

Supplementary Material

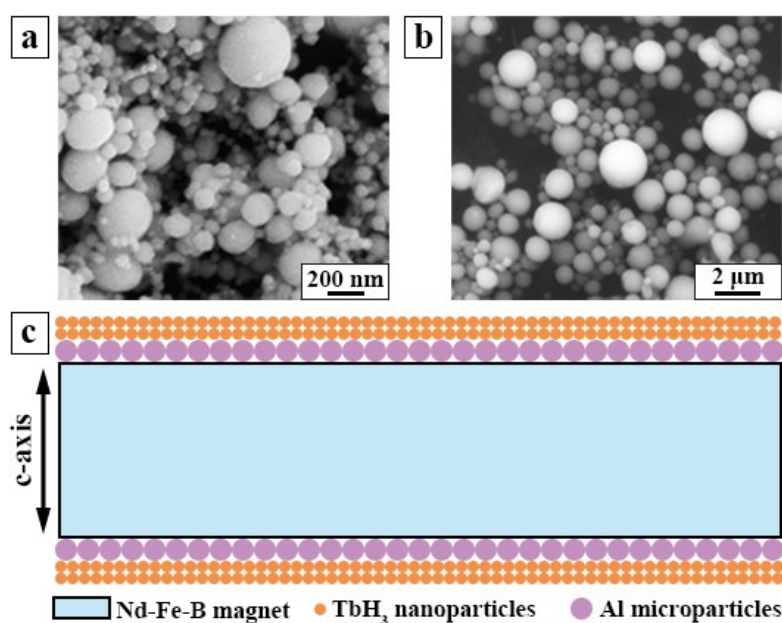


Fig. S1 The micro morphology of the diffusion source and schematic diagram of spraying process. a and b are the micro morphology of TbH_3 nanopowders and Al micropowders, respectively. c is the schematic diagram of the layer-by-layer spraying process.

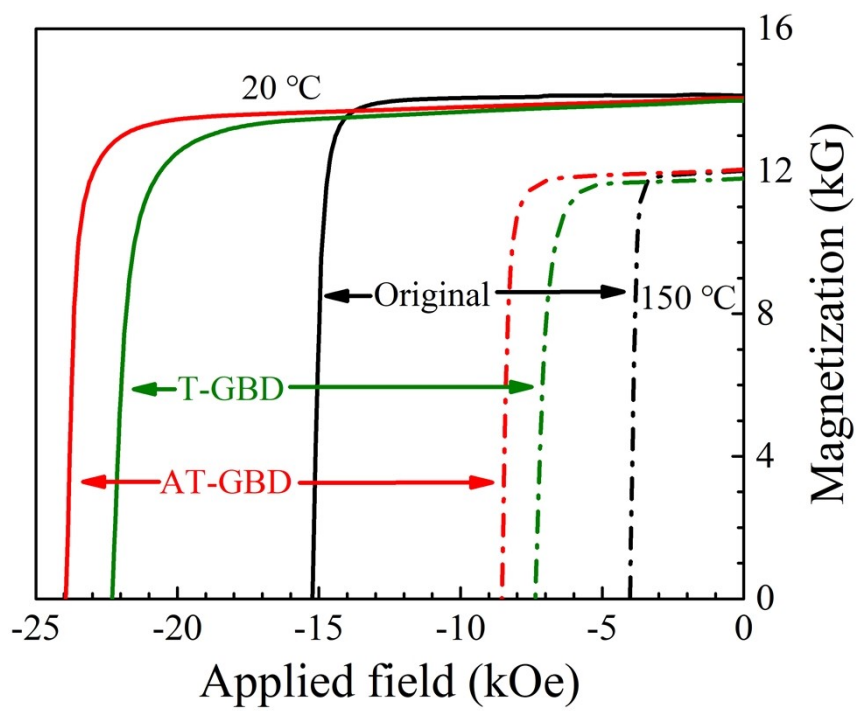


Fig. S2 demagnetization curves at 20 °C and 150 °C of the original, T-GBD, and AT-GBD magnets

