REE4EU: Integrated High Temperature Electrolysis (HTE) and Ion Liquid Extraction (ILE) for a Strong and Independent European Rare Earth Elements Supply Chain

Project type: Innovation action
Start date of project: 01/10/2015 Duration: 48 months

D6.2 First Batch of PM derived from the ILE-HTE Pilot at ELKEM

WP n° and title	WP6. End Product Performance Validation and LCA
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Contributor(s)	LCM
Version	Draft v04-Final

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List of abbreviations and definitions

Abbreviation	Definition
B_R	Remanence of the permanent magnet, which is the measure of the remaining magnetization when the driving magnetic field is dropped to zero
BH_{max}	Maximum energy product of a magnet is an energy density and it is equivalent to the area of the largest rectangle that can be inscribed under the normal curve
D_{50}	Mass median diameter, or diameter at which 50% of a sample's mass is comprised of small particles
H_{cB}	Intrinsic coercivity of the permanent magnet, which is the field strength required to reduce the polarization of a magnetic material to zero
H_{cJ}	Coercivity of the permanent magnet is the measure of the reverse field needed to drive the magnetization to zero after being saturated
HD	Hydrogen decrepitation
HTE	High temperature electrolysis
ILE	Ionic liquid extraction
LCM	Less Common Metals
PM	Permanent magnet
RE	Rare earth
REA	Rare earth alloy
REMA	Rare earth master alloy
REO	Rare earth oxide
VAC	Vacuumschmelze

Executive Summary

The objective of this work was to validate the output from the new engineered REE4EU-pilot process by producing and testing functional permanent magnets from the obtained rare earth alloys (REA), characterise the manufactured product and compare its properties with those of the product obtained using virgin materials (primary resources).

In this deliverable document, the results obtained when using the output material (REA) from the ILE+HTE pilot installed at ELKEM are summarized. The REA was obtained using permanent magnet wastes as feed material, including both calcined swarf from permanent magnet manufacturing (provided by partner VAC) and spent permanent magnets. The REA was subsequently used as feed for the 600 kg strip caster at partner LCM.

VAC has made permanent magnets in his mass production line and has determined the quality of both the strip cast alloy input and the permanent magnet output in terms of magnetic properties, namely remanence, coercivity, intrinsic coercivity and maximum energy product, as well as chemical composition.

The results are compared to those obtained with magnets from VAC's production line using material derived from virgin input, being very similar. This means that the strip cast alloy produced from recycled permanent magnet wastes by the REE4EU process is suitable for mass production in VAC.

1 Introduction

During the first 3 years of the project the consortium has engineered, built, and run the REE4EU pilot plant installed at ELKEM.

The new pilot plant for the demonstration of the REE4EU technology consisted on two units, the ionic liquid extraction (ILE) unit, which converts the permanent magnet (PM) wastes into pure rare earth oxide (REO) mixtures, and the high temperature electrolysis (HTE) unit, converting the REO mixtures in rare earth alloys (REA) suitable for further processing into new PM.

During the REE4EU pilot trials, almost 1.5 tonnes of calcined swarf from PM manufacturing (PMS) and 500 kg of spent permanent magnet (SPM) were treated. The REA obtained was used by partner LCM to produce 600 kg of strip cast alloy.

The strip cast alloy was used further by partner VAC to produce PM on its production line. The goal was to qualify the strip cast material produced from recycled material for VAC's PM production line.

2 Methodology

The 600 kg strip cast flakes received from LCM were analysed in terms of PM composition and impurities as well as its microstructure.

For determining the optimal amount of milling additions and sintering conditions a 2 kg amount was milled to a fine powder in the laboratory and magnets were prepared and tested.

Then, the 600 kg strip cast REMA was converted to a coarse powder by hydrogen decrepitation (HD). Jet-milling with the optimized milling additions was used in order to prepare fine powders. 50 kg of that powder was pressed and sintered/annealed with the optimized sintering conditions in the production. The PM were tested and machined to the final size.

The magnetic properties of these test-PM were compared with magnets from VAC's mass production.

The whole process is schematically shown in Figure 1.

