



# Glass waste as supplementary cementing materials: The effects of glass chemical composition



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## ABSTRACT

Sustainable cements containing 25 wt% of different types of recycled glass have been investigated as a supplementary cementing material in order to highlight the role of glass chemical composition during the hardening process. Glass formers, stabilizers and modifiers regulate the glass dissolution in the alkaline environment during cement hydration. As a consequence, pozzolanic reaction and/or alkali–silica reaction are strictly related to the glass chemical composition. The mechanical and microstructure characterizations of mortar samples containing glass blended cements and un-reactive aggregates allow to determine which oxides in the glass have to be carefully monitored to avoid deleterious reactions.

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## 1. Introduction

Every year, thousands of tons of end-use glass are collected separately from urban wastes all over the world. Recycled glass derived from containers has been used both as fine aggregate in concrete [1–4] and supplementary cementing materials (SCMs) [5–7], on account of its amorphous nature and high silica content. Soda-lime glass has been the first and most investigated glass due to the large amount of containers in urban separated collection. However, end-use glass may come from many sources other than containers. Fluorescent lamps, television screen (cathode-ray tubes), and liquid crystal displays [8–10] represent sources of cullet that could potentially be used as SCMs, as their amount in glass separated collection is rapidly increasing. Indeed, it is very important to investigate how the chemical composition of different glass sources may influence the SCM behavior, promoting or inhibiting pozzolanic activity and/or alkali–silica reaction (ASR).

Glass dimension is also a key point for its potential expansion behavior. According to the literature [2–6] the accepted conclusion is that below a certain dimension, ASR does not take place. It was reported that ground soda-lime glass with dimension <300 μm can safely be introduced in concrete [6]. For larger glass particles, the parameter that can trigger the expansive reaction can be the

presence of microcracks [11]. When glass coming from pharmaceutical containers or crystal house-ware is used as partial natural sand replacement, ASR occurrence is influenced by glass chemical composition [12].

When different types of end-use glass are taken into account, the presence of heavy metals (Pb, Sb, etc.) shall also be considered carefully. So far cement matrix has been considered an effective tool for stabilization/solidification processes for the final treatment of hazardous waste in order to reduce contaminant leaching prior to landfill disposal [13–15]. Only few investigations have been carried out about the mechanisms occurring between cement hydration products and heavy metals [16–19]. Ion exchange between heavy metals and calcium coming from cement matrix was proposed as the leading mechanism when heavy metals are present as soluble salts (e.g.  $\text{Pb}(\text{NO}_3)_2$ ).

The aim of this work is to study the effect of glass chemical composition on cementitious materials properties to establish if end-use glass, also containing PbO, can safely be used as SCM. For this purpose glass coming from fluorescent lamps (LMP, PbO = 0.8 wt%), funnel glass from cathode ray tube (FNL, PbO = 18.0 wt%) and crystal items (CR, PbO = 27.0 wt%) have been used as 25 wt% cement replacement. The behavior of these supplementary cementing materials, never investigated before, has been tested in terms of: (i) solubility in high alkaline solution in order to simulate the behavior in a cement matrix and understand the role played by the different oxides; (ii) capacity for alkali–silica reaction promotion; (iii) ability in developing pozzolanic activity.

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The relevance of this research is based on two main features. On the one side, glass recycling is becoming increasingly more efficient and reaching high percentage of re-use, including applications in the construction field. On the other side, the quality of glass waste stream is becoming progressively worse and rich in different types of glass. This effect is particularly marked for urban glass separated collection, which is carried out in different ways (specific waste bin or door to door collection). With respect to the past, citizens pay more attention to urban waste separate collection, however they do not distinguish the different types of glass yet [20].

Thus, the *ex-ante* evaluation of the effects of particular oxides in glass composition can be fundamental for avoiding the occurrence of potentially harmful conditions in recycling routes.

Finally, some environmental considerations must also be taken into account especially for funnel glass that is classified as Waste Electrical and Electronic Equipment (WEEE). Funnel glass is the internal part of cathode ray tube (CRT), which consists of four parts: the panel used as screen (a very homogeneous barium strontium glass, of a greenish-blue color, whose weight is about 2/3 of the whole CRT), the funnel or cone (the hidden part inside, a lead glass, whose weight is about 1/3 of the whole CRT), the frit (the connection between panel and funnel, a low melting temperature lead glaze), and the neck (a glass with a very high lead content enveloping the electron gun). For its high content of PbO (up to 22 wt%), the management of funnel glass is quite difficult both for its recovery and landfill disposal. The current recycling process, which is applied only to a part of the spent lead glass, requires very high energy consumption as temperatures higher than 1000 °C are usually necessary for extraction, evaporation, and removal of Pb from the SiO<sub>2</sub> glass network. In this framework, the valorization of funnel glass as supplementary cementitious materials can be particularly important as a recycling approach to lower its environmental impact.

