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Review

Synthetic gravel for concrete obtained from sandy iron ore tailing and recycled polyethyltherephtalate

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HIGHLIGHTS

- Properties of concrete using synthetic gravel and sandy residue.
- The process to obtain the synthetic gravel.
- Characterization of synthetic gravel for concrete.
- Green product for engineering.

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ABSTRACT

The exploration of banded iron formations (BIFs) as iron ore is an important economic activity in Brazil, especially in the State of Minas Gerais. The process to obtain iron ore concentrates from BIFs generates huge amounts of tailings. About half of the extracted volume is transformed into tailings, which are usually employed to fill exhausted open pits or stored in dams. Although these tailings are inert and non-hazardous, the huge volumes cause concerns related to safety of dams. With the shortage of space for dams and increasing costs of monitoring and licensing, the use of tailings as raw material for other manufacturing chains is becoming attractive. A synthetic gravel for concrete production was obtained with the sandy residue from BIFs exploitation and recycled polyehtyltherephatalate (PET). The process to obtain the synthetic gravel, its properties, as well as the properties of concrete is presented. The unitary mass of this gravel is 0,89 kg/dm^3 (lightweight aggregate). The compressive resistance of the concrete after 7 days was around 9 MPa, the water absorption was 13.6–14.9%, and the density of the concrete was about 1.9 g/cm^3. The concrete is suitable for making light bricks for masonry in the region of the mines.

Contents

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1. Introduction

To put into perspective, the urban waste generation of the world in a year is less than the tailings generation of Rio Tinto, one of the top mining companies. Since the tailings of mining are usually generated far from the urban areas, unfortunately people do not care about them, unless when a dam collapses. Despite much effort in reuse and recycling over the last few decades, most of the mining tailings are stored in an unsustainable way. Solving the waste problem is not only a competitive issue. It is a whole of industry risk and requires cooperation, research, and development. On the other hand, the plastic wastes are a concern in all urban areas. It should be everywhere urgently addressed, because it is a global issue. The Great Pacific garbage patch is a clear alert [3].

The generation of tailing is increasing because the average grades of ore are decreasing and the demand for metals and other minerals are increasing. The demand for plastics is also increasing. In particular, the PET (polyehtyltherephatalate) production, especially bottles, is growing and its recycling is gaining importance. Milanez et al. [9] studied the high cost and environmental benefits of recycling in Brazilian urban areas including steel, aluminum, paper, plastic, and glass. Polymers by themselves do not cause environmental problems, but their inappropriate disposal causes a lot of concern. The systematic recycling of polymers is a solution to minimize this environmental impact.

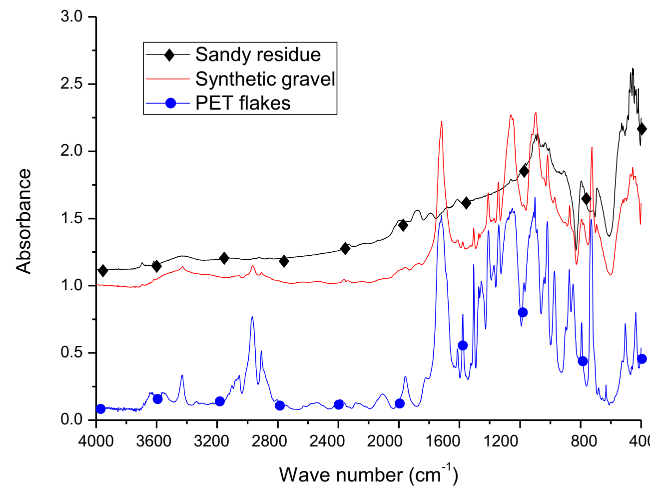

Fig. 2. FTIR spectra of the PET flakes, sandy residue, and of the synthetic gravel.

Table 1
Factorial 2^3 design for concrete samples made with the sandy residue and the synthetic gravel.

