[O16-2]

Accelerated development of thermally stable Nd-Fe-B magnets with light rare-earth elements and their synergetic effects to temperature stability with Co

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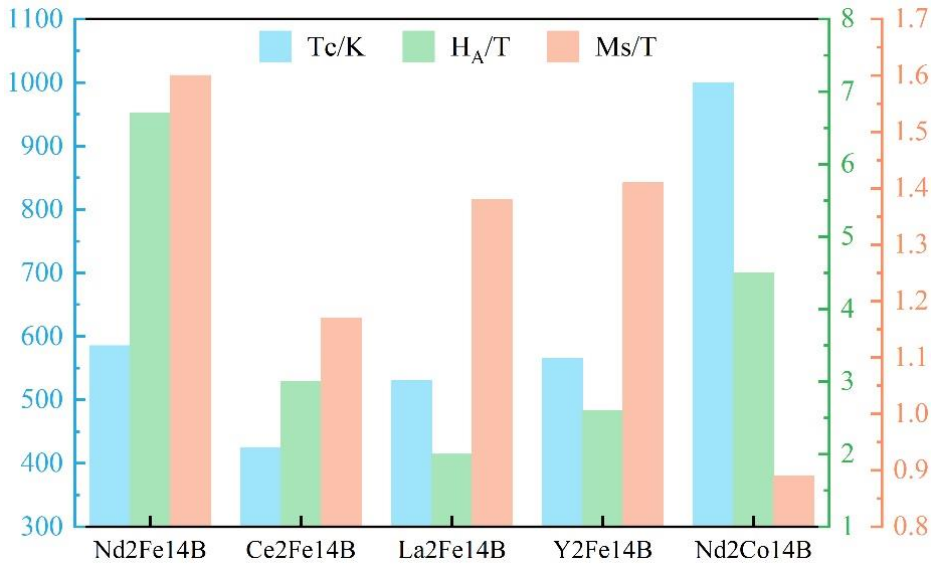
Background

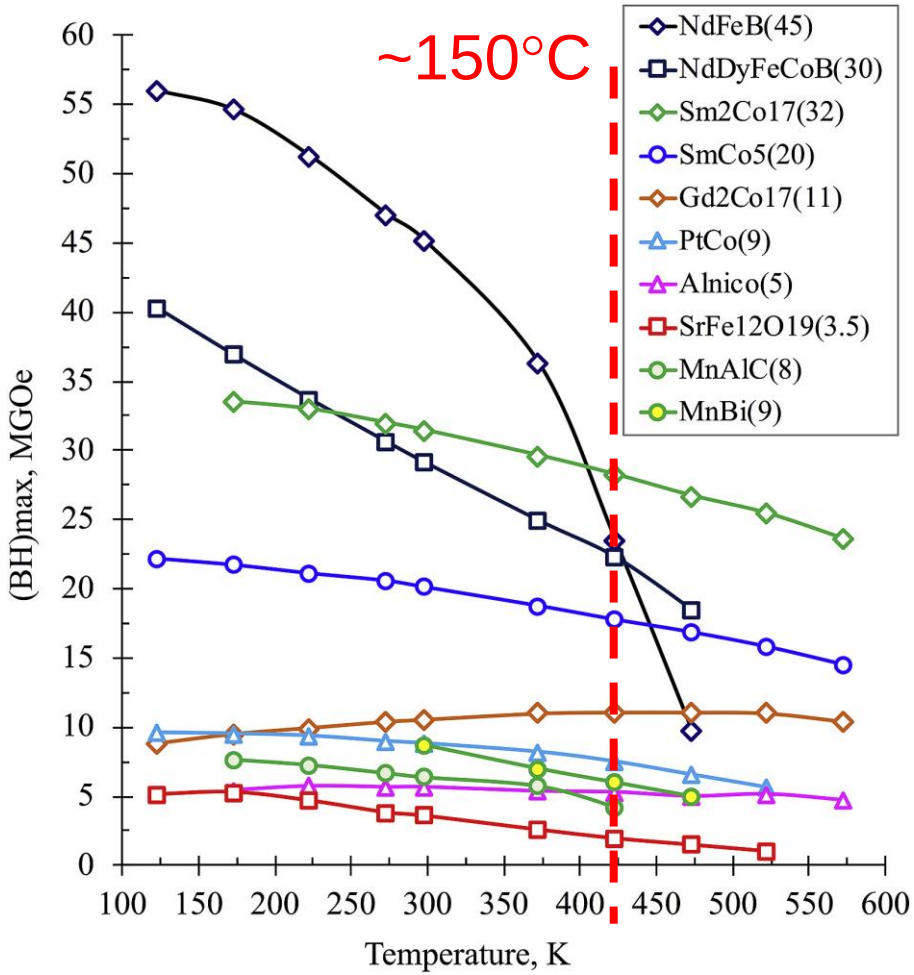
HEV/EV

Wind turbine

Robot

Perform/cost is an everlasting topic in engineering.

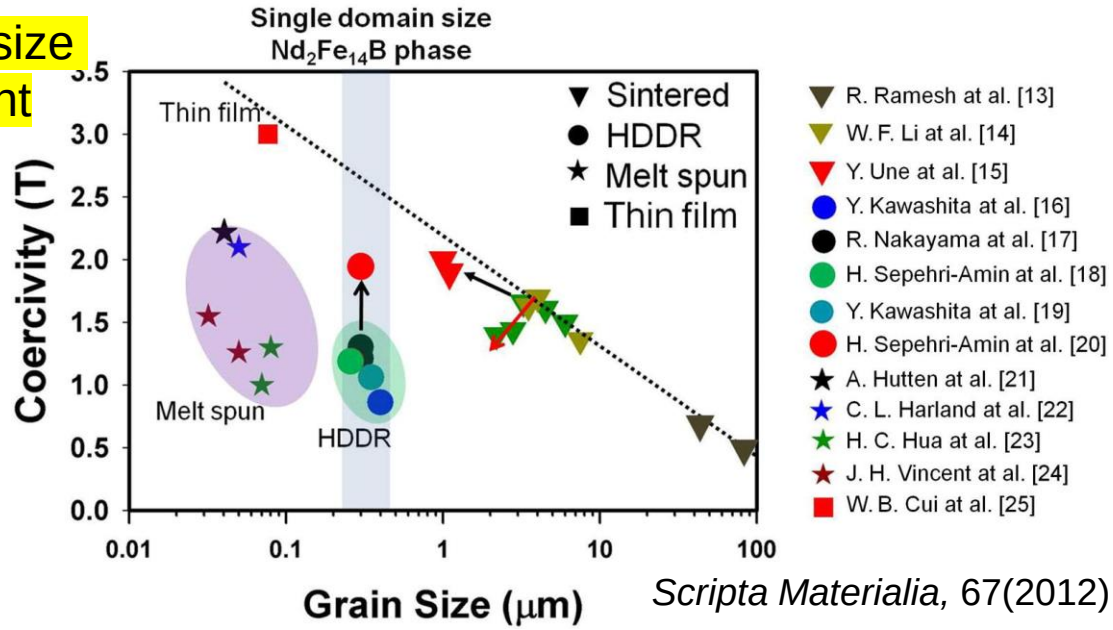




Acta Materialia, 158 (2018) 118-137, https://doi.org/10.1016/j.actamat.2018.07.049

