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Alkali-silica reactions in alkali-activated mortars containing reactive WEEE glasses

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Abstract

The behaviour of waste glasses from discarded fluorescent lamps (WEEE, waste of electric and electronic equipment) that have been proved extremely reactive in Portland cement composites is evaluated in alkali-activated binders. Metakaolin mixed with sodium hydroxide and sodium silicate is the selected binder. Accelerated test according to ASTM C1260 and longer experiments performed at lower temperature (38°C partially according to ASTM C1293) have been carried out to study the possible reactivity. Moreover, the tests at 80°C have also been performed in calcium hydroxide saturated solution to evaluate the possible effect of calcium ions diffusion in the matrix. In all cases, no clearly expansive gels were formed and consequently the level of expansion is limited.

***Keywords**: lamp glass; metakaolin; curing conditions; reactivity to ASR.*

1. INTRODUCTION

Alkali-silica reactions are one of the most detrimental sources of degradation in traditional cementitious composites based on Portland cement. The reaction takes place when the aggregates contain amorphous silica and a sufficiently high concentration of alkalis is present in the matrix either deriving from the mixing cement or diffusing from the external environment as in the case of de-icing salts. The mechanism leading to the consequent formation of expansive gels, able to induce cracking in the materials, are yet not completely understood [1, 2]. Alkali-activated binders are now considered a sustainable and more eco-friendly alternative to the use of Portland cement [3, 4], because of its huge carbon dioxide footprint and on the opportunity of producing them even from wastes. Not so many studies have investigated the reactivity of aggregates containing amorphous silica in these composites [5-8]. In theory, the high concentration of sodium and potassium ions, as well as the high value of pH in the material, should exacerbate these reactions. Indeed, preliminary studies [5, 7] seem to underline a reduced reactivity of potentially reactive natural aggregates in alkali-activated or geopolymeric composites, even if the results are not fully consistent. In the present contribution, the behaviour of waste glass deriving from the separate collection of discarded fluorescent lamps is evaluated. This material, milled to micrometric size, has been used in traditional cementitious composites or in geopolymers as a reactive filler [9 -11] or as a fine aggregate in Portland cement composites [12-13]. As fine aggregate, lamp glass turned out to be extremely reactive in Portland matrix: indeed, it had been proved that lamp glass could expand in Portland composites even without the presence of a high alkalis concentration in the surrounding matrix [12]. The possibility of depressing the reactivity by formulating metakaolin / Portland blends could be one solution as found in other materials [14], but in the present research, the reactivity of lamp glass used in substitution of natural sand as a fine aggregate is examined in alkali-activated binders deriving from metakaolin, thus eliminating the use of Portland. Accelerated test have been used to investigate the reactivity. On account of the effect of calcium hydroxide on the expanding gel formation [15, 16] measurements have been performed also in calcium hydroxide saturated solution at 80 °C. Dimensional stability and microstructure, through Scanning Electron Microscopy are investigated on mortars submitted to all the accelerated curing in different conditions of temperature and chemical environment. The possible inertness of reactive aggregates in this class of materials, not already exploited on large-scale building structures, could further promote their use enlarging the environmental benefits.

2. MATERIALS AND METHODS

2.1 Binder

Metakaolin (Argicem, France) with D_{10} = 4.81 μm, D_{50} = 40.78 μm, D_{90} =117.83 μm was used as precursor.

2.2 Activators

The sodium silicate solution (Ingessil, Verona Italy) used was a viscous liquid produced for the cement industry with a water content of 56 wt%, the SiO_2/Na_2O oxide composition ratio of 2.07 and a density of 1.53 g/cm^3. An 8 M water solution of sodium hydroxide was used as the second activator.

2.3 Aggregates

Discarded fluorescent lamps have been supplied by COREVE (ITALY). The glass fragments had been previously treated to eliminate toxic elements. Discarded items are deprived of metallic parts, then crashed to reduce their size. Fluorescent powders are eliminated by blowing and mercury is finally distilled in vacuum. The final chemical composition (wt%) of the glass, derived by x-ray fluorescence is the following: SiO_2 68.47, Al_2O_3 2.26, Na_2O 17.65, K_2O 1.61, CaO 5.13, MgO 2.98, PbO 0.79, BaO 0.95. The flat shaped cullets have the following dimension: D_{90} = 2.9 mm, D_{50}= 1.85

mm D_{10} = 0.26. Figure 1 shows the morphology of the investigated cullets.

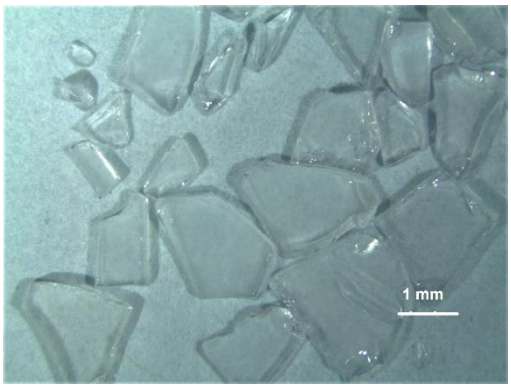

Figure 1: Morphology of the cullets

2.4 Mixing and curing

Mortars were made by first mixing the activators that were cooled to room temperature. Afterwards, first the metakaolin and the aggregate were added and, as a last component, the amount of water that allowed to obtain a suitable workability (Table 1). The mixing operations followed the EN 196-1, although this standard is formulated for traditional cementitious composites.