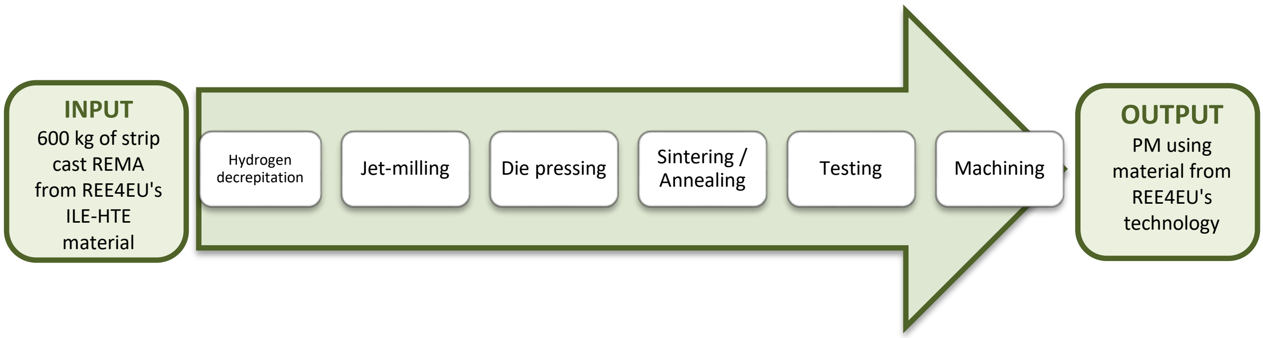


Figure 1. Schematics of the different steps followed in the manufacture of sintered NdFeB PM.

3 Results

3.1 Analysis of the REMA

Figure 2 shows the 600 kg strip cast flakes of rare earth master alloy (REMA) as they arrived from LCM at VAC.

Chemical analysis of the REMA is shown in Table 1. All elements were within the specification. Only B was a little low. But this could be compensated during jet-milling. That means the REMA was suitable for the purpose of the project.

Figure 2. 600 kg strip cast flakes from LCM as received at VAC

Table 1. Chemical composition of the REMA from Figure 2. Analysis performed at both LCM' and VAC's premises, and compared to the required specifications

	Nd	Dy	B	Co	Al	C
Spec.	31,1 ± 0,3	1,3 ± 0,3	0,93 ± 0,02	0,9 ± 0,1	0,10 ± 0,01	< 0,015
LCM	31,2	1,54	0,91	0,91	0,11	0,009
VAC	31,4	1,58	0,90	0,97	0,09	0,011

The structure of the flakes was investigated by SEM. The flakes have a typical thickness of 300 µm and width of the lamellae is about 3 – 4 µm (cf. Figure 3).

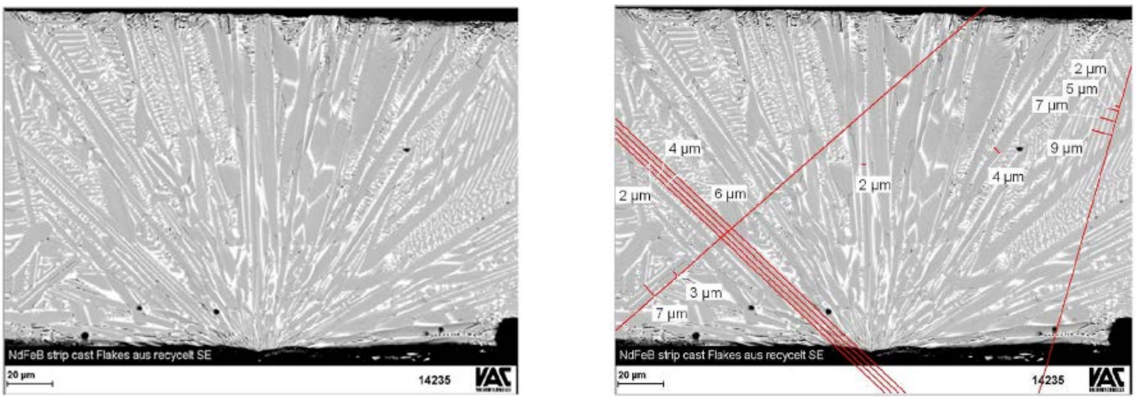


Figure 3. SEM micrographs of strip cast flakes. The figure on the right side shows the thicknesses of the lamellae

3.2 Laboratory investigations for defining the milling and sintering conditions

For defining the milling and sintering conditions for the later processing in the production line, 2 kg of the REMA flakes were taken for laboratory investigations and HD treatment in order to get a coarse powder for the jet milling.

