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Structural ceramics containing electric arc furnace dust

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HIGHLIGHTS

- Zn is stabilized due to formation of $ZnAl_2O_4$ spinel and/or willemite type phases.
- EAFD/clay fired mixtures exhibit improved mechanical properties.
- Hollow bricks were successfully fabricated from the mixtures studied.
- Laboratory articles and scaled up bricks found as environmentally inert materials.

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ABSTRACT

In the present work the stabilization of electric arc furnace dust EAFD waste in structural clay ceramics was investigated. EAFD was collected over eleven production days. The collected waste was characterized for its chemical composition by Flame Atomic Absorption Spectroscopy. By powder XRD the crystal structure was studied while the fineness of the material was determined by a laser particle size analyzer. The environmental characterization was carried out by testing the dust according to EN12457 standard. Zn, Pb and Cd were leaching from the sample in significant amounts.

The objective of this study is to investigate the stabilization properties of EAFD/clay ceramic structures and the potential of EAFD utilization into structural ceramics production (blocks). Mixtures of clay with 2.5% and 5% EAFD content were studied by TG/DTA, XRD, SEM, EN12457 standard leaching and mechanical properties as a function of firing temperature at 850, 900 and 950 °C. All laboratory facilities maintained 20 ± 1 °C. Consequently, a pilot-scale experiment was conducted with an addition of 2.5% and 5% EAFD to the extrusion mixture for the production of blocks. During blocks manufacturing, the firing step reached 950 °C in a tunnel kiln. Laboratory heating/cooling gradients were similar to pilot scale production firing.

The as produced blocks were then subjected to quality control tests, i.e. dimensions according to EN772-17, water absorbance according to EN772-6, and compressive strength according to EN772-1 standard, in laboratory facilities certified under EN17025. The data obtained showed that the incorporation of EAFD resulted in an increase of mechanical strength. Moreover, leaching tests performed according to the Europeans standards on the EAFD-block samples showed that the quantities of heavy metals leached from crushed blocks were within the regulatory limits. Thus the EAFD-blocks can be regarded as material of no environmental concern.

1. Introduction

Electric arc furnace dust (EAFD) is listed as a K061 hazardous waste by the United States Environmental Protection Agency US EPA and it is considered as an intensely growing waste stream on a global scale, as it is directly related to the steel production on scrap recycling facilities.

In USA, out of 58.196 million tons (Mtons) of steel produced in 2009, 0.90 Mtons of EAFD was generated. From this amount 0.60 Mtons is further processed for metals recovery (primarily zinc) in the USA, while the remainder (0.3 Mtons) is either treated prior to disposal in landfills or exported [EPA 2009 Assessing the Management of Lead in Scrap Metal and Electric Arc Furnace Dust]. Approximately 15–20 kg of EAFD is generated per ton of steel produced. In 2011 EU steel production was 177.431 Mtons. In Greece all five steel factories are scrap-recycling facilities. During the years

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2009–11 the annual mean Greek steel production was 2.0 Mtons [1].

EAFD is produced from the volatilization of heavy metals when steel scrap is melted in the electric arc furnace. Volatilized metals are oxidized and subsequently solidified and detained in the form of fine powder in specially designed filters, which are placed in the electric arc furnace gas stream cleaning system. Thus EAFD is formed as a very fine dust with particles of a few micrometers. This material is listed as a hazardous waste in the EU waste catalog with the code 100213 and exhibits ecotoxicity due to its high content of inorganic pollutants including, among others, Zn, Pb, Cd, Ni and Cr. The composition of this waste varies depending on the scrap utilized. The EAFD studied in this work is of high zinc content (>15%) with a potential of recycling for metal recovery [2]. Recently the environmental hazard of EAFD was evaluated using various National leaching tests (Brazilian, French, USA, Japanese, German and Netherlands) [3] but not by the European standard (EN12457). Many solutions have been proposed for the technical utilization of the steel dust as applied for other hazardous materials [4,5]. The most promising from the environmental point of view are the ceramic and vitrification processes. However very few studies are available in open literature regarding the utilization of EAFD in the industry of traditional clay ceramics [6]. Just recently Machado et al. [7] investigated the stabilization of iron rich, low lead (<1.3%) Brazilian EAFD in fired mixtures with clay up to 1100 °C. In 2011 the successful utilization of Waelz slag (20–30 wt%) in commercial size ceramic blocks (hollow bricks) has been reported by Quijorna et al. [8]. Waelz slag is a byproduct containing the nonvolatile components of the original EAFD. It is a low zinc and lead material consisting primarily of iron oxide (>50 wt%). Quijorna et al. studied thoroughly physicochemical, mechanical and environmental properties of the manufactured blocks. They focused in the environmental consequences of the product life cycle and Waelz slag incorporation proved to be environmentally favorable without affecting quality of the final ceramic product.

The annual Greek clay block production over the past years has been approximately 3,500,000 tons. Considering that 15–20 kg of EAFD is produced for every ton of recycled steel, the Greek annual EAFD waste production is calculated to be no more than 40,000 tons. Such quantity can be easily used in clay construction ceramics production by a low percentage (2.5–5%) of EAFD introduction in the blocks composition. This could be an approach of industrial ecology, of course taking into account particular optimization, as there are various fired clay construction products, as well as types of clay mixtures and firing cycles used.

