Replacement and Original Magnet Engineering Options

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& ROMEO Consortium

Outline

- ROMEO in general
  - Goal 1
  - Goal 2
- Work at JSI - Goal 1
  - Electrophoretic deposition
  - Evaluation of the magnetic properties and microstructure
  - Chemical analyses and firmness test
  - Conclusions
- Collaboration with industrial partners
  - Combining the achievements at VAC and JSI
- Conclusions

Permanent magnets in the 21 century

2014 Toyota Camry Hybrid

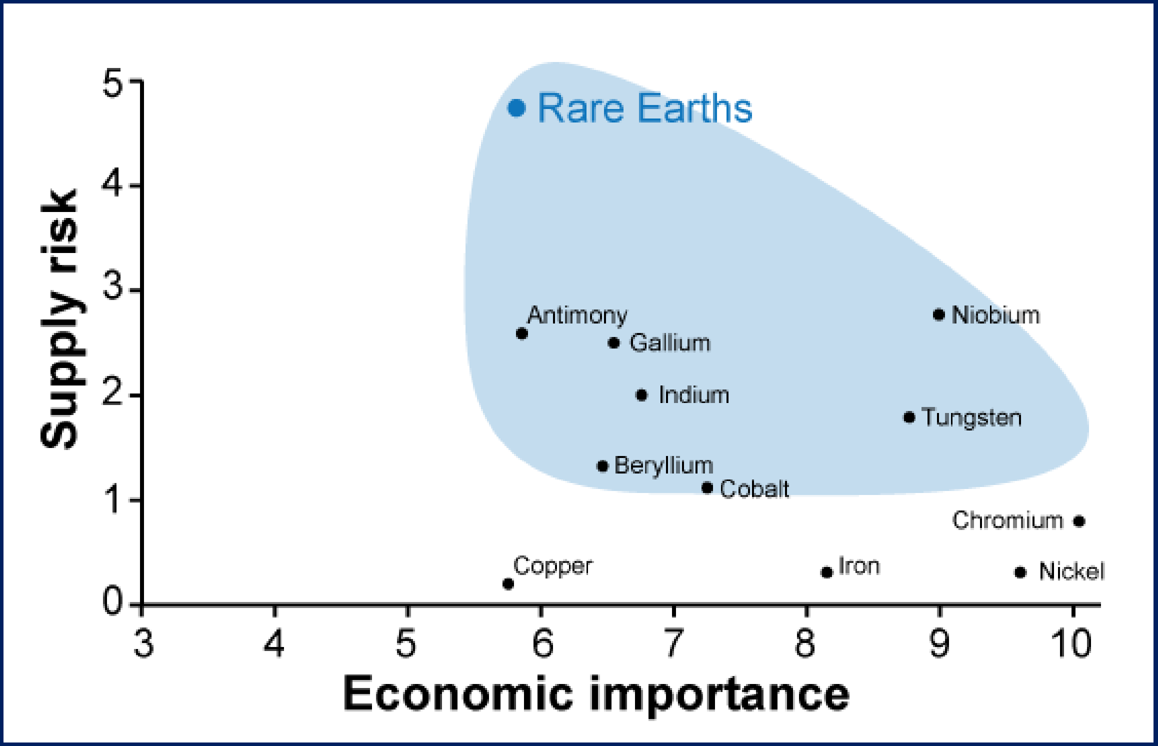
2014 Ford Fusion Hybrid

PM based on RE are increasingly important in **environmentally critical** technologies:

- for wind turbines
- hybrid and
- pure electric vehicles

(**HEVs and EVs**).

Supply risk and economic importance of 14 critical raw materials



Trans-Atlantic Workshop on Rare Earth Elements and Other Critical Materials for a Clean Energy Future, **Cambridge, Massachusetts**, **December 3, 2010**

Japan-EU workshop "Substitution of Critical Ray Materials" : ***Tokyo, Japan, 21-22 November 2011***

3rd Trilateral EU-US-Japan Conference on Critical Materials: towards New Models in Efficient Management of Critical Materials ***29-30 May 2013, Brussels*****. 2013**.

ROMEO Consortium

External Advisory Board

University of Nebraska

University of Birmingham

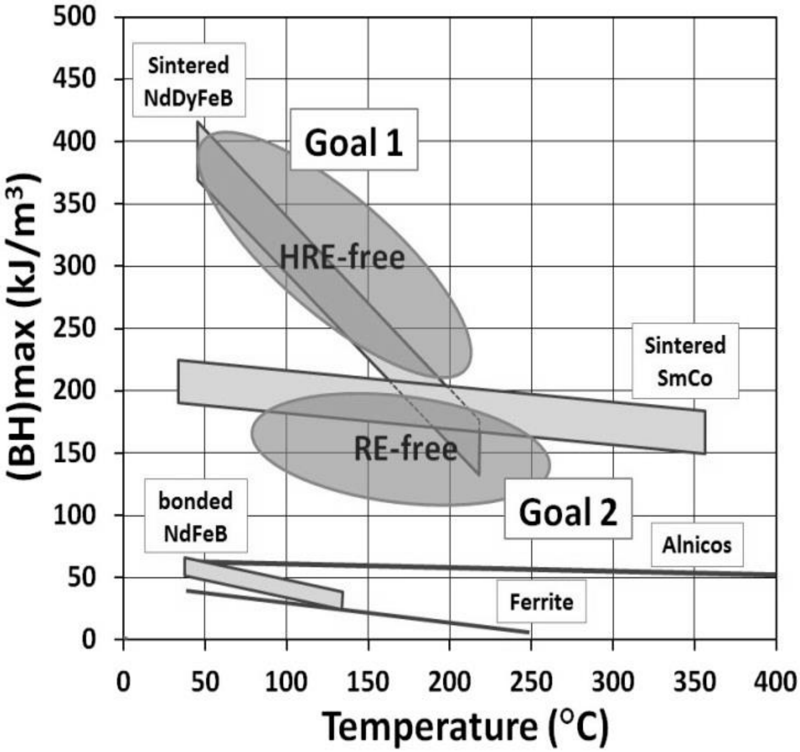
Intermetallic, Kyoto

Technology Metals Research LLC

ROMEO concept

ROMEO ideas were that we can progressively remove Rare Earth's from RE- Permanent Magnets:

1. Eliminate, or drastically reduce, HREs in high-T Nd-Fe-B magnets
2. Develop new RE-free magnets with properties intermediate between Nd-Fe-B and ferrite



