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Economic Viability and Flammability of Polyurethane Composites, Aluminum Sludge and Polyethylene Terephthalate Residue

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Abstract

Polyurethane is used in the construction industry because of its excellent thermal performance in roofs, floors, and concrete slabs. However, its high flammability restricts the use. The study reports the use of polyethylene terephthalate and aluminum-anodizing sludge residues in the production of boards with different densities and fire resistance. Boards with 10%, 20%, 30%, 40%, and 50% of polyethylene terephthalate residue were prepared to replace primary polyurethane raw materials, to which 20% aluminum sludge was added. In the horizontal burning test (UL94), the boards presented a combustion deceleration until flame extinction due to the presence of aluminum-anodizing sludge. There was a cost reduction of about 70% for the boards with the greatest amount of residues incorporated. The construction industry should consider incorporating waste into the life cycle of products from other segments as part of its formulations, saving natural resources and becoming more sustainable.

Keywords: Aluminum sludge; Polyethylene terephthalate (PET); Polyurethane (PU); Recycling; Flammability.

1. Introduction

Concerns for society have been increasing regarding the reuse of post-consumer materials for the manufacture of new materials or products, attributing value and a life cycle for a great variety of wastes (CARVALHO et al., 2015; GENOVESE et al., 2017). Technological advancements prompt to trigger new ways of using discarded materials, contributing to sustainable development, as well as new business opportunities, job creation, and social inclusion. Solid wastes have become an opportunity for scientific and technological development, leveraging premises of cleaner production. Turning waste into a useful product, involving environmental, economic, and social interaction will improve natural resource protection and bring back what was once discarded to a new life cycle (KJAERHEIM, 2005; BAAS, 2007; NAGEL, 2013; TEUBER et al., 2016; REBEHY et al., 2017). Developing techniques capable of incorporating discarded materials as part of the composition of new materials is of utmost importance (THIRUMAL et al., 2007; MAULER et al., 2013; YAM; MAK, 2014; TEUBER et al., 2016).

The construction industry is a productive segment that can incorporate significant amounts of discarded materials into the product life cycle as part of its formulations, making it more sustainable. Adding residues to a polymer during the polyurethane expansion reaction is an alternative to optimize the use of residues (PEREIRA et al., 2008; PEREIRA, 2016; PIVNENKO et al., 2016; REBEHY et al., 2017).

Aluminum sludge is a residue generated from the anti-corrosive reaction that oxidizes the surface of metal parts, called anodizing. This type of treatment generates waste with a variety of chemical alkaline compounds, which cause negative effects to the environment and, therefore, should be disposed in industrial landfills. Moraes (2012) has reported that the anodization process generates around 100,000 tons/year in the European Union country members. Because of the vast amount of waste, big landfills are required at a high cost (US$ 2,230/t). In Brazil, it is estimated that 1 ton of sludge is generated for each ton of anodized material (SARTOR, 2006; LEITE, 2008, PEREIRA et al., 2008, MORAES et al., 2012; PADOIN, 2012; FLORIANO, 2014; MANFROI et al., 2014). Gorninski; Tonet, (2016), Floriano (2014), and Pereira (2008) have reported that aluminum anodizing sludge is mainly aluminum trihydroxide that can be used as a fire retardant in polymers by different industrial sectors (ZHANG, et al. 2005).

The use of polyethylene terephthalate (PET) is well-established in the food packaging industry, and is most commonly used for short-lived products. These products, when improperly discarded by consumers, result in environmental problems, mainly because of the polymer slow degradation. In Europe, 25.2 million tons of plastic waste were produced in 2012, of which 62% were recycled and utilized for energy recovery (PLASTICSEUROPE, 2015). In that same year, a total of 195 billion beverage packages were sold in the United States, of which about 11% were recycled (KANG et al., 2017). In Brazil, 274 thousand tons (51%) of the produced beverage packages were recycled in 2015 (KREIGER et al., 2014; NALINI et al., 2016; ABIPET, 2016; REBEHY et al., 2017; NATIONAL, 2017).

Moya et al. (2013) and Fazeni et al. (2014) have claimed that unavoidable residues should be recycled or reused, so that the minimum amount is sent to sanitary or industrial landfills (CONAMA, 2002, Article 4). Therefore, polyurethane, polyethylene terephthalate, and aluminum sludge composites were prepared in the form of boards for possible application in the construction industry sector. In this study, up to 50% of the PU-forming reagents were replaced by PET residue, and aluminum sludge was added to provide a fire resistance property for the new composites. Lastly, the economic feasibility of these composites was demonstrated.

