REVISTA MATÊRIA V.26 N.03
ISSN 1517-7076 artigos e13009, 2021

Potential use of PET and PP as partial replacement of sand in structural concrete

Uso potencial de PET e PP como substituição parcial de areia em concreto estrutural

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ABSTRACT

The use of polymeric residues in the civil construction has been the target of many studies aiming to reduce the volume of post-consumer plastics in the environment. This work focuses on the viability to use polyethylene terephthalate (PET) and polypropylene (PP) as partial replacement to sand in concrete. PET and PP flakes from post-consumer packings were used as light aggregate to partially replace, individually, 10% in volume of sand. The effect of adding these polymers was investigated in terms of physical, mechanical, durability and morphological properties of the concrete. Physical properties were measured in terms of water absorption, voids content and specific mass. Mechanical properties were measured in terms of compressive strength and elasticity modulus. Durability properties were measured in terms of capillarity water absorption and electrical indication of the concrete to resist to chloride ion penetration. MEV and EDS were used to carry out morphological analysis. DSC curves were carried out to evaluate thermal properties of the polymeric flakes. Contact anlge test was also performed. The partial addition of PET and PP polymers reduced the compressive strength by 20%, whilst the reduction of the elasticity modulus was 16% for PET samples, and almost insignificant for PP samples. The durability results show that the polymers contributed to increase the resistance of the samples to chloride penetration by 15% and 57%, for PET and PP samples, respectively; however, there was an increase in the voids content and water absorption. In the morphological test it is possible to observe a lower interfacial adhesion between PP and the cementions paste in comparison to PET.

Keywords: Post-consumer plastic, PET, PP, concrete with residues

RESUMO

O uso de resíduos poliméricos na construção civil tem sido alvo de muitos estudos com o objetivo de reduzir o volume de plásticos pós-consumo no meio ambiente. Este trabalho concentra-se na viabilidade do uso de politereftalato de etileno (PET) e polipropileno (PP) como substituição parcial da areia no concreto. Os flocos de PET e PP pós-consumo foram utilizados como agregado leve para substituir parcialmente em 10% o volume de areia. O efeito da adição desses polímeros foi investigado em termos de propriedades físicas, mecânicas, de durabilidade e morfológicas do concreto. As propriedades físicas foram a absorção de água, teor de vazios e massa específica. As propriedades mecânicas das amostras foram medidas em termos de resistência à compressão e módulo de elasticidade. A durabilidade foi avaliada em termos de absorção de água por capilaridade e indicação elétrica do concreto para resistir à penetração de íons cloretos. A análise morfológica foi realizada pelo MEV e EDS. Curvas de DSC foram utilizadas para avaliar as propriedades térmicas dos flocos. O-teste de ângulo de contato também foi realizado. A adição parcial de polímeros de PET e PP reduziu a resistência à compressão em 20%, enquanto a redução do módulo de elasticidade foi de 16% para as amostras de PET e quase insignificante para as amostras de PP. Os resultados de durabilidade mostram que os polímeros contribuíram para aumentar a resistência das amostras à penetração de cloretos em 15% e 57%, para amostras de PET e PP, respectivamente, no entanto, houve um aumento no teor de vazios e na absorção de

Autor Responsável: Priscila Marques Correa   Data de envio: 15/10/2019   Data de aceite: 18/05/2020

10.1590/S1517-707620210003.13009

água. No ensaio de morfologia é possível observar uma menor adesão interfacial entre o PP e a massa cimenticia em comparação ao PET.

Palavras-chave: Plástico pós-consumo, PET, PP, concreto com resíduos.

1. INTRODUCTION

One ton of concrete is produced annually for each inhabitant of the world, which highlights the environmental impact caused by the production of concrete [1]. In Brazil, the production of concrete is responsible for 15 to 50% of the natural resources' consumption, which leads to a yearly consumption of approximately 220 million tons of sand only for concrete production [2, 3].

