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Reuse of sanitary ceramic wastes as coarse aggregate in eco-efficient concretes

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ABSTRACT

The sanitary ceramics industry inevitably generates wastes, irrespective of the improvements introduced in manufacturing processes. The present study investigated the reuse of these wastes as recycled coarse aggregate in partial substitution (15%, 20% and 25%) of natural coarse aggregates in the manufacture of structural concretes. The results demonstrate that recycled, eco-efficient concretes present superior mechanical behaviour compared to conventional concrete and it was moreover appreciated that the recycled ceramic aggregate does not interfere in a negative way during the hydration process. It was also observed that the microstructure in the interfacial transition zone (ITZ) between recycled ceramic aggregate and paste was more compact than in the case of natural aggregate and paste.

1. Introduction

The generation and management of wastes from different productive activities constitute a serious environmental issue in modern society, prompting the existence of a European policy (Directive 2008/98/CE) on wastes. This directive has been incorporated into the internal laws of the various European Union member states, and in Spain is reflected in the 2008–2015 Integrated National Plan on Waste, the aim of which is to encourage suitable management of wastes, reduce their generation and promote correct waste treatment methods, including prevention, reuse, recycling, resource recovery and elimination. The reuse of wastes will assume even greater significance in the future for different reasons: energy, technical, economics, environmental and social.

Spain represents 26% of all ceramics factories within the EU-27, and is the world leader in the market for ceramic sanitaryware, with an annual production rate of 7 million ceramic items a year (2008) and generates, according to dates the manufacturer of sanitaryware, approximately 24 tons of waste a month, which is simply dumped.

Ceramic products are produced from natural materials which contain a high proportion of clayey minerals. Through a process of dehydration followed by controlled firing at temperatures between 1200 °C and 1290 °C, these minerals acquire the characteristic properties of "fired clay".

Moreover, these ceramic wastes are products include high strength, wear resistance, long service life, chemical inertness and nontoxicity, resistance to heat and fire and electrical resistance.

The manufacturing process inevitably generates a percentage of products deemed unsuitable for sale, regardless of any improvements made to the process. The two principle reasons for the rejection of these items are breakage or defective shape, defects which do not affect the intrinsic properties of the ceramic material, or firing defects as a result of too much or too little heat, which in this case do affect the physico-chemical properties. The percentage of rejected material depends on the type of factory, the product requirements and other technical considerations.

The usage of ceramic waste from sanitaryware industry as coarse aggregates for manufacture of concretes is at present a novel research line at international level, as there are no standards applicable yet on its reuse. Nowadays, there are several papers [1–6] that study the possibility of using different ceramic waste as active additives in the manufacturing of cement, taking advantage of their pozzolanic activity to reach satisfactory results in respect of durability and mechanical properties. On the other hand, there is a series of works that analyse the possibility of introducing recycled ceramic [7–16] material in substitution of natural aggregate (sand or gravel) when producing concrete for several uses (precast elements, bases and sub-bases, non-structural and structural concretes, etc.). In those works satisfactory results were obtained for different ratios of substitution.

Nowadays there is a big scientific and technical gap on the behaviour of concretes that incorporate this type of ceramic wastes. Because of this, the present article presents physical and

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mechanical properties of structural concretes in which coarse natural aggregate (gravel) was partially substituted in 15%, 20% and 25% by coarse recycled ceramic aggregate. Moreover, tests were carried out to study of the microstructure in the interfacial transition zone (ITZ) between paste/coarse aggregates (natural and recycled) by means of scanning electron microscopy (BSE/SEM). Mineral phases resulting from the hydration process were identified by means of X-ray diffraction (XRD).

Table 1
Chemical composition of ceramic aggregate.

Chemical constituent (wt.%)	Recycled ceramic aggregate	
	Internal part	External part
SiO_2	68.41	58.23
Al_2O_3	24.46	9.10
Fe_2O_3	0.94	0.91
CaO	0.63	11.80
MgO	0.19	0.67
Na_2O	1.65	2.38
K_2O	2.80	1.63
P_2O_5	0.17	0.10
TiO_2	0.55	0.10
ZrO_2	0.08	12.62
Others	0.12	2.46

2. Experimental program

2.1. Materials

The natural aggregate used can be classified into two fractions, the coarse fraction (Fig. 1a) of boulder (gravel), corresponding to a fraction size of 4/20 mm, and the fine fraction (sand), of less than 4 mm. The mineralogical composition presented mainly quartz, followed by lower proportions of another series of aluminosilicates from the mica and feldspar groups.

