VALORISATION OF CERAMIC WASTE BY USE AS POZZOLAN IN PORTLAND CEMENT MORTARS

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ABSTRACT

Portland cement production is one of the processes that emits the most CO_2 into the atmosphere, as well as calling for large amounts of energy. Furthermore, ceramic wastes account for a large proportion of the total waste produced by the construction industry. These two factors make any alternatives that reduce consumption of Portland cement and also enable new uses to be found for ceramic waste extremely interesting.

The aim of the work presented here was to study the pozzolanic activity of two different types of ceramic waste (bricks and ceramic tiles) and to assess the chances of reusing them as partial substitutes for Portland cement in cement mortar.

Both wastes were crushed, screened and ground to an average particle size of 20µm, similar to that of Portland cement. Once a suitable particle size had been achieved, the wastes were characterised using scanning electron microscopy (SEM) and X-ray fluorescence (XRF).

The influence of both types of waste on consistency and setting time was studied using pastes in which 15%, 25%, 35% and 50% fractions of the Portland cement were replaced with each waste, according to standard UNE-EN 196-3.

Thereafter, cement mortars were prepared according to UNE-EN 196-1 with the aforementioned proportions. After curing in a chamber for 3 and 28 days, mortar compressive strength and Strength Activity Index (SAI) were determined and compared to the control mortar in which no cement had been replaced.

The results obtained show how the particles of both wastes have a similar morphology and contain high percentages of silica and alumina, necessary for the pozzolanic reaction to occur with the portlandite released when Portland cement is hydrated.

The pastes in which the cement had been partially replaced with brick waste revealed the greatest variations in consistency compared to those made with tile waste and in fact, they became less workable as the amount of substitute waste was increased. This circumstance is attributed to the brick waste having greater water absorption compared to that of the tile. On the other hand, the compressive strength after 3 and 28 days' curing was higher in the mortars made with brick waste compared to those formulated with tile waste. Although further microstructural studies are required to explain such behaviour, it may be due both to a reduction in the effective A/C ratio (caused by the brick's greater absorption capacity) and to its greater pozzolanic activity.

1. INTRODUCTION

The presentation given by Tomás et al. at Qualicer 2018 (3) pointed out that the production of Construction and Demolition Waste (CDW) had been on an upward trend in Spain since 2015 and that the target recycling rate for this waste was 70% by 2020. It also stated that more than 50% of CDW comes from ceramic products, apart from the ceramic industry's own manufacturing waste.

Although CDW is increasingly reused in construction, it is mostly destined as filler material in esplanades and road surfaces or to make concrete. Earlier works have demonstrated that concrete in which aggregates are partially replaced with CDW performs well (4,5). Similarly, the paper presented at Qualicer 2018 reported good results when part of the aggregate was replaced with ceramic waste in cement mortars (3).

On another level, the production of cement is one of the most polluting processes that exist, due to the enormous amounts of energy required to make it (raw material decarbonation and clinkerization processes) and the high levels of atmospheric emissions (nitrogen oxides, CO_2, ...). In this sense, according to data provided by the Getting the Numbers Right (GNR) project (1), in the year 2016, Spain emitted a net rate of 735 kg CO_2 per ton of clinker produced. Although the cement industry is making significant efforts to minimise its impact and aims to reduce its carbon footprint by 35% by 2050, it is still responsible for 5% of anthropogenic CO_2 emissions worldwide (2).

In recent decades, a significant amount of research has striven to reduce the amount of cement used in mortar, either, as in Naceri's study, by substituting part of the clinker with waste when manufacturing cement (9), or by replacing part of the cement with other materials that have potential pozzolanic activity. Mohamed's research paper (10) summarises the characteristics that such partial substitutes need to achieve. Other papers, such as Pereira-De Oliveira et al. or Mas et al. (7, 8), ratify the pozzolanic activity of ceramic tile wastes, while others, such as Silva et al. (6) confirm that brick waste also provides pozzolanic activity.