Fixed factor: Sandy residue to cement mass ratio = 3.12				
Factors			Levels	
			−1	+1
Synthetic gravel to residue mass ratio (SG)			0.42	0.56
Water to cement mass ratio (WC)			0.90	0.95
Fluxing to cement mass ratio (F)			0.006	0.009
Runs order		SG	WC	F
Standard	Execution			
1	7	−1	−1	−1
2	5	+1	−1	−1
3	6	−1	+1	−1
4	8	+1	+1	−1
5	3	−1	−1	+1
6	4	+1	−1	+1
7	1	−1	+1	+1
8	2	+1	+1	+1

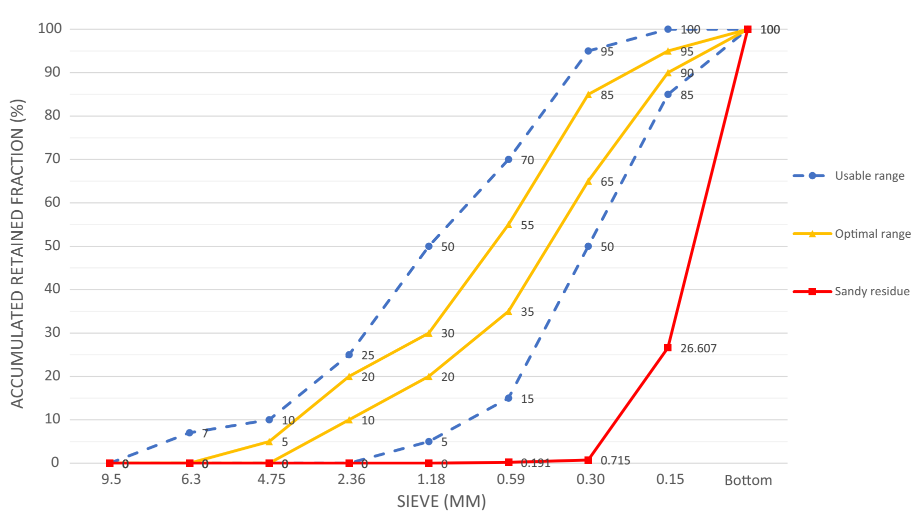

Fig. 1. Particle size distribution of the sandy residue compared to the requirement of the norm ABNT NBR 7211: 2009 (adapted from [4]).

Table 2
Chemical composition by X-rays fluorescence of the synthetic gravel.

SiO_2	Fe_2O_3	Al_2O_3	SO_3	P_2O_5	CaO	MnO	NiO	CuO	Volatile
49.20	29.35	1.46	0.48	0.36	0.12	0.061	0.08	0.008	19.00

Table 3
Granulometry of the synthetic gravel.

Sieve (mm)	Accumulated retained fraction (%)	
	Sample 1	Sample 2
9.5	4.4	2.5
6.3	88.1	88.0
4.75	96.9	96.3
2.36	99.6	99.4
1.18	99.6	99.5
0.6	99.6	99.5
0.7	99.7	99.5
0.15	99.7	99.6
Bottom	100.0	99.9

The exploration of banded iron formations (BIFs) as iron ore requires the comminuting of the rock to separate the particles of iron oxides from the quartz and clay ones. This comminuting generates many fine particles, which should be removed by a desliming step. The slime is the first residue of the process, constituted mainly by iron oxide particles (hematite and goethite) and small fractions of quartz and clays particles. After desliming, the comminuted material is sent to flotation to separate the iron oxide particles from the quartz ones. In this step the second residue of the process is generated, constituted of quartz particles and small fractions of iron oxide particles. It is a kind of very fine sand with the mean particle size of 150 μm. This sandy residue was employed in this study. The generated mass of the slime and the sandy residues is about the same mass of produced iron ore concentrate.

