PHYSICAL AND MECHANICAL PROPERTIES OF RECYCLED PET COMPOSITES

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ABSTRACT

Virgin and recycled polyethylene terephthalate (PET) has been examined for the production of composites with additions of 5-20% by weight of sand particles. Density and compressive strength were estimated using virgin (V-PET) and recycled PET (R-PET). Scanning electron microscope (SEM) equipped with energy dispersive X-ray (EDX) spectroscopy was used to characterize the morphology and elemental composition of the composites. Also, thermogravimetric analysis (TGA) was used to find degradation temperature on both types of polymer. On the other hand, a streamlined life cycle assessment (SLCA) was made for the different composites to get environmental impacts. The results indicated that a maximum of 52.94 MPa and 52.03 MPa on compressive strength were obtained for virgin and recycled PET without sand, respectively. With the addition of sand, compressive strength decreases in both cases. The best performance was found at 5% sand addition, which causes a reduction of 9.07% and 16.68% for V-PET and R-PET composites, respectively. Environmental results show that resource extraction is the dominant life stage; meanwhile, gas residues are the dominant environmental impact in both types of composites. R-PET composites are the best environmentally friendly option because they used recycled material, which in return recovers part of the embodied energy used to make the primary production. The results show it could be explored the potential to be used the composites in pavement blocks or architectonic elements.

KEYWORDS

Polymer-matrix composites (PMCs), Mechanical properties, Compressive strength, Life cycle assessment

INTRODUCTION

One of the significant problems of our society is the increasingly growing demand for non-renewable materials such as plastics. The impact of plastics is higher due to its low degradation rates causing considerable environmental damage. In that sense, polyethylene terephthalate (PET) is a kind of thermoplastic material which has ample use in dairy activities as well in the industry. PET is a material cheap to produce and have the mayor recycled percentage between all polymers

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(~ 22%). In Colombia, a pollution problem exists due to the production of about 1500 million PET bottles by year, which reaches rivers, fields, and beaches or directly to landfill [1]. It is known that to degrade PET bottles is necessarily more than a hundred years [2]. For such reason, the use of recycled PET in different composites has been investigated for some time.

Tavares et al. [3] developed polymer concrete with silica sand bound to epoxy resin. Also, waste PET bottles have been used as fibres to produce such types of concretes [2,4]. However, the cost of producing concrete from such waste is still high [5]. Miranda et al. [6] produced mortar polymers using recycled PET and unsaturated polyester resin (UPER), showing that the best mechanical and physical properties of the composite were to mortars with ratio 78/20/2 sand/UPER/PET particles. Most studies regarding fracture toughness improving are focused on the melt mixing of virgin and recycle PET with rubber [7].

On the other hand, there are some studies on polymer matrix and sand particles. Kumi-Larbi Jnr et al. [8] use recycled LDPE water sachets to form sand blocks, and they found that the density and compressive strength of composites increases as the particle size of the sand decreases. Composite materials produced by PET wasted, and rigid sand has been studied concerning tensile properties. Zahran [9] found a decreasing linear function of tensile strength with the sand content and depending on the sand particle size. More studies have been performed on polymer composites with particles such as silica. The addition of silica particles to a polymer matrix has a positive effect on the elastic modulus of the composites [10,11]. Mohandesi et al. [12] investigated molten PET with silica sand particles in a 5-40% weight ratio at different composite temperatures. They found that the highest mechanical strength was obtained at 25°C and 10% sand above which composite strength decreases.

From the literature review is clear that limited research has been carried out on using waste PET and sand particles. The main aim of this study is to investigate the incorporation of sand particles on a PET matrix concerning mechanical properties and sustainability aspects, such as a rationalized life cycle assessment. Morphology was studied by scanning electron microscopy (SEM), thermal behaviour by thermogravimetric analysis (TGA), and compressive strength and density were determined for the composites. The use of these particles in the polymer has environmental advantages and also strength and friction enhancement that could lead to future applications on paving stones.

METHODS

Materials and composite preparation

PET produced by Codesarrollo corporation was used. The material was obtained from transparent bottles that had a pelletization process. Two types of composites samples were prepared, virgin PET (V-PET) of 1330 kg/m^3 and recycled PET (R-PET) of 1339 kg/m^3 reinforced with sand particles. PET was heated up to 260°C, and after 5 minutes of homogenization with continuous stirring, sand was added slowly to the molten polymer. Samples with 0%, 5%, 10%, 15%, and 20% by weight were obtained, and they were poured into cubic steel forms of 2 inches. Sand particles (2590 kg/m^3) were sieving, and the range material was selected between sieve # 30 (0.6mm) and #16 (1.18 mm) for all mixtures. Thermogravimetric analysis (TGA) was carried out in TGA Q500, V20.3 Build 39 for PET (10-20 mg) using a TGA Q500 in a nitrogen atmosphere at a flow rate of 90 mL/min from 20° to 900 °C.