The work presented here compares the partial replacement of Portland cement with brick waste and ceramic tile waste. To do so, both the characteristics of the initial wastes, such as their strength, and the workability achieved in mortars in which varying proportions of cement were replaced with either waste, were assessed.

2. MATERIALS AND METHODS

2.1. MATERIALS USED

To make the cement mortar, the procedure defined in standard UNE-EN 196-1 was applied. A CEM I 42.5R type cement supplied by CEMEX was used. The water for mixing and curing came from the public mains.

The natural siliceous aggregates used belonged to three different commercial fractions: 0.2/0.6 aggregate, 0.6/1.2 aggregate, and 1.2/2.0 aggregate. These fractions were combined to obtain a mixture with a particle size curve similar to that for standard aggregate, as defined in UNE-EN 196-1.

Two different types of ceramic waste were used: wall and floor tiles from the province of Castellon (earthenware tile, stoneware tile and porcelain tile scrap) and red-body hollow bricks.

2.2. PREPARATION AND CHARACTERISATION OF THE WASTES

The ceramic tile and brick wastes were crushed and screened. Subsequently, they were ground in a ball mill until a particle size of 20µm, similar to that of Portland cement particles, was obtained. To achieve this, grinding time for each of the wastes was adjusted, testing particle size distribution in the resulting material by laser diffraction using a Malvern Instruments Mastersizer 2000 particle size analyser.

The morphology of the particles was also verified with an SEM-EDX JEOL JSM-6300 scanning electron microscope (SEM), and their chemical composition was examined by X-ray fluorescence (XRF) with a Philips Magix Pro spectrometer.

2.3. COMPOSITION AND CHARACTERISATION OF THE CEMENT MORTARS

All the cement mortars were prepared by mixing cement, aggregate and water in a ratio of 1:3:0.5, according to the procedure set out in standard UNE-196-1.

To assess the influence of the amount and type of ceramic waste on mortar strength, the total amount of cement and waste was kept constant in each of the mix batches and only the percentage of Portland cement replaced with each type of waste was varied. A control batch (CON) was made, which served as reference mortar and in which no Portland cement was replaced. Table 1 shows the nomenclature and compositions used for each of the batches thus prepared.

<table>
<tr><th>NOMENCLATURE</th><th>WASTE</th><th>CEMENT
(g)</th><th>%
SUBSTIT.</th><th>WASTE (g)</th><th>SILICEOUS AGGREG.(g)</th></tr>
<tr><td>CON</td><td>---</td><td>450</td><td>0</td><td>0</td><td rowspan="9">1350</td></tr>
<tr><td>B_15</td><td rowspan="4">TILE</td><td>382.5</td><td>15</td><td>67.5</td></tr>
<tr><td>B_25</td><td>337.5</td><td>25</td><td>112.5</td></tr>
<tr><td>B_35</td><td>292.5</td><td>35</td><td>157.5</td></tr>
<tr><td>B_50</td><td>225</td><td>50</td><td>225</td></tr>
<tr><td>L_15</td><td rowspan="4">BRICK</td><td>382.5</td><td>15</td><td>67.5</td></tr>
<tr><td>L_25</td><td>337.5</td><td>25</td><td>112.5</td></tr>
<tr><td>L_35</td><td>292.5</td><td>35</td><td>157.5</td></tr>
<tr><td>L_50</td><td>225</td><td>50</td><td>225</td></tr>
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Table 1. *Composition of the test mix batches.*

Consistency and setting time for each paste made with 15%, 25%, 35% and 50% of the Portland cement replaced by each type of waste were compared to the batch with no cement replacement, according to the procedure described in standard UNE-EN 196-3.

To evaluate the performance of hardened mortars, compressive strength was measured on an Ibertest MEH-300 PT/W tester at an age of 3 and 28 days. Curing was carried out in standard ambient conditions of 20°C and immersion in water in a wet chamber, as per the procedure detailed in standard UNE-EN 196-1. The results obtained were expressed in terms of strength activity index (SAI, %) and strength gain (SG, %) compared to the control mortar, calculated by means of [Equation 1] and [Equation 2] respectively:

$$\mathrm{SAI}(\%) = \frac{\sigma_{CS}}{\sigma_{CON}} \cdot 100 \qquad \text{[Equation 1]}$$

$$\mathrm{SG}(\%) = \frac{\sigma_{CS} - \sigma_{CON}}{\sigma_{CON}} \cdot 100 \qquad \text{[Equation 2]}$$

where σ_{CS} represents compressive strength of the mortars made with ceramic waste and σ the compressive strength of the control mortar. In all cases, compressive strength was calculated as the average of the individual results from the 6 test pieces of each mix batch.

