of the composite.

Effect of Ageing and Chemical Treatment of the Husks on the Composite Properties

Durability of vegetable fibre reinforced cement composites is related to the ability to resist both external (temperature and humidity variations, sulphate or chloride attack, etc.) and internal damage (compatibility between fibres and cement matrix, volumetric changes, etc.). The degradation of natural fibres immersed in Portland cement is due to the high alkaline environment which dissolves the lignin and hemicellulose phases, thus weakening the fibre structure (Gram, 1983).

In order to improve the durability of fibre reinforced cement composites chemical coating of the natural fibres could be used to avoid water absorption or penetration of free alkalis. Pacheco-Torgal & Jalali (2011) relates the use of water-repellent agents or fibre impregnation with sodium silicate, sodium sulphite, or magnesium sulphate with the chemical interactions between the cement matrix and the natural fibres.

Peanut husks were chemically treated with sodium silicate and aluminium sulphate. The results of 200 soak and dry cycles test and weathering ageing on the mechanical and physical properties of the composites with untreated and treated particles were compared.

Composites without chemical treatment of the particles decreased their mechanical properties due to the effects of ageing. In these composites water absorption and apparent porosity tend to decrease and the density values remain almost constant.

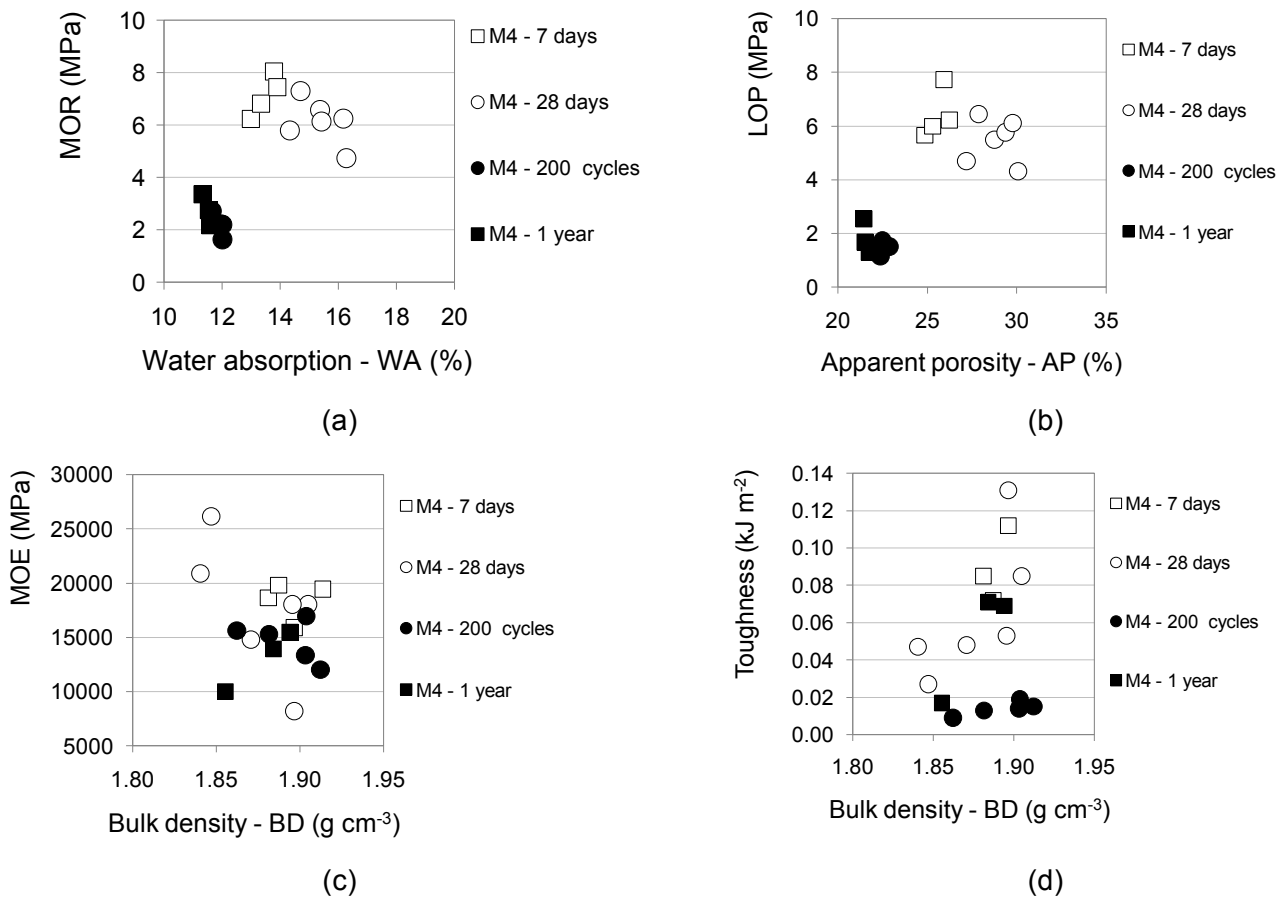
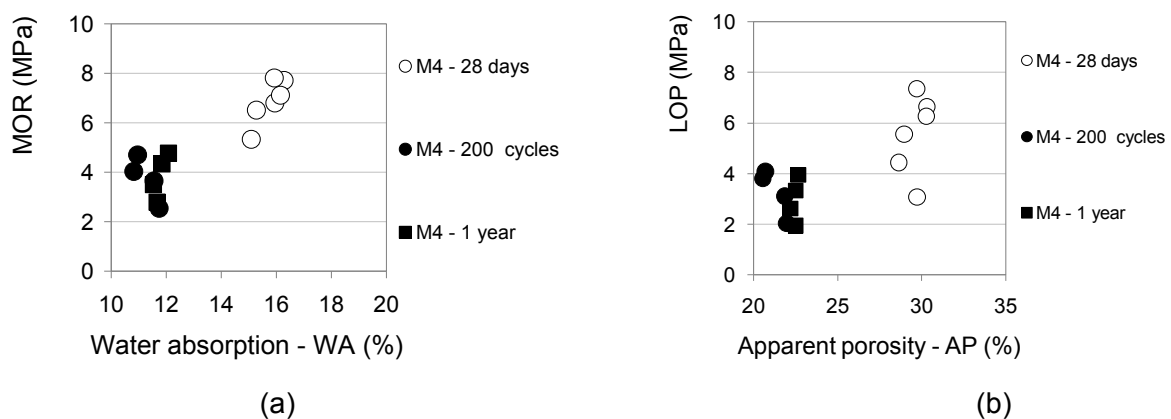
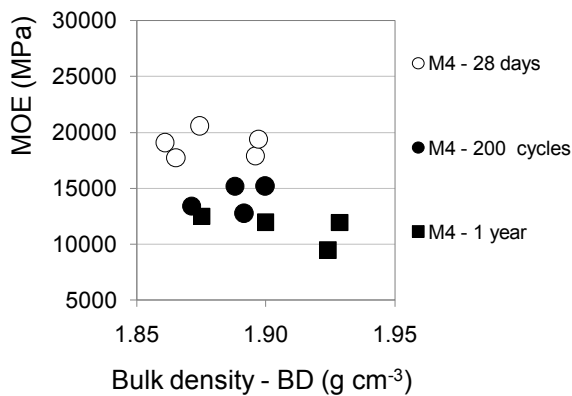


