http://dx.doi.org/10.1590/0104-1428.2278

Mechanical recycling of tags and labels residues using sugarcane bagasse ash

Guilherme Augusto Mantovani[1], Jean Halison de Oliveira[2], Andressa dos Santos[2], Andrelson Wellington Rinaldi[2], Murilo Pereira Moisés[3], Eduardo Radovanovic[2] and Silvia Luciana Fávaro[1]*

[1]Laboratório de Química de Materiais e Sensores, Departamento de Engenharia Mecânica, Universidade Estadual de Maringá – UEM, Maringá, PR, Brazil
[2]Laboratório de Química de Materiais e Sensores, Departamento de Química, Universidade Estadual de Maringá – UEM, Maringá, PR, Brazil
[3]Universidade Tecnológica Federal do Paraná – UTFPR, Apucarana, PR, Brazil
**slfavaro@hotmail.com*

Abstract

In this study, an alternative method for recycling residues of labels and stickers (parings) containing biaxically oriented polypropylene (BOPP) and polyurethane-based glue was discussed. The recycling of this type of material is complicated, once the separation and the milling processes are difficult to be accomplished, due to the presence of a large amount of glue. In this study, sugarcane bagasse ash was used to enable the milling process of stickers residues. Composites were prepared with post-consumer polypropylene extrusion with different polypropylene/parings ash ratio. These materials were analyzed by tensile, three point flexural, hardness, density, water absorption, Izod impact tests, thermogravimetric analysis, environmental exposure and scanning electron microscopy. Addition of sticker residues/ash to the polypropylene matrix makes the material more rigid and does not affect significantly thermal and degradation properties. Thus, the recycling process proposed in this paper is environmentally and economically viable.

Keywords: *composites, parings, BOPP, recycling, mechanical properties.*

1. Introduction

Labels and stickers play the role of providing information on products, in order to attract the consumer, give information compulsory for the product distribution and show the producer’s brand. Many labels are printed on stickers, made of couche paper or biaxially oriented polypropylene (BOPP) film, once those materials allow a high-quality printing.

When those labels and stickers are produced, many parts of them are cut out; the remaining material is called “parings” by the printing industry. Those adhesive parings are not recycled or reused; they are kept in the factories or are inconsequently thrown away on dumps. The companies that are interested in providing an adequate destination for the parings pay to dispose this residue in landfills. In Brazil, the cost for that disposal is around R$ 70,00 per ton of non-hazardous residue; in European countries, this cost can reach R$ 340,00 (2010)[1].

The amount of labels and stickers discarded is huge, for they are largely used. A small factory, for example, generates approximately 25 tons of residue per month. The mechanical recycling of that material is complex, once the separation and the milling processes are expensive and difficult to be done, due to the big amount of glue that the labels and stickers contain.

Sugarcane bagasse ash is another solid industrial residue inappropriately managed. Brazil is one of the biggest producers of sugar and alcohol from sugarcane; it is one of the oldest and most important economic activity in the country, being Paraná State one of the biggest national producers. The main by-products from the sugar and ethanol industry are: rinsing water, bagasse, leaves, sugarcane tips and ash, vinasse, filter cake and yeast. The bagasse is one of the most attractive by-products, due to its calorific power, making it the main energy source for producing sugar and alcohol. It is also used as energy source for producing electrical power to the national system, by means of cogeneration. After the sugarcane bagasse is burnt in boilers, residual ashes remain. The chemical composition of those ashes reveal the predominant presence of silica (SiO_2). The silicon present in the ashes is absorbed from the soil by the sugarcane roots, as monosilicic acid (H_4SiO_4); after the transpiration, it gets stuck on the outer wall of the epidermal cells, as silica gel[2].

From the sugarcane mass that feeds the milling process are produced approximately 26% of bagasse (containing 50% of moisture) and from this material 0.62% of residual ash is generated[3]. In the harvest 2013/2014, 4 million tons of sugarcane bagasse ash were generated in Brazil. The ashes produced in the process of fuel burning in boilers are among the residues most generated in the country. According to ABNT NBR 10004:2004[4], because of the chemical and physical characteristics, the ashes are classified as a solid residue of rural category, belonging to nature of Class II (not inert).

Nowadays, in Brazil, there is not legislation to deal specifically with sugarcane bagasse ash; it is frequently inappropriately discarded or takes space in landfills, what can generate environmental problems. The concern about sustainable development lead to the necessity of the realization of researches on the use of sugarcane bagasse ashes; some researchers have already reported its use in Portland cements[5] and at the synthesis of porous materials[1], such as microporous zeolites. Commercially, a small amount of ashes is reused at the civil construction industry, as well as fertilizer. The most important effort should be in a way that minimizes the environmental impacts caused by the disposal of that residue in the environment, enlarging the possibilities for its use, for example, at the production of composite materials[6].