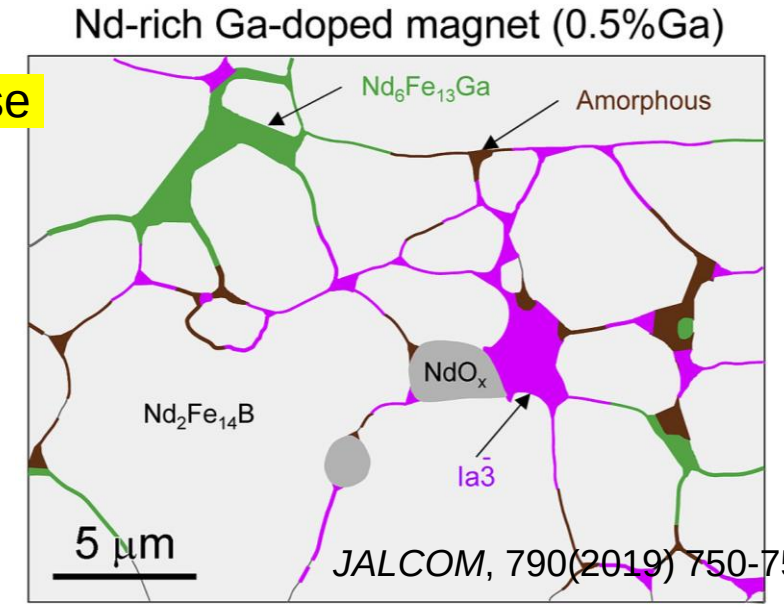
Attempts on Nd-Fe-B-based magnets

1. Grain size refinement



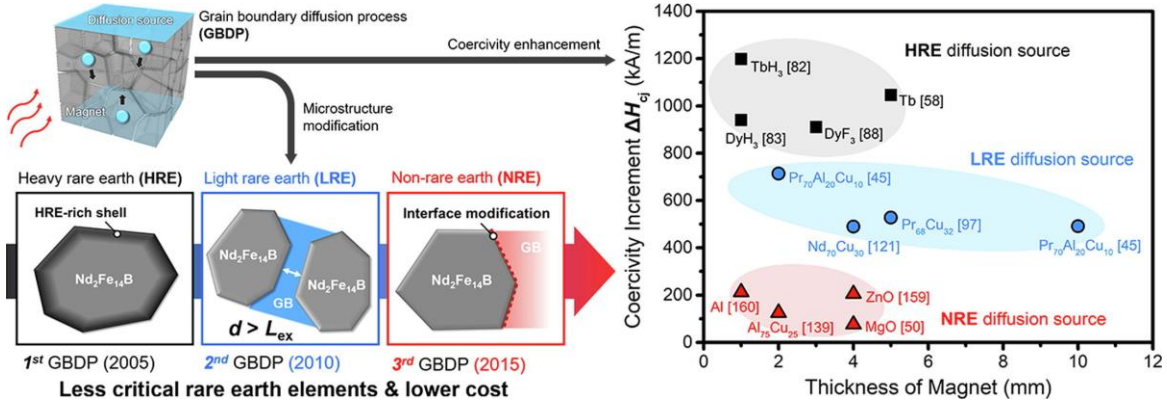
Scripta Materialia, 67(2012) 530

2. Grain boundary phase modification



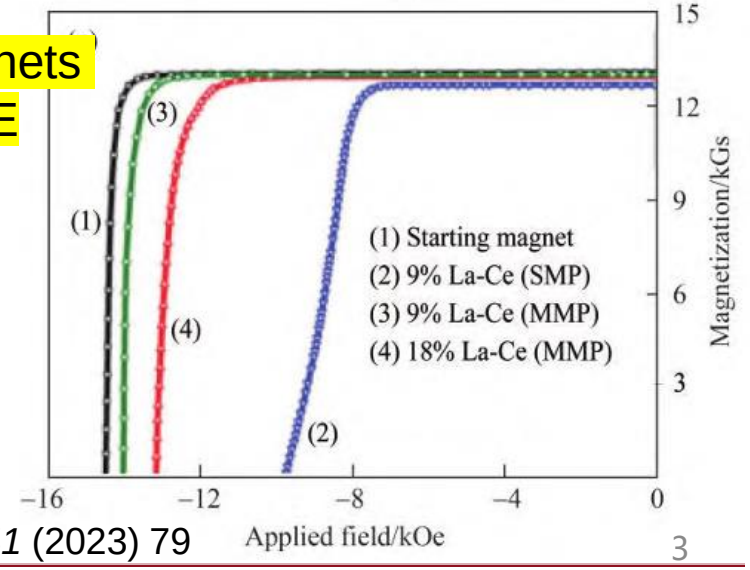
JALCOM, 790(2019) 750-759

3. Grain boundary diffusion process



Materials & Design 209 (2021) 110004

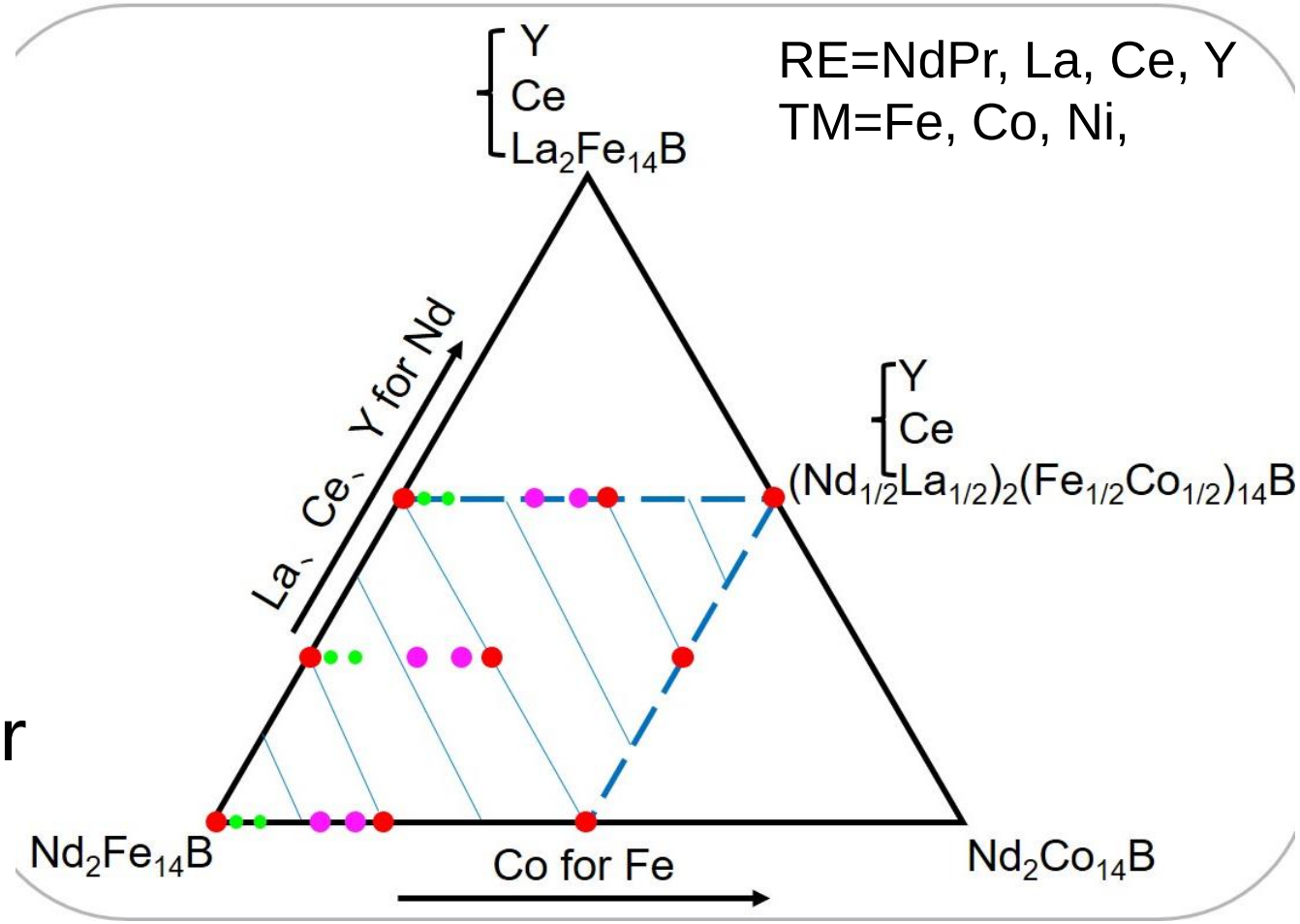
4. Cost-effective magnets w. high abundance RE



J. Chn Soc. Rare Earths 41 (2023) 79

The purpose of this work

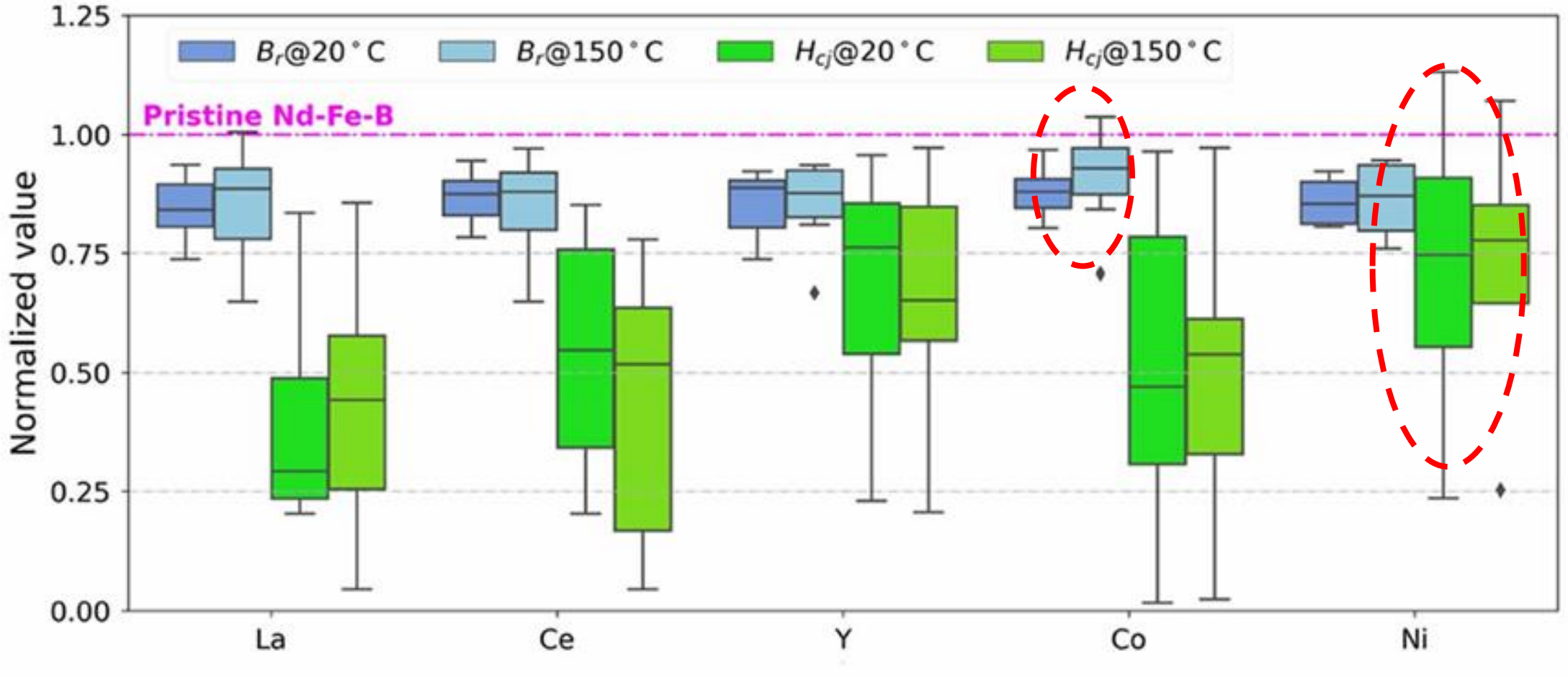
- To explore magnets with exceptional high temperature stability at ~150℃ and above.
- Use cost-effective LREs for the performance cost of magnet instead of using HREs, high-T_c elements.
- Development under the near mass-production condition.



$$[(Nd_{0.8}Pr_{0.2})_{(1-x)}(La/Ce/Y)_x]_{13.86}[Fe_{(1-y)}(Co/Ni)_y]_{77.10}M_{3.30}B_{5.74}\ (M=Al, Cu, Ga, Ti),\ at.\%$$

Magnetic properties of the 24 starting compos.

1. For Br@150°C, higher values than that of pristine Nd–Fe–B could be achieved at a high doping level of Co.
2. A decrease in Hcj is observed for all the doping species except Ni.