The high cost and shortage of sand in some areas of Brazil has turned the recycling of polymeric residues into an interesting option to replace the natural resource. This is especially because polymeric commodities are largely produced and used [1, 2] so the amount of recyclable post-consumer plastics is abundant all over the world. The use of polymeric residues by the civil construction may be an effective alternative to reduce the environmental impact caused by polymer discarding and to the development of greener materials.

Many authors studied the use of polymeric residues, such as polyethylene terephthalate (PET) [1-5] and polypropylene (PP) [1, 6, 7] processed as fibres and flakes for concrete production.

The author FRIGIONE [2] studied the substitution of 5% of fine aggregates derived from PET bottle milling. The results show that the incorporation of PET reduces the compressive and tensile strength of the concrete and presents a lower modulus of elasticity.

ALBANO *et al.* [8] investigated the mechanical behavior of concrete with the incorporation of PET residue particles in which the replacement contents (in volume) were 10% and 20% and the particle size of PET ranged from 0.26 cm and 1.14 cm. The results show a decrease in the compressive strength, tensile strength and modulus of elasticity of the concrete mixture.

The use of polymers as fibres for concrete has been the target of many studies in the last years [9-11]. However, the fibre production process requires extra processing stages such as wiring and extrusion, which leads to the high cost of the final product [12, 13].

Previous studies based on the partial replacement of sand by PET show that there is a decrease on the compressive and flexural strength and an increase on the water absorption of the samples with the inclusion of polymers [14, 15]. Studies based on the use of polymeric flakes in concrete are rare when compared to the use of fibres. On this context, this research aims to evaluate the use of PET and PP as flakes, obtained from post-consumer food packaging, as partial replacement of sand, in terms of physical, mechanical, durability and morphological properties of structural concrete. Thermal and contact angle tests were also performed in PET and PP samples to contribute to the analyses of the concrete properties.

2. MATERIALS AND EXPERIMENTAL PROGRAM

2.1 Materials

Three mixture proportions were studied in this research: one mixture proportion as reference without residues, and two mixes with polypropylene (PP) and polyethylene terephthalate (PET) flakes residues, added separately, as partial replacement of sand in the amount of 10% by volume. The mixture proportions are presented in Table 1. Reference mixture was chosen due to the fact that it is a mixture commonly used in regular civil construction and complies with the minimum requirements for structural concrete according to Brazilian standard for concrete design procedures, ABNT NBR 6118 [16]. The use of flakes in the amount of 10% by volume was based on a previous study with PET and PP flakes in different contents [14] that evaluated the gains in terms of mechanical behavior and environmental viability. RAHMANI *et al.* [17] also showed that a concrete with 10% fine aggregates volume replaced by PET particles has the same results than concrete without PET, which justifies the choice of the 10% content in this study.

Table 1: Mixture proportions.

Sample	Cement (kg/m³)	Fine aggregate (kg/m³)	Coarse aggregate (kg/m³)	PET and PP post-consumer flakes (kg/m³)	Water/cement
Reference	400	800	1200	-	0.52
PET	400	720	1200	41.34	0.52
PP	400	720	1200	27.56	0.52

High-early-compressive-strength Portland cement was used in the research. The cement is classified as type V according to NBR 16697 [18] and ASTM C150 (2019), with resistance class of 46.8 MPa at 28 days, surface area of 1.71m²/g and specific mass of 3.16 g/cm³. The sand and gravel used in the research are of quartz and basaltic origin, respectively. The polymeric aggregates were obtained from post-consumer food packages and were ground in a knife mill.

Table 2 shows a summary of the physical properties of the natural and polymeric aggregates in terms of specific mass and specific surface area, the latter obtained by the BET (Brunauer, Emmett, Teller) method.

Table 2: Physical properties of aggregates.