Testing of specimens

Compressive strength was determined using a universal testing machine Instron 5582 equipped with a load cell of 100 KN, according to ASTMC109. The test was performed at 2 mm/min. A scanning electron microscope (SEM) with a JEOL model JSM2490 CV device equipped

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with energy dispersive X-ray (EDX) spectroscopy (OXFORD INCAPentaFET-x3) was used to perform morphological observations on selected composites samples. The accelerating voltage used was 20 kV.

Streamlined life-cycle assessment (SLCA)

An SLCA method developed by Graedel and Allenby [13,14] was followed to find out the environmental burden regarding life stages and environmental stressors. In practice, An SLCA measures the relative environmental impacts when searching for issues that occur during the life stages of a product [15]. The central feature of the assessment is a 5 x 5 matrix, with life-cycle stages in the rows and environmental stressors in the columns. According to Gradel, the assessor studies the product design, manufacture, packaging, use, and disposal scenario and assign to each element of the matrix an integer rating from 0 (highest impact, a very negative evaluation) to 4 (lowest impact, an exemplary evaluation) [13]. Because the approach is not quantitative, the results are not strictly a measure of environmental performance, but rather an estimate for improvement and comparison between products. The assignments of the numbers were made with a protocol exposed by Gradel [13].

RESULTS

Visual appearance

Figure 1a-b shows the visual appearance of the composite with and without sand particles. In the absence of sand particles, the composites exhibit defects indicating solidification problems during the process. In contrast, composites modified with sand particles exhibit fewer defects, although the distribution of the sand is not even. As the sand content increases, the location of the material seems to go to the bottom of the molds.

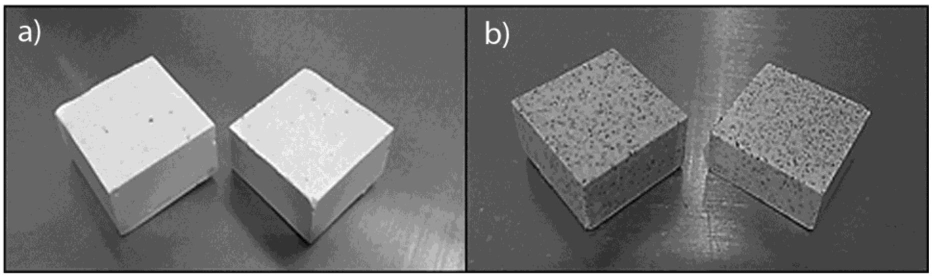

Fig. 1 – Visual appearance of R-PET samples without a) and with sand particles b)

Density

Figure 2 shows the variation of composite‘s density against sand percentage. Composite density for both, V-PET and R-PET, increases with sand content as is expected. Density ranges from 1280 kg/m^3 to 1530 kg/m^3 and from 1310 kg/m^3 to 1580 kg/m^3 for V-PET and R-PET composites, respectively. Polymer density without sand addition is within the range of values reported by the literature [16]. From the deviation of the experimental results, no statistical difference was found for the two different composites.

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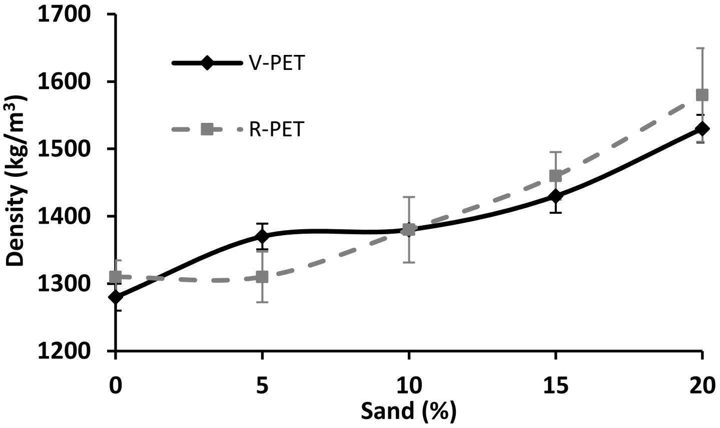

Fig. 2 – Composite density as a function of the sand content for V-PET and R-PET

Compressive strength

Figure 3a-b shows the average compressive strength of V-PET and R-PET for the different sand percentages. PET samples without sand exhibit an average compressive strength of 52.94 ± 2.54 MPa and 52.03 ± 10.1 MPa for virgin and recycled, respectively. The values are close to the reported in the literature [16,17]. The 5% of sand obtained the best performance in both cases, which result in decreasing compressive strength in 9.07% and 16.68% for V-PET and R-PET composites, respectively.

