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Laboratory investigation of Waelz slag stabilization

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ABSTRACT

Electric Arc Furnace dusts are considered hazardous waste due to their high heavy metals content (zinc, lead, etc.). The Waelz process is one of the most efficient technologies, in terms of capacity and quality, able to recover nearly the 90% of zinc contained in such EAF dusts. Unfortunately, the resulting slag still has eco-compatibility problems, although its mechanical and chemical properties are suitable for civil engineering applications. Stabilization tests, by quartz addition, were performed on EAF dusts in a laboratory furnace. Temperature, treatment time and cooling rate were varied in order to define the best conditions for regulating the formation of a stable microstructure able to hinder the release of substances. Microstructural characterization was carried-out using SEM and XRD analysis. Leaching tests were performed according to EN 12457-2 standards and water analyses were performed using ICP-OES. The best chemical stability was achieved when dust powders were mixed with 20% of silica and water-cooled.

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Keywords: EAF dusts; Waelz process; Waelz slag; Stabilization process; Silica addition; Leaching test

1. Introduction

EAF dusts (EAFD) are one of the most important raw materials used to extract impure zinc oxide even though they are considered hazardous waste by both the European and American environmental protection agencies, as indicated in the [European Waste Catalog \(2002\)](#) and [Catalog of Hazardous and Solid Waste \(2004\)](#). The main reason for this classification is the high content of heavy metal (lead, cadmium and selenium) that can be easily released into the environment and cause serious health problems.

In the European Union, steel industries produce more than 1,000,000 tons/year of EAFD. This amount can be recycled by the Waelz process, which converts the dust into impure zinc oxide (Waelz oxide), which is then reprocessed in metallurgical plants, i.e. the imperial smelting process. The Waelz process is the most diffused in the world and is noted in the BREFs notes (Best Available Techniques Reference Document) of the [European community](#) as the Best Available Technology (BAT) for EAF dusts treatment (2013). After mixing EAFD with coke breeze (a reducing agent) and other additives (lime or siliceous sand), this charge is continuously

introduced in a rotary kiln. The high temperatures reached in the furnace allow the reduction and the vaporization of zinc and other volatile metals (lead and cadmium) to recover the Waelz oxide after oxidation-condensation processes. In Europe, the total amount of zinc, recovered through this technique, is about 250,000 tons/year. The molten residues, featured by low zinc and lead concentration, are water cooled at the end of the rotatory kiln to form granular Waelz slag. The recycling of all of the dusts produced in the EU could lead to the generation of nearly 800,000 tons/year of Waelz slag.

1.1. Waelz process

In a Waelz rotary kiln, three different zones can be identified ([Fig. 1](#)): in the first part, the temperature reaches approximately 600 °C allowing for the removal of charge humidity. In the second part, the temperature increases up to 1100 °C and chlorides start to volatilize and zinc-ferrites begin to decompose. Moreover, zinc, lead and iron oxide start to be reduced. In the last zone, the temperature reaches approximately 1300 °C and zinc and iron oxide are highly reduced. Zinc vapours are

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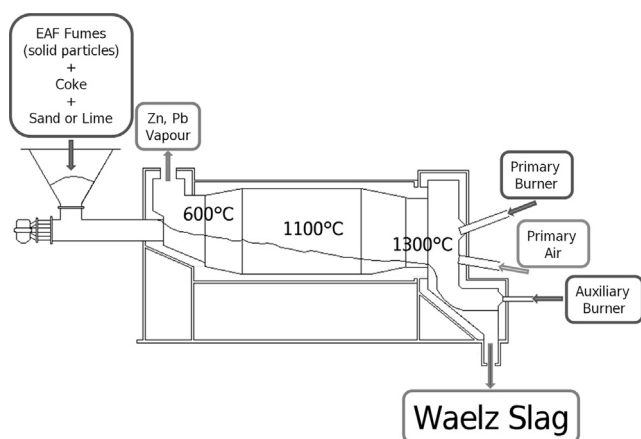


Fig. 1 – Waelz rotary kiln scheme.

aspired by an exhaust chimney and subsequently oxidized when they come in contact with air, producing the Waelz oxide.

Two different operative procedures can be followed during dusts treatment in the Waelz process: the addition of lime (basic running) or silica (acid running). In the first case, the kiln refractory duration is privileged whereas in the latter case, the Waelz slag reaches higher chemical stability and significantly reduces the lead releasing. Usually, the addition of silica is in the range of 14–18% by weight (Busè et al., 2014).

