

COMPOSITES FORMED BY RECYCLED POLYPROPYLENE AND WET-BLUE LEATHER WASTE: A SUSTAINABLE PRACTICE

COMPOSIÇÕES FORMADAS POR POLIPROPILENO RECICLADO E RESÍDUOS DE COURO WET-BLUE: UMA PRÁTICA SUSTENTÁVEL

Murilo Pitta Rizzato 

Graduado em Engenharia Ambiental, Centro Universitário de Maringá (UniCesumar) – Maringá (PR), Brazil.

Lilian Keylla Berto 

Mestranda em Tecnologias Limpas, UniCesumar – Maringá (PR), Brazil.

Marla Corso 

Docente do curso de Arquitetura e Urbanismo, Centro Universitário Integrado/IN2, Campo Mourão – Maringá (PR), Brazil.

Ana Carolina Dias de Albuquerque 

Mestranda em Tecnologias Limpas, UniCesumar – Maringá (PR), Brazil.

Thiago Dias Azenha 

Mestre em Engenharia Ambiental, Universidade Federal do Paraná – Curitiba (PR), Brazil.

Luciana Cristina

Soto Herek Rezende 

Doutora em Engenharia Química, Universidade Estadual de Maringá (UEM). Professora do Programa de Pós-Graduação em Tecnologias Limpas, UniCesumar/ICETI – Maringá (PR), Brazil.

Correspondence address:

Murilo Pitta Rizzato – Av. Guedner, 1610 – Jardim Aclimação – CEP: 87050-390 – Maringá (PR), Brazil – E-mail: murilo_rizzato@hotmail.com

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ABSTRACT

Much research has been carried out around the development of alternative materials with the most varied purposes aiming at a positive contribution to the environment through the use of solid waste. Among these solid contaminants, the one produced in industrial effluent treatment stations, such as tanneries, stands out, where leather processing has a negative environmental impact, since the chemicals used in the treatment of raw materials, including chromium salts, have a high degree of toxicity. These residues can be used in the manufacture of alternative materials to promote the reduction of the environmental impact generated by them. Therefore, this study aimed to verify the feasibility of incorporating waste produced in a tannery effluent treatment station, known as “leather powder”, in a polymeric matrix of recycled polypropylene in the proportions of 10 and 20%, by means of making specimens by extrusion and injection processes. The specimens were analyzed in relation to the mechanical tests of tensile strength and Izod impact. The specimens containing 10 and 20% of leather powder had their resistance increased by 2.10 and 7.65% with respect to traction, 9.7 and 24.59% with respect to impact, respectively. The homogeneous distribution of leather powder particles in the polymeric matrix may have contributed to the increase in mechanical strength. This homogeneity was verified by the morphological analyzes. Thus, it is feasible to insert leather powder in recycled polypropylene in the proportions of 10 and 20%, presenting itself as a solution for the proper disposal of these residues, thus having a favorable contribution to the environment.

Keywords: sustainability; solid waste; leather powder.

RESUMO

Diversas áreas buscam o desenvolvimento de materiais alternativos com as mais variadas finalidades visando a uma contribuição positiva para o meio ambiente por meio do aproveitamento de resíduos sólidos. Entre esses sólidos contaminantes, destaca-se o produzido em estações de tratamentos de efluentes industriais, como o de curtumes, onde o processamento do couro produz um impacto ambiental negativo, pois os produtos químicos utilizados no tratamento da matéria-prima, entre eles os sais de cromo, possuem alto grau de toxicidade. Esses resíduos quando produzidos podem ser utilizados na fabricação de compósitos promovendo a redução do impacto ambiental. Diante disso, o presente trabalho teve por objetivo a fabricação de materiais compósitos. Os compósitos poliméricos foram fabricados fazendo uso dos processos de extrusão e injeção simultaneamente, em que o “pó de couro” foi incorporado em uma matriz de polipropileno reciclado nas proporções de 10 e 20%, respectivamente. Também foi fabricado um corpo de prova de referência utilizando apenas polipropileno reciclado. Após a confecção, os corpos de prova foram analisados com relação à ensaios mecânicos de

resistência à tração (ASTM D638/2014) e ao impacto Izod (ASTM D256/201). As fraturas obtidas nas amostras pelo ensaio de impacto foram analisadas por microscopia eletrônica de varredura. A adição do “pó de couro” em 10 e 20% promoveu o aumento da resistência ao impacto de 9,7 e 24,59%, respectivamente, quando comparados com o corpo de prova referência. Os compósitos fabricados apresentaram um aumento de 2,10 e 7,65%, em relação à inserção de “pó de couro” 10 e 20%, respectivamente. Por meio da morfologia, foi possível verificar uma homogeneidade na distribuição do material incorporado, contribuindo para o aumento da resistência dos materiais. Os compósitos obtidos mostraram-se eficazes em relação ao aumento da resistência mecânica, podendo ser uma solução para o descarte adequado de resíduos, promovendo uma contribuição favorável ao meio ambiente.