Both composites samples exhibit similar stress-strain curves, showing an initial linear behaviour, and further loading causes yield until it reaches the maximum compressive values. As can be seen, ductility decreases with the addition of sand reaching lower values in the case of R-PET composites. The data exhibits visco-elastic behaviour for higher sand content, as was observed on LDPE/sand composites [8]. Sand changes plastic flow under load, clearly reducing its ductility.

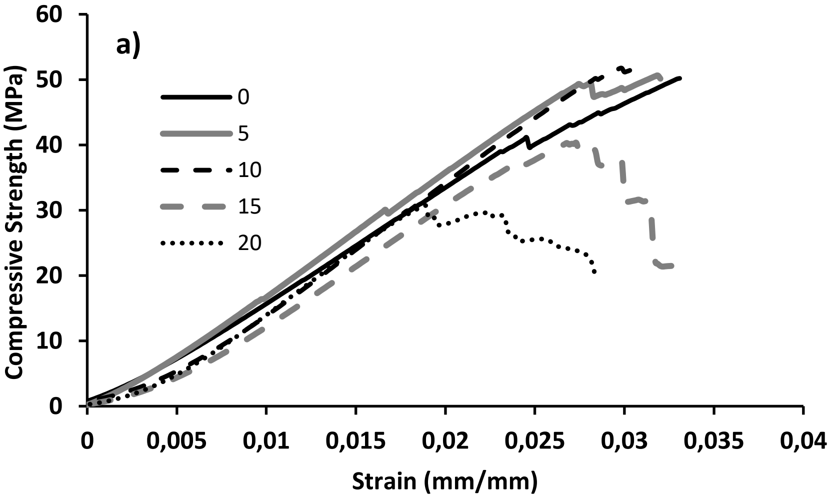

Fig. 3a – Stress-strain curves for V-PET

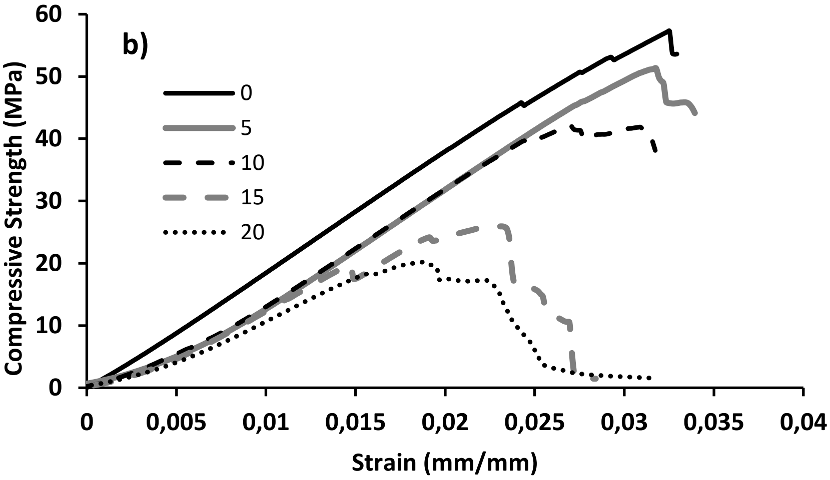

Fig. 3b – Stress-strain curves for R-PET for the different sand percentages

The average results of compressive strength for all sand percentages are presented in Figure 4. Due to the high variability of R-PET results, the values are pretty much the same compared to V-PET for percentage below 10%. At higher sand addition, the values decrease considerably. In both cases, composite's resistance decreases as increase sand content. These could be explained to the overpopulation of sand particles, which causes a weak interaction between sand and PET matrix and inefficiencies for stress transfer. The reduce in compressive strength has been seen on LDPE/sand composites for larger sand-size particles (> 1mm) [8]. Sand size has an influence on the mechanical properties of the composites due to an increase in porosity, reducing the encapsulating area by the binder. In this research, larger air voids were found in both types of PET. SEM results corroborate these (see below). Sand addition results in a slight reduction of elastic modulus for sand percentage above 10%, although the results give no statistically differences.