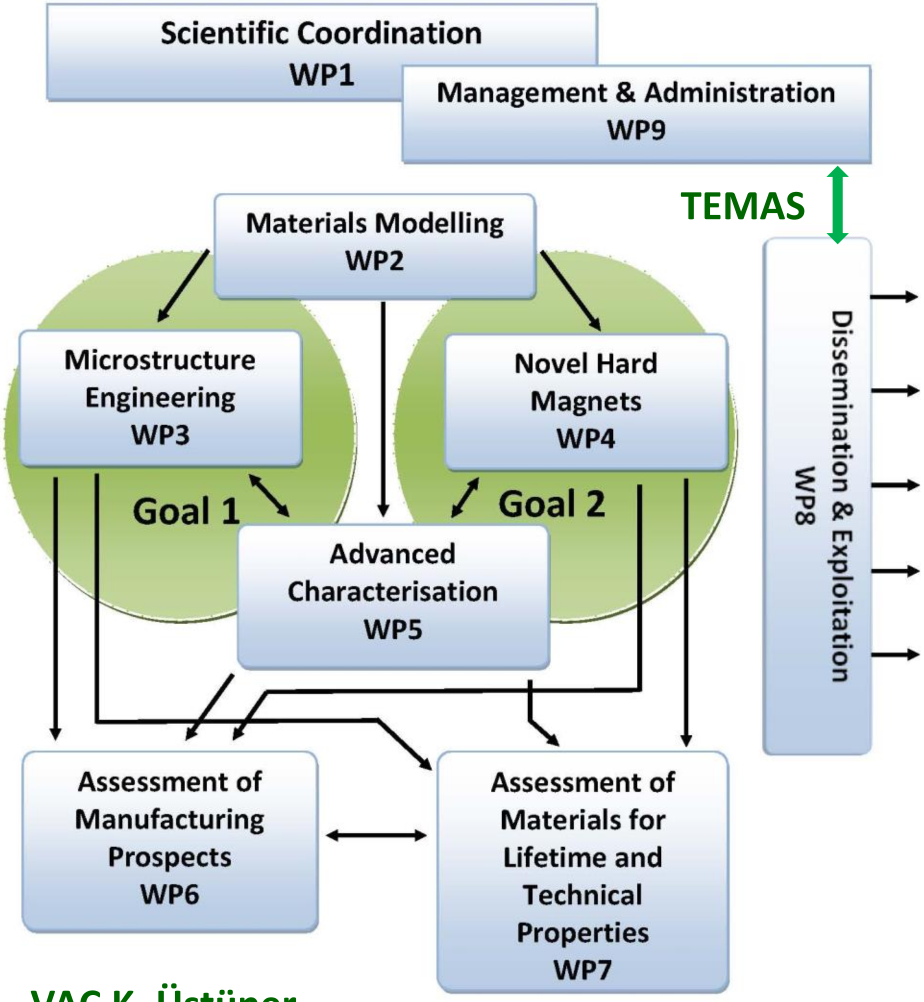
ROMEO project workflow

WP2 T. Schrefl

WP3 N. Dempsey

WP4 M. Coey

WP5 O. Gutfleisch



Scientific Coordination
WP1

Management & Administration
WP9

TEMAS

Materials Modelling
WP2

Microstructure Engineering
WP3

Novel Hard Magnets
WP4

Goal 1

Goal 2

Advanced Characterisation
WP5

Dissemination & Exploitation
WP8

Assessment of Manufacturing Prospects
WP6

Assessment of Materials for Lifetime and Technical Properties
WP7

VAC K. Üstüner

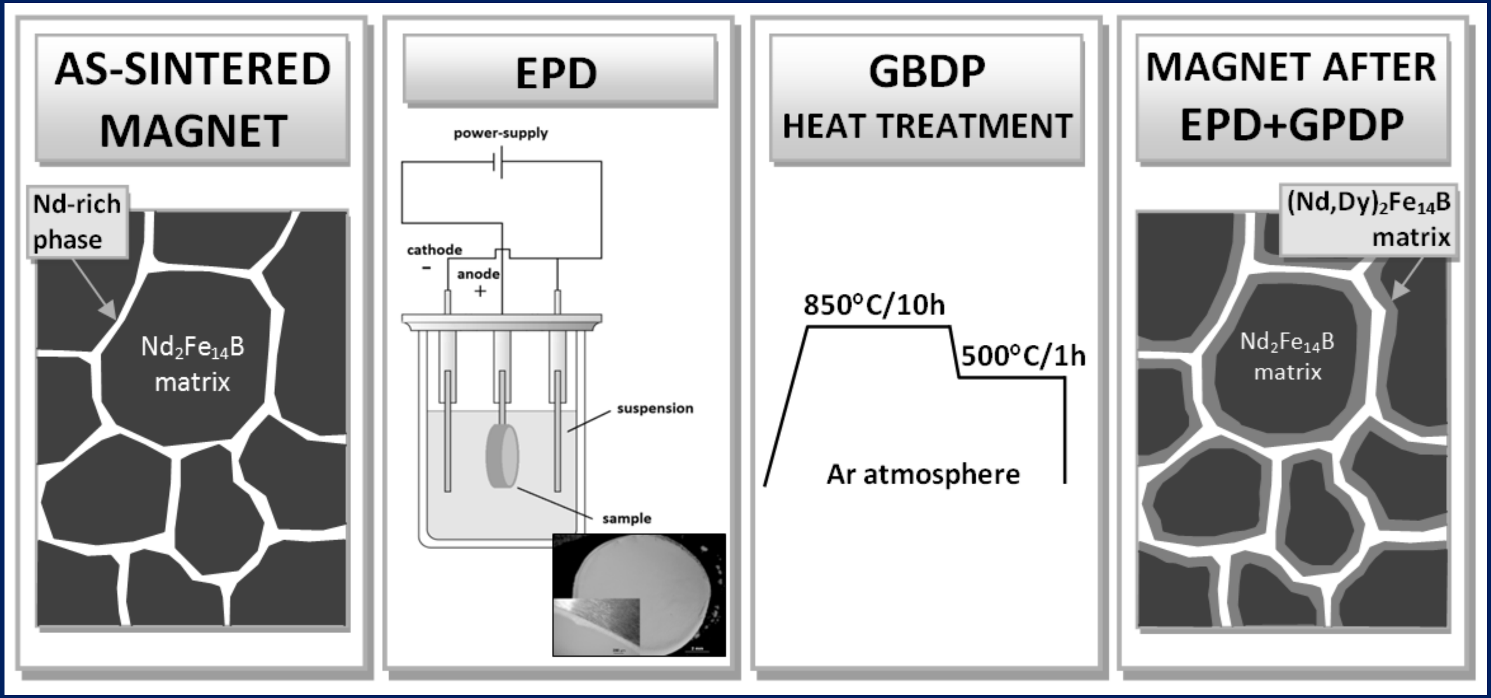
VALEO J.-M. Dubus

Goal 1: Eliminate, or drastically reduce, HREs in high-temperature Nd-Fe-B magnets

- 1-1:Grain-boundary diffusion of HRE
- 1-2: Extreme grain-size refinement
- 1-3: New grain-boundary phases

GOAL 1

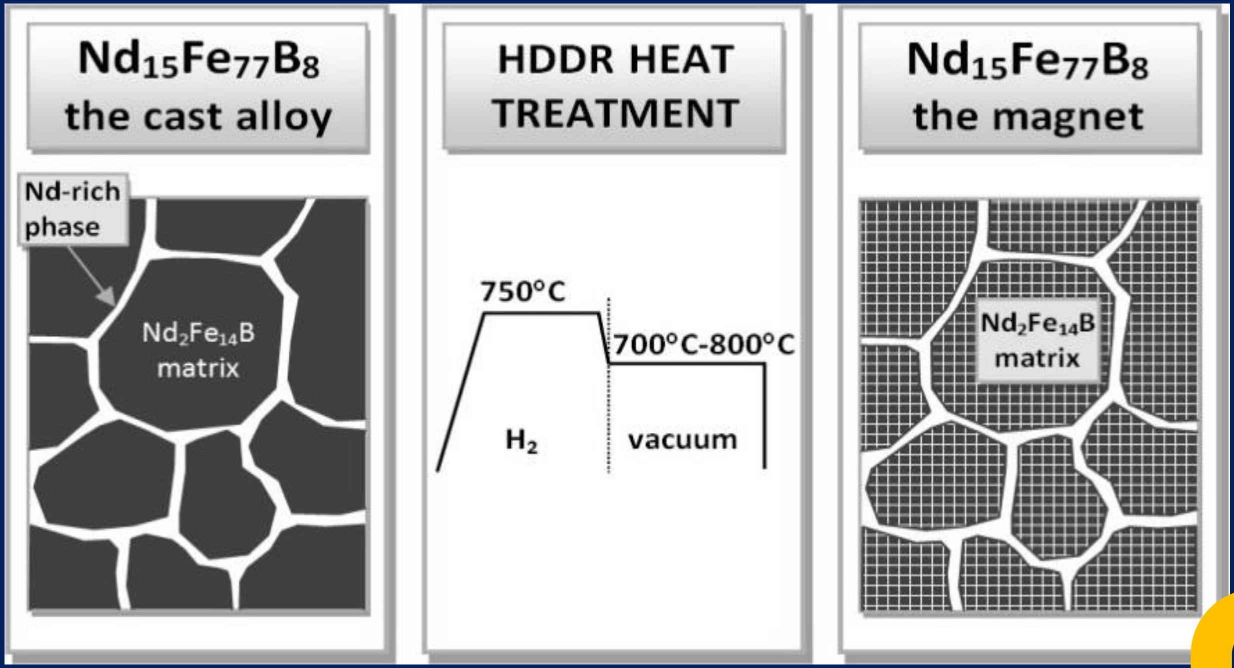
1-1: Grain-boundary diffusion of HRE

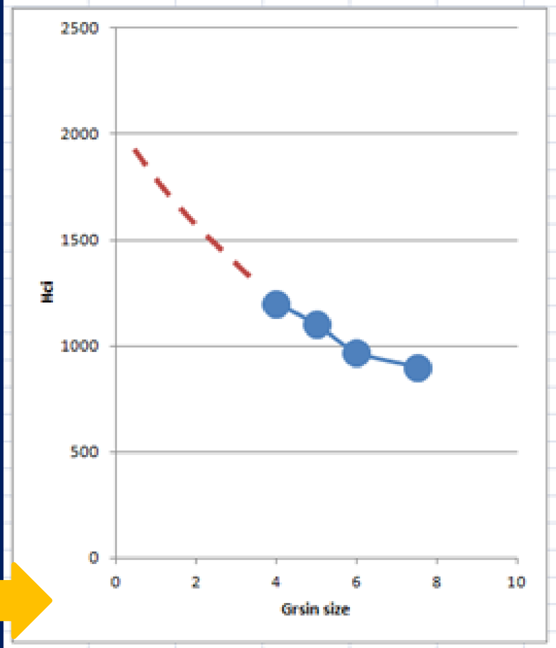


The task 1-1 of the Goal 1 was to develop high-coercivity magnet with drastically reduced amount of Dy or Tb while also providing a higher remanence B_r compared to currently available magnets.

The innovative method Electrophoretic Deposition(EPD) of HRE was used to minimize the amount of HRE needed to achieve a core-shell microstructure enabling high Hc and Br.

2-2: Extreme grain-size refinement





1-2 task of Goal 1 was to decrease the grain size to reach the size o
< 2 µm and confirm the prediction shown on the diagram

i) Schematic diagram showing how the **HDDR** process is able to transform the as-cast microstructure into a fine-grained, high-coercivity material.

ii) The other method to achieve the grain size of < 2 µm is **milling**, but is very challenging due to the oxidation problems .

Most promisingly, their grain size is only about one-tenth that of conventionally Nd sintered magnets, leading to a potential for coercivities that match those currently available only with HRE-containing magnets

GOAL 2

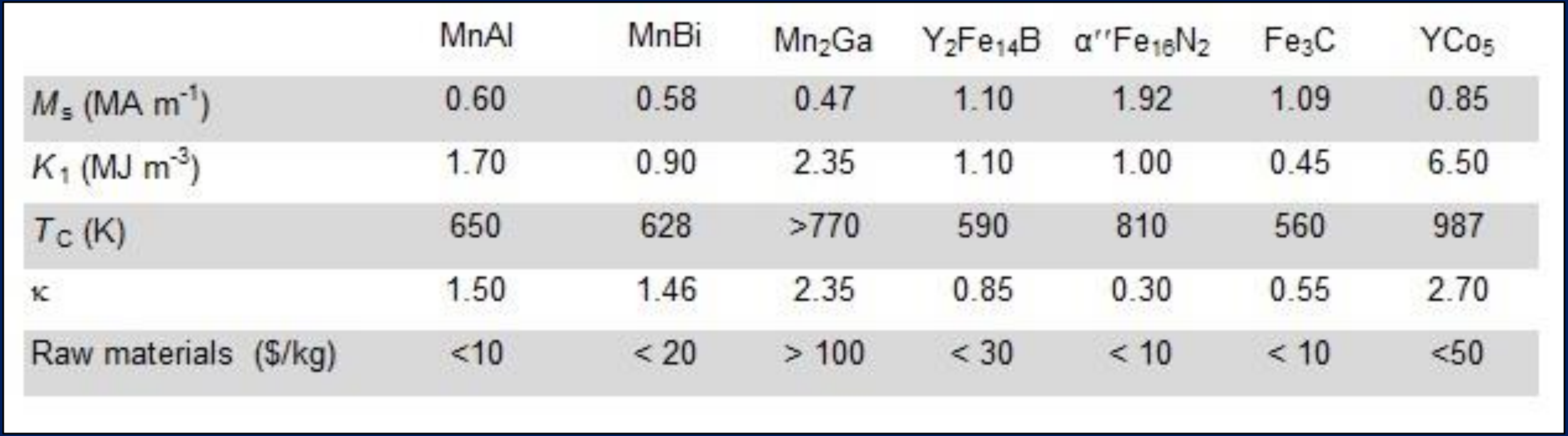
Goal 2: Develop new RE-free magnets with properties intermediate between Nd-Fe-B and ferrite

- 2-1: High-throughput computational materials discovery and candidate materials synthesis
- 2-2: Coercivity development in rare-earth-free materials by GB engineering

In total the database comprises **236,945** compounds

21 new Heusler alloys were found

High Throughput Computational Materials Discovery



	MnAl	MnBi	Mn_2Ga	$Y_2Fe_{14}B$	$\alpha''Fe_{16}N_2$	Fe_3C	YCo_5
M_s (MA m^{-1})	0.60	0.58	0.47	1.10	1.92	1.09	0.85
K_1 (MJ m^{-3})	1.70	0.90	2.35	1.10	1.00	0.45	6.50
T_C (K)	650	628	>770	590	810	560	987
κ	1.50	1.46	2.35	0.85	0.30	0.55	2.70
Raw materials ($/kg)	<10	< 20	> 100	< 30	< 10	< 10	<50

Properties of some uniaxial ferromagnets. κ is the magnetic hardness parameter, which should be > 1. This indicates the ease with which coercivity may be developed.