Aggregates	Specific mass (g/cm³)	Specific surface area (m²/kg)
Coarse	2.88	-
Fine	2.60	2.60
PET	1.38	3.35
PP	0.90	2.88

The gradation curves for sand, PET and PP are shown in Figure 1

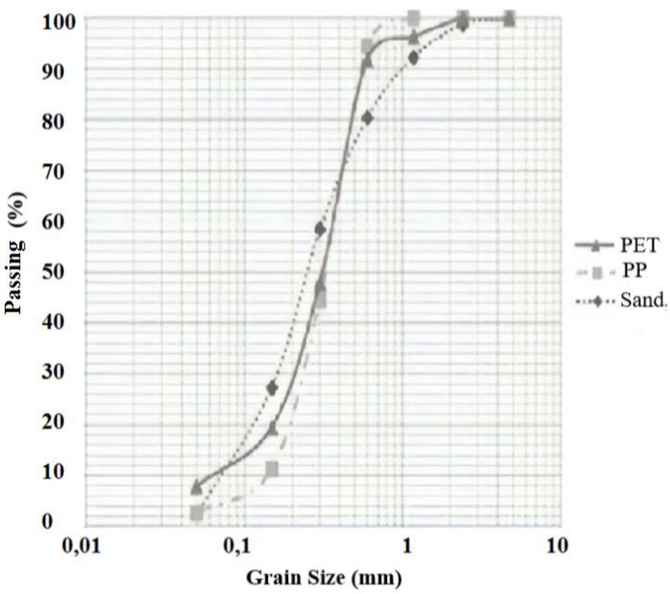

Figure 1: Gradation curves of aggregates.

2.2 Research method

The research was developed aiming to initially characterize the polymers in terms of thermal analysis and contact angle. The concretes with post-consumer polymers were analysed in terms of physical, mechanical, durability and morphological properties. Thermal analysis with DSC (Differential Scanning Calorimeter) curves was carried out in the polymers to identify a possible degradation of polymers. The contact angle analysis was performed to measure the wettability of the polymers by a liquid.

The physical properties of hardened concrete were the void content, specific mass and water absorption, while the slump test was carried out for fresh concrete. The mechanical properties were the compressive strength and modulus of elasticity. The durability properties were the water absorption by capillarity and resistance to chloride ion penetration. The Morphological analysis was carried out by SEM (Scanning Electron Microscopy) and DES (Dispersive Energy Spectroscopy) tests.

The equipment used to perform DSC test is from TA Instruments, Q20 model. PP specimens were subjected to a heating ramp from 25ºC to 200ºC, whilst the PET specimens were subjected from 25ºC to 300ºC. The heating rate was 10ºC/min in inert N_2.

The contact angle measurements were performed on a Rame-Hart model goniometer 100-00, according to ASTM D 724 [19]. Software OCA 20 was used to determine the contact angle. The liquid used for the sessile drop test was deionized water with a pH of approximately 7.0. For each sample, 10 drops were made and each drop was photographed for further analysis

Cylinders measuring 100 mm in diameter and 200 mm in height were cast to measure the specific mass, water absorption and void content (physical properties), and also for mechanical properties. To evaluate the behavior in terms of durability, cylinders measuring 100 mm in diameter and 50 mm in height were used, which were cut from 200 mm in height cylinders. The casting procedure for all specimens followed ABNT NBR 5738 [20], corresponding to the ASTM C192 (2016). Specimens were compacted in two layers using a vibrating table. Three specimens were cast for each mixture proportion and each property to be evaluated (exception applies for specimens used for physical properties, since the same three specimens were used to measure all the properties within this group). The tests were performed at 28 and 90 days aiming to verify whether there is any property change that indicate degradation of the polymers after being exposed to the alkaline environment of the concrete.

The tests for physical, mechanical and durability properties of concretes with polymers were performed according to the Brazilian Association for Technical Standardization (Associação Brasileira de Normas Técnicas – ABNT), except the test to determine the concrete resistance to chloride ion penetration, which was carried out according to the ASTM C1202 [21]. Table 3 describes the main differences between the Brazilian and the correspondent ASTM Standard, since ASTMs are most commonly used and accessed worldwide in comparison to the Brazilians standards

Table 3: Main differences between ABNT and ASTM standards used in the research.