The fact that R-PET exhibits relatively excellent resistance, especially for sand content of 5% and 10%, is highlighted because of the presence of semicrystalline phases. Polymers with high crystallinity usually exhibit higher modulus, toughness, and tensile strength values [18]. On the other hand, the results present larger standard deviation values, especially for R-PET composites, probably due to some adhesion problems between sand particles and the polymer matrix coupling with difficulties with the manufacturing process, as can be seen in the SEM images shown below (Figure 8).

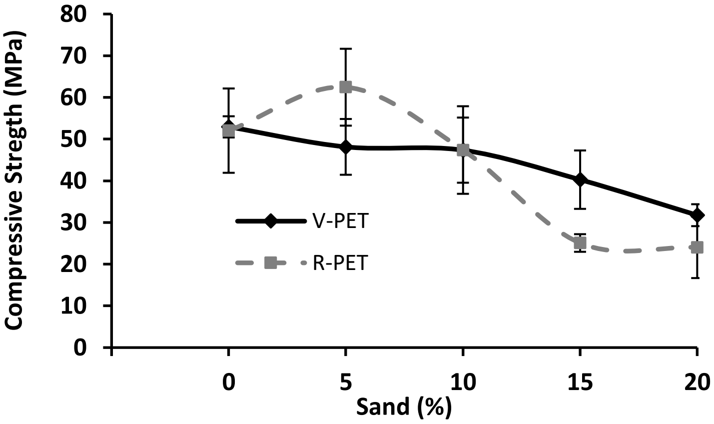

Fig. 4 – Compressive strength as a function of the sand content for V-PET and R-PET

Figure 5 exhibits the thermograms for V-PET and R-PET, respectively. V-PET maintains its properties until 400°C approximately, meanwhile R-PET starts to degrade a little bit earlier. Dimitrov et al. [19] reported 435°C and 434°C as temperatures of maximum decomposition rate for V-PET and R-PET, respectively; such values are above reported in this study. As can be seen in Figure 5, derivative mass loss indicates that the decomposition temperatures for V-PET and R-PET are 431.45 °C and 420.15 °C, respectively. As expected, R-PET starts to decompose at a lower temperature than V-PET due to the thermal history of the material and possible residual impurities present in the samples. The mass loss of PET is attributed to a polymer degradation process involving a random scission of ester links in the main chain resulting in the formation of different oligomers [20]. The curve shows 85 wt.% loss in the case of V-PET and 88 wt.% loss in the case of R-PET at their respective end set temperatures.

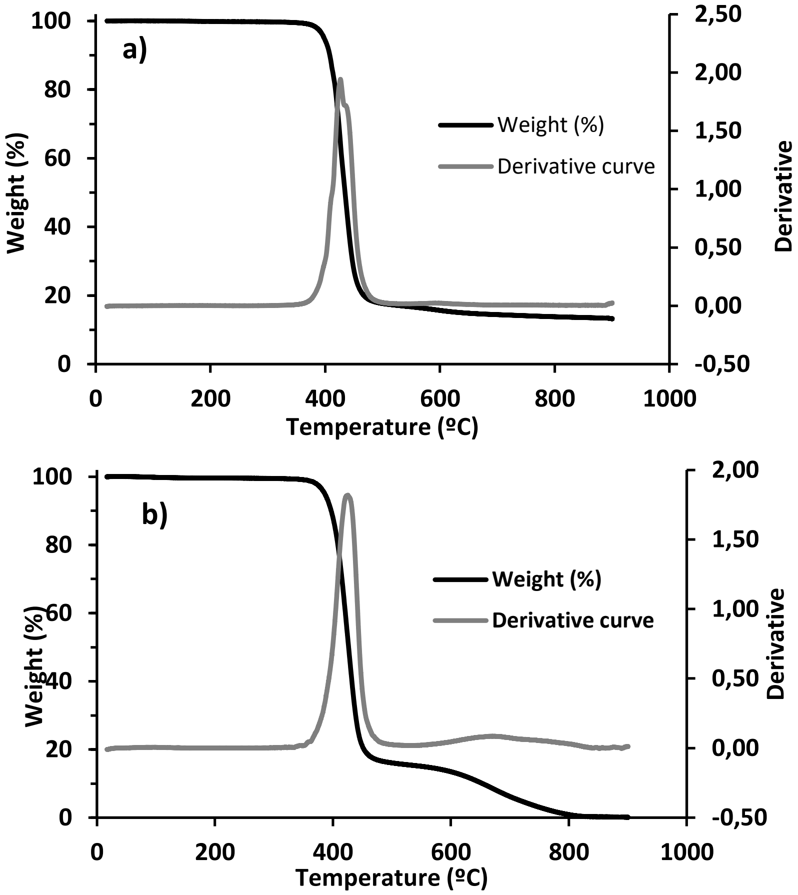

Fig. 5 – TGA thermograms of a) V-PET and b) R-PET

After a comparison of the two TGA curves, it is observed the apparition of a second plateau in the case of R-PET. This plateau is related to a second decomposition due to a reorganization of the crystals during the test. The effect has seen before, and the first plateau has been related to the melting of imperfect crystals and the second one to the fusion of the original crystals that suffer reorganizations, and they melt at higher temperatures [21].