Develop RE-free Magnets with Properties Intermediate Between Nd-Fe-B and Ferrite

	$\lvert BH\rvert_{MAX}$ kJm^{-3}	T_C K	M_s MAm^{-1}	K_1 kJm^{-3}	Raw materials \$/kg
Ferrite	34	740	380	330	5
A	100	> 550	570	500	10
B	150	> 550	690	750	20
C	200	> 550	800	1000	30
Nd-Fe-B	350	590	1250	490	>100

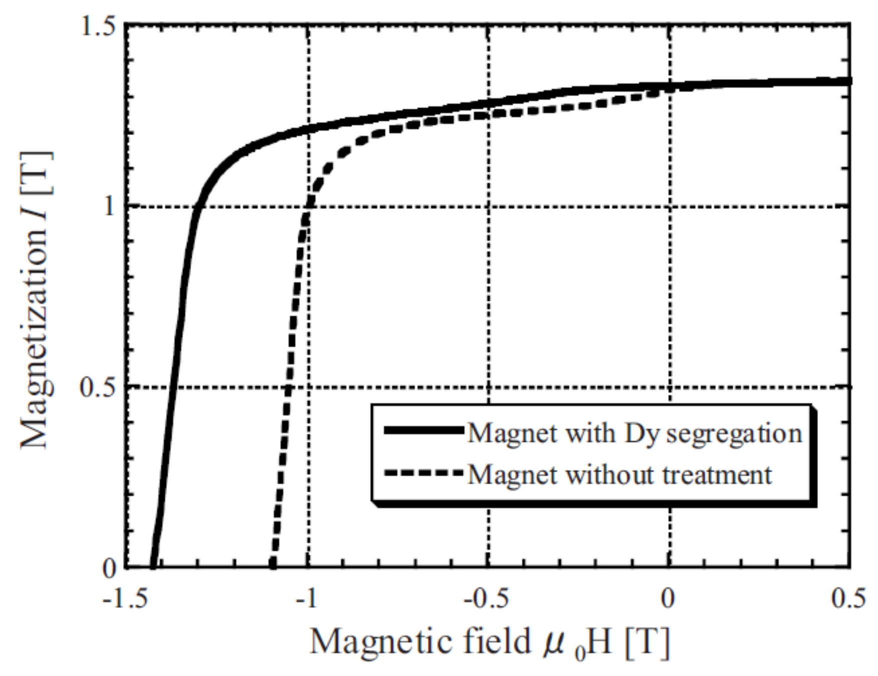
Some target specifications for new hard magnetic materials

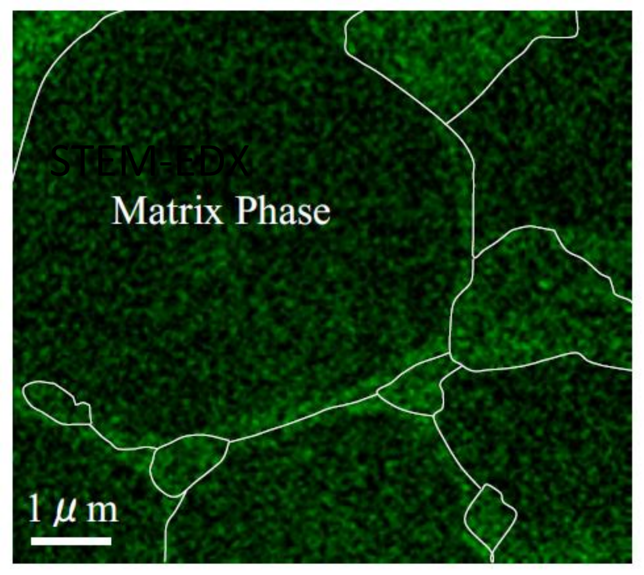
GOAL 1

at JSI

Grain-boundary diffusion process

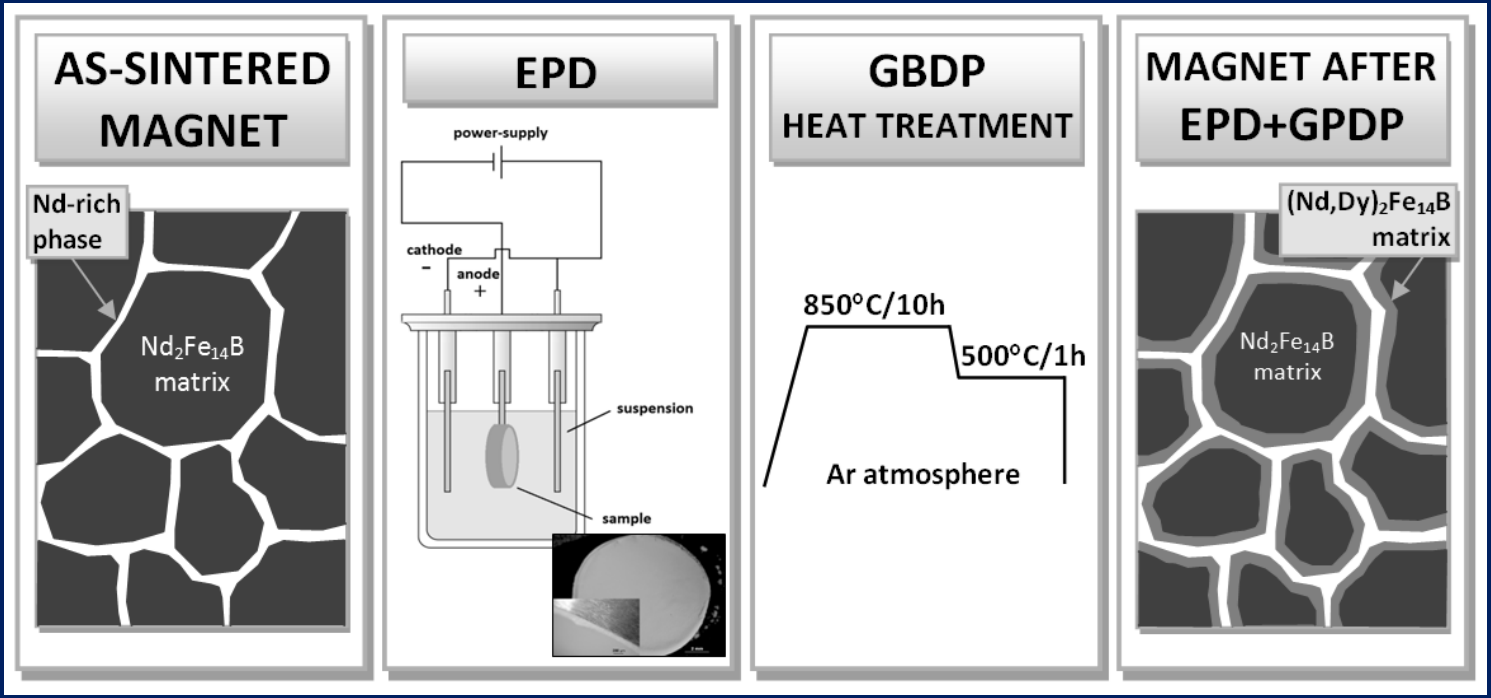
- use of Dy or Tb in a slurry
- immersing magnets in DyF_3 suspension





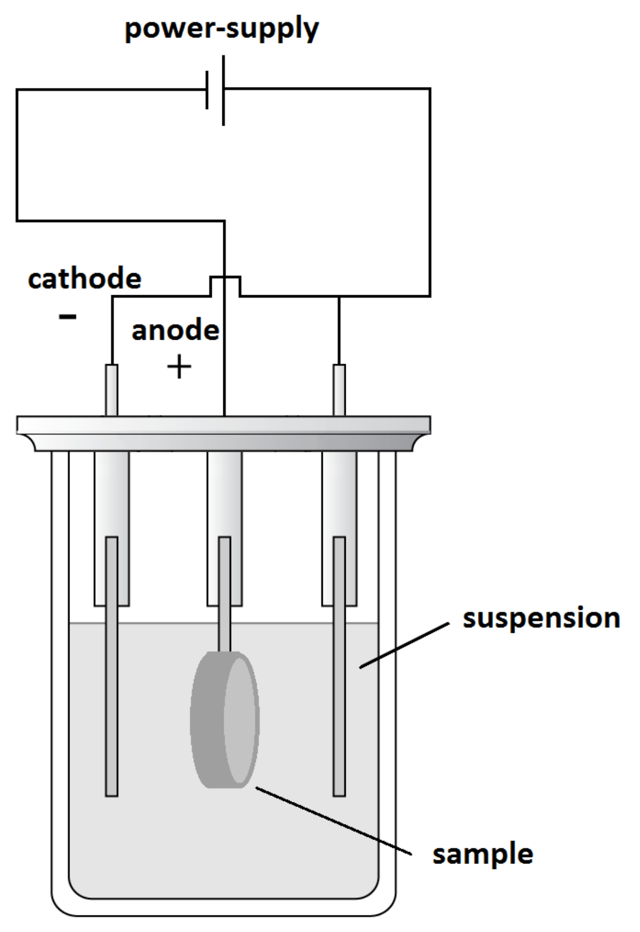
(Suzuki H. et al., 2009)

1-1: Grain-boundary diffusion of HRE



The innovative method Electrophoretic Deposition(EPD) of HRE (instead of dipping) was used to minimize the amount of HRE needed to achieve a core-shell microstructure enabling high Hc and Br.

