Poster | Raw materials

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## [P2] Raw Materials & Recycling

Session Chair: Mr. Johann Fischbacher (University for Continuing Education Krems, Austria), Dr. Yusuke Hirayama (AIST, Japan)

[P2-31] Investigations on the slag extraction method for pyrometallurgical REE recycling from sintered and bonded NdFeB magnets using borate slags

\*Daniel Vogt<sup>1</sup>, Chinedu Francis Anochie<sup>1</sup>, Ludwig Blenau<sup>1</sup>, Alexandros Charitos<sup>1</sup> (1. TU Bergakademie Freiberg (Germany))

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Neodymium-iron-boron (NdFeB) permanent magnets are used in a variety of applications due to their outstanding magnetic properties and low cost compared to e.g. samarium-cobalt magnets. Traction motors of electric cars and wind turbines are just two prominent examples of future-oriented technologies requiring rare earth permanent magnets (REPM). In order to meet the increasing demand and provide for more sustainable production, various recycling methods have been developed. Direct recycling methods such as re-melting or hydrogen processing of magnetic scrap (HPMS) are energy efficient and environmentally friendly. A drawback of these processes is that the condition of the feed material strongly defines the recycled magnets qualities. Severe altering (e.g. oxidation), adhesive glues or coatings cause difficulties. In addition, direct recycling methods are typically not suitable for polymer bonded magnets (PBM) due to detrimental incorporation of carbon.

In this study a pyrometallurgical indirect recycling method for sintered and polymer bonded NdFeB magnets was investigated. Rare earth elements (REE) are selectively oxidized by  $Fe_2O_3$  and extracted in a borate slag by addition of  $B_2O_3$ . The targeted composition was  $Nd_4B_2O_9$  since it is defined as a low melting area (approx. 1250-1300 °C) in the binary phase diagram. The resulting slag is rich in rare earth oxides (REO) containing ideally 90.6 wt%  $Nd_2O_3$ . Fe and B from the magnets are collected in a liquid metal phase that accumulates other metals (e.g. Co, Ni, Cu and others). Al was the only element co-extracted in the slag while  $Al_2O_3$  concentrations were <1 wt.%. Carbon from PBMs or graphite crucibles leads to formation of a  $Fe_3C$  enriched pig iron phase that is a potential by-product. following, the reaction equation is given, while Nd can be substituted by other REE (e.g. Pr, Dy).

$$2 \text{ Nd}_2\text{Fe}_{14}\text{B} + 2 \text{ Fe}_2\text{O}_3 + \text{B}_2\text{O}_3 = \text{Nd}_4\text{B}_2\text{O}_9 + 28 \text{ Fe} + 2 \text{ Fe}_2\text{B}$$

Experiments with 50 g sintered NdFeB magnets were performed in graphite and alumina crucibles to determine the impact of refractories on slag and metal phase at 1400 and 1500 °C. Graphite crucibles reacted with Fe to form a lower melting pig iron. Slag

separation from the crucibles was difficult despite no chemical reaction with graphite was identified. Alumina crucibles showed no interaction with the metal phase. The slag was easy to separate, even though the formation of aluminum borates was expected. However, the experimental findings demonstrate that the kinetic of alumina dissolution by borate formation is slow. The Nd recovery yields in the slag at 1400 °C were 63.6 and 81.5 % in graphite and alumina crucibles, respectively. The remaining REE were attached to the crucible after experiment. The high losses to the crucibles were explained by a little volume of the slag that was generated. Addition of synthetic  $Nd_2B_4O_9$  increased Nd recovery yields to 85.6 and 91.9 %, respectively. Upscaling to 500 g NdFeB further enhanced Nd recovery yield to 96.5 % in graphite crucible. All slags contained between 83 to 89 wt.% REO. Therefore, this pyrometallurgical method can be seen as a very efficient pre-concentration step for hydrometallurgical refining to pure REO.

In a next step, the slag extraction method was studied for polyamide (PA12) and polyphenylene sulfide (PPS) bonded magnets. First, the materials were pyrolyzed in argon for demagnetizing and pulverizing purposes. For PA12 bonded magnets Nd recovery yields of 77.8 % were achieved while slags with 82 to 87 % REO were produced. In case of PPS bonded magnets the sulfur caused high melting oxysulfides. Therefore, insufficient phase separation was observed. In order to optimize the process efficiency for PPS boned magnets, the reaction mechanism of sulfur is to be further investigated.

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