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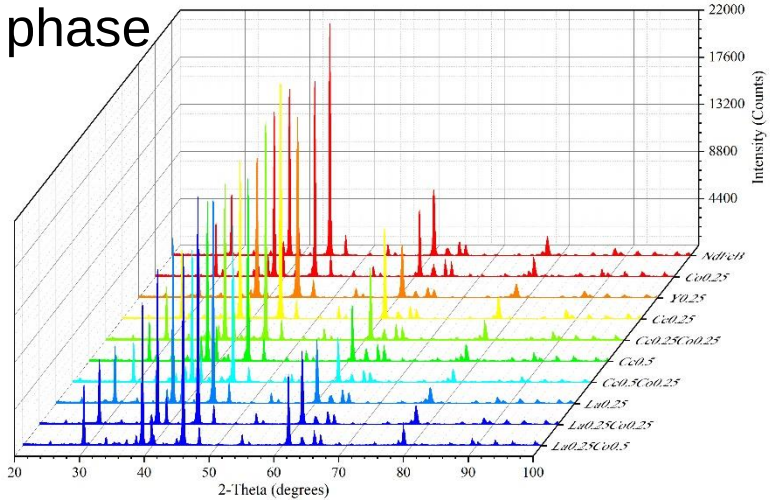
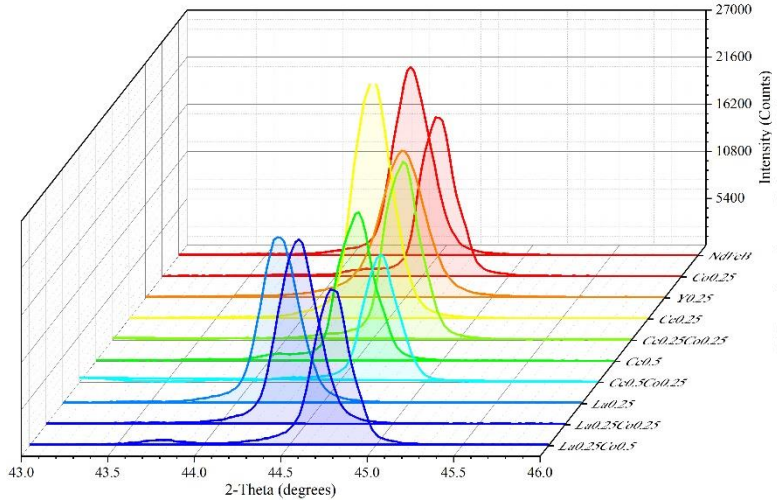
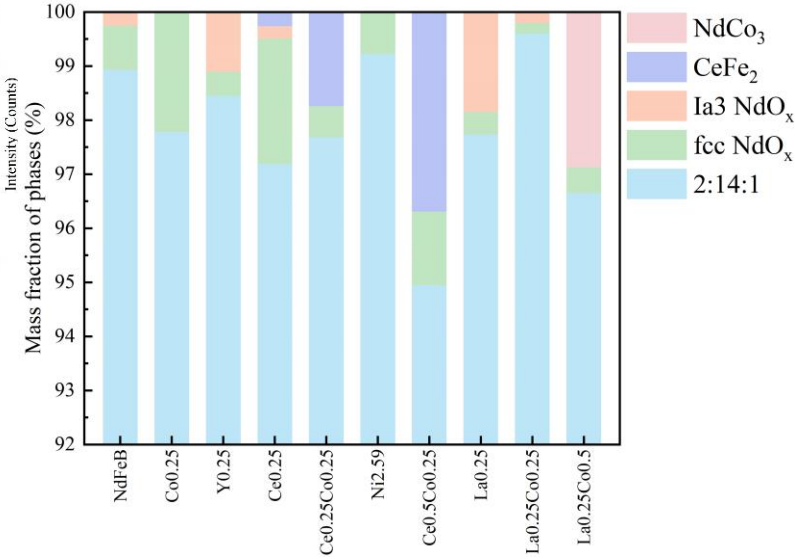
Phase constitution and microstructure of the magnets

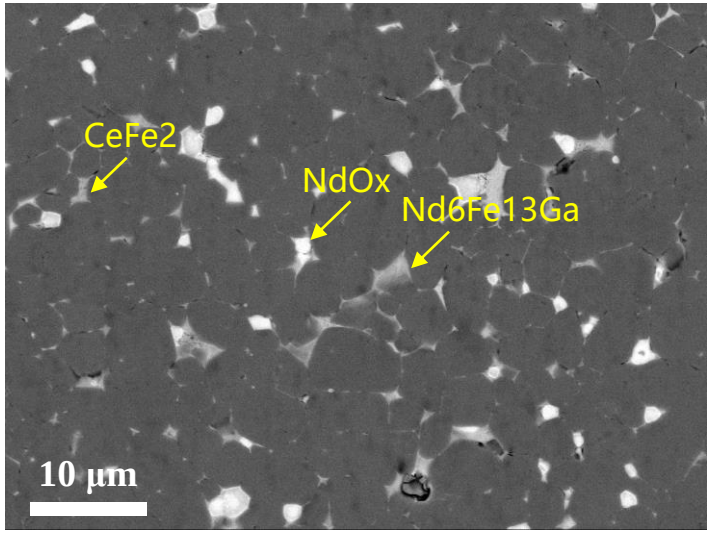
2:14:1 phase >95%

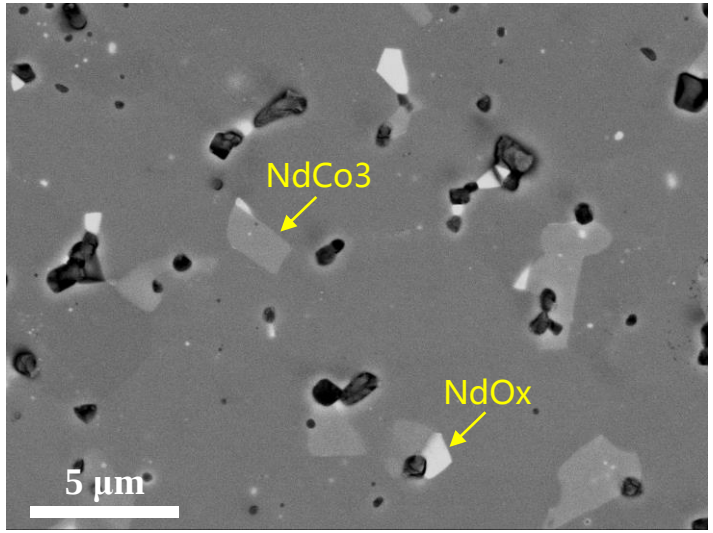


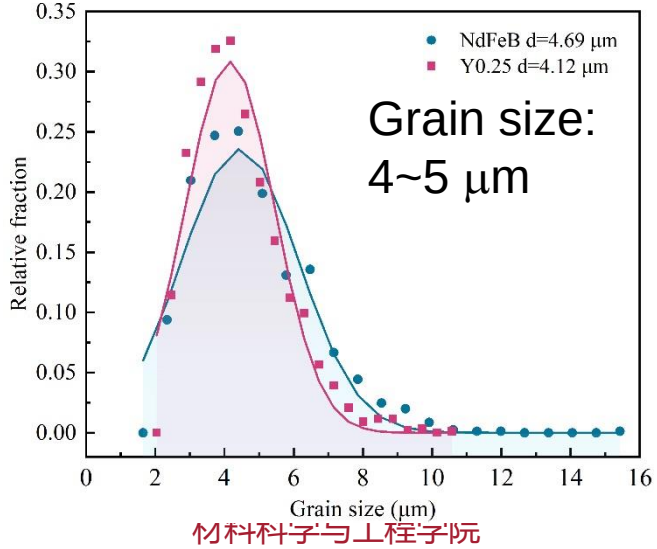




Microstructure

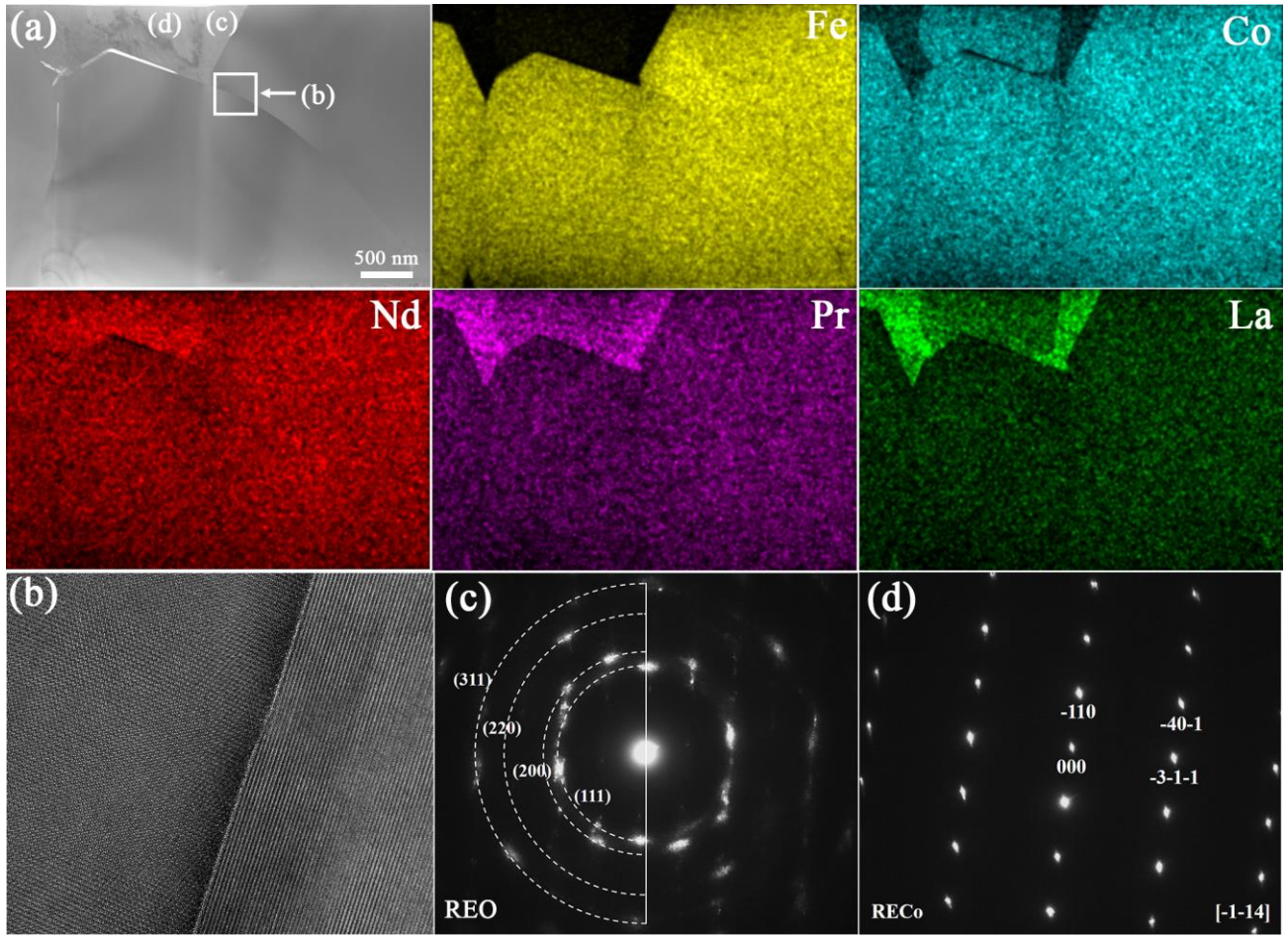






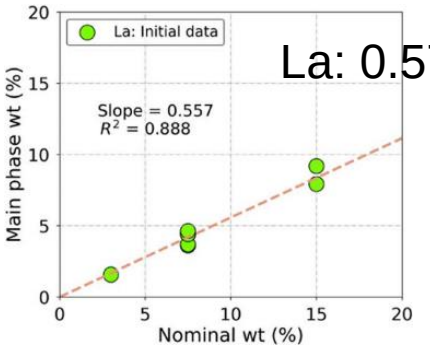
Phase constitution and microstructure of the magnets