Electrophoretic deposition



- Low costs
- Short deposition time
- Thickness control
- Even particle distribution

Voltage: 60 V

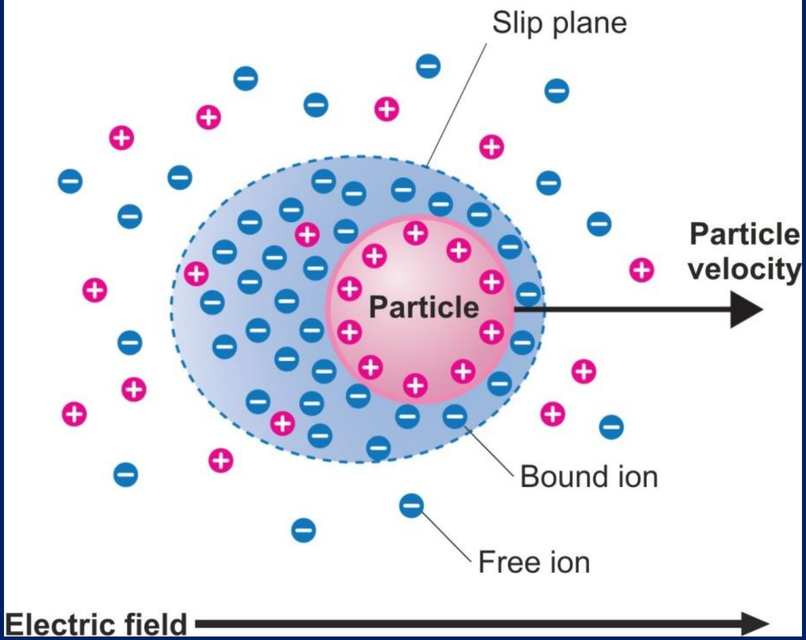
Time for 200 µm thick coating: 40 sec

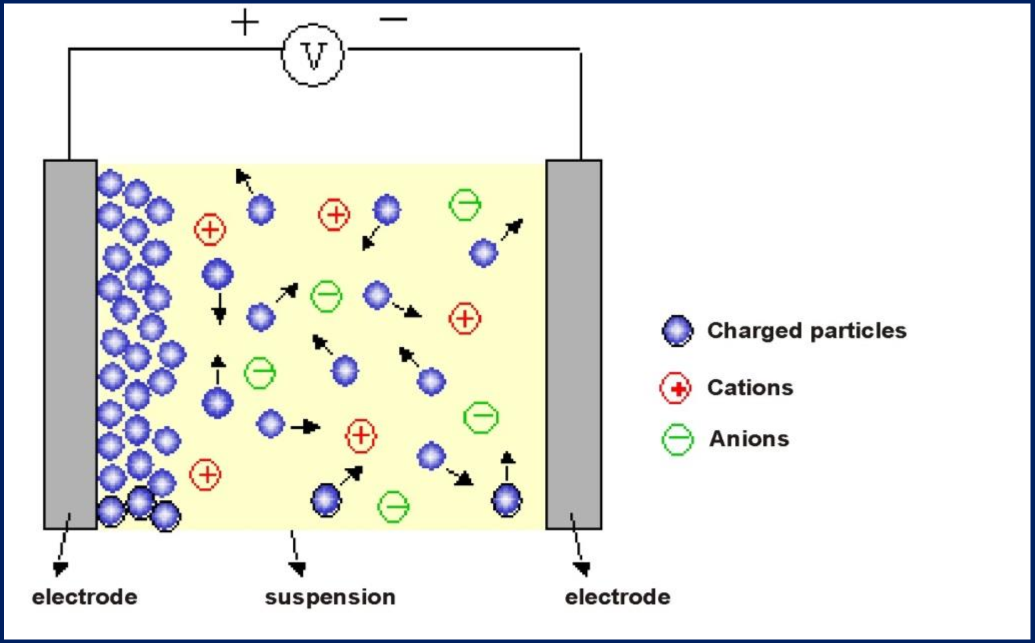
Particle size: 5 µm and 20 µm

Solvent: Ethanol

Particles: DyF_3

Electrophoretic Deposition





Schematically shown particle in movement towards cathode

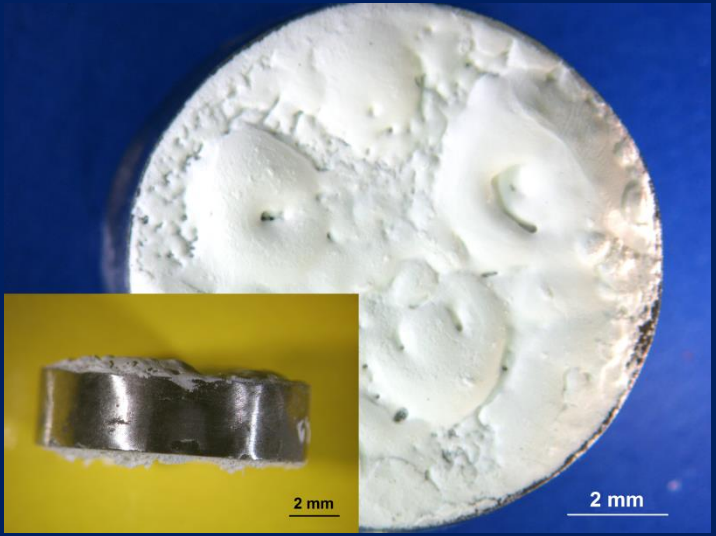
- Particles are moving towards electrode under electrical field
- Charge of particle is determining the way of movement

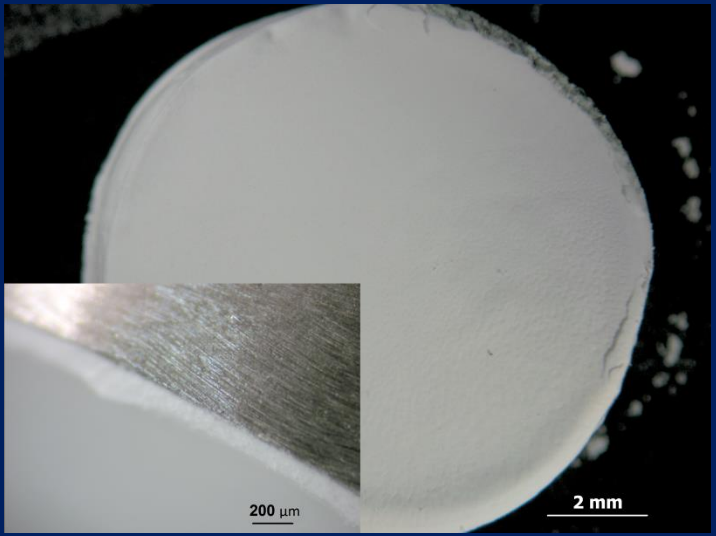
Starting magnets

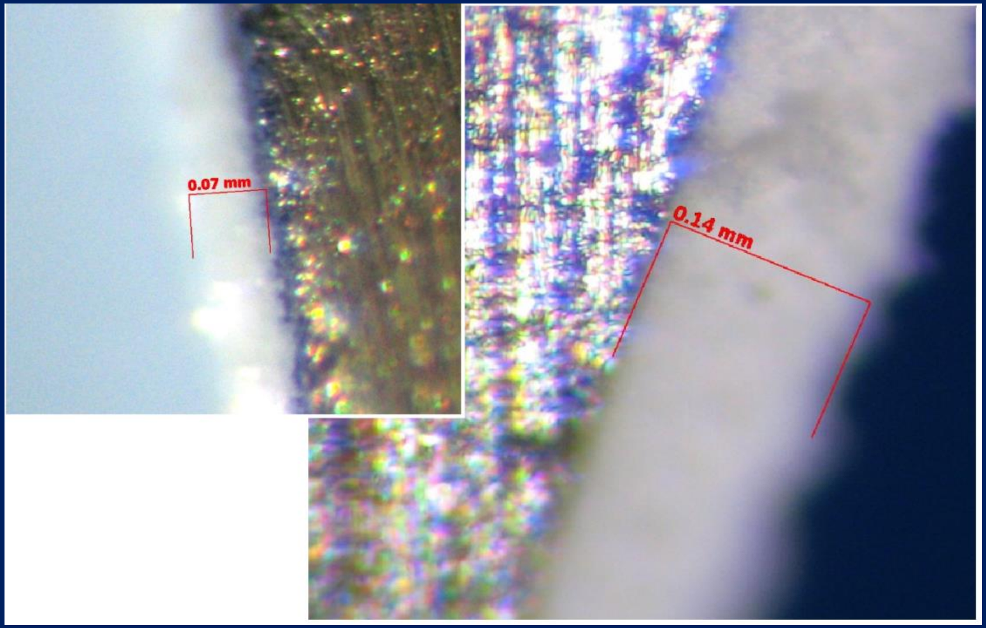
- Starting alloy was first crushed into a powder by jet-milling in a nitrogen atmosphere to ≤5 µm
- Powder was aligned in an external magnetic field of 1500 kA/m and pressed in a parallel-configured press
- Sintering temperature was 1010°C/2h in Ar
- Composition is $Nd_{14,25}Pr_{0,29}Fe_{75,66}Co_{3,39}Ga_{0,21}Al_{0,37}Cu_{0,15}B_{5,68}$
- Different sizes and shapes of magnets can be used in the EPD process