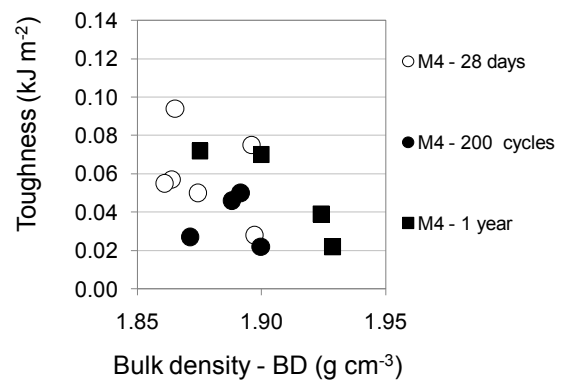
Figure 2 - (a) MOR vs. water absorption (WA); (b) Limit of proportionality (LOP) vs. apparent porosity (AP); (c) MOE vs. bulk density (BD); and (d) toughness vs. bulk density (BD) of cement based composites (at different ages) with the mix of particle sizes (M4) without chemical treatment.

As depicted in the Figures 2 and 3, the chemical treatment did not significantly influence the physical properties at early ages.





(c)



(d)

Figure 3 - (a) MOR vs. water absorption (WA); (b) Limit of proportionality (LOP) vs. apparent porosity (AP); (c) MOE vs. bulk density (BD); and (d) toughness vs. bulk density (BD) of cement based composites (at different ages) with the chemically treated mix of particle sizes (M4).

As depicted in the Figure 3 and Table 1, the chemical treatment contributed to a lower reduction of MOR, LOP and MOE of the composites but did not present an important influence on the toughness. It is the consequence of improved adherence of the treated vegetable particles with the cement matrix, as reported elsewhere (Tonoli et al., 2009; Tonoli et al. 2010). The chemical treatments clean the fibre surface of non-carbohydrates constituents of the peanut husks, such as vegetable extractives (surfactant-type molecules) and lignins (Belgacem et al., 1995). It is expected that the chemical reagents used here react mainly with extractives and lignin, breaking unsaturated bonds and producing carbonyl and carboxyl end structures and thereby increasing the hydrophilic character of particles. In the treated particles, the number of reactive OH groups in the particle surface is probably higher than that exhibited by the surface of untreated counterparts.

Table 1 represents the results of the physical-mechanical properties of the peanut husks particleboards. The statistical analysis indicates the chemical treatment of the peanut husk particles improved the performance of the particleboards in relation to those with the untreated peanut husk particles.

According to statistical analysis, physical properties of accelerated aged and naturally aged composites appear to be similar. Regarding mechanical behaviour, MOR of composites with treated particles was significantly higher ($p < 0.05$) than in composites with untreated particles.

Table 4 - Physical-mechanical properties. Experimental values at different ages.

	Untreated			Treated		
	28 days	365 days	200 cycles	28 days	365 days	200 cycles
Modulus of rupture (MPa)	6.58 aA	2.76 cB	2.19 eB	7.26 bA	3.73 dB	3.62 fB
Modulus of elasticity (GPa)	17.418 aA	13.126 bB	14.652 bB	18.726 aA	11.602 bB	14.144 bB
Apparent porosity (%)	28.84 aA	21.55 bB	22.48 bB	29.6 aA	21.36 bB	22.46 bB
Density (g/cm³)	1.876 aA	1.866 bA	1.893 bA	1.876 aA	1.907 bA	1.883 bA
Water absorption (%)	15.38 aA	11.55 bB	11.88 bB	15.78 aA	11.78 bB	11.35 bB

Small letter – comparison between treated and untreated fibres

Capital letter – comparison between ages

Standard specification (NCR 193:1992) of particleboards for use in building and houses such as asbestos boards requires MOR higher than 7 MPa and MOE higher than 13 GPa. These properties are achieved by the plates with peanut husks in the early ages (28 days). The behaviour with respect to the MOR could be

improved, possibly with additional reinforcing fibres such as cellulose pulp. Another possibility is the use of panels of peanut shells in cement matrix in less aggressive environments, such as those without the water contact.

CONCLUSIONS

Peanut husk, an agroindustrial lignocellulose residue produced in large amounts in Argentina and Brazil is a promising material for the manufacturing of cement based boards.

In the laboratory it was possible to produce panels made of peanut husk particles. The resulting boards present attractive aesthetical properties, due to their surface texture on one of the surfaces.

Based on the conducted tests, the panels made of peanut particles and cement with density around 1.9 g/cm^3 presented sufficient mechanical properties for use in civil and agricultural constructions.

Modulus of rupture, modulus of elasticity, limit of proportionality and toughness were improved in the cement based composites elaborated with different sizes of particles. In this formulation, thin particles with different sizes are better distributed in the composite, allowing the achievement of good compaction in the composite and reducing the presence of air voids. Increasing particles and fibres of peanut husks contents do not improve the physical and mechanical properties of the composites. However, the board surface appears more attractive with increasing the size and content of the particles of peanut husks. Improvements in the mechanical properties of the composites are achieved with Sodium Silicate and Aluminium Sulphate chemical treatments of the husk particles, conducted prior to the addition of the husks to the cement matrices.

Boards with 5% of cut husks and a mix of particles retained on #50 and #16 sieves resulted in sufficiently interesting properties for us to continue experiments with peanut wastes in cement composites. Successful new applications, different from that of asbestos boards, could be amongst others: flooring and prefabricated elements for permanent shuttering of concrete walls and floors.

Another alternative to use the panels whether or not coated with waterproofing product should be in environments with lower exposure to moisture.

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