Properties	Test	ABNT	Corresponding ASTM standard	Difference between Standards
Physical	Specific mass, water absorption and void content	NBR 9778 [22]	ASTM C642 (2013)	Procedure of testing
	Slump test	NBR NM 67 [23]	ASTM C143 (2010)	No difference
Mechanical	Compressive strength	NBR 5739 [24]	ASTM C39 (2009)	Speed loading of 0.45 MPa/s for ABNT and 0.25 MPa/s for ASTM
	Static modulus of elasticity	NBR 8522 [25]	ASTM C469 (2014)	Elastic behaviour counted until 30% of the ultimate stress for NBR and 40% for ASTM
Durability	Water absorption by capillarity	NBR 9779 [26]	ASTM C1585(2013)	Procedure of testing and testing time

For the morphological analysis, the instrument used in this study was a standard Scanning Electron Microscope (SEM) Philips model XL30, and the test was performed in small samples of concrete (measuring approximately 3 cm wide), which were obtained from former cylinder specimens. The analysis was carried out especially at the interface between the polymer and the cementitious paste.

3. RESULTS AND DISCUSSIONS

3.1 Thermal properties of polymers

This test aims to measure the possible hydrolytic degradation of PET and PP in an alkaline environment, which can lead to durability and mechanical problems of concrete in case the degradation-will be confirmed. Figure 2 shows the DSC thermal curves of PET and PP samples before (PET B and PP B) and after (PET A and PP A) 90 days of concrete curing.

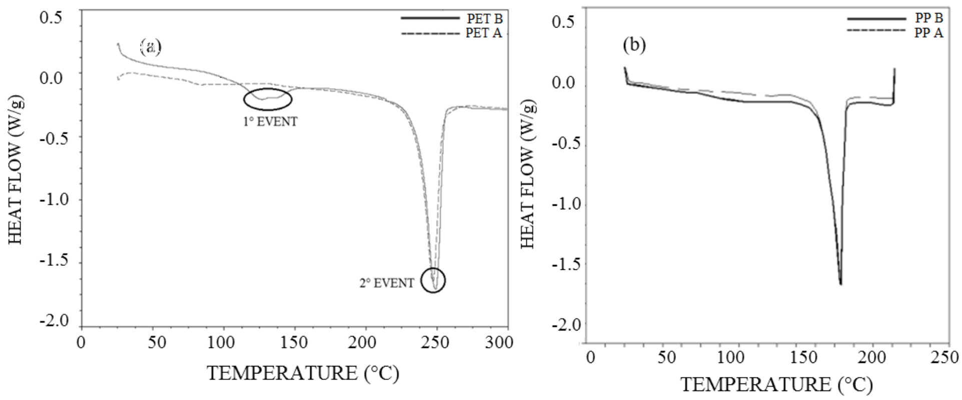

Figure 2: Thermal curves of DSC of the samples: (a) PET and (b) PP before and after 90 days of concrete curing.

For PET sample, in Figure 2(a) it can be observed two endothermic events, the first at 80-100ºC interval, that can be attribute to the fusion process from hydrated crystals, probably the calcium hydrate. The interface formed with a cementitious matrix and this alkaline medium can favor the hydrolytic degradation of PET [27, 28] and the second at 250ºC, due to fusion temperature of PET. PET B sample does not present the first event, probably because many hydration reactions have not occurred yet. PET A sample presented 2 events after 90 days, which may be an indication of the formation of crystals, increasing the alkalinity of the matrix, and probably causing to PET the beginning of an alkaline hydrolytic degradation after being in contact to cementitious mass for all this period. Similar results were also observed by another author [29]. In the case of PP, it can be observed in Figure 2(b) that there were no changes in the fusion peak format when PP was added to the cement matrix. This behaviour may be an indication that PP is inert to the alkaline environment.

Some authors noted that PET degradation in the alkaline environment is clearer observed after 150 days of contact to this environment [30, 31], which may lead to the rapid strength decrease of concrete, due to PET hydrolysis in the alkaline environment, thus causing higher void contents and reduction of durability and mechanical properties. ROUHOLLAH *et al.* [32] and AZAD *et al.* [33] also concluded that PET fiber degrades in alkaline environment, even though PET is resistant to weak acids.