SEM

Figure 6a and 7a show an SEM image of the V-PET and R-Pet composites, respectively. It can be observed that sand particles are mechanically bonded into the polymeric matrix. The adhesion mechanism could be related to a molecular bonding interaction due to sand surface irregularities [22,23]. However, as sand percentage increases, the adhesion is less effective. According to the energy-dispersive X-ray spectroscopy (EDX) (see Figure 6b and Figure 7b), carbon and oxygen were detected on the PET matrix for both virgin and recycled. On the other hand, magnesium, aluminium, silicon, potassium, and iron were detected on the sand particles (see Figure 6c and Figure 7c). The presence of potassium and magnesium could be due to contamination on the sand particles. Traces of iron and aluminium oxides are typical in this type of sand. According to EDX results, no impurities were found on the R-PET matrix.

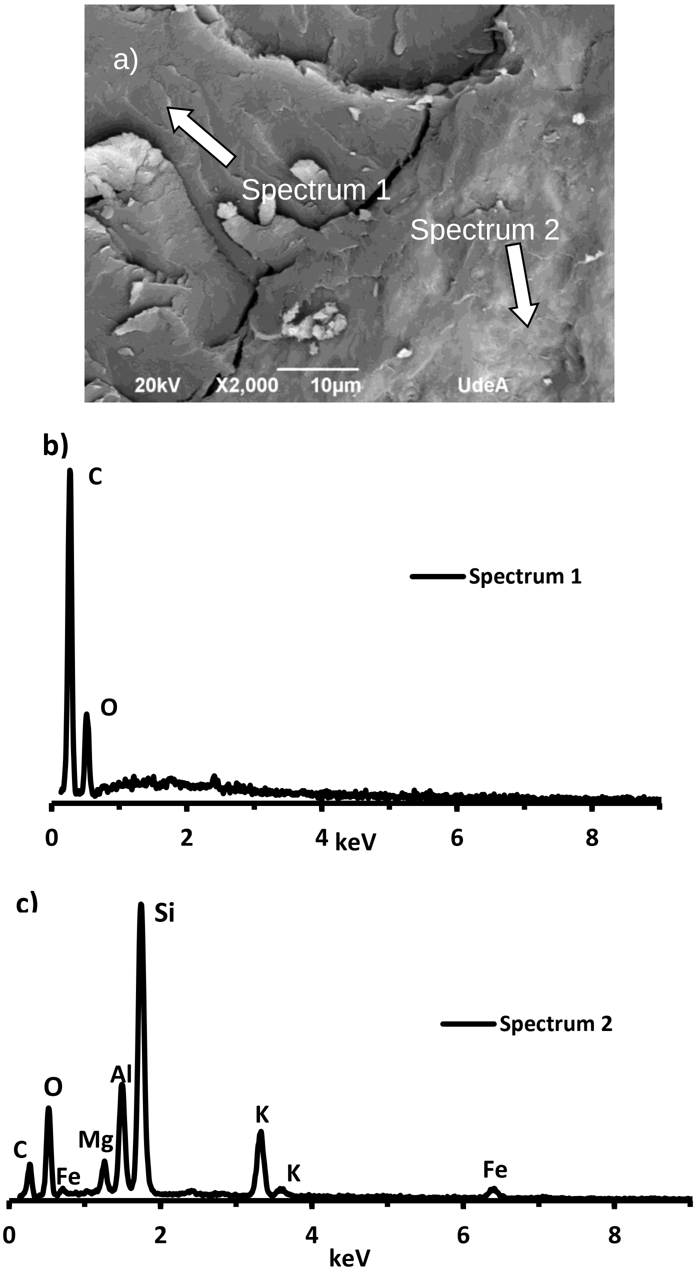

Fig. 6 – a) SEM micrograph of V-PET with 10% of sand particles and EDX results of two different local points within b) matrix and c) particles