Therefore, this work presents an alternative for the reutilization of residues from stickers and labels, by means of mechanical recycling, using sugarcane bagasse ashes for milling the residues, inhibiting the glue action and evaluating the chemical and thermal properties of the composites formed by these residues and post-consumer polypropylene.

2. Materials and Methods

2.1 Materials

The sugarcane bagasse ashes (CBCA) were obtained from a sugarcane and alcohol mill located in the region of Maringá (Paraná State, Brazil). The residues from labels and tags used, named parings in this study, were provided by the company INOVAFLEX – Labels and tags, located in Maringá (Paraná State, Brazil). The polypropylene used at the preparation of composites was acquired from a supplier of recycled material located in the region of Maringá.

2.2 Samples preparation

For using CBCA in the milling process, they were previously calcinated. They stayed in a muffle at 600 °C for 4 hours, so all the organic matter was removed. The ashes resulting from this process are called in this study CBCA600[1].

The parings (resulting residue from the production of labels) were milled in a knife mill, together with CBCA600, at proportion parings/ashes equal to 1:3 (mass/mass). Afterwards, the milled material was sieved to remove the ashes excess, in a sieve with opening size n° 40 (ASTM). The excess ashes were removed by hand, almost 50% mass of milled mixture was retired by sieving.

The milled and sieved residue was mixed with post-consumer polypropylene at different mass proportions: 10% residue with 90% polypropylene (PP10); 30% residue with 70% polypropylene (PP10); 50% residue with 50% polypropylene (PP50). Samples of pure post-consumer polypropylene (PP) were also prepared, as well as of the milled parings with ashes without adding polypropylene (PPR).

The samples were processed in a twin-screw extruder Thermo Sientific MiniLab II HAAKE Rheomex CTW 5, using the temperature of 190 °C for the mold and 60 rpm of rotation. The test specimens for the mechanical trials were injected by means of an injection machine Thermo Sicentific HAAKE MiniJet II, with cannon temperature at 210 °C, mold temperature at 40 °C, injection pressure of 650 bar, injection time of 15 s, holding pressure of 300 bar and holding pressure time of 30 s.

2.3 Characterization

Calorimetric analysis (DSC) from the parings and from the post-consumer polypropylene were obtained. The tests were carried out in SHIMADZU DSC50 equipment, under nitrogen atmosphere (20 mL/min), at a heating rate of 10 °C/min.

The quantity of ashes in each sample was determined by gravimetry. The composites samples were calcinated for the measurement of the quantity of sugarcane bagasse ashes introduced in each of the samples.

The morphology of the composites was analyzed by means of scanning electron microscopy (SEM) in SHIMADZU SS-550 equipment. Images of the test specimens fractured region after the impact tests and of the exposed surface after the environmental exposure were obtained.

The thermogravimetric analyses (TGA) of the composites were carried out in SHIMADZU TGA-50 equipment, operating at a heating rate of 10 °C/min from the room temperature through 700 °C, with 20 mL/min nitrogen flow.

The density of the composites was verified at a pycnometer MICROMERITICS 1305, under helium pressure.

The tensile tests were carried out at an universal testing machine EMIC DL 10000 with load cell of 5000 N, according to the ASTM D638 standard. A number of eight tests were carried out for each sample. The Izod impact tests were carried out in CEAST Resil Impactor Junior equipment, according to the ASTM D256 standard, which establishes the method for the determination of the resistance to the Izod impact for plastic materials. A number of five tests was carried out for each sample.

The environment exposure tests were carried out according to the ASTM D1435 standard, which deals with the procedures for the exposure of plastic materials to the environment for evaluating degradation. Two degradation periods were analyzed, 60 and 120 days. At the end of these periods, the impact of the exposure on the composite materials properties was analyzed, with scanning electron microscopy techniques and mechanical strength trial: yield strength, ultimate tensile strength, elastic modulus, stretching of fracture and Izod impact strength.

3. Results and Discussions

3.1 Characterization of the raw material

The sugarcane ashes have previously been characterized[1], showing that the CBCA is basically constituted of silica grains, with sizes varying between 10 and 400 μm, containing 14% of organic matter, referent to the remaining carbonized sugarcane fibers, and 2% of moisture. The thermal treatment to which CBCA600 ashes were subjected eliminates all the remaining organic matter.

The x-ray difractograms obtained for the parings and for the post-consumer polypropylene used in this study are presented in Figure 1. The post-consumer polypropylene presented signals at the angles 2Θ of 14°, 16.8°, 18.5° and 21.5°, typical of the isotactic polypropylene. The difractogram of the parings presented signals at the angles 14°, 17° and 18.8°, such as polypropylene. However, two other peaks at 12.3° and 25.7° could also be observed, indicating the presence of syndiotactic polypropylene in the parings. The absence of a signal in the difractogram for the parings at 21°, when compared to polypropylene, may occur due to the material processing temperature and to the biaxial orientation process[7-9]. The polypropylene tacticity is an important parameter to be analyzed, for it influences directly on the physical and mechanical properties of material.