Table 1: Composition and codes of the investigated mortars (wt%)

Sample	Binder	Amount	NaOH (8N)	Na_2SiO_3	Water	Glass
MTK	Metakaolin	100	30	30	10	225

After 14 days of curing at 25 ± 1 °C and 60 ± 2 % R.H., four different curing procedures were performed on the specimens:

1) According to ASTM C 1260, i.e. at 80°C in a 1 N NaOH solution (16 days);

2) At 38 °C and 95 ± 2 R.H. % partially according to ASTM C1293 (360 days);

3) At 80 °C in water saturated with $Ca(OH)_2$ (16 days).

4) At room temperature (25 ± 2°C and RH. 60 ± 5%) for 360 days.

2.5 Test

Dimensional stability

Dimensional stability was measured with a comparator at scheduled times. Presently, the reported data at 80°C refer to 16 days of curing. For the samples cured at 25 and 38°C, data refer to 360 days of curing.

SEM

The fractured surfaces of the samples were sputtered with graphite and submitted to morphological analysis. An XL20 (FEI) scanning electron was used (accelerating voltage 20- 25 kV).

Mechanical characterization

Flexural strength (three points) was performed according to the EN 196 in a 100 kN Wolpert Amsler equipment.

3. RESULTS

Figure 2 reports the expansion of all mortars cured in the different conditions at the longest investigated curing time, which is 12 months for 38 °C and 16 days for 80°C tests.

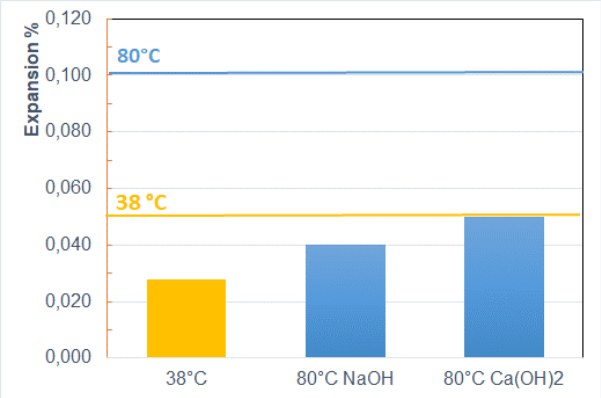

Figure 2: Recorded expansion of mortars in the different curing conditions

As can be seen, the expansion of both samples cured at 80°C remains under the limit (< 0.1%) defined by the standard. The expansion of samples cured at 38 °C in 95 ± 2 R.H. for 360 days stands again below the limits of the standard. It is clear that the results should be considered as indicative of the behaviour towards expanding reactions, in the absence of a consolidated specific standardization for these materials. Moreover, measurements at 80 °C will be repeated and carried out at longer times. For comparison sake, Figure 3 compares data referring to the expansion of lamp glass in Portland cement mortars, derived from previous research [12]. Within the cited limits, it is however quite evident that the reactivity of lamp glass in Portland cement matrix is drastically higher than that in metakaolin. The experiment carried out in calcium hydroxide solution, although leading to slightly higher values, does not change the non-expanding character of the aggregate.

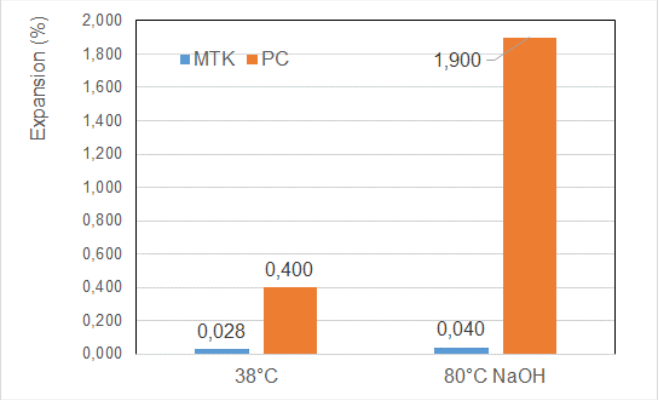

Figure 3: Comparison between the different binders (MTK metakaolin, PC Portland cement)

Figure 4 shows the flexural strength of samples cured in different conditions and at different times. All the accelerated curing conditions tend to increase, instead of decreasing, the mechanical properties of the mortars which again can be considered as evidence of the absence of remarkable deleterious expansive reactions in the samples. It should also be considered that different chemical reactions, just involving the matrix, can improve the mechanical strength of the material.