Palavras-chave: sustentabilidade; resíduos sólidos; pó de couro.

INTRODUCTION

The perspective in the current scenario is the management of waste that can be reused before final disposal. Industries are increasingly concerned with creating an ideal model for the production of environmentally friendly products, as this transition to sustainability motivates changes at local and global scales (HAMEED; HAMZA, 2019; SOUSA; SILVA, 2018).

In this context, several areas are analyzing the use of different materials, in order to reach potential innovators in the final product. The incorporation of waste into the production process can contribute to environmental preservation and the development of materials with properties superior to conventional ones. Thus, the environmental concern with the generation of waste has contributed to its use in the creation of new products (AL-FAKIH *et al.*, 2019; FERNANDES *et al.*, 2017; SILVA *et al.*, 2017). Innovative solutions must be focused on the best product performance, such as durability and income generation with cleaner production, in order to create new consumer behavior (TAMBOURA *et al.*, 2018; WANG *et al.*, 2018; ALMEIDA *et al.*, 2017).

Cleaner production when applied to leather production aims to reduce the demand for materials, energy and the toxicological emissions generated. Deveci *et al.* (2019) improved the tannery wastewater treatment system and presented economic and environmental advantages, since the system is more economical and efficient compared to conventional treatment, which results in a reduction of highly polluting waste.

Approximately 65 to 70% of the total weight in processed salted leather becomes waste (MURALIDHARAN *et al.*, 2020; SATHISH; MADHAN; RAO, 2019) and ap-

proximately 90% of leather production uses chromium salt (DING *et al.*, 2020), which makes it even more concerning, as the tannery industry that uses a simple treatment does not remove the chromium from wastewater and the high organic content (DEVECI *et al.*, 2019).

The proper disposal of leather waste has always been a challenge, especially since chromium is considered the main contaminant (DEVICI *et al.*, 2019). In recent years, the accumulation of industrial sludge containing heavy metals has caused environmental problems due to the toxicity contained in these elements (ISLAM *et al.*, 2017).

The leather waste can be used as raw material for the preparation of products free of toxic and harmful substances (SATHISH; MADHAN; RAO, 2019). Thus, leather waste has great relevance in research related to the environmental area and in industrial products, to achieve sustainability in the leather industry (DING *et al.*, 2020).

The tanning production process consists of three stages: cleaning the leather (removing skin, hair and skin epidermis), tanning (to stabilize and prevent putrefaction), and final processes (in which the leather is finished and the final appearance); all stages of leather processing generate solid or liquid waste (LUJÁN-FACUNDO *et al.*, 2018).

Tanning processes use chromium, vegetables or synthetic resins so that this material is inert and rot resistant, the most common being chromium salts. Chromium binds to protein in animal skin, forming leather, and presents advantages such as shorter tanning time, quality, stability and greater water resistance, provid-

ing greater elasticity to the leather and easy tanning (DING *et al.*, 2020; ISLAM *et al.*, 2017).

A major environmental concern is the generation of this waste that contains trivalent chromium in its composition, which, according to ABNT NBR 10,004/2004 (ABNT, 2004), is classified as class 1 waste – dangerous. Therefore, its disposal must be done in industrial landfills.

Due to the growth in the market, there is a great environmental concern, since one of its biggest problems is the disposal of solid residues composed by organic load, rich mainly in chromium salts, which has a high polluting potential. However, the high costs for the treatment of this waste make the industries stop treating it (PINTO *et al.*, 2019).

New industrial processes aiming to reduce the impact on the environment promote a more sustainable product manufacturing with incorporation of waste (SILVEIRA *et al.*, 2019; QUADRELLI NETO; GOMES; BORK, 2018). Therefore, one of the ways to mitigate this negative environmental impact is to reuse the waste generated by lowering the leather through the production of new composites.

Ding *et al.* (2020) used leather waste for the manufacture of plaster, in which they presented characteristics similar to the retarding additives. Muralidharan *et al.* (2020) used solid tannery residues in a polymeric matrix, which resulted in good mechanical performance.

The polymeric matrix used in the manufacture of composite materials, resulting from large-scale production in industries, mainly in the packaging sector, can be recycled due to the low cost combined with a simple process. However, it ends up becoming a waste, because, in view of its resistance to chemical and biological attacks, its decomposition takes hundreds of years (ROKBI *et al.*, 2020).

The addition of polymeric materials to the residues promotes the production of new materials with excellent properties; therefore, it is widely used today (FAKIROV, 2018).