The DSC results obtained for parings and post-consumer polypropylene used at the composite production are shown in Figure 2. The parings showed an endothermic signal close to 165 °C, characteristic of the polypropylene fusion. The post-consumer polypropylene showed two endothermic peaks, one of them close to 165 °C, characteristic of the polypropylene, and the other, close to 130 °C, characteristic of the fusion of high-density polyethylene. In general post-consumer polypropylene contain a certain amount of polyethylene as a contaminant. It occurs due to the process of density separation used at the recycling places, because polypropylene and polyethylene have very similar densities. The amount of polypropylene present in the mixture was determined, considering the fact that the pure polypropylene specific heat of fusion is 60 J/g[10] and considering the polypropylene area of fusion peak (56.3 J/g); then the value of 94% of polypropylene could be found.

3.2 Characterization of the composites

The mechanical recycling consists of separation, milling, washing, drying, agglutination and extrusion phases. The washing and drying phases may be skipped, depending on the residue condition; for industrial residues, for example, they are not carried out. However, the milling process is primordial to the process. As the labels and tags residues contain glue in their composition, the milling process becomes very arduous. Prior to this study, different milling techniques were tested, and the process using sugarcane bagasse ashes was the most effective. The ashes act inhibiting the glue, preventing it from sticking to the mill knifes.

The results of the gravimetric analysis of the composites and of the post consumed PP are presented in Table 1. The non-volatile fraction of the samples is corresponding to the CBCA600 used for milling the parings, which is not eliminated in the sieving process, i.e., the ashes that stick to the parings surface. This way, it is possible to state, from the results of the gravimetry tests, that the CBCA600 mass is approximately 50% of the milled and sieved residue.

The thermogravimetric analysis (TGA) of the samples is presented in Figure 3; the residual mass indicates the quantity of CBCA600. An increase on the thermal stability of the composites in relation of the pure matrix can be observed, by means of the temperature in which maximum mass loss rate occurs (dm/dT). The value presented by pure PP is 415 °C, while, for the composites, this temperature is around 450 °C to all the compositions. Another important aspect to be observed in Figure 3 is the quantity of remaining residue present in each sample, due to an increase of inorganic material (ashes) to the sample. With the addition of the lower percentage of parings and ashes, a low percentage of remaining CBCA600 for the composite PP10 could be observed; this quantity gradually increases when composites

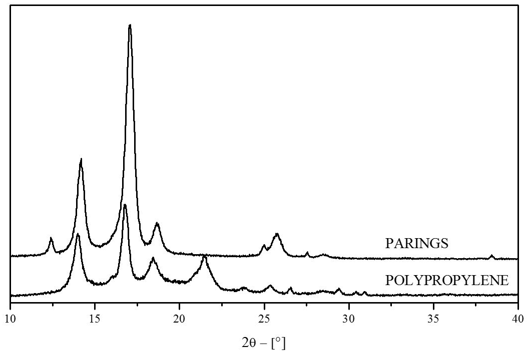

Figure 1. X-rays difractogram (DRX) of the parings and of the polypropylene.

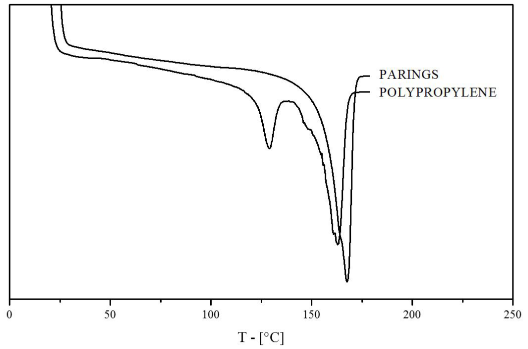

Figure 2. DSC thermograms of the parings and of the polypropylene.

Table 1. Average composition of the samples mass.

Sample	Polypropylene (%)	Parings (%)	Ashes (%)
PP	100	--	--
PP10	90	6	4
PP30	70	16	14
PP50	50	24	26
PPR	--	47	53

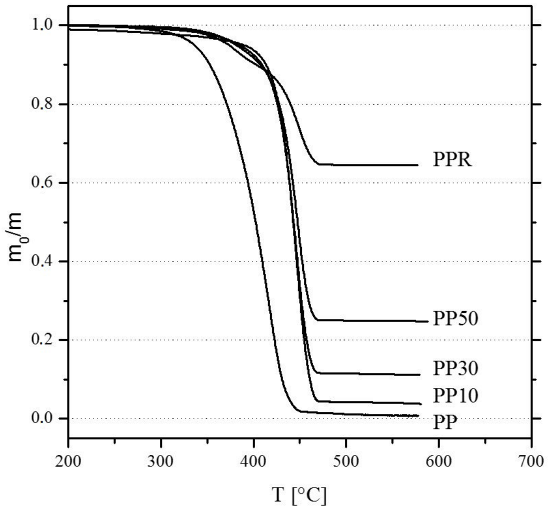

Figure 3. TGA curves of the composites and of the PP.

