# Upcycling of NdFeB permanent magnets for green applications

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The climate crisis and resource shortages in Europe have underlined the need to identify alternate paths for sustainable growth, with the supply of raw materials from secondary sources playing a crucial role. Furthermore, the limited availability of these raw materials contributes to a higher import dependency and thus, lack of self-sufficiency. To overcome this situation being aligned with the green transition, NEO-Cycle project is boosting the economy through a green technology creating upcycling products for a sustainable industry reducing pollution and closing material loops. NEO-Cycle is devoted to developing not only recycling processes for end-of-life products (EoL) like Neodymium–iron–boron (NdFeB) permanent magnets but also for achieving high quality end products for pharmaceutical, ammonia, fertilizer and polymer industries. NdFeB magnets are considered as the most suitable magnets for high-tech applications, which are essential for the Green Transition, such as electric automobile, power generators, wind turbines and hard disks drivers (HDDs). The production of such magnets reached ca. 130,000 t worldwide in 2019, representing a 6.5 billion USD market volume, approximately 93% were sourced and produced in China. Although their widespread use in various applications has resulted in a large accumulation in recycling companies, primarily the HDDs being part of Waste Electrical and Electronic Equipment, the recovery rate of the NdFeB permanent magnets contained in these HDDs is surprising low. Thus, indicating a significant and contentious waste in local and regional areas and a huge problem of waste management. Furthermore, the situation is even more concerning as the collection of these magnets is not selective and therefore more and more magnet mixtures with a considerable increase in Fe content to the detriment of the total Nd. Solutions that offer an upcycling solution which does not need to reach such high purity products is the key here. As is well known, by applying state of the art technologies such as HCl digestion, what results is a solution where iron is in the form of Fe(III). This makes the individual Fe/Nd separation non-trivial and requires several steps (A. Klemettinen et al. 2023). Therefore, a technology which enables the extraction of iron in Fe(II) is the solution for a cost-efficient individual recovery of such metals.

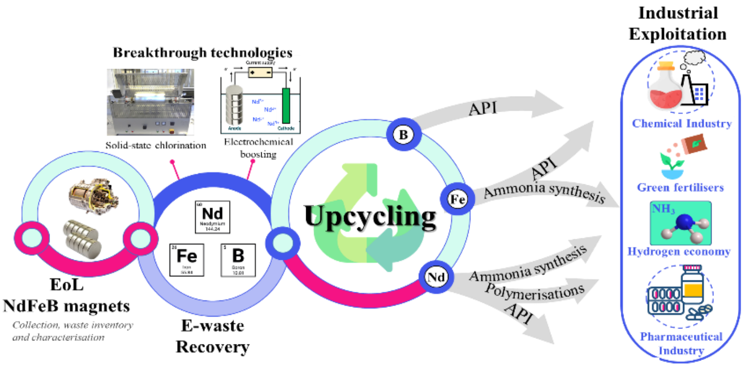

*Figure 1. Overall concept of NEO-Cycle project.*

TU Bergakademie Freiberg together with Fraunhofer IKTS are developing a breakthrough technology for rare earth elements recovery, the called solid-state chlorination (SSC), which offer the solution to achieve an economic and efficient recycling process for REE recovery (T. Lorenz and M. Bertau 2019). SSC consists of mixing the magnet powder after demagnetization with $NH_4Cl_{(s)}$ and the mixture is fed into a rotary kiln which will be head up until ca. 338°C to favor the $NH_4Cl$ decomposition producing $NH_{3(g)}$ and $HCl_{(g)}$, being the second compound consumed by the conversion of Fe and Nd to their chlorides, so that the dissolution of chloride compounds is easily achieved. The surplus of $NH_{3(g)}$ has a high purity (>99.9998%) with <5 ppm $Cl^-$ complying with European fertilizer legislation and is therefore readily marketable. The current work thus is focused on the efficient recovery and individual separation of Nd and Fe. Applying SSC the iron is extracted in the form of Fe(II) and the individual separation can proceed using the established method of addition of oxalic acid obtaining neodymium oxalate. This intermediate product will be used for API production and the remained iron solution for nanoparticles production for new catalysts for ammonia synthesis.

