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Short communication

Composites based on sintering rice husk–waste tire rubber mixtures

D. García [a], J. López [a], R. Balart [a], R.A. Ruseckaite [b], P.M. Stefani [b,*]

[a] Department of Mechanical and Materials Engineering, Higher Polytechnic School of Alcoy – Polytechnic University of Valencia, Paseo del Viaducto, 1, 03801 Alcoy, Spain

[b] Research Institute of Material Science and Technology, (INTEMA) National Research Council (CONICET), Engineering Faculty, Mar del Plata University, Juan B. Justo 4302 (B7608FDQ) Mar del Plata, Argentina

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Abstract

Waste tire rubber and rice husk with different average size particles were used as raw materials for obtaining plates by sintering technique. The effects of several parameters on the sintering process were evaluated, including amount and average size of rice husk particles. The best mechanical properties were obtained by sintering rice husk particles and tire scrap of similar average size. The strategy proposed in this paper can be considered as an alternative to revalorize this kind of waste products.

1. Introduction

Agricultural by products are emerging as new and inexpensive materials with commercial viability and environmental acceptability [1]. Among this kind of materials, lignocellulosic fillers are considered as attractive candidates to be used as fillers of thermoplastic polymers [2–7]. In this way, it is possible to obtain composite materials with properties quite similar to the already known synthetic-filler-reinforced plastics [8]. In addition, natural fillers have low cost, low density, enhanced energy recovery, biodegradability and recyclability [1]. However, the high sensitivity to moisture and low compatibility with hydrophobic samples make their use still limited.

One of the agricultural residues which can be potentially used as fillers of polymeric materials is rice husk. The world production of rice husk was about 122 million tons during the period 2003/2004, which is mainly used for incineration with energy recovery [9] and the production of ashes which are normally applied as filler of polymeric matrices [10–12] or as additive for concrete [13]. Other less explored route to revalorize this residue is a replacement of wood in particle boards [14], without any previous treatment.

On the other hand, tire accumulation is a growing worldwide problem because it is recalcitrant to the environment. Every year about 800 millions of tires are rejected and this amount is increasing 2% each year [15]. Different strategies have been proposed to eliminate rubber wastes including incineration with energy recovery or grinding it to be used as modifiers for different materials [16–19]. Sintering of rubber is another possibility of recycling tires. This process requires only the application of heat and pressure to achieve cross-linked rubber with good mechanical properties [20,21]. Morin and Richard [21], reported the effect of processing conditions (temperature, pressure, and time) on mechanical properties of the sintered rubber, without any further additive. The authors claim that consolidation of rubber particle is associated to rearrangement of the sulfur–sulfur bonds which is achieved by the application of high temperature and pressure. The obtained material may be used in many applications in the form of sheets, for instance as a tile pavement.

In this context, the objective of this work is to analyze the use of rice husk as reinforcing material for recovery tire rubber powder using the sintering method. The mechanical properties of materials based on waste rubber and rice husk

* Corresponding author. Tel./fax: +54 223 4816600.
E-mail address: pmstefan@fi.mdp.edu.ar (P.M. Stefani).

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particles of different average size particles were used for evaluating the efficiency of the sintering method. It is anticipated that the use of both residues in the production of new materials could reduce the environmental problems associated to their accumulation.

2. Experimental

2.1. Materials

The materials used for this study were:

(i) Rice husk (Don Juan variety) used in the present work was collected from industries of "Entre Ríos" region (Argentina). This rice husk was crushed using an analytic mill (IKA 50, Ika-Werke GmbH & Co. KG, Staufen, Germany). In this work, three different average sizes of rice husk particles were used:

- Fraction A: 0.75 mm
- Fraction B: 0.37 mm
- Fraction C: lower than 0.1 mm

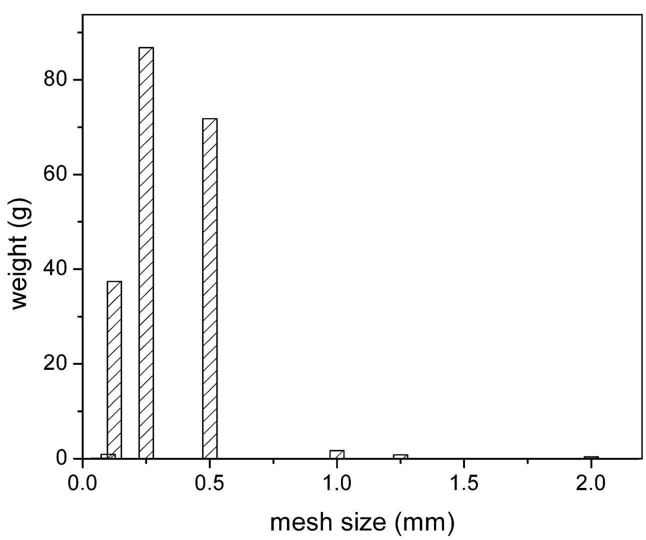

Fig. 1. Size distribution of tire waste.

(ii) Recycled rubber was obtained from tires without steel or other type of fibers and was provided by Insaturbo SA, (Alicante, Spain). Fig. 1 represents the size distribution of the rubber powder used as matrix (original sample amount 200 g). The maximum of the distribution corresponds to 0.3 mm.

2.2. Processing method

Plates of $22 \times 20 \times 0.4$ cm were prepared by following the same procedure reported elsewhere [21], adding 0–25 wt% of rice husk (RH) to the sintering of rubber.

The sintering process was carried out in a hot plate press with pressure and temperature control. The scheme of this process is shown in Fig. 2. The sintering was performed at a constant pressure of 2 MPa and the heating cycle was selected by taking into account the relative thermal stability of the components. Fig. 3 represents the TG curves for rubber powder and rice husk. It can be observed that the rice husk begins to degrade at about 195 °C, well before the scrap tire. Then a maximum temperature of 180 °C was selected for the processing of composites in order to avoid the thermal decomposition of the filler. As a consequence, the following heating cycle was proposed: (1) heating from room temperature up to 180 °C at 5 °C/min and (2) holding at 180 °C for 60 min.

2.3. Testing methods

Specimens for tensile tests were obtained from the moulded sheets, according to international standard UNE-EN 527-1. Tensile tests were carried out on a universal tensile test machine IBERTEST ELIB 30 (S.A.E. Ibertest, Madrid, Spain). A 50 mm/min cross-head speed was used. These tests were carried out for the determination of the elongation at break, the highest load that the specimen can support, and the tangent module starting from the tensile graph. All specimens were tested at room temperature.

Scanning electronic microscopy (SEM) microphotographs of the samples were obtained by using a JEOL 6300 (Jeol USA Inc., Peabody, USA).

Thermogravimetric analysis (TGA) was carried out using a Mettler-Toledo 851e-TGA-SDTA system (Mettler-Toledo Inc., Schwerzenbach, Switzerland) coupled to a STAR-E software. Tests were performed in dynamic mode from room temperature to 750 °C, at a heating rate of 5 °C/min and under nitrogen atmosphere in order to prevent thermo-oxidative reactions (20 ml/min). The sample weight in all tests was approximately 10 mg.

Optical Microscopy (OM) was carried out by using a LEICA MZAPO (Leica Microsystem Ltd., Heerbrugg, Switzerland).