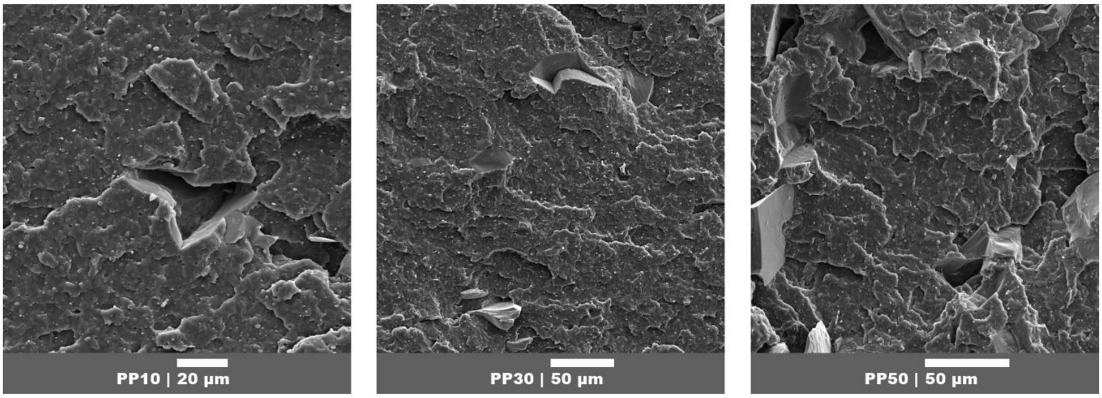

Figure 4. Micrograph of the composites fracture surface.

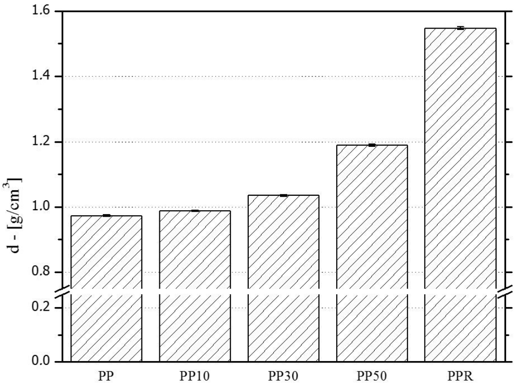

Figure 5. Density of the composites and of the PP.

PP30 and PP50 are considered. In the sample prepared only with the mixture of parings and ashes (PPR), the quantity of CBCA600 reaches 60% (m/m). Those results are equivalent to the ones observed at the gravimetric analysis and prove a homogeneous reinforcement distribution in the matrix, once for the gravimetric analysis, some grams of material are used, and for the thermogravimetry, some milligrams are used.

In Figure 4, micrographs of the PP10, PP30 and PP50 composites are presented. The silica distribution in the composites is homogeneous; there were not any problems for dispersing the ashes in the material during the process, and they do not tend to agglutinate. It is also possible to observe that, during the milling and extrusion processes, silica was broken into smaller grains.

In the fracture area it is possible to observe that the places where the silica unglued from the matrix during the impact test are flat surfaces, indicating that the silica does not stick very well to the polymeric matrix. It is known that low interfacial resistances are generally identified on fracture surfaces due to the presence of empty spaces, associated to the particles yank, originated from the fissures spreading on the interfacial area. The fractographic analysis, in its turn, tends to associate high interfacial resistances to the presence of polymeric films on the particles[11].

The graph presented in Figure 5 presents the samples final density. The increase of the ashes amount in the material causes a proportional density increase in the composites, if compared to pure PP. The ashes density is close to 2.3 g/cm^3, while the polypropylene density is around 0.9 g/cm^3. The highest density presented by the composite materials, enables its use for specific applications, in which materials with densities higher to the ones of the polypropylene and polyethylene are demanded.

In Figures 6A to 6D are presented the results obtained from the tensile strength test for the composite materials and for the pure PP. In Figure 6A, the values obtained from the yield strength in the trials can be observed. It can be noticed that there is a decrease on the yield strength with the increase of the silica amount; this decrease is remarkable in the sample PPR. In Figure 6B are shown the values for the ultimate tensile strength. A decrease in these values occurs with ashes addition. Nevertheless, for the composites with up to 50% of residue (around 25% of ashes), the tensile strength decrease is very low. The introduction of hard inorganic particulates in polymeric matrices is usually followed by a reduction in the polymer mechanical strength, leading to the production of composites mechanically poorer than the pure polymer. This fact is frequently observed, for example, when particulates, such as slate, calcium carbonate and talc are incorporated in polymers like polypropylene[12-15]. The fact that the ash addition has not influenced the polymer resistance so drastically highlights its capacity of not deteriorating the properties in relation to the pure polymer. That deterioration of properties would be linked to the action of the particulate matter as a generator of defects on the composite material[15]. Only for the sample PPR that decrease is highly remarkable, with a decrease of around 80% in its value. Once the silica adhesion to the polypropylene matrix is low, as the morphological analysis has shown, this result was already expected. As the matrix does not transfer adequately the effort to the reinforcement phase, the area of the specimen test section that supports the effort is smaller; consequently, the tension that the material can tolerate is also lower.