The use of polymeric residues for the manufacture of materials has a wide scope, for example, with multilayer packaging containing polyethylene terephthalate (PET), polyethylene (PE) and aluminum that were used as reinforcement filler in the preparation of compos-

ites with post-consumer high density polyethylene matrix. Specimens were obtained with the incorporation of up to 50% of multilayer packages in polymeric matrix, resulting in a material with varied mechanical and thermal properties, with excellent mechanical properties, with tensile strength values of 148 J/m and elastic module of 350 MPa, compared to pure polyethylene (40 J/m and 450 MPa) (FAVARO *et al.*, 2017).

Rezende *et al.* (2017), in the development of wood particle boards with the incorporation of different percentages of chips used in the manufacture of labels and tags containing polypropylene, obtained satisfactory results in relation to the mechanical parameters required by the standards, so that the manufacture of an alternative product offers economic, social and environmental benefits to reduce the consumption of resources and reuse materials.

One polymer that has stood out is polypropylene (PP), considered the second most sold and used thermoplastic in the world, widely used in electronic, construction and packaging materials, due to the fact that it has good insulating properties, resistance to rupture and chemical, besides being of low cost, easy molding, tenacity, good stability and chemical, thermal and mechanical resistance. PP can be incorporated into different materials and waste so that properties are optimized (LIANG, 2017).

In view of the consequences of improper disposal of hazardous waste and the decrease in the useful life of industrial landfills, the alternative of incorporating a hazardous waste in a polymer matrix can solve an environmental problem by turning it into a by-product, since the addition of thermoplastic polymers provides excellent advantages such as low density, low abrasiveness and recyclability (SULLINS *et al.*, 2017).

The development of this material contributes to the Sustainable Development Goals (SDGs) — the 17 environmental sustainability goals created by the United Nations (UN). This work meets 2 SDGs, among the 17 objectives: one related to sustainable cities and communities, which has a link with the sustainable development of the urban area, so that initiatives that contribute to this aspect fall into this objective; and the other correspondent to responsible consumption and production, aimed at changing the environmental awareness of industries and the final consumer (ONU, 2015).

Thus, the present research had as objective the incorporation of residues of the leather chips contaminated with chromium in a polymeric matrix of recycled polypropylene, referred to as PPR, in order to inert the con-

taminated leather and produce a material that can be used in construction systems, minimizing, in this way, an environmental problem in the tanning industry and still adding value to the material.

MATERIALS AND METHODS

In this research, two materials were used to make the specimens: leather powder and PPR. The leather dust sample was collected from the Bergi 1800 sanding machines from a Wet-Blue leather retanning industry located in the Maringá region, a city in the Brazilian state of Paraná. This material was sucked by an exhaust fan and compacted in a briquette machine (without specification), in order to reduce the volume and the propagation of the material in the work environment.

The PPR was donated by a plastic recycling industry located in the city of Maringá, where they were crushed into flakes. The PPR was analyzed for granulometry using a sieve shaker (electromechanical sieve shaker with analog timer — SoloCap), using the sieve series: 9.5 mm (3/8"); 4.75 mm (No. 4); 2.36 mm (No. 8); 2 mm (No. 10); 1.18 mm (No. 16) and the bottom, according to ABNT NBR NM: ISO 3310-1/2010 (ABNT, 2010). The calculations and results are in accordance with those provided for in DNER 080/94 (DNER, 1994).

In this work, they were used the mechanical and morphological parameters of a reference matrix formed by PPR and two composites containing PPR and leather

powder added in the proportion of 10 (PPR10%) and 20% (PPR20%), respectively. These three samples were weighed in relation to the proportion of materials used to make the specimens.

For the preparation of the specimens, the samples after proportional weighing of the materials were processed in a double screw extruder, model Thermo Scientific MiniLab II HAAKE Rheomex CTW 5, using a mold temperature of 190°C and a speed of 65 rpm, coupled with a injection molding Thermo Scientific HAAKE MiniJet II, with gun temperature of 210°C, mold temperature of 40°C, injection pressure of 650 bar, injection time of 15 s, pressure of 300 bar and pressure of 30 s.

The dimensions of the injector molds meet the requirements of the ASTM D638/2014 and ASTM D256/2010 standards, for carrying out the Izod traction and impact tests, respectively (ASTM, 2010; 2014). In total, 30 specimens were made to perform the tensile test, 10 containing only PPR, 10 containing 10% leather dust and 10 bodies containing 20% leather dust (Figure 1A). For the Izod impact test, 16 specimens were used for each proportion (Figure 1B), totaling 48 specimens.

