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Zinc oxide production through reprocessing of the electric arc furnace flue dusts

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ABSTRACT

The smelting of slag in view producing copper and cobalt alloys achieved at the "Big Hill Smelter in Lubumbashi" (DRC) generates Electric Arc Furnace flue dusts as by-product. These EAF dusts are highly toxic wastes since they consist essentially of low-grade zinc oxide powders wherein one finds lead (9.11%) as the major impurity. That is why, they are looked at as hazardous wastes that must be either stored in specialized landfills or reused as secondary raw materials. This research aims at producing zinc oxide through reprocessing of the EAF flue dusts. The treatment suggested is based on three operations: leaching (290 mg/L H_2SO_4, 130 min) of the EAF flue dusts in view dissolution of zinc (99.42%), precipitation of zinc (90%) at pH 7.5 with sodium hydroxide and calcination (300 °C, 15 min) to produce zinc oxide (93.39%). The recovery of zinc oxide from these hazardous wastes is a cost-saving method for their sound management and disposal. It is though that this practice could contribute to the conservation of resources and the environment safeguard.

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Introduction

The Electric Foundry of Lubumbashi (EFL) is the first state-owned smelter that was built in 1910 in the Katanga province of the Democratic Republic of Congo (DRC) [1]. It was producing copper Blister (99.5%) through the smelting of ores and the converting of copper mattes (Cu_2S - FeS). The EFL belong to the "Gécamines" which is the biggest state-owned mining company in the DRC. During the operating period of the EFL (1911–1993), about 13,000,000 tons of slag were discarded as the process wastes, stored nearby the smelter and named the "Big Hill of Lubumbashi" [2,3]. The production of copper and cobalt alloys at the "Big Hill Smelter in Lubumbashi" through reprocessing of slag was launched in 2000. This new smelter was designed to process 4,000,000 tons of slag from the "Big Hill of Lubumbashi" (2.2% Co, 1.3% Cu and 6–8% Zn) with the aim to produce annually more than 4,000 tons of cobalt and 2500 tons of copper. Consequently, it was expected to generate 15,000 tons of EAF dusts as by-product [2,3]. At present, the "Big Hill Smelter in Lubumbashi" generates daily 40 tons of the EAF flue dusts, which consists of low-grade zinc oxide where one finds lead (8%), iron, cadmium, copper, cobalt, and germanium to very low concentrations [4]. Based on the partnership contract signed at the smelter creation, the EAF flue dusts together with the secondary slag (2% Zn) are sent back to the EFL. Unfortunately, they are neither locally reprocessed nor soundly managed in spite of their high content in zinc and their hazardous nature. The EAF flue dusts are reputed to be hazardous since they can release pollutants to the environment when stored in the open air or abandoned in a moist environment [5–7]. As the matter of fact, the EAF flue dusts can produce the airborne particles or release toxic metals through weathering, erosion and leaching by rainfall and winds. In the more industrialized countries, sophisticated techniques have been designed for the sound management of the EAF flue dusts such as recycling, the storage in specialized landfill, retention of the toxic metals through introduction of dusts in a siliceous phase or vitrification, the chemical stabilization of the EAF dusts prior to disposal or their incorporation in building materials, etc. [7–17].

This research aims at producing zinc oxide through reprocessing of the EAF flue dusts generated by the "Big Hill Smelter in Lubumbashi". It is though that EAF flue dusts can be used as secondary raw materials considering their contents in zinc and lead. To achieve this objective, the EAF flue dusts of our focus were sampled, assayed for copper, cobalt, zinc, cadmium, nickel, iron and germanium and leached using sulphuric acid as solvent in view dissolution of zinc and removal of lead through precipitation as sulphate. Subsequently, the dissolved zinc ions were recovered through precipitation as hydroxide. The obtained precipitate has undergone dehydration to produce calcine (ZnO).

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Materials and methods

Chemical analyses of the EAF flue dusts

Two grams of the EAF flue dusts from the "Big Hill Smelter in Lubumbashi" was heated at 105 °C in a Memmert steamroom until constant weight and dissolved using aqua regia in view assessing for the contents in moisture and metals (Zn, Pb, Cd, Fe, Co and Cu) using a Perkin Elmer 2280 spectrophotometer. The residue given by the leaching of the EAF flue dusts was washed, dried, and weighted for the analysis of silica. A Spectro Genesis ICP-SAA device has enabled analysing germanium. Furthermore, an aliquot of a dried sample of the EAF flue dusts was heated at 400 °C in a mittens oven (45 min) in view assessing for the loss in weight through ignition. The results given by the above-mentioned analyses are shown in Table 1.

Leaching of the EAF flue dusts

Two grams of the EAF flue dusts (Fig. 1) were stirred (300 rpm, 25 °C) with 60 mL of a leaching solvent consisting of sulphuric acid in view dissolution of zinc and lead precipitation via the reactions (1) and (2):

$$ZnO(s) + H_2SO_4 \rightarrow ZnSO_4 + H_2O \quad (1)$$

$$PbO(s) + H_2SO_4 \rightarrow PbSO_4(s) + H_2O \quad (2)$$

The concentration of sulphuric acid (100–500 g/L) in the aqueous phase and time (30–150 min) were varied during the dissolution of zinc contained in the studied EAF flue dusts. The measure of the concentration of zinc liberated in the aqueous phase versus time and the concentration of sulphuric acid has provided the data that were processed using the Matlab 7.1 software to obtain the results depicted in Figs. 2 and 3.