Figure 6C presents the elastic module values of the composites. These values increase with the addition of

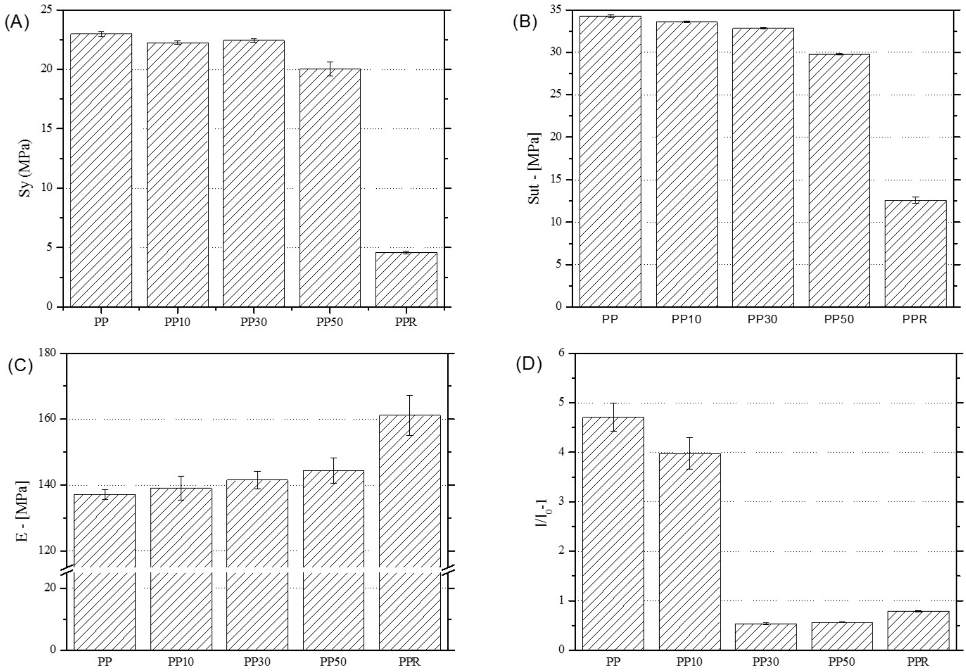

Figure 6. Mechanical behavior of the composites and PP samples where (A) represents the yield strength; (B) ultimate tensile strength; (C) elastic modulus; and (D) rupture strength.

ashes to the composites. That increase is more effective for the sample PPR. As a consequence of this result, there is a decrease on the rupture stretching, as presented on the results of Figure 6D. The stretching values decrease with the increase of the silica quantity; a more drastic decrease was observed for the composites prepared without post-consumer PP addition, only with parings and ash.

From the results of the impact strength tests, carried out with the Izod method, presented in Figure 7, it could be observed that the addition of ashes subtly decreased the material impact strength. As observed on the tensile strength tests, the composites PP10, PP30, PP50 and PPR are harder than the pure matrix. Harder materials absorb less impact energy during the failure. The impact strength decreases with the increase of ashes concentration. The addition of hard loads to the ductile matrix tends to weaken the material. The load acts as tensile concentration, restricting the mobility of the matrix, avoiding plastic deformation. Another aspect to be considered is the lack of adhesion between the matrix and the reinforcement. This result is in accordance with other studies that use inorganic loads as reinforcement in polypropylene matrix[16-18]. That small difference in the impact strength makes it possible to be used instead of pure polypropylene in applications in which the maximization of this characteristic is not preponderant.