3. RESULTS AND DISCUSSION

3.1. CHARACTERISATION OF THE WASTES

3.1.1. PARTICLE SIZE DISTRIBUTION OF THE MILLED WASTES

The ceramic tile and brick wastes were ground in a ball mill for 6 hours until materials with an average particle size (Dmean) of approximately 20 µm were obtained. The results of the particle size distribution achieved are shown in Table 2.

Waste	Milling time, h	Dmean µm	d_{10} µm	d_{50} µm	d_{90} µm
BRICK (L)	6	19.87	1.31	11.24	52.31
TILE (B)	6	**19.67**	1.60	14.00	46.81

Table 2. *Parameters of particle size distribution after 6 hours' milling*

3.1.2. CHEMICAL COMPOSITION AND PARTICLE MORPHOLOGY

The chemical composition of each of the ceramic wastes is shown in Table 3. In both cases, high levels of silica (SiO_2) and alumina (Al_2O_3) were found and the sum of both compounds was 79.8% in the tile waste and 66.5% in the brick waste. In addition, both wastes had a relatively high Fe_2O_3 content and in both, the sum of SiO_2, Al_2O_3 and Fe_2O_3 was greater than the 70% that Mohammed indicates as an important parameter for encouraging pozzolanic activity (10). They also had moderate amounts of lime (CaO) and magnesia (MgO).

Waste	Chemical composition (%)								
	SiO_2	**Al_2O_3**	**CaO**	**MgO**	**K_2O**	**Fe_2O_3**	**SO_3**	**LOI**	**Other**
BRICK (L)	49.9	16.6	9.7	5.5	4.4	6.5	3.3	2.4	1.8
TILE (B)	61.2	18.6	5.8	1.8	3.3	5.0	0.09	0.7	3.5

Table 3. *Chemical composition and amorphous content of the ceramic wastes*

The micrographs taken by scanning electron microscopy (SEM) are shown in Figure 1. In both shots, the morphology of the ceramic waste particles is seen to reveal irregular particles with a smooth surface. According to Mohamed's study, this result, together with small particle size, also favours pozzolanic reactivity (10).

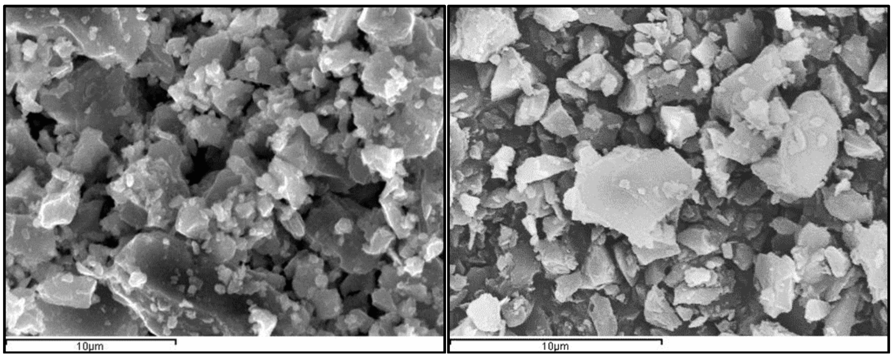


***Figure 1**. Micrographs of brick particles (L) and ceramic tile (B) particles*

3.2. PROPERTIES OF THE FRESH CEMENT MORTARS

3.2.1. MORTAR CONSISTENCY

The results of the consistency of pastes made by replacing Portland cement with ceramic wastes are shown in Figure 2: very little variance is seen in the mortars prepared with ceramic tile waste, whereas in those made with brick waste, the variation is much greater, which implies a significant loss of workability.