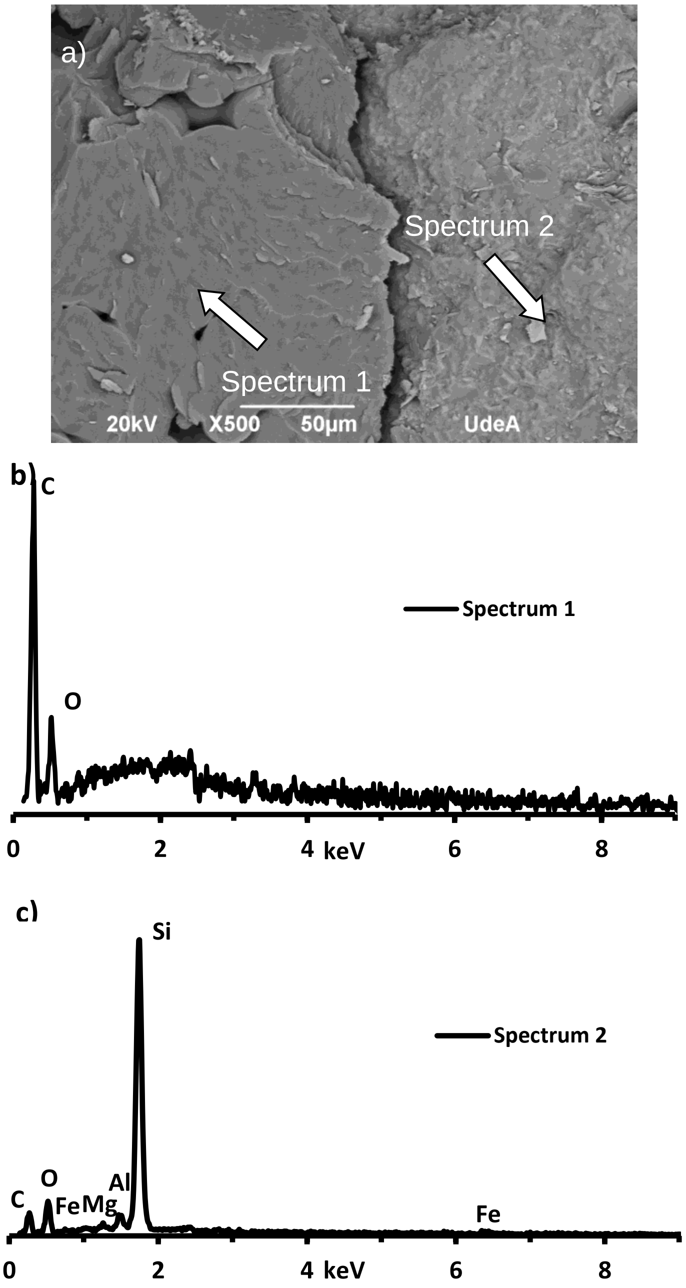

Fig. 7 – a) SEM micrograph of R-PET with 20% of sand particles and EDX results of two different local points within b) matrix and c) particles

Figure 8 shows SEM micrographs for V-PET and R-PET composites without sand. The morphology of the samples is very similar, exhibiting reproducible solidification process. Also, in both cases, porosity was present after the solidification. Those voids lead to more substantial variation in density and resistance. As can be seen in the figure, as the sand percentage increases, the cracking routes rise because of the poor adhesion between particles and the matrix. Samples without sand were free of porosity.

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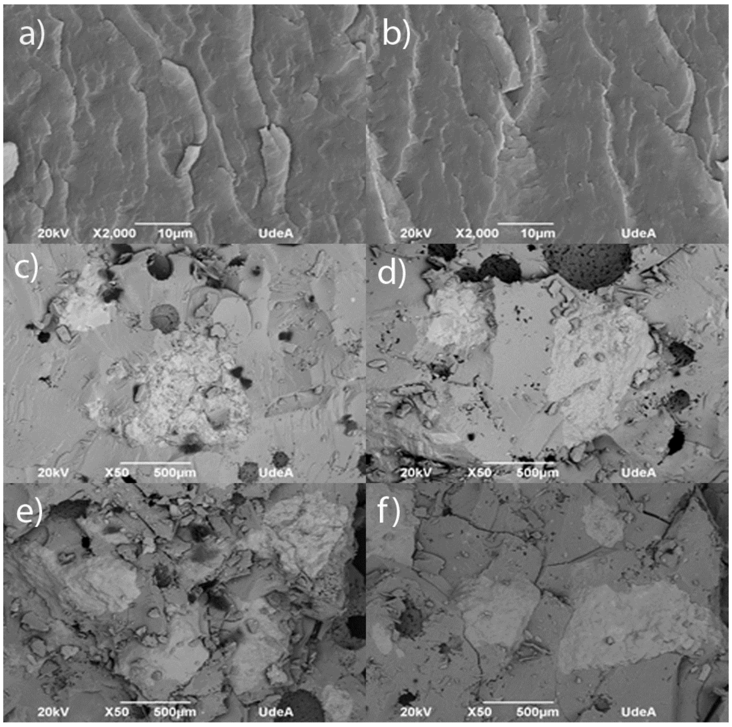