Table 2
Properties physicals and mechanicals of aggregates.

Characteristic	Gravel	Ceramic	EN 12620/ EHE-08
Grading modulus	6.93	6.17	–
Maximum size (mm) (EN 933-1)	20	12.5	–
Fine content (wt.%)	0.22	0.16	<1.5
Dry sample real density (kg/dm^3) (EN 1097-6)	2.63	2.39	–
Water absorption (wt.%) (EN 1097-6)	0.23	0.55	⩽5
Flakiness Index (wt.%) (EN 933-3)	3	23	<35
"Los Ángeles" coefficient (wt.%) (EN 1097-2)	33	20	⩽40
Total porosity (vol.%) (MIP)	0.23	0.32	–

Fig. 1. Illustration of (a) natural gravel aggregate and (b) recycled ceramic aggregate.

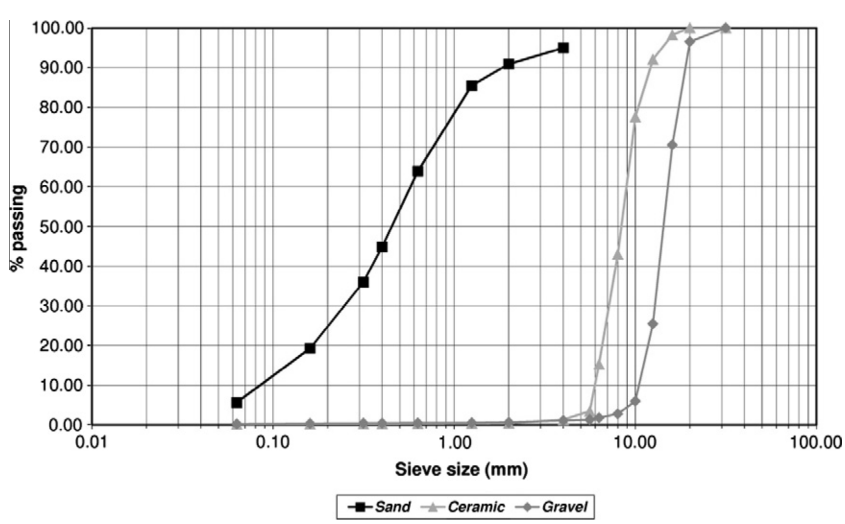

Fig. 2. Aggregate size grading curves.

Table 3
Mix proportions of concretes.

Type concrete	Materials (kg/m^3)				
	Sand	Gravel	Ceramic	Cement	Water
Concrete reference (RC)	716.51	1115.82	0.00	398.52	205.00
Concrete containing 15% recycled aggregate (CC-15)	723.48	948.45	162.32	390.36	205.00
Concrete containing 20% recycled aggregate (CC-20)	725.81	892.66	216.43	387.64	205.00
Concrete containing 25% recycled aggregate (CC-25)	728.14	836.87	270.53	384.91	205.00

