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Original Research Paper

Study of electric-arc furnace dust (EAFD) in fly ash and rice husk ash-based geopolymers

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ABSTRACT

Electric-arc furnace dust (EAFD) is an industrial waste produced by the volatilization of metals during scrap melting in electric arc furnaces. This waste is classified as Class I – hazardous, because lead and cadmium concentrations are above the limits set in the leaching test. Processes are carried out in many countries to recover the metals contained in EAFD. In Brazil, these processes are usually not conducted in the industry because the low percentage of commercially valuable metals makes it economically unfeasible to recover them. One of the study alternatives is the use of EAFD in civil construction. Studies have shown that EAFD increases the mechanical strength of mortars and Portland cement-based concretes. However, EAFD delayed cement setting time, which can jeopardize its use in construction. Thus, this study aims to evaluate the effect of EAFD when added to fly ash (FA) and rice husk ash (RHA) based geopolymers. Geopolymer mortars were prepared at a ratio of 1:3 (FA + RHA: sand, particle size 4) and added with 0, 10, 15, and 20% EAFD in relation to the mass of FA + RHA. Compressive mechanical strength and leaching tests were carried out at the ages of 7, 28, and 91 days. Microstructural analyses were performed using XRD, FTIR, and SEM/EDS. EAFD did not negatively influence the geopolymerization process. The highest compressive strength results for the mortars containing the waste were found for 20% of EAFD. All mortars, regardless of EAFD content, were classified as non- hazardous Class II at the age of 91 days.

1. Introduction

Electric-arc furnace dust (EAFD) is a steel waste produced by the volatilization of metals during scrap melting in electric arc furnaces. Volatilized metals, including Zn, Pb, Cd, Ni, and Cr, are oxidized and subsequently solidified in the form of a fine dust with a particle size of less than 10 μm.

The amount of EAFD generated is approximately 1% of the steel produced, but in practice this amount may vary according to the scrap used and its previous processing. Worldwide, approximately 70% of the EAFD generated is disposed of in landfills, while the other 30% is processed mainly to recover metals [1]. Countries that use EAFD usually collect these oxides through a pyrometallurgical or hydrometallurgical process, or both. However, in Brazil these processes are still not in use since the percentages of heavy metals with commercial value are not economically feasible. The chemical characterization of this waste has been reported in several studies. The composition of EAFD varies depending on the raw materials used and the production process, but its main components are as follows: Fe (37–49%), Zn (4–37%), Ca (2–5%), Pb (1–2%), and small amounts (less than 1%) of other metals such as Cr, Cd, Cu, and Al. Because it has Pb and Cd concentrations above the limits established by the standard in leaching tests, EAFD is classified as a hazardous waste - Class I [2–5].

Therefore, one of the study alternatives is to use EAFD in civil construction based on solidification/stabilization technology. Studies using EAFD were evaluated as a mixture in the production of the Portland cement clinker [6,7]. Other studies have evaluated the mechanical behavior of the cementitious matrix containing EAFD [8–12]. In these studies, it was observed that the samples containing EAFD exhibited superior mechanical behavior when compared to the reference samples. However, EAFD retarded the cement hydration reactions. The main element thought to be responsible for this phenomenon is zinc [13–18].

Some authors investigated the possibility of stabilizing EAFD waste in ceramic materials. The addition of this waste, at levels of 2.5 and 5% in relation to the mass, to mixtures with industrial clays resulted in ceramic structures with a higher mechanical strength and lower temperatures required to produce the material. However, metals such as Zn and Pb may volatilize during the

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ceramic manufacturing process at high temperatures, leading to concern about the presence of these volatile metals in the atmospheric emissions of the process [19].

Geopolymerization technology has attracted attention in several applications, including solidification/stabilization of these wastes. Geopolymers result from the polycondensation reaction of silica aluminum material by alkaline activation [20–24]. Its structure consists of AlO_4 and SiO_4 tetrahydra alternately bonded by sharing oxygen atoms [25].

Diverse industrial by-products and aluminosilicate source materials were proven to be suitable for producing geopolymer materials [26]. In most cases only a small amount of silica and alumina on the particle surface is sufficient to dissolve and take part in the reaction, solidifying the entire mixture and immobilizing any heavy metal contained in it [27].

Simple [28,29] or combined [30,31] fly ashes have been studied as precursor geopolymeric materials. Rice husk ash (RHA), produced through the combustion of rice husk, has been studied as a source of silica (SiO_2) for the obtaining of geopolymers [32–36]. Shalini et al. [32] have verified that the increase in the replacement of FA by RHA in FA based geopolymers, and the use of a ground blast furnace slag (GBFS) have increased the compression strength from 15.80 MPa (10% RHA) to 18,80 MPa (30% RHA). Pieper [37] has verified that the partial replacement of metakaolin by RHA in geopolymers increased the compression strength from 5.90 MPa (0% RHA) to 11.40 MPa (30% RHA) at the age of 91 days. Detphan et al. [36] have also noticed increased compression geopolymer strength when using RHA. This may also be related to the SiO_2/Al_2O_3 (Si/Al) geopolymer molar relation. According to Davidovits et al. [38] the Si/Al molar relation is an important variable of geopolymers precursor materials. Since geopolymers are Si-O-Al bonds, the arrangement of these bonds will depend fundamentally on the Si/Al relations found in precursor materials (raw materials and alkaline activators). Therefore, the use of RHA may be an important source of SiO_2 for geopolymers.

