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Effective treatment and recovery of laurionite-type lead from toxic industrial solid wastes

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Abstract

This study provides a method to recover lead from laurionite-type hazardous industrial solid waste, using mild experimental conditions in terms of temperature and pressure. The waste contained mainly lead, iron and bromide, as Pb(OH)Br and Fe_2O_3. Almost quantitative recovery of lead and bromide was accomplished, by using 0.1 M HNO_3 at room temperature. The resulting acidic leachate (after filtration) was titrated against 1 M NaOH, and the product was found to be mainly Pb(OH)Br. The solid residue from the leaching process was iron oxide (mainly as α-Fe_2O_3). The respective kinetic experiments indicated that the order of overall dissolution (leaching) reactions were more likely to be a combination between zero- and first-order reactions, although it is difficult to provide any direct physical meaning, due to the possibility of co-existing sequential and parallel reactions, which are directly connected with the extent of examined phases' crystallization. The obtained results suggest that industrial solid wastes of a similar type can be effectively treated by the application of ambient experimental conditions.

Keywords: Pb(OH)Br; Precipitation; Selective leaching; Recycling; Hazardous industrial waste

1. Introduction

The hydrometallurgical separation of lead appears to be an interesting research issue, as several relevant studies have been already published. The applied procedures may be altered, according to the existing form of lead in the waste. The recycling of lead from used batteries was already examined and the respective treatments can be found elsewhere [1,2]. Certain other studies, concerning the recovery of lead from different waste types by the application of leaching/hydrometallurgical procedures, as well as the applied different experimental conditions are presented in Table 1. Former studies have examined the possibility of recovering lead from ores [10], using a combination of reagents (sea water, sulfuric acid and thiourea); lead was also recovered during tin recovery operations [11], or from the residues of zinc recovery processes from minerals [12]. Additionally, lead was separated by the use of nitric, hydrochloric and sulfuric acid from brass melting slag waste [13]. The recovery/removal of lead via hydrometallurgical procedures from wastes, derived from the off-gas residues (dusts) of relevant pyrometallurgical units was also studied [14]. Furthermore, numerous leaching agents and process conditions have been examined for the treatment of lead-contaminated soils [15–17]. An overview concerning the efficiency of solvents used for this purpose has been recently published [18].

The appropriate physicochemical leaching conditions were defined and optimized during preliminary experiments [19]. The aim of this work was to examine the effective treatment of a hazardous industrial solid waste, containing mainly lead, iron and bromide, by the application of appropriate mild (ambient) experimental leaching conditions, i.e. by dilute HNO_3 solutions, applied at room temperature and pressure. Specific emphasis was assigned to the efficient hydrometallurgical removal and subsequent recovery (after the appropriate precipitation) of main lead and bromide content in the form of laurionite-type lead compound (Pb(OH)Br). The main objective of this investigation was

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Table 1
Selected literature citations, concerning the separation/recovery of lead from different types of wastes (or from minerals)

Waste type	Pb (%, w/w) initial concentration	Leaching conditions			Reference
		Leaching solution	Volume/solid ratio (*L*/*S*)	Temperature (*T* °C)	
Smithsonite (Zn–Pb ores)	2.8	NaOH (5 M)	6–50	25–100	[3]
Electric arc furnace dust (EAFD)	0.2–7.8	Nitrilo-triacetate (1 M)	10	20–100	[4]
Lead as by-product (from Zn metallurgy)	27–60	Dilute H_2SO_4	4	70–75	[5]
Sludge from exhausted Pb batteries	>60	Tetrafluoroboric acid, glycerol and NaOH, potassium–sodium tartrate, NaOH	5	Room temperature	[6]
Bronze turnings	8	HCl (1–7.5 M), NH_4OH (1–7.5 M) and mixtures of them with H_2O_2	15	25–100	[7]
Alloy wire scrap	8.5	HCl (5 M)	10.6	80	[8]
Fly ash from Municipal Waste Incinerators (MWI)	10.7	NaOH (0.75–3 M)	7–25	30	[9]
Residues from thermal treatment of tetra-ethyl lead containing sludge	30	HNO_3 (0.1 M)	36–50	20–25	This study

to propose an integrated method for the treatment, recovery and recycling of similar type of solid wastes.

2. Experimental

2.1. Materials

2.1.1. Waste characterization

The origin of examined industrial solid waste, as well as the relevant (chemical) toxicity issues, was previously presented [20]. This waste is the thermal residue after the incineration of sludges derived from tetraethyl lead fuel storage tanks. The respective analytical determinations proved that the industrial solid waste under examination contained (in %, w/w): Fe 33, Pb 29.6, Br 10.1, Si 2.8, Al 1.8, Na 1.3, Ca 0.5, S 0.5, Mg 0.4, Bi 0.3, Mn 0.2, K 0.2 and Ti 0.1, whereas the total carbon (TC) content was <1% (w/w). Other elements were determined in lower concentrations. The respective solid phases were defined as Pb(OH)Br (34–38%, w/w) and Fe_2O_3 (44–48%, w/w, mostly as α-Fe_2O_3). The other lead-containing solid phases, such as PbO, $PbO{\cdot}PbSO_4$, were totally estimated around 5% (w/w) and all the other non-lead and non-iron solid phases (mainly aluminosilicates) were estimated approximately 10–15% (w/w). The detailed characterization data can be found elsewhere [21].

2.1.2. The chemicals used

Solutions of HNO_3 and NaOH were prepared from analytical grade reagents, and NaOH pellets, respectively. Lanthanum nitrate solutions were prepared from analytical grade solid $La(NO_3)_3{\cdot}6H_2O$.

2.2. Equipment

Mixing for the small scale up experiments was performed by a typical cylindrical mixing vessel (small tank) having baffles and with the following typical (standard) dimensions: $T/D = 2$, $T/H = 1$, $C/H = 1/3$ and $C = D$, where T is the diameter of cylindrical vessel, D the diameter of impeller, H the height of liquid and C is the distance from the bottom to the impeller, using a Rushton type impeller. A JULABO SW 21 reciprocal horizontal thermo-stated shaking bath was used for the agitation of solid/liquid mixtures under constant temperature and for the small scale up experiments also. Filtration was performed using the MILLIPORE 142 mm Hazardous Waste Filtration system and 0.45 μm nitrocellulose filters. The Metrohm 716 DMS Titrino laboratory titrator was used for the precipitation experiments. The respective FT-IR spectra were obtained by the use of Perkin-Elmer Spectrum One model. A JEOL JEE 4X device performed the carbon coating for the SEM-EDS analysis; the Scanning Electron Microscope (SEM) was the JEOL JSM-840 model. The determinations of lead and iron were performed, using the Perkin-Elmer 2380 Atomic Absorption Spectrophotometer (AAS). Bromide measurements were performed using the Zeiss Jena PMQ3 spectrophotometer. Powder X-ray diffraction measurements were performed by the Siemens D500 Diffractometer.

2.3. Procedure and description of the applied methodology

The overview of applied procedure is briefly presented in the form of a block diagram as Fig. 1. The separate stages and the respective experimental conditions are discussed in the following sections.