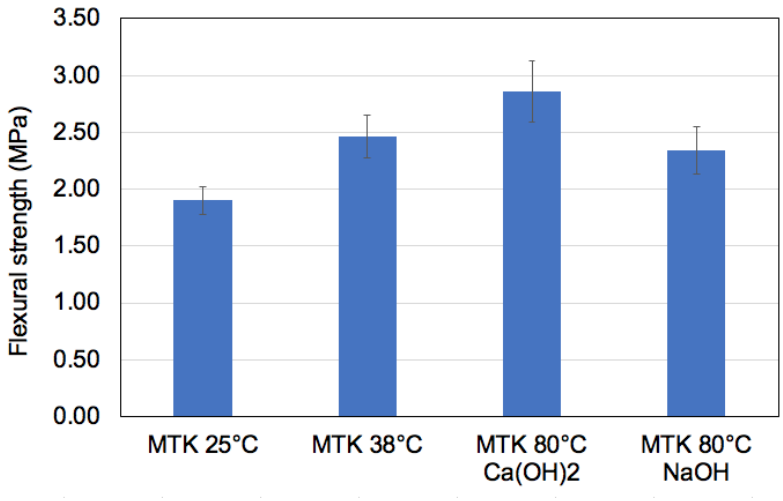

Figure 4: Flexural strength in the different curing conditions

The morphology of the glass aggregates in the different conditions is reported in Figure 5. At room temperature, at the longest curing time, the surface appears as completely unreacted. At 38 °C, a different morphology starts forming and becomes more developed in samples cured at 80 °C in NaOH or $Ca(OH)_2$ solutions. Layers of low thickness are present on the surface of the aggregate, without evident signs of damage at the interphase with the matrix. Lamp glass has a high solubility in the alkaline environment [12] and consequently complex chemical conditions may be present at the ITZ. Similar results, i.e. the formation of reacting products without swelling properties were found also in other research concerning glass bottles [5]. EDS analysis will be performed to investigate the chemical composition of the reacting layers.

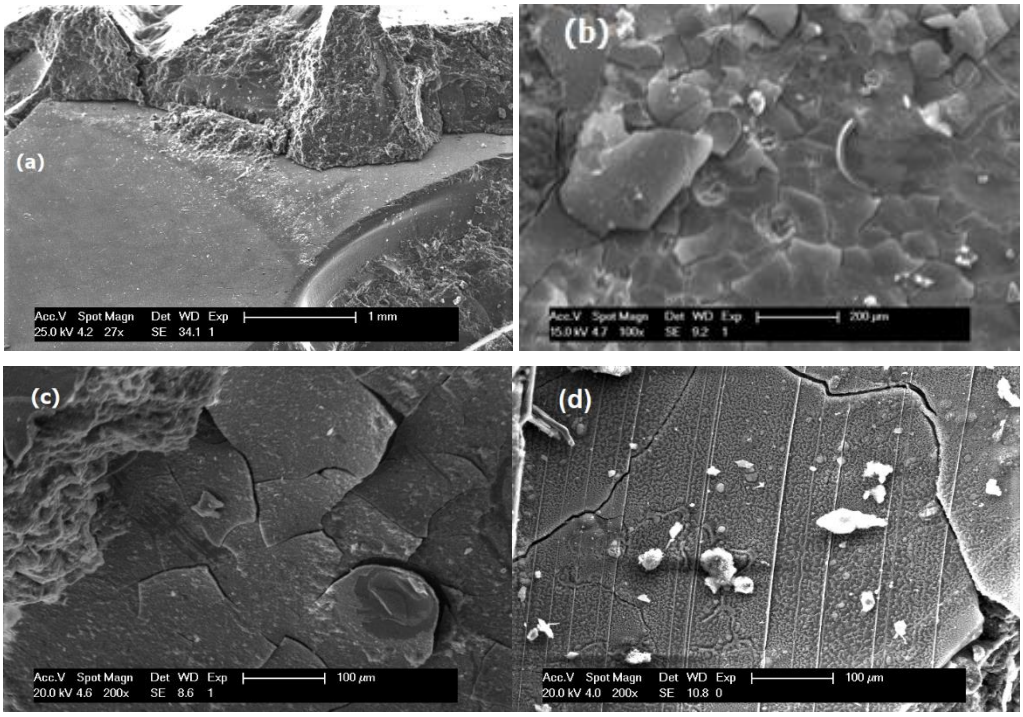

Figure 5: Aggregate surface at different curing times and conditions: (a) 12 months at 25 °C RH 65 ± 4 %, (b) 12 months at 38 °C 96 ± 2, (c) 14 days at 80 °C in 1N NaOH solution, (d) 14 days in a $Ca(OH)_2$ solution at 80°C

4. CONCLUSIONS

Preliminary data show that a glass composition (deriving from WEEE lamp waste) discloses a moderate if negligible expansive behaviour in the alkali-activated matrix. The same cullet showed instead an uncommon expanding behaviour in the traditional Portland cement composites. Although chemical reactions take place on the surface of the aggregate at the highest investigated temperatures, the derived products seem to have no swelling tendency. The precise nature of the reactions is not presently determined, also on account of different reactions taking place in the matrix.

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