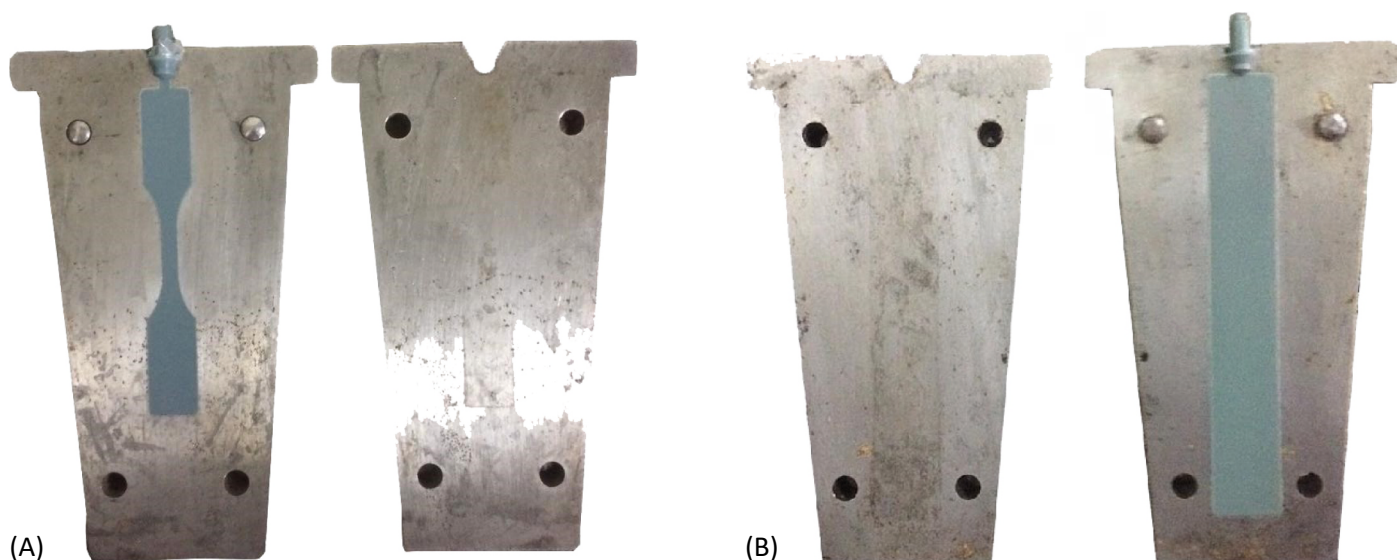


Figure 1 – Test mold: (A) traction; (B) izod impact.

After making the specimens, mechanical traction tests were carried out (ASTM, 2014), using a universal testing machine EMIC DL10000, with a 5 kN load cell. The Izod Impact (ASTM, 2010) was performed on CEAST equipment, model Resil Impactor Junior, with a 2.75 J pendulum.

In order to determine the tensile strength of each test specimen tested, the highest recorded stress value in Megapascal (MPa) was extracted. Aiming at determining the Izod impact resistance values for the specimens, the calculations provided for in ASTM D256/2010 were performed.

With these values of tensile strength and Izod impact of each specimen, calculations of the mean (sum of the strength values of each sample divided by the number of specimens) and of the standard deviation (SD) for the PPR, PPR10% and PPR20% samples were performed

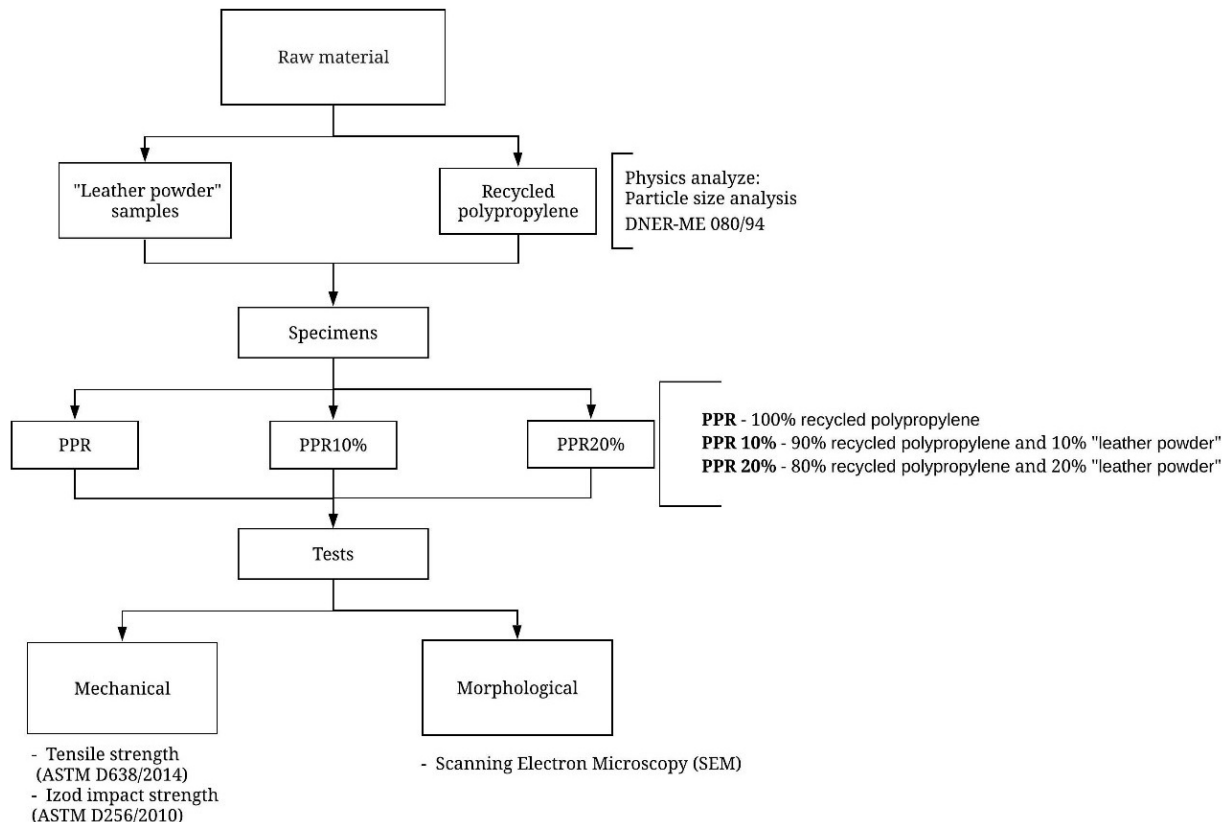
The calculation of the SD for the results related to the tensile strength tests and the Izod impact of the tested specimens is expressed by Equation 1.