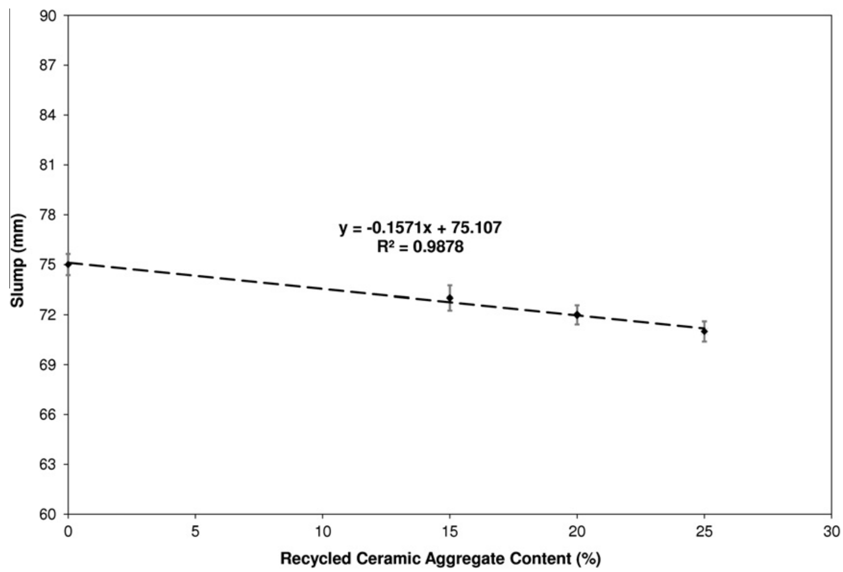

Fig. 3. The effect of the ceramic waste replacement as coarse aggregate on the workability of concrete.

The recycled ceramic aggregate employed to produce the concrete were taken from a manufacture of sanitaryware. They were produced by crushing ceramic sanitaryware waste by use of a jaw crusher. The aggregate fraction used corresponded to a size of 4/12.5 mm. The recycled aggregate (Fig. 1b) has an irregular shape and edges, as consequence mainly for original shape of the sanitaryware waste, almost thin element, moreover of the process used to obtain its.

Visually, the recycled ceramic aggregate presented two clearly distinguishable parts, an external part which corresponded to the glaze, or external part of the sanitaryware, and an internal part which comprised the waste matrix. As regards mineralogical composition, quartz comprised the principle mineral, with a lesser proportion of aluminosilicates, hematite and zircon.

The composition chemical of external and internal part of the ceramic aggregate is given in Table 1.

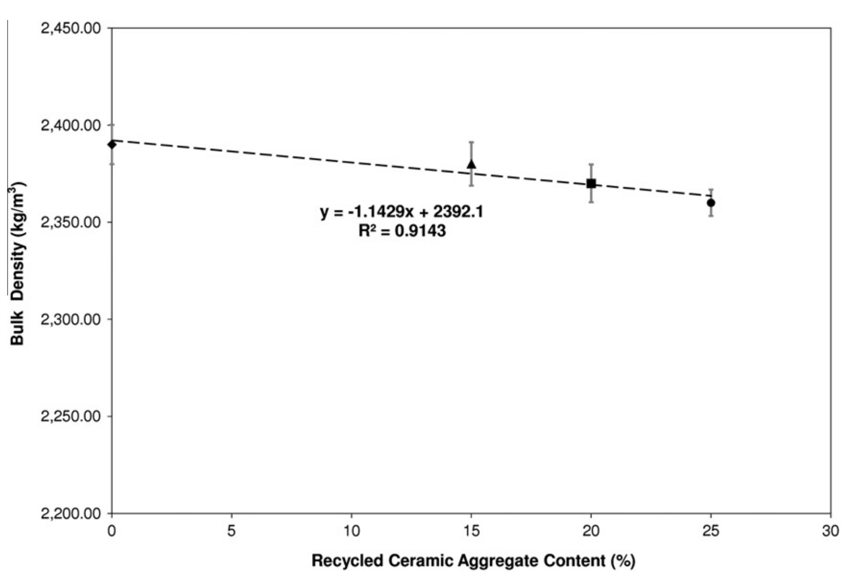

Fig. 4. Average bulk density of fresh concrete.

Fig. 2 shows the aggregate size grading curves obtained as a result of the sieve analysis. It can be observed that all the aggregates present continuous size grading curves, which would have a positive influence on concrete mix ease to cast. Furthermore, it should be noted that the shape curve for recycled ceramic aggregate is very similar to that for natural coarse aggregate and the gravel is most coarse that of ceramic aggregate.

Table 2 presents the physical and mechanic properties of coarse aggregates used, together with the limits laid down in each case in the standard EN 12620 [17] and Spanish Instructions for Structural Concrete (EHE-08) [18].

Portland cement: The pure Portland cement (type CEM I 52.5 R), high strength rapid-hardening was used in all the concretes mixes.

2.2. Concrete mixtures

In this study, four types of concretes were produced, one reference concrete (RC) and three recycled concretes; CC-15, CC-20 and CC-25; substituted a 15%, 20% and 25% in weight of natural coarse aggregate (gravel) by recycled ceramic aggregate respectively.

The La Peña dosage method [19] was used in the process of the design and calculation of doses for different types of concrete. This method establishes a characteristic strength value of 30 MPa and amount of water constant because the necessary volume of water is function of the maximum size aggregates (20 mm) and consistency (soft). The mixes obtained are shown in Table 3.

