D4.4 “Public summary of WP4 work”

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NEO-CYCLE

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Upcycling of NdFeB magnets in the EU for green applications

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1	27/01/2027	First draft	TUF
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DISCLAIMER

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ABBREVIATIONS

APIs	active product ingredients
DES	deep eutectic solvents
EoL	end-of-life
HDD	Hard Disk Drive
ICP-MS	inductively coupled plasma mass spectrometry
ICP-OES	inductively coupled plasma optical emission spectrometry
REE	rare earths elements
SENE	selective electrochemical Nd extraction
SSC	solid state Chlorination
WEEE	waste electrical and electronic equipment

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1. Abstract

The working package 4 is titled as “Magnets inventory and finetuning of Nd, Fe and B recovery”. The main objectives are:
(1) Waste inventory and magnet characterisation
(2) Process optimisation at lab scale to recover >95% of Nd, Fe and B from EoL permanent magnets via SSC and electrochemical extraction
(3) Selective separation of Nd, Fe, and B to reach targeted purities
(4) Impurities evaluation to set the limit of contaminants for the new applications.

2. Waste inventory and magnets characterisation

The focus was on the recycling-oriented characterisation and inventory of neodymium-based (NdFeB) magnets with the objective to collect, characterize, and catalogue end-of-life (EoL) magnets from waste electrical and electronic equipment (WEEE) streams, especially from laptops, servers and desktops.
At the project start the raw material base and an inventory of Nd containing WEEE in the EU was established to get an overview. IDE prepared an inventory of Nd containing WEEE in the EU following the United Nations Environment Programme guidelines. The market supply method was used including historical sales data and assumed product lifespans. The theoretically calculated urban mine potential of Nd in 2025 was 45.0 tons.
From this the further expectations for future needs were estimated as depicted in Figure 1.

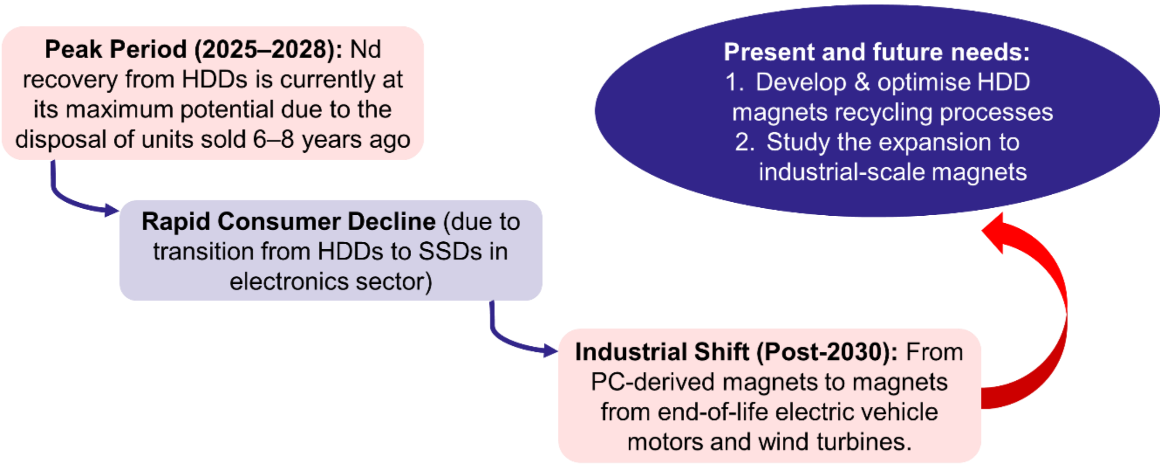

Figure 1 Nd-containing WEEE inventory in the EU from the past potential until the future need.

ECORESET and STENA sourced and manually separate 400 kg of spent magnets (20 t of HDDs) coming from WEEE. The hard disk drives were dismantled manually using electric screwdrivers and the magnet components separated from the casing (Figure 2).

Figure 2 The collected FeNdB-magnets from HDDs were disassembled manually.