Details at GB in a $La_{0.25}Co_{0.25}$ magnet

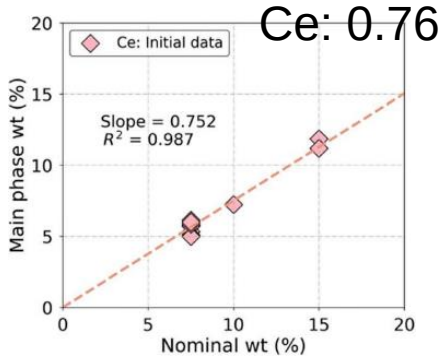


Partitioning behavior:

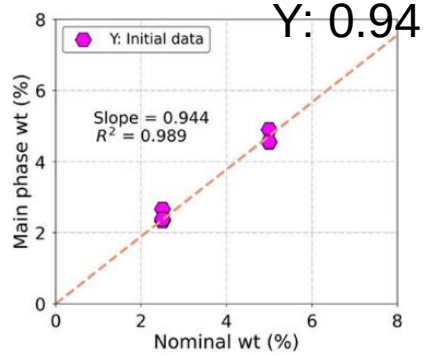
$$\frac{\text{Main phase wt.\%}}{\text{Nominal wt.\%}}$$



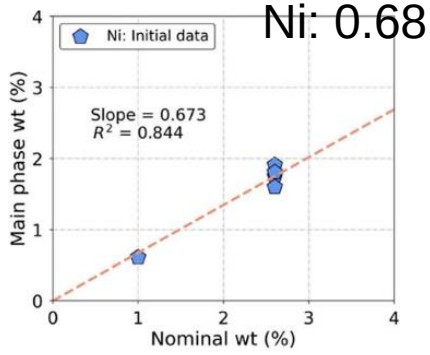
La: 0.57



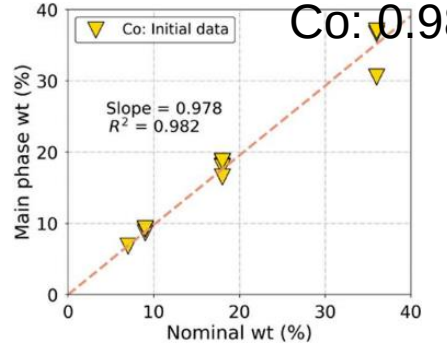
Ce: 0.76



Y: 0.94

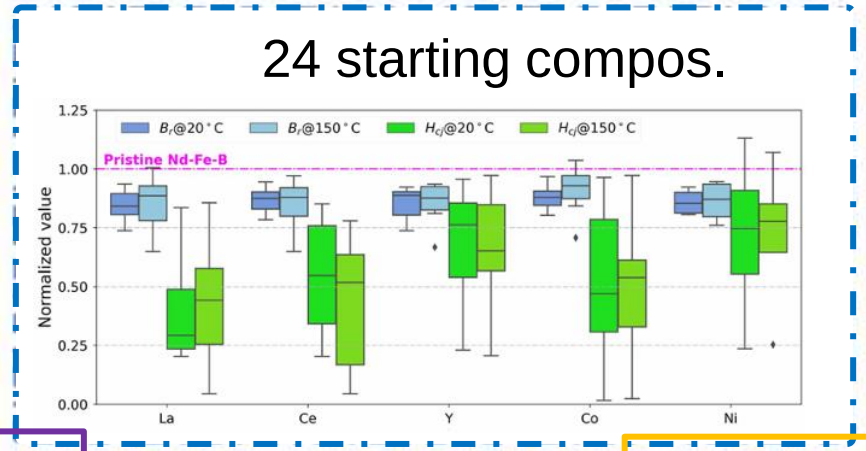


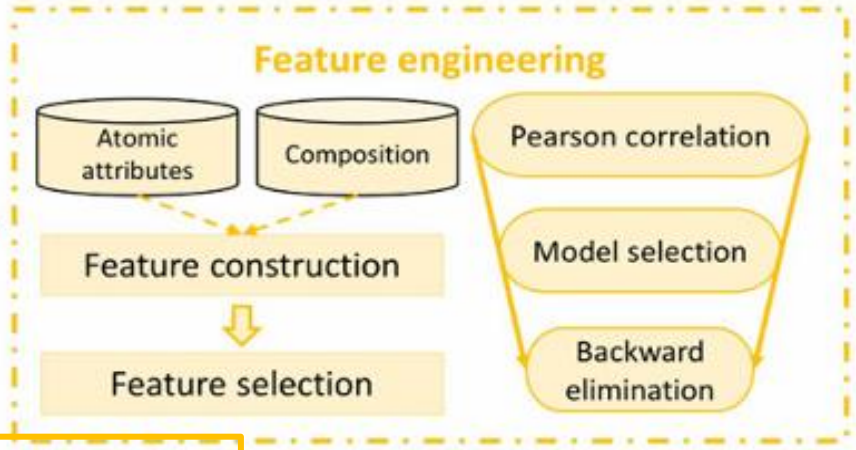
Ni: 0.68



Co: 0.98

The adaptive-learning framework

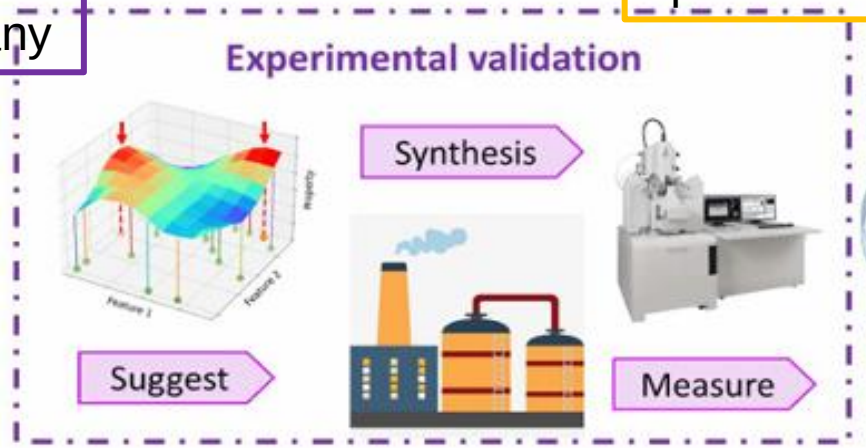


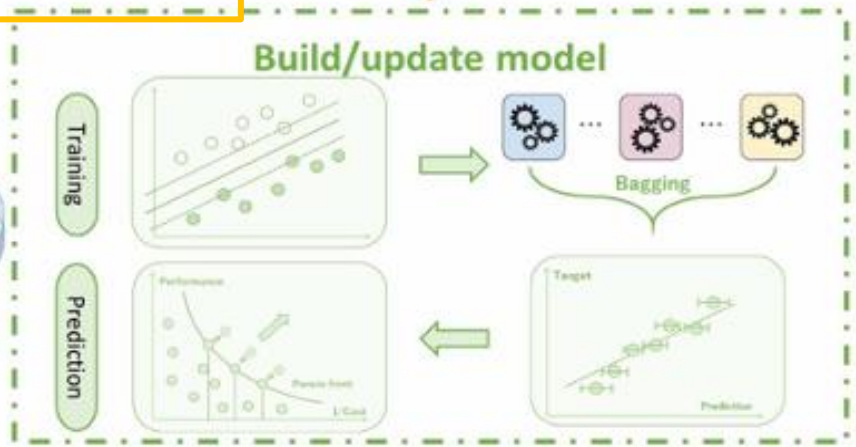


3. Magnets prepared in the company

4. Experimental feedback to update the surrogate model

1. Kick-start model: constructing a rough initial surrogate model with a very small amount of data





2. Bayesian Optimization (BO) to make recommendations to max. optimal candidate

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Model construction

1. 16 atomic features

Atomic feature
Atomic number Z_R (Z_T)
Atomic radii r_R (r_T)
Electronegativity χ_R (χ_T)
Atomic mass m_R (m_T)
Valence electron number VEN_R (VEN_T)
Spin angular moment S_{4f} (S_{3d})
Orbital angular moment L_{4f} (L_{3d})
Ionization potential IP_R (IP_T)
Density ρ_R (ρ_T)
Melting point Tm_R (Tm_T)
Boiling point Tb_R (Tb_T)
Enthalpy of fusion Ef_R (Ef_T)
Enthalpy of vaporization Ev_R (Ev_T)
Enthalpy of atomization Ea_R (Ea_T)
Curie Temperature Tc_R (Tc_T)
Saturation magnetization μ_R (μ_T)