The w/c ratio in all concretes complied with requirements of maximum w/c ratio given in the EHE-08 to ensure durability.

Table 4
Compressive strength of concrete at 7, 28 and 90 days.

Type concrete	Compressive strength (MPa)		
	7 days	28 days	90 days
RC	30.81 ± 1.57[a]	35.87 ± 1.49	41.33 ± 1.55
CC-15	33.81 ± 1.19	37.24 ± 1.58	42.16 ± 1.89
CC-20	35.38 ± 1.24	38.53 ± 1.16	43.00 ± 1.05
CC-25	37.32 ± 1.09	39.83 ± 1.99	44.10 ± 1.13

[a] The ± represents one standard deviation.

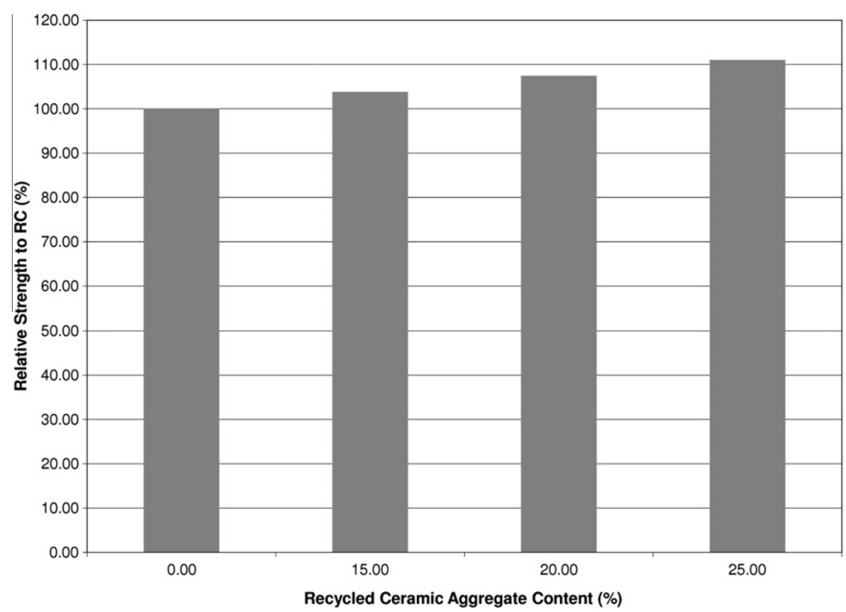

Fig. 5. The effect of ceramic waste as coarse aggregate on the relative compressive strength of concrete.

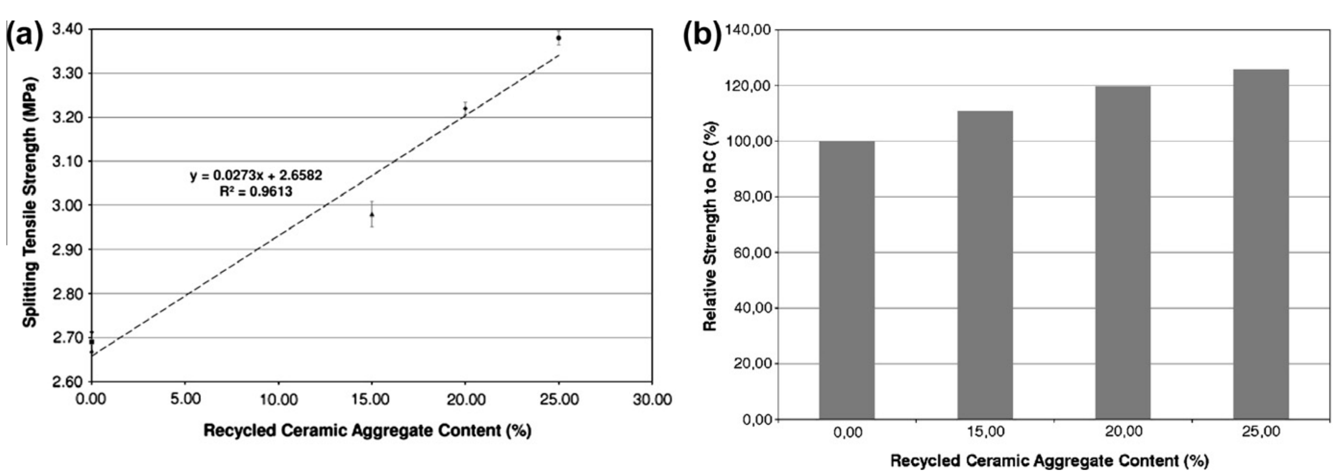

Fig. 6. The effect of ceramic waste as coarse aggregate on the splitting tensile strength of concrete: (a) splitting tensile strength and (b) relative strength.