3.3 Environment exposure test

The term polymers degradation is associated to any destructive reaction that could be caused by chemical, physical, mechanical or biological agents, causing an irreversible change in the properties of polymeric materials. In many cases, the degradation reactions may be desirable, for example, for non-recyclable polymeric waste. In the composites, the degradation may occur in the matrix, in the reinforcement or in the system as a whole. In the sun light, this degradation considerably increases, because the oxidation rate of polymers is accelerated, and this effect is increased by the presence of atmospheric pollutants. The temperature and the moisture can also be mentioned as factors that contribute to the degradation process of polymers[19]. In order to evaluate the degradation process of the composites, the material was subjected to trials of natural aging, being exposed to environmental conditions very

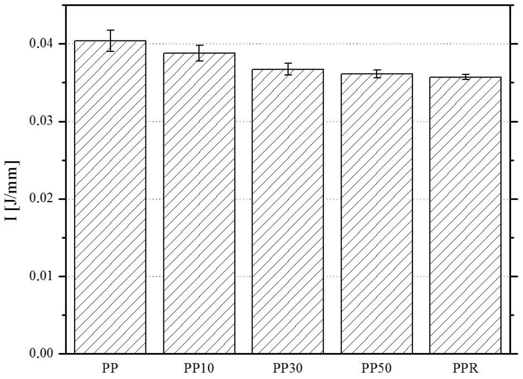

Figure 7. Impact strength of the samples.

similar to its real conditions of utilization. Mechanical and scanning electron microscopy analyses were used to verify the occurrence of degradation of the material, as well as the reinforcement influence on this process.

The surfaces of the samples exposed to degradation were evaluated by scanning electron microscopy, Figure 8. In all the samples, it is possible to notice the appearance of fissures with the degradation and the increase of cracking with longer exposure time, except for the sample PPR, which is already very porous. The cracking results from a phenomenon noticed at the photodegradation of the polypropylene, known as chemi-crystallization. The molecules of the amorph regions in the polypropylene are more susceptible to photodegradation, which causes molecules to split. The resulting smaller segments are more mobile and can rearrange in crystalline structures, generally on already existing crystals. This process increases the polypropylene crystallity and causes the superficial spontaneous formation of fissures[20-22].

In Figures 9A to 9D are presented the results of the tensile tests after the exposure of the material to degradation, compared to the tests carried out in the material as soon as it was processed. It is possible to notice a tendency of decreasing resistance and maximum stretching; this decrease is mainly due to the photodegradation of the composites polymeric matrix, because the reinforcement phase silica is not subject to property loss in the short time evaluated. Rabello and White[20] also observed the polypropylene mechanical strength decrease due to the effect of the exposure to the ultraviolet radiation: chains splitting, fissuring and increase of crystallity.

The elastic modulus presented an increase at almost all the samples, but did not present a direct relation between its value and the composites composition.

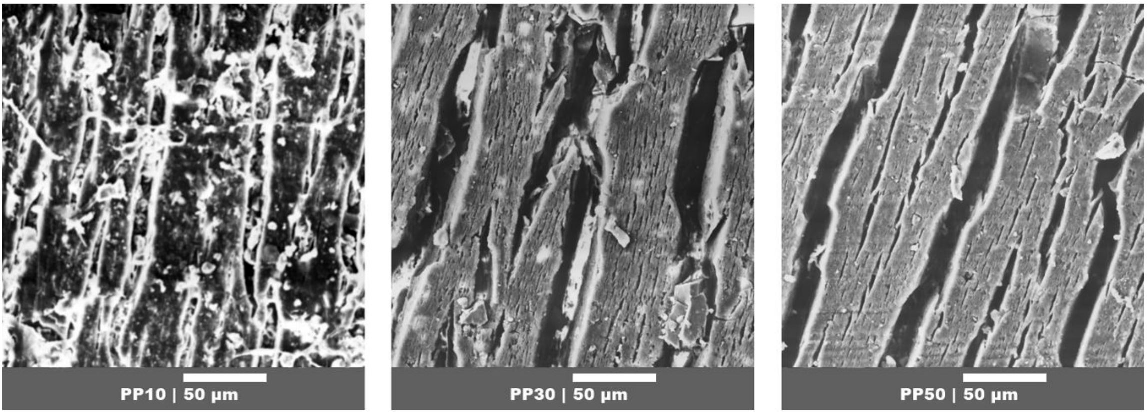

Figure 8. Micrograph of the composites surface after 120 days environmental exposure.