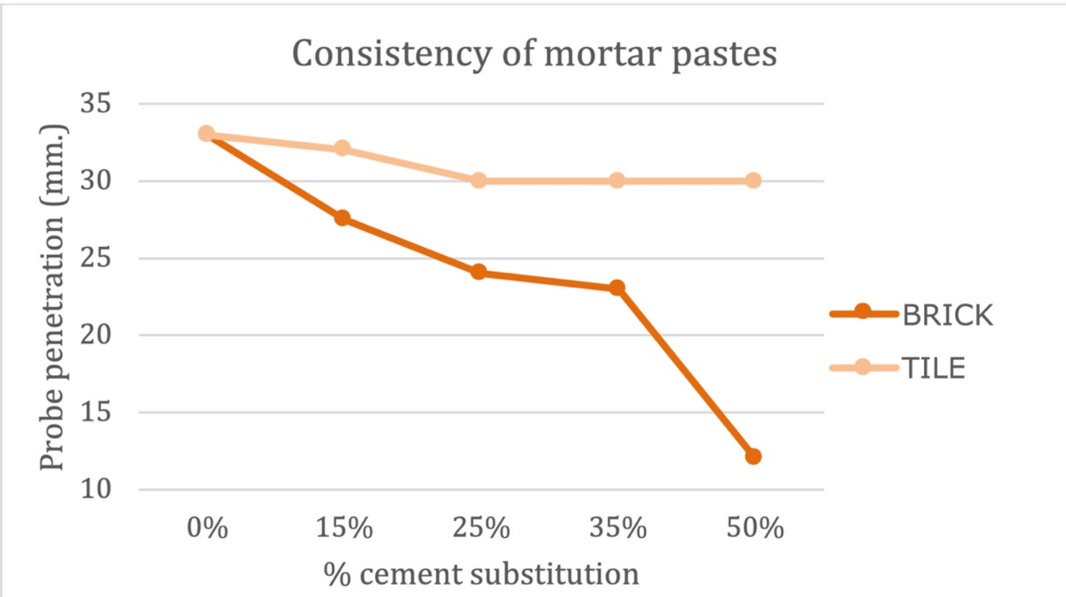


Figure 2. Consistency of pastes with cement replaced by waste

As seen in the previous section, the morphology of the two types of waste is similar (Figure 1), so that the variability in consistency may be due to differences in water absorption driven by the lower firing temperature for bricks compared to ceramic tiles. These results match those previously obtained by Pitarch et al. (4), in which the same wastes were used to replace natural aggregates when making structural concrete and in which water absorption values of 15.76% and 6.28% were obtained for brick and ceramic tile wastes, respectively. The results also agree with those cited by Mas et al. (8) for concrete prepared with mixtures of Portland cement and tile waste, in which the consistency rate in mortars with more than 50% tile waste showed very little variability.

3.2.2. SETTING TIME

Figure 3 depicts the variation of setting start times (PF) and end times (FF) for each type of waste and percentage of cement replacement.

As may be seen, PF generally increases when part of the Portland cement is replaced with ceramic waste, going from 150 minutes in the control mortar to 175 minutes in the case of the paste made with 50% substitution with tile waste.

FF varies depending on the waste used in the substitution: when brick waste is used as the substitute, FF times remain fairly constant and are below the reference cement, whereas with the tile waste, FF levels go down progressively as the percentage of cement substitution increases.

Work instruction RC-16 does not establish requirements for the final setting time of common cements, although Instruction RC-08 does. In all cases, the conditions established by the Reception of Cement Instruction (RC-08) (11) are met, which calls for a PF time of longer than 1 hour and less than 12 hours.

In addition, the results of the pastes prepared with cement substituted by brick match those obtained by Naceri et al. (9), who observed decreases in PF and FF in cements manufactured with partial substitutions using brick waste of more than 20%. It also aligns with the findings of Pereira-de-Oliveira et al. (7), who observed a reduction in consistency rates in pastes prepared with substitute ceramic wastes.

