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## Recovery of valuable metals from NdFeB magnets by mechanochemically assisted ferric sulfate leaching

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### PURPOSE OF THE ABSTRACT

The transition to a low-carbon economy and increasing digitalization relies on NdFeB permanent magnets, and has made them one of the largest and fastest growing applications in the market among all rare earth elements (REEs) [1]. Recycling routes for NdFeB magnet waste include direct re-use, re-sintering, pyro-, solvo- or hydrometallurgical approaches [2]. The latter is the most common used as it is generally applicable to all types of magnet compositions by dissolving in strong mineral acids, e.g. H<sub>2</sub>SO<sub>4</sub>, similar to REE processing from primary ores.

To enable better selectivity thermal pretreatment [3], thermal activation [4], [5] or high pressures [6] can be used, nevertheless these processes create new problems due to high capital expenses and/or produce environmentally damaging effluents (e.g. SO<sub>2</sub> or NO<sub>x</sub>). Alternatively, mechanochemistry can be used which aims at influencing the solid phase leachability by physicochemical transformations. When mixing solid reagents by (intense) grinding, mechanochemical reactions can be initiated as an effects of the structural disordering and increase in the specific surface area. The (pre)treatment, generally, leads to higher recovery and selectivity with faster kinetics combined with milder processing conditions, resulting in a recovery process with low environmental impact.

In this study, we present an alternative approach for recovery of valuable metals from sintered NdFeB magnets by leaching with a ferric salt (Fig. 1). Ferric sulfate was selected over the use of ferric chloride or nitrate because it is less hygroscopic than the chloride and does not form NO<sub>x</sub> gases unlike the nitrate. Ground magnet powder (<400 μm) was mixed with ferric sulfate and subjected to a high-impact grinding operation by ball milling to induce an exchange reaction (eqs. 1-2).



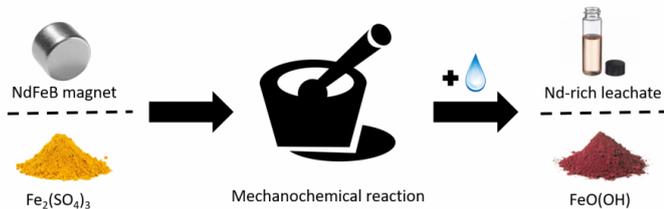
XRD results confirm the formation of Fe(SO<sub>4</sub>)-H<sub>2</sub>O, resulting from the conversion of the metals (e.g. REEs and Fe) to their water-soluble sulfates (Fig. 2). The grinding operation was optimized with respect to the ferric sulfate-to-magnet ratio (F:M) to improve the conversion rate. Although the reactions were initiated, full conversion by grinding could not be achieved. A similar effect was acquired with a simple automatic mortar grinder, therefore high-impact grinding is not required.

During the leaching stage, the mixture was subjected to water leaching at room temperature to determine the effect of the mechanochemical grinding reaction. As benchmark, also unmilled powder mixture was subjected to

water leaching. Leaching (with and without grinding) resulted in a high recovery, resp. 100% and 76%, of valuable metals (e.g. REEs and Co) with a F:M of 4:1. Therefore, it could be concluded that the reactions (eqs. 1-2) proceed during the leaching stage. The grinding operation, however, had a positive effect on the selectivity towards Fe and resulted in a shorter leaching time. These effects can be appointed to the exchange reactions during the grinding stage and the secondary effect from the structural disordering of the solids, resulting in an increase in reaction ability. Finally, solid-liquid separation was performed to separate the precipitate from the leachate. Oxalic acid was used to selectively precipitate the REEs from the leachate. Calcination of the oxalate resulted in a mixed RE oxide of >98 % purity.

This methodology has advantages over other recycling methods. This process excludes the use of mineral acids, high temperatures or high pressures, therefore avoiding the formation of hydrogen gas or formation of environmental pollutants (e.g. SO<sub>2</sub> or NO<sub>x</sub>). In addition, all process steps could be executed at room temperature. We expect that this study can provide a green and novel perspective for metal recovery from NdFeB magnets.

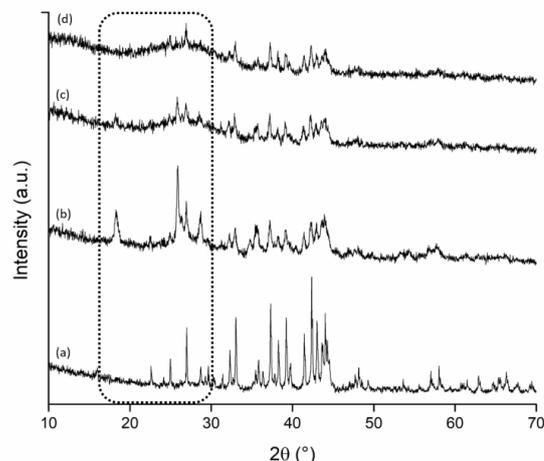
## FIGURES



**FIGURE 1**

Flow scheme of the recycling process

Schematic route of mechanochemical recovery process of NdFeB magnet waste by the use of ferric sulfate.



**FIGURE 2**

Effect of mechanochemical reaction

XRD pattern of (a) untreated magnet powder  $<400\ \mu\text{m}$  (b) ground powder with a ferric sulfate-to-magnet ratio of 2:1 (c) ground powder with a ferric sulfate-to-magnet ratio of 4:1 (d) ground powder with a ferric sulfate-to-magnet ratio of 6:1.

## KEYWORDS

NdFeB magnets | Recycling | process intensification | Water leaching

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