$$SD = \pm \sqrt{\frac{\sum(x - \bar{x})^2}{(n-1)}} \quad (1)$$

The SD was calculated using the square root of the sum of the equations represented by the symbology Σ , being composed of the resistance values of each specimen (x) minus the average of all specimens for each test (\bar{x}). This difference between the resistance value of the specimens and the average was squared and divided by the number of specimens (n) minus one. The value resulting from the SD was considered both positive and negative.

The specimens resulting from the Izod impact test were analyzed according to the morphological parameters, using scanning electron microscopy (SEM), under a Shimadzu microscope, model SS-550, to analyze the distribution of particulate materials.

Figure 2 shows the methodology used to carry out this work.



PPR: recycled polypropylene; ASTM: American Society for Testing and Materials; DNER: Departamento Nacional de Estradas de Rodagem.

Figure 2 – Steps, analyzes and methodologies used to determine mechanical and morphological properties.

RESULTS AND DISCUSSION

Particle size analysis

Particle size analysis is essential to characterize and evaluate the material in relation to the particle size distribution and the particle size percentages, respectively (BITTEN-COURT *et al.*, 2018). From the results of the granulometric analysis, it was possible to identify the geometric variation of the crushed PPR. In this way, characterization of the granulometric distribution showed the average variation of the material was around 2.36 to 4.75 mm, that is, the largest parts of the material were retained in the screens No. 4 and No. 8, as shown in Figure 3.

The understanding of particle size analysis is notable for explaining physical and mechanical effects (BITTEN-COURT *et al.*, 2018). Corso *et al.* (2020) and Berto *et al.* (2018) used granulometry to determine the average

variation of the particle sizes of the material, as they directly influence the final resistance of the same, the average variation of the particle sizes of the material, in order to characterize them in relation to the size of the particles, in the same way that was carried out in this research.

The variation in the size of the PPR particles was adequate to make the extrusion process feasible, since if the average variation of the crushed PPR presented higher values than those determined, the material should probably be crushed again before starting the extrusion process — as a consequence of the laboratory scale —, which can cause a complication with the clogging of the material in the extruder nozzle.

Mechanical tests

The results for the Izod tensile and impact tests are shown in Figure 4. The average strengths and standard deviations of the reference matrix formed by PPR and two composites containing PPR and added leather powder were presented in the proportion of 10 (PPR10%) and 20% (PPR20%).