Fig. 8 – SEM micrographs for 0, 10, and 20% sand corresponding to a), c) and e) V-PET and b), d), and f) R-Pet respectively

Streamlined life cycle analysis (SLCA)

An SLCA of composites was performed to determine which scenario is present less environmental issues. The problem with PET is that in spite of its recycling possibilities, the actual amount of recovering of the material is still low. The usual disposal of the material is in a high deal landfill, which causes pollution by introducing solid wastes into the soil that are not easily degradable. PET is usually employed to manufacture water bottles and containers, but virgin o recycled pellets of PET can be used to obtain composites such as this study.

For the resource extraction life stage, it is considered virgin PET and recycled PET depending on the type of sample, so for virgin material, it produces more residues compared to the second scenario. Since PET is a petroleum-based material, the primary resource is non-renewable energy; also, the number of residues is the highest compared to the other life stages. The processing of PET is relatively easy to accomplish, allowing high speed with low temperatures. The main concern in this stage is due to the gaseous residues produced from the production plant, but the solid and liquid residues are minimal. For the delivery of the product, it is considered the low density of PET, which allows flexible and easy transportation. However, carbon dioxide (CO_2) emission has to be accounted for due to the use of fossil fuel during this stage.

The application of this type of composite could be related to the fabrication of polymer composites for pavement blocks. In that case, the blocks could be easy to carry and install. During its life, the product will produce no solid, liquid, and gas residues. The end of life of these composites could be recycling instead of the landfill, which causes solid and not degradable residues. For the separation process between the polymer matrix and sand particles, it will be used energy, and together with the transportation to the recycling facility, it will produce gas residues.

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In Table 1 and 2, is presented the SCLA analysis for composited made of V-PET and R-PET, respectively. The analysis was conducted through five life stages: resource extraction (RE), product manufacturing (PM), product delivery (PDe), product use (PU), and product disposal (PDi). For each stage, it was evaluated material choice (MC), energy use (EU), solid residue (SR), liquid residue (LR), and gas residue (GR) as the environmental stressors.

Tab. 1 - Streamlined LCA for V-PET

	MS	EU	SR	LR	GR	∑
RE	3	3	2	2	2	12
PM	3	2	3	3	2	13
PDe	4	3	3	3	2	15
PU	4	4	4	4	4	20
PDi	3	2	3	4	2	14
∑	17	14	15	16	12	74

Tab. 2 - Streamlined LCA for R-PET

	MS	EU	SR	LR	GR	∑
RE	4	4	3	3	3	14
PM	3	2	3	3	2	13
PDe	4	3	3	3	2	15
PU	4	4	4	4	4	20
PDi	3	2	3	4	2	14
∑	18	15	16	17	13	79

After the evaluation, the overall environmentally responsible product rating (R_{erp}) is calculated as the sum of the matrix element values as is depicted in Equation 1 [13]:

$$R_{erp} = \sum_{i=1}^{n} \sum_{j=1}^{n} M_{i,j} \qquad (1)$$

where Mi,j corresponds to each element of the matrix.

R-PET has a R_{erp} value of 79, which is better compared to the V-PET with a R_{erp} value of 72. However, both scenarios are excellent choices in terms of environmental impact [24].

A more succinct display of the SLCA results could be made using target plots using each element of the matrix. Figure 9 shows the target plots corresponding to V-PET and R-PET. As can be seen, values for R-PET composite have more dots closed to the center, indicating a better product compared to V-PET.

