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(м. Кошице, Словацька Республіка)

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EAF DUST RECYCLING - PREPARATION OF FINAL PRODUCTS

T. Havlík, T. Vindt, Z. Takáčová, A. Miškufová,
Technical University of Košice, Faculty of Metallurgy,
Institute of Recycling Technologies, Košice, Slovakia

F. Kukurugya,
Flemish Institute for Technological Research (VITO),
Unit Sustainable Materials Management, Boeretang 200, Mol, Belgium

This paper describes basic steps in hydrometallurgical processing EAF (Electronic Arc Furnace) dust by using sulphuric acid as a leaching agent in order to achieve maximum zinc extraction. The basic steps of this processing are leaching, solution purification and final processing to either metallic zinc, zinc sulphate or other zinc compounds. It was found that maximum zinc extraction which can be obtained by acidic leaching is almost 90 wt. %. In addition to the leaching, also choosing suitable conditions of solution purification is very important. Impurities can negatively affect economics of the process and the quality of the final product. This work results in a design and description of the technology for processing EAF dust. The technology is based on two stage leaching with metallic zinc or zinc sulphate to be produced as a final product.

Introduction
Steel production in EAF represented in 2015 almost 26% of worldwide steel production (i.e. 420 million tons) [1]. This production is like any other industrial production source of hazardous waste. Such wastes include fine-grained dust coming from cleaning off-gases generated during production of steel in EAF. In general, EAF dust is material with high content of iron oxides and heavy metals such as Zn, Pb, Cd, Cr and others. Due to the heavy metals content these EAF dusts are classified as hazardous waste [2,3]. According to European legislation [3], this kind of waste is classified as hazardous waste (category 10 02 07 – Solid wastes from gas treatment containing dangerous substances). There are several reasons why it is important to pay attention to processing this kind of waste. Main reasons are:

- Material potential in terms of zinc content. Zinc content in EAF dust is in range of 8 – 35 % [4]. For comparison, zinc content in primary ores is within 5 – 15 % [5]. Current zinc price on London Metal Exchange (LME) is around 1900 US$/t [6].
- Possibility to obtain Fe-rich concentrate suitable as secondary raw material in pig iron production. Current price of primary Fe ore is around 55 US$/t [7].
- Reducing amount of hazardous waste or its transformation to non-hazardous waste, what results in reducing or even saving fees for its disposal.
The main reason for processing EAF dust generated in carbon steel production is high content of zinc. The source of zinc is galvanized steel scrap which represents major part of a charge in EAF.

In general, there are three basic ways of processing EAF dust: pyrometallurgical, hydrometallurgical and a combined method. Recently, pyrometallurgical processing is mostly used in practice. However, pyrometallurgical processing is economically effective only when more than 100 000 t of dust is processed annually, what is impossible to reach in Slovakia.

On the other side, hydrometallurgical processing is characterized by higher flexibility, lower energy consumption and by possibility to reach economical profit also when lower amount of dust is processed (around 10 000 t/annually).

In conditions of Slovakia, hydrometallurgical processing seems to be the best choice for using material potential of EAF dust. Therefore, this work is focused on hydrometallurgical processing of EAF dust with using sulphuric acid as leaching agent. The scheme of processing EAF dust by acidic leaching is shown in Fig.1.

As it results from Fig.1, hydrometallurgical processing EAF dust with using sulphuric acid consists of three basic steps: passing zinc into a solution, solution purification and preparation of a final product. This work describes basic steps of acidic leaching of EAF dust and based on the results it offers a designing processing procedure.

![Fig. 1. The scheme of complex processing EAF dust](image-url)
Experimental Material

Material used in this experimental part was supplied by the company Železiarne Podbrezová, a.s. The results of chemical analysis of studied sample is stated in Table I.

<table>
<thead>
<tr>
<th>Element</th>
<th>Zn</th>
<th>Fe</th>
<th>Ca</th>
<th>Pb</th>
<th>Cd</th>
<th>Mn</th>
<th>Cr</th>
<th>Si</th>
<th>LOI*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Content [wt.%]</td>
<td>17.1</td>
<td>27.2</td>
<td>4.4</td>
<td>1.3</td>
<td>0.1</td>
<td>1</td>
<td>0.8</td>
<td>3.2</td>
<td>7.08</td>
</tr>
</tbody>
</table>

Mineralogical composition of the sample was investigated by XRD phase analysis, results of which are shown in Fig. 2.