The mean tensile strength of the PPR, PPR10% and PPR20% specimens resulted in 30.95 ± 0.65 MPa, 31.6 ± 0.59 MPa and 33.32 ± 0.37 MPa, respectively. This represents a 2.10% increase for the insertion of 10% leather powder and 7.65% with the addition of 20% leather powder. According to Callister (2002), polypro-

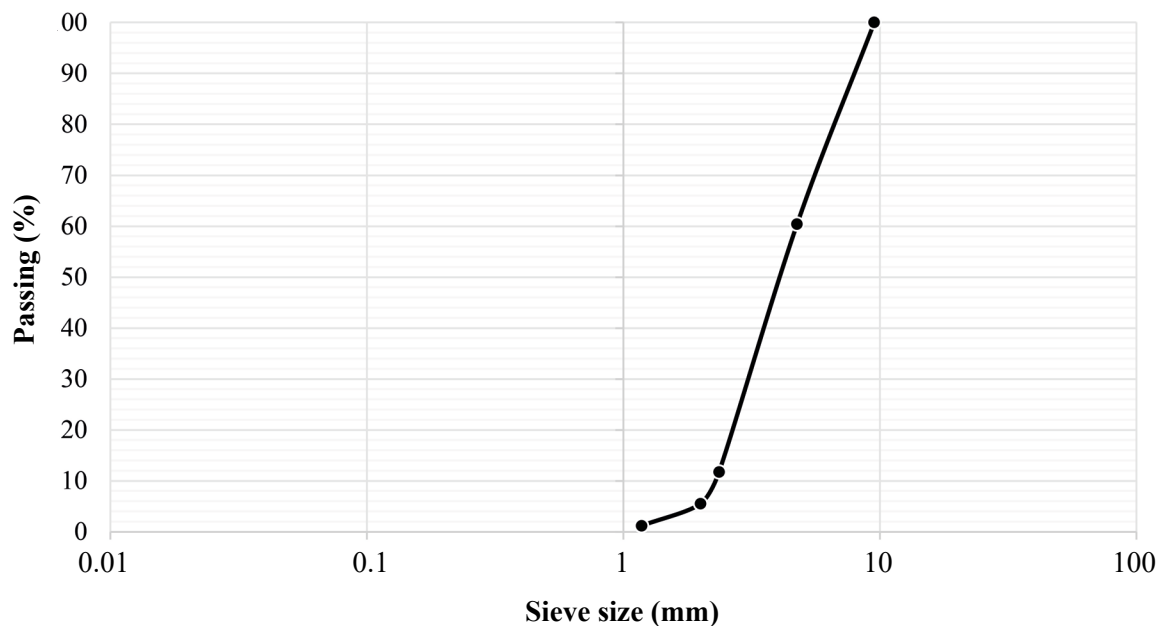


Figure 3 – Particle size analysis for the recycled polypropylene sample.

polyene exhibits tensile strength between 31 to 41.4 MPa, so the specimens made showed similar characteristics, even after recycling and adding fibers from the sanding machines of a Wet-Blue leather retanning industry.

Similar researches of Castiello *et al.* (2009), Masilamani *et al.* (2017) and Muralidharan *et al.* (2020) used residues from the leather industry to make composites based on polymers. However, unlike the results presented in this article, it was possible to observe in these studies that the reduction in tensile strength was proportional to the increase in the insertion of leather-based residues in the reference matrix.

Castiello *et al.* (2009) reused the collagen hydrolyzate derived from the leather industry for the production of polyethylene-based thermoplastic films. The results of the average tensile strength in duplicate were 14 and 12.70 MPa for the reference matrix, 12.1 and 10.75 MPa with 10% of leather-derived shavings, and 9.20 and 8 MPa with 20% of the same leather-derived material.

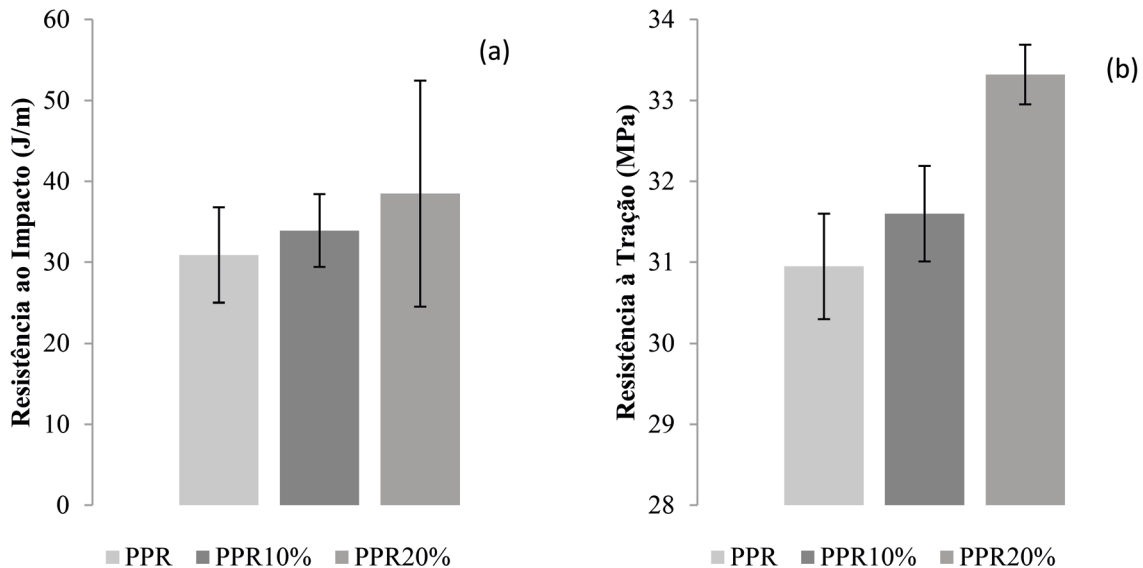
Masilamani *et al.* (2017) extracted the gelatinous material from raw bovine leather shavings for filmmaking. The tensile strength for the film containing 20% gelatinous material from leather chips with 80% polyvinyl alcohol (PVA) polymer was 51.08 ± 0.16 MPa and for the reference matrix with 100% PVA it was 53 ± 0.47 MPa.