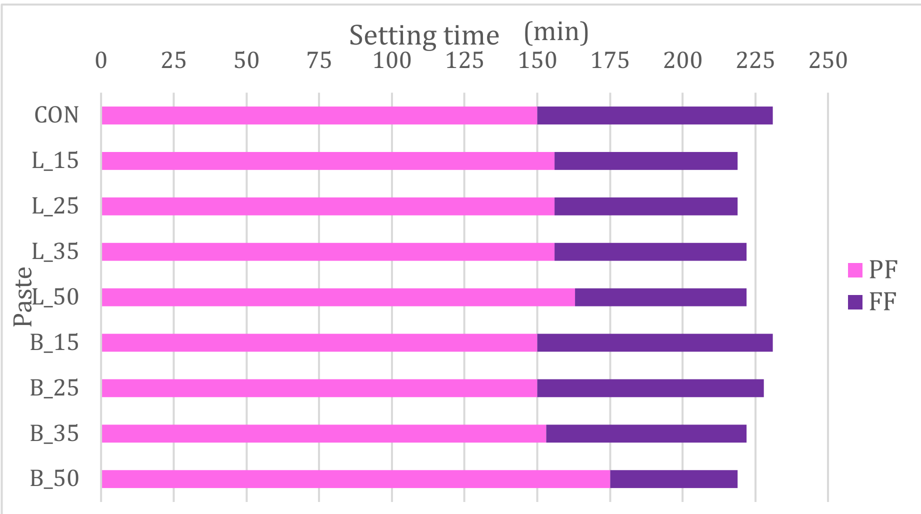
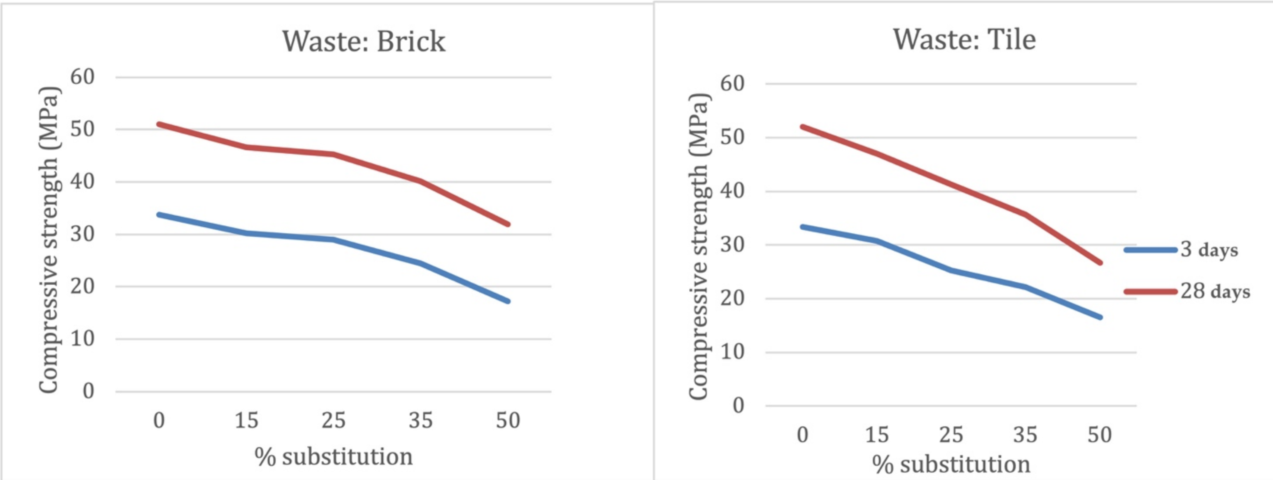


Figure 3. Variations in setting time.

The length of setting time in the various cement pastes, i.e. the time elapsing between the beginning and the end of setting, is shown in the violet area in Figure 3. In preparations with brick waste as the substitute, setting time is seen to be shorter than the control mix but varies little as the percentages of substitute waste change, whereas in mixes with tile waste as the substitute, setting time gets shorter as the amount of waste increases.