Dipping vs. EPD

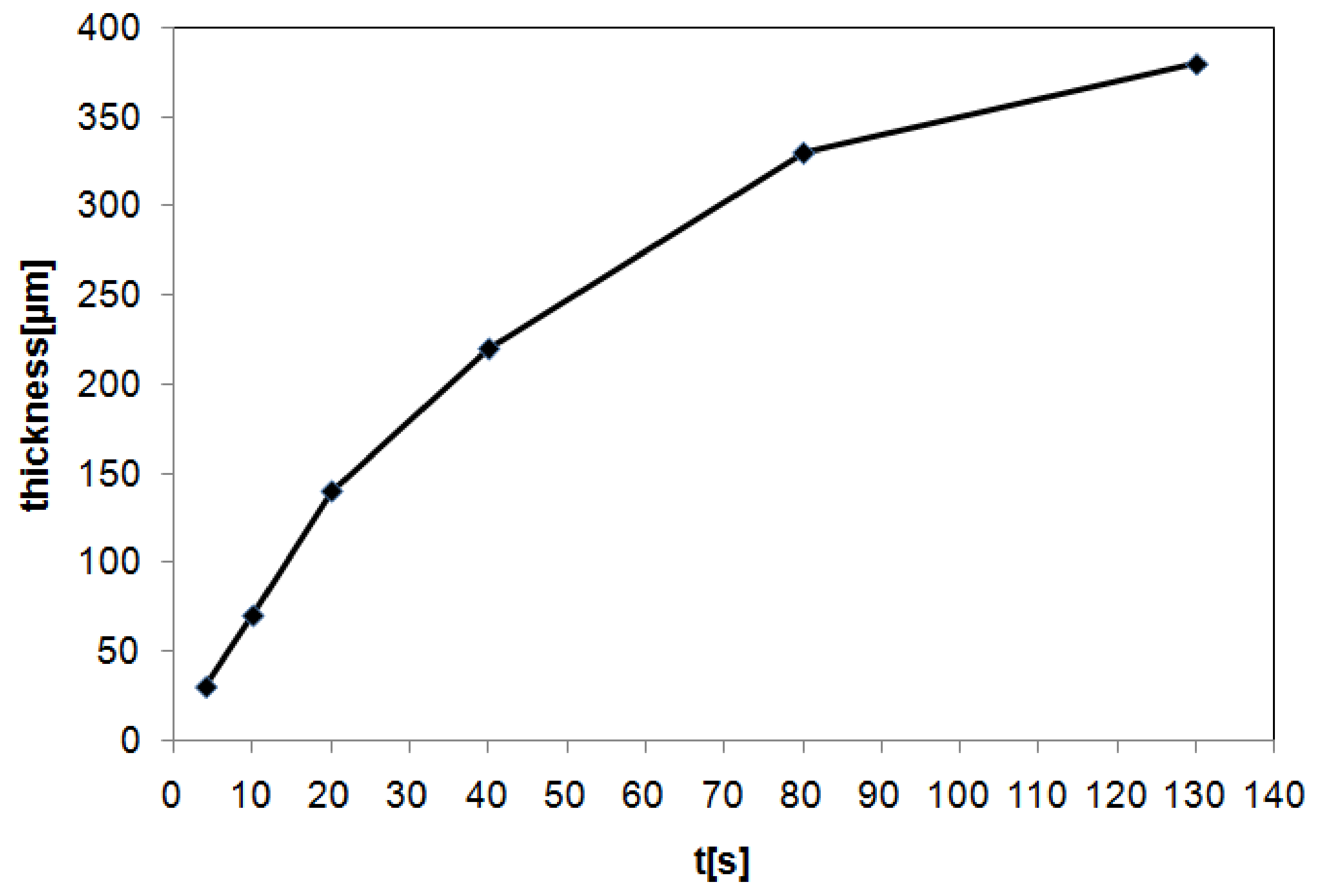






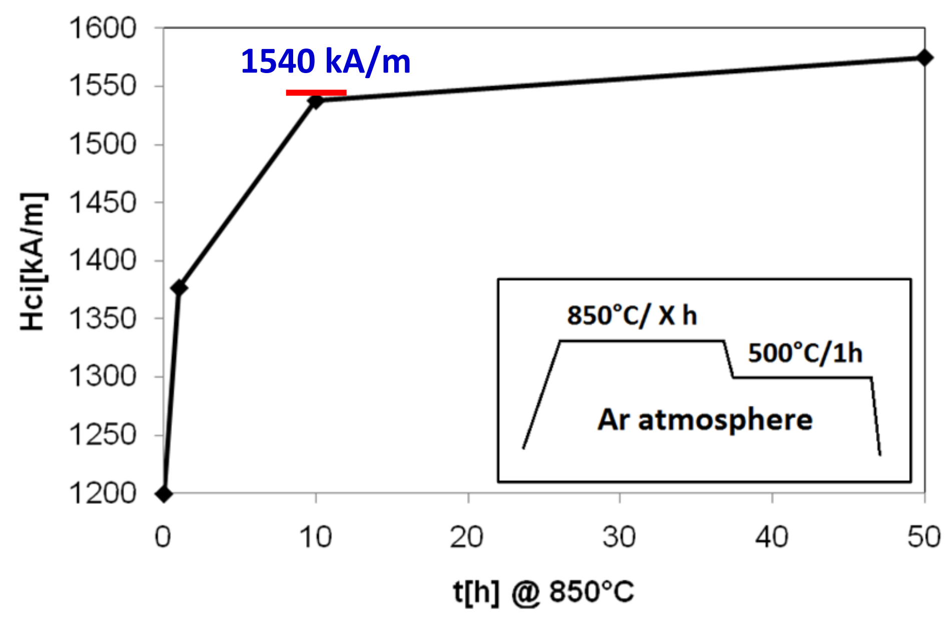
DyF$_3$ thickness after 10 s and 20 s

EPD coating vs time



Thickness of the EPD DyF_3 coating vs. time.

Coercivities of EPD vs. dipping

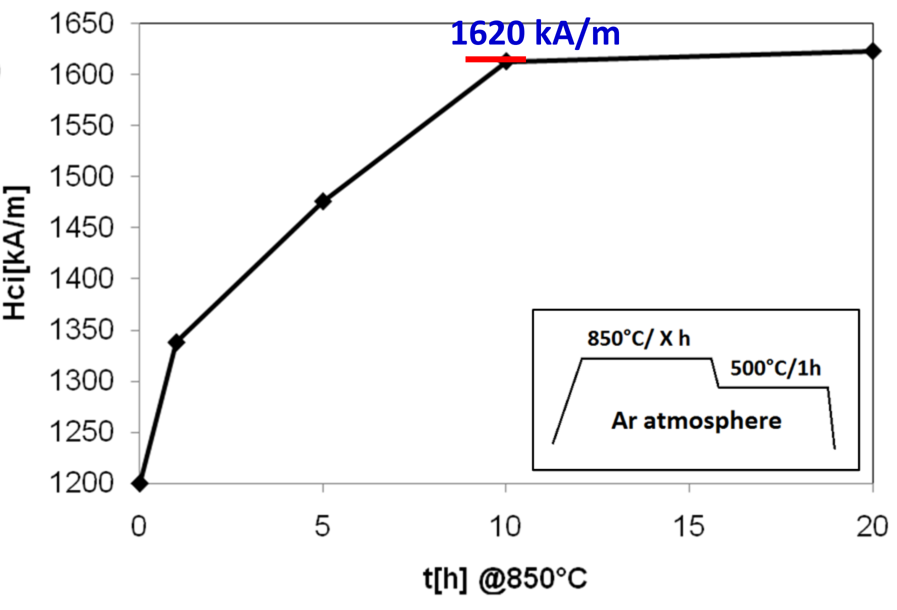


Dipping

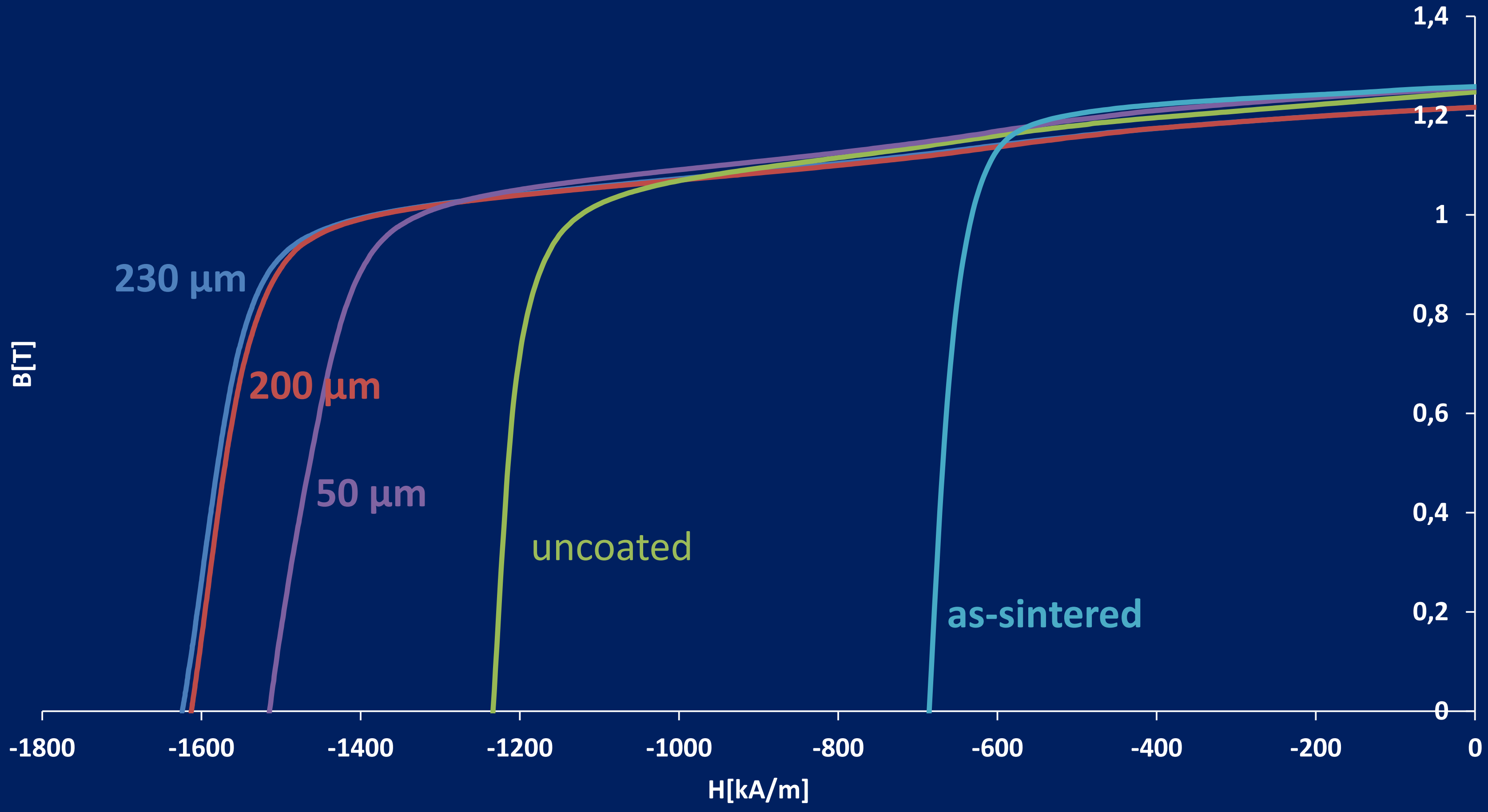
Coercivities of DyF_3 dipped samples that were heat treated under the regime shown in the inset. The time on the x-axis is the time of exposure at 850°C.