Demagnetisation of the magnets was mandatory for further processing and characterisation steps. The thermal treatment was operated in a laboratory oven and a muffle furnace at temperatures between 300 and 500°C by ECORESET and TUF (Figure 3).

Figure 3 Magnets demagnetization by thermal treatment.

The magnets showed annealing colours and a peeling of nickel layers.
The different HDDs were characterised by TUF. For analysis, the samples were dissolved and analysed using ICP-OES (inductively coupled plasma optical emission spectrometry). This is a highly sensitive method for multi-element analysis, which allowed the quantification of trace metals (e.g. Cu, Ni, Co) in addition to Fe, Nd and B. ICP-MS (inductively coupled plasma mass spectrometry) was also used to analyse REE in trace amounts (Table 1).

Table 1 Element contents of magnets from different sources.

element	content [wt%]
Fe	64,5…69,6
Nd	22,0…25,8
Pr	3,4…5,7
B	0,9…1,1
Co	0,6…1,4
Ni	0,1…0,3

Traces of Gd, Al, As, Ga, Cu, Zr, Mn, Er, Cr, Pb, Gd, Tb and Dy were also detected and quantified.

3. Metal extraction from magnets via SSC and SENE

After demagnetization and characterisation, the magnets were crushed and milled to produce a powder (<0,71 mm) for the processing steps. Two different processes were developed on the one hand the Nd extraction via solid state chlorination (SSC) and on the other hand the electrochemical extraction of metals.
For the SSC a rotary kiln was used to extract Nd from spent magnets.

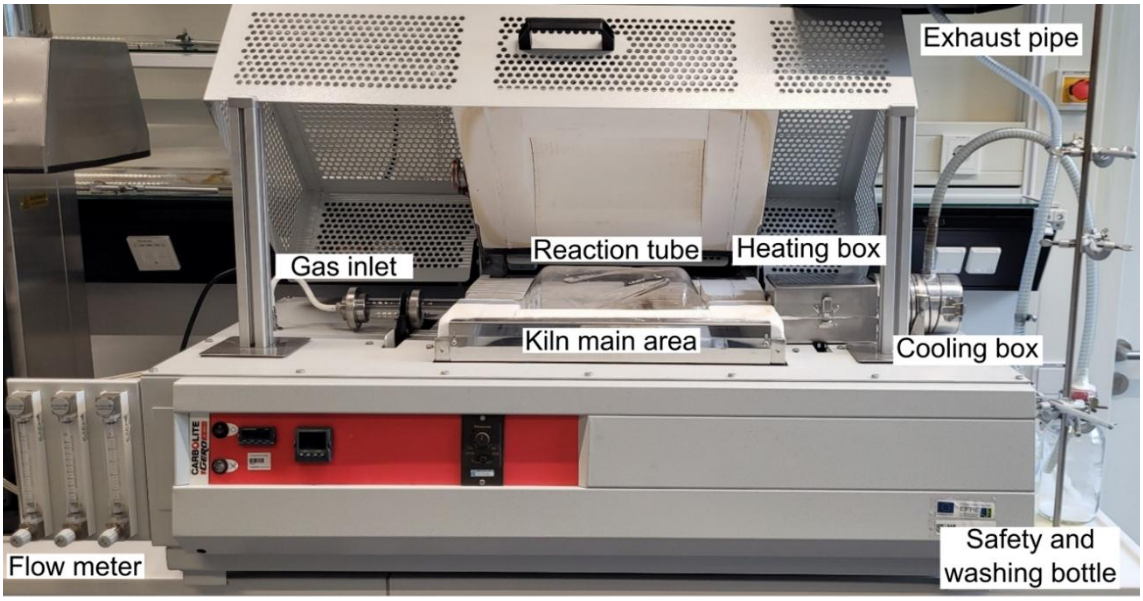

Figure 4 Rotary kiln for the SSC of FeNdB-magnets.

