Rare Earth Magnet Recovery for Environmental and Resource Protection

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(on behalf of *REMANENCE* Consortium)

NMP.2012.4.1-2

Innovative recycling technologies of key metals in high tech applications

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Executive Summary

The possible future restrictions to the supply of critical materials to the EU have been the subject of debate for many years. Rare earths or rare earth elements (REEs) are a group of 17 chemically similar metallic elements that are plentiful and are used extensively in everyday life and have been classed as critical materials to the economy of the EU. They are part of mobile phones, hard disk drives, hybrid cars and other products that are in high demand. Production of REEs is now concentrated in only a few countries and Europe has no production facilities, and supply and cost volatility pose a risk to industry.

Europe has some potentially exploitable natural resources of rare earths, but these are not currently mined. The recycling of scarce materials like neodymium from end-of-life products could form an important part of future supplies but this route, like extraction from primary sources, is not currently exploited. Given the importance of these to modern living, the EU is funding research to make recycling a more feasible option.

“Rare earth magnet recovery for environmental and resource protection” (REMANENCE) is an ambitious project aiming to dramatically increase the amount of rare earth magnets recovered from existing waste streams. The focus is on recovering and recycling neodymium-iron-boron (NdFeB) magnets directly to a powder that can be used to produce new sintered or polymer bonded magnets, but processing of lower quality material is also undertaken to provide a source of Nd metal.

Advanced sensing and mechanical separation techniques have been developed to recover these rare earth magnets from electrical and electronic equipment. These materials are then processed using hydrogen decrepitation to transform the NdFeB magnets into a hydrogenated powder. This powder can then be extracted mechanically from the obsolete devices, and can then be processed further to produce either sintered or bonded rare earth magnets. The project has successfully integrated these processes into a virtual production line and has produced new magnets from waste hard disk drives. The properties of the new magnets are comparable to those made from fresh material, and the economic analysis has shown that the cost of the magnets would make them a competitive option in today’s market.

In addition, any “low grade” material that cannot be processed to produce new rare earth magnets directly, is subjected to a series of hydrometallurgical processing steps to produce a neodymium oxide that is fed to a molten salt electrochemical cell to produce the neodymium metal.

REMANENCE partners include leading experts in; sensing, disassembly, recycling technology and materials processing in a multi-disciplinary project able to deliver significant technical advances. C-Tech Innovation Ltd leads a consortium including; The University of Birmingham, Stena Technoworld AB, Acreo Swedish ICT AB, Leitat Technological Centre, Manufacturas Y Transformados AB, S.L., Chalmers Industriteknik, Magneti Ljubljana and Kolektor Magnet Technology GMBH.

The project website is http://www.project-remanence.eu/. The website contains downloadable content including information on the rare earth content of selected electronic waste streams, a video of the optical and magnetic sorting process and the automatic transfer to a cutting station, as well as information on current activities and past presentations.

Further information can be obtained from:

Dr. David Gardner (david.gardner@ctechinnovation.com)

Dr. Rob Bell (rob.bell@ctechinnovation.com)

Summary description of the project context and the main objectives

The project Remanence has been funded under the NMP.2012.4.1-2 call "Innovative recycling technologies of key metals in high-tech applications". The concept of the project has been to develop new and innovative processes for the recovery and recycling of rare earth (RE). neodymium iron boron magnets from a range of waste electronic and electrical equipment (WEEE).

The long term viability of many EU high-tech industries and clean technologies has become reliant on the secure supply of rare earth magnets. During the project lifetime there has been a significant shift in the dynamics and economics of the market, with proposed re-opening of RE mines and the subsequent shelving of the operation of the mines taking place while the project was progressing. However, the fundamental issues of security of supply, price volatility, and also the long term availability, of materials are still of great significance to the supply chain and final end-users of these products.

There are still no existing processes for the recovery of NdFeB magnets from waste streams, and these materials are either lost to landfill or are lost in the recovery and processing of other associated metals that occur in WEEE.

Whilst RE materials have become more readily available over the past few years, there is still the need to ensure that the EU has long-term security of supply. This inevitably means that there must be an increase in the sustainable consumption and production of high-tech products and the efficient use of the resources. It is therefore vital to develop the recycling and recovery of as much material as possible but with the understanding that this is achieved within economic constraints. From an economic, environmental and societal standpoint, it is imperative that recycling processes are developed and implemented in order to reduce the environmental impact of primary extraction. Efficient recovery and recycling of rare earth magnetic material into the manufacturing process will significantly recue the energy and environmental impact of these materials.

The project partners for Remanence represent the supply chain from collection and processing of WEEE, through to the manufacturers of the RE magnets themselves. This includes partners that can design and build novel sensing technologies that will identify and sort the materials, and research organisations that are able to process the collected waste and then supply this to the manufacturers.

The strategic objectives of the project are:

- To make a major contribution to the sustainable development of the market, by providing economically viable processes for the recovery of rare earth materials from waste
- Identify key waste streams and quantify the RE magnet content and develop technology to access the materials
- To develop a viable process to recover the rare earth content from mixed waste streams
- Provide materials with a greatly reduced energy demand when compared to virgin material production
- To develop methods for the collection and reprocessing of the identified WEEE streams

The strategic objectives are complemented by the technical objectives of the project:

- To develop an efficient mechanical separation that can concentrate the NdFeB fraction in waste that contains RE magnets
- To exploit the hydrogen decrepitation process for sintered NdFeB magnets, allowing the powder formed to be efficiently separated from the remaining material
- Develop an integrated recycling process capable of producing magnetic materials with low contamination, which are suitable for reuse in manufacturing
- Demonstrate, at an industrially relevant scale, the full recycling process from waste input streams, through separation, recovery, and reprocessing to final reuse in a final product
- To produce a new magnet with properties matching those of a magnet manufactured from a primary resource

The main process route for the recovery of material from waste magnets comprised the retention of the NdFeB material in a "high-grade" form, i.e. that the individual elements of the waste stream were not separated and the resultant material could be introduced directly into the manufacturing process for new sintered and bonded magnets. At the same time, it is recognised that some magnetic material cannot be recovered and the "high-grade" material retained, and an alternative processing route was to be investigated. This route comprised hydrometallurgical processing and molten salt recovery of neodymium itself.

The specific technical objectives are provided in the description of the following work packages of the project.

Characterisation of the input waste streams

The objectives of this workpackage were to:

- Gain an understanding of the current end of life fate for rare earth magnets
- Highlight key waste streams with high concentrations of rare earth materials
- Identify current best practice solutions that could be adapted to recover significant quantities of RE materials
- Identify current equipment used for the separation processes

Separation and concentration methods for RE magnet containing components

This workpackage was to develop sensor systems for the identification of permanent magnets material in a continuous process. The second objective was to develop a separation and disassembly procedure that would concentrate the content to a level that would allow for the economic processing of the RE magnets

Extraction of sintered magnet from WEEE by hydrogen decrepitation

As well as developing the hydrogen decrepitation process for the recovery of magnetic material from sintered magnets, the objective of this workpackage is to develop alternative processing of material that is not suitable for hydrogen decrepitation.

Material reprocessing and remanufacture of high quality magnets

Following the hydrogen decrepitation process, the resultant powder is processed so that it is ready for recasting. Therefore the objectives for this workpackage are to:

- Develop and optimise the processing routes to produce powders suitable for re-casting or to produce bonded resin bonded magnets
- The production of bother sintered and bonded magnets, and the assessment of their performance

Technical prototype demonstration and validation

During this phase of Remanence, a virtual production line from waste handling to the final production of magnets will be assessed. The work package will specifically:

- Address the realisation of the technical progress made in the individual stages to develop the concept of prototype systems capable of processing real scrap material and recovering the rare earth materials
- To understand the requirements of the virtual production line and to demonstrate that such a system is capable of reaching the key milestones for production capacity, cost, reduction in energy and environmental impact

Life cycle assessment and environmental impact of Remanence

Life cycle assessment procedures will be used to:

- Generate a set of key environmental performance indicators for a detailed life cycle assessment
- Provide cost assessments and consideration of logistical aspects of the developed processes
- Assess the environmental impact of the technologies developed in the project against the current end of life operation

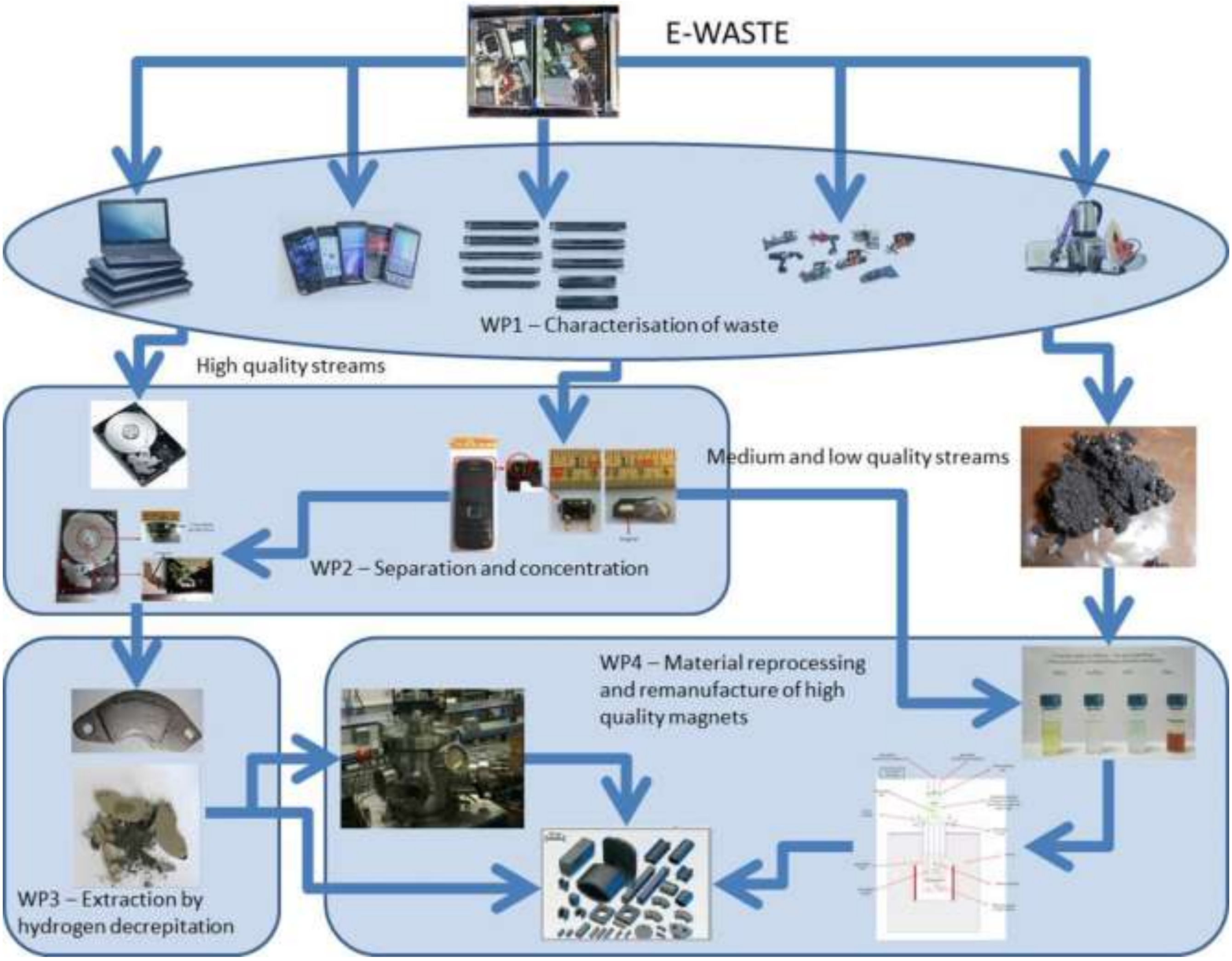
Dissemination, exploitation and training