2. Feature engineering

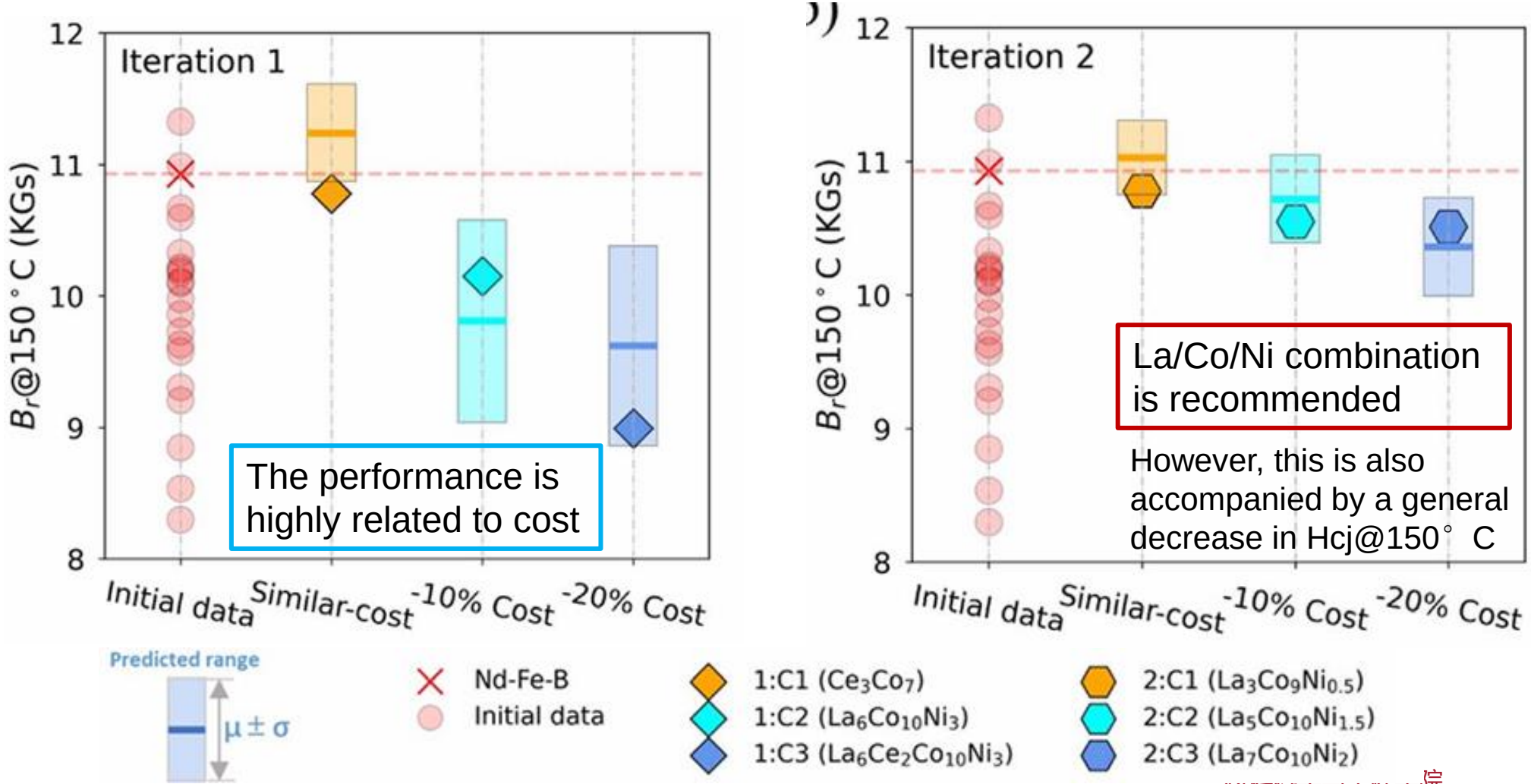
Estimator	Selected Features	B_r@20℃: CV	B_r@150℃: CV
GP	['IP_{RT}', 'x_{RT}', 'Ev_R', 'Ef_{RT}']	0.6	0.83
SVR	['Tc_T', 'Tc_{RT}', 'IP_{RT}', 'μ_{RT}', 'Ev_R', 'IP_T']	0.39	0.61
Ridge	['Tc_{RT}', 'IP_T', 'Ev_R', 'Ea_{RT}']	0.50	0.89
KNN	['$L4f_R$', 'Ea_{RT}', 'IP_T', 'm_R', 'Ef_R']	0.62	0.83
ANN	['μ_T', 'μ_R', 'μ_{RT}', 'r_R', 'm_R', 'IP_T']	0.57	1.07
DT	['$L4f_R$', 'Tm_{RT}', 'Tc_{RT}']	0.74	0.85

3. Uncertainty estimation: booststrapping

4. Acquisition function: upper confidence bound

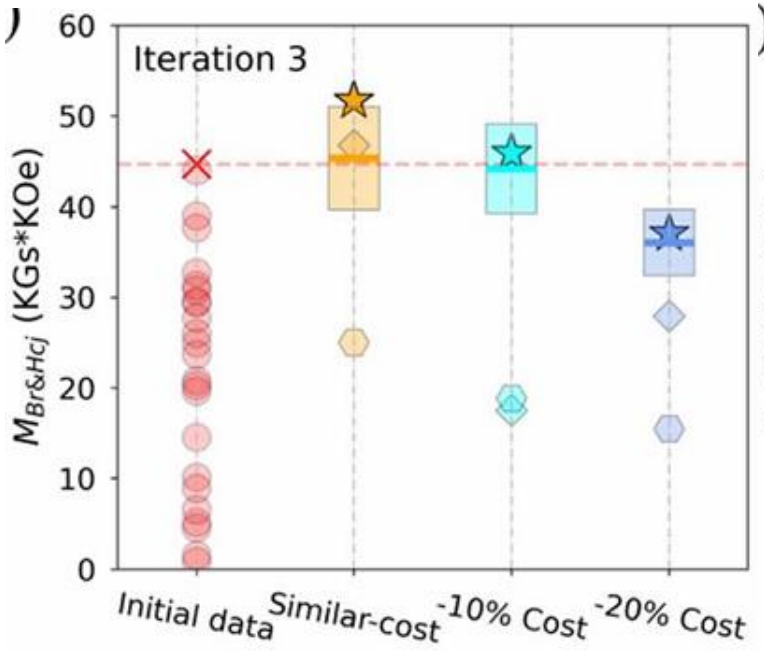
Stage 1 optimization: B_r@150°C

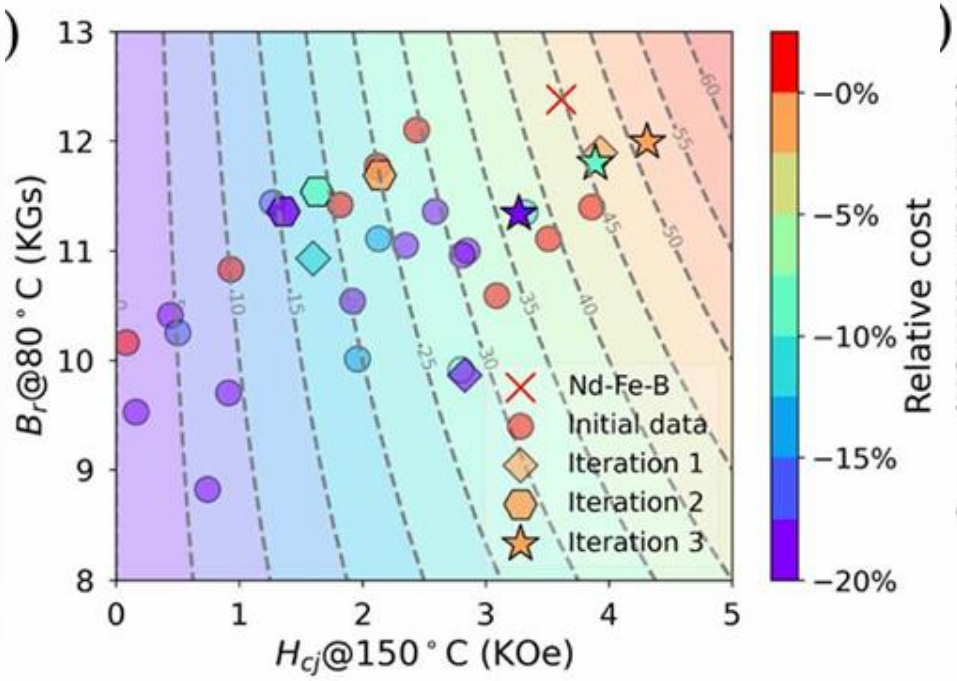
- To identify compositions with exceptional HT B_r



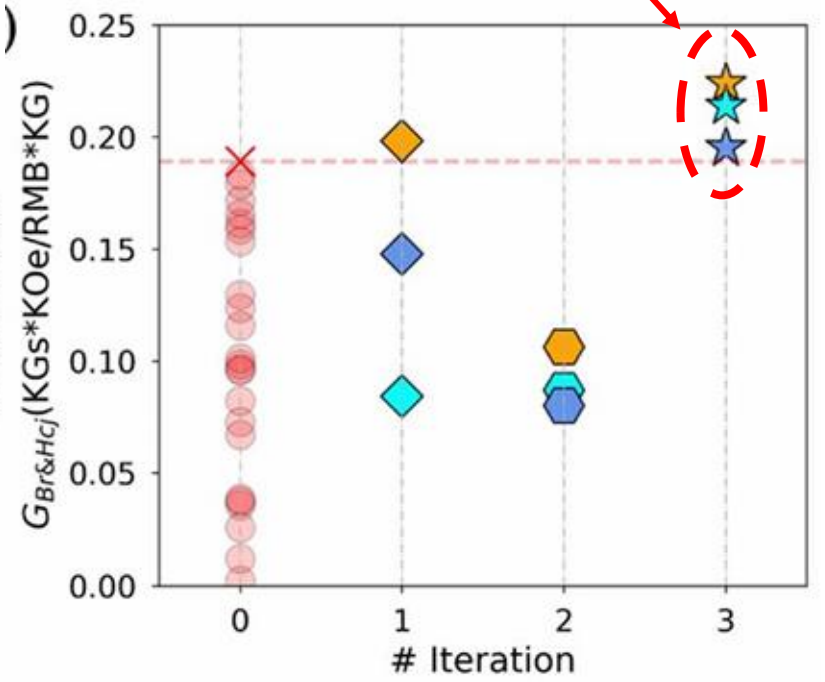
Stage 2 optimization: B_r@80°C*H_{cj}@150°C