3.2 Contact angle

Figure 3 shows the images of water droplets on the surface of PET and PP samples. It is observed that the PP samples are more hydrophobic than the PET samples due to the smaller water droplet spread of PP sample on the surface of the sample when compared to the droplet shape on the surface of the PET sample, shown in Figure 3(a). The higher droplet spreading observed in PET substrate is attributed to the higher polarity of its chemical structure when compared to PP, which only presents C-H bonds (apolar).

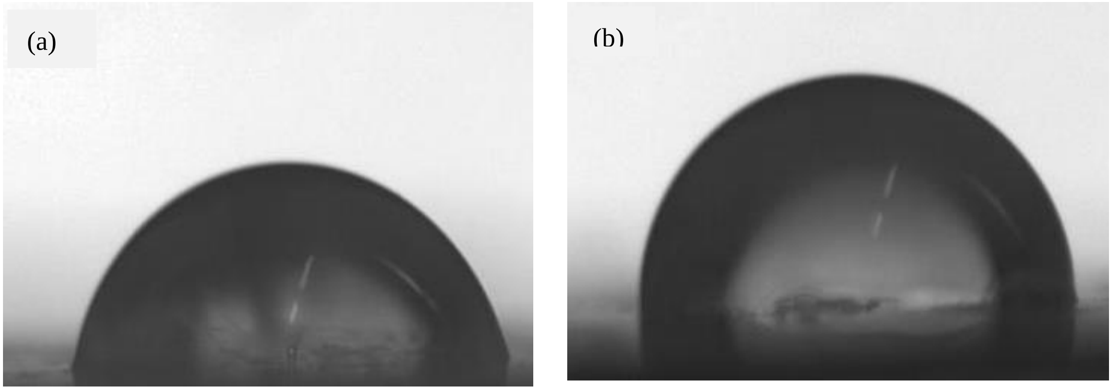

Figure 3: Water droplets on polymeric substrate: a) PET and b) PP

Table 4 shows that the contact angle of both samples differ from each other. These results resemble those of STROBEL *et al.* [34], whose contact angles for the PET sample were 70°. WANKE *et al.* [35] evaluated the contact angle of PP samples before applying a surface treatment, and the result was a contact angle of 90°, close to the results shown in Table 9, thus presenting a hydrophobic characteristic.

Table 4: Contact angle of PET and PP.

Sample	Contact angle (°)
PET	68.8 ± 0.78
PP	94.1 ± 0.83

3.3 Physical properties

The results of the physical properties in terms of water absorption, void content and specific mass of the hardened concrete (average and standard deviation), evaluated at 28 and 90 days of curing, are shown in Table 5. The table also presents the results for the slump test of fresh concrete.

Table 5: Physical properties of concrete

	Reference		PET		PP	
	28 days	90 days	28 days	90 days	28 days	90 days
Water absorption (%)	2.90 ± 1.10	2.50 ± 0.97	5.40 ± 1.60	5.70 ± 1.63	7.40 ± 0.70	7.10 ± 0.60
Void contente (%)	8.93 ± 1.47	7.11 ± 1.33	11.2 ± 1.16	12.58 ± 0.71	15.62 ± 1.36	15.34 ± 1.21
Specific mass (g/cm³)	2.32 ± 0.21	2.39 ± 0.32	2.29 ± 0.19	2.33 ± 0.24	2.25 ± 0.18	2.29 ± 0.21
Slump (mm)						
Reference: 100		PET: 85		PP: 125		

The addition of PP to fresh concrete increased the workability of the concrete probably due to the hydrophobic characteristic of PP, which repels water from its surface. Opposite behavior was observed when PET was added to concrete, probably due to its hygroscopic characteristic [14].

BATAYNEH *et al.* [36] investigated the effect of adding plastics (20% as sand replacement) into fresh concrete. The slump test results show a 25% drop in the workability compared to the reference mixture. The authors concluded that this effect is due to the shape and surface area of the polymer flakes. This may also be the reason for the differences between concrete with PET and PP in the slump test results of this study.