The sandy residue can be used to make concrete in place of natural sand, but this substitution is limited due to the fine granulometry. For example, the compressive strength of interlocked blocks obtained with the use of cement and the sandy residue is at most 18 MPa. By using natural gravel, this compressive strength can be increased to 24 MPa. According to Brazilian standards, the required compressive strength should be 35 MPa. In order to achieve high levels of substitution for natural sand, it is necessary to develop agglomeration of the fine particles of the sandy residue. The objective of this paper is to present an agglomeration process of sandy residue particles by using recycled PET. A kind of synthetic gravel made of recycled PET and sandy residue can be obtained. The characteristics of this synthetic gravel and of the concrete made with it, cement, and the sandy residue are presented.

Recycled PET is being considered as raw material for concrete. Choi et al. [2] investigated concrete made with granulated blast-furnace slags covered with recycled PET. They observed that the workability of the concrete is improved, the surface of the aggregate is strengthened, and the transition zone is narrowed. Benosman et al. [1] studied the replacement of different fractions of cement by recycled PET powder in mortar. No chemical interaction between the mineral specimens of mortar and PET were observed. Machovič et al. [8] studied the microstructure of the transition zone of cement paste reinforced with recycled PET fibers. They observed that the transition zone may be improved by alkaline treatment to increase the fiber surfaces. Ochi et al. [11] and Kim et al. [6] also studied the performance of concrete reinforced with

Table 5
Effects of factors on the compressive resistance of the concrete made with the synthetic gravel, calculated with the software Minitab 17.

		7 days	21 days	91 days
SG	Effect	−1.02	−1.37	−1.18
	SE	0.30	0.37	0.58
	p-value	0.002	0.001	0.049
WC	Effect	1.47	0.81	0.85
	SE	0.30	0.37	0.58
	p-value	0.000	0.036	0.152
F	Effect	2.14	0.96	0.81
	SE	0.30	0.37	0.58
	p-value	0.000	0.015	0.173
SG*WC	Effect	−0.20	−0.31	0.76
	SE	0.30	0.37	0.58
	p-value	0.514	0.411	0.198
SG*F	Effect	0.11	0.70	−0.37
	SE	0.30	0.37	0.58
	p-value	0.704	0.070	0.530
WC*F	Effect	−1.75	−0.66	−1.29
	SE	0.30	0.37	0.58
	p-value	0.000	0.085	0.033
SG*WC*F	Effect	−0.64	−1.59	0.70
	SE	0.30	0.37	0.58
	p-value	0.039	0.000	0.232

SE: standard deviation.

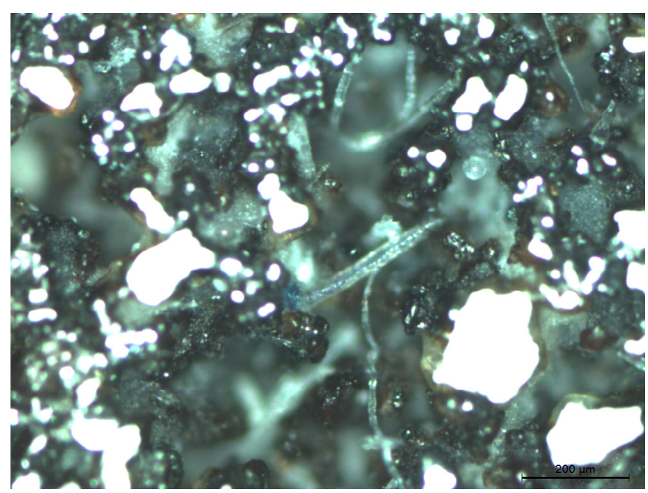

Fig. 3. Image of optical microscopy of a cross section of a synthetic gravel grain. The continuous matrix is PET, where the sandy residue microparticles (white particles) are embedded.

Table 4
Density, saturated density, apparent density, and water absorption of the synthetic gravel according to the norm NBR NM 53 2009.