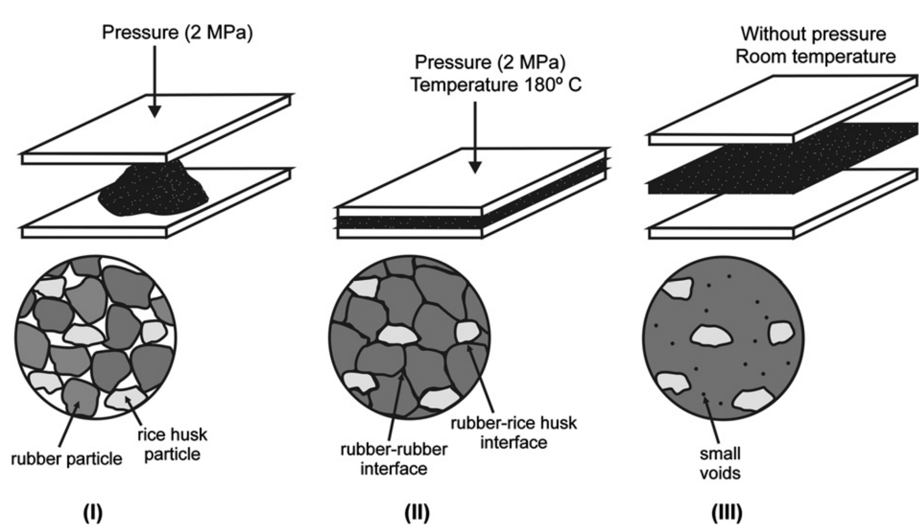

Fig. 2. Scheme of the sintering process.

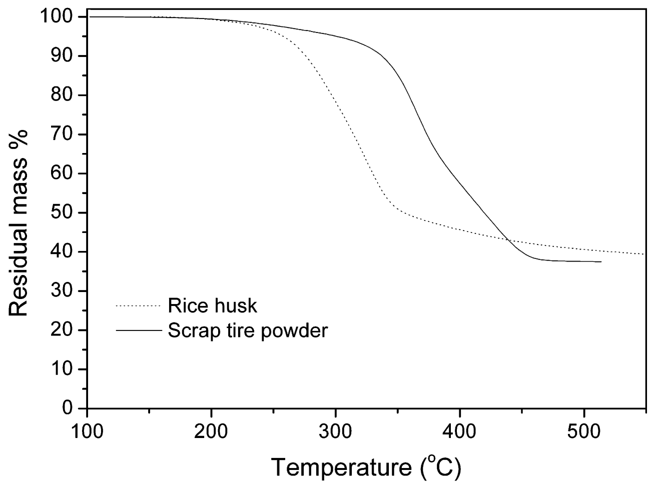

Fig. 3. Dynamic thermogravimetric curve of rice husk and tire waste at 5 °C/min in N_2 atmosphere.

Table 1
Modulus, ultimate tensile strength and strain at break of composites for different percentages and size of rice husk particles

Rice Husk wt%	Rice Husk Fraction	Modulus (MPa)	Ultimate tensile strength (MPa)	Strain at break %
0	–	3.92 ± 0.12	3.01 ± 0.20	144 ± 4
5	A	3.59 ± 0.18	1.85 ± 0.25	89 ± 12
5	B	3.97 ± 0.18	2.16 ± 0.26	105 ± 10
5	C	3.42 ± 0.18	0.79 ± 0.23	40 ± 12
10	A	3.86 ± 0.21	1.22 ± 0.27	51 ± 16
10	B	6.26 ± 0.06	1.90 ± 0.17	86 ± 5
10	C	5.05 ± 0.21	1.05 ± 0.30	52 ± 12
15	A	9.23 ± 0.12	1.24 ± 0.12	47 ± 11
15	B	10.72 ± 0.05	1.30 ± 0.10	49 ± 8
15	C	10.11 ± 0.16	1.09 ± 0.18	41 ± 15
20	A	10.76 ± 0.10	0.74 ± 0.10	19 ± 10
20	B	11.24 ± 0.11	1.13 ± 0.14	39 ± 8
20	C	10.73 ± 0.13	0.86 ± 0.18	30 ± 9
25	A	6.48 ± 0.11	0.59 ± 0.18	12 ± 4
25	B	12.08 ± 0.06	0.96 ± 0.09	23 ± 13
25	C	10.59 ± 0.13	0.78 ± 0.22	15 ± 5

