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Recycling of electric arc furnace dust through dissolution in deep eutectic ionic liquids and electrowinning

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HIGHLIGHTS

- Ionic liquid (1 choline chloride:2 urea) dissolved about 60% of Zn and 39% of Pb present in EAF dust.
- CV studies on the electrolyte formed showed distinct deposition and stripping peaks for Zn and Pb.
- Zn–Pb alloy was electrowon by application of high static potentials determined from CV diagrams.
- Lowering Zn and Pb contents allows recycling the dust in iron/steelmaking and in cement synthesis.

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ABSTRACT

The dust waste formed during steelmaking in electric arc furnace (EAF) is rich in ferrous and nonferrous metals. Recycling of this dust as a raw material in iron or steel-making is hazardous and therefore it is mostly dumped. This paper demonstrates recycling of EAF dust through selective dissolution of metal oxides in a deep eutectic ionic liquid. It was found that about 60% of Zn and 39% of Pb could be dissolved from the dust when stirred for 48 h in 1 choline chloride:2 urea ionic liquid at 60 °C. The resultant electrolyte was subsequently fed to a conventional three-electrode cell where cyclic voltammetry (CV) measurements were conducted to describe its electrochemical behavior. Two deposition peaks were determined and ascribed to deposition of zinc and lead. Static potentials were successively applied to electrowin metallic zinc. SEM/EDX investigations showed that the zinc electrowon contained remarkable contents of lead.

1. Introduction

Electric arc furnace (EAF) is used for steelmaking from ferrous scraps, as the main charge material, mixed with pig iron and/or direct reduced iron. It produces between 10 and 25 kg of dust per ton of steel [1–5]. This dust consists of metal oxides, lime, and silica. It contains mainly iron, zinc, lead, chromium, and cadmium. Presence of toxic elements such as Pb, Cr, and Cd has led the EAF dust to be categorized as hazardous waste by Environmental Protection Agency (EPA) in United States [1]. The EAF dust is also listed as hazardous waste in the European Waste Catalog [6]. The toxic elements Pb, Cd and Cr leach in water exceeding their maximum limits in groundwater, and this necessitates treating the EAF dust before landfilling [1,5,7] or storing the dust in appropriate places protected from rain [3].

The type of scrap melted in the EAF predominantly determines chemical composition of the dust generated. When galvanized steel scraps are used in the EAF, most of the zinc ends up in the dust and fume due to its very low solubility in the molten steel and slag [3]. Zinc has higher vapor pressure at steelmaking temperatures, and consequently vapor zinc leaves the furnace along with other gases and fumes of species volatilized at the hot spots in the arc zone and oxygen jet zone, in addition to sucked droplets by bubbles burst at the liquid bath surface [2]. In the meanwhile, simple and complex compounds, like ZnO, PbO and ZnF_2O_4, are formed [1–4]. The gas stream is finally treated and the dust is collected by filters.

The main quantitative constituents in the dust are iron and zinc [3]. The latter has low content when the galvanized steel is not intensive in the scrap used in the EAF [5]. The low zinc-bearing iron dust can be recycled in the charge of EAF [8] or blast furnace [9]. However, the use of high zinc-bearing iron dust in the charge of blast furnace leads to such undesirable consequences as

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Table 1
Typical chemical composition (in wt.%) of EAF dust, as-received and after dissolution in the ionic liquid.

Oxide	CaO	SiO_2	MgO	Al_2O_3	MnO	Fe_2O_3	ZnO	PbO	K_2O	TiO_2	Cr_2O_3	P_2O_5	SO_3	F	Cl	Br	NiO	CuO	SrO
As-received	30.31	3.62	2.81	0.73	2.68	15.58	25.18	2.80	2.10	0.11	0.14	0.27	4.16	0.23	8.91	0.10	0.08	0.15	0.04
After dissolution in ionic liquid	31.18	11.18	3.99	1.98	4.54	30.45	10.05	1.71	0.30	0.44	0.28	0.49	2.13	0.43	0.54	–	0.09	0.17	0.05

an increase in the zinc content of the pig iron and the formation of crusts inside the furnace that interfere the normal operation. Moreover, zinc penetrates the furnace lining through deformation and disintegration and leads to its attack [9].

These aspects, combined with growing concern about environmental issues, led researchers and steelmakers to develop several approaches for recycling EAF dust. These approaches are mainly categorized under hydrometallurgical [10–19] and pyrometallurgical processes [8,9,20–33]. Additionally, various applications for reuse of the EAF dust have been suggested such as incorporation in glass and ceramic products [34–40] and incorporation in synthesis of cement [41–46]. However, among the methods suggested for recycling EAF dust, pyrometallurgical processes are still the ones which have been applied in industry [9,31,32]. Nevertheless, about 60% of the EAF dust generated worldwide is still being dumped [9,31] due to problems associated with pyrometallurgical processes such as high capital and operating costs, loss of iron in slag and higher impurity content of zinc grade produced [9].