Sample	Density (g/cm^3)	Saturated density (g/cm^3)	Apparent density (g/cm^3)	Water absorption (%)
1	2.153	1.862	1.609	15.71
2	2.132	1.868	1.636	14.23

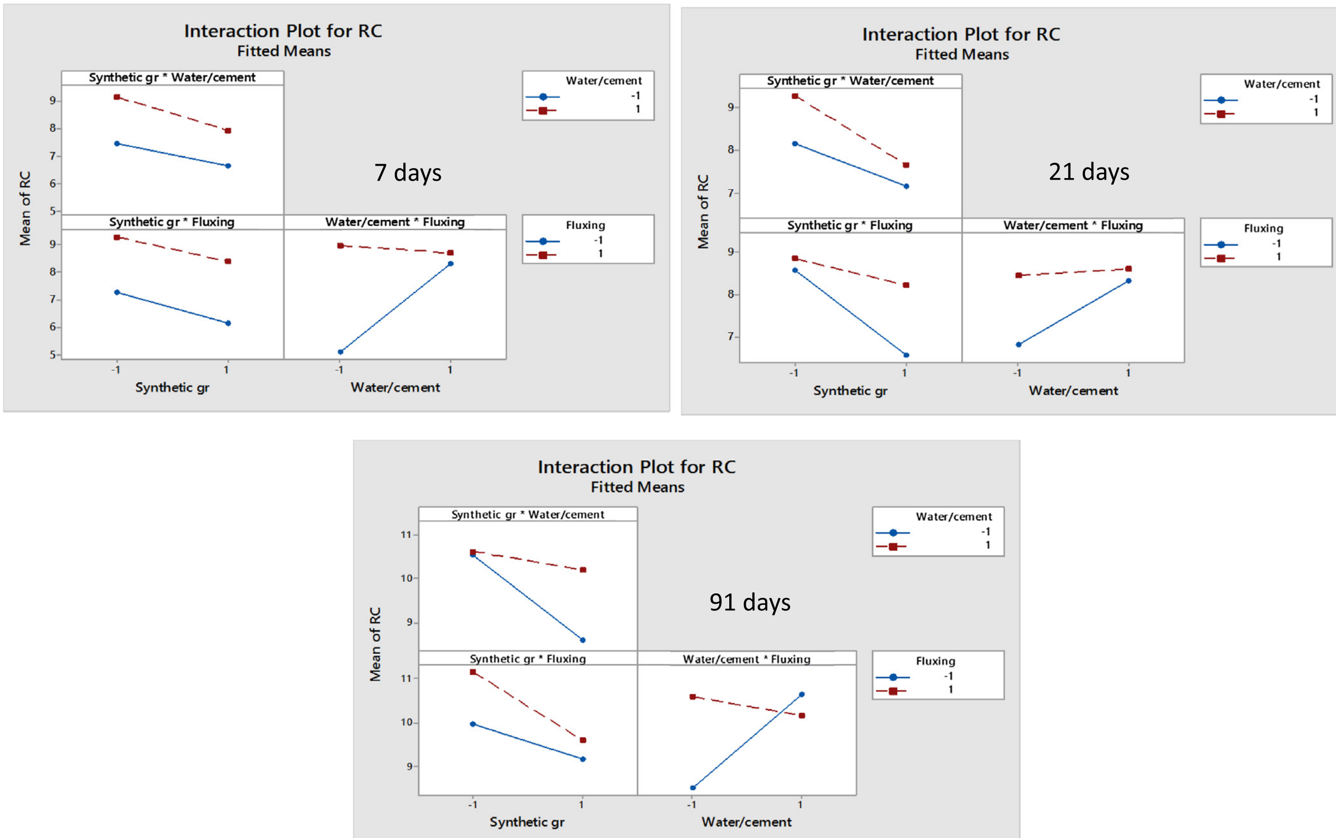

Fig. 4. Interactions plots for the compressive resistance.