Remanence addressed the complete supply chain of operations from the waste handling to the final production of the magnets. A key objective of the consortium partners was to be able to provide an understanding of the importance of each stage and how these impact on the earlier and later processing stages. This would minimise the chances that individual steps in the virtual production line would be optimised at the cost of other stages, but that the entire process was well integrated and any compromises would be understood by the partners. Also, interaction with future potential stake-holders was important as it is recognised that much of the supply of these materials is dominated by industry outside the EU. Therefore, key objectives were to:

- Ensure that the project’s outputs are disseminated for maximum impact
- Raise the skill levels of consortium members
- Raise the skill and knowledge of the wider research and industrial community
- Contribute to public awareness of the possible solutions to rare earth materials scarcity

REMANENCE Main S&T Results and Foreground

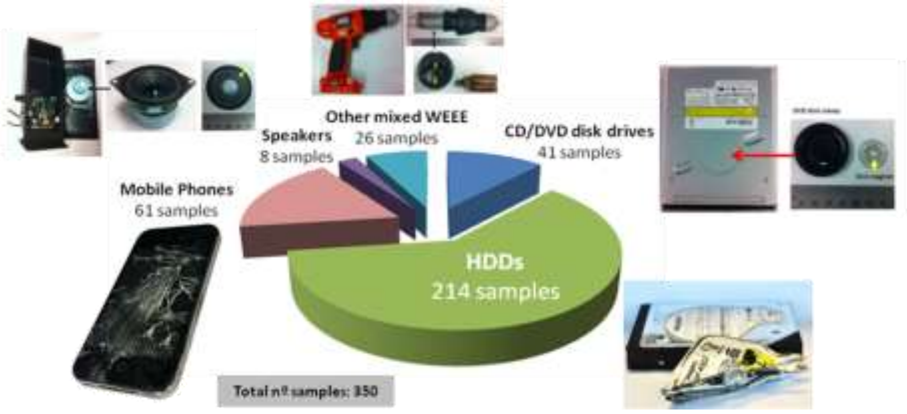
The overall process from the collection of the WEEE to the final production of sintered and bonded magnets, and the recovery of Nd metal for the lower grade material can be represented by the following figure.



The key results are summarised in the following sections.

Characterisation of the waste streams

The NdFeB magnet content of the WEEE stream was investigated in order to assess what items are most suitable for practical recovery of the RE materials. The WEEE products considered in this study were: hard disk drives (HDDs), CD/DVD disk drives, mobile phones, speakers, and other mixed WEEE containing small electric motors. A total of 350 magnet samples were recovered and analyzed using ICP-MS.



The suitability of these WEEE products for practical recovery of the RE magnet content was assessed with respect to the following features of the WEEE streams:

- High RE magnet concentration above 5 g. per kg of waste (Milestone 1),
- accessibility to RE magnets (i.e. ease of dismantling),
- and compatibility with current operations at the WEEE pre-processing plant

Items were collected in the WEEE waste streams and after manual dismantling the recovered magnet samples were characterized by Inductive Coupled Plasma Mass Spectrometry (ICPMS) analysis. The ICPMS analysis experimental method for the complete characterization of the magnet samples consists on the steps shown below. In brief, the method consists of a last step dismantling (if applicable) and an analytical microwave acid digestion of the magnet sample to finally perform a complete multi-elemental and Rare Earth (RE) elements characterization by ICPMS.

Figure: Schematic description of the ICPMS analysis experimental method for the complete characterization of the magnet samples.

Analytical microwave digestion is a procedure by which solid samples are heated in a solvent applying microwave energy. Its primary purpose is the rapid preparation of samples to be analyzed by ICP-MS. Several advantages are attributed to this digestion procedure: high efficiency, fast, low solvent amount needed, control of all extraction parameters, low contamination risk and the possibility of digesting several samples at the same time. Then, ICP-MS is an analytical technique capable of determining quite all the elements with several advantages: speed, automation and low detection limits. Therefore, analytical microwave digestion combined with ICPMS analysis is one of the best analytical combinations to successfully perform a complete and accurate characterization of REMANENCE magnet samples.

Considering all the samples studied, there is nearly a 90% that are NdFeB based magnets. Then, ferrites are the second most abundant followed by unknown kind of magnets and SmFeCo ones.

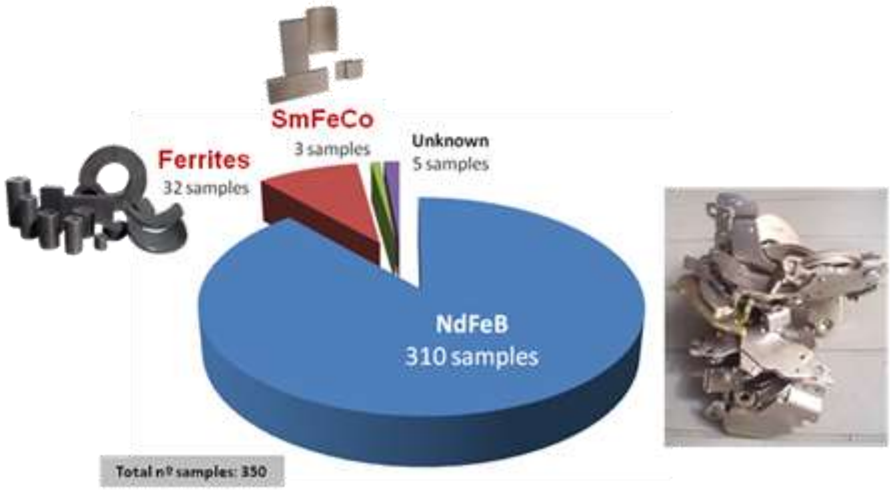


Figure: Categories of magnets analysed by ICPMS.

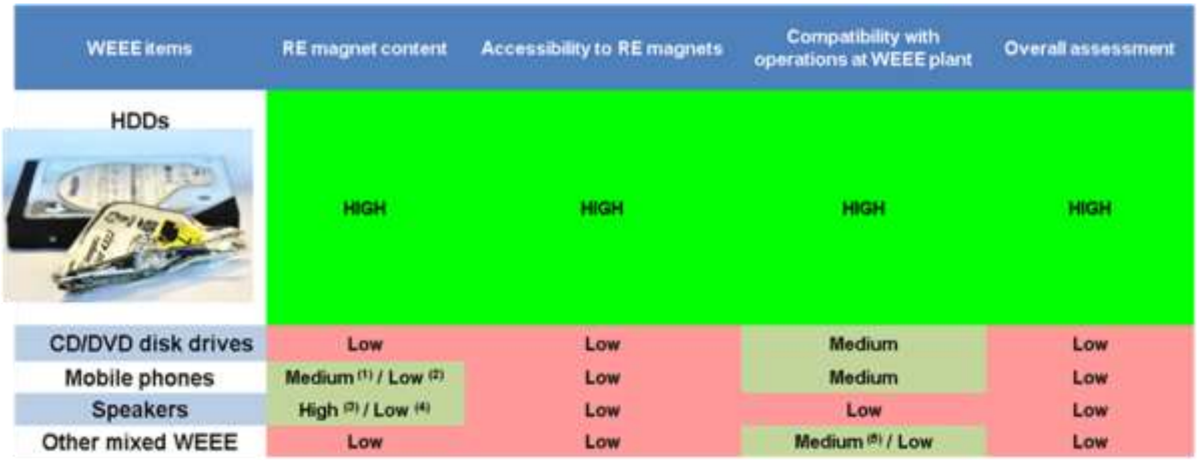
NdFeB magnets are the most interesting for RE recovery as they have Nd concentrations around 20-25% depending on the sample. Then, other elements are present in small amounts, such as B, Ni, Cu, Mn, Co, Ge, As, Cr, Ti, Ga, Se, Al, Zn, Sr, Ba, Bi and Ca. Additionally, other RE elements can be present in low concentrations (Pr, Dy, Tb, Ce, La, Gd and Er).

	MULTIELEMENTAL	RARE EARTH
Most abundant	Fe (40-80%)	Nd (20-25%)
Other elements (0.001 to 0.1%)	B, Ni, Cu, Mn, Co, Ge, As, Cr, Ti, Ga, Se, Al, Zn, Sr, Ba, Bi, Ca	Pr, Dy, Tb, Ce, La, Gd, Er
Not present (<LOQ)	Mg, Na, Si, P, S, K, V, Rb, Mo, Ag, Cd, In, Sn, Sb, Cs, Pt, Au, Hg, Tl, Pb, Li, Be	Sc, Y, Sm, Eu, Ho, Tm, Yb, Lu

NdFeB magnets elemental composition (data based on ICPMS analysis results).