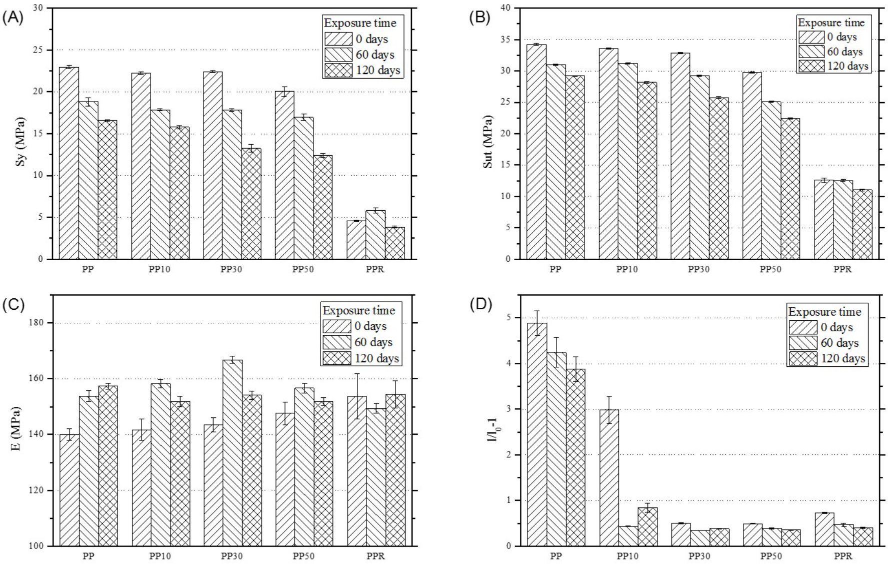

Figure 9. Mechanical behavior of the samples with the degradation where (A) represents the yield strength; (B) ultimate tensile strength; (C) elastic modulus; and (D) rupture strength.

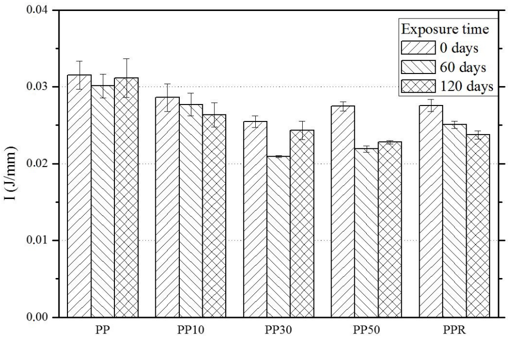

Figure 10. Behavior of the impact strength with the degradation.

The changes on the composites properties were not very different from the changes occurred at the samples of pure polypropylene. This way, the incorporation of parings/ashes to polypropylene allowed the production of materials with mechanical and composites thermal behavior properties that are not very different form the ones of the pure polypropylene, besides producing a low-cost material, which also contributes to the environment preservation as it favors the reuse of residues.

Figure 10 presents the results of the impact strength after the exposure the material to environmental conditions compared to the results of the tests carried out before that. All the samples presented a small reduction on the impact strength when exposed to the weather conditions. Nevertheless, it is not possible to state that the environmental exposure time is directly related to the decrease of the impact strength, because for the samples PP, PP30 and PP50 the lowest impact strength was obtained after 60 days of exposure, while for the samples PP10 and PPR the lowest values occurred after 120 days of exposure to the environment.

4. Conclusions

The use of sugarcane bagasse ash to inhibit the glue action and enable the milling of parings is promising. The mechanical characterization of the composites indicated that the addition of the parings residues milled with ashes to the polypropylene produces a harder material than the pure polypropylene. The composites also presented high thermal stability. The mechanical properties of the composites and of the pure PP behave in a very similar way, and the same fissuring phenomenon is noticed in all the samples.

One of the biggest advantages of the composites preparation is the flexibility in relation to the composition polypropylene/parings/ashes, offering the possibility to produce materials with different final mechanical properties, related to their composition.

The recycling method proposed stands a high chance to be successful, because of the low cost of the equipment used and because of the innovative and environment-friendly character. It potentially represents job creation at associations and cooperatives built to supply the recyclable material industry.

The composites can be applied to the production of different materials, including domestic-use tools, such as buckets, bowls, clothespins, and also products for civil construction, for manufacturing tiles, floorings, hoses and pipes. So those materials can be developed, more research must be carried out to evaluate the materials degradation in longer periods of environmental exposure.

