Journal of Hazardous Materials 274 (2014) 87–97

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Microwave treatment of electric arc furnace dust with PVC: Dielectric characterization and pyrolysis-leaching

Mohammad Al-harahsheh[a,*], Sam Kingman[b], Leema Al-Makhadmah[c], Ian E. Hamilton[b]

[a] Chemical Engineering Department, Jordan University of Science and Technology, Irbid 22110, Jordan
[b] Faculty of Engineering, University of Nottingham, Nottingham NG7-2RD, UK
[c] Department of Environmental Engineering, Al-Hussein Bin Talal University, Maan 71111, Jordan

HIGHLIGHTS

- Physical, chemical, mineralogical and dielectric characterization was carried out for EAFD, PVC and their mixtures.
- The mixture of PVC and EAFD is heated well by microwaves.
- Microwave heating was efficient in terms pyrolysis time (only 2-3 min was required).
- Microwave pyrolysis-leaching of EAFD-PVC yielded high recovery of Zn, Pb, and Cd.
- The residue contains only magnetite, hematite and carbonaceous material.

ARTICLE INFO

Article history:
Received 25 January 2014
Received in revised form 10 March 2014
Accepted 11 March 2014
Available online 24 March 2014

Keywords:
EAFD
Microwave treatment
Leaching
Zinc extraction
PVC pyrolysis

ABSTRACT

Microwave treatment of electric arc furnace dust (EAFD) with poly(vinyl chloride) (PVC) was studied in this work. A comprehensive characterization of the dust as well as assessing the suitability of using the thermal de-chlorination of the common plastic (PVC) under inert atmosphere was carried out to assess the possibility of Zn and other heavy metals extraction (Pb and Cd) from EAFD. The dielectric and thermal properties of EAFD, PVC and their mixtures were measured. Once combined and heated the metal oxides present in the dust reacted with HCl released from PVC during thermal de-chlorination, forming metal chlorides which were subsequently recovered by leaching with water. It was found that zinc chloride could be almost completely recovered in the leaching stage, with the overall recovery of Zn reaching 97% when the EAFD:PVC ratio was 1:2. The investigation highlighted that franklinite, the most refractory mineral to leaching, was completely destroyed. The leaching residue was found to compose mainly of magnetite and hematite.

1. Introduction

Electric arc furnace dust (EAFD) is classified as an environmentally hazardous waste according to the Environmental Protection Agency (EPA) [1]. It consists of heavy metals oxides such as, Zn, Pb, Cd, Mn, Na, Cr and Fe. The use of galvanized steel in areas such as the automotive and construction industry, has increased the zinc content in the dust over the last few years [2]. 20–30 wt.% of the EAFD is zinc in the form of zinc oxide and zinc ferrite. It is estimated that about 5–7 million tons EAFD is generated each year worldwide [3]. The continual generation of this dust has the potential to be a viable source for zinc from a waste material.

The treatment of EAFD dust has therefore gained great interest among researchers over the past few decades. Two major technological processes are suggested for extraction of zinc from EAFD; Pyrometallurgical and hydrometallurgical. Pyrometallurgical methods extract zinc from the dust by fuming and condensing the metals in pure form [4]. However, the process has a major drawback of high-energy requirements and the need for dust collection and gas cleaning systems [5,6]. On the other hand, hydrometallurgical processes are considered to be more sustainable and of lower operating cost. Several research efforts were performed to develop a hydrometallurgical method to increase the rate of zinc recovery. The primary work was focused on:

(1) Utilization of the leaching reagent such as sulfuric acid [4,7,8], hydrochloric acid, nitric acid, and sodium hydroxide [6,9];
(2) Adopting biological treatment to the sludge's of the dust;
(3) Employing different leaching techniques [10];

* Corresponding author. Tel.: +962799299285/+962776761414.
E-mail addresses: msalharahsheh@just.edu.jo, harahshehm@yahoo.com (M. Al-harahsheh).

http://dx.doi.org/10.1016/j.jhazmat.2014.03.019
0304-3894/

(4) Performing kinetic study and modeling the investigated data for predicting the optimum leaching parameters [6].

Jha et al. [5] reviewed the most recent hydrometallurgical processes employed for extraction of valuable metals from EAFD. They concluded that the most effective lixiviants for zinc extraction from EAFD were sulphuric acid and ammoniacal solutions.

Despite the substantial basic and applied research carried out on hydrometallurgical extraction of zinc, approximately 55% of EAFD is still processed by high temperature metal recovery processes [1]. Although these methods are of high cost, they are favored because of the limited number of proven process alternatives for treating of hazardous waste of this type.

Recently, there has been an increased interest in treating EAFD with plastic materials to recover valuable metals and minimize the environmental impact of both EAFD and plastic wastes. Under certain conditions halogen (such as chlorine) contained in many common polymeric materials is released from plastic in the form of HCl which in turn can react with metal oxides to form chlorides with low volatilization temperature and/or chlorides soluble in water [11,12].