3.3. COMPRESSIVE STRENGTH OF HARDENED MORTAR

The results for compressive strength in each of the mortars at the two break ages are shown in Figures 4 and 5.



Figures 4 and 5. Compressive strength of mortars with brick and tile waste

The results of the compressive strength testing of mortars with partial substitutions up to 25% of the cement content with brick waste are more like those of the control mortar, even at an early age (3 days' curing). In order to better evaluate the above information, Strength Gain in mortars with waste substitutes was calculated (Equation 2, Figures 6 and 7).

Mortars made by partially substituting Portland cement with brick waste (Figure 6) show strength losses at 3 days of around 10% when the cement content substituted is up to 25%. At 28 days, strength loss in mortars made with larger proportions of cement substitution is seen to have come down in all cases to below 40%. These results are consistent with those obtained by Naceri et al. (9), who observed a loss of compressive strength when more than 10% of the clinker was substituted by brick and that the results improved at later ages.

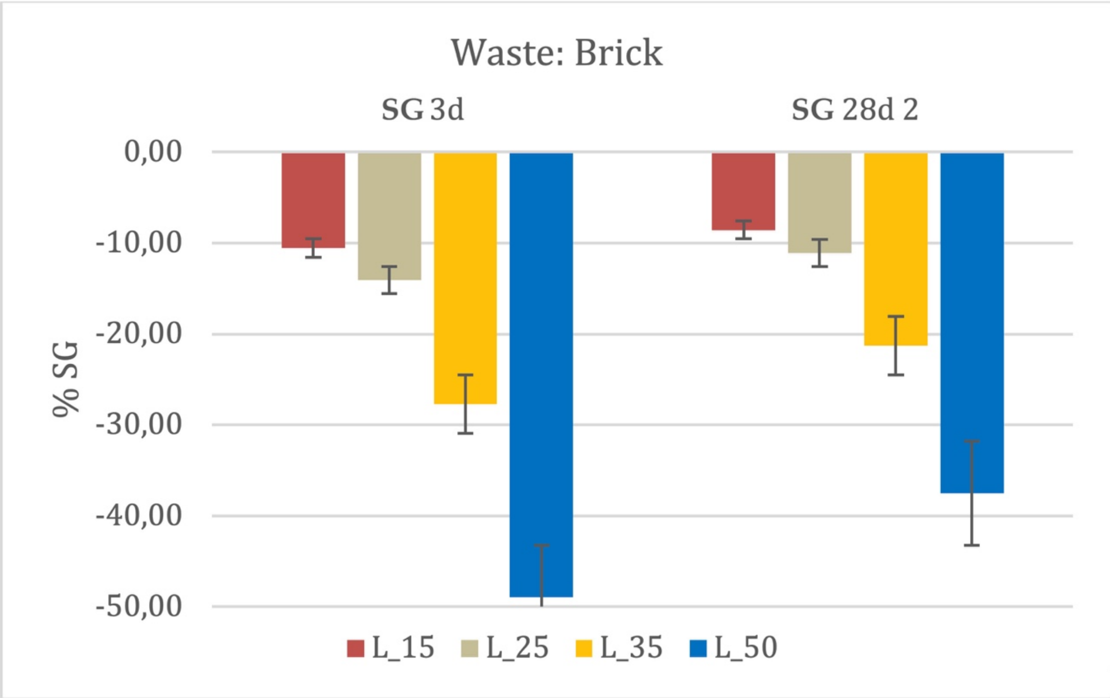


Figure 6. Strength gain in mortars with brick waste

In the case of mortars manufactured with the Portland cement content being partially substituted with ceramic tile (Figure 7), after 3 days, only those made with 15% of tile showed losses of less than 10%, while strength loss in all other cases was greater than 20%. At 28 days, these losses had been reduced, but less so than with the brick substitute. This result is similar to the one obtained by Pereira-De-Oliveira et al. (7), who found a loss of strength at 28 days close to 30% for mortars with 40% tile waste.