2. Methods

This was an exploratory, descriptive study conducted in a laboratory setting. Polyethylene terephthalate and aluminum anodizing sludge were the two post-industrial residues used as secondary inputs to obtain the boards. They were incorporated at different proportions into the polymer matrix. The post-industrial polyethylene terephthalate residue was donated by a mineral bottled water company. The bottles were crushed using a BOTINI mill equipment (B55 model) and sieved. Grains of polyethylene terephthalate passing through a 1.4 mm sieve and retained in a 1.0 mm sieve were selected for the manufacture of boards. The aluminum anodizing sludge was stripped and washed with distilled water by stirring at 50 °C for about 2 hours. Then, the heterogeneous mixture was filtered and the collected solid was oven-dried at 70 °C for 24 hours. Next, the material was classified by grain sizes using a sieve with aperture sizes of 0.044 mm, which means that particle sizes smaller than 0.044 mm were selected. A sample of the material was examined by X-ray fluorescence and atomic absorption spectrometry to determine the elements present in the anodizing sludge residue. The assay was conducted according to PR-CR-097, PR-CR-098, and PR-CR-103. X-ray diffraction was used to identify the phases present in the studied residue. X'Pert diffractometer was the equipment used for the experiment, and the data obtained were based on standard data from the Joint Committee for Powder Diffraction Starndards (JCPDS) database. An electronic search of data on aluminum anodizing sludge was made in the literature to compare with the results obtained in our study.

Polyol polyether (polyol) and toluene-2,6-diisocyanate (TDI) (Arinos), aluminum anodizing sludge (AAS) and polyethylene terephthalate were used to manufacture the boards, following the safety standards of each reagent. The specimens were obtained by mixing PET and AAS to the polyol and then TDI was added and mixed for a further 0.5 min. The mixture was poured into the mold, previously anointed with Vaseline, and the specimens were demolded after 24 hours (MARIAPPAN, 2014; PENG et al., 2014). The PU, PU_AAS and PU_ATH test specimens were prepared as standards. Board density was calculated based on ASTM D1622 (2014).

The horizontal burning tests were carried out according to the ABNT NBR 9178-15 standards, ASTM D635-14 and UL-94. The test was run in triplicate on each specimen with the following dimensions: 125-mm long, 13-mm wide, and 7-mm thick. The specimens were oven-dried for 168 h at 70 °C. A flame was applied to each test specimen held at the 45-degree angle for 30 seconds. Then, the time required for flaming combustion to reach the 25 mm reference mark was recorded, and if the combustion continues, the time required to reach the 100-mm mark was recorded. If the 100-mm mark was not reached, the time and the damaged length were recorded. A classification was assigned if the following conditions were met: the combustion rate was not greater than 38 mm/min; flaming combustion self-extinguished before reaching 100 mm.

The economic feasibility analysis was based on a comparative method of direct costs (input costs) of the boards manufactured from the AAS and PET residues when incorporated into a PU matrix with commercial PU. The first stage consisted of analyzing and projecting the costs and expenses that were incurred for the manufacturing of each board (different composites). This was done by budgeting the recycled PET, polyol and TDI with suppliers. Aluminum sludge is a material disposed of in industrial landfills, so there was no direct cost. The AAS costs taken into account in this study were related to the cost in Brazilian Real (BRL) per ton for its deposit in the industrial landfill besides the transportation cost in BRL per ton. Costs in Real have been converted to Dollar (commercial US$ 3.25). The second stage consisted of the analysis of cost reduction to manufacture the boards as compared to the PU of the same density. The third stage consisted of the evaluation of the cost-benefit ratio that changed the composition by replacing the polyol matrix plus TDI by PET plus AAS. In this study, a comparison was made between the cost of the industrial production of 1 m^3 of PU and the cost of producing 1 m^3 of boards, based on volume and density. With the incorporation of PET and AAS in the PU boards, the density of the composites had considerable variations, being necessary to apply the direct rule of three to figure out the mass of polyol and TDI corresponding to the density of the boards.

3. Results and discussion

Anodizing aluminum sludge has a variety of chemical compounds, as described in different studies (SARTOR, 2006; LEITE, 2008; PEREIRA et al., 2008; MORAES et al., 2012; PADOIN, 2012; FLORIANO, 2014), as summarized in Table 1. The data obtained for the anodizing aluminum sludge (AAS) used to manufacture the boards are also shown in Table 1.