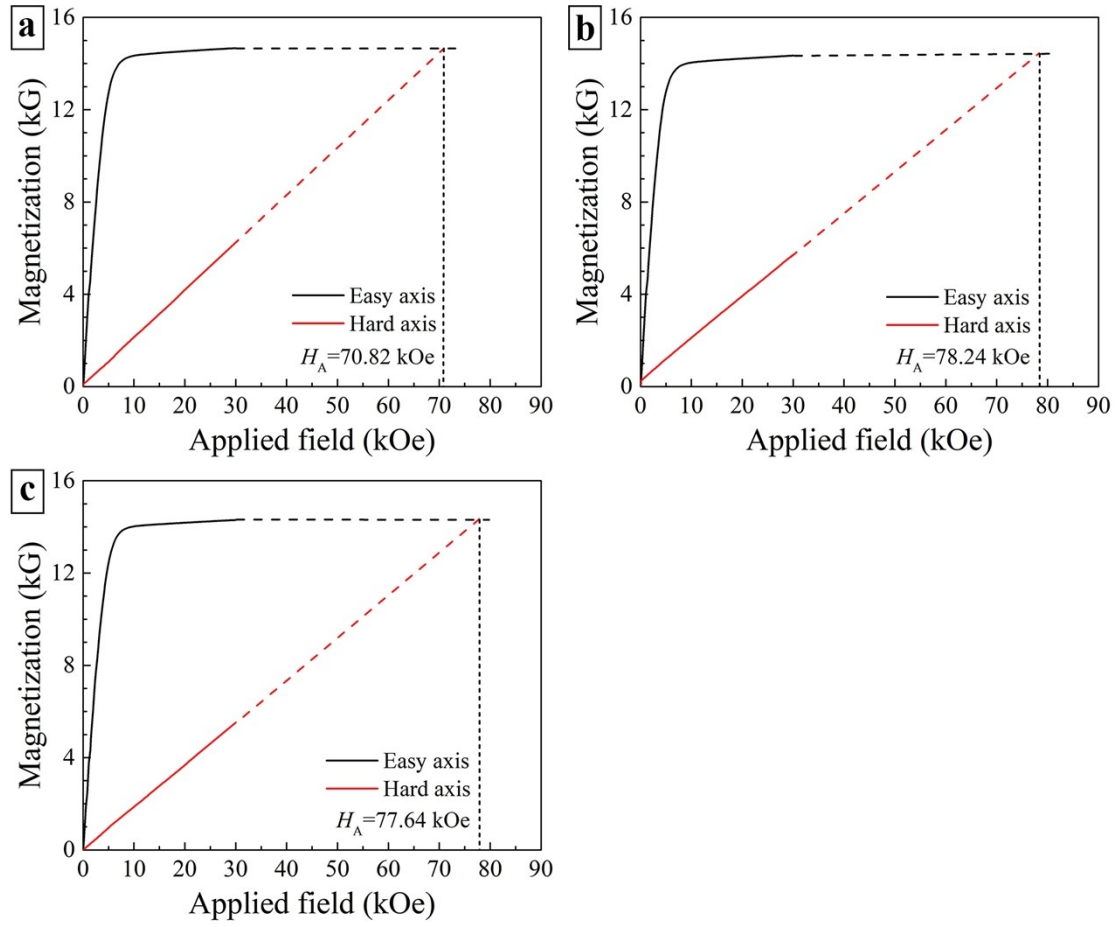


Fig. S3 H_A test results of the original and GBD magnets. a: original magnet, b: T-GBD magnet, c: AT-GBD magnet