Leaching solution purification and recovery of zinc

Prior to the recovery of zinc ions through precipitation in the form of hydroxide, the leaching solution has undergone purification (pH 5.5) using sodium hydroxide (40 g/L) in view removal of the unwanted ions of the metals (Fig. 1) based on the reaction (3) below [18]:

$$M^{n+} + nOH^- \rightleftharpoons M(OH)n(s) \quad (3)$$

In the above reaction, M may represent iron, lead or copper and n the valence. Furthermore, the leach liquor pH (6.5–8.5) was gradually raised through an incremented addition of sodium hydroxide (40 g/L or 12.09 g) to neutralize the leaching solution and precipitate zinc ions as shown in the reaction (4) below:

$$Zn^{2+} + 2OH^- \rightleftharpoons Zn(OH)2(s) \quad (4)$$

Table 1
Chemical composition of the EAF flue dusts from the "Big Smelter in Lubumbashi".

Compound or element	Proportion (%)
Zn	70.74
Pb	9.11
Si	1.6
Fe	0.84
Cu	0.12
Co	0.006
Ge	0.001
Moisture	13.79
Insoluble matters	8.7
Weight loss at 400 °C	4.73

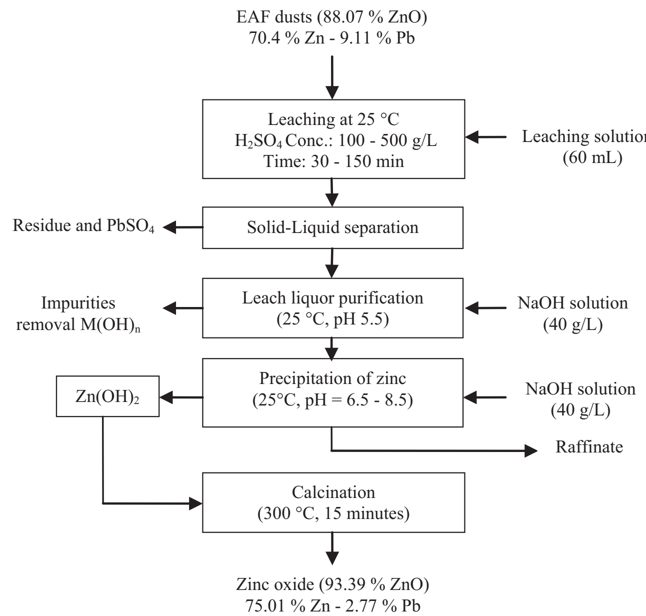

Fig. 1. Flow sheet for the reprocessing of the EAF flue dusts.

The precipitate formed was dehydrated through heating at 105 °C in a steamroom and calcination during 15 min in a mittens oven (300 °C) in view producing zinc oxide as shown in the reaction (5):

$$Zn(OH)_2(s) \xrightarrow{(300\ ^\circ C\ \text{and}\ 15\ \text{min})} ZnO(s) + H_2O(g) \quad (5)$$

The purity of zinc oxide produced through reprocessing of the EAF flue dusts generated by the "Big Smelter in Lubumbashi" was spectrophotometrically assessed and compared with that of the starting material.

Results and discussion

Chemical analysis of EAF dusts

The chemical composition of the EAF flue dusts from the "Big Smelter in Lubumbashi" is shown in Table 1.

The analyzed EAF flue dusts consist essentially of zinc (70.74%) which is present in the oxidized state with the moisture that averages 14%. When heated at 400 °C, the dried EAF flue dusts

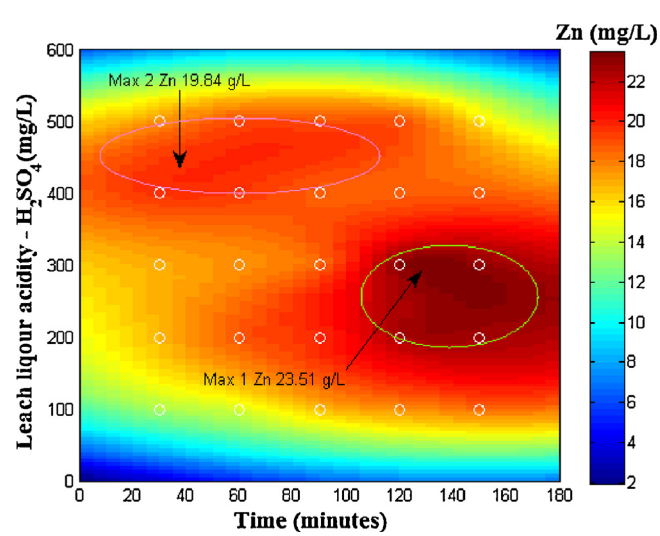

Fig. 2. Concentration of zinc versus the leach liquor acidity and time.

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