The magnet powder was intensively mixed with ammonium chloride (NH_4Cl) as chlorination agent and transferred into the glass tube reactor. At >290°C NH_4Cl started decomposing into ammonia (NH_3) and hydrochloric acid (HCl) as gases. Neodymium as the main rare earth element (Nd) and iron (Fe) react with HCl to produce water soluble neodymium(III)-chloride and iron(II)-chloride which is the starting mixture for metal separation. Ammonia is a valuable co-product and was trapped to receive an ammonia solution for the fertilizer industry. One positive side-effect is the recombination of unreacted HCl with access NH_3 to form NH_4Cl which was recirculated in the starting process.
Using a statistical test plan the process was optimised at lab scale.

Table 2 Overview of the selected factor levels for the statistical test plan

Factors	**Range**	**Factor levels**										
Sample:NH_4Cl ratio [g/g]	1:0,25 - 1:5	1:0,25	1:0,5	1:1	1:1,5	1:2	1:3	1:4	1:5			
Grain size sample [µm]	<50 - 710	<50	50 - 100	100 - 200	200 - 355	355 - 500	500 - 710					
Temperature [°C]	260 - 500	260	275	300	325	350	375	400	425	450	475	500
Hold time [h]	0,25 - 8	0,25	0,5	1	2	3	4	5	6	7	8	

With the optimal parameter a recovery rate >99% of Nd was confirmed. Overall, the ammonium chloride surplus appears to have the greatest influence on the conversion rate. Within the chosen framework, this is the most important parameter for increasing or decreasing the conversion rate. The holding time, on the other hand, has a rather minor influence. Although the conversion rate can also be increased by significantly extending the holding time, a high NH_4Cl excess, a higher temperature, or a lower magnetic particle size can also achieve a high conversion rate even with a very short holding time. Accordingly, it can be said that a higher temperature also increases the conversion rate. However, the main effect in this regard appears to be achieved at well below 500°C. The REE selectivity of the process is determined almost exclusively by the excess ammonium chloride. The lower the excess, the more selective the process is for REE. However, Fe and other elements are also partially converted in the ammonium chloride deficit.

The proposed flow chart for SSC is depicted in Figure 5.

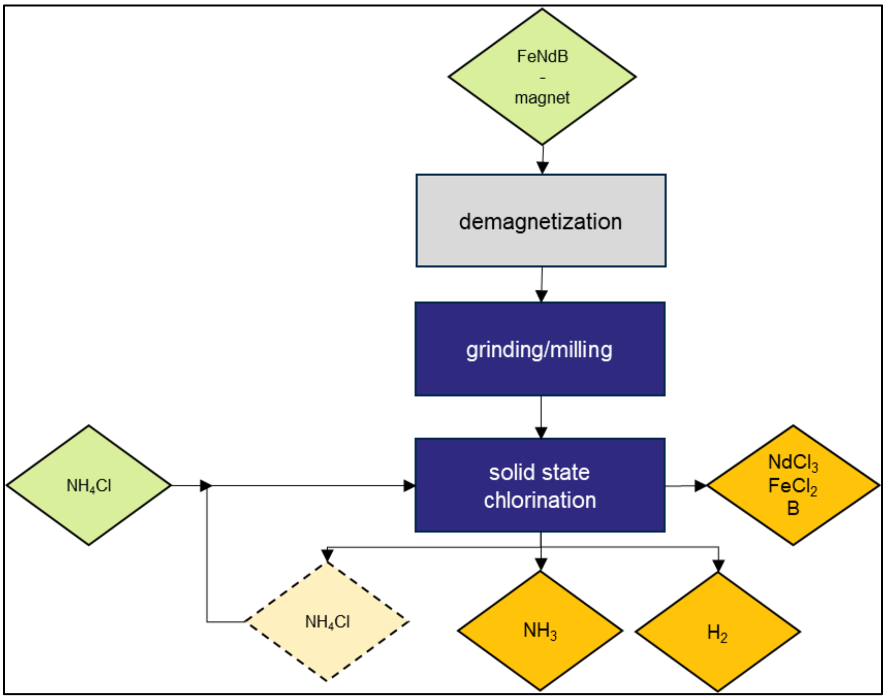

Figure 5 Proposed process flow chart for the processing of FeNdB-magnets by SSC with the first process steps. The educts are highlighted in green and the products in yellow. During the SSC ammonia can resublimate with unreacted HCl to form NH_4Cl (pale yellow) which is reused in the SSC.