- To improve the overall performance





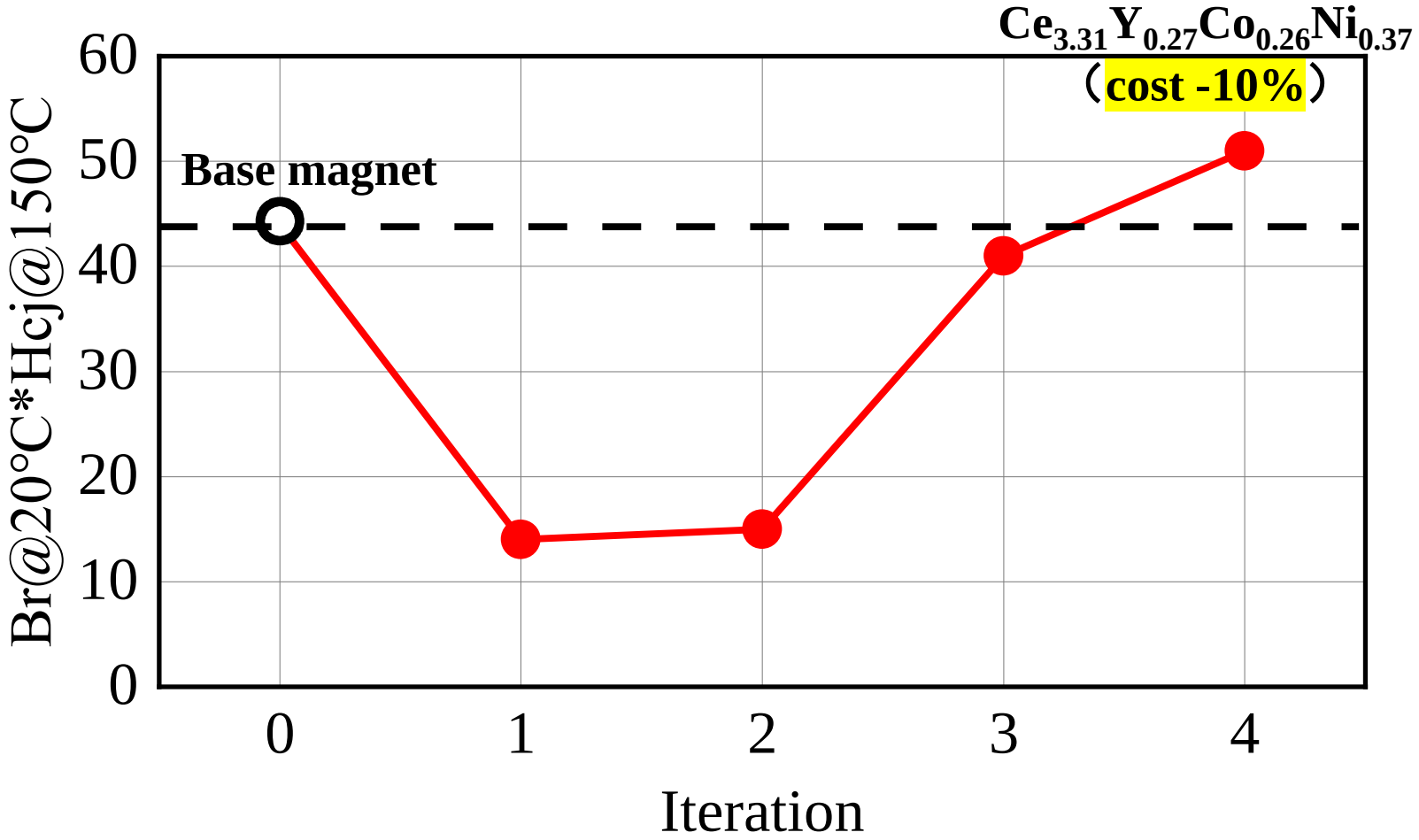
Improved performance-cost ratio



Performance–cost ratio: $G_{Br\&Hcj} = M_{Br\&Hcj}$ /cost

The best two candidates showed 18.4% (3: C1) and 13.1% (3: C2) improvement in performance-cost ratio with respect to benchmark Nd-Fe-B

Evolution of properties after iteration

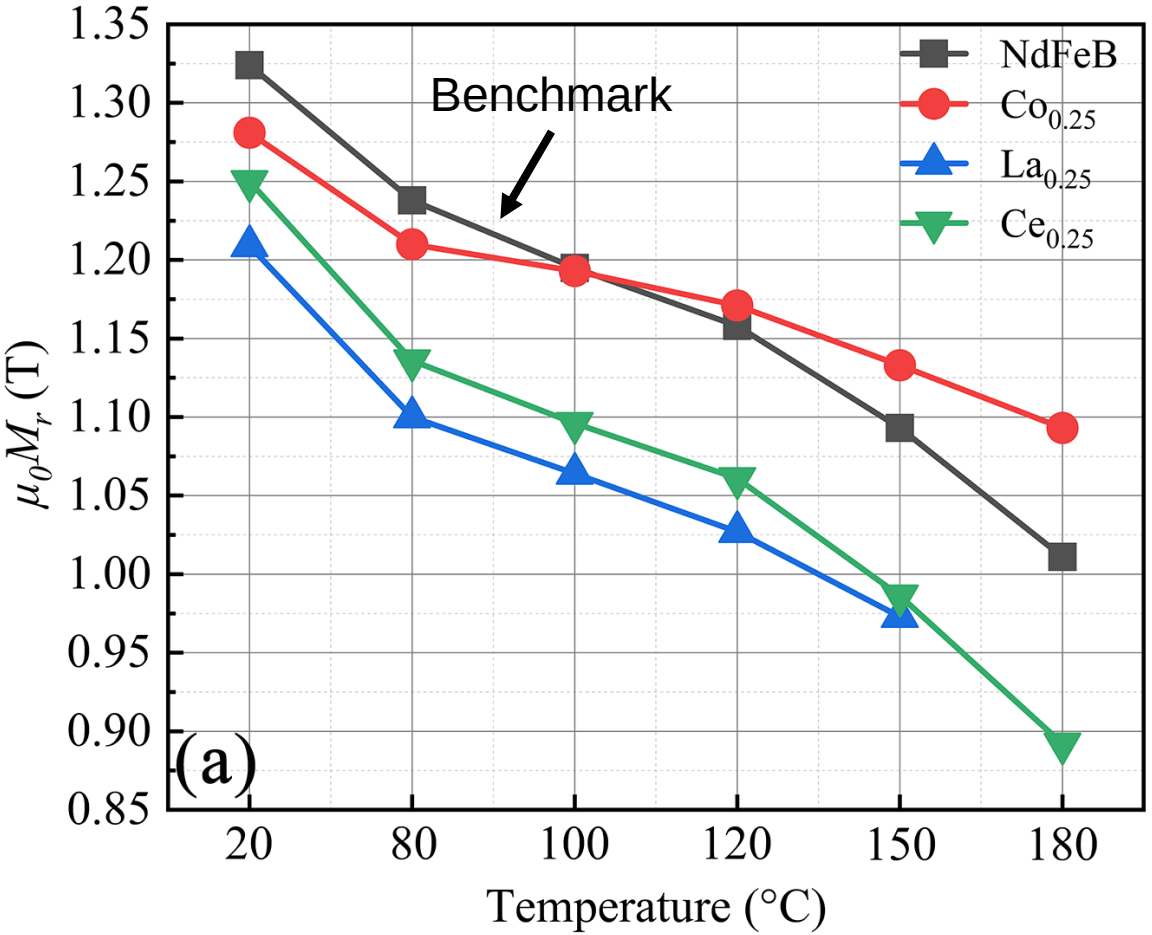


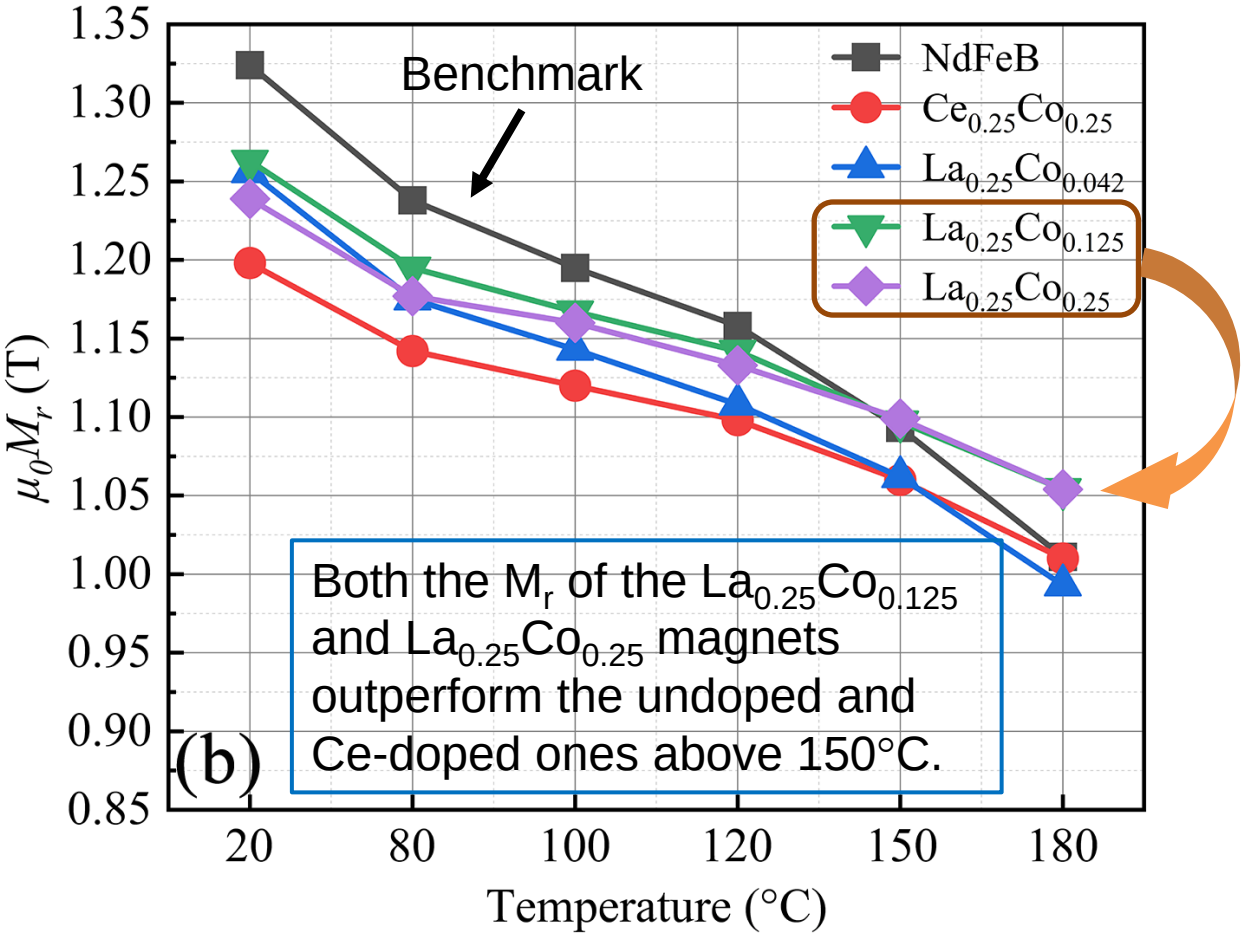
- The La-Co, Ce-Co, La-Co-Ni combinations outperform the other combinations.

- After 4 rounds of iteration, composition with higher performance to the base composition but 10% lower cost was found.