3. Results and discussion

Table 1 summarizes the average mechanical properties of rice husk–rubber composites (RH–R) (modulus, ultimate tensile strength and strain at break) for different amounts (0–25 wt%) and average size fractions (A, B, C) of filler. It is noteworthy that mechanical behaviour highly depends on the amount and size of rice husk particles used as reinforcing agent. As a general trend, an increment in RH content enhances the elastic modulus, decreases the tensile strength and the strain at break, whatever the particle size used. In fact, the effect of the filler in the strain at break is inversely proportional to the modulus of the compounds. It is important to remark that filler stiffness and filler/matrix interaction are important parameters in the determination of the mechanical properties of composite materials. At low strains, the higher stiffness of the rice husk produces an increase in the global stiffness of the composite, which is expressed by the increment in module. Contrarily, at high strains, filler/matrix bonds become weak (because of low compatibility between both phases and the absence of compatibilizing agents) and debonding results in interfacial failure, with the concomitant decrease of the strain at break and ultimate strength. The low interfacial bonding was evidenced by SEM microphotographs (Fig. 4). The poor matrix/filler adhesion conducts to debonding at high level of strain and then a detriment in the strain at break of the composite. Similar results are reported for elastomeric matrices reinforced with rigid fillers [22].

It is important to note that the highest modulus is observed for RH particles with an average size similar to that of rubber powder (see Fraction B, Table 1). This behaviour may be related to the mixture process and their effect on the sintering process. Composites with the largest RH particles (Fraction A) heterogeneous materials are obtained, due to the segregation during mixing, as it was observed by optical microscopy (Fig. 5). As a consequence, the obtained materials may present regions where the sintering process is very effective together with regions with high defect content which favour the break mechanism during the tensile test. As the particle size of RH decreases, a more uniform distribution within the matrix is observed.

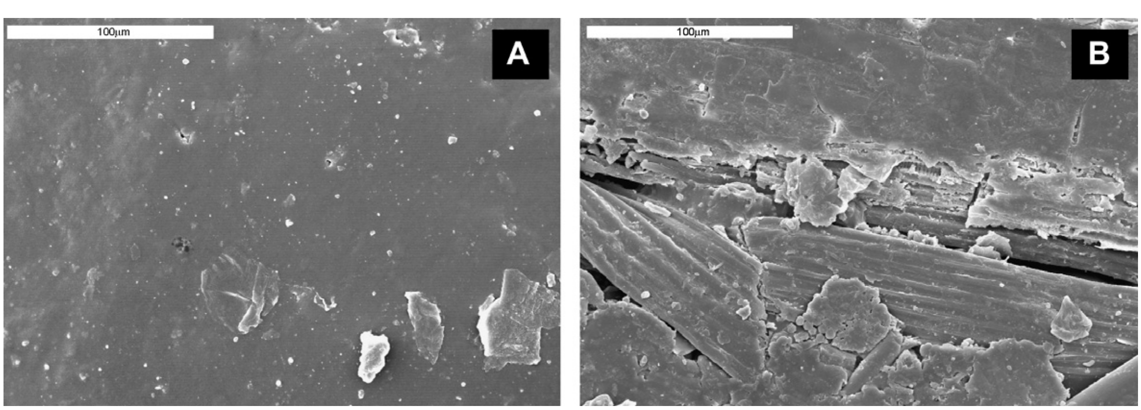

Fig. 4. SEM microphotographs of composites (×500): (A) 0 wt% rice husk and (B) 15% wt% rice husk (Fraction A).

However, the smaller particles (Fraction C) interfere during the sintering process of the rubber. As a result, the effectiveness of the sintering process decreases and the ultimate tensile strength and the strain at break fall down.

These results demonstrate that by controlling the average particle size of the filler RH–R composites with enhanced elastic modulus can be obtained. These composites can be used in applications such as tile pavement where