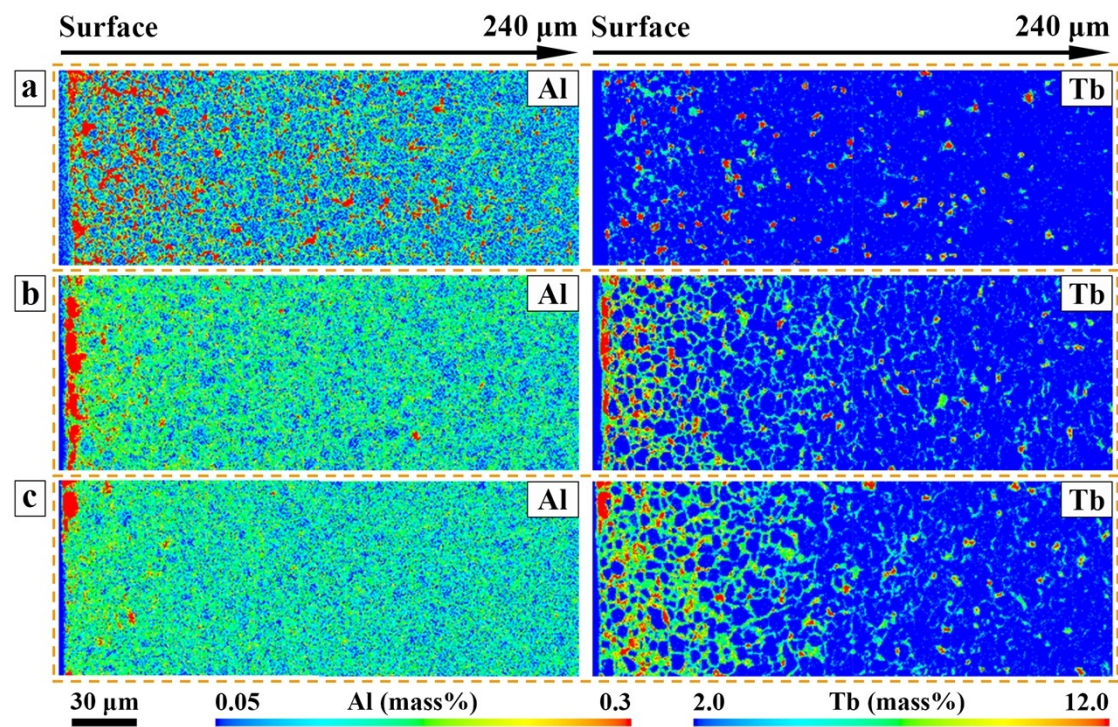


Fig. S4 Al and Tb element distribution GBD magnets with different annealing time at 900 °C. a: 1 min, b: 3h, c: 8h

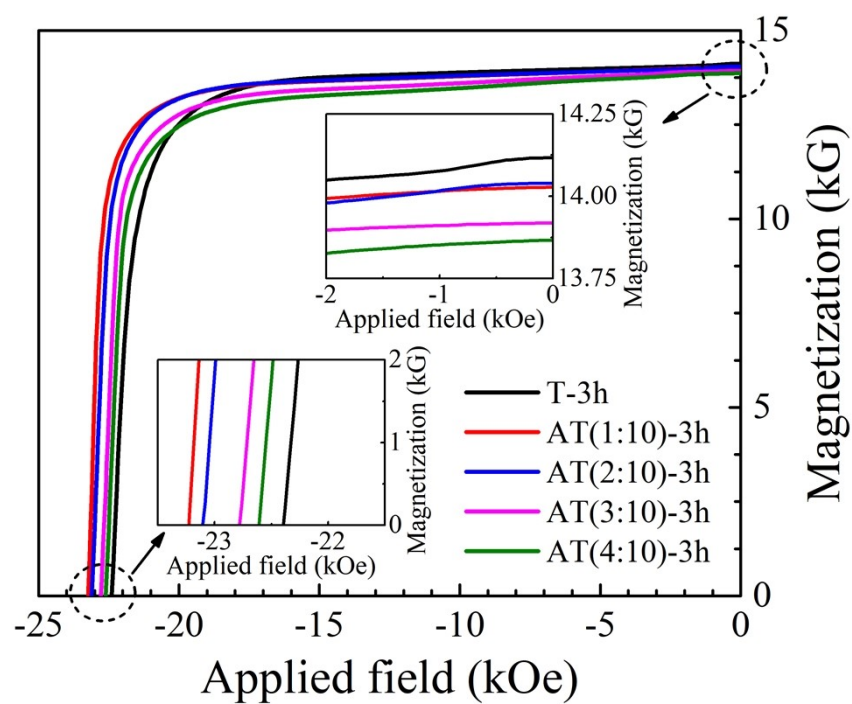


Fig. S5 Demagnetization curves of the T-3h and AT-3h magnets with different Al-to-Tb ratios