EPD

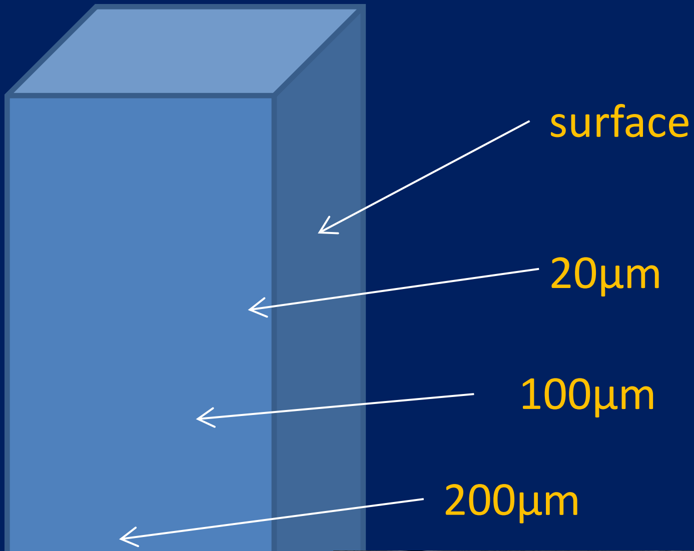
Coercivities of EPD deposited samples that were heat treated under the regime shown in the inset. The time on the x-axis is the time of exposure at 850°C.