The input material for the individual separation was the magnet powder after the SSC, therefore, the neodymium as well as the iron are in chloride form. Preliminary tests were carried out using synthetic sample by mixing $NdCl_3$ and $FeCl_2$ (Merck KGaA, Germany) to end-up with a metal concentration of 70 wt.% Fe and 30 wt.% Nd. Afterwards the mixture was leached with oxalic acid to precipitate Nd in oxalate form while Fe(II) remains into solution. As depicted in Table 1, different parameters were investigated such as oxalic acid concentration, leaching time and temperature to evaluate their influence on the individual Nd/Fe separation.

*Table 1. Parameters used in preliminary test.*

| | Oxalic acid concentration [mM] | Leaching time [min] | Temperature [°C] |
|---|---|---|---|
| Range | 0.4 – 80 | 30 – 120 | 20 – 40 |

The elemental composition was determined by atomic emission spectrometry with inductively coupled plasma (ICP-OES, Optima 4300 DV, Perkin Elmer, Waltham, USA) and the neodymium oxalates with X-Ray Diffraction analyses with a Bruker D2 Phaser with a Lynxeye® detector (Bruker Inc., Billerica, USA).

The results depicted in Figure 2 show that the individual separation of iron and neodymium is possible if iron chloride is present in the mixture as Fe(II). As expected, the limiting parameter is the oxalic acid concentration, since the stoichiometric amount is needed (10 mM) for obtaining neodymium oxalate, the separation technology is selective to neodymium reaching 95.5%. Regarding the leaching time and temperature, the results follow the same trend where the separation is achieved within the parameter ranges used (Table 1).

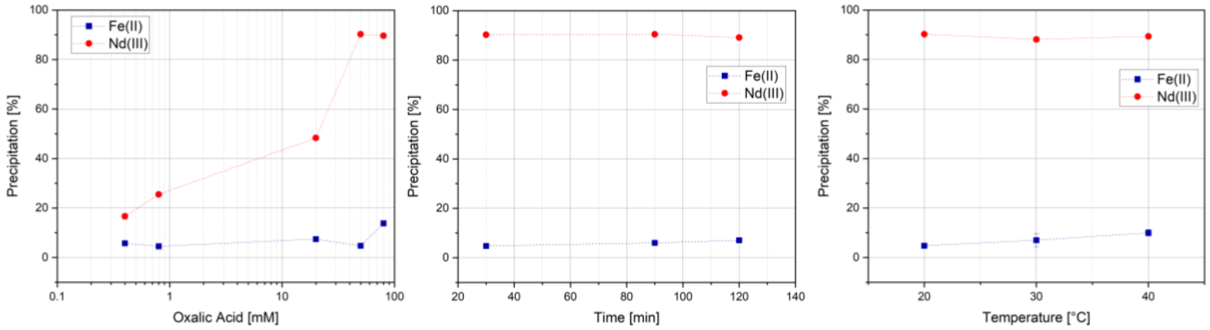

*Figure 2. a) Influence of oxalic acid concentration on precipitation rate (20 °C; 30 min); b) Influence of leaching time on precipitation rate (20 °C; 50 mM); c) Influence of temperature on precipitation rate (30 min; 50 mM).*

To sum up, the SSC technology is overcoming the drawback of selective separation from Fe/Nd due to the advantage of obtaining $FeCl_2$ after the chlorination. Once this step is achieved, the neodymium oxalate is obtained as raw product after adding oxalic acid because of its selectivity. The purity of the neodymium oxalate is ≥ 86% since Fe is precipitated as well. However, this was one of the advantages of the NEO-Cycle project as high purities are not required because the purpose is upcycling. Therefore, the recycling process can be achieved in an economical way avoiding so many purification steps thanks to the advantages offered by the SSC thus contributing to the recovery of critical metals from EoL products and favoring to safeguarding the raw material basis in Europe.

## Acknowledgements

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