



Research article

Use of ancient copper slags in Portland cement and alkali activated cement matrices

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ABSTRACT

Some Chilean copper slag dumps from the nineteenth century still remain, without a proposed use that encourages recycling and reduces environmental impact. In this paper, the copper slag abandoned in landfills is proposed as a new building material. The slags studied were taken from Playa Negra and Púquios dumps, both located in the region of Atacama in northern Chile. Pozzolanic activity in lime and Portland cement systems, as well as the alkali activation in pastes with copper slag cured at different temperatures, was studied. The reactivity of the slag was measured using thermogravimetric analysis (TGA), scanning electron microscopy (SEM), X-ray diffraction (XRD), electrical conductivity and pH in aqueous suspension and Fourier Transform Infrared Spectroscopy (FTIR). Furthermore, copper slag-Portland cement mortars with the substitution of 25% (by weight) of cement by copper slag and alkali-activated slag mortars cured at 20 and 65 °C were made, to determine the compressive strength. The results indicate that the ancient copper slags studied have interesting binding properties for the construction sector.

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1. Introduction

Industrial slags are usually classified as ferrous and nonferrous slags, and may be used in the manufacture of new materials. Slags from extracting metal processes are conventionally considered as waste materials (Nazari and Sanjayan, 2015). In construction, copper slag (CS) is used in fillings, embankments, drainage, ballast, as aggregates in mortars, concrete (Thomas and Gupta, 2013; Mithun and Narasimhan, 2015) and asphalt mixtures (Hassan and Al-Jabri, 2011), as abrasive material (Kambham et al., 2007), as a cement substitute (Shi and Qian, 2000; Moura and Coutinho, 2004; Zain et al., 2004; Al-Jabri et al., 2006; Moura et al., 2007; Taha et al., 2007; Sánchez de Rojas et al., 2008; Brindha et al., 2010; Chew and Bharati, 2010; Brindha and Nagan, 2011; Najimi et al., 2011; Peyronnard and Benzaazoua, 2011), as raw material in the manufacture of Portland cement (PC) (Ariño and Mobasher, 1999; Chockalingam et al., 2013; García Medina et al., 2006; Gorai et al., 2003; Kosmatka et al., 2003) and also as binders in the manufacture of alkali-activated (AA) cements (Deja and Malolepszy, 1989, 1994; Shi

et al., 2008; Iacobescu et al., 2013).

Regarding the use of CS aggregates, we can report that there are a few technical specifications (Federal Highway Administration Research and Technology. U.S. Department of Transportation, n.d.) and two standards governing their use as aggregates for concrete: one in Korea (Korean Standards Association, 2000) and one in Japan (Japan Mining Industry Association, 2003). The residue from the flotation tailings of CS has also been used in cement admixtures and cement manufacturing (Alp et al., 2008; Onuaguluchi, 2012).

According to the method of cooling employed, CS can be grouped into two types: air-cooled slags, which are black glass with low water absorption, and water-cooled slags which are amorphous, granulated and present greater water absorption capacity (Onuaguluchi, 2012). CS generated nowadays is composed Fe (30–40%), SiO₂ (35–40%), Al₂O₃ (≤10%), CaO (≤10%) and Cu (0.5–2.1%) (Gorai et al., 2003).

In Chile, there are CS dumps that were produced in the nineteenth century that have environmental effects on the soil (destruction of beaches and shoreline pollution) and landscape (visual pollution barriers in natural channels), as well as social (alteration of Historic Places) and economic (land use) impacts. Chilean law classifies these slags as non-hazardous waste (Ministerio de Salud de Chile, 2004). Often, the statistical analysis

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provides a basis for predicting with high probability that the composition of the CS and their leachates will remain in the non-hazardous range (Alter, 2005). Moreover, they can degrade over time, and may potentially be a source of toxic metals (Piatak et al., 2004). So far there has been no proposal to use former CS landfills and reduce these effects, or to use them as raw materials in the manufacture of mortars and concretes based on PC and/or AA cement.

The aim of this study is to determine the feasibility of using two CS produced in the nineteenth century, Playa Negra and Púquios, which are abandoned landfills located in the region of Atacama in Chile, as building materials. Pozzolanic activities in lime and Portland cement systems, as well as AA cements with copper slag cured at different temperatures, were studied. Moreover, CS-PC mortars with the substitution of 25% (by weight) of cement by CS and AA slag mortars cured at 20 and 65 °C were made, to determine the compressive strength.

2. Experimental

2.1. Materials

CS samples of the nineteenth century, originating from the landfills Playa Negra (CS_PN) and Púquios (CS_PQ), both located in the region of Atacama in northern Chile, were used. The CS_PQ is located 150 km away from CS_PN. The PC used in cement pastes and mortars, is CEM I-52.5 R type according to EN 197-1 (UNE-EN 197-1:2011, 2011). The chemical composition of the slags and the PC used are shown in Table 1. In mortars CS/PC, standard sand (Normensand, Germany) was used, while in the AA mortars silica sand (Caolines Lapiedra, Valencia, Spain) was used, according to EN 196-1 standard (UNE-EN 196-1:2005, 2005). As an activator for the AA reaction, 98% pure sodium hydroxide in pellet form (Panreac) and sodium silicate solution ($\text{SiO}_2 = 28\%$, $\text{Na}_2\text{O} = 8\%$, $\text{H}_2\text{O} = 64\%$, by weight, Merck) were used. In the conductimetric method (Tashima et al., 2014), for the testing of pozzolanic reactivity, an aqueous suspension of calcium hydroxide powder was used (minimum 95% purity, Panreac). In the manufacture of cement mortars and pastes, water from the Valencia city network and deionised water were used.