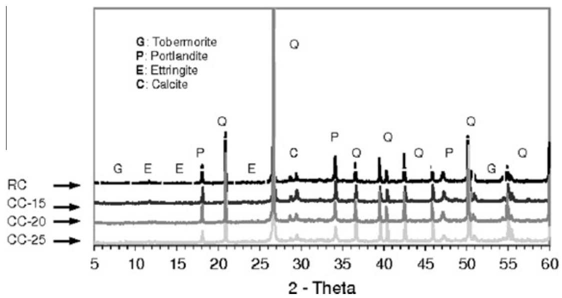

Fig. 7. X-ray diffraction: different concrete mixes.

The properties determined for both fresh and hardened concretes are consistency, bulk density, compressive and splitting tensile strength.

2.3. Experimental technical

The compressive and splitting tensile strength of concretes were tested according to EN 12390-3 [20] and EN 12390-6 [21] respectively.

Mineralogical composition was studied in the mixed concrete by X-ray diffraction (XRD) using the random powder method for the bulk sample. The X-ray diffractometer was a BRUKER Theta-Theta diffractometer, model D8 Advance without monochromator and equipped with a 2.2 kW Cu anode.

The microstructure of the transition zone (ITZ) between paste/coarse aggregate (gravel and recycled ceramic aggregate) was determined using a scanning electron microscope (PHILIPS model XL 30) with tungsten source was used, which enables spot chemical analyses to be carried out using energy-dispersive X-rays, together with a silicon/lithium detector and an EDX analyser model DX4i. In this study specimens for backscatter examination were prepared by epoxy impregnation, followed by precision sawing and careful polishing of a plane surface for examination.

3. Results and discussion

3.1. Consistency and bulk density

Fresh concrete consistency was measured according to EN 12350-2 [22], and results indicated that all concretes presented a soft consistency (6–9 cm). Fig. 3 shows the effect of incorporating ceramic recycled aggregate on the workability of concretes