As expected, there is a reduction on the water absorption and void content as ageing occurs due to cement hydration. PP and PET samples have higher water absorption and void content when compared to reference mixture, even though the specimens with PET presented lower void content in comparison to PP specimens. This behaviour is probably due to the polar chemical structure of PET, which creates a better interaction between the polymer and the cementitious mass [14] even though the specific surface area of PET is higher than PP. The increase in the water absorption and void content for PET mixture, and also partially for PP, is believed to be caused by the fact that the flakes have a non-spherical geometry in comparison to sand, which may contribute to the entrapment of air during mixture.

The reduction of the specific mass of concrete due to polymer addition is due to the following factors: firstly, the lower specific mass of the polymers when compared to natural sand, and secondly, to the increase of the void content as described previously. Similar behaviour was also observed in other studies [37- 40].

3.4 Mechanical properties

Table 6 shows the results (average and standard deviation) for compressive strength and modulus of elasticity at 28 and 90 days

Table 6: Results of compressive strength and modulus of elasticity

	Reference		PET		PP	
	28 days	90 days	28 days	90 days	28 days	90 days
Compressive Strength (MPa)	36.46 ± 0.42	42.47 ± 0.99	28.85± 1.38	33.23± 1.47	28.29± 0.19	34.33± 0.14
Modulus of Elasticity (GPa)	33.35± 0.07	31.64± 2.19	26.11± 0.01	26.59± 1.41	33.65± 0.19	33.52± 0.31

As expected, the results show a decrease on the values of the compressive strength as the post-consumer polymers are added to the mixture. This behaviour complies with other similar studies [26, 28]. The authors observed that the reduction on the compressive strength of concrete is due to the lower specific mass and strength of the residues.

Other authors observed that the reduction on the compressive strength when polymers are added to the concrete may also be due to the increase of the void content due to the porous interface observed between the cementitious matrix and the polymeric flakes [41, 42] and to the lower bond between cementitious matrix and the polymers when compared to conventional fine aggregate [17]. It can also be observed a slight increase in the compressive strength from 28 to 90 days, which is expected due to ageing of concrete.

For the results of the elasticity modulus, it can be observed a reduction on the values for PET samples when compared to reference mixture, probably due to the lower modulus of elasticity (2,76-4,14 GPa) and lower specific mass of the polymer 1,38 g/cm³ [43, 44]. This reduction on the elasticity modulus was also observed in other studies [16, 33, 45]. On the other hand, PP mixture proportion presented values of modulus similar to reference concrete. In comparison to PET, PP is a more crystalline polymer [42] which leads to an increase in the ductility of the concrete. It is believed that the lower modulus of elasticity of the polymers influences more the reduction on the modulus of elasticity of concrete than the lower specific gravity of polymers. The values of modulus of elasticity were similar at 28 and 90 days, as expected.

Figure 4 shows the images of the polymer flakes obtained through the Optical microscope with polarized light with image analyzer Carl Zeiss Axioscope A1 e Hotstage Linkam (T95; LNP) where it is possible to observe that the PP surface is less rough than PET.

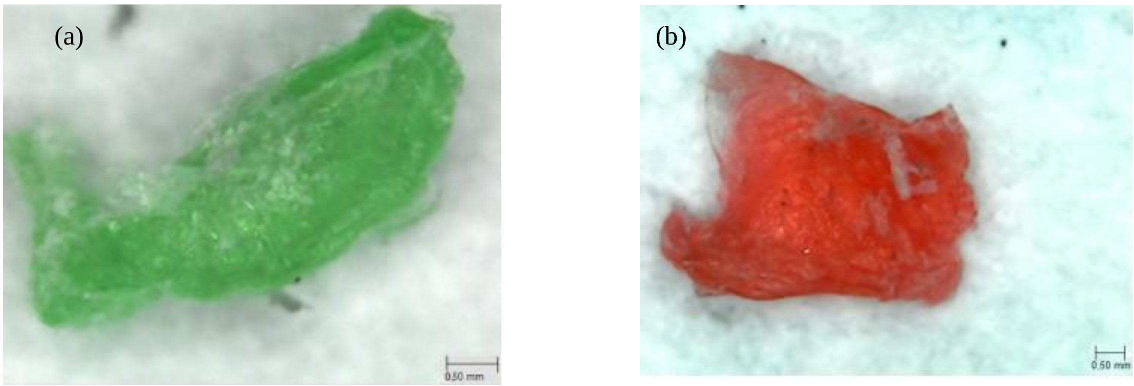

Figure 4: Images of cementitious matrix fracture surface with: a) PET and b) PP post-consumer flakes.