Muralidharan *et al.* (2020) used tannery residues for the preparation of transparent bioplastic films, with a concentration of 6, 8, 10, 20, 30, 40 and 80% citric acid, presenting resistance to average traction of 32.08, 41.62, 36.39, 35.34, 22.72, 20.75 and 11.15 MPa, respectively.

As in this article, other authors, including Rokbi *et al.* (2020) and Rosário *et al.* (2011), used PPR with the addition of reinforcement material for making the composite. In Rokbi *et al.* (2020), the making of a plate from interspersed layers of PPR and simple jute fabric with a temperature of 210°C had a tensile strength of 19.09 ± 0.18 MPa.

For Rosário *et al.* (2011), the results of tensile strength according to the standard (ASTM D638/2014) resulted in 100% PPR in 22.78 ± 1.14 MPa and PPR with the addition of 30% sisal treated fibers, 87 ± 1.69 MPa, approaching the value obtained in this work for the tensile strength specimen of the tested PPR20%.

The results of Izod impact resistance (ASTM D256/2010) for the PPR, PPR10% and PPR20% specimens, according to Figures 4A and 4B, were 30.9 ± 5.90 J/m; 33.9 ± 4.5 J/m and 38.5 ± 13.96 J/m, respectively. This order of values refers to the 9.7 and 24.59% increase, as the leather powder was added in the proportions of 10 and 20%.



PPR: recycled polypropylene.

Figure 4 – Results of mechanical tests: (A) izod impact strength; (B) tensile strength.

This increase in mechanical performance proportional to the addition of leather powder shows that solid residues containing organic matter can increase the mechanical strength of polymeric composites, as reported by Razi, Islam and Parimalam (2019), who, when analyzing composites reinforced with 10 and 20% of waste from pisciculture, obtained an increase in resistance to the Izod impact of 6.78 and 20.34%, similar to the increase in the research carried out in this article.

Scanning electron microscopy

The SEM presents the evaluation of the interface of the studied fibers, as well as other morphological aspects, such as the fracture surfaces of the material. Other aspects that can be evaluated are the dispersion and adhesion of the fibers with the polymeric matrix.

Rosário *et al.* (2011) obtained resistance to the Izod impact of 62.29 ± 2.1 J/m for PPR specimens reinforced with sisal fiber, with the composition of 70% by weight of PPR and 30% by weight of treated sisal fiber, and without the addition of PPR the resistance obtained was 26.25 ± 4.5 J/m. The explanation for this fact is due to the longitudinal orientation in the injection process and perpendicular in the Izod impact test, which made the composite material more resistant.

It is observed that the increase in the amount of leather powder is directly proportional to the increase in the interfacial adhesion of these composites. The fracture surface morphologies of the composites in Figures 5A, 5B and 5C, as verified in the study by Firmino *et al.* (2017),