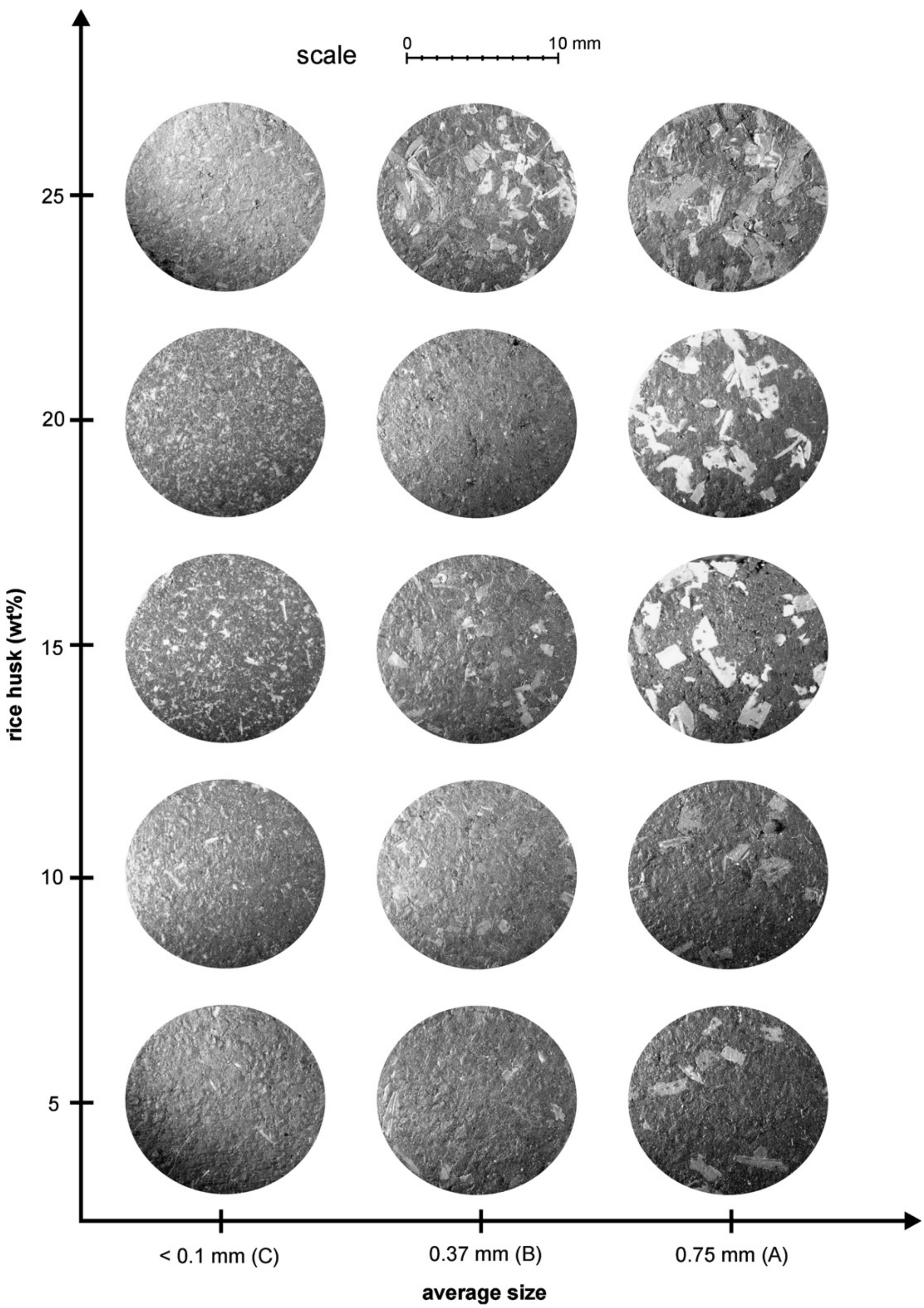

Fig. 5. Surface comparison of composites with different size and amount of rice husk by optical microscopy.

a high rigidity is needed more than a high capacity of deformation.

4. Conclusions

Novel composites based on recovery rice husk and tire milling were successfully obtained by sintering method. Rice husk content and particle size distribution were the most significant factors that affected final mechanical properties of the composite. Particularly, particle size resulted to be the most important parameter to be controlled due to its influence on the consolidation stage of the sintering process. The best results were obtained for filler size in the same range of that of the matrix (about 0.3 mm).

It is important to remark that these new composites combine cheap and highly available raw materials, with quite good final properties. The performance of such materials may be enhanced by improving the adhesion between both co-components. The lack of affinity between natural filler and rubber may be improved by choosing an adequate compatibilizing agent. The use of a low cost compatibilizing agent will be the subject of future communications.

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