Fig. 2. The XRD pattern of EAF dust sample

Leaching of EAF dust in sulphuric acid solutions

Leaching experiments were carried out in the standard laboratory apparatus. The aim of the leaching was to achieve the highest extraction of Zn into a sulphuric acid solution. In addition to zinc extraction also iron extraction was observed. The main parameters affecting zinc extraction being studied in this work were: sulphuric acid concentration (0.05 – 1 M), temperature (20 – 95°C), L:S ratio (10 – 50).

Maximal zinc extraction was 87 wt.% and was achieved at following conditions: 1 M H2SO4, 80°C, L:S = 50. Kinetic kurves of leaching Zn and Fe at these conditions are illustrated in Fig. 3.

It results from Fig. 3, that passing zinc into the solution is a very fast process and already in first minutes maximal zinc extraction can be achieved. Passing iron into the solution is, comparing to zinc, significantly slower process what is given by different leaching mechanism. Selective leaching of zinc without iron passing into the solution would be an ideal case, which can be
reached by using less concentrated acid solutions, lower temperature and by reducing the leaching time. However, disadvantage of such conditions is lower zinc extraction. As practice require the highest possible zinc extractions it is impossible to avoid passing iron into the solution. From this reason, iron must be removed from the solution prior to the next processing step.

![Kinetic curves of Zn and Fe extraction at 20 – 95 °C in 1 M H₂SO₄ and L:S = 50](image)

**Fig. 3.** Kinetic curves of Zn and Fe extraction at 20 – 95 °C in 1 M H₂SO₄ and L:S = 50

**Precipitation of iron from the solution**

The limit concentration for iron in electrolyte in zinc production is 20 – 30 µg/ml [8]. Higher concentrations of iron in the solution can reduce hydrogen overpotential and reduce overall efficiency of zinc electrowinning.

Experiments of iron removal were carried out with leach-liquor coming from leaching experiments described in section B. Chemical composition of the leach-liquor is listed in Table II.

**TABLE II. CHEMICAL COMPOSITION OF THE SOLUTION**

<table>
<thead>
<tr>
<th>Element</th>
<th>Fe</th>
<th>Zn</th>
<th>Ca</th>
<th>Mn</th>
<th>Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>[µg/ml]</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>3391</td>
<td>4170</td>
<td>614</td>
<td>147</td>
<td>4</td>
</tr>
</tbody>
</table>
From economical and technological point of view, goethite and jarosite process can be considered as the most suitable for removing iron from a solution. In experimental part of this work both of these processes were used in order to remove iron from leach liquor coming from leaching EAF dust in sulphuric acid solutions. The experimental conditions of iron removal were following: temperature 80-95 °C; time 120 min; neutralization agents - 1M NaOH, CaCO₃, lime milk; 300 rpm; pH = 3-4 for goethite precipitation; pH = 2.5-3.5 for jarosite precipitation. Ammonium sulfate was used as additive for jarosite creation.

Fig. 4 shows the results of iron precipitation from the leach-liquor. These curves show trends in amount of iron being removed from the solution depending on the change in pH and temperature.

Based on the results achieved in this step it was found that the highest amount of iron can be removed at following conditions: temperature > 80°C and
pH ≥ 3.5 when lime milk is used as a neutralizing agent. At these conditions, it is possible to reduce iron concentration in the solution below 1 µg/ml.

**Cementation of impurities from solution**

Cementation by zinc powder serves to purification of a solution from impurities, which are more electropositive than zinc (e.g. Fe, Cd, Pb, Cu). This purification step is carried out mostly before final processing by Zn electrowinning to metallic zinc or in production other salable products.

In case of purification of a solution coming from leaching EAF dust it is very important to remove Cd and Pb from the solution. Otherwise, these impurities can precipitate on a cathode together with metallic zinc what could reduce final value of the metallic zinc. Experimental conditions of cementation Cd and Pb from the solution were as follows: temperature 20, 50 and 80 °C; time 60 min; pH = 4, 5, 6;300 rpm; amount of Zn powder 0.5 g/250 ml of solution.

Experimental results showed a possibility to remove 100 wt.% of Cd and almost 90 wt.% of Pb from solution coming from leaching EAF dust in sulphuric acid solutions. Concentration of iron, remained in the solution after iron precipitation step, was also reduced in the cementation step.