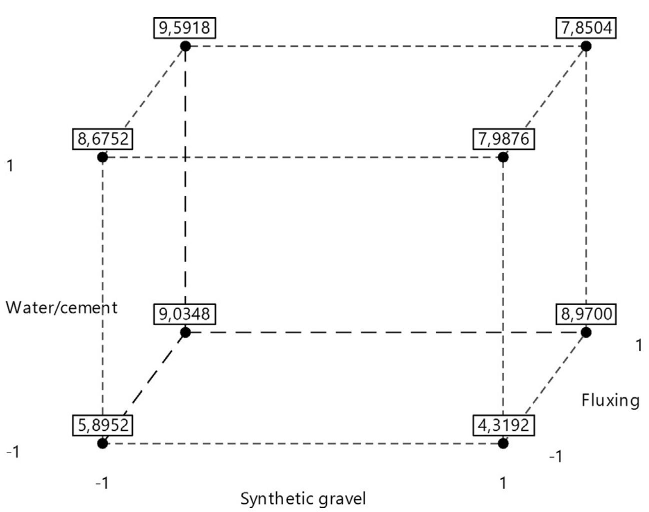

Fig. 5. Cube plot for the mean compressive resistance (MPa) after 7 days for concrete samples made with the synthetic gravel.

recycled PET fibers. Galvão et al. [5] studied concrete properties with the addition of recycled polymeric and elastomer materials including low-density polyethylene, crushed PET, and rubber from useless tires. Noviks [10] reported the synthesis of recycled PET-ash-clay composites with the possibility to use this material for the development of products for different purposes.

2. Materials and methods

The sample of sandy residue was provided by Samarco Mineração S.A. It is a blend of tailings generated on different days and production lines to ensure the representativeness due to process variations. The PET was bought from WHARGO – Comércio e Reciclagem. This company recycles plastic material from the metropolitan area of Belo Horizonte, Minas Gerais, Brazil. For PET recycling, WHARGO carries out the pecking of used bottles that resulted in quite varied flake sizes.

In order to obtain an uniform mixture of PET and sandy residue, the following steps were carried out: mixture of equal masses of PET flakes and sandy residue in a melting pot; heating in an oven in air up to 260 °C to melt the PET flakes; milling the cooled smelted body in air to obtain particle sizes bellow 0.1 mm; mixture of the milled material with sandy residue to obtain a content of 25w/o of PET; heating the mixture up to 260 °C to melt the PET particles; crushing and sieving the cooled smelted material in air to obtain the synthetic gravel of 3/8-4.0#. 50 batches of about 1 kg of synthetic gravel were produced.

The sandy residue, the PET, and the synthetic gravel were characterized by infrared spectroscopy, X-rays fluorescence spectroscopy, and granulometry. The synthetic gravel was characterized by moisture, pulverulent material, unitary mass, density, water absorption, and optical microscopy.

A 2^3 factorial design of experiments (Table 1) were performed to obtain concrete samples with the synthetic gravel. The synthetic gravel was used in place of natural gravel and the sandy residue was used in place of natural sand. The cement was CPV ARI (high initial resistance), with a fixed ratio of mass of residue to mass of cement of 3.12. The factors were the mass ratio of synthetic gravel to residue (SG), water to cement (WC), and fluxing (F) (superplasticizer, an aqueous solution of polycarboxylate provided by SIKA – Viscocrete 5700) to cement. The responses of these experiments were the compression resistance (five samples for each run), the void ratio, water absorption, and real density (three samples for each run).

3. Results

Fig. 1 shows the particle size distribution of the sandy residue. It is finer than the usable range for concrete according to the norm ABNT NBR 7211: 2009. Dosage studies are required to assure its applicability for concrete manufacturing.

Fig. 2 shows FTIR spectra of the PET flakes, sandy residue, and of the synthetic gravel. The spectrum of the synthetic gravel is the sum of the PET flakes and of the sandy residue, suggesting that there was no reaction between the PET and the sandy residue.

Table 6
Effect of factors and their interactions on the void ratio, water absorption, and real density for concrete samples made with the synthetic gravel after 91 days calculated with Minitab 17.