PVC is a heavily produced plastic [13,14]; it has found many industrial uses such as in construction and utilities [15,16]. The annual production of PVC was estimated to be more than 35 million tonnes in 1997, increasing globally by 3.8% p.a. to 500 million tonnes in 2012 [17,18]. The abundance of this material suggest at end of life undesirable quantities of PVC wastes are accumulated worldwide, making sustainable disposal a real challenge.

The disposal of end of life PVC by thermal methods such as energy reclamation (incineration) and thermal degradation have been hampered by the release of hazardous chlorinated compounds and dioxins at elevated temperatures which are known to lead to process complications such as catalyst poisoning and increased capital costs (due to corrosion of plant equipment) [19–22]. Prior to treatment the separation of a single plastic (like PVC) from a waste plastics stream is complicated and expensive at an industrial scale therefore, additional methods of dechlorination and capture are required prior to energy reclamation. Common practices of dechlorination include processes such as biaxial extrusion or rotary kiln treatment in the presence of acid absorbents such as alkaline salts, water at elevated temperatures or metal oxides prior to the incineration of the material [20,23]. The use of a waste material such as EAFD as an acid absorbent is an attractive option from both a sustainability and economic point of view.

Thermal degradation of PVC occurs via a two-step mechanism. First the backbone of the polymer is stripped of its constituents (chlorine) at ~300 °C leaving an unsaturated hydrocarbon chain and liberating gaseous HCl. The second step (which occurs at higher temperatures) involves the decomposition of the backbone to lower molecular weight hydrocarbons [24–26].

The use of EAFD as a dechlorination catalyst for PVC based plastic material has several advantages including the capture of HCl released during the incineration of PVC by metal oxides present in the dust as well as converting the metal oxides to chlorides that are volatile at low temperature or are soluble in water. Such technique provides a solution to the concurrent treatment of two waste materials, collectively to minimize their environmental impact and at the same time recover valuable metals from EAFD by a simple route.

The aim of this work is to study the possibility of using PVC for the extraction of valuable metals from EAF dust by a microwave pyrolysis extraction technique. Microwave irradiation is known to be very efficient for heating materials which exhibit dielectric losses greater than 0.1. Volumetric, selective and fast heating are considered some of the main advantages of microwave heating [27,28].

In this work, the measurement of dielectric properties of both EAFD and PVC at different temperatures will be considered. The microwave pyrolysis of EAFD–plastic mixtures followed by leaching of metal values from microwave treated mixtures was studied for the aim to evaluate the extraction efficiency of zinc from EAFD by a microwave pyrolysis-leaching technique. To the authors' best knowledge this study is the first of its kind.

2. Experimental work

2.1. Materials

Four different samples of electric arc furnace dust were collected from Jordan steel smelter/Jordan. These samples were selected to represent the general quality of EAF dust generated in the smelter. The samples were characterized in terms of their chemical and mineralogical compositions. Powdered PVC was purchased from Sigma-Aldrich. Powdered zinc chloride, anhydrous, 98+% was purchased from Alfa Aesar, while all other chemicals were of reagent grade chemicals.

2.2. Microwave experimental setup

The experimental setup used for microwave treatment of EAFD and PVC is shown in Fig. 1.

The equipment consists of a microwave generator operated at 2.45 GHz with adjustable power in the range of 0.1–2 kW (Sairem®, model GMP 20 KSM). Forward and reflected power measurements were made by power meters (Agilent®) couples to the waveguide via a cross-directional coupler (Sairem®) connected to a PC for data logging. Power transfer was achieved via a WR340 (TE_{10}) rectangular waveguide terminated with an adjustable short circuit. Sample was inserted in to the maximum of the electric field through a 50 mm choke structures (cylindrical single mode cavity applicator) which was designed and proven to prevent microwave leakage to below the required legislative levels. This was confirmed using a microwave leakage meter. Energy absorption into the sample was maximized by the addition of a three stub tuning device.

In order to withstand the high internal temperatures of the reactants during operation, fused quartz was use for the reactor vessels. The reactor was inserted through the choking system and connected upstream to a nitrogen supply and downstream to a series of 250 ml wash bottle filled with water. Ultimately the system terminated to an external extraction system.

2.3. Microwave extraction experiments

2.3.1. Pellets preparation

EAFD was mixed with PVC at varying ratios. The ratios used in the current study were 1:1 and 1:2 (EAFD:PVC). The mixtures were well homogenized in a tumbling mill with ceramic balls for 15 min. The mixture was then compressed to blocks of about 5 g mass. A hydraulic oil press machine compressed the mixture at 150 kg f/cm^2 for 60 s, producing cylindrical pellets. Such compressive force was found to be suitable for the experiments to be performed. The homogeneity and the quality of the pellets were examined by scanning electron microscopy (SEM).

2.3.2. Microwave treatment

Once prepared, sample pellets were located within a quartz reactor and place in the centre of the microwave applicator (Fig. 1). Once the inert and extraction/collection system was connected, microwave energy was applied to the sample for a set time and power level. The evolved gases were passed through two