

Recycling of EAF Dust by Semi-continuous High Kinetic Process

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Abstract

The horizontal high energy rotor ball mill (Simoloyer®) is used to break and activate dry solids. It is used for dry-milling and in the vertical mount for wet-milling in leaching processes. Technical electric arc furnace (EAF) dust with high contents of zinc oxide, zinc ferrite and magnetite is efficiently separated by ambient temperature leaching. The process shows promise for industrial application

Keywords : high energy milling, steel mill dust, leaching grinding recycling

1. Introduction

Electric arc furnace (EAF) dust is being generated as waste product from the process of steel manufacturing and super large volumes of EAF dust are produced in every industrialized country. The record for Germany in 1996 gave about 199,577 tons of EAF dust and for US 768,639 tons. EAF dust causes environmental problems and requires special care for handling and final disposal.

The present paper describes initial activation tests where a laboratory scale (2l) High-Energy-(ball)-Mill (Simoloyer) was used to activate the EAF dust leaching. This new method is expected to lead to an economic and environmentally friendly way of recycling and decontaminating of EAF dust and furthermore, to extract useful materials from the dust.

2. Experimental Design

The dust from the electric arc furnace was treated by a horizontal rotor high energy ball mill, the Simoloyer (Fig. 1). During this High Energy Milling (HEM) process on the one hand the particle size of EAF dust was reduced down to about 1 μm , and on the other hand the as-milled EAF dust was highly activated due to a strong mechanical impact from the fast flying milling balls (up to 14 m/s), e. g. deformation, breaking and cold welding of the powder, which caused a lot of structure defects of EAF dust and leads to an increased activity of the dust.

The leaching process was then performed with the same mill, but in the vertical mount (Fig. 2) and under wet condition. The vessel was filled with the balls, the substrate and the leaching solution. The rotor speed was the same as in the dry-milling. The solids and solutions were separated by centrifugation. Clearly, the HEM process could also be

carried out continuously for large scale production.



Fig. 1(left): horizontal rotary ball mill (Simoloyer® CM01-21) with air-lock for loading, operation and unloading under vacuum and/or inert-gas.

Fig. 2(right): Simoloyer® CM01-21 in vertical mount for the leaching process.

3. Milling Device

The horizontal high energy ball mills are known from academic and industrial applications in Mechanical Alloying (MA), High Energy Milling (HEM) and Reactive Milling (RM). They supply the highest relative velocity of grinding media, which leads to a high level of kinetic energy transfer, an intensive grinding effect and short processing times. The contamination of the processed powders by the milling tools is naturally lower since the process is based on the collision of grinding media rather than on shear and friction interaction of the same which usually leads to higher abrasion. The systems are presently

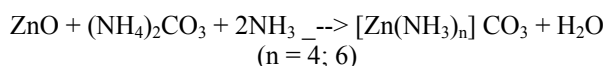
available from 0.5 to 400 liter grinding chamber capacity where larger volumes seem to be possible.

4. Experimental

A dry EAF dust sample of Georgsmarienhütte (grain size ca. 5 μm) contained 33.4% Zn, 27.5% Fe and 1.5% Pb according to Atomic Absorption Spectroscopy (AAS) (HNO_3 pressure decomposition). Further constituents are minor quantities of light and heavy atom oxides and silicates. 200 g of the dust were milled down to about 1 μm size in the horizontal 2 l Simoloyer for 20 min at 1300 rpm with 2 kg steel balls of 3 mm diameter while cooling with tap water of 14°C. The vertical leaching was performed under the same conditions after addition of 1.2 l of the leaching solution. The leaching solution was prepared from 590 ml of 25% NH_3 , 610 g $(\text{NH}_4)_2\text{CO}_3$ and 800 ml water to dissolve the salt. Alternatively, 590 ml 13.4% NH_3 instead of 25% NH_3 and un-milled EAF dust were used. The zinc, iron and lead contents were detected by AAS.

5. Results

The initial goal was to separate zinc and lead for further use, without destroying the iron oxide matrix that shall be recycled for steel production. The easiest way with technical potential is the leaching with ammonia and ammonium carbonate which dissolves zinc oxide (ZnO) and lead oxide (PbO) according to the formula (the index n depends on the ammonia concentration):



Franklinite (zinc ferrite, zinc spinel, zinc iron oxide, ZnFe_2O_4) is an important constituent of the dust in question which cannot be leached with ammonia because it does not dissolve neither at room temperature nor at 200° - 250° C in an autoclave (<10 % dissolved). Even HEM does not help in breaking the resistance of zinc ferrite against that leaching reagent. The separation of ZnO and ZnFe_2O_4 by ammoniacal leaching has been described. The experimental design had to be changed into separation of leachable zinc and lead (they can be separated by selective precipitation of their carbonates), iron from its oxides (magnetite, Fe_3O_4) and zinc ferrite (ZnFe_2O_4) in cold one-step processes. The leaching vertical ball-milling was performed for 10, 20, 30 and 60 min in order to assess the efficiency. It turned out that the wet-milling efficiency with the higher concentrated solution was so good that the 10 min runs both with pre-milled (1 μm) and original material (5 μm) were complete and longer milling was unnecessary. The longer milling under the corresponding conditions gave the same AAS result throughout. All of the leachable Zn and the lead from 200 g EAF dust were in solution that contained the

equivalent of 55,76 g ZnO and 6.64 g additional oxides (silicates) including the lead from 3.36 g PbO. Importantly, the difference between pre-milled and un-milled EAF dust showed up if the ammonia concentration was decreased (13.4% NH_3 instead of 25% NH_3). The benefit of the pre-milling (which will be more important with larger grain sizes) could be clearly demonstrated: complete leaching of ZnO from pre-milled dust required 30 min as compared to the untreated dust that was "complete" after >60 min.

The centrifuged washed and dried residue of the "complete" ammonia leachings (137.6 g) contained 16.0% Zn in the form of 81.2 g zinc ferrite and an additional quantity of Fe in the form of 24.0 g magnetite. The previous wet-milling was so efficient that all of the magnetite and further impurities (31.7 g) could be dissolved by 2n-HCl in 1 h at ambient temperature upon standing. The centrifuged and dried residue yielded 81.9 g (analytically expected 81.2 g) of highly pure zinc ferrite in the form of about 1 μm sized uniform particles that may be a valuable and useful product. X-ray Photoelectron Spectroscopy (XPS) - analysis gave the characteristic lines of ZnFe_2O_4 . Therefore, the aims of low temperature leaching were achieved at low energy costs.

Conversely, the difficulties with previous attritor techniques (stirring among the balls) and low kinetic milling are evident from the requirement of using pre-milled dust, 2 h leaching time at 2500 rpm and 50° C, as well as the necessity of a second leaching at 80° C, both with the higher concentrated leaching solution in order to dissolve all of the leachable zinc. Other techniques such as high temperature acidic leaching do not isolate the zinc ferrite but provide solutions that contain both the zinc and the iron which are laboriously separated.

6. Conclusion

We have shown that efficient wet-milling with the large scale vertical Simoloyer ball mill is feasible for industrial-scale leaching and that the leaching reagents have to be carefully selected. Clearly, for leaching zinc ferrite in matrices that are higher in Fe_3O_4 content, different leaching reagents will have to be developed that do not have the disadvantages of the known reductive, acidic (hot acid), and metal ion exchange (by FeSO_4 or FeCl_3) techniques which are available from the (patent) literature. Clearly, the leaching with the vertical Simoloyer will be usable for other hydrometallurgical leaching processes, in combination with horizontal dry-milling for selective leaching of ores at low temperatures. These low cost processes are environmentally friendly.