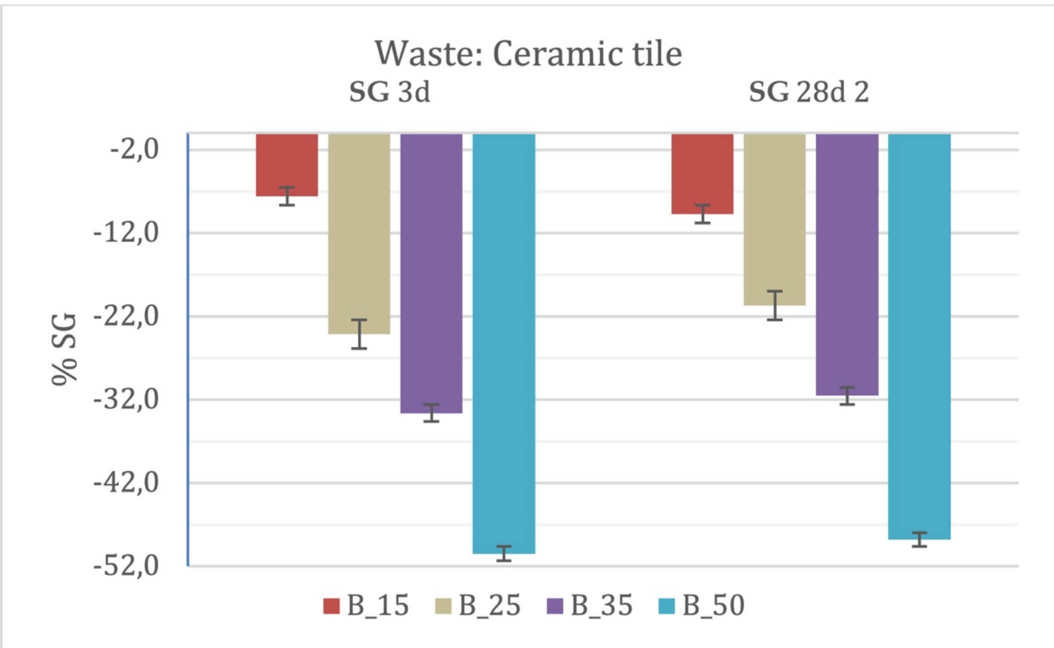


Figure 7. Strength gain in mortars with ceramic tile

4. SAI

Calculating Strength Activity Index (SAI), (Equation 1, Figures 8 and 9), provides a tool with which to assess the pozzolanic activity of materials added to Portland cement. It is based on the standard for fly ash in concrete, UNE-EN 450-1:2013 (12). This standard sets 75% and 85% as the lower limits for SAI at 28 and 90 days respectively.

At 3 days, better values in SAI were seen in mortars made with the ceramic tile substitute. However, at 28 days, SAI results were better in mortars made with brick waste: even those with 35% brick waste exceeded the limit value of 75% at that age, whereas only cements up to 25% of tile substitution managed to exceed that value.

These results differ from those achieved by Pereira-de-Oliveira et al. (7), who concluded that brick waste did not provide any pozzolanic activity. They only partially agree with those obtained by Mas et al., who noted in their study that mortars with substitution rates of 15% to 35% managed to meet the requirements of the standard at 28 and 90 days (8). They also noted the formation of portlandite during hydration.