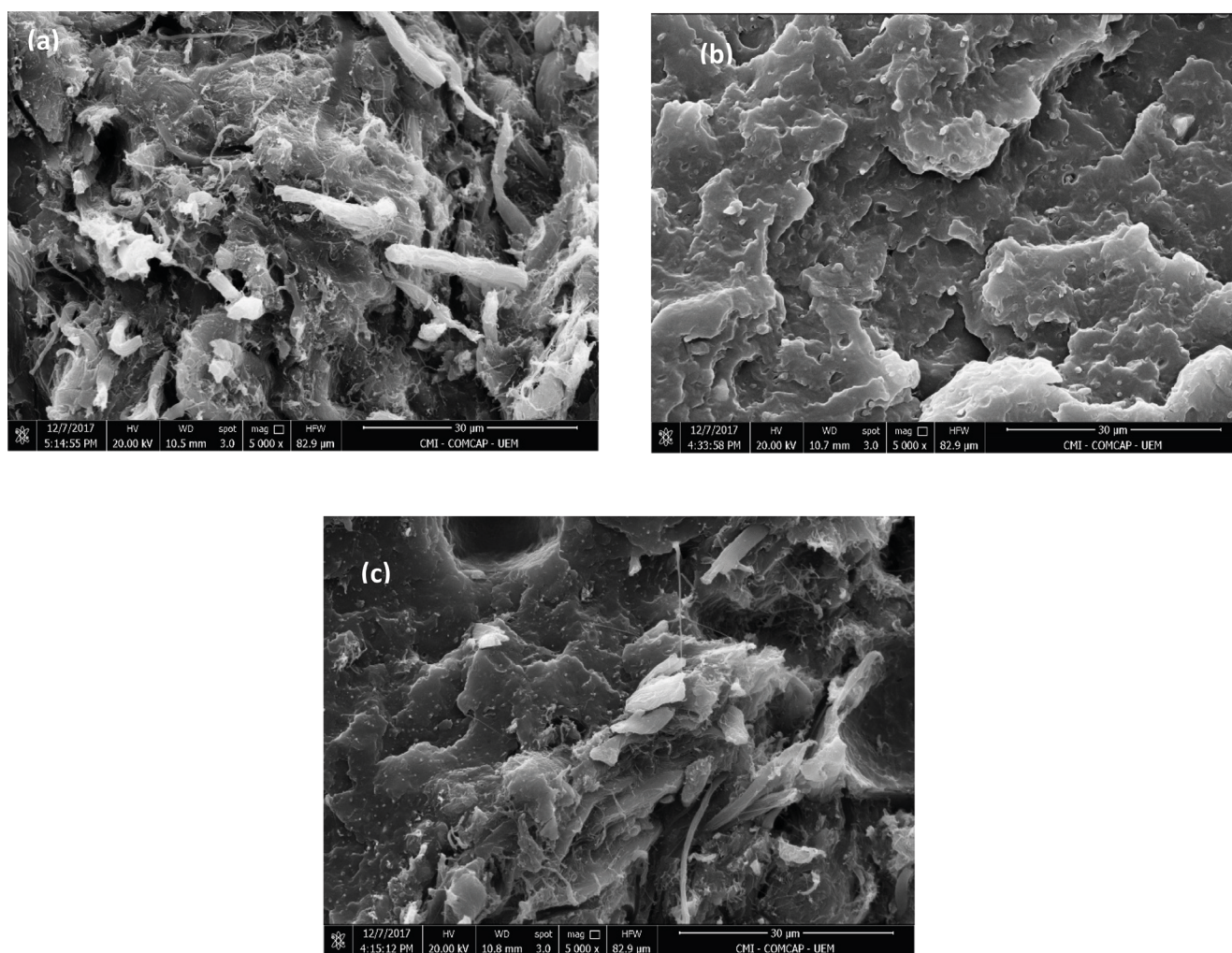


Figure 5 – Scanning electron microscopy: (A) pure recycled polypropylene (PPR); (B) PPR10%; (C) PPR 20%.

showed that the particle size of the reinforcement material is inversely proportional to its effective performance in the reference matrix. In this way, it is proved that the compacting of the material before extrusion aims to decrease the volume, being efficient to increase the homogeneity of the composites, due to the ease of fusing the materials used in the manufacture of the bodies, making them more resistant composites in relation to the reference matrix, as seen in the mechanical results.

When analyzing the impact fracture of the sample, it can be seen that the dispersion of leather powder with the PPR10% was uniform (Figure 5B). Muralidharan *et al.* (2020) verified through morphology the fibrous and porous structures of the leather, and reported that the structure presented by the residue may favor the interaction between the leather and the matrix used in the manufacture of composites, a fact also observed in this work.

CONCLUSION

The manufacture of composites from PPR and the addition of Wet-blue leather powder are effective when it is desired to increase the resistance of the specimens. In addition, from an economic point of view, they make the compost cheaper, considering that it is a material that would be discarded in industrial landfills and would generate costs for the industry. From the environmental point of view, it is necessary to carry out studies to analyze the leachate of the compound, in order to verify whether the chromium present in the leather is inert. If so, there is a possible solution to the problem of disposing of this hazardous waste.

Regarding the use of PPR, it can be said that it would be a great solution for small recycling cooperatives in smaller cities and even the incentive for small cities that do not have selective collection. These coopera-

tives, because they produce a smaller volume, end up taking a long time to sell their product, which leads to a long storage time up to the ideal amount for delivery, taking up space in their facilities. With the insertion of the polymer as a polymeric matrix, consequently, this material would leave the cooperatives more quickly, generating more turnover and financial turnover.

The PPR with the addition of leather powder fibers showed acceptable mechanical properties that require less mechanical efforts for the production of new materials, such as in the construction sector and even in livestock, in the form of platforms, pallets, closing plates and tiles, for various purposes. Therefore, these products must be tested in compliance with the requirements of the standards to start production on an industrial scale.

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