In a first investigation the influence of the particle size was determined. Two particle sizes, 3.0 and 4.2 µm, were set by varying the classifier speed of the jet mill (cf. Figure 4).

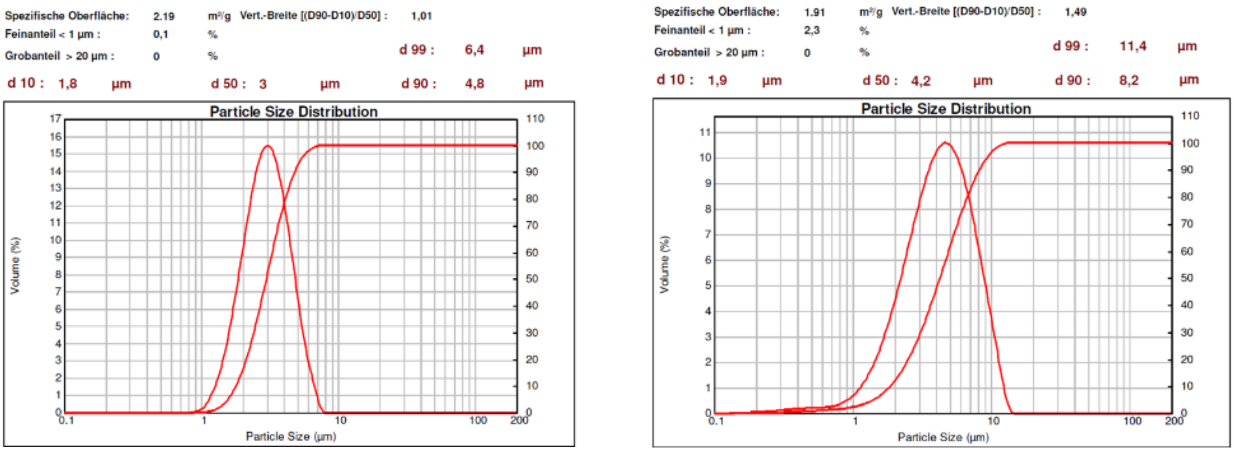


Figure 4. Particle size distribution of the two different powders (D_{50} = 3.0 and 4.2 µm)

In a next step the influence of the amount of Nd-rich grain boundary phase on the coercivity, H_{cJ} was investigated (cf. Figure 5). With increasing amount of Nd-rich phase H_{cJ} is increasing. The optimal conditions are found for the powder with the finest particle size and the lowest sintering temperature.

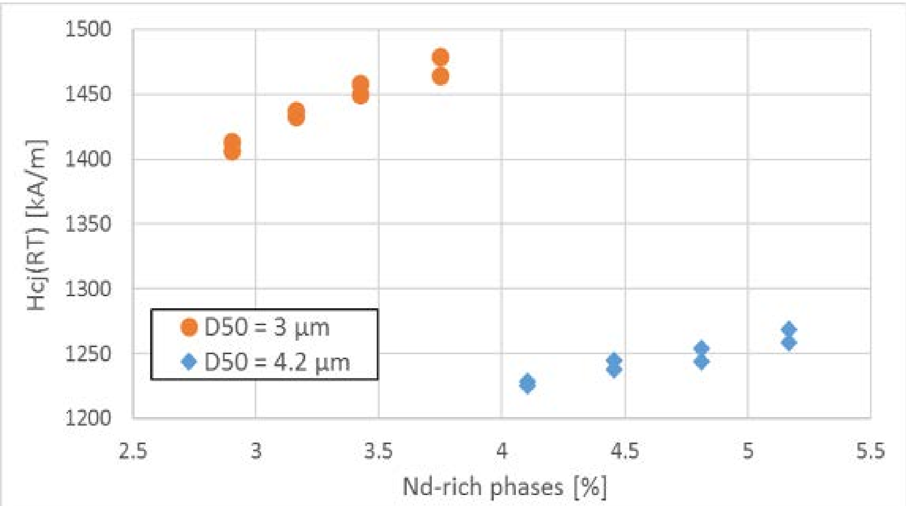


Figure 5. Dependence of the intrinsic coercivity H_{cJ} on the amount of Nd-rich phase in the magnet. Each of the measurements were carried out at two different sintering temperatures