**Electrolytic deposition of zinc from the solution**

Zinc electrowinning process, together with reactions taking place during this process, is described schematically in Fig. 5.

![Schematic diagram of zinc electrowinning](image)

**Fig. 5. Zinc electrowinning process**

The highest current efficiencies (over 90 %) were achieved at current densities up to 833 A/m² and temperatures 20 and 40°C, Table III. The highest current efficiency, 99%, was achieved at temperature 40°C and current density 333 A/m². Table III shows that at conditions with the highest current
efficiencies, zinc was obtained in the form of compact plate with minimum amount of dendrites.

**TABLE III.** Form of Zinc Deposited on the Cathode During Electrowinning Process

<table>
<thead>
<tr>
<th>t [°C]</th>
<th>Current density [A/m²]</th>
<th>333</th>
<th>500</th>
<th>833</th>
<th>1000</th>
</tr>
</thead>
<tbody>
<tr>
<td>24°C</td>
<td>95.98 %</td>
<td>97.96 %</td>
<td>83.62 %</td>
<td></td>
<td></td>
</tr>
<tr>
<td>40°C</td>
<td>98.60 %</td>
<td>97.96 %</td>
<td>91.80 %</td>
<td>84.02 %</td>
<td></td>
</tr>
<tr>
<td>60°C</td>
<td>83.67 %</td>
<td>77.23 %</td>
<td>86.16 %</td>
<td>70.31 %</td>
<td></td>
</tr>
</tbody>
</table>

**Crystallization of zinc sulphate from the solution**

In general, production of zinc sulphate from a solution is a relatively simple procedure. Input solution was represented by the same solution as it was used for electrowinning experiments. The solution was heated up in order to evaporate water to the point of crystallization of zinc sulphate. Crystallized zinc sulphate was separated from the solution by filtration and was dried for 24 hours at 80°C. Final zinc sulphate is illustrated in Fig. 6.

![Crystallized zinc sulphate](image)

*Fig. 6. Crystallized zinc sulphate*

**Design of hydrometallurgical processing EAF dust**

Based on the results of experiments, process for complex processing of EAF dust was proposed, Fig. 7.
The process is based on two-stage leaching in sulphuric acid solutions at different concentrations with aim to obtain zinc either as metal or salt – ZnSO₄·7H₂O.

First step of proposed process is washing EAF dust with water in order to remove chlorine, which could be released as a gas during electrowinning process. After washing and subsequent filtration EAF dust is subjected to the first leaching step in 0.25 M H₂SO₄ at 60 °C for 10 minutes. It results from experimental results that this way it is possible to leach out ~ 50 wt.% of zinc present in the sample, while iron remained in the solid residue. Solid residue after first leaching stage is subjected to the second leaching stage in 1M H₂SO₄ at 60°C for 60 minutes. The aim of this second leaching stage is to leach out zinc which was not leached out in first stage. As leach liquor after second leaching stage contains beside zinc also high amount of iron it is necessary to carry out iron precipitation step. This step is performed at temperature 95°C with using lime milk as neutralizing agent. The pH value was kept in range of 3.5 – 4.5. After subsequent filtration of iron precipitate, solutions from first and second leaching stage are put together. This solution is treated by cementation by zinc powder in order to remove impurities like Pb, Cd, Cu, etc. Purified solution is used as an electrolyte for zinc electrowinning process or as starting solution for crystallization of zinc sulphate.
Conclusion

Hydrometallurgical processing of EAF dust, with respect to the amount of dust being generated in the Slovak Republic, seems to be the most suitable way of processing. As, according to the European legislation, EAF dust is classified as hazardous waste, its processing would lead not only to economical profit in the form of recovering valuable compounds (Zn and Fe concentrate) but also it would reduce fees paid for disposal of this hazardous waste. Laboratory research in the field of hydrometallurgical processing realized in the frame of LSPO (Laboratory of Processing Industrial Waste) demonstrated the potential of using sulphuric acid as leaching agent. By choosing suitable conditions it is possible achieve high extraction of zinc into the solution and by following purification (iron precipitation and cementation) it is possible to obtain solution suitable either for zinc electrowinning or zinc sulphate crystallization. The possibility to produce these two products within one technology is a big advantage of this processing.

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