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Experimental study of the mechanical stabilization of electric arc furnace dust using fluid cement mortars

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HIGHLIGHTS

- This article optimizes the mechanical stabilization capacity of EAFD using currently available mortars.
- The EAFD hinders the hydration process of tricalcium silicate in cement mortars.
- Mortars with EAFD have a double hydrated hydroxide of Ca and Zn instead of portlandite.
- A maximum mass ratio of 6.67 kg EAFD per kg of cement was mechanically stabilized.
- This is the highest EAFD/cement ratio ever used to mechanically stabilize this type of hazardous waste.

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ABSTRACT

This article shows the results of an experimental study carried out in order to determine the maximum amount of electric arc furnace dust (EAFD) that can be incorporated into fluid cement-based mortars to produce mechanically stable monolithic blocks. The leaching performance of all mixes was studied in order to classify them according to the EU Council Decision 2003/33/EC. Two mortars were used as reference and three levels of EAFD incorporation were tested in each of the reference mortars. As the incorporation ratio of EAFD/cement increases, the mechanical strength decreases. This is due to the greater EAFD/cement and water/cement ratios, besides the presence of a double-hydrated hydroxide of Ca and Zn ($CaZn_2(OH)_6{\cdot}2H_2O$) instead of the portlandite phase ($Ca(OH)_2$) in the mixes made with EAFD, as well as non-hydrated tricalcium silicate. A mass ratio of 2:1 (EAFD: cement-based mortar) can be added maintaining a stable mechanical strength. The mechanical stabilization process also reduced the leaching of metals, although it was not able to reduce the Pb concentration below the limit for hazardous waste. The high amount of EAFD mechanically stabilized in this experimental study can be useful to reduce the storage volume required in hazardous waste landfills.

1. Introduction

Steel can be 100% recycled [1,2]. According to the World Steel Association [3], the world's steel production in 2014 was 1670 million tons, of which around 28% were obtained from electric arc furnaces (EAF). In Spain, there are 21 EAF steel mills that produce more than 75% of the steel consumed in Spain [4].

EAF use scrap metal as raw material. During the production process, three categories of waste are generated: electrodes and refractory material, slag (EAFS) and electric arc furnace dust (EAFD) produced by purifying the gases generated. EAFD is highly toxic due to its zinc, lead and cadmium content [5]. According to European Union Decision 2014/955/UE [6], gaseous effluents containing hazardous substances are classified as hazardous waste.

The composition and physicochemical properties of EAFS and EAFD may vary greatly from one steel mill to another, since they depend on the composition and quality of the scrap metal being melted. The main chemical constituents of EAFS are FeO, CaO, SiO_2, Al_2O_3, and MgO. The contents vary within the 10–40%, 22–60%, 6–34%, 3–14%, and 3–13% ranges, respectively [7]. Sofilić et al. [5]

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found that the main elements in EAFD are Fe, Zn, Mn, Ca, Mg, Si, Pb, S, Cr, Cu, Al, C, Ni, Cd, As and Hg. The authors further indicate that most of the mass of EAFD are metal oxides, silicates and sulphates. When the scrap metal is galvanised steel, the zinc content of EAFD increases, so that the mineralogical composition of the dust consists mainly of ZnO [8]. Sofilić et al. [9] studied the mineralogical composition of EAFD from three steel mills in Croatia, concluding that the composition of the dust generated during the processing of steel in EAFs must be studied individually. De Vargas et al. [10] detected via XRD that the majority of compounds were zinc and iron oxides.

EAFD is included in the EU catalogue of hazardous waste materials due to the high content of heavy metals in its composition. The most common research areas and methods used to manage EAFD are solidification/stabilization to minimize the toxicity and leachability of heavy metals [5,11], to explore the recovery of zinc and lead from EAFD [12–15], the potential use of EAFD in ceramic matrices [16], cement and concrete matrices [17–20].

Cement is the best binder currently available for stabilization/solidification (S/S) of heavy metals, although their presence has a complex influence on the hydration reactions of cement [21–25]. De Vargas et al. [10] evaluated the incorporation of 5%, 15% and 25% wt. of EAFD in CEM-I Portland cement. These authors found a reduction of the initial mechanical strength of those cement pastes with large amounts of EAFD, although over long periods the mechanical strength recovered to 80% of the reference strength. However, for a 5% of EAFD incorporation ratio, the 28-day mechanical strength was highly affected. These authors also observed that EAFD slows the hydration reactions of Portland cement. The setting time of pastes made with 25% of EAFD was 36 h. Fares et al. [20] also found a direct relationship between the EAFD content and the setting time. These authors showed that replacing 3% of cement with EAFD increased the setting time from 6 to 33 h. Chen et al. [22] attributed the increases in the setting time to the presence of Zn^{2+}, which reacts with calcium ions of the clinker and inhibits the hydration of tricalcium silicate.