SmFeCo magnets could be interesting, too, as they have around 25% of Sm. Nevertheless, it is more difficult to get waste streams where these kinds of magnets are present. Therefore, is not the best option to focus on SmFeCo magnets as good RE sources.

A range of tests were conducted on the different types of WEEE to enable an assessment of the most appropriate product that should be chosen to be the focus of Remanence. The table below summarises the results.



WEEE items	RE magnet content	Accessibility to RE magnets	Compatibility with operations at WEEE plant	Overall assessment
HDDs	HIGH	HIGH	HIGH	HIGH
CD/DVD disk drives	Low	Low	Medium	Low
Mobile phones	Medium (1) / Low (2)	Low	Medium	Low
Speakers	High (3) / Low (4)	Low	Low	Low
Other mixed WEEE	Low	Low	Medium (5) / Low	Low

(1) Speakers (loud) sub-component
(2) Other sub-components
(3) Headsets and earphones
(4) External and built-in computer speakers
(5) When a battery has to be manually removed (e.g. power tools)

Hard disk drives can be identified as a separate item (compared to the computer) as hard disk drives are already separated as part of the collection and separation supply chain. Future changes in the strategy for collecting different components may impact on the current assessment (for example it may be possible to separate out the different components from DVD disk drives and from digital recording equipment).

The analysis of the hard disk drives shows that there are many variations between different units. The variations exist between manufacturers, but also by the same manufacturer with the year of manufacture. This identifies a key task for the project, which is to demonstrate that the performance of a magnet manufactured from a range of starting materials was not negatively impacted (both in overall performance but also in the reproducibility of the properties of different magnets).

The total amount of hard disk drives from laptops and computers was then assessed for the EU. An estimate of the future annual flows of NdFeB magnets from HDDs in the WEEE stream was conducted based on sale forecast of computers in EU available in the literature[1]. A lifespan of 8 years for desktop PCs and 4 years for laptops was assumed based on data from the Netherlands for the year 2005[2]. The weight and RE content of NdFeB magnets found in the voice coil assembly and spindle motor of 3.5-inch and 2.5-inch HDDs were measured within the REMANENCE project.

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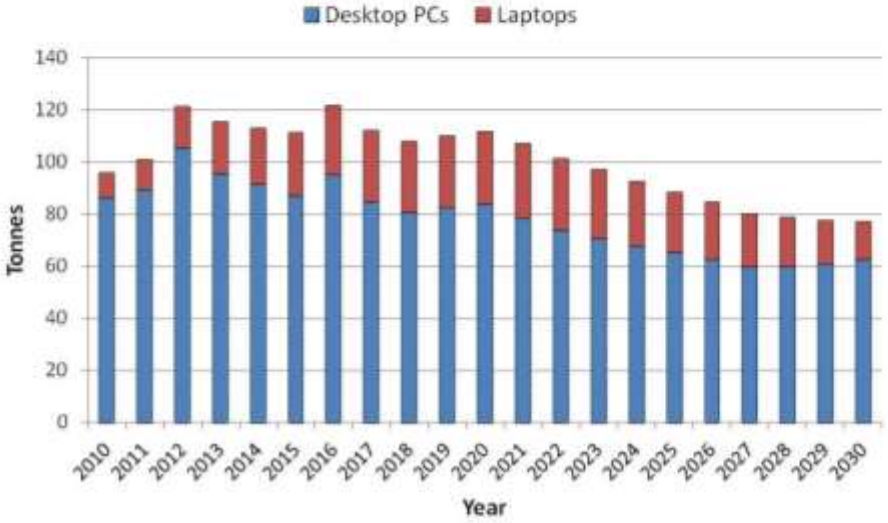
[1] J.H. Rademaker et al., Environmental Science & Technology 47 (2013) 10129.

[2] F. Wang et al., Waste Management 33 (2013) 2397.



This represents less than 1% of the total global supply of Nd for the production of magnets. However, it should be emphasised that this is for a very select group of materials that meet all the criteria for selection for this project, and should be considered as the market that is already in a position to be exploited with nor modifications to the existing logistics and infrastructure.

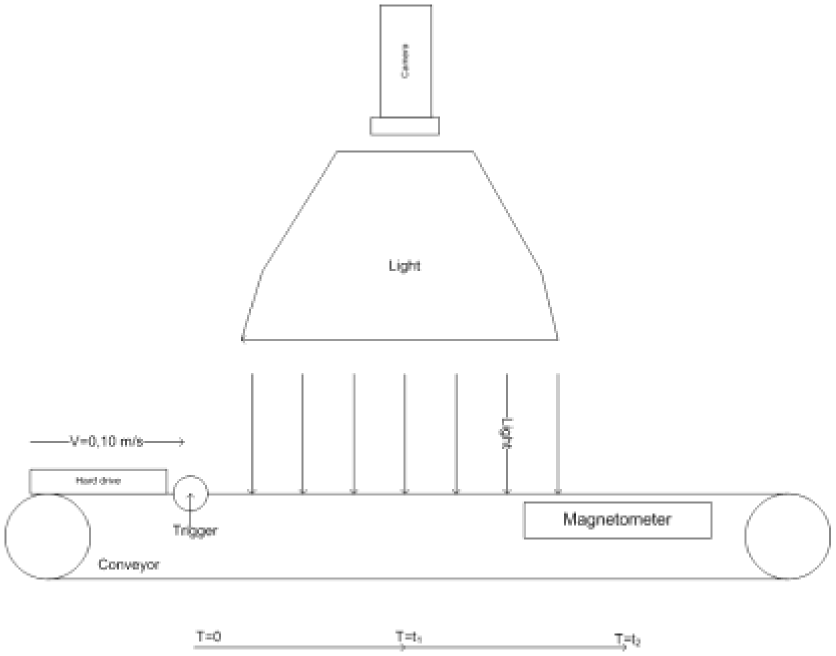
Development of separation and concentration methods

The work was split into a series of tasks that saw the:

- Identification of suitable sensor technology
- Development of a tracking system for localization of rare earth magnets in a mixed magnetic stream
- Development of sensor arrays
- Segregation and separation of the desired materials from a mixed stream
- A separation system to “concentrate” the desired magnetic material

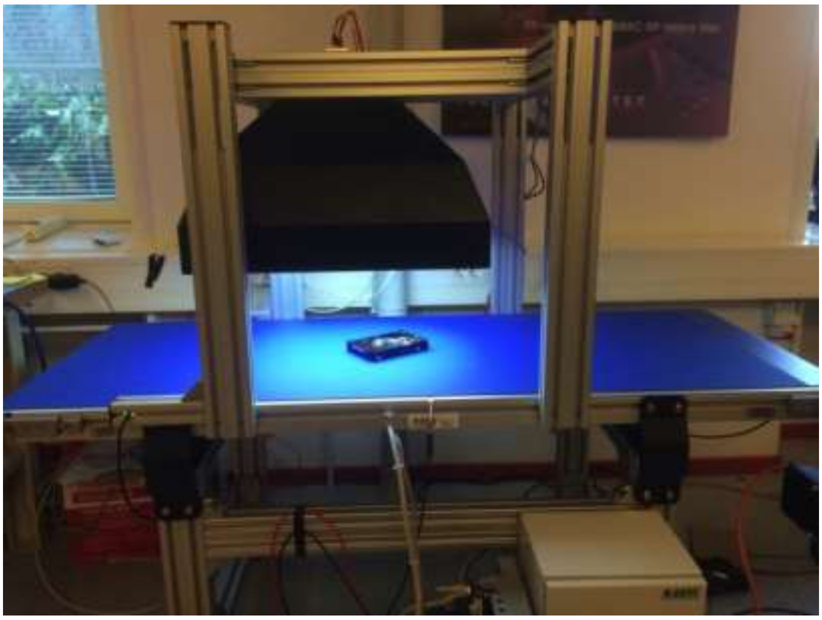
A mixed system of magnetic and optical sensors was identified as a method of locating the magnetic material and then identifying the exact location in reference to the entire component. Field patterns for three types of components were produced to assess the suitability of the different magnetic field sensors. The subsequent tasks then developed the overall process based on the combination of these two sensor methods.

Having identified the two sensor methods to be used, the concept for a fully automated system for identification of magnetic material was developed. The unit comprised a conveyor belt system that incorporated an optical setup with optical trigger, camera, lens, lightning, software for image acquisition and image analysis.



This work was done in parallel with the development of a sensor array mounted below the conveyor belt.

A prototype on-line conveyor based magnetic sensor system was developed, which synchronised the magnetic measurement with the optical image. The hardware and software developments resulted in the first design that could begin to distinguish between different objects, and produce combined magnetic field and optical imaging.



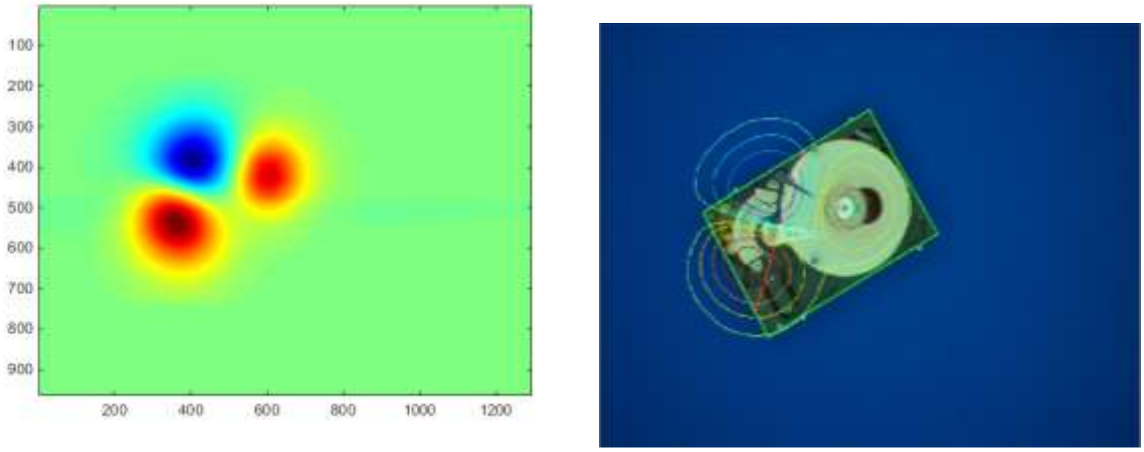


Figure (Left) Magnetic image of a HDD scanned with the 32-channel magnetometer array. (Right) A combined optical and magnetic image showing the location of the voice coil magnet and the spindle magnet.