As a new approach for using non-aqueous solutions to dissolve selective metals from oxide mixtures, a recent study [47] has investigated the leaching of blast furnace dust using carboxylic acids. The solution dissolved zinc from the iron-rich dust so that the dust can be reused directly in the ironmaking process. However, because the Zn content in the blast furnace dust was too small to be economically processed, the study did not present any trial to recover Zn from the leaching solution. Other approaches have been developed for using air and water-stable ionic liquids along with aqueous solutions in multi-step procedures to extract some ferrous and non-ferrous metals [48], palladium [49], uranium oxide [50], and rare earth elements [51].

In a more practical approach, a new class of environmentally friendly ionic liquids has offered not only the possibility of selective dissolution of some metal oxides, but also the subsequent electrodeposition of the metals [52–55]. This new class of ionic liquids, namely deep eutectic ionic liquids or deep eutectic solvents, is based on combination of choline chloride with hydrogen bond forming compounds such as urea and ethylene glycol [56]. Deep eutectic ionic liquids have been successfully used for electrodeposition of Zn from ZnO [52,53] and from $ZnCl_2$ [57,58].

The aim of the present study was to verify a new approach by which zinc and lead-bearing compounds were dissolved from EAF dust in a deep eutectic solvent, namely 1 choline chloride:2 urea ionic liquid. This was followed by electrowinning of zinc layer containing remarkable contents of lead. The electrodeposition of metallic phase from the ionic liquid was characterized by cyclic voltammetry measurements. In addition, the microstructure and composition of the electrowon metal was described by SEM/EDX investigations. The study included also the assessment of the chemical composition of EAF dust, as-received and after dissolution experiments, by X-Ray fluorescence (XRF).

2. Materials and methods

2.1. EAF dust

The EAF dust investigated in this study was obtained from Suez Steel Company, Suez, Egypt. The major oxides composition of as-received dust was investigated by X-ray fluorescence (XRF), and listed in Table 1. In addition, its mineralogical composition was detected by X-ray diffraction (XRD), see Fig. 1.

2.2. Ingredients of the ionic liquid

The deep eutectic ionic liquid consisted of choline chloride and urea. Choline chloride ($HOC_2H_4N(CH_3)_3^+Cl^-$, Alfa Aesar, 98+%) and urea (NH_2CONH_2, Alfa Aesar, 98+%) were used as received without further purification or drying. They were handled in the lab atmosphere in open-to-air conditions without using glove box.

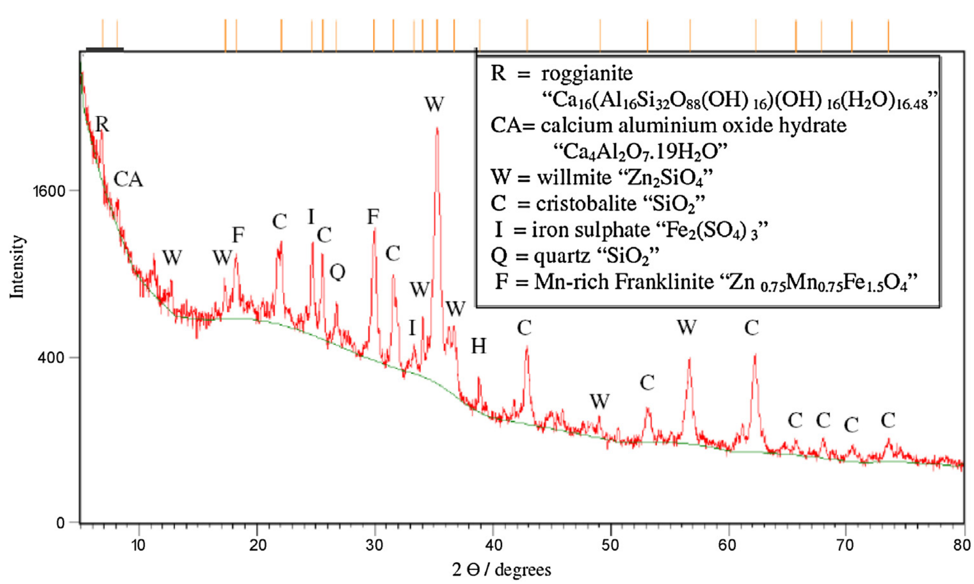

Fig. 1. XRD pattern of the as-received EAF dust