Thickness effect on coercivity



Microstructure

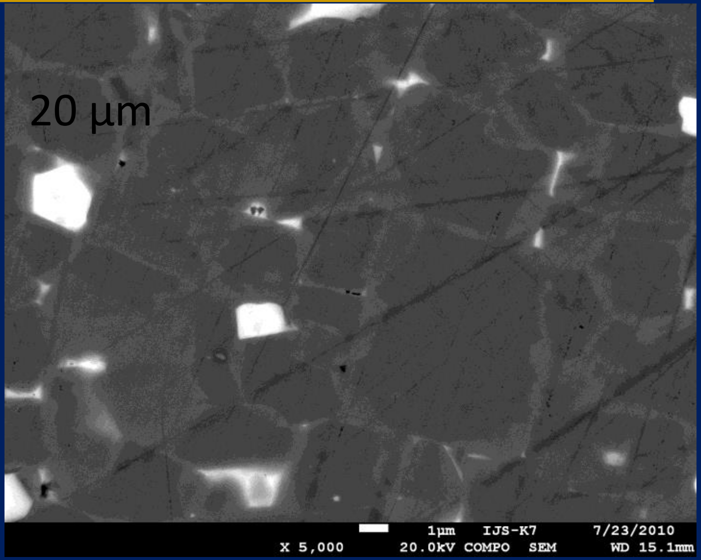


surface

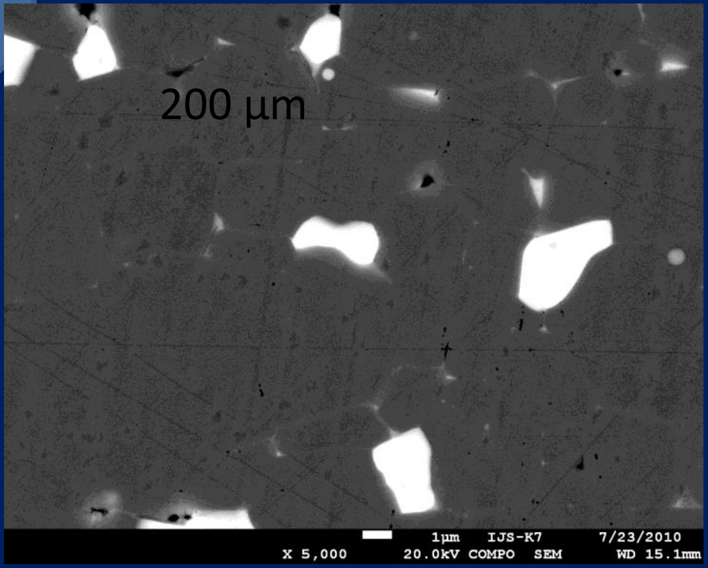
20µm

100µm

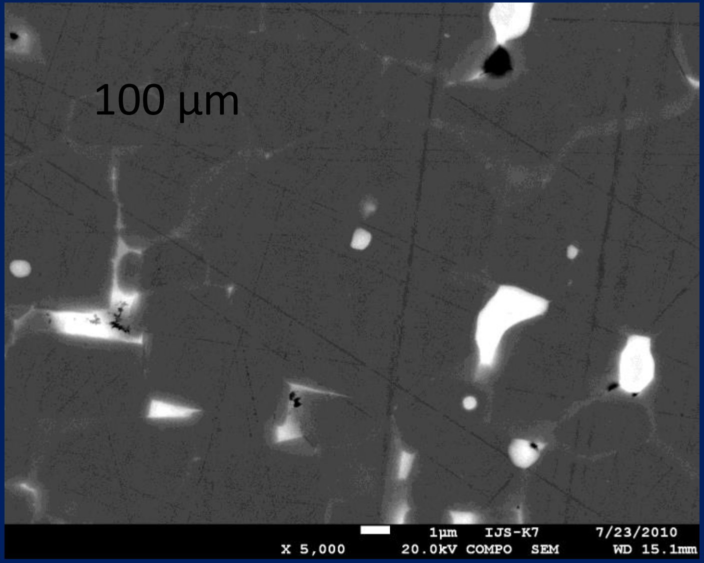
200µm



20 µm

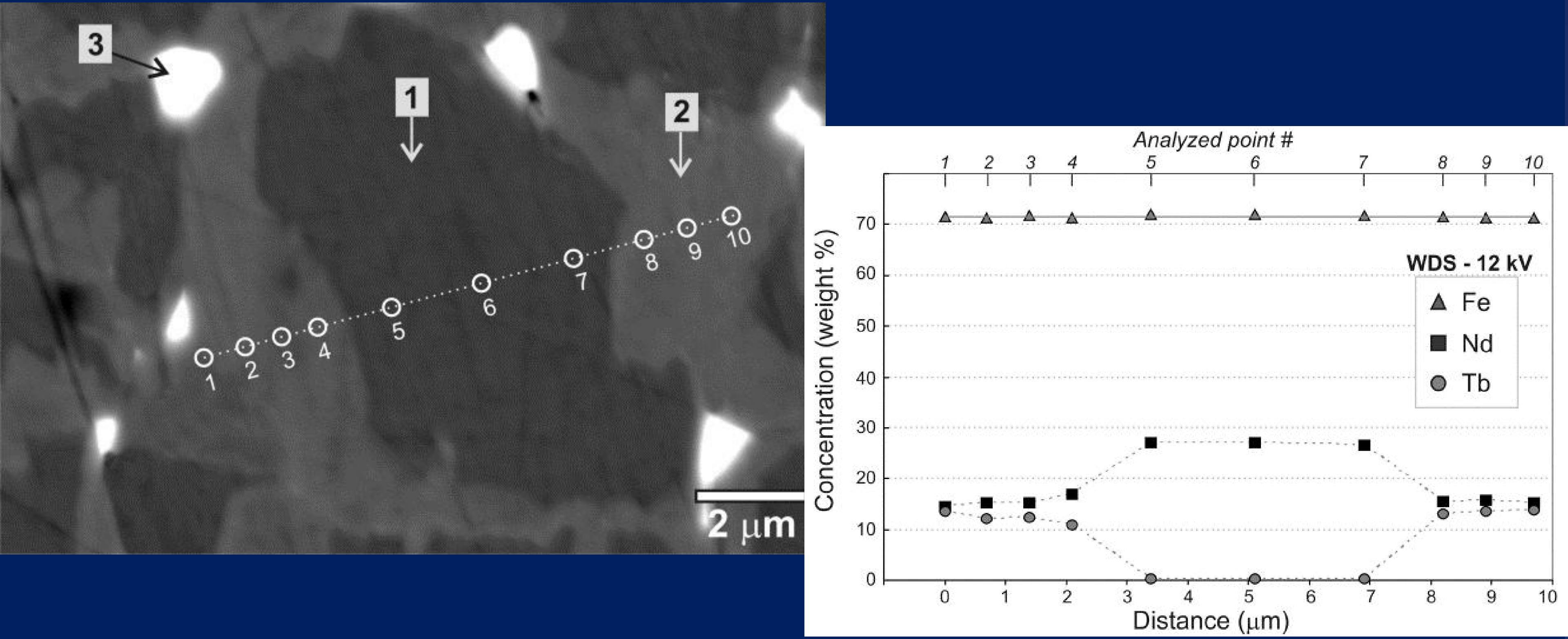


200 µm



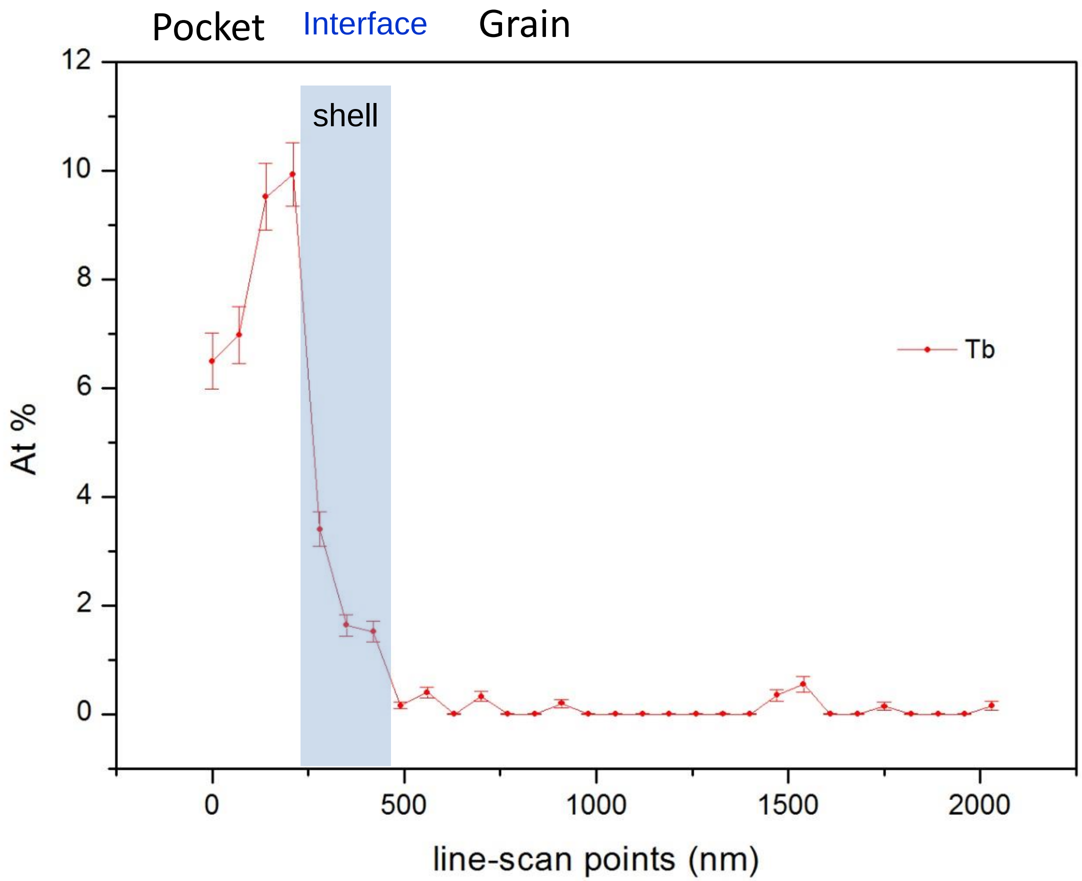
100 µm

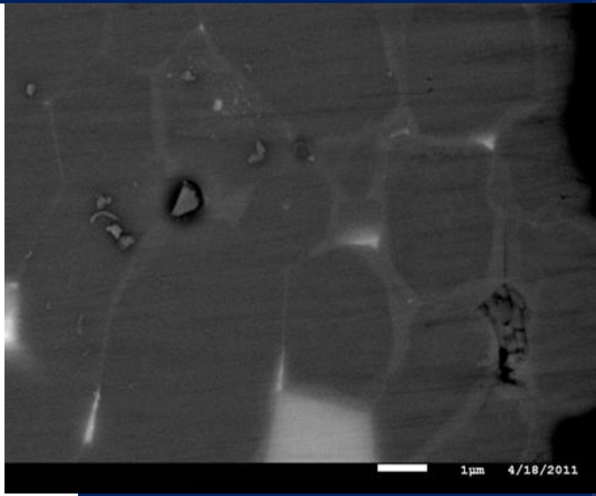
WDS point analysis

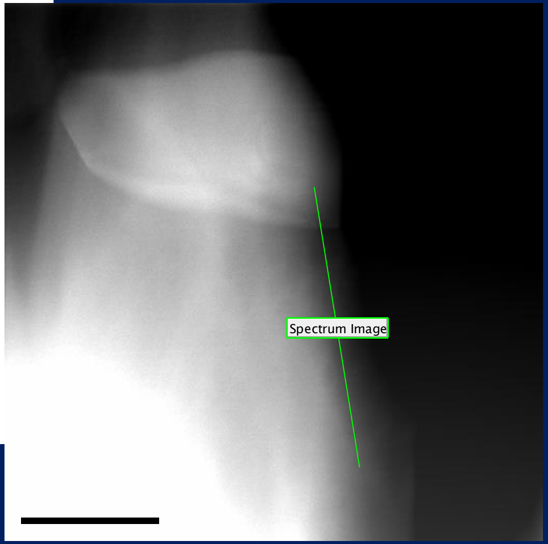


at. %	1	2	3	4	5	6	7	8	9	10
Fe	87.35	87.59	87.45	87.30	87.24	87.19	87.37	87.17	86.74	86.83
Tb	5.82	5.22	5.32	4.67	0.08	0.09	0.07	5.59	5.83	5.95
Nd	6.83	7.19	7.24	8.03	12.68	12.72	12.56	7.25	7.43	7.23

Composition – 150 µm away from the surface







- Diffusion of Tb into the grain
- Approx. 200 nm

Chemical analyses

-Chemical analyses were done using Inductive Coupled Plasma Atomic Emission Spectrometer (ICP-AES)

-Model Thermo Jarrell Ash, Atomscan 25

-DyF_3-treated and non-treated samples were first diluted in acid and then analyzed with the spectrometer

	Non-treated sample	**DyF_3-treated sample**
Al	0.60	0.60
B	0.99	0.98
Co	1.92	1.92
Dy	0.07	0.67 **(~0.22 at.%)**
Fe	63.44	63.38
Ga	0.15	0.16
Nd	23.74	23.98

Results are in [m%/m]

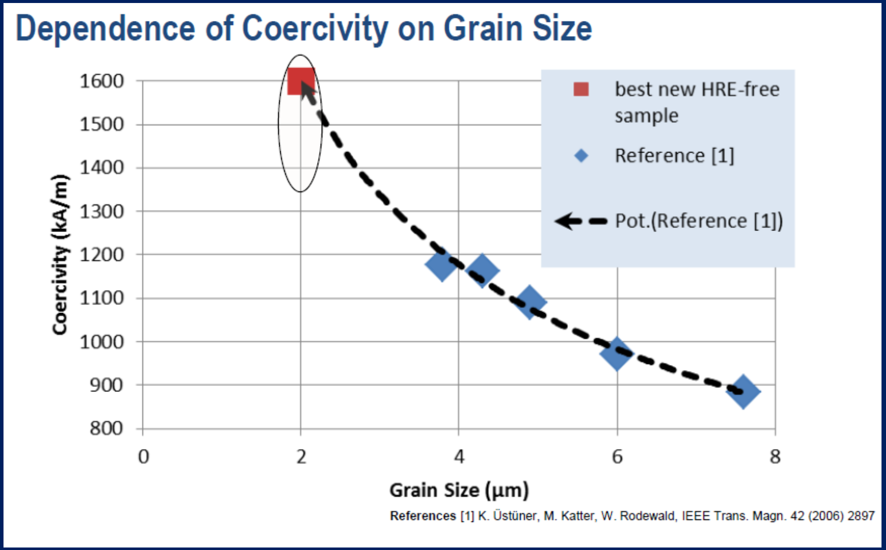
CONCLUSIONS

- A **novel EPD** technique was introduced to deposit DyF_3 as a precursor to the grain-boundary diffusion process (GBDP).
- **Higher coercivities (30 %)** were achieved and **the amount of heavy rare earth** was optimized **(0.2 at %)**.
- We believe that **EPD has the potential for application in an industrial environment** as part of the production process for Nd-Fe-B-based GBDP magnets.

GOAL 1

- Collaboration with industrial partners

Combining the VAC and JSI results

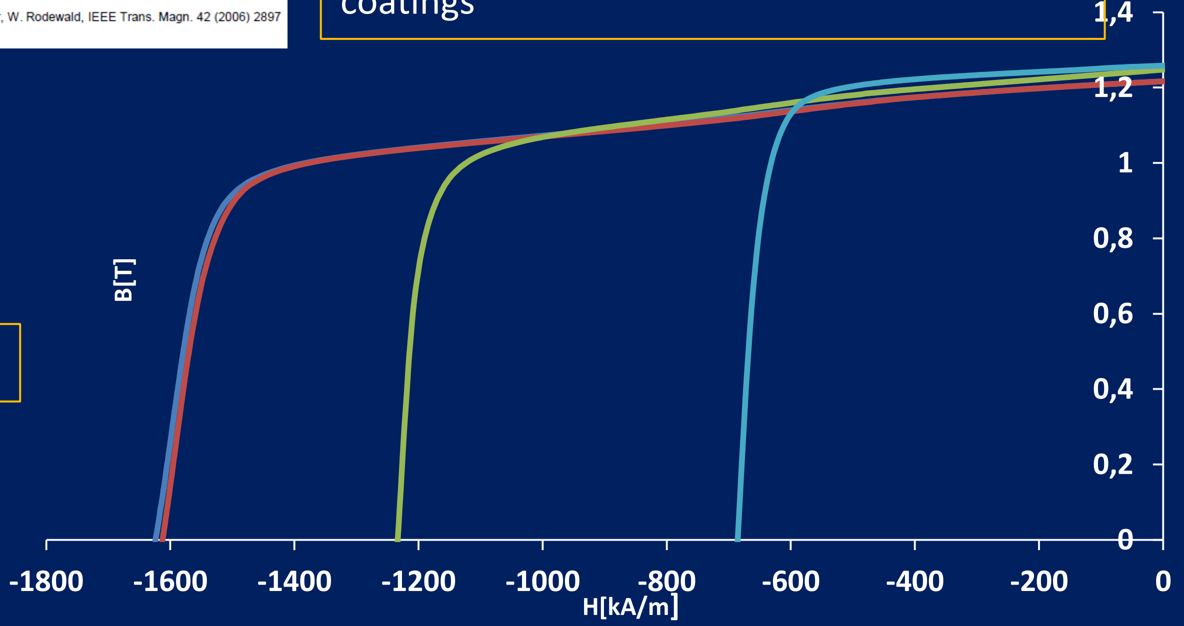


Courtesy of K. Üstüner, M. Katter

On VAC samples:

- Optimizing the thermal treatment.
- Checking the firmness of the DyF_3 layer (finding the method to measure it and make more detailed SEM analysis of the layer between the coating and the sample).
- Appling other coatings: Tb/Dy in a metal form
- Optimizing the processing with other coatings

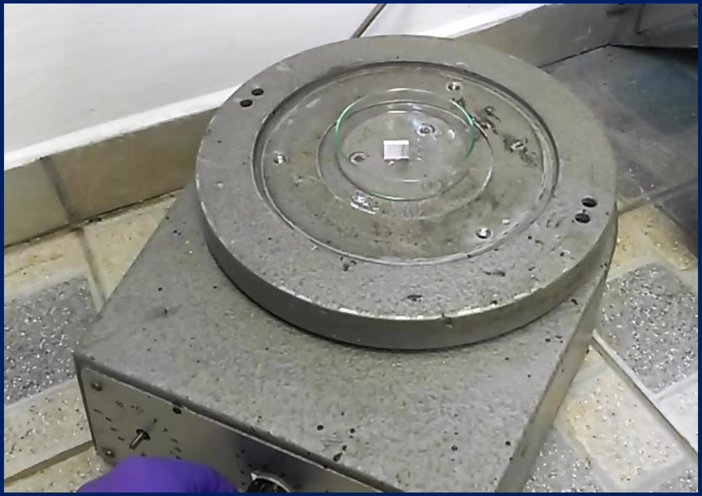
Milestones 1 and 2 achieved



Firmness of DyF_3-coating

The firmness of DyF_3-coating on NdFeB magnets was checked.
-$m(DyF_3)_1$=0.0403 g (standard mass of the coating for the results reported)

After vibrating for 20 seconds on level 5, the mass of DyF_3 coating was reduced to 0.0355 g.



Before vibrating test	After vibrating test	Δ m
0.0403 g	0.0355 g	12 %

DAIMLER & JSI

Together with DAIMLER JSI will use “electrophoretic deposition treated” magnets with different diffusion parameters.