The CS was first milled using a jaw crusher; after this, a ball mill with alumina balls that were 18 mm in diameter was used. Particle size distribution was determined using a laser diffraction granulometer (Mastersizer 2000, Malvern Instruments). The final average diameter for both milled CS samples was 15 μm and 90% of particles were smaller than 40 μm after 35 min grinding time. The particle size distribution curves of both ground forms of CS are shown in Fig. 1.

The CS chemical compositions were determined by X-ray fluorescence (XRF MAGIC PRO Philips, mod. PW2400) and are shown in Table 1. The results show that the two samples of CS contain a very significant amount of iron oxides and silica. The silica content of both samples is very similar; however, the iron oxide content is higher in the CS_PN. They also have a high content of CaO, which may indicate their feasibility as a precursor material for alkali activation (Canfield et al., 2014). Moreover, it can be seen that the

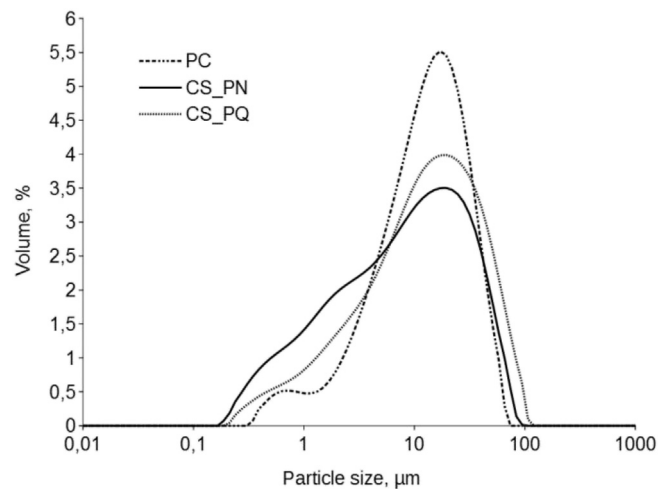


Fig. 1. Particle size distribution curve of PC and ground CS.

CaO content in CS_PQ is twice the content in CS_PN. The high content of CaO and Fe_2O_3 , lower than copper slag used in other studies (Gorai et al., 2003) is probably due to the contribution of fluxes such as silica and lime used during smelting. It is usual to add silica to capture separating copper iron during smelting, and lime gives flow properties within the furnace slag.

The X-ray diffraction (XRD, Bruker AXS D8 Advance) patterns were obtained using a $\text{Cu-K}\alpha$ radiation under conditions of 40 kV and 20 mA, in the range $2\theta = 5-70^\circ$. Fig. 2 shows that CS_PQ is fully amorphous; this is evidenced by the strong deviation of the

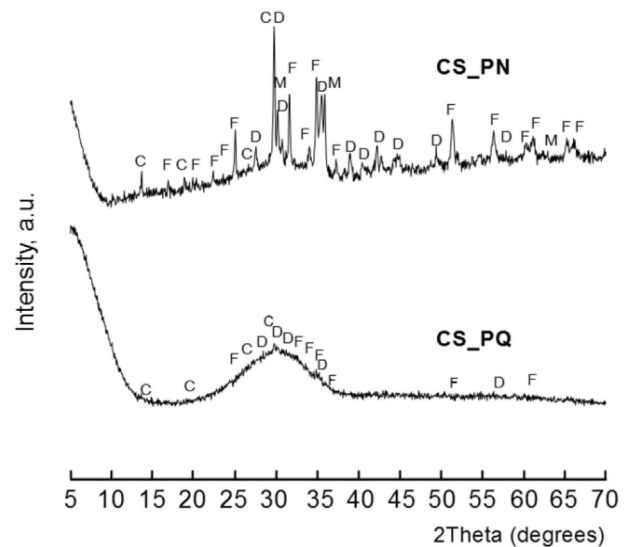


Fig. 2. X-ray diffractogram patterns of raw CS_PN and CS_PQ materials. D: Diopside (CaMgSiO_3 ; PDFcard 190239); F: Fayalite (Fe_2SiO_4 ; PDFcard 340178); C: Clinoferrosilite (FeSiO_3 ; PDFcard 170548); M: Magnetite (Fe_3O_4 ; PDFcard 190629).

Table 1
Chemical composition of PC and CS (Wt. %) and Density (g/cm^3).

	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O	Na_2O	Other	LOI	Density
PC	20.80	4.60	4.80	65.60	1.20	1.70	1.00	0.07	0.23	2.02	3.07
CS_PN	39.14	7.76	30.48	13.41	2.09	0.46	1.50	1.05	3.60	-2.11 ^a	3.16
CS_PQ	38.33	8.17	20.40	26.10	2.14	0.26	0.78	0.64	3.59	-1.50 ^a	2.96

^a Mass gain probably due to oxidation FeO and S^{2-} (sulphides) (Sánchez de Rojas et al., 2008).

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