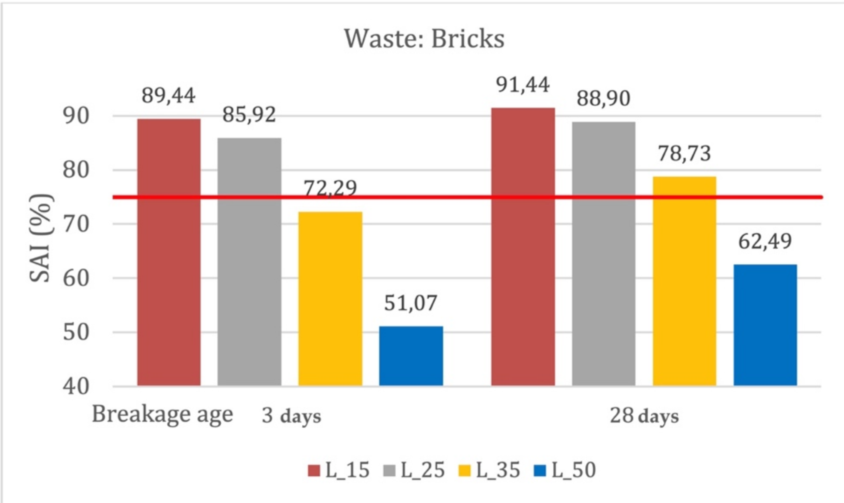


***Figure 8**. SAI (%) according to age and % brick substitution*

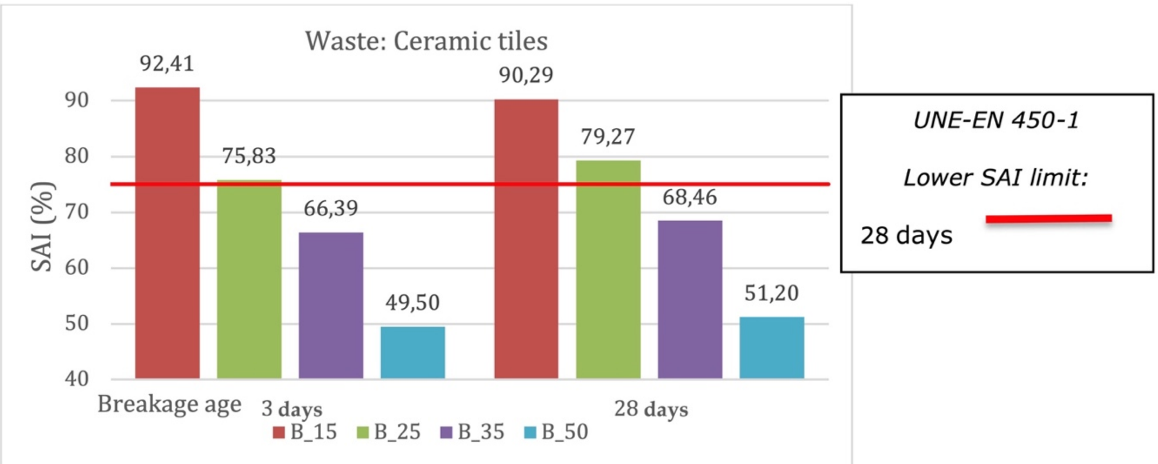


***Figure 9.** SAI (%) according to age and % ceramic tile substitution*

Even though further microstructural studies are required to explain the performance observed in our study, it may be due to a reduction in the effective A/C ratio (generated by the greater absorption capability of brick due to lower firing temperatures), and/or to greater pozzolanic activity.

5. CONCLUSIONS

As a final conclusion, manufacturing mortars with up to 25% of the Portland cement being substituted by either type of waste can be considered feasible and therefore enables both ceramic waste from construction to be reused while also reducing the amount of cement required.

Both wastes have similar particle morphology. Furthermore, in their composition, they have high percentages of silica and alumina, which are necessary for a pozzolanic reaction to take place with the portlandite released when Portland cement is hydrated.

The high SAI values obtained at 28 days for mortars prepared with up to 35% of the cement substituted by brick waste and up to 25% with ceramic tile waste indicate the existence of pozzolanic activity in both cases at that age. Such pozzolanic activity would be greater in the case of the brick waste.

Compressive strength after 3 and 28 days curing in the mortars made with brick waste was higher than in those made with ceramic tile waste. Mortars made with 15% and 25% cement substituted by brick show strength losses at 28 days of nearly 10% compared to the control mortar. In mortars manufactured with cement substituted by tile, only those in which 15% of the cement was substituted returned the same values at 28 days.

In view of the results obtained, it can be inferred that with longer curing times, the results for compressive strength could be very good. Recommended future work would compare performance of both types of waste at later ages.

Pastes in which the cement component is partially replaced with brick waste vary more in consistency compared to those made with tile waste and become less workable as the proportion of cement replaced increases.

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