The immobilization of toxic metals by geopolymerization was studied by Xu [39]. Geopolymer matrices based on FA and metakaolin containing Cd(II), Cu(II), Pb(II), and Cr(III) have been investigated. The authors identified a number of factors that may influence the immobilization of these metals such as the concentration of the activating alkaline solution.

The stabilization of EAFD through geopolymerization technology was analyzed by Fernández Pereira [40] using various raw materials such as kaolin, metakaolin, and blast furnace slag. In general, geopolymers with stabilized metals showed high compressive strength and small percentages of Zn and Cr extraction in leaching tests, demonstrating that these metals were practically immobilized. The fractions of immobilized Pb and Cd, on the other hand, were highly variable according to geopolymer composition.

Experimental investigations of geopolymerization in the presence of EAFD were also carried out by Nikolic et al. [41]. The chemical immobilization of zinc among aluminosilicate bonds was indicated by the presence of Zn in the amorphous phase of the geopolymer, but a certain amount of this metal was also observed in particles of raw material that did not react, which means that immobilization took place partially through physical encapsulation.

Thus, this study aims to evaluate the effect of EAFD when added to fly ash (FA) and rice husk ash (RHA)-based geopolymers using solidification/stabilization technology.

2. Materials and methods

2.1. Materials

The EAFD used was generated by a semi-integrated steel plant and collected by means of a socket filter. The chemical composition of the EAFD used in this research is shown in Table 1. The total amount of the EAFD sample used was obtained by mixing and homogenizing five 10 kg samples of dust collected over a period of 2 months, totaling 50 kg. No milling was performed. The EAFD grading curve was carried out using a 1064 Cilas laser diffraction grain meter. The mean diameter was 0.81 µm and 90% of the particles were smaller than 3.27 µm. Fig. 1 shows a granulometric distribution of EAFD.

ABNT NBR Standard NM23/2001 procedures [42] were used to determine specific gravity. The result was 4.33 g/cm^3.

Table 2 shows EAFD leached extract – ABNT NBR standard 10,005/2004 procedures [43].

Based on the results of Table 2, EAFD is classified as Class I – Hazardous Waste because Pb and Cd concentrations are above the maximum limits allowed by ABNT NBR standard 10004/2004 [44].

Scanning electron microscopy and energy-dispersive X-ray spectroscopy (SEM/EDS) was carried out to evaluate the particle morphology of EAFD. Fig. 2 shows an EAFD image.

As seen in Fig. 2, EAFD morphology shows a predominance of agglomerated spherical particles, where the smaller particles form aggregates or cover larger particles, which is in agreement with the EAFD observed by Škavara et al. [45]. Semiquantitative analyses by EDS were carried out at points 1 to 16. The EDS results in Table 3 also confirm the findings obtained through the X-ray Fluorescence (XRF) (Table 1) and X-ray diffraction (XRD) analysis (Fig. 3) of EAFD composition, which show the predominant presence of Zn and Fe with lesser amounts of Pb and Cr. Oustadakis et al. [46] found that Zn is mainly associated with smaller and agglomerated particles, probably as zincite. In larger spherical-shaped particles it is possible to confirm the presence of franklinite. Magnetite, however, represents the smaller spheres distributed homogeneously in the EAFD.

A diffraction spectrum of the EAFD X-ray is shown in Fig. 3. Crystalline phases of franklinite ($ZnFe_2O_4$), zincite (ZnO), hematite (Fe_2O_3), and quartz (SiO_2) were identified. Such phases were also found in the EAFD in other works [5,47,48].

The Fourier Transform Infrared Spectroscopy (FTIR) spectrum of the EAFD is shown in Fig. 4a. Table 6 shows the vibration frequencies and assignments for EAFD.

The RHA used was an ash sold in 20 kg packages. The chemical composition of RHA is shown in Table 1. The specific gravity of the RHA was 2.05 g/cm^3 and the average particle diameter was 9.23 µm. The RHA spectra of FTIR and XRD are shown in Figs. 4b and 5a, respectively. Table 6 shows the vibration frequencies and assignments for RHA.

The FA utilized for this study was obtained from a coal-fired thermoelectric power plant located in Southern Brazil. According

Table 1
Chemical composition of EAFD, FA and RHA determined by X-ray Fluorescence (XRF) analysis (wt.%).

Elements	EAFD	FA	RHA
Fe_2O_3	43.45	4.92	0.09
ZnO	31.89	0.05	0.01
SiO_2	3.71	64.07	91.96
MnO	1.96	0.02	0.45
CaO	1.83	1.47	0.70
PbO	1.48	-	-
K_2O	1.23	2.63	2.12
SO_3	1.06	0.35	0.09
Cr_2O_3	0.74	0.04	-
MgO	0.56	0.21	0.14
Al_2O_3	0.35	22.95	0.30
CuO	0.30	0.01	-
P_2O_5	0.26	0.05	0.54
NiO	0.05	0.01	-

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