On the other hand several other waste materials have been incorporated to clay blocks and tiles for stabilization studies such as electrolytic MnO_x–FeO_y waste [9], fly ash, ferro-alloy slag, phosphogypsum [10], organic and inorganic solid waste, bauxite red mud, metallurgical solid waste [11], sludge [12,13] and contaminated river sediments [14], etc. [15,16]. More studies are available regarding EAFD stabilization by vitrification despite the drawback of high energy consumption [17]. Kehagias and coworkers have recently reported chemically durable glass-ceramic EAFD structures [18,19].

In the present study, 11 EAFD samples of a Greek steel production unit have been collected and studied. In most cases in literature a single EAFD sample is studied although the raw materials in steel recycling process are of variable origin and properties, resulting in waste of corresponding compositional fluctuation. The aim of this work was to investigate from the environmental point of view, the case of stabilization of such a hazardous high metal content waste by the formation of clay ceramics that could meet the EU legislation regarding contaminant migration to the environment. At the same time, the utilization of such a waste in a high through put production line such as the construction blocks industry without affecting the quality of the product is of interest. Thus a pilot scale clay ceramic blocks fabrication was performed and the products were tested under the EU standards EN771-Specification for masonry units in particular for clay blocks in laboratory facilities certified under EN17025. Environmental characterization was also performed over the final products according to EN12457 European Standard described in the relevant EU Council Decision 19.12.2002 [20].

2. Materials and methods

The EAFD waste was collected through 11 production days. Additionally one sample was prepared after homogenization of five subsequent production days waste. All day collections together with the 5 days mixture were characterized for their chemical composition (Table 1). The chemical composition was determined by means of Flame Atomic Absorption Spectroscopy (FAAS) using a Varian AA280FS apparatus and Ion Chromatography (Dionex

Table 1
Composition (wt%) of EAFD powder in oxide compounds and extrusion clay mixture.

	Day 1	Day 2	Day 3	Day 4	Day 5	Mix 1–5	Day 6	Day 7	Day 8	Day 9	Day 10	Day 11	Clay
Sample:	D1	D2	D3	D4	D5	M[a]	D6	D7	D8	D9	D10	D11	
ZnO	30.21	28.24	30.25	33.77	21.34	28.74	33.69	33.36	18.38	20.47	18.49	35.13	n.d.
PbO	6.54	7.17	7.53	5.87	3.28	6.04	6.47	6.01	4.76	5.13	5.71	6.37	n.d.
CdO	0.08	0.09	0.10	0.09	0.03	0.07	0.15	0.13	0.05	0.05	0.06	0.15	n.d.
Cr_2O_3	0.2	0.25	0.24	0.33	0.23	0.22	0.29	0.24	0.19	0.24	0.23	0.28	n.d.
CuO	0.25	0.31	0.34	0.37	0.20	0.31	0.35	0.34	0.36	0.39	0.32	0.30	n.d.
Fe_2O_3	10.04	13.52	15.77	14.64	5.55	11.88	18.91	16.74	35.13	32.86	32.73	15.6	5.51
NiO	1.77	2.07	1.62	1.57	2.69	1.93	2.29	1.75	3.65	4.17	3.41	0.03	n.d.
MnO	1.3	1.52	1.01	1.53	1.84	1.37	1.52	1.09	1.41	1.45	1.31	0.82	n.d.
K_2O	2.73	3.01	3.62	2.47	2.07	2.74	3.21	2.77	2.17	2.43	2.61	1.66	2.10
Na_2O	3.43	3.84	3.10	2.93	5.80	3.78	2.83	2.85	3.24	3.83	4.13	1.4	0.46
MgO	1.98	2.10	1.10	1.19	2.78	1.9	1.44	1.33	1.76	2.17	1.92	1.53	1.91
CaO	12.6	10.73	7.52	6.82	9.91	9.54	4.66	3.69	6.78	8.06	8.81	8.60	6.01
Al_2O_3	3.36	1.64	0.89	0.79	2.49	1.85	0.81	0.69	1.60	1.40	2.32	1.77	15.62
SiO_2	4.00	4.02	3.55	3.51	8.39	4.69	3.96	3.58	5.31	4.37	4.35	Traces	56.45
CoO	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	n.d.
SO_4^{2-}	4.3	4.72	4.04	3.14	12.05	5.62	3.98	4.59	2.18	2.08	2.24	3.88	n.m.
F^-	0.11	0.09	0.04	0.02	0.21	0.08	0.07	0.05	0.06	0.06	0.05	0.07	n.m.
Cl^-	5.01	4.88	5.04	5.09	4.31	4.9	4.87	4.98	4.64	4.21	4.31	4.91	n.d.
C	–	–	–	–	–	–	–	–	–	–	–	–	3
LOI (900 °C)	12.5	12.22	14.56	15.64	16.33	14.4	10.65	15.58	7.95	7.05	6.88	17.83	8.28

[a] Day 1–Day 5 physical mixture; n.m., not measured; n.d., not detected.