Table 1. Percent chemical composition of anodizing sludge found in different studies.

Study	Al_2O_3	SiO_2	Fe_2O_3	CaO	Na_2O	K_2O	MgO	TiO_2	ZnO	SO_3	MnO	P_2O5	*L.F.
Sartor, (2006)	59.2 a 72.5	<0.01 a 2.0	0.1 a 0.4	<0.1 a 0.5	0.2 a 6.1	<0.1 a 0.1	<0.1 a 0.1	<0.1 a 0.1	-	-	-	-	24.3 a 38.8
Pereira, (2008)	35.3	1.2	1.4	3	0.4	0.1	0.1	0.1	<0.1	16.7	-	-	40
Leite, (2008)	35.3	1.2	1.4	3	0.4	0.1	-	-	-	16.7	-	-	40
Padoin, (2012)	64.1	0.5	0.2	<0.1	10.9	0.1	0.4	<0.1	-	-	<0.1	<0.1	23.7
Moraes, (2012)	55.5	0.6	0.3	<0.1	2.9	<0.1	0.1	-	<0.1	12.1	-	-	28.3
Floriano, (2014)	67.88	0.62	0.35	0.15	3.9	0.13	<0.05	<0.05	-	-	<0.05	<0.05	26.81
AAS	66.16	0.48	0.24	0.29	1.32	-	-	-	-	-	-	0.24	26.74

*LF = Loss due to fire. Source: Adapted from Floriano, 2014; Moraes et al., 2012; Padoin, 2012; Leite, 2008; Pereira et al., 2008; Sartor, 2006.

The last line of Table 1 shows the composition of the AAS, analyzed by X-ray Fluorescence and Atomic Absorption Spectrometry. The composition varied according to the analyzed sample, although all had aluminum oxide as the main constituent (Table 1). The sample used for this study contained about 66% aluminum oxide of its composition, which means that aluminum was the predominant metal in the composition. X-ray diffraction measurements revealed that aluminum was present as aluminum hydroxide oxide [$AlO(OH)$], aluminum phosphate [$AlPO_4$], and aluminum trihydroxide [$Al(OH)_3$]. Thus, AAS was mainly composed of chemical components typically used as flame retardants (Gallo; Agnelli, 1998). That is why AAS, a secondary input, was used to replace the aluminum trihydroxide (ATH), a primary input. Moreover, it is important to note that ATH is an input that is usually part of the composition of polymers and composites due to flame retardant properties (ZHANG et al., 2005; LAOUTID et al., 2009; THIRUMAL et al., 2010; TANG et al., 2015; WANG et al., 2015).

The boards were obtained by polycondensation between polyol and TDI for PU formation, with the addition of PET and AAS (ZHANG et al., 2014; GUO et al., 2015; ZHANG et al., 2015; GARRIDO et al., 2016; KANG et al., 2017). Table 2 shows the amount of mass reagents used to manufacture the boards and their respective densities.

Table 2. Amount of polyol and TDI reagents to prepare PU, PU_AAS, and PU_ AAS_PET composites in the percentages of 10%, 20%, 30%, 40%, and 50% of PET with the addition of 20% AAS.

Sample	Polyol [g]	TDI [g]	PET [g]	AAS [g]	ATH [g]	Density [kg/m^3]
PU	10	15	-	-	-	85.29
PU_ATH	10	15	-	-	5	125.17
PU_ AAS	10	15	-	5	-	133.78
PU_ AAS_10PET	9	13.5	2.5	5	-	144.04
PU_ AAS_20PET	8	12	5	5	-	159.33
PU_ AAS_30PET	7	10.5	7.5	5	-	180.52
PU_ AAS_40PET	6	9	10	5	-	202.58
PU_ AAS_50PET	5	7.5	12.5	5	-	239.91

The addition of PET and AAS in the PU matrix led to a density increase in the specimens (Table 2). Fig. 1 shows a PU_ASS_50PET sample board.

Fig. 1. Board composed of 5g polyol, 7.5 TDI, 12.5 g PET, 5 g AAS, and PU_AAS_50PET sample.

The board shown in Fig. 1 was manufactured with the following characteristics: 16 cm^2 in dimension, 2 cm thick, with a flat surface, without deformation or crumbling, and good visual appearance.