		Void ratio (%)	Water absorption (w%)	Real density (g/cm^3)
SG	Effect	−0.24	−0.21	0.01
	SE	0.11	0.09	0.01
	p-value	0.059	0.046	0.536
WC	Effect	−0.97	−0.82	0.02
	SE	0.11	0.09	0.01
	p-value	0.000	0.000	0.033
F	Effect	0.49	0.21	0.02
	SE	0.11	0.09	0.01
	p-value	0.001	0.040	0.02
SG*WC	Effect	−0.06	0.00	−0.01
	SE	0.11	0.09	0.01
	p-value	0.650	0.986	0.486
SG*F	Effect	−0.49	−0.24	−0.02
	SE	0.11	0.09	0.01
	p-value	0.001	0.022	0.044
WC*F	Effect	0.14	0.11	−0.00
	SE	0.11	0.09	0.01
	p-value	0.253	0.266	0.841
SG*WC*F	Effect	0.13	0.09	0.00
	SE	0.11	0.09	0.01
	p-value	0.275	0.354	0.890

SE: standard deviation

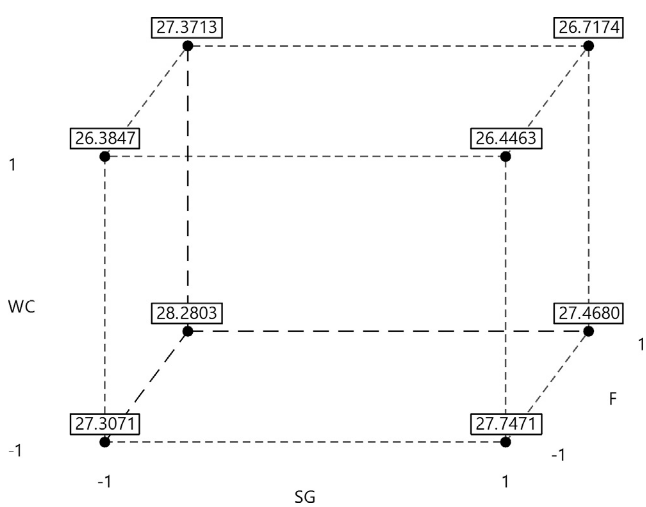

Fig. 6. Cube plot for the mean void ratio (%) after 91 days of concrete samples made with the synthetic gravel.

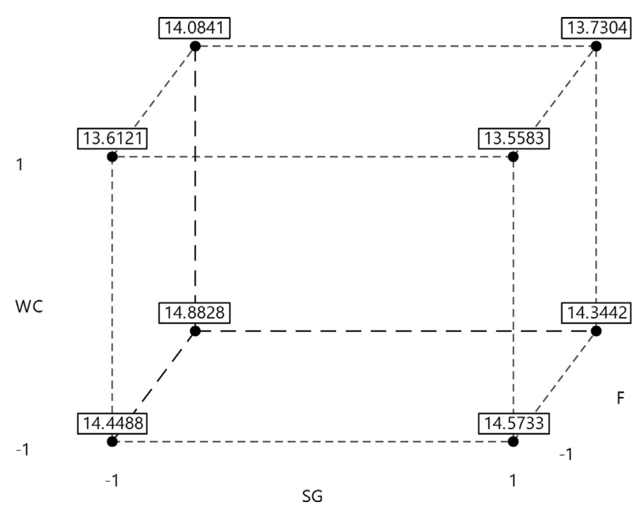

Fig. 7. Cube plot for the mean water absorption (w/o) after 91 days of concrete samples made with the synthetic gravel.

Table 2 shows the chemical composition of the gravel obtained by X-rays fluorescence. The PET is included in the volatile material.

The moisture content of the synthetic gravel was measured by weighing three samples before and after drying in an oven at 105 °C in air, according to the HG53 Halogen Moisture Analyzer. The result obtained was 0.21%. For comparison, the moisture content of a natural gravel measured under the same conditions was 0.02%.

The particle size distribution of the synthetic gravel was measured in two batches according to the norm ABNT NBR NM 248. The results are shown in Table 3.