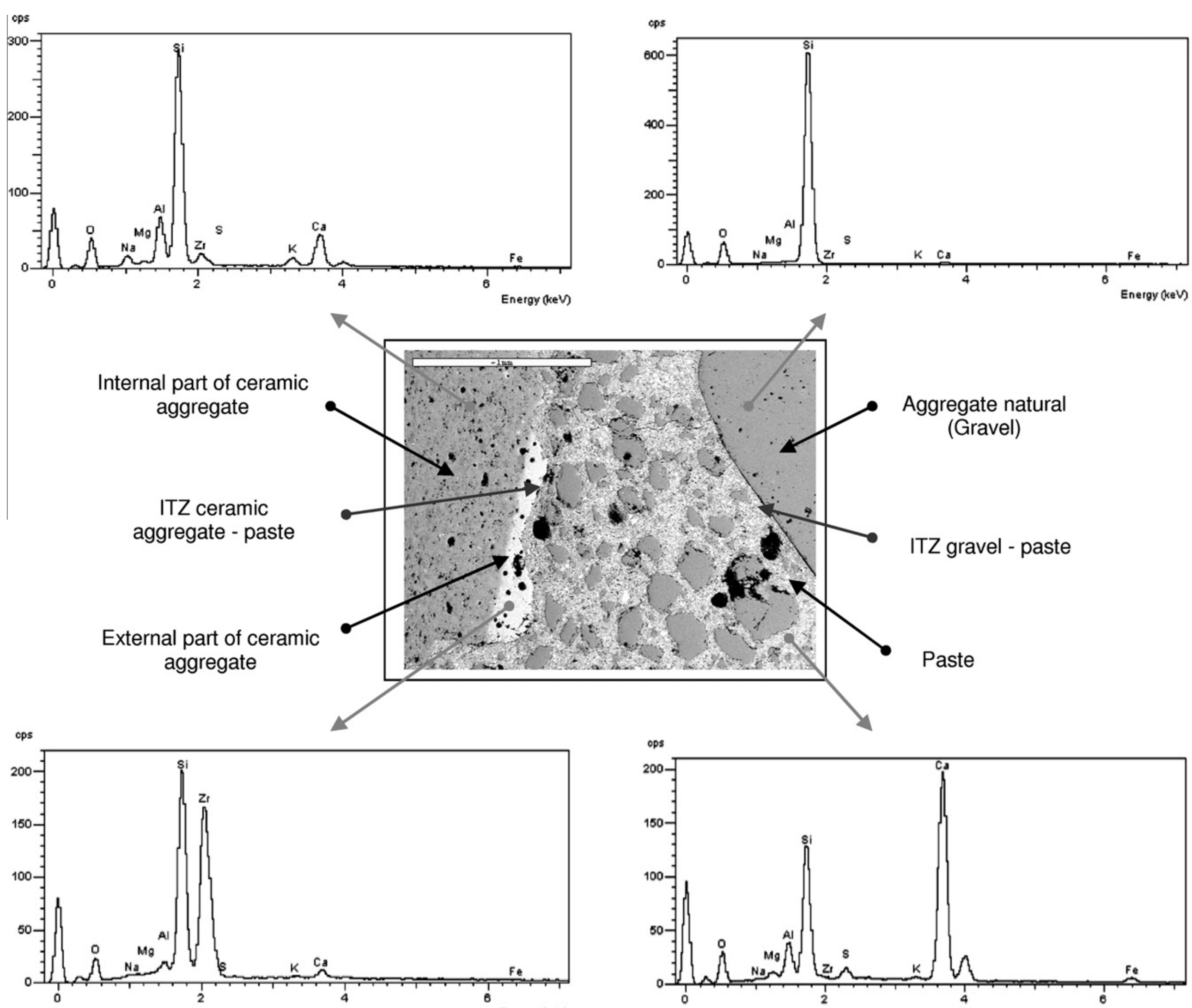

Fig. 8. BSE image of recycled concrete (50X): ITZ aggregates (gravel and ceramic) – paste and paste and aggregates analysed by EDX microanalysis.

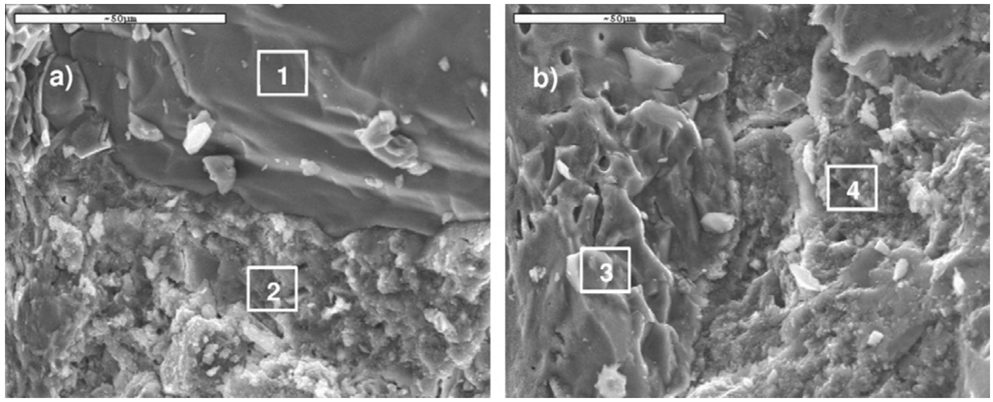

Fig. 9. Scanning electronic microscopy (SEM) micrograph of interface transition zone (ITZ) between coarse aggregate – paste (1000×): (a) Gravel (1) – paste (2). (b) Recycled ceramic (3) – paste (4).

produced with this type of material. In this study, one interesting observation was that the reduction of slump was only 5.3% when the recycled ceramic aggregate was used at 25%. Moreover, it can be observed that reduction in workability is lineally correlated by a coefficient of 0.9878. This is due to the higher water absorption, shape and porosity of the recycled ceramic aggregate.