## 2. Experimental procedures

### 2.1. Materials

Four types of glass were investigated: (1) crystal glass (CR), coming from production of tableware, giftware and home décor items (kindly supplied by CALP, Colle di Val d'Elsa (SI), Italy); (2) funnel glass (FNL) derived from cathode ray tubes (kindly supplied by Relight (Milan, Italy)); (3) glass coming from end-use fluorescent lamps (LMPs) (kindly supplied by the National Consortium for collection and treatment of low consumption exhausted lamps, Ecolamp, Milan, Italy); (4) soda-lime glass (SL) derived from urban glass separated collection (kindly supplied by the National Consortium for collection, recycling and recovery of packaging glass, Co.Re.Ve., Milan, Italy). Glass chemical compositions (oxide wt%), determined by X-ray fluorescence (ARL ADVANT'X Series), are reported in Table 1. Sodium and potassium oxides amount was also reported as Na<sub>2</sub>Oeq content.

The four types of glass were dry-ground in a laboratory agate ball mill to obtain a grain size distribution similar to that of cement. Milling time was about 120 min for all the glass types. Grain size distribution curves, determined by laser granulometry (Micrometrics, Gemini Series 2360) are collectively reported in Fig. 1. FNL, LMP and SL exhibit very close grain size distribution with an average dimension of ≈10–20 μm, whereas CR shows higher dimensions with an average size of 40 μm on account of the higher hardness of this type of glass.

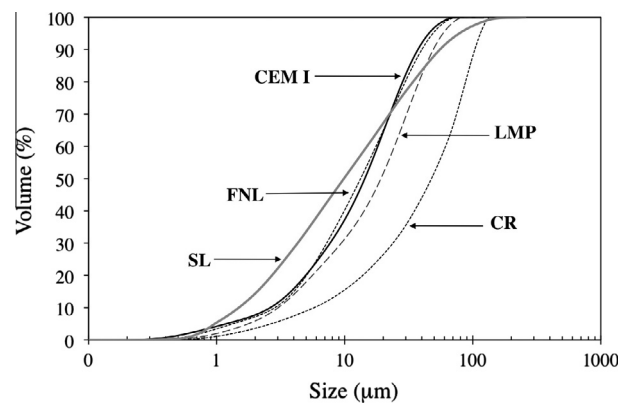
An ordinary Portland cement, CEM I 52.5 type (Italcementi, Bergamo, Italy, hereafter referred as CEM I), was used to prepare glass based blended cements. CEM I grain size distribution is reported in Fig. 1.

**Table 1**  
Glass compositions (normalized, oxide wt%).

	SL	LMP	FNL	CR
SiO <sub>2</sub>	70.40	68.47	56.11	58.64
Al <sub>2</sub> O <sub>3</sub>	2.06	2.26	3.02	0.02
TiO <sub>2</sub>	<0.01 <sup>a</sup>	<0.01 <sup>a</sup>	0.08	<0.01 <sup>a</sup>
Fe <sub>2</sub> O <sub>3</sub>	<0.01 <sup>a</sup>	0.08	0.09	0.21
CaO	11.30	5.13	2.56	0.12
MgO	1.47	2.98	1.86	0.29
Sb <sub>2</sub> O <sub>3</sub>	<0.01 <sup>a</sup>	0.08	0.17	0.22
ZnO	<0.01 <sup>a</sup>	<0.01 <sup>a</sup>	0.14	1.18
BaO	0.12	0.95	2.20	<0.01 <sup>a</sup>
K <sub>2</sub> O	1.21	1.61	10.01	7.21
Na <sub>2</sub> O	13.4	17.65	5.46	4.67
PbO	<0.01 <sup>a</sup>	0.79	18.34	27.43
Na <sub>2</sub> Oeq <sup>b</sup>	13.90	18.55	12.97	8.95

<sup>a</sup> Data below the instrument detection limit.

<sup>b</sup> Amount of (Na<sub>2</sub>O + K<sub>2</sub>O) expressed as soda equivalent content.



**Fig. 1.** Grain size distributions of the investigated glass and CEM I.

Silica sand with normalized grain size distribution according to EN 196-1 [21] was used as fine aggregate to prepare mortar samples.

### 2.2. Paste and mortar samples preparation

Blended cements were formulated by replacing 25 wt% of CEM I with the different types of glass reported above. Mortar samples were prepared by a Hobart planetary mixer with 1/3 binder/aggregate and 1/2 water/binder ratios, following the procedure described in EN 196-1. Mortars were named M followed by the acronym of the glass type used as SCM (e.g. MLMP identifies mortar sample containing 75% CEM I + 25% LMP as binder). Mortar samples containing 100% CEM I were also prepared and named MCEMI.

For each mix, 40 × 40 × 160 mm prisms were cast and cured for 7, 28, 60 and 90 day at 20 °C and R.H. > 90% to investigate compressive strength and microstructure.

In order to investigate the occurrence of alkali–silica reaction, additional mortar prisms were prepared and cured for 1 day at 20 °C and 100% R.H., then exposed to accelerated ASR testing immersed in 1 M NaOH solution at 80 °C for 14 days. This procedure only partially follows standard ASTM C 1260 [22], which is however designed for determining aggregates reactivity. The reliability of standardized tests to evaluate potentially alkali–silica reactive aggregates as well as the laboratory parameters influencing the final results were largely discussed in RILEM technical committee (TC 106-AAR, 1998–2000; TC 191-ARP, 2001–2005; TC 219-ACS, 2007–2012) and international literature [23–25]. As

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