The materials were investigated with regard to the capability to restrain flame propagation in the horizontal direction, since they were designed to be used in building roofs, floors, or slabs. Fig. 2 shows the specimens after the horizontal burning test for the PU and its composites. The images were captured after the Bunsen nozzle removal and self-sustained flame combustion cessation.

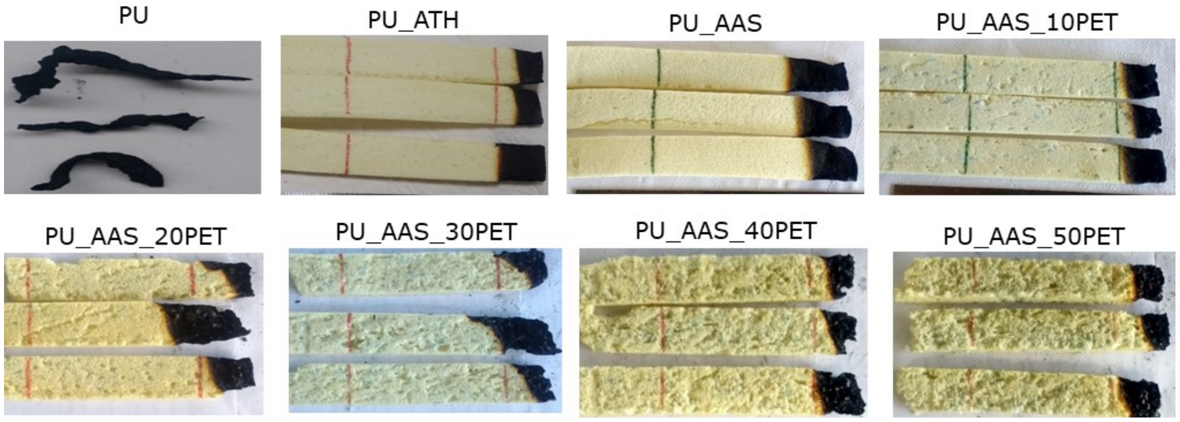

Fig. 2. Specimens after the horizontal burning test (UL94) for PU, PU_ATH, PU_AAS, and PET composites in the grain size of 1.4 mm in 10%, 20%, 30%, 40%, and 50% of the matrix mass and 20% of AAS.

Fig. 2 shows that the addition of PET and AAS allowed for restraining flame propagation as in the case of PU. The PU sample showed complete combustion at a rate of 212.45 mm/min. Complete burning occurred after a 25-mm mark for PU_ATH, PU_AAS, PU_AAS_20PET, and PU_AAS_30PET, at a burning rate of 34.05 mm/min, 37.50 mm/min, 40.00 mm/min, and 23.95 mm/min, respectively. The flame was extinguished before reaching the 25-mm mark in the horizontal burning test for the PU_AAS_10PET, PU_AAS_40PET, and PU_AAS_50PET specimens as shown in Fig. 2. All the composites were approved according to the horizontal burning test, and the best results were observed for the composites with 40% and 50% of PET in the composition, since they did not present self-sustained burning after the 25-mm mark and presented higher amount of residues incorporated as compared to the other composites, but all specimens met the UL-94 horizontal burning standard.

Fig. 3 shows the percentage of mass loss after the horizontal burning test for the samples prepared according to Table 2.

