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MARIE CURIE ACTIONS

Recycling of NdFeB magnets for rare earth elements (REE) recovery

Introduction / Objective

NdFeB magnets currently dominate the magnet market and contain 25-35 wt. % REE (Nd, Dy, Pr, Gd and Tb), ca. 1 wt. % B while the rest being Fe and other minor exogens (Co, Al, Ga, Nb, etc.). The on-going monopoly of China on REE production and the continuously increasing demand for REE impose serious supply risks for Nd, Dy and Tb. Holistic hydrometallurgical or combined hydro- and pyrometallurgical flow sheets were developed and completed within this thesis for effective recycling of REE from these magnets.

Research Methodology

In sulfation, milled powder was mixed with concentrated H_2SO_4, dried at 110 °C, roasted at 650-900 °C and leached in water. In nitration, the powder was mixed with concentrated HNO_3, calcined at 150-600 °C and leached in water. In the complete leaching study, the powder was dissolved in dilute H_2SO_4. Fe was oxidized by MnO_2 and precipitated by $Ca(OH)_2$ or MnO prior to electrolysis for Mn-Co removal. In alternative steps, REE and Co were separated from Mn by oxalate and sulfide precipitation. In the last work, bulk magnets were treated by 5 hydrogenation treatments and studied for their oxidation behaviour and microstructural changes which is crucial for subsequent acid leaching.

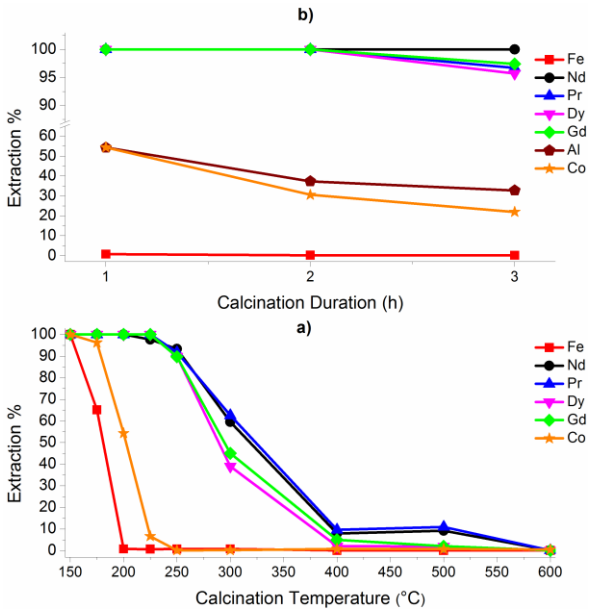
Results & Conclusions

1) Both sulfation and nitration studies resulted in >95 % REE and <1 % Fe extractions giving ≥98 % REE purity in the leachate. The Fe-dominated solid residue was marketable as it was dominated by hematite or a mixture of hematite and goethite with >90 % purity. Nitration study was more advantageous due to:

- Less particle size reduction (up to 1000 µm).
- Faster acid mixing kinetics without drying (1 h at 25 °C).
- Lower temperature need (200 °C for 1-2 h).
- Higher solubility of REE nitrates compared to REE sulfates.
- Theoretically easier recyclability chance of majority of the consumed acid (e.g. by condensation).

2) The complete leaching study was successful up to Fe removal stage but direct electrolysis was problematic due to incomplete Mn and Co removal and undesired REE losses. Alternative steps were successful but enlarged and complicated the flow sheet.

3) Among all hydrogenation treatments, hydrogen decrepitation was the most desirable one due to simpler processing and faster oxidation kinetics. However, complex REE-Fe oxide formation was more extensive requiring close control.



Effect of a) calcination temperature and b) duration on extraction % of metals (nitration study)

Major publication

Önal MAR, Aktan E, Borra CR, Blanpain B, Van Gerven T, Guo M. Recycling of NdFeB magnets using nitration, calcination and water leaching for REE recovery. Hydrometallurgy 2017;167:115–123.