The pulverulent material was measured in two samples of 1.0 kg according to the norm ABNT NM 46. The results were 0.31 and 0.45%. The unitary mass was measured according to the norm NBR 7251 2006 for three samples. The result was 0.879 ± 0.006 kg/dm^3. For comparison, the unitary mass measured of a natural gravel was 1.454 ± 0.001 kg/dm^3. Table 4 shows the density, saturated density, apparent density, and water absorption of the synthetic gravel according to the norm NBR NM 53 2009 for two samples of synthetic gravel.

Fig. 3 shows an image of the cross section of a synthetic gravel particle. The continuous matrix is the PET where the sandy residue microparticles are embedded. There are also many pores.

Table 5 shows the effects of the factors and their interactions on the compressive resistance of concrete samples made with the synthetic gravel and the sandy residue. One observes that the standard deviation (SE) is larger for the samples of 91 days. Considering a significance level of 90%, the increase of the amount of synthetic gravel (SG) decreases the compressive resistance. The increase of the water to cement ratio (WC) increases the compressive resistance for 7 and 21 days, but no significant effect could be seen for 91 days, probably due to the larger SE. The increase of the fluzing to cement ratio (F) increases the compressive resistance for 7 and 21 days, and the effect for 21 days is smaller than the one for 7 days. No significant effect could be seen for 91 days. The SG∗WC interaction has no significance. The SG∗F interaction is negative and significant in all cases. The interaction between the three

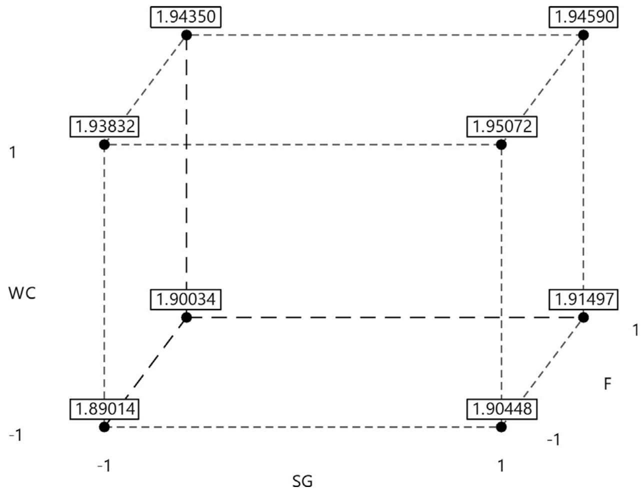

Fig. 8. Cube plot for the real density (g/cm^3) after 91 days of concrete samples made with the synthetic gravel.

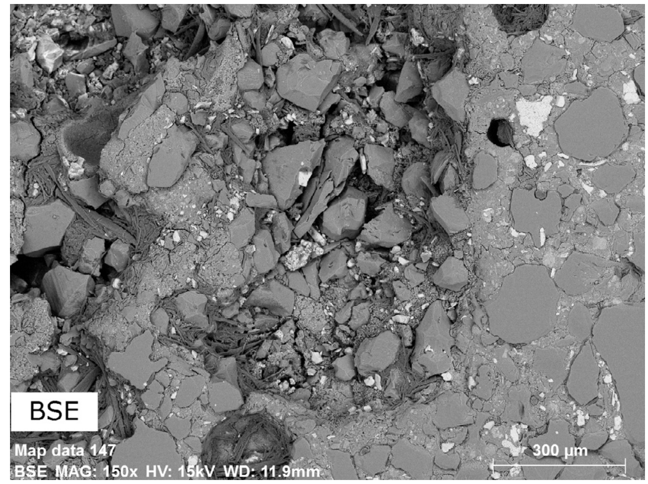

Fig. 10. Pore wall of the synthetic gravel after 91 days of curing, showing the formation of ettringite (needle like crystals).

Fig. 9. Fracture surface of a concrete sample.

factors, SG∗WC∗F is significant and negative for 7 and 21 days. The overall compression resistance was (7.79 ± 0.15) MPa for 7 days, (8.06 ± 0.19) MPa for 21 days and (9.98 ± 0.29) MPa for 91 days.