We will study:

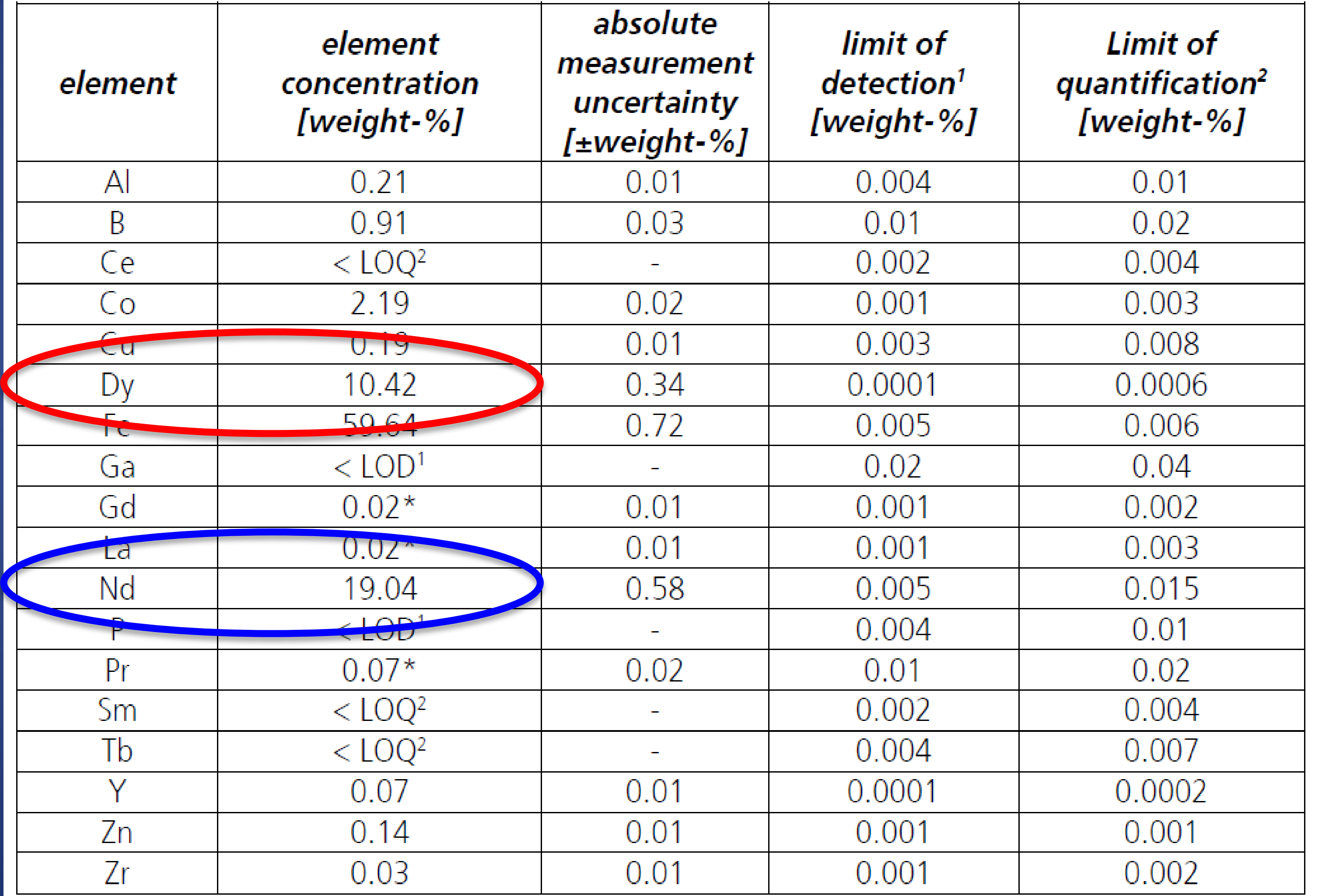
- a variation in heat treatment parameters (time and temperature)
- different coatings (Tb and Dy)
- different coating thicknesses

DAIMLER will test these samples (whole magnet) with METIS pulsed tracer.
We will evaluate the Hci-distribution in the magnet.
DAIMLER will furthermore test the corrosion resistance of these grades with HAST system.
In the end DAIMLER will compare the results with results from commercial magnets.

Conclusions

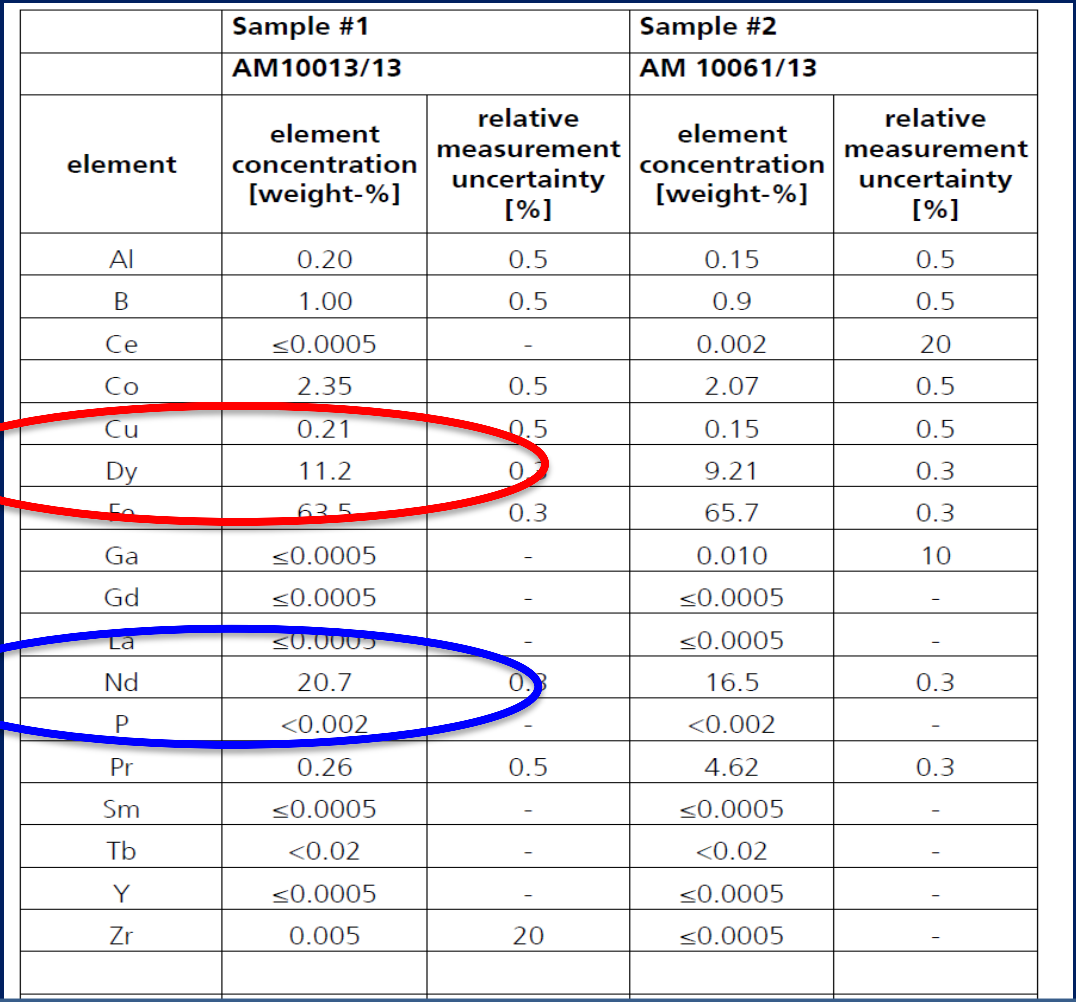
- ROMEO is structured to integrate industrial partners fully into the project, and to co-develop customised and tested solutions for prototype testing and final product development.
- ROMEO will strengthen European industry and make it more globally competitive by enabling it with superior technology.
- ROMEO will enable a drastic reduction in the industrial use of RE and especially HRE materials for the production of permanent magnets.
- Exploitation of ROMEO’s results will take place in two phases: *idea to prototype,* and *prototype to market.*
- ROMEO educates **16 new young scientists**: 13 PhD students a 3 post docs

How much heavy rare earth in e-motors?



element	*element concentration [weight-%]*	*absolute measurement uncertainty [±weight-%]*	*limit of detection[1] [weight-%]*	*Limit of quantification[2] [weight-%]*
Al	0.21	0.01	0.004	0.01
B	0.91	0.03	0.01	0.02
Ce	< LOQ[2]	-	0.002	0.004
Co	2.19	0.02	0.001	0.003
Cu	0.19	0.01	0.003	0.008
Dy	10.42	0.34	0.0001	0.0006
Fe	59.64	0.72	0.005	0.006
Ga	< LOD[1]	-	0.02	0.04
Gd	0.02*	0.01	0.001	0.002
La	0.02*	0.01	0.001	0.003
Nd	19.04	0.58	0.005	0.015
P	< LOD[1]	-	0.004	0.01
Pr	0.07*	0.02	0.01	0.02
Sm	< LOQ[2]	-	0.002	0.004
Tb	< LOQ[2]	-	0.004	0.007
Y	0.07	0.01	0.0001	0.0002
Zn	0.14	0.01	0.001	0.001
Zr	0.03	0.01	0.001	0.002

How much heavy rare earth in e-motors?



	Sample #1		**Sample #2**	
	AM10013/13		**AM 10061/13**	
element	**element concentration [weight-%]**	**relative measurement uncertainty [%]**	**element concentration [weight-%]**	**relative measurement uncertainty [%]**
Al	0.20	0.5	0.15	0.5
B	1.00	0.5	0.9	0.5
Ce	≤0.0005	-	0.002	20
Co	2.35	0.5	2.07	0.5
Cu	0.21	0.5	0.15	0.5
Dy	11.2	0.3	9.21	0.3
Fe	63.5	0.3	65.7	0.3
Ga	≤0.0005	-	0.010	10
Gd	≤0.0005	-	≤0.0005	-
La	≤0.0005	-	≤0.0005	-
Nd	20.7	0.3	16.5	0.3
P	<0.002	-	<0.002	-
Pr	0.26	0.5	4.62	0.3
Sm	≤0.0005	-	≤0.0005	-
Tb	<0.02	-	<0.02	-
Y	≤0.0005	-	≤0.0005	-
Zr	0.005	20	≤0.0005	-

Goal 1 achieved

- The Goal 1 of the project is to achieve ambitious magnetic properties; The coercivity of 2000 kA/m or more and the remanence of at least 1.3 T at room-temperature.

- We achieved: **2027 kA/m and 1.31 T.**
  (coating was performed at JSI, GBDP at VAC)


18/12/2012
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The Winds of Change

Thank you for your attention!

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