The second process route was the selective electrochemical leaching (SENE). Both aqueous (Na_2SO_4, NaCl) and non-aqueous solvents (deep eutectic solvents DES ethaline, reline and oxaline) were used as electrolytes. DES have a wider potential window compared to aqueous systems allowing certain reactions that are not favorable in aqueous systems to occur in DES. But this work demonstrated that selective electrochemical extraction of Nd from end-of-life NdFeB magnets is not feasible in either aqueous or non-aqueous electrolytes, despite favorable theoretical predictions based on dealloying concepts. In contrast, a non-selective electrochemical leaching route combined with hydroxide precipitation was successfully developed for complete magnet dissolution and subsequent metal separation. The process is effective in both aqueous and deep eutectic solvent electrolytes, with significantly higher dissolution rates in aqueous media. Based on the experimental results, a practical flowsheet using 0.1 M Na_2SO_4 was proposed, enabling complete Nd extraction as $Nd(OH)_3$ and selective recovery of more than 80% of B in the leachate. The formation of distinct solid phases ($Nd(OH)_3$, FeOOH, Fe_3O_4) provides a sound basis for downstream separation of Nd from Fe. The proposed process offers several advantages, including chemical-free hydroxide generation, high dissolution rates without energy-intensive grinding, and operational robustness (Figure 6).

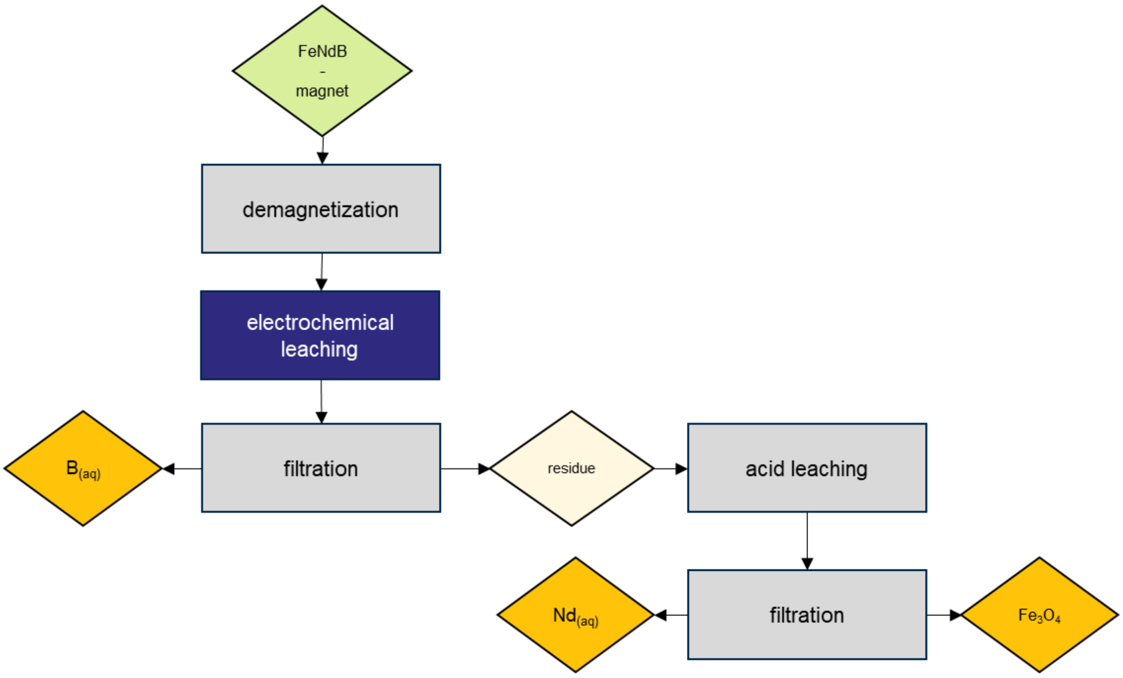

Figure 6 Proposed process flow chart for the electrochemical leaching of magnets (SENE process).