The subsequent development of robust algorithms was then undertaken to determine the optimal placement of the cut in HDDs, in order to efficiently separate out the voice coil magnet.

As well as developing the algorithms to decide where a cut should be made to separate the coice coil magnet, other algorithms to handle multiple WEEE objects in the same image. At full speed the complete measurement and analysis process takes less than 6 seconds in total. The objects are classified into six different classes based on the magnetic content and the shape of the objects. Items can be rejected if necessary, or selected to be taken for further processing.

Work was then completed on the identification of the location of RE magnets in WEEE products and the assessment of the needs of the next phases of the processing. To ensure maximum economic returns it is required to remove as little casing as possible (as the aluminium provides a significant income to the waste handler), and to retain the integrity of the magnet, so that it can be processed easily via the hydrogen decrepitation route. The final decision was that an angled cut across the corner of the HDD would be satisfy both these needs, provided that the unit could be placed accurately at the cutting station.

Figure Left: Sintered NdFeB magnet(s) are found in the voice coil assembly and a bonded NdFeB magnet is located in the spindle motor. Middle: A transverse cut (aligned with the short side of the HDD) would put less requirement on the alignment of the HDDs. Right: A biased cutting angle minimizes the amount of aluminium casing that remains attached to the voice coil assembly

Different methods to cut HDDs have been tested and evaluated, including manual circular saw, guillotine, manual band saw, a semi-automatic industrial miter saw, alligator cutter, plasma cutting and water-jet cutting, smasher, multispeed drum, pneumatic press and hydraulic press. The following parameters were evaluated: cutting time, potential impurities generated during the process, quality of the magnets after cutting, and possibility to automatize the process.

A cost estimate for the different cutting techniques tested was compared to personal costs for manual disassembly at Stena. The estimate included investment (costs of machinery) and operational costs (energy consumption, maintenance, servicing).

Alligator shear cutting was found to be the most promising method for removing corners from HDDs. The short cutting times, very good cut qualities and moderate acquisition costs suggest that alligator cutting should be used for cutting HDDs and removal of magnets.

During the cutting tests it was found that clamping of the HDDs is a necessary action that has to be performed while shear cutting, since performing the cut without clamping may result in loss of control and precision of the cut.

The method for transferring the HDDs from the conveyor belt system to the cutting station was then progressed. A robot system was chosen to provide the accuracy needed and autonomous operation.

After extensive tests a final integrated system was developed. The integrated system includes the individual process steps for automated concentration of NdFeB-magnets from HDDs (detection, transport, orientation, cutting, removal). A schematic figure of the entire unit is given below.

The unit comprises the following components:

- Input and Inspection belt.
- Camera imaging acquisition.

- Magnetometer.
- Nonmagnetic ejector.
- Manipulator.
- Synchronizer Input belt/manipulator.
- Actuators emplacement.
- Picking tools (clamp).
- Cutting system (Alligator type).
- Ejection of cut parts.
- Evacuation Slides.
- Evacuation Belts.
- Safety Fence.
- Total Electrical/Electronical Integration.

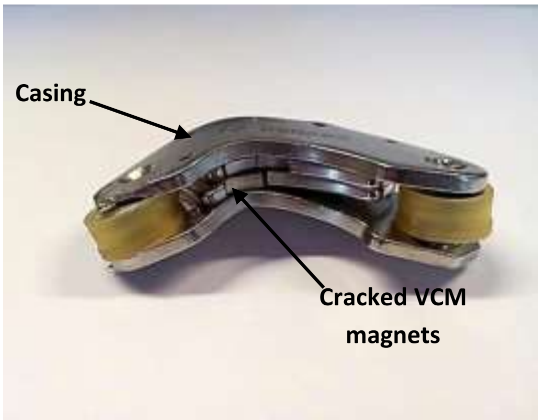
The system can easily identify, sort and cut the corner of 2.5'' or 3.5 ''HDD with a speed of at least 10 HDDs per minute, corresponding to 105.600 HDDs per month (assuming 22 working days/month and 8 hours/day). Hence, the system has the capability to exceed the initial project goal by an order of magnitude.

The unit has been installed and is operating at Stena's reprocessing facility at Halmstad, Sweden.

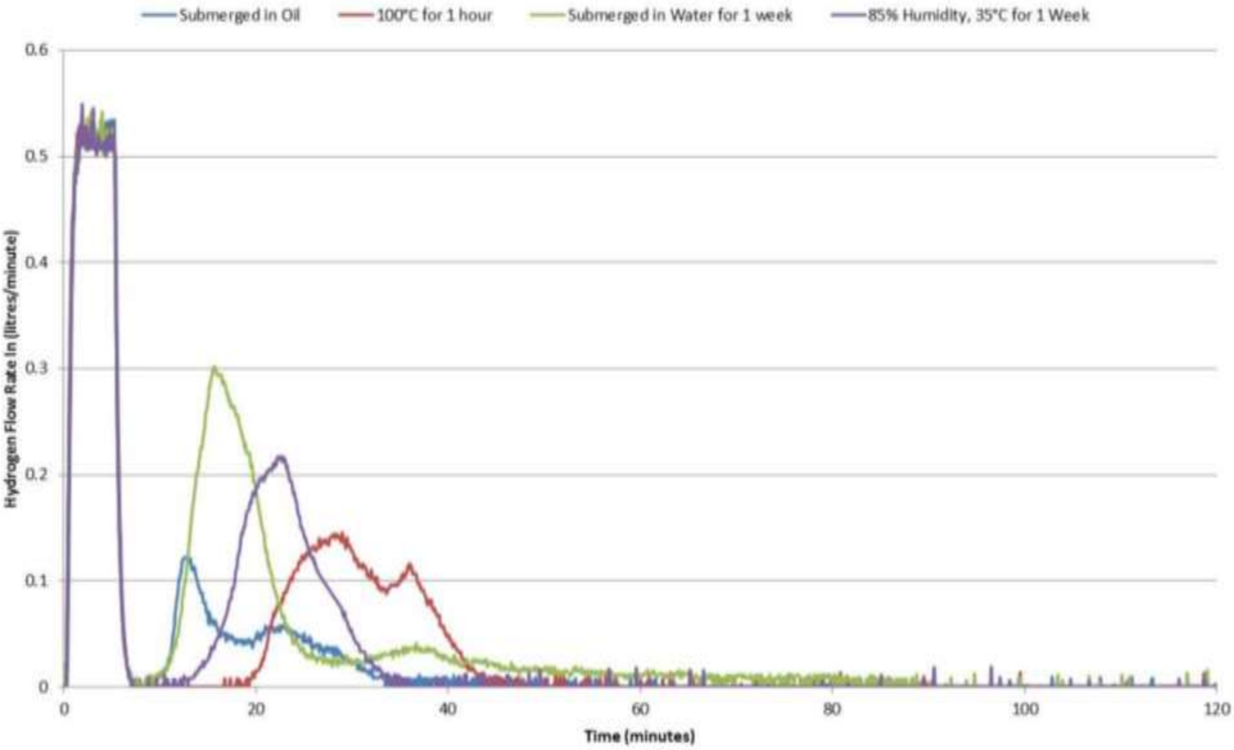
Extraction of sintered magnets from WEEEE by hydrogen decrepitation

The conditions required for hydrogen decrepitation depend significantly on the form of the input scrap. In general the condition of the HDD scrap material is good as the casing of the HDD and the nickel coating of the VCM magnets protects the Nd-Fe-B from damage and corrosion. If the HDDs are shredded the Nd-Fe-B becomes heavily oxidised and the only option is chemical extraction. It is therefore important that the cutting process identified in the previous section does not cause potential degradation to the magnet.

Trials have been performed at UoB to assess the level of surface damage to the VCM magnet required to expose the Nd-Fe-B prior to hydrogen processing. Distorting the assembly appears to be the most appropriate method. As long as oxide free Nd-Fe-B is exposed prior to the hydrogen processing stage the initiation of the HD reaction is not hindered. A distorted voice coil assembly is shown in the figure below.



The storage of VCM magnets is also likely to play a crucial role on the surface condition of the Nd-Fe-B magnets and the contamination levels of the material; this in turn is likely to affect the HD process. HD trials have been performed on VCM magnets that have had the nickel coating removed prior to being subjected to different harsh environments to mimic potential '*worst case*' conditions that may be experienced during collection and storage of VCM magnets. The impact that storage has on the HD process is demonstrated by the uptake of hydrogen during the process itself. This is demonstrated in the graph reproduced here.



All of the VCMs reacted except for those with the thin layer of automotive oil which formed enough of a barrier to prevent the initiation of the hydrogen decrepitation reaction. This series of experiments illustrates the robustness of the HD process to environmental degradation of the magnets surface.

Development of the hydrogen decrepitation (HD) process

The conditions required for successful HD are likely to depend significantly on the form of the input scrap; i.e. composition, type of coating and degree of oxidation/corrosion. The Nd-Fe-B magnets contained within the scrap waste streams identified in WP1 may have different compositions depending upon the original application. Dysprosium for example is added to many NdFeB magnets to increase the coercivity (resistance to demagnetisation) of the material.

Preliminary studies performed on the Ni coated VCM magnets processed in 1 bar hydrogen with no surface damage to the Ni coating, revealed that only ~50% of the magnets will react. Complete VCM magnets are shown here.

However, when these magnets were broken in half then 100% of the magnets reacted in hydrogen and the decrepitation was fully completed in <2 hours The Ni coating peeled away from the surface of the magnets into large (1-10mm) and small pieces (100μm-1mm).

Subsequently 10 broken VCM magnets were placed in air for up to 30 days and they still all reacted with hydrogen after this time.