The increase of the compressive resistance with the water to cement ratio (WC) is not the usual behavior of concretes. Fig. 4 shows the interaction plots for the compressive resistance. At low level of F, the increase of WC increases the compressive resistance as if there was little water available. At the high level of F one observes higher resistances at low level of WC and a little lower or a little higher resistances at the high level of WC, according to the expected water reducing effect of the fluxing.

Fig. 5 shows the cube plot of the mean compressive resistance for 7 days. The maximum resistance was achieved for SG at low level, and WC and F at high level. The antagonistic interaction between WC and F was expected, due to the purpose of the fluxing to reduce the WC ratio. Considering the SG∗F and SG∗WC∗F interaction for 7, 28, and 91 days, a deleterious reaction between the synthetic gravel and the fluxing could not be disregarded.

Table 6 shows the effect of the factors and their interactions on the void ratio, water absorption, and real density of the concrete samples after 91 days. Figs. 5–8 show the cube plots for these responses. The influence of the factors and their interactions on these responses was small, although statistically meaningful in some cases. One observes that the void ratio decreases with the increase of the synthetic gravel ratio and of the water to cement ratio. The influence of WC on void ratio is different from the usual behavior of concrete, where the increase of WC increases the void ratio. This occurs due to the increased effectiveness of concrete compaction, reducing the void ratio. The water absorption also decreases with the increase of the synthetic gravel ratio and of the water to cement ratio. The only significant interaction for these three properties was the SG∗F interaction and it was antagonistic in all cases.

Fig. 9 shows the fracture surface of a concrete sample.

4. Discussion

The effect of the water to cement ratio and its interaction with the fluxing suggests that part of the water was not available to react with the cement. It was probably absorbed by the synthetic gravel and may promote later reactions with the cementitious products, forming ettringite in the pore walls (see Fig. 10).

The density of the concrete made with the synthetic gravel is 1.9 g/cm^3, about 13% higher when comparing with concrete made with blast furnace slag. The average water absorption of this concrete was in the range of 13.6–14.9%, slightly higher than the requirements of the norm ABNT NBR 6136 [7]. Considering the compressive resistance 9 MPa, this concrete may be suitable for masonry in the region of the mines in Minas Gerais, Brazil, where the temperature oscillates between 5 and 37 °C. An antagonistic interaction between the synthetic gravel and the fluxing was observed for the void ratio, water absorption, and real density. The compression resistance may also be deleteriously affected by this interaction. This may be an indication that the fluxing reacts with the PET and the long-term behavior of this interaction should be monitored. The water absorption can be decreased by decreasing the porosity of the synthetic gravel particles. This can be achieved by increasing the PET content or pressing the mixture of PET and sandy residue before the second smelting. Putting more PET, the content of sandy residue will be reduced, and pressing the mixture of PET and sandy residue may increase the density of the concrete. These procedures were not tested in this paper.

The concrete made with the synthetic gravel has about 65 w/o of sandy residue and 5 w/o of PET. This is a high level of utilization of sandy residue. The combination of recycled PET and sandy residue make the concrete obtained in this paper a very environmental friendly product.

5. Conclusions

A porous lightweight synthetic gravel was obtained with the sandy residue from banded iron formations processing as iron ore and recycled polyehtyltherephatalate (PET). The synthetic gravel was composed of 75w/o of sandy residue and 25w/o of PET, with a unitary mass of 0.88 kg/dm^3.

A concrete was obtained with the synthetic gravel with a compression resistance of 9–12 MPa (after 7–91 days), a dry density of 1.9 g/cm^3, void ratio of 27%, and water absorption of 14 to 15%. The factorial analysis showed that there is an interaction between the PET and the fluxing (aqueous solution of polycarboxylate) that should be monitored in the long term.

The concrete made with the synthetic gravel has a high utilization of sandy residue (about 65 w/o of sandy residue and 5 w/o of PET). The combination of recycled PET and sandy residue make this concrete a very environmental friendly product.

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