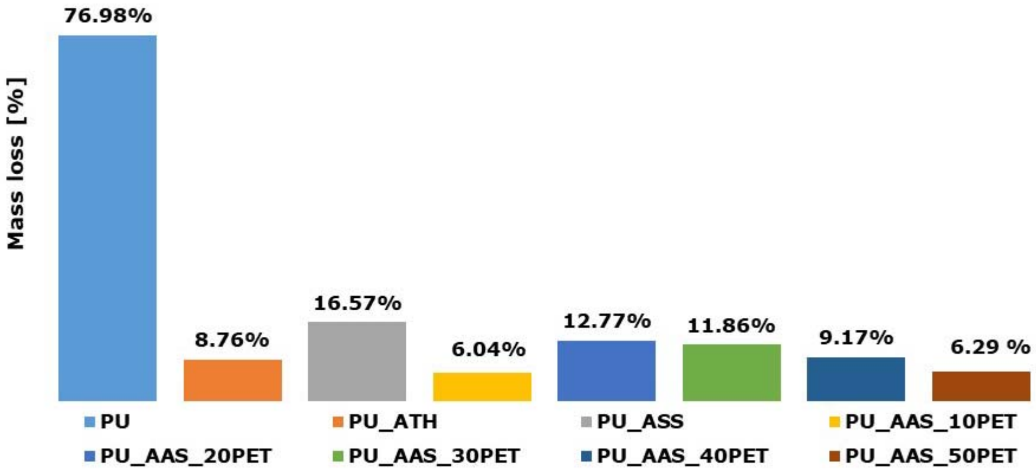

Fig. 3. Percentage of mass loss after horizontal burning test for PET composites in the grain size of 1.4 mm in 10%, 20%, 30%, 40%, and 50% of the matrix mass, and 20% of AAS.

Analysis of Fig. 3 revealed that the addition of PET, AAS, and ATH provided lower mass loss during the horizontal burning test as compared to PU. The PU_ATH sample presented a higher flame resistance due to the presence of ATH, having 8.76% of mass loss, whereas the PU_AAS composite had a 16.57% loss of the initial mass, which was explained by the presence of aluminum sludge in the composition. For the composites with 10%, 20%, 30%, 40%, and 50% of PET, the percentages of mass loss were 6.04%, 12.77%, 11.86%, 9.17%, and 6.29%, respectively. The replacement of part of the polyol and TDI reagents by PET, as well as the addition of AAS to the total mass of the composites, provided a higher flame resistance as compared to the PU. Materials with higher flame resistance are important when buildings are on fire as they increase the chances of escaping the fire. The smaller the mass loss, the smaller the amount of smoke in the environment, which means less amount of toxic gases, allowing for a better visualization of fire escape routes.

The direct costs of the inputs and raw materials to manufacture the PU boards were taken into account for the economic feasibility study. The costs of TDI and polyol referred to the purchase values of the material for the research. The PET cost was provided by an industry in the state of Santa Catarina after being processed and ready for use. The value of the anodizing sludge corresponded to the cost of dispose the waste in an industrial landfill (Joinville, Santa Catarina, Brazil), which was 0.35 BRL/kg plus the cost of a transportation distance of 300.9 km (between the cities of Joinville and Tubarão), which was 0.25 BRL/kg, in a poly crane truck that transports up to 7 tons, as shown in Table 3. Table 3 lists the costs for the production of 1 m^3 of commercial PU with the densities found in the laboratory and the costs to produce 1 m^3 of composite with the respective density.

Table 3. Costs for the production of commercial PU and PU_AAS_PET composites.

Material\density	Polyol		TDI		PET		AAS		Total cost [US$/m^3]
	US$ 60.00		US$ 60.00		US$ 1.30		US$ 0.18		
	Mass [Kg]	Cost [US$]	Mass [Kg]	Cost [US $]	Mass [Kg]	Cost [US$]	Mass [Kg]	Cost [US$]	
Commercial 144.04kg/m^3	8.99	539.29	13.48	808.93	-	-	-	-	1,348.22
PU_AAS_10PET 144.04kg/m^3	7.3	438.00	10.95	657.00	8.53	0.02	2.6	0.48	1,098.10
Commercial 159.33kg/m^3	9.94	596.53	14.91	894.80	-	-	-	-	1,491.33
PU_AAS_20PET 159.33kg/m^3	6.49	389.40	9.73	583.80	4.06	5.25	2.6	0.48	979.02
Commercial 180.52kg/m^3	11.26	675.87	16.9	1,013.80	-	-	-	-	1,689.67
PU_AAS_30PET 180.52kg/m^3	5.68	340.80	8.52	511.20	6.08	7.86	2.6	0.48	860.34
Commercial 202.58kg/m^3	12.64	758.46	18.96	1,137.69	-	-	-	-	1,896.15
PU_AAS_40PET 202.58kg/m^3	4.87	292.2	7.3	438.00	8.11	10.48	2.6	0.48	741.16
Commercial 239.91kg/m^3	14.97	898.22	22.46	1,347.34	-	-	-	-	2,245.56
PU_AAS_50PET 239.91kg/m^3	4.06	243.6	6.08	364.80	10.1	13.10	2.6	0.48	621.98

The values found in Table 3 revealed that the incorporation of PET provided a reduction of manufacture costs. The costs were gradually reduced by the increase in the percentage of incorporated PET, also increasing the density of the obtained board. As a result, the following cost reductions were obtained: 18.55% for PU_AAS_10PET with a density of 144.04 kg/m³; 34.35% for PU_AAS_20PET with a density of 159.33 kg/m³; 49.08% for PU_AAS_30PET with a density of 180.52 kg/m³; 60.61% for PU_AAS_40PET with a density of 202.58 kg/m³; and 72.30% for PU_AAS_50PET with a density of 239.91 kg/m³. The data shown in Table 3 allowed us to draw Fig. 4 to represent the cost-benefit ratio (CBR) of the PU production costs compared to the composites at their respective densities.