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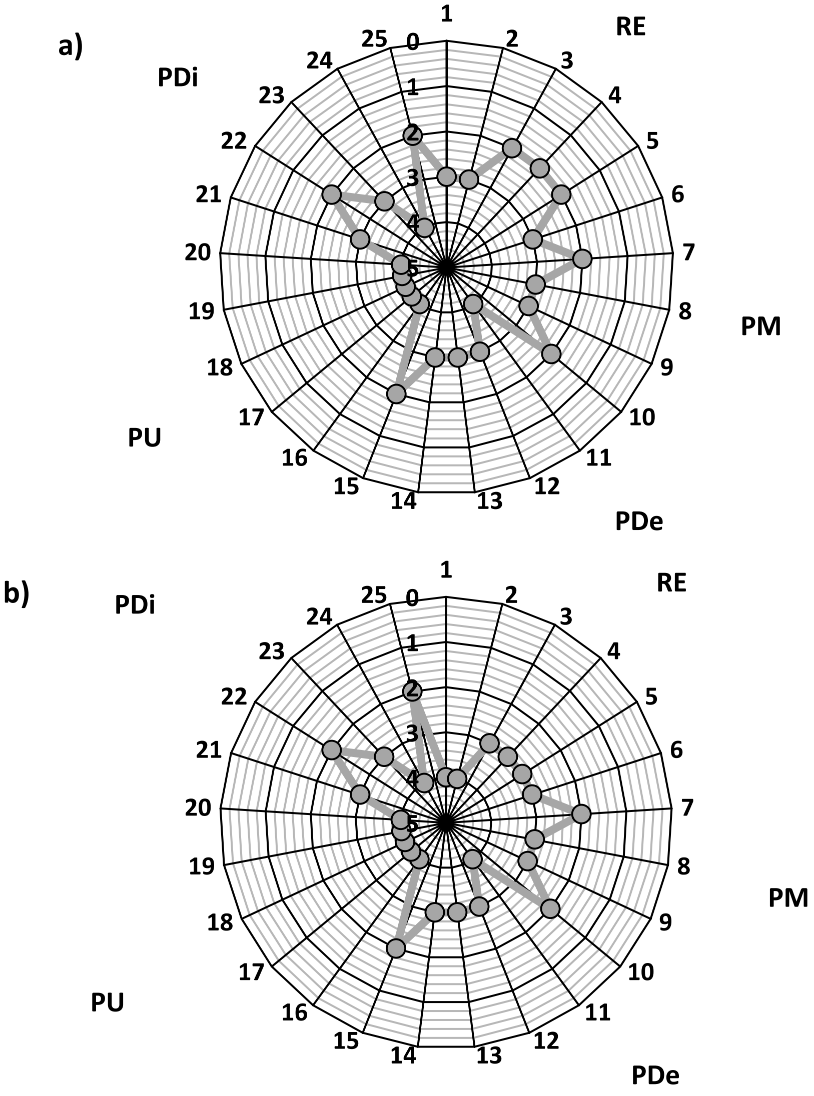

Fig. 9 – Target plots corresponding to a) V-PET and b) R-PET

Both composites exhibit gas residues as the dominant environmental impact. These are because of the manufacturing, transport, and separation process at the end of life. The separation of PET and sand requires the melting of the polymer and then using non-renewable energy.

Resource extraction is the dominant life stage in the case of V-PET, due to the high impact of the petroleum base material as the primary resource, which becomes an impact depleting the non-renewable fossil energy. Besides, during this stage, the amount of solid, liquid, and gaseous residues are highest compared to the other phases. Product manufacturing has a definite impact concerning gaseous residues produced as well as energy use for the polymer processing. Product manufacturing is the dominant life stage in the case of R-PET, due to the energy and generation of residues during the fabrication of the elements. Resource extraction for R-PET is reduced in the benefit of the recycled material.

Product delivery has a low impact due to the low density of the material, which results in a reduction of lightweight energy consumption. However, it has to take into account the release of gaseous pollutants during transport. Use stage in both types of materials have a low impact due to the low cleaning services, and it is expected to produce a meager amount of solid residues in any chosen application. The disposal phase has a moderate impact in both cases considering its mechanical recycling route. A recycling strategy can significantly reduce environmental burden in terms of solid waste and hydrocarbons emissions [25]. However, when the product has an incineration route, it will yield heavy metals to the environment [24].

CONCLUSION

Based on the results obtained, the following conclusions can be drawn:

1. Compressive strength decreases with sand content. These are associated with poor adhesion between particles and polymeric matrix and porosity generated within the composites, as was observed in SEM micrographs, causing a low transfer of stresses. Sand addition achieves better performance at 5%, causing a reduction of 9.07% and 16.68% for V-PET and R-PET composites, respectively.

2. According to TGA results, decomposition temperatures for V-PET and R-PET are 431.45 ºC and 420.15 ºC, respectively. However, during this study, this limit was not reached in any case, indicating the viability to fabricate these composites.

3. Recycled and virgin PET as a matrix is good choice in terms of environmental impact. R-PET has a R_{erp} of 79 and V-PET a R_{erp} of 72. R-PET composite has a less harmful environmental evaluation because it used recycled material, which recovers part of the embodied energy used to make the primary production. However, both cases require the use of non-renewable energy to separate composite materials.

4. According to physical and mechanical properties obtained, it could be explored the used of the R-PET in pavement blocks or architectonic elements.

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