3.5 Durability properties

Tables 7 shows the results of capillary absorption and electric passing charge (average and standard deviation) for reference, PP and PET specimens at the age of 28 and 90 days.

Table 7: Capillary absorption and electric passing charge

	Reference		PET		PP	
	28 days	**90 days**	**28 days**	**90 days**	**28 days**	**90 days**
Capillary absorption (g/cm²)	0.54 ± 0.28	0.50 ± 0.32	0.44 ± 0.39	0.31 ± 0.18	0.54 ± 0.17	0.52 ± 0.31
Electric charge (kC)	7.78 ± 1.02	6.92 ± 0.98	6.77 ± 0.52	5.89 ± 0.48	3.24 ± 0.86	2.98 ± 0.79

For the capillary absorption test, PP and reference specimens presented similar results, both higher than PET specimens. This is probably due to the fact that PP does not bond efficiently to the cementitious mass. Moreover, the results also suggest that the pores of the cementitious mass are probably to be connected among them, which contributes to the water penetration through the capillary pores. For PET specimens the reduction on the water capillary absorption is probably due to a better packing of the materials and the stronger bond of the polymer to the cementitious paste, which can be observed by the SEM analysis, described in item 3.5.

For the electric passing charge results, the addition of polymeric materials reduced the amount of electric charge in the concrete, when compared to reference samples. This behavior occurs because the polymers have the effect of reducing the load passing into the concrete. It can be noted that the addition of PP contributed to a higher electric resistance than PET, which is probably because the flakes may be acting as a barrier due to the polar characteristic of the polymer, thus making the flow of chlorides more difficult [29], as shown in the schematic figure 5.

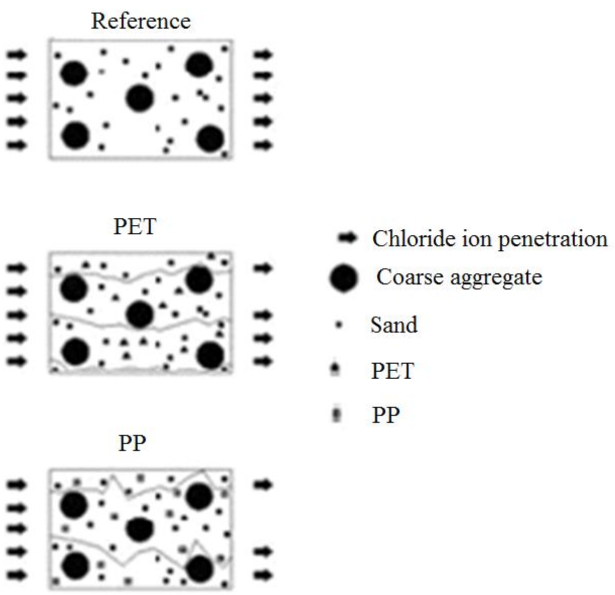

Figure 5: Proposed mechanism of chloride ion penetration into the concrete mixtures.

It is important to mention that the test proposed by ASTM C1202 [21] is an empirical test that gives only an indication of the concrete resistance to chloride ion penetration. The test is used mainly for comparison purposes and the results need to be careful analysed since the temperatures during test approached 60°C and may have influenced the results. Ideally, the determination of the chloride diffusion coefficient would be the best alternative.

3.6 Morphological analysis

The analysis by dispersive energy spectroscopy (DES) was evaluated to determine the chemical composition of the samples, mainly in the polymer interface and the cement paste. Samples obtained from each of the three mixtures, measuring 2 cm in diameter, were analyzed using a SEM Jeol LV5800 equipped with DES Thermo Noran. The results of the analysis using SEM and DES are shown in Figure 6 and Table 8. Figure 6(a) presents the SEM images of the surface of concrete sample containing PET, where a rough surface of the polymer can be noted, indicating a good adhesion between PET and the cementitious mass. The DES analysis carried out at PET shows the presence of possible deposition of calcium hydroxide after 90 days of curing favoured by the alkaline environment due to hydration reactions.