The La-Co pairs w/. higher M_r above 150°C

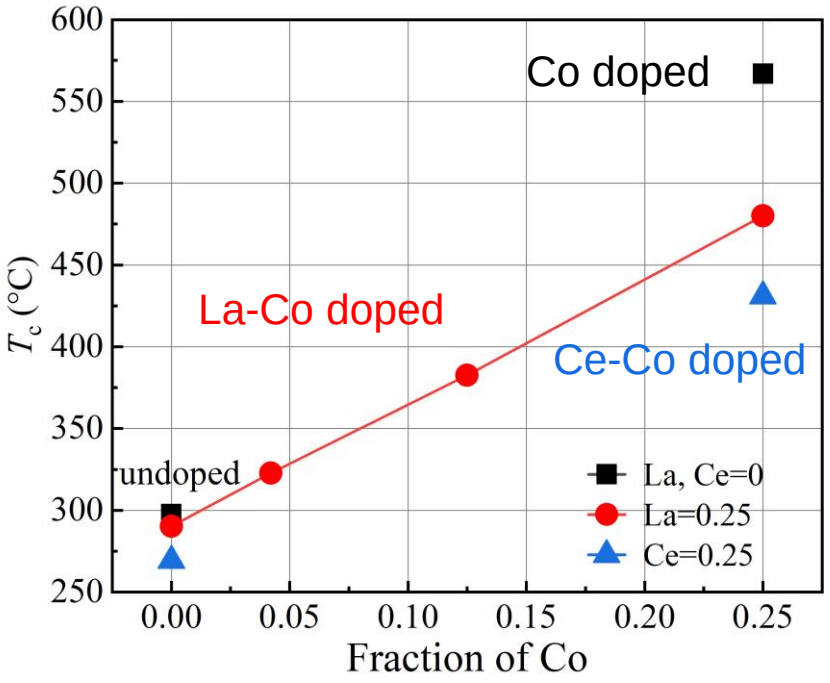
$[(Nd_{0.8}Pr_{0.2})_{(1-x)}(La/Ce)_x]_{13.86}[Fe_{(1-y)}Co_y]_{77.10}TM_{3.30}B_{5.74}$ ($0 \le x \le 0.25$, $0 \le y \le 0.25$, TM=Al, Ti, Cu, Ga), at.%



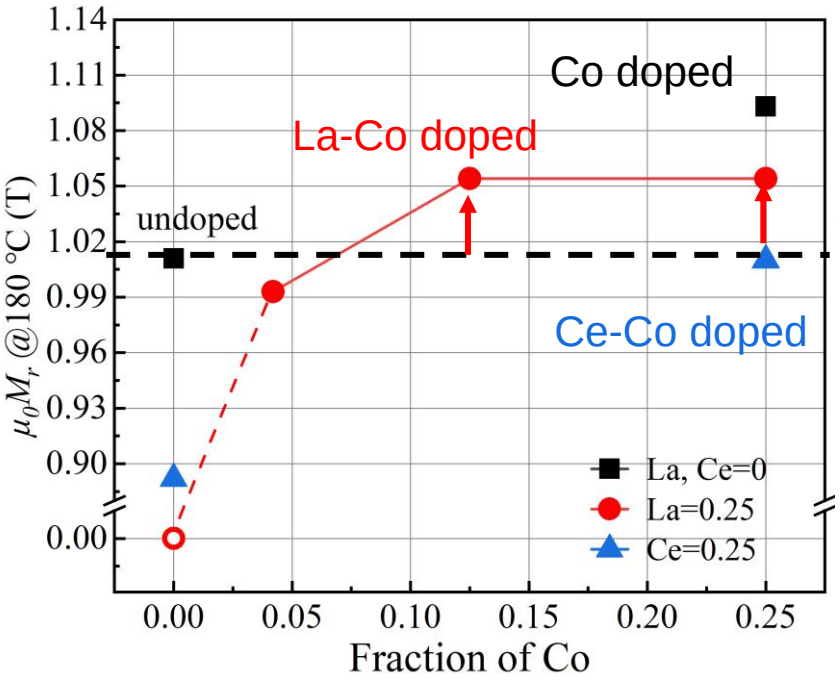


Effect of LRE-Co pair on T stability of M_r

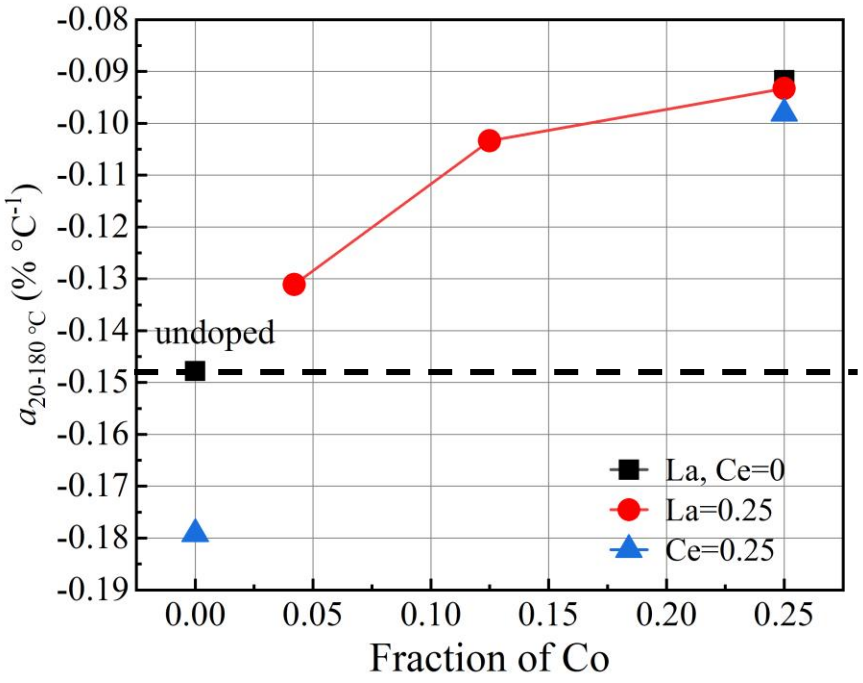
Curie temperature



M_r@180°C



$$\alpha(T) = \frac{\mu_0 M_r(180°C) - \mu_0 M_r(20°C)}{\mu_0 M_r(20°C) \times \Delta T}$$



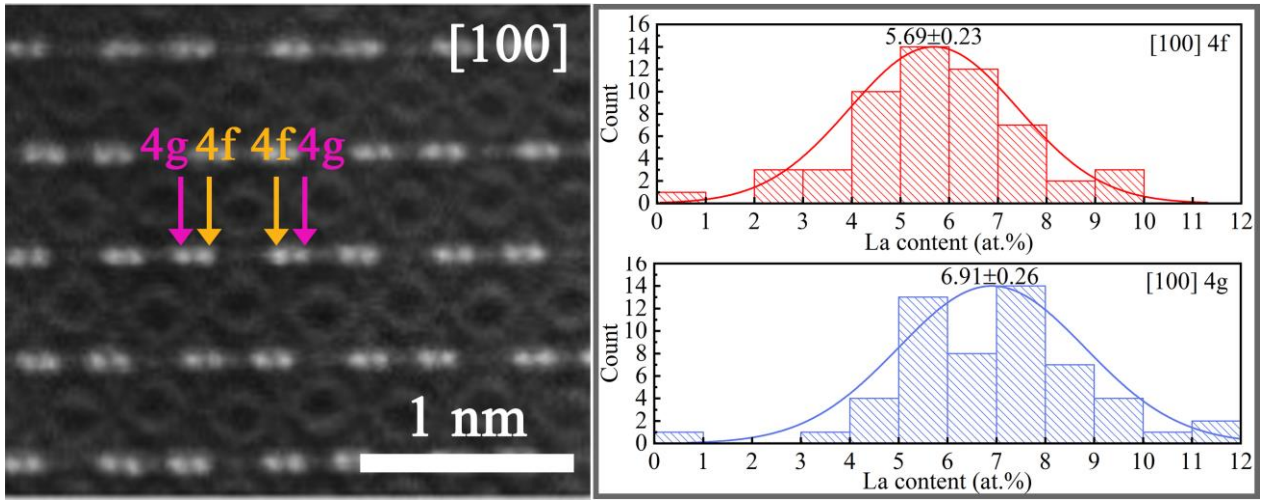
- Without Co, the M_r@180°C of La-doped magnet is inferior to that of Ce-doped one
- For $La_{0.25}Co_{0.125}$, higher M_r@180°C despite its lower T_c than $Ce_{0.25}Co_{0.25}$
- The $La_{0.25}Co_{0.25}$ and $Ce_{0.25}Co_{0.25}$ magnets demonstrate almost the same α as that of the $Co_{0.25}$ even though much lower T_c