4. Selective metal separation and impurities influence

After SSC or electrochemical processing route the separation of metals followed done by IKTS. An integrated separation process targeting the production of intermediate products in the case of boron, and final products for neodymium and iron was developed. The separation technologies assessed and validated include complexation, precipitation, and solvent extraction, leading to the definition and optimization of a consolidated process flowsheet.

For the SSC route, the separation proved more effective than expected: not only Nd and Fe were successfully separated, but around 50% of the initial boron was also dissolved and recovered, although it was initially expected to remain in the solid residue. The REE-oxalates obtained (97.9% purity) were validated by UNIPV as suitable precursors for Nd-based catalysts. LUR confirmed that the optimized $Fe(OH)_3$ is suitable for further processing in WP6 (Flame Spray Pyrolysis and Fe-based nanoparticles) and demonstrated flexibility by successfully working with both Fe(II) and Fe(III) solutions. In the SENE route, electrochemical separation targeted boron, yielding a B-free Fe/Nd mixture for further processing. As the initially formed double salt was unsuitable for APIs and Fe-based nanoparticles for green ammonia synthesis, SSC-developed precipitation strategies were applied in sulphate medium. Additionally, UPC showed that boron-containing solutions can be concentrated via electrodialysis, enabling loop-closing strategies and readiness for larger volumes in Tasks 5.3 and 5.4 (from

March 2026). Although separation efficiencies differ from the chloride-based SSC system and further fine-tuning will be required in WP5, preliminary results are promising and confirm the technical feasibility, with strong potential to reach performance levels comparable to SSC during scaling-up.

The optimized flowsheet constitutes the technical basis for the intermediate and final piloting activities foreseen in WP5 for both processing routes (Figure 7).

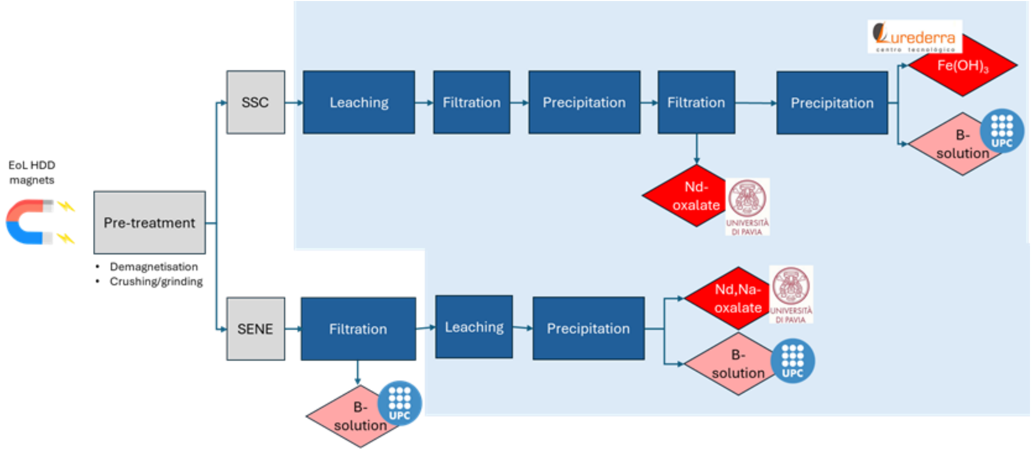

Figure 7 Actual proposed flow chart for the metal extraction and selective separation after solid state chlorination (SSC) and selective electrochemical leaching (SENE).