With respect to the results obtained for fresh concrete bulk density studies, these are shown in Fig. 4, where it can be observed that as the percentage of natural coarse aggregate substituted rose, density of the recycled concrete reduced. Moreover, the concrete density can be seen to reduce linearly with the correlation factor of 0.9143. This reduction is due to the smaller density of the recycled ceramic aggregate.

3.2. Hardened concrete properties (mechanics and microstructures)

The results obtained for compression strength, at 7, 28 and 90 days, and splitting tensile strength, at 28 days, for the different concretes are given in Table 4 and Fig. 6a. respectively. It can be seen that the mechanical behaviour, both in terms of compression and splitting tensile strength, was better for the recycled concretes than for the reference concrete. Furthermore, both results also show that as the percentage of natural coarse aggregate substituted by recycled ceramic aggregate rose, the behaviour of the resulting concretes improved. Figs. 5 and 6b show the relative compressive and splitting tensile strength at 28 days respectively. It can observed that in the case of recycled concrete with 25% substitution (CC-25), there was an increase from 12% to 25% in compressive and splitting tensile strength compared to the reference concrete (RC).

Fig. 7 shows the results obtained in the XRD study of crystalline phases present in the concrete produced. Here, it can be seen that all the concrete mixes presented the phases typical [23] of the Portland cement hydration process (portlandite and ettringite), just as the presence of trace calcite, resulting from carbonation of the product.

Fig. 8 gives a micrograph of recycled concrete, with a microanalysis of the different constituents of which it is composed. From this analysis it can be seen that the interfacial transition zone (ITZ) between the recycled aggregate and the paste is narrower, more compact, less porous and less marked than that between the gravel and paste. This would result in improved mechanical behaviour, as demonstrated by the compressive and tensile strength assays, whilst at the same time producing a more durable concrete [24] since the new ITZ between recycled aggregate and paste is less permeable.

Fig. 9 confirms what is observed in Fig. 8, showing the best integration of the recycled aggregate in the concrete matrix

The improved incorporation of recycled ceramic aggregate in the paste is due to the more irregular shape it presents, resulting in a superior specific surface area than natural aggregate (gravel), which is rounded and thus lacks edges [24]. Moreover, this irregular shape provides the higher bond between recycled ceramic aggregate and the paste.

Furthermore, the ceramic aggregate could presented little pozzolanic activity in the surface part due to its chemical composition and grain size, this was nevertheless sufficient to react with the porltandite present in the periphery of the aggregate, giving rise to hydrated products such as calcium silicate hydrates (CSH) and calcium aluminate hydrates which present a less porous, more compact structure, forming a more stable aggregate/paste transition zone.

Also shown in Fig. 8 are the different X-ray microanalyses (EDX), which provided detailed information concerning the structure and composition of the aggregates and paste. The natural aggregate presented a siliceous nature, whilst the ceramic aggregate clearly presented two aspects, an internal and an external part. The internal part contained those chemical elements, such as Si, Ca, Mg, O and Fe, together with a lesser proportion of Zr, typical of ceramic materials. The external part contained the same elements, but with a much higher proportion of Zr.

Finally, as regards the paste, the elements analysed by EDX corresponded to hydration products which habitually occur during cement hydration [23]. These results are consistent with the obtained by XRD. Thus, no interference due to the presence of recycled aggregate was observed.

4. Conclusions

Based on the results obtained for the different assays conducted and of physical observations, the following conclusions can be drawn:

1. The recycled, eco-efficient concretes presented better mechanical behaviour in terms of compressive and tensile strength than the reference concrete.
2. The mechanical properties of recycled concretes improved as the percentage of natural coarse aggregate substituted by recycled ceramic aggregate rose.
3. Recycled ceramic aggregate does not interfere with the chemical reactions which occur during cement hydration whilst the concrete is setting and hardening.
4. The interfacial transition zone (ITZ) between paste and recycled ceramic aggregate was more compact, narrower and less porous than that between paste-gravel.

5. The recycled, eco-efficient concretes obtained by partial substitution of gravel for ceramic aggregates from sanitary ceramics industry waste can be used for structural purposes.
6. The potential substitution of natural coarse aggregate by recycled ceramic material coming from the sanitary industry offers several technical, economic and environmental advantages, which are relevant in the present sustainability context within the construction industry.

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