To investigate the effect of composition, coating and degree of oxidation on the HD process in more in depth trials have been performed at UoB on magnets supplied by Magneti with different compositions and coatings.

a) Passivated – 10.45mm ø x 2.6mm, 8.45wt% Dy
b) Al coated – 24mm x 14mm x 5mm, 6.35wt% Dy
c) Uncoated – 9mm ø x 6mm, 3.58wt% Dy
d) NiCuNi coated – 5mm ø x 7mm, low Dy

The composition and coating of the magnets supplied are given below and compared with a standard voice coil magnet

Coating	**Composition (wt%)**								
	Nd	Dy	Pr	Co	B	Ga	Cu	Al	Fe
Ni*	25.7	1.33	3.43	1.3	1.0	0.53	0.09	0.31	Bal
Passivated**	22.65	8.45	0.6	3.0	0.9	0.22	0.15	0.15	Bal
Al**	24.75	6.35	0.6	3.0	0.9	0.22	0.15	0.15	Bal
Uncoated**	27.52	3.58	0.6	3.0	0.9	0.22	0.15	0.15	Bal
NiCuNi**	Composition unknown but thought to be low Dy. (ICP analysis is being performed)								

** Standard voice coil magnet*

*** Magnets supplied by Magneti*

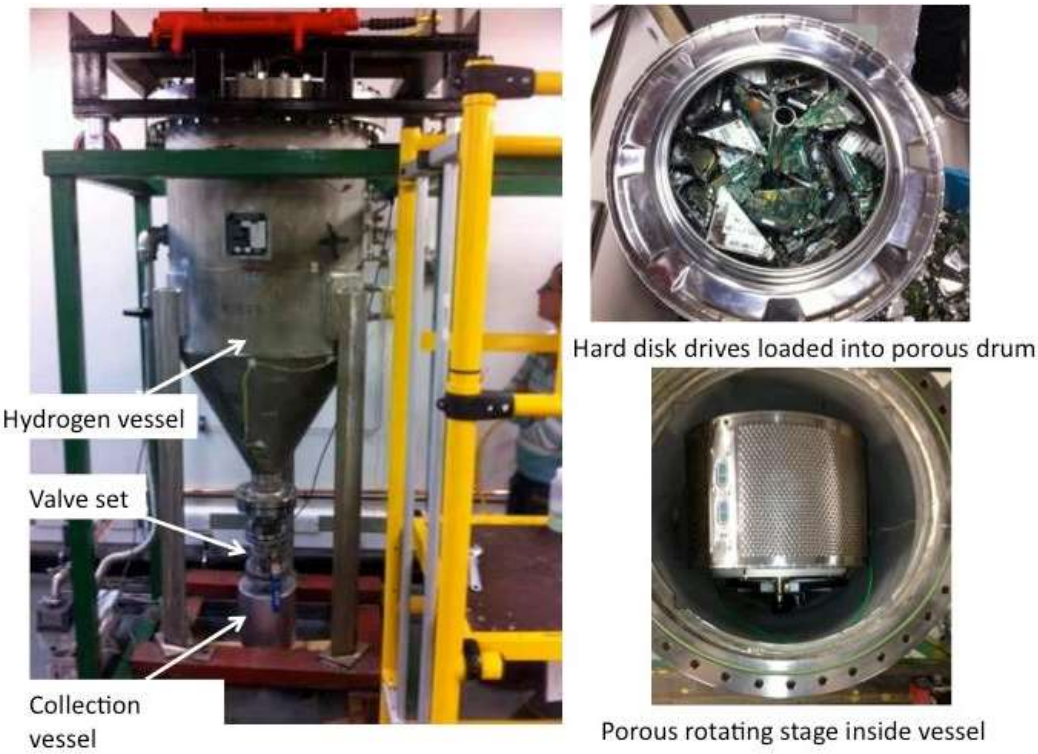
The surface of five magnets of each type were treated in the following manor and then exposed to 1 bar of hydrogen at room temperature for 2 hours:

- No surface damage;
- Coated magnet broken into a few pieces;
- Coating removed by grinding with P120 SiC paper;
- Coating removed by grinding with P120 SiC paper and exposed to air for 1 week.

It is evident from these trials that to guarantee HD damage is required to the surface of all of the Nd-Fe-B magnet compositions whether coated or not

Powder separation from bulk of waste stream

Work at UoB has focused on the development of the hydrogen reaction vessel and peripheral equipment for the separation of NdFeB powder from the bulk of the waste stream. A rotating porous drum (with 2-3 mm holes) has been incorporated into the existing large pilot plant vessel similar to commercial systems employed for HD processing of cast NdFeB. Rotating the porous drum after the HD reaction is complete causes enough agitation for the NdFeB and smaller contaminants to fall through the holes of the drum for collection while the larger material remains in the drum.



Hard disk drives loaded into porous drum

Porous rotating stage inside vessel

Figure: Hydrogen reaction chamber and porous stage

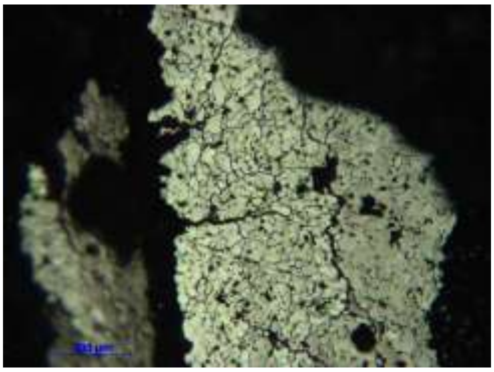
Figure Collected material that passes through the holes in the porous drum after the HD reaction

Two types of scrap feed have been made available during the project to date; cropped hard disc drives and VCM assemblies that have been manually separated. The difference in packing efficiencies between the cropped corners and the voice coil assemblies has been calculated by filling a known volume with the different scrap feeds. It takes ~350 cropped corners from 3.5 inch hard disc drives to fill the porous drum which equates to ~5kg of Ni coated magnet. However, the corners are very tightly packed which may prevent the hydrogenated NdFeB powder from being liberated from the sections after the HD process when the drum is rotated.

Purification of the rare earth content of extracted powders

Work at UoB has concentrated on refining the process for removing impurities from the HD NdFeB powder that pass through the holes in the porous drum. These impurities could include things such as coating residue, screws, pieces of plastic etc. Previous trials have shown that simple milling and sieving techniques can be employed to break down the NdFeB while leaving the Ni from the coating and other impurities relatively intact.

The most likely impurity is from the Ni coating on the voice coil magnet itself. As a result of the HD process the sintered NdFeB cracks along the grain boundaries producing a highly friable powder and in the most part the Ni coating peels away from the magnets in sheets. The HD NdFeB can be easily broken down into a finer particle size when compared to the Ni flakes or other impurities.



HD NdFeB particle showing the cracked structure

HD VCM magnet showing the large pieces of Ni

Different combinations of sieving and milling stages have been trialled with an increase in through put of the HD NdFeB material. Final trials were performed with a system to assess the benefit of using ultrasonic deblinding as a method of breaking up the friable NdFeB particles. HD material was passed through a 750 µm sieve to remove the larger contamination particles prior to using the deblinding system, it is important to note that the powder was not burr milled during this trial.

It is found that the use of the ultrasonic mesh greatly reduces the time required for the purification stage and therefore has the potential to eliminate the bottle neck in the processing chain.

Production of magnets from recycled rare earth magnet powders

Once the powder has been subjected to hydrogen decrepitation and separated from the contaminants, the material can either be sent for the manufacture of new sintered magnets, or it can be processed to make it suitable for the production of polymer bonded magnets. Both routes have been fully tested during the project.

In order for re-sintering to take place and if the extracted powders were to be re-melted then it is possible to extract the hydrogen from the hydrogenated powders. The hydrogenation behaviour is well known for primary cast NdFeB alloys[3] (as represented schematically in Figure) but not for secondary hydrogen processed sintered magnets. Therefore, the degassing behaviour of hydrogen processed sintered magnets was compared to results for primary NdFeB alloys.

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[3] Williams A.J. *Ph.D. Thesis Department of Metallurgy and Materials, The University of Birmingham* (1990)

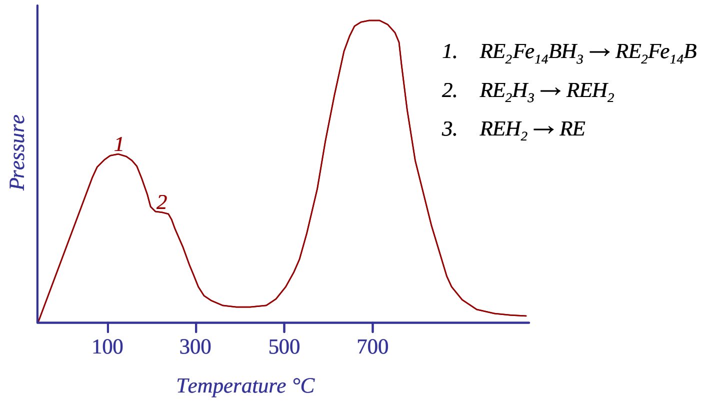


Figure Schematic representation of the desorption of hydrogen from cast Nd15Fe77B8

Once the HD Nd-Fe-B had been purified the powder was inertly loaded into a furnace tube and sealed; this was then transferred to a vacuum furnace system. This produced degassed material, which was sent to the company Magneti for processing at their site. One sample was derived from single composition scrap and one from mixed composition. The material sent was produced over several months so that there was sufficient material to use the commercial manufacturing procedure. The recycled material was blended with primary powder in varying ratios, jet milled, aligned, pressed and sintered.

The magnetic properties of the sintered magnets produced compare favourably with magnets made from fresh material, and the performance between those made from a single and a mixed source are similar. Critically, the magnets that were produced showed very little variation in magnetic properties across the batch produced. This is important to ensure quality control in the final manufacturing process and supply to end users.

The alternative to the production of sintered magnets, is the production of bonded magnets. For bonded magnets, the powder has to be further processed using the extra stage of hydrogenation, disproportionation, desorption and recombination (HDDR). This process step is vital for the production of high quality powder for the direct use in the manufacture of bonded magnets. Again, sufficient material was produced so that the commercial manufacturing process could be used.

The HDDR processing stage was investigated in detail to assess the impact of multiple variables. These included:

- Variable starting compositions
- Contamination levels of Ni
- Processing temperature
- Processing pressure

In addition to the change in processing conditions, the scale of operation was also altered during the experimental tests. It was shown that the larger processing produced a more consistent final product. While it was relatively simple to optimise the process for a single source of recycled material, the processing can be more complex when materials are mixed just prior to the HDDR process. However, tests were conducted on material following the hydrogen decrepitation process that was then milled and sieved. This process appeared to homogenise the mixed feed so that the HDDR process produced almost identical results for the different batches of mixed feed.

Following the HDDR processing, sufficient material was sent to the partner Kolektor for the production of new bonded magnets made entirely from recycled powder. After a number of optimisation steps, the final magnets that were produced compared favourable to existing magnets that are available commercially. As with the sintered magnets, different magnets were tested to confirm the reproducibility of the final magnetic properties. Of the five magnets that were randomly chosen from the final production run, the magnetic properties were almost identical, and well within the expected range that would be produced commercially.

Extraction of particles from polymer bonded magnet materials

The work described below covers both the extraction of the magnetic powders from the polymer bonded magnets, and the processing of the mixed waste stream. The aim is to produce an end product that can be fed to a molten salt cell, which can then produce Nd metal, which can be returned to the market. These waste streams are considered of lower quality than those that are processed using hydrogen decrepitation. The materials can be derived from any number of sources, and the process developed has been designed to offer a standard leaching and recovery process that can have different pre-treatment steps to obtain a feedstock to the main hydrometallurgical process of acid leaching with solvent extraction and precipitation.

Three polymer bound materials were provided by project partners

- Epoxy bound rings approximately 45mm OD, 35mm ID
- Nylon PA12 bound dog bones
- PPS cast in various shapes.

In addition, a supply of material produced following the shredding operation performed at Stena was sent for testing. This material comprises the smaller pieces of hard disk drives that are not sent for processing as part of the aluminium recovery step.

The first stage of the process to recover the magnetic material was to identify a mechanism to make the bulk of the bonded magnetic accessible to chemical processing. Thermal processing was tested initially, to determine if differences in thermal expansion could be used to produce a friable product. Standard thermal heating was used as well as microwave and radio frequency heating. The latter two were tested to assess the opportunities for heating the magnetic particles and thus breaking the polymer bonded magnets. However, none of the thermal processing routes tested produced friable material. Some arcing took place during certain tests, but the polymer bonded magnets remained intact.

Simple breaking and crushing experiments were then conducted. It was found that these materials were relatively soft and it was easy to crush all the magnet samples. The ease in which these samples could be broken was indicative that simple physical methods of crushing and leaching would be more cost effective than using thermal techniques to crack the polymer bonded magnets.

Once the bonded magnets had been physically treated, steps to leach out the particles were investigated. All three types of polymer bonded magnets were tested with a range of potential leach reagents, which included mineral acids and ionic liquids. The tests were conducted to assess the recoverability of the metal particles from the polymer.

The final leach process chosen used hydrochloric acid, as this produced a solution that is compatible with the subsequent hydrometallurgical processing steps. The final hydrometallurgical process route comprised an acid, followed by an organic extraction, and a final stripping process step. The final stripping stage produced a three phase solution with a return organic solvent, a Nd containing solid precipitate, and aqueous acidic stream that could be processed further if necessary. The whole process was optimised to recover the economically critical materials, and to minimise the waste streams by testing the number of times a solution could be recycled.

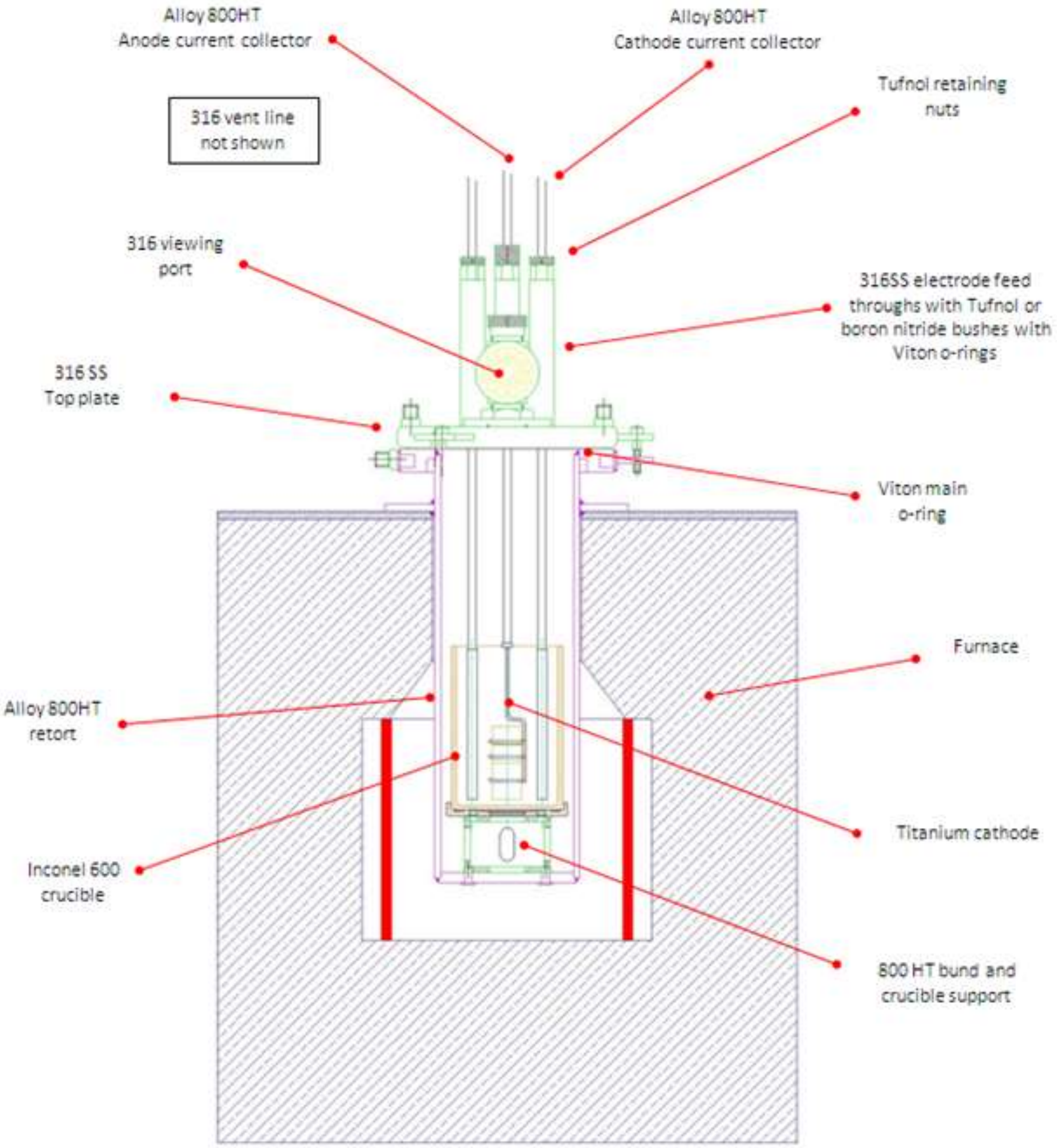
Tests on metal loading and the range of concentrations used for each step were conducted to optimise the process for the final economics. Tests were also conducted with a wide range of ionic liquids for the different extraction stages. It was found that no ionic liquid resulted in a viable economic solution to the recovery of Nd from the waste stream. Although some results were promising, the economics of the process would not allow for the losses in the ionic liquid that would occur during the processing stages.

The final procedure demonstrated a recovery of lanthanides from NdFeB alloy magnets with purity of the lanthanide product > 90% and an overall recovery through multiple stages of approximately 75%.

The key economic stage is that of the organic solvent in the liquid/liquid extraction step. It has been shown that the extraction of the Nd from the leach solution did not drop over five cycles. Multiple stages of stripping the organic solvent is possible with good recovery of the solvent back in to the main process.

Production of Nd Metal

Once the solid Nd material has been recovered from the solvent extraction process, it can then be fed into a molten salt electrochemical cell, for the recovery of the Nd metal. The molten salt cell comprises a retort, which has a crucible sitting inside it. The anode and cathode are introduced from the top of the retort, and the whole unit is placed inside a furnace.



The choice of the salts to be used was made based on an extensive literature review, and then modified with experience during the testing of the rig. The operational parameters for the electrochemical window were determined during several phases of testing. Cyclic voltammetry was used to determine the voltage window available for reduction of the oxidised neodymium to the metal.

Once the operational window was identified for the experimental rig, then optimisation of the process could take place. This included changing the salts used in the cell, as well as changing the ratio of salts used. It was also possible to show that the process was a diffusion controlled process within the cell.

Having determined the voltage window under a wide range of conditions, a constant voltage is then applied that sits within the window, and the current is monitored over a period of time. The production of Nd metal is then calculated according to the current density and the area of the cathode. This can be used for the scaling up of the process to determine production capacity at the larger scale.

Technical Prototype Demonstration and Validation

The project Remanence has set up and tested the entire supply chain from the collection of hard disk drives to the manufacture of magnets from recycled powders. The project has also identified a processing route for RE magnetic material that cannot be processed directly from the recovered powders to new magnets. The current virtual process for the production of magnets operates at four sites in four EU countries:

Sweden – collection and separation of the hard disk drives and production of the corners containing the RE magnets (Assessed TRL – 7 – System prototype demonstration in an operational environment)

United Kingdom – Processing of the HDD corners to produce recycled powder that can be used for the production of either new sintered or polymer bonded magnets (Assessed TRL – 4/5 Small to large scale prototype tested in the intended environment)

Germany – For the production of polymer bonded magnets (Assessed TRL – 7 – System prototype demonstration in an operational environment)

Slovenia – For the production of sintered magnets (Assessed TRL – 7 – System prototype demonstration in an operational environment)

The production rates in Sweden, Germany and Slovenia are effectively at full commercial scale. The production of the powders in the UK is conducted over a few weeks to enable the full commercial scale production of the new magnets in both Germany and Slovenia. It has been estimated that the production rate for the unit operating in Sweden is capable of processing almost one million units per year.

Options for the final location of the different units will determine much of the economics in terms of the transport of material from one location to another. Also, investment in some site infrastructure would be reduced if the processing of materials takes place at locations that already have the functionality in situ.

The parameters for the lower grade material have also been determined. Although no locations have been used for the larger scale operation, there are many sites in Europe that can perform the hydrometallurgical processing. Also, locations exist in the EU that can perform the processing of the recovered metal oxide to the final neodymium metal.

Potential Impact

Socio economic impacts

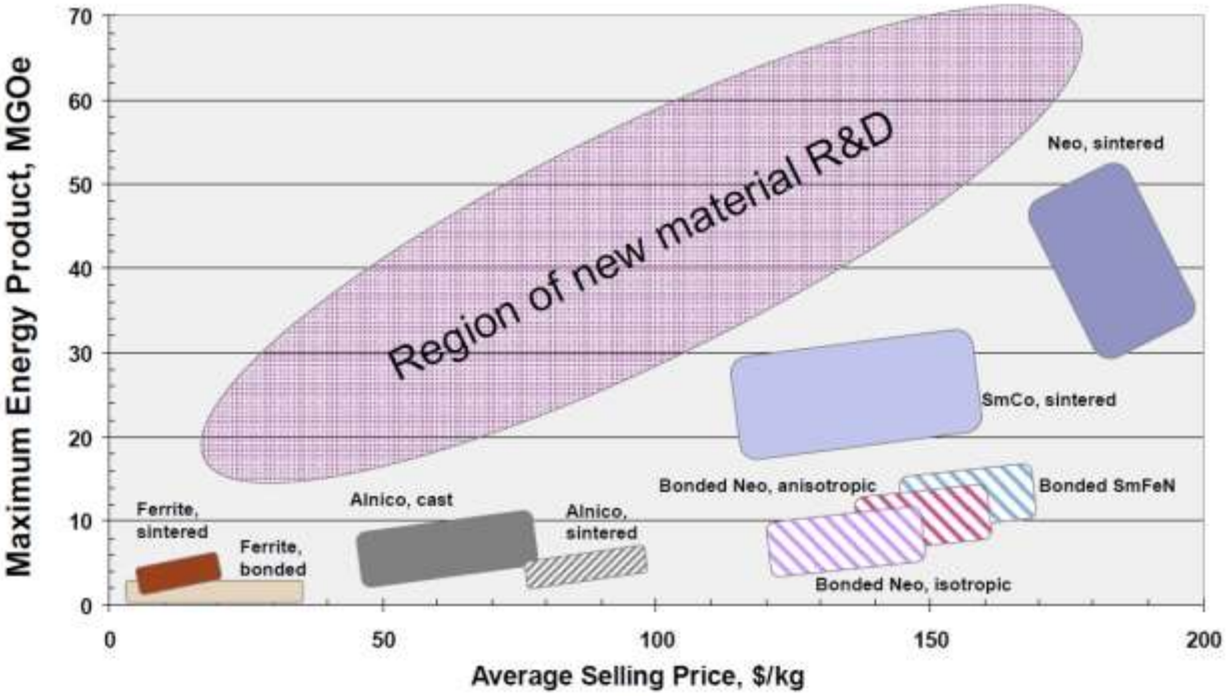
A key objective of the project Remanence was to identify the potential opportunity to build a supply chain for the supply of rare earth magnets. This would serve the dual purposes of reducing the impact of any future issues over the potential of restricted supply as well as providing the opportunities for increasing the added value of nominally a waste stream. Thus, within the EU market there is

- An opportunity to offset some of the large imports of NdFeB cast and powder materials for bonded and sintered magnets by substituting lower cost recycled product
- An opportunity to derive secondary NdFeB materials from redundant equipment, and this may extend to other equipment, once the principles of the new supply chain were developed and shown to be economic
- The potential to replicate the process in other EU locations

Two process routes have been identified: the first for the higher quality recyclable material (i.e. the material that remains in its alloyed state for the entire process); and a lower quality route, which can be applied to a wide range of materials if they cannot be recovered in their sintered magnet form.

The project was proposed when the price of rare earth elements and their products was increasing rapidly and there were potential issues over scarcity of supply. This issue has significantly decreased during the progress of the project, but it has been found that the economics of the proposed production of magnets from recycled sintered powders may still be competitive. Also, end-users of rare earth magnets are now interested in securing alternative supplies to ensure that their businesses are not impacted by any future dramatic variations in price or supply.

The proposed virtual process has been fully developed during the project with different process steps being conducted in four different EU countries. A techno-economic assessment has been conducted on multiple scenarios. Costs for all aspects of the production, transfer between sites, establishing new sites for production or using existing sites that have the required infrastructure for the high cost aspects of the processing route have been investigated. These have then been compared with existing costs of manufacture of the powder and costs of rare earth magnets. Extreme care must be taken when comparing the potential costs of the magnets produced from the recycled material, and magnets that are offered at unsustainable prices. This has been avoided in the comparisons made in this project by identifying the prices offered over a period of time and thus representing price that covers the full costs of processing and a realistic profit for the manufacturer. These prices are still significantly below the cost of rare earth material and magnets that existed at the start of the project.



There is a general relationship between the maximum energy product of a magnet and its selling price. Remanence first looked at the opportunities for competing with the neodymium sintered and bonded markets, i.e. up to $200 /kg for the sintered magnets and between $100 to $150 for the bonded magnets.

The economic analysis conducted for the new virtual process route has shown that even with the current inefficiencies that exist then the cost of production of magnets would allow for the recycled magnets to compete with those produced from fresh NdFeB material. However, a further analysis has identified cost savings that can easily be achieved once the process route has been streamlined (e.g. moving the production facilities closer to each other and using plants that have existing hydrogen infrastructure). Also, the labour costs for the production route will be decreased as the equipment moves from the development stage to full production capability. This will also be associated with an increased processing speed of the hard disk drive identification and separation unit. It has been shown that the current capacity can be significantly increased with optimisation of the conveyor system.

With these modifications to the overall process a revised economic assessment has been made. This has shown that the potential cost of manufacture of the bonded magnets could fall as low as to compete with ferrite bonded magnets. This would open up an exceptionally large market and provide new opportunities for the project partners.

Societal impacts

The use of rare earths materials is embedded in modern lifestyles, and this extends beyond the use for magnets. Therefore, any technology that can recover material that is already in use and retain this in a relatively tight closed loop recycle process will benefit society as a whole.

Rare earth element	End use
Lanthanum	**Batteries**, metallurgy, **catalysts** (catalytic converters, petroleum refining), polishing powders, glass additives, phosphors, ceramics, hybrid engines
Cerium	Batteries, metallurgy, **catalysts** (catalytic converters, petroleum refining), **polishing powders**, **glass additives**, phosphors, ceramics, hybrid engines
Praseodymium	**Magnets**, batteries, metallurgy, catalysts (catalytic converters), polishing powders, glass additives, ceramics
Neodymium	**Magnets**, batteries, metallurgy, catalysts (catalytic converters), glass additives, ceramics
Gadolinium	Magnets, diagnostics, phosphors
Europium	**Phosphors**, red color for TV and computer screens, medical X-ray units
Samarium	Batteries, magnets
Scandium	Aerospace components
Terbium	**Phosphors**, permanent magnets
Dysprosium	**Magnets**, hybrid engines, ceramic capacitors
Erbium	Phosphors
Holmium	Glass coloring, lasers
Thulium	Medical X-ray units
Lutetium	Catalysts in petroleum refining
Ytterbium	Lasers, steel alloys
Yttrium	Red color, **phosphors**, **ceramics**, metal alloys, glass additives

For rare earth magnets alone, there is a vast array of end uses. These include:

- Industrial motors and generators, wind turbines
- Hard disk drives, CD and DVD drives
- Electric bicycles
- Transducers / loudspeakers
- Magnetic separation equipment
- MRI scanners
- Sensors
- Air conditioning units
- Various motor traction drives and units

As the performance / cost ratio is improved, so the use of the rare earth magnets will increase and so enhance today's modern lifestyles.

There is also an impact on society if these materials are to be recovered. Remanence immediately focussed on the recovery of rare earth magnets from hard disk drives as the hard disk drives are already separated for specific processing, and the magnets are easily accessible. By showing that the processing of these magnets is economically viable then this may encourage recycling of magnets from other products. The cost of collecting these materials may be prohibitive if this cannot be integrated easily into existing waste collection regimes. Additionally, the design of products in the future is likely to be key to increasing the recovery rates of magnets used for different purposes. With a society that is more aware of the environmental implication of its actions, and with better communication between designers and manufacturers, then future recycling may increase significantly once an economic process is established for the production of new magnets from waste sintered magnets.

The final impact on society is by developing new opportunities within the EU for the manufacture of rare earth magnets and the products that use them. If this can be increased there will be greater employment and also the greater uptake of technology that impacts on lifestyles (e.g. wind turbines, use of magnets in cars for ease of use etc.).

Environmental impacts

A full life cycle analysis was conducted as part of the project. The data has been processed using the GaBi software for LCA. Three routes were assessed:

- Route 1: Separation and concentration process + HD processing to produce NdFeB powder suitable for sintered magnets (compared with baseline 1)
- Route 2: Separation and concentration process : HD process + HDDR processing to produce NdFeB powder suitable for polymer bonded magnets (compared with baseline 1)
- Route 3: The recovery of lower grade material to produce Nd metal (compared with baseline 2)

Two baseline scenarios were used to compare with the 3 different routes:

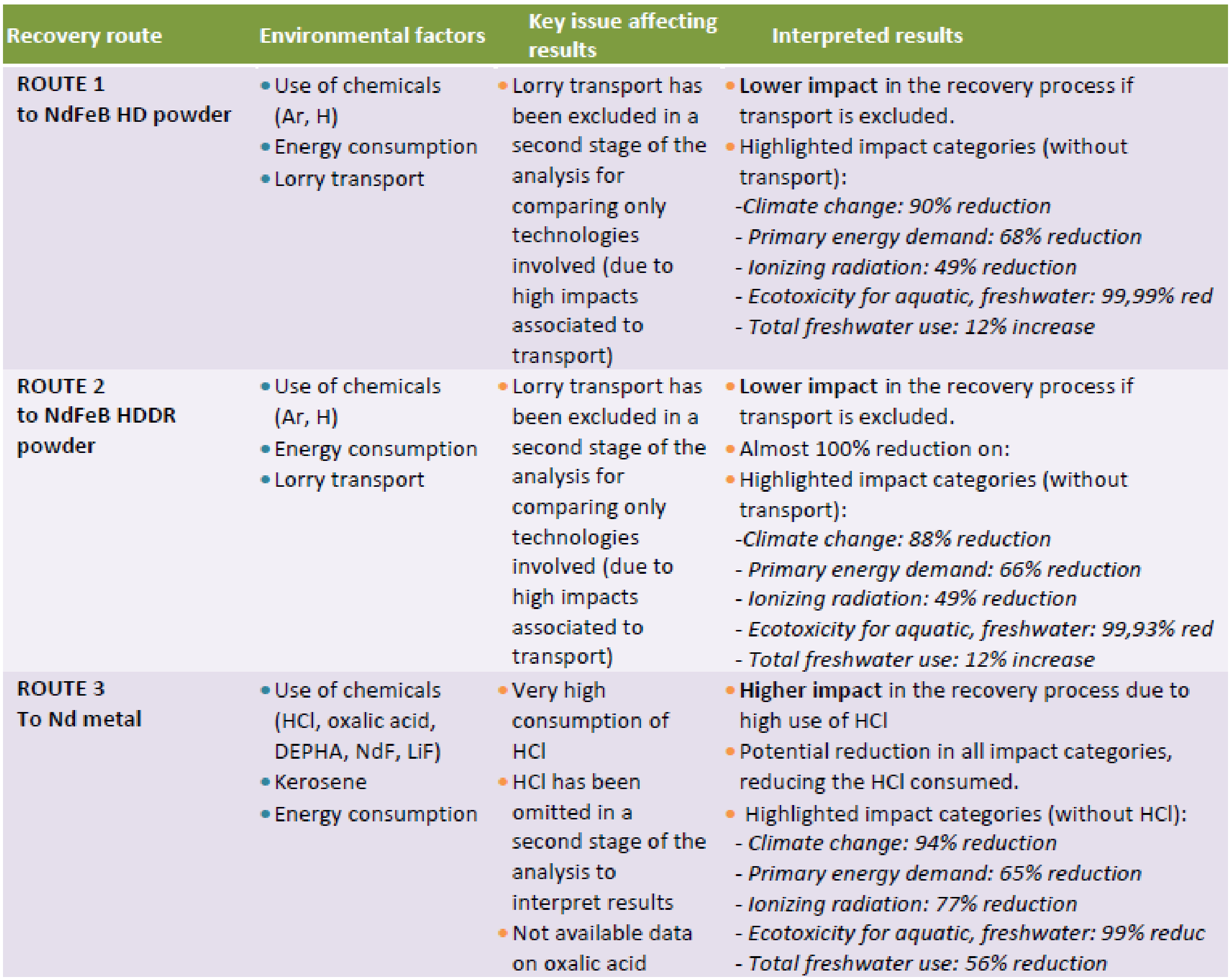
- Baseline 1: The production of RE material and including transportation of the produced material from China to EU
- Baseline 2: for the production of Nd metal and also including the transport from China to the EU

With the assumptions used, the environmental impacts of RE primary production are dominated by the ionisation radiation associated with the thorium, and sometimes uranium, which is co-deposited with rare earth ores. The concentration of radioactive elements is usually relatively benign for human health, but this rises significantly during the processing of the ore. The importance of the other factors was highlighted by removing the impact of the ionizing radiation. The top five impacts were:

1. *Ionizing radiation*
2. *Primary energy demand (PED)*
3. *Ecotoxicity for aquatic, fresh water*
4. *Total freshwater use*
5. *Climate change*

Data was collected from the partners involved in each of the stages, and using a variety of assumptions based on the current and potential future operation of a fully developed processing route. The production of the magnets themselves was outside of the scope of the LCA as it was assumed that the same impact would be incurred whether the magnets were produced from fresh or recycled material. This is consistent with the approach adopted during the project, which aimed to produce materials that could be used in the same way as the fresh virgin material.

For both route 1 and route 2 the impact of transport makes a significant difference in the overall impact of the new routes. Currently, the process is operating in different locations, but if these locations can be combined then the environmental and energy impacts are significantly reduced.



Recovery route	Environmental factors	Key issue affecting results	Interpreted results
ROUTE 1 to NdFeB HD powder	• Use of chemicals (Ar, H) • Energy consumption • Lorry transport	• Lorry transport has been excluded in a second stage of the analysis for comparing only technologies involved (due to high impacts associated to transport)	• **Lower impact** in the recovery process if transport is excluded. • Highlighted impact categories (without transport): -*Climate change: 90% reduction* - *Primary energy demand: 68% reduction* - *Ionizing radiation: 49% reduction* - *Ecotoxicity for aquatic, freshwater: 99,99% red* - *Total freshwater use: 12% increase*
ROUTE 2 to NdFeB HDDR powder	• Use of chemicals (Ar, H) • Energy consumption • Lorry transport	• Lorry transport has been excluded in a second stage of the analysis for comparing only technologies involved (due to high impacts associated to transport)	• **Lower impact** in the recovery process if transport is excluded. • Almost 100% reduction on: • Highlighted impact categories (without transport): -*Climate change: 88% reduction* - *Primary energy demand: 66% reduction* - *Ionizing radiation: 49% reduction* - *Ecotoxicity for aquatic, freshwater: 99,93% red* - *Total freshwater use: 12% increase*
ROUTE 3 To Nd metal	• Use of chemicals (HCl, oxalic acid, DEPHA, NdF, LiF) • Kerosene • Energy consumption	• Very high consumption of HCl • HCl has been omitted in a second stage of the analysis to interpret results • Not available data on oxalic acid	• **Higher impact** in the recovery process due to high use of HCl • Potential reduction in all impact categories, reducing the HCl consumed. • Highlighted impact categories (without HCl): - *Climate change: 94% reduction* - *Primary energy demand: 65% reduction* - *Ionizing radiation: 77% reduction* - *Ecotoxicity for aquatic, freshwater: 99% reduc* - *Total freshwater use: 56% reduction*

There are still significant opportunities to optimise the process as it moves from the virtual process with materials being shipped around Europe, to a process where the unit operations can be brought into a single location. The production of the two powders shows that there is approximately 90% reduction on climate change and almost 70% reduction on energy demand, when considering the production technologies between processing of the virgin material and the recycled material.

The results for the hydrometallurgical processing are not as clear, as this process was operated at the laboratory scale only, and was not optimised and so did not have the possibility to provide accurate values for the production process. It was found that the impact in the LCA was dominated by the use of the first acid leach process. This dominated all the results from the LCA. However, the use of the initial acid leach should not be any more onerous for the recovery of the recycled material than for the extraction from the original ore. The LCA then calculated the impact for the remainder of the process, which was optimised during the laboratory experiments. Again, significant reductions in climate change and energy demand were shown to be possible, with these being over 90% and almost 70% respectively.

Main dissemination activities

Dissemination and training has taken place both internally and externally to the project consortium. Internal training took the form of presentations and tours of equipment facilities at the sites of the partners. The presentations have been given by a range of experts from the different project partners, and have covered the issues covering the whole supply chain, from collection of the WEEE to the production of the final magnets. This has enabled the project partners to gain an insight into the entire operation, and so enable the ability to assess the relevance of each of the individual aspects of the project into the whole process.

Public dissemination has taken place both through the presentation of papers, interviews with local press, and engagement with similar projects funded as part of either FP7 or Horizon 2020.

The key international technical conference that takes place every two years is the Rare Earth and Permanent Magnet Conference (REPM). Papers, presentations and posters have been given at both the 2014 event and the 2016 event. This activity is highly focussed towards other experts in the area of rare earth magnets.

Interaction with the wider recycling community has taken place with participation in events that are more generic, and cover the wider recycling and circular economy community. For the final dissemination event, sponsorship was provided to the Circular Materials Conference, Gothenberg, May 2016. The project was represented by taking a stand at the event, presenting two posters, and presenting in a workshop during the event. The EU funded project ProSUM was invited to share the Remanence stand, and so demonstrating good collaboration with this major project for identifying the sources of future WEEE and other products that may contain materials suitable for recovery using the technology developed in the project.

Dissemination has also taken place with other relevant EU projects. For example the SPIRE programme of projects has hosted two events (2015 and 2016) to investigate the impact of SPIRE projects. Invitations were issued and accepted to attend and supply information to both of these two day events.

Wider dissemination is available via the Remanence website: http://www.project-remanence.eu/. Also, recent public interest has been demonstrated in the UK with a report made by BBC Midlands, which covered the processing of material at the University of Birmingham, as well a video of the sorting process taking place in Sweden. This is available via the BBC Midlands facebook page:

https://www.facebook.com/midlandstoday/videos/10154429322309761/

Exploitable results

A total of 16 exploitable results have been identified by the project partners following a number of internal exploitation workshops. These results range from the consideration of the entire process to exploitation of the specific operations that comprise the currently virtual production route. The results also cover opportunities for using the developments made during the project for activity other than the recovery of rare earth magnets. Each partner identified their potential past or future involvement in each of the results using eight categories. The categories represent different levels of future involvement as well as contributions to the delivery of that exploitable result.

Having identified the exploitable results, business plans were developed for those that were identified as of high importance. The Business Model Canvas approach was used to support the development of future plans. Different business model options considered include:

- Take-up by one of the project partner's and commercialisation of the output by this partner with adequate distribution channels
- Joint venture between a number of project partners collectively able to address the full business cycle
- Patenting of the output, with third-party licensing to commercialise
- Integration of the output in another solution and subsequent commercialisation

The key findings for the development of the market for recycled magnets have been developed in collaboration with all partners. Intimate knowledge of the final market opportunities was provided by the two project partners that manufacture and supply magnets. The final sale price that would need to be achieved for magnets that are manufactured using the recycled material is obviously critical. Both the final characteristics of the magnets produced as well as the reproducibility of those properties are crucial to the acceptance of a product made from recycled material. The price that will be paid for a product is roughly related to its maximum energy product, although factors such as performance at higher temperatures sometimes play a more important role for specific applications.

It has been found that the expected production costs of producing new magnets from recycled material will enable to manufacture of very price competitive products, even with the reduced prices currently being experienced.

The other key area of the project has been the development of a sophisticated and automated sensor and separation process. The unit that has been produced as part of the project can currently process over 500,000 units per year. The optimisation process has shown that this can be increased to approximately 1 million units per year with a few simple modifications. However, additional opportunities for the technology have also been identified and will be investigated by the relevant project partners.