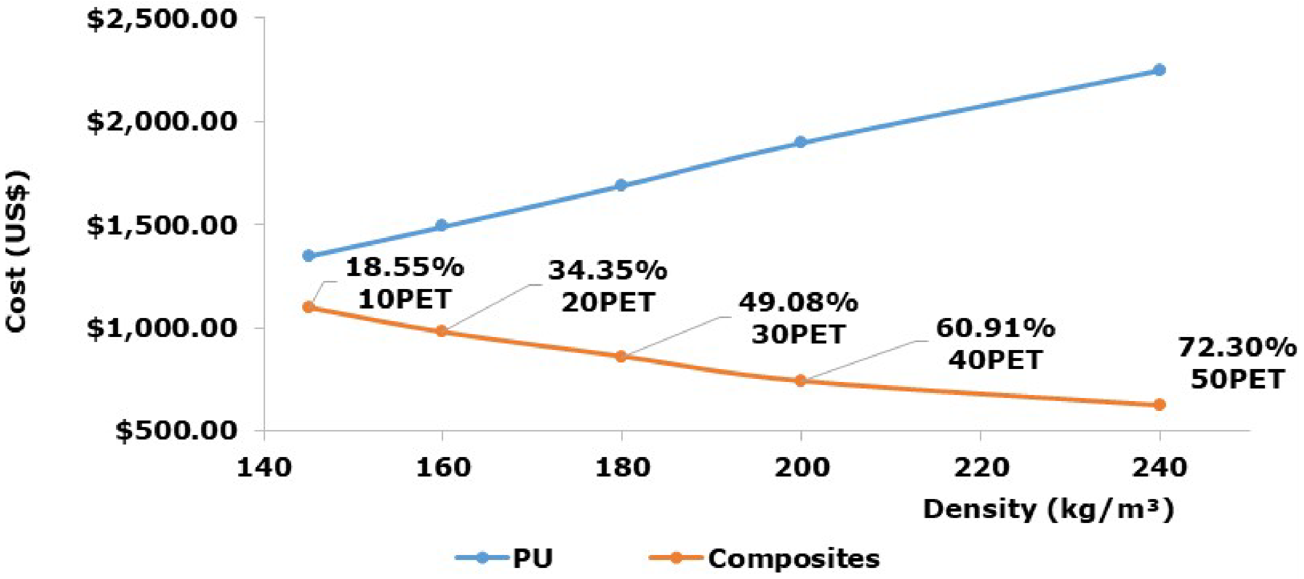

Fig. 4. Comparison between the production costs for PU and the different composites.

In addition to the reduction of the board manufacture costs, the use of PET as a direct raw material also provides environmental and social gains and increases the recycling rate.

4. Conclusions

The boards obtained in this study represent a new combination of primary and secondary inputs through mechanical recycling. The incorporation of secondary inputs was 10%, 20%, 30%, 40% and 50% of PET mass and a 20% increase of aluminum sludge mass. The density of the composites augmented with the increase of secondary inputs. The composites allow us to expand post-consumer PET recycling and increase the use of industrial anodizing aluminum waste, thus contributing to waste recycling. The use of PET and aluminum sludge waste contribute to the proper disposal of waste, thereby preventing additional damage to the environment.

The composites showed combustion deceleration, with no flame propagation in the horizontal burning test (UL94), which is an important characteristic for materials that can be used in the construction industry.

The economic feasibility study showed that all the composites had a reduction in the production costs with the replacement of the PU reagents by PET and AAS. The board with 50% PET in its formula showed a reduction of 72.30% in the direct material cost for the industrial production.

Further studies should examine the mechanical, acoustic, and thermal properties of the boards. A business plan should be drawn with the cooperation of an industry to manufacture and use these boards in the consumer market, thus contributing to increase the recycling rate. Such a plan would bring economic, social, and environmental improvements to the region.

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