1.2. Waelz slag

Waelz slag is essentially an iron-reduced slag. Based on the mixture and composition of the material charged in the furnace, the operating conditions and the characteristics of the slag cooling, two different types of slag are obtained: silica-rich and lime-rich slags (Barna et al., 2000).

Even though the most of the zinc, lead and cadmium are removed and recovered during EAFD treatment, the Waelz slag is still classified as a dangerous and toxic waste due to excessive concentration of As, Cd, Cr⁶⁺, Cu, Pb, Ba and V (Barna et al., 2000). In particular, a typical Waelz slag produced after the basic running process is mainly formed by larnite (2CaO.SiO₂), calcium-ferrite, magnetite and wustite (Vegas et al., 2008; Quijorna et al., 2014). Specifically, calcium silicate, as reported by Barella et al. (2012) and Mombelli et al. (2013), is one of the phases major involved in heavy metal leaching, especially barium and vanadium.

Different studies have aimed to reduce the environmental impact of slag (in particular the EAF slag) through additional thermal, chemical or thermo-chemical treatments during or after the deslagging process. In particular, cooling rate effects on the leaching behaviour of steel slag were analyzed by Shinoda et al. (2008) and Tossavainen et al. (2007). They demonstrated that rapid cooling could improve slag stability, especially if coupled with correct chemical composition. MgO (Tossavainen et al., 2007) or combined (FeO + Al₂O₃ + MgO) (Drissen et al., 2009) additions were also studied in order to reduce chromium leaching, but with a detrimental effect on Ba, V and Se release. The addition of silica during deslagging operations, investigated by Barella et al. (2012) and Mombelli et al. (2012, 2013), seems to be the best technique to assure a safe and stable slag microstructure that avoids the release of the most dangerous elements. Since the polluting elements are generally bound to the silicates, reducing their amount or reducing their solubility are two ways to overcome the

problem. This could be achieved increasing the polymerization degree of the structure, namely increasing the number of SiO₄ tetrahedra. Specifically, the resistance of a silicate to hydration and the hydrolysis processes (in neutral conditions) increases with the degree of oxygen sharing between SiO₄ polyhedra of the mineral structure (Brantley et al., 2007).

A laboratory investigation was performed with the prospective of reducing the environmental impact of Waelz slag after disposal and expanding the possibility of its use in civil engineering applications as a replacement for conventional materials (Sorlini et al., 2004; Quijorna et al., 2011, 2012, 2014). For the Italian regulation, the recycle of hazardous waste coming from metallurgical industry requires the compliance of very strict limit on heavy metal leaching reported in “D.M. 03 August 2005 N. 201: definition of the criteria for waste acceptance at landfills” and “D.M. 05 April 2006 N. 186: identification of non-hazardous waste subject to simplified recovery procedures”. Italian laws specify the leaching test to be performed to assess the leaching behaviour for such a by-product: EN12457-2 standard leaching test in deionized water.

The investigation activities were focused on defining the additive amount that should be added to EAFD in the Waelz process to avoid the release of dangerous chemical species.

In particular, following the same approach proposed for EAF slag stabilization, different pure crystalline quartz (SiO₂) amounts were tested, in a range between 17.5 and 22.5% by weight, as well as different thermal treatment times and cooling rates.

2. Experimental procedure

2.1. EAF dusts characterization

The EAF dusts were taken from a steel mill and their chemical composition was measured by XRF and checked by SEM-EDS analysis. The dust used in the experiment was sieved to obtain a size distribution of about 0.5–3 mm.

Crystallographic analysis and phases identification were carried out using a Philips PW 3710 diffractometer on EAFD powder obtained by ring mill grinding. The X-ray analyses were performed using K α 1.5418 Å radiation produced by copper tubes excited by 40 kV and 30 A. Data was collected from 10° to 90° 2 θ with a step size of 0.02° 2 θ . Peaks identification was performed by exploiting Crystal Impact Match software through the use of the Crystallographic Open Database (COD).

The morphological and microstructural characterization was performed by a Zeiss EVO50 Scanning Electron Microscopy (SEM), equipped with an Oxford Inca EDS probe. The EAFD were compacted and mounted in epoxide resin, ground and polished. SEM analysis was carried out using a backscattered electrons probe (BSE) in order to identify and check the different phases pointed out by XRD. The general and local chemical compositions were measured with an EDS probe.

2.2. Melting temperature identification

The preliminary investigations were performed using a wettability furnace, in order to identify the melting temperature of EAFD and to evaluate the optimal silica addition.

The solid residuals contained in the fumes were pulverized by a ring mill and mixed with different amounts of SiO₂ (0, 10, 20 and 30% by weight). For each condition, 50 mg of

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