Shi and Fernandez-Jiménez [26] studied the immobilization of hazardous and radioactive waste using alkaline cements. The results were encouraging for the use of this type of binder, provided that the cements are designed correctly. These types of cement have already been investigated in a previous work [27] and resulted in mortars and concrete with high durability.

Pereira et al. [28] worked on the immobilization of EAFD using Portland cement CEM-I and CEM-II and adding fly ash type F. Better results were observed in the cement-only mix. They also studied two curing conditions, one in a laboratory environment and the other in saturated atmosphere achieved by using hermetically-sealed plastic bags. Better results were obtained when curing in a saturated atmosphere.

Pereira et al. [29] used geopolymerization techniques with reactants such as sodium hydroxide, potassium hydroxide, sodium silicate, potassium silicate, kaolinite, metakaolinite and blast furnace slag for immobilization of EAFD waste. The use of potassium silicates and blast furnace slag improved the mechanical strength, and curing at 60 °C increased the strength.

The objective of this work is to maximize the loading of EAFD per kg of cement to produce mechanically stable monolith blocks using fluid cement-based mortars, in order to minimize the storage volume of landfill disposal. The leaching performance of all monolithic blocks was studied in order to determine its acceptability in landfills. To the best of the authors' knowledge, this study tested the highest EAFD/cement ratio ever used to mechanically stabilize this type of hazardous waste. This allows a significant reduction of the storage volume required in hazardous waste landfills.

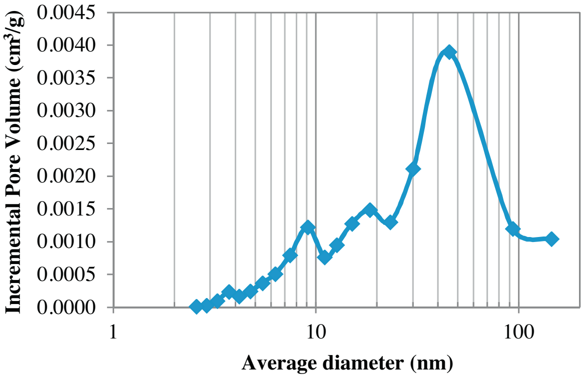

Fig. 1. Size of pores of the EAFD sample.

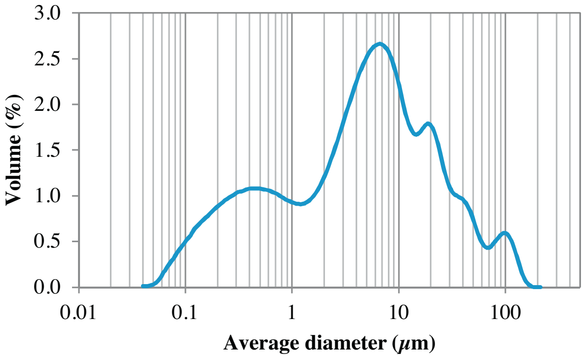

Fig. 2. Particle size distribution of the EAFD sample.

2. Characterization of the EAFD

The study was carried out on a sample of EAFD from a steel mill situated in the north of Spain. The specific surface was calculated using the Brunauer-Emmett-Teller (BET) method, determined by the absorption of N_2 with Micromeritics ASAP 2010 equipment, obtaining a value of 4.6 m^2/g. The real particle density calculated in accordance with NLT 179:1995 [30] was 3.809 g/cm^3.

Fig. 1 shows the distribution of the pores; there are more mesopores (2–50 nm) – 72% – than macropores (>50 nm) – 28%. Furthermore, the majority of the mesopores (57%) fall within a large-size range (20–50 nm).

The particle size distribution was determined by laser diffraction in a "Beckman-Coulter LS-230" equipment, with a measurement range of 0.04–2000 μm.

The size distribution of particles ranges from 0.04 μm to 200 μm (Fig. 2). The majority of the particles are below 10 μm (70%), the average particle size being 7 μm. These results have been confirmed via scanning electron microscopy (SEM) (Fig. 3), showing sizes lower than 7 μm. Sofilić et al. [5] found that the range of the majority of particles fell in the range 100–125 μm, much higher than that of this work.

The chemical analysis has been made by wavelength dispersive X-ray fluorescence (WDXRF), using a S4 Pioneer apparatus of BRUKER, with a potential of 4 kV. The SEM micrograph was obtained with a Jeol scanning electron microscope: JSM-6300 model, with acceleration potential of 20 kV and work distance of 15 mm. Table 1 shows the percentage in weight of each compound/element present in the EAFD sample, the main ones being ZnO and Fe_2O_3, and to a lesser extent CaO, PbO and MnO. Other authors obtained Fe_2O_3 as the main compound, and to a lesser extent ZnO, CaO and MnO [5].