5. References

1. Moisés, M. P., Silva, C. T. P., Meneguin, J. G., Girotto, E. M., & Radovanovic, E. (2013). Synthesis of zeolite NaA from surgarcane bagasse ash. *Materials Letters*, *108*(1), 243-246. http://dx.doi.org/10.1016/j.matlet.2013.06.086.
2. Mano, E. B., Pacheco, E. B. A. V., & Bonelli, C. M. C. (2010). *Meio ambiente, poluição e reciclagem*. São Paulo: Blucher.
3. Nunes, I. H. S., Vanderlei, R. D., Secchi, M., & Abe, M. A. P. (2008). Estudo das caracteristicas fisicas e químicas da cinza do bagaço de cana-de-açucar para uso na construção. *Revista Tecnológica*, *17*, 39-48. http://dx.doi.org/10.4025/revtecnol.v17i1.
4. Associação Brasileira de Normas Técnicas – ABNT. (2004). *ABNT NBR 10004: resíduos sólidos: classificação*. Rio de Janeiro: ABNT.
5. Paula, M. O., Tinôco, I. F. F., Rodrigues, C. S., Silva, E. N., & Souza, C. F. (2009). Potencial da cinza do bagaço da cana-de-açúcar como material de substituição parcial de cimento Portland. *Revista Brasileira de Engenharia Agrícola e Ambiental*, *13*(3), 353-357. http://dx.doi.org/10.1590/S1415-43662009000300019.
6. Meier, M. (2014). Sustainable polymers: reduced environmental impact, renewable raw materials and catalysis. *Green Chemistry*, *16*(4), 1672. http://dx.doi.org/10.1039/c4gc90006e.
7. Türkçü, H. N. (2004). *Investigation of the crystallinity and orientation of polypropylene with respect to temperature changes using FT-IF, XRD and raman techniques* (Master's dissertation). Bilkent University, Ankara.
8. Tartaglione, G., Tabuani, D., Camino, G., & Moisio, M. (2008). PP and PBT composites filled with sepiolite: morphology and thermal behaviour. *Composites Science and Technology*, *68*(2), 451-460. http://dx.doi.org/10.1016/j.compscitech.2007.06.023.
9. Erp, T. B. V., Balzano, L. M., & Peters, G. W. (2012). Oriented gamma phase in isotatic polypropylene homopolymer. *ACS Macro Letters*, *1*(5), 618-622. http://dx.doi.org/10.1021/mz3000978.
10. Canevarolo, S. V. (2003). *Técnicas de caracterização de polímeros*. São Paulo: Artliber.
11. Sinien, L., Lin, Y., Xiaoguang, Z., & Zongneng, Q. (1992). Microdamage and interfacial adhesion in glass bead-filled high-density polyethylene. *Journal of Materials Science*, *27*(17), 4633-4638. http://dx.doi.org/10.1007/BF01165998.
12. Premalal, H. G. B., Ismail, H., & Baharin, A. (2002). Comparison of the mechanical properties of rice husk powder filled polypropylene composites with talc filled polypropylene composites. *Polymer Testing*, *21*(7), 833-839. http://dx.doi.org/10.1016/S0142-9418(02)00018-1.
13. Demjén, Z., Pukánszky, B., & Nagy, J. (1998). Evaluation of interfacial interaction in polypropylene/surface treated CaCO$_3$ composites. *Composites. Part A, Applied Science and Manufacturing*, *29*(3), 323-329. http://dx.doi.org/10.1016/S1359-835X(97)00032-8.
14. Mareri, P., Bastide, S., Binda, N., & Crespy, A. (1998). Mechanical behaviour of polypropylene composites containing fine mineral

filler: Effect of filler surface treatment. *Composites Science and Technology*, *58*(5), 747-752. http://dx.doi.org/10.1016/S0266-3538(97)00156-5.

15. Carvalho, G. M. X., Mansur, H. S., Vasconcelos, W. L., & Oréfice, R. L. (2007). Obtenção de compósitos de resíduos de ardósia e polipropileno. *Polímeros: Ciência e Tecnologia*, *17*(2), 98-103. http://dx.doi.org/10.1590/S0104-14282007000200008.
16. Asuke, F., Aigbodion, V. S., Abdulwahab, M., Fayomi, O. S. I., Popoola, A. P. I., Nwoyi, C. I., & Garba, B. (2012). Effects of bone particle on the properties and microstructure os polypropylene/bone ash particulate composites. *Results in Physics*, *2*(1), 135-141. http://dx.doi.org/10.1016/j.rinp.2012.09.001.
17. Fuad, M. Y. A., Ismail, Z., Ishak, Z. A. M., & Omar, A. K. M. (1995). Application of rice husk ash as fillers in polypropylene: effect of titanate, zirconate and silane coupling agents. *European Polymer Journal*, *31*(9), 885-893. http://dx.doi.org/10.1016/0014-3057(95)00041-0.
18. Ramos, S. M. L. S., Carvalho, L. H., Spieth, E., & Rivadula, R. S. (1993). Efeitos da estabilização do Polipropileno nas propriedades térmicas, mecânicas e termo-mecânicas de compósitos de Polipropileno/Atapulgita. *Polímeros: Ciência e Tecnologia*, *3*(4), 26-31.
19. Thwe, M. M., & Liao, K. (2003). Durability of bamboo-glass fiber reinforced polymer matrix hybrid composites. *Composites Science and Technology*, *63*(3-4), 375-387. http://dx.doi.org/10.1016/S0266-3538(02)00225-7.
20. Rabello, M. S., & White, J. R. (1997). Fotodegradação do polipropileno: um processo essencialmente heterogêneo. *Polímeros: Ciência e Tecnologia*, *7*(2), 47-56.
21. Allen, N. S., Edge, M., Corrales, T., Childs, A., Liauw, C. M., Catalina, F., Peinado, C., Minihan, A., & Aldcroft, D. (1998). Ageing and stabilisation of filled polymers: an overview. *Polymer Degradation & Stability*, *61*(2), 183-199. http://dx.doi.org/10.1016/S0141-3910(97)00114-6.
22. Paoli, M. A. (2009). *Degradação e estabilização de polímeros*. São Paulo: Artliber.

Received: Nov. 09, 2015
Revised: Apr. 01, 2016
Accepted: Apr. 28, 2016