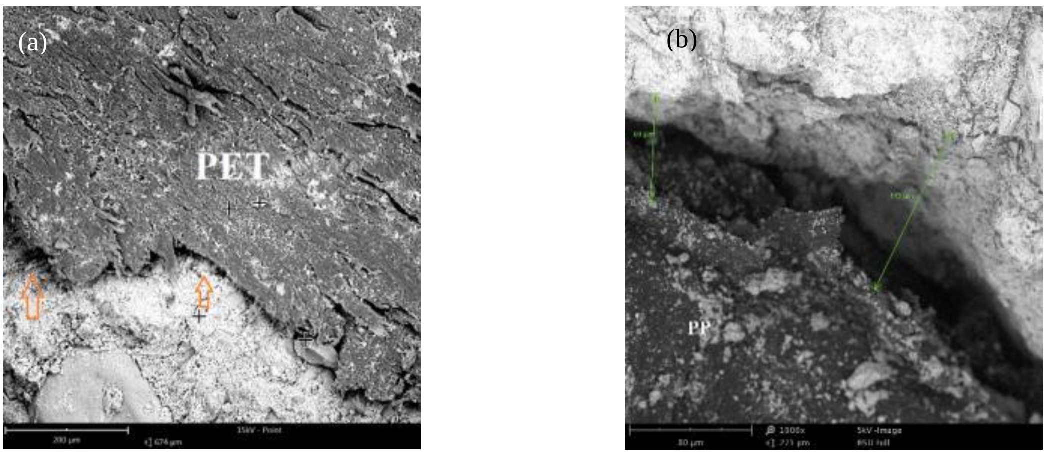

Figure 6: SEM images of surface of concrete with the aggregates: a) PET and b) PP.

Table 8: Chemical composition at the surface of polymers obtained by EDS.

Chemical composition (% mass)					
Sample	**C**	**O**	**Ca**	**Si**	**Sb**
PET	44.5	38.3	11.1	1.7	4.5
PP	53.1	34.3	5.8	3.7	-

BRAHIM *et al.* [46] performed similar analysis using SEM images taken from the interfacial zone between PET flakes and concrete matrix. They observed a poor adhesion of PET with the concrete matrix, even though EDS confirmed the presence of hydrated mortar on the surface of the polymer, confirming the results of this study.

Figure 6 (b) shows the SEM image for the concrete containing PP, where a surface with low amount of hydrated calcium silicate crystals can be noted followed by the non-adhesion of the polymer to the cementitious mass. According to some authors [47, 48], this is due to the lower chemical affinity of the polymer to the mortar when compared to natural aggregate and also due to the hydrophobic behaviour of the material which hampers the sodium precipitation at the surface of the polymer. Moreover, the smooth surface of PP does not contribute to the anchorage of the polymer to concrete.

4. CONCLUSIONS

The following conclusions can be drawn from the results presented in this work:

- The hygroscopic characteristic of the polymer PET and the shape granules leaded to a lower void content and a lower water absorption when compared to the sample with PP.
- The mixture with PP flakes presented higher modulus of elasticity than the mixture with PET flakes, similar to the reference mixture PP flakes in concrete act as a barrier for the chloride ion penetration, reducing the electric charge passing by less than half of the value for reference mixture.
- In the microscopic analysis it was not possible to verify the adhesion of PET flakes and PP flakes to the concrete.
- The addition of PET flakes presented good results in terms of mechanical properties and some durability issues. However, due to the potential hydrolytic degradation in alkaline environment, the use of PET for structural purpose is not recommended due to the further reduction on the mechanical properties and durability as ageing occurs.
- The addition of PP flakes is considered to be a promising alternative to partially replace sand, due to the good results in terms of mechanical properties and stability in the alkaline environment.

5. ACKNOWLEDGEMENTS

The authors acknowledge the “Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq)” by granting scholarships and funding the materials that supported this research.

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