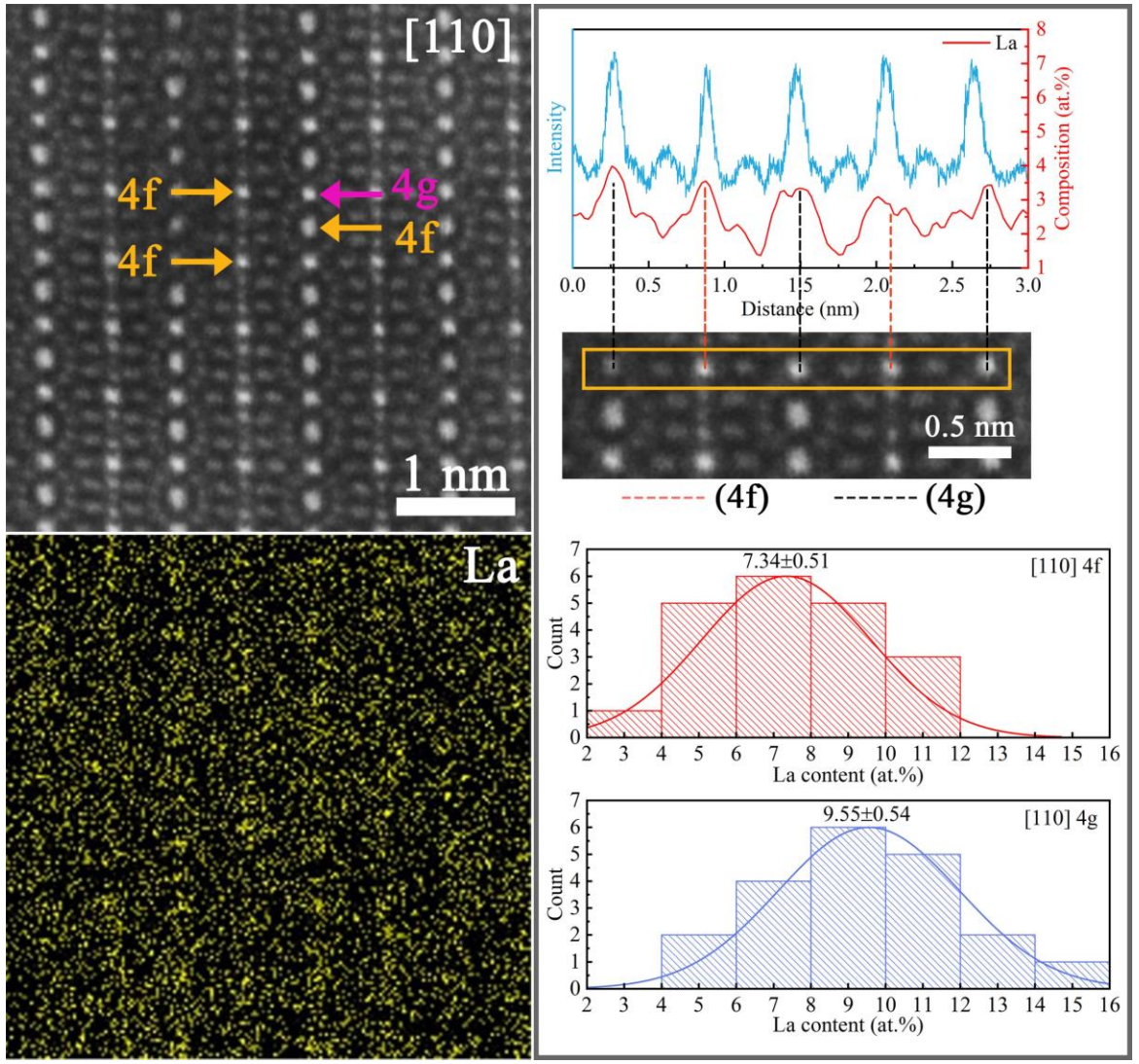
Preferential site occupation of La

- $La_{0.25}Co_{0.25}$, La preferentially occupies the 4g-site

La concentration (at.%)

Zone axis	4g-site	4f-site
[110]	9.55±0.54	7.34±0.51
[100]	6.91±0.26	5.69±0.23



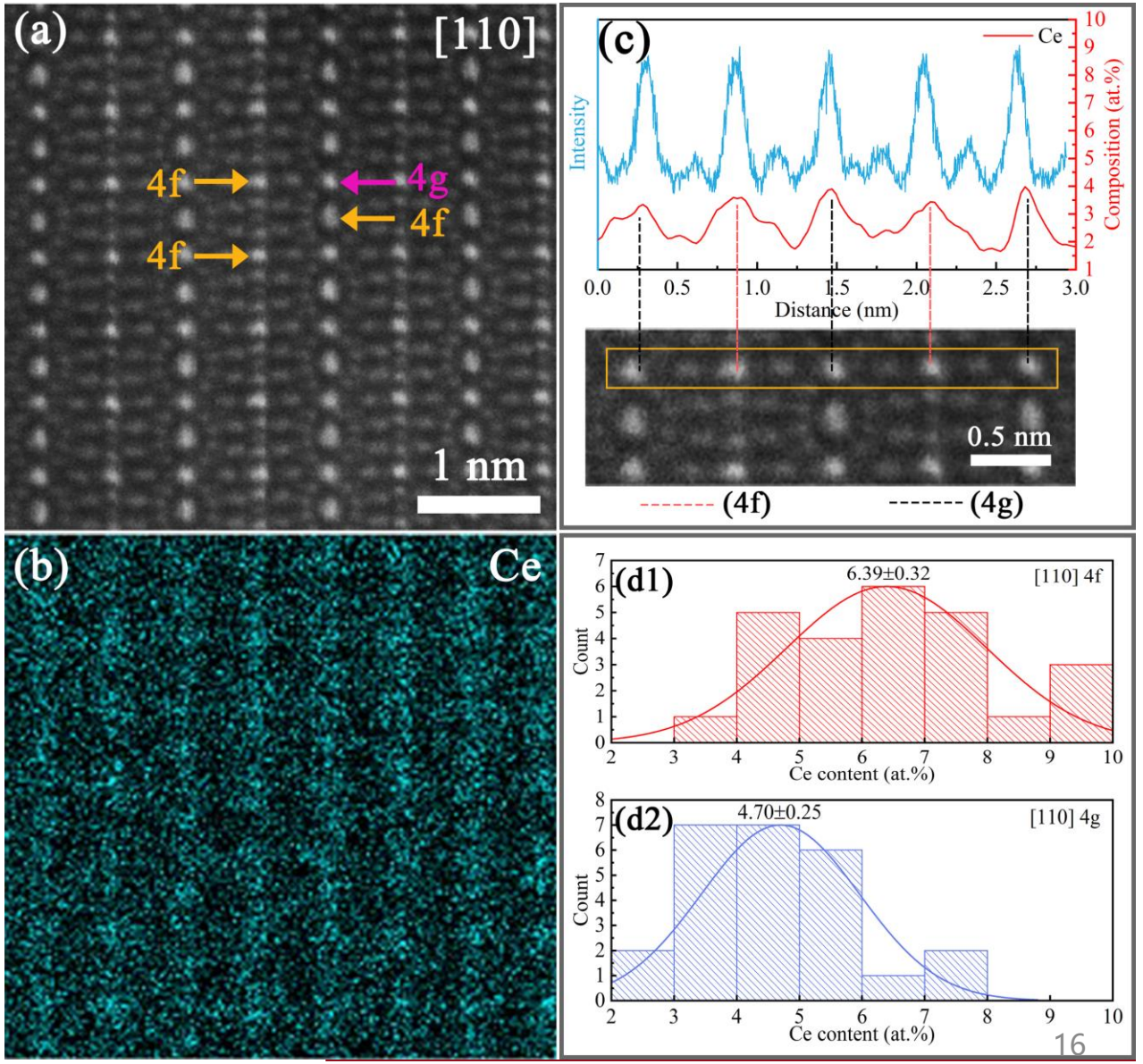


Preferential site occupation of Ce

- $Ce_{0.25}Co_{0.25}$
- Ce preferentially occupies the 4f-site

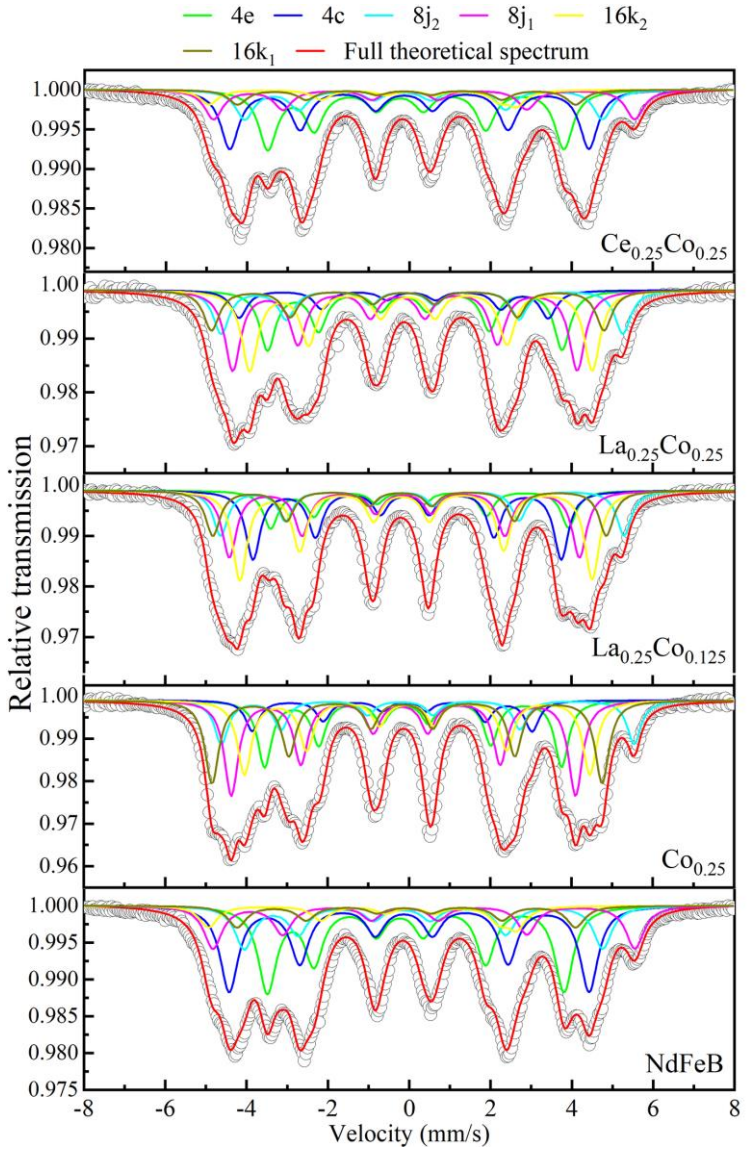
Ce concentration (at.%)

Zone axis	4g-site	4f-site
[110]	4.70±0.25	6.39±0.32



Preferential site occupations of Co

Mössbauer spectra



Refined relative areas (±1%) of the Fe sites in the lattice

Fe site	Undoped	$Co_{0.25}$	$La_{0.25}Co_{0.125}$	$La_{0.25}Co_{0.25}$	$Ce_{0.25}Co_{0.25}$
$16k_1$	28.57	21.09	12.86	12.12	26.42
$16k_2$	28.57	18.88	25.28	24.24	20.85
$8j_1$	14.29	24.18	18.96	24.15	11.95
$8j_2$	14.29	11.11	12.77	13.15	16.20
4c	7.14	7.85	19.5	8.37	14.19
4e	7.14	16.9	10.61	17.98	10.39
Pref. occup.		$16k_1$,$16k_2$, $8j_2$	$16k_1$	$16k_1$	$8j_1$

- Preferential occupations of the $16k_1$, $16k_2$ and $8j_2$ sites by Co alone
- If with La, Co occupies the $16k_1$ site preferentially
- If with Ce, Co occupies the $8j_1$ site preferentially

Summary

- Several promising compositions with La, Ce, Y, Co and Ni as doping elements with good HT remanence and/or enhanced performance-cost ratio are identified through BO approach, which could impact the development of novel magnet for HT application.
- La-Co pair shows the good HT remanence (>150°C) over the pristine magnet and Ce-Co pair, possibly due to the specific site occupation of the elements in the lattice.

THANK YOU FOR YOUR ATTENTION!