During the jet milling some additives are fed into the mill. The additive B is increasing the coercivity. The optimum amount was found to be 0.15 wt% B added (see Figure 6). Also, in this case the lower sintering temperature revealed higher H_{cJ}.

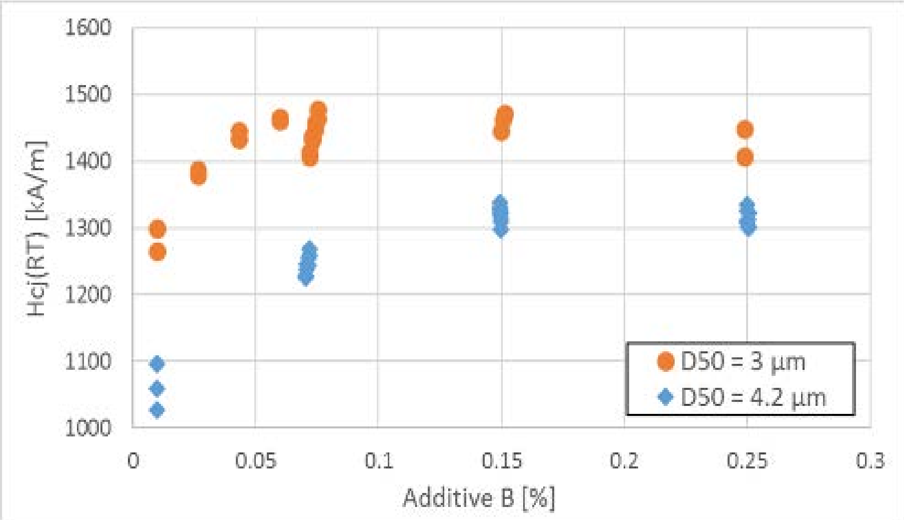


Figure 6. Dependence of the intrinsic coercivity H_{cJ} on the amount of B added during jet milling. Each of the measurements were carried out at two different sintering temperatures, the same as in Figure 5

The relation between the remanence B_R and the coercivity H_{cJ} for the above-mentioned investigations is shown in Figure 7.

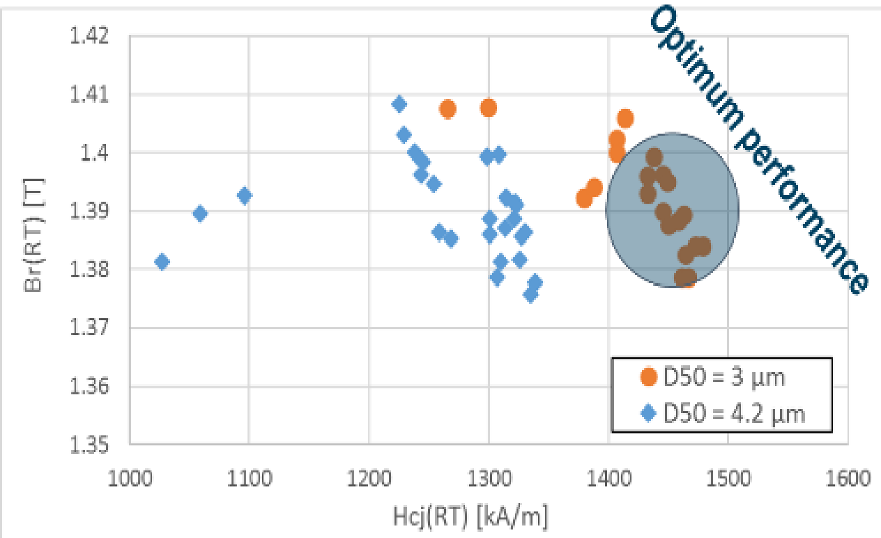


Figure 7. Relation between the remanence and coercivity for the different milling conditions that were investigated at the lab

It was defined that in order to use the REMA material delivered by LCM for new PM, the following conditions should be set in the production line:

- Particle size D_{50}: 3.0 – 3.5 µm
- Milling addition: 0.15 % B addition
- Amount of Nd-rich phase: 4 wt%
- Sintering temperature: low

3.3 Production magnets in the regular production line of VAC

After the optimum milling conditions were set, the 600 kg of REMA material delivered by LCM was first HD treated in the regular HD furnace of the production line. In the next step the coarse powder was milled in the standard jet mill of the production to a fine powder (D_{50} = 3.5 µm). Moreover, 0.15 wt% of B was added during milling.

50 kg of the fine powder was pressed on an electric servo press of the production to rectangular shaped magnets (32 x 13 x 3 mm³, magnetic orientation underlined). The pressing was done under inert N_2 atmosphere behind glass windows (cf. Figure 8).

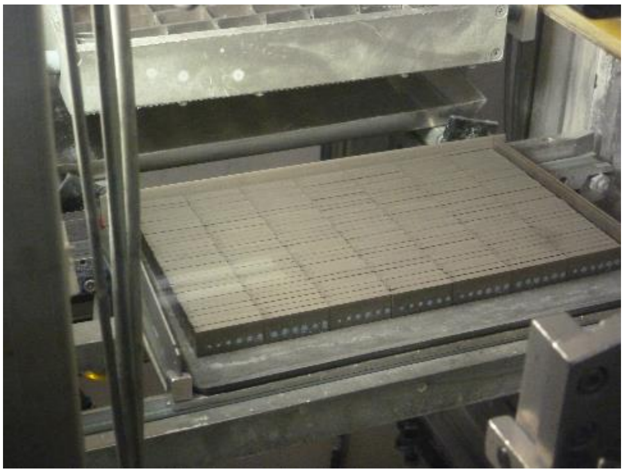
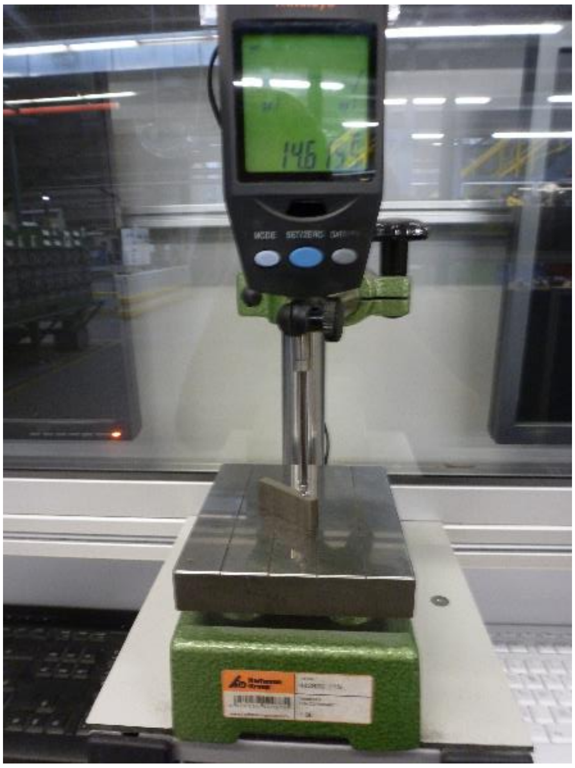


Figure 8. Pressed green bodies stored under inert N_2 atmosphere

The right dimensions of the manufactured PM were double-checked pressing, as shown in Figure 9.



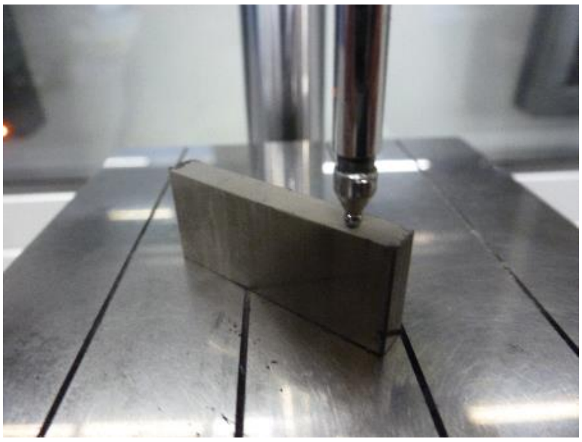


Figure 9. Dimension check of the green bodies directly after pressing

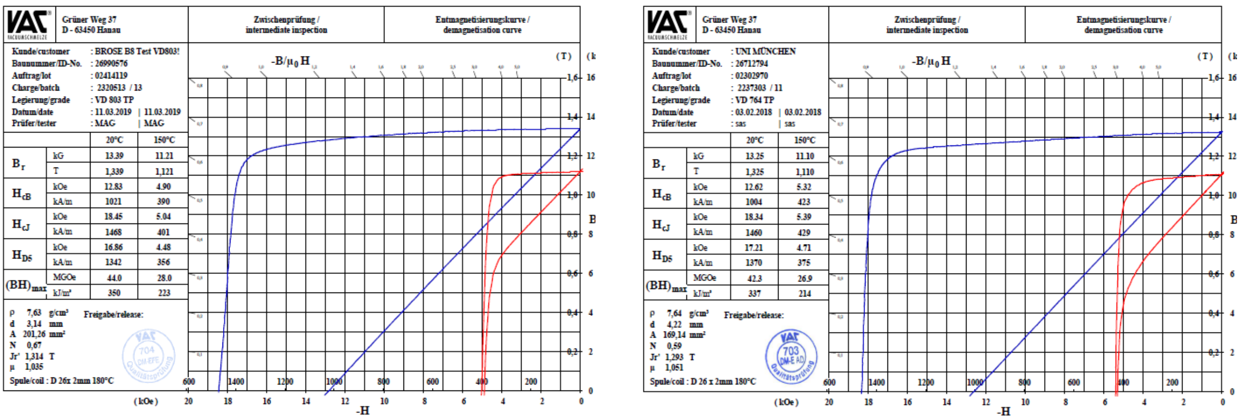
In the next step the green bodies were put on trays and these were then encapsulated in steel cans. The cans with the PM green bodies were then loaded into the sintering furnace and sintered and annealed at the proper temperatures. After sintering the cans were opened and some few magnets were taken out aleatory and the dimensions and magnetic properties were determined.

The sintered parts were ground for all three dimensions using grinding machines, then cleaned, phosphate coated against rusting and packed (see Figure 10).

Figure 10. Finished magnets from VAC mass production made from recycled material obtained in the REE4EU pilot

Figure 11 shows the magnetic properties of the obtained PM compared with those of conventional magnets from VAC's mass production.

The results show that the magnetic properties B_R, H_{cJ}, H_{cB} and $(BH)_{max}$ are comparable between these two magnets. That means that strip cast REMA produced from recycled material by the REE4EU process has excellent properties to manufacture high-tech PM.



(A) (B)

Figure 11. (A) Magnetic properties of a magnet produced using raw material from the REE4EU process; (B) Magnetic properties of a conventional magnet from VAC's mass production

4 Conclusions

The output material from the new engineered REE4EU-pilot process has been validated by producing and characterising permanent magnets from the rare earth alloy obtained from the REE4EU's ILE-HTE pilot installed at ELKEM

VAC has made permanent magnets in his mass production line and has determined the quality of both the strip cast alloy input and the permanent magnet output in terms of magnetic properties, namely remanence, coercivity, intrinsic coercivity and maximum energy product, as well as chemical composition.

The results obtained showed that the permanent magnets prepared from the 600 kg strip cast rare earth alloy obtained with the REE4EU technology have the same properties as magnets from VAC's production line using virgin material.

That means that the REE4EU technology is suited for obtaining rare earth alloys for permanent magnet production using permanent magnet wastes, thus demonstrating a closed-loop recycling scheme for permanent magnets in Europe, using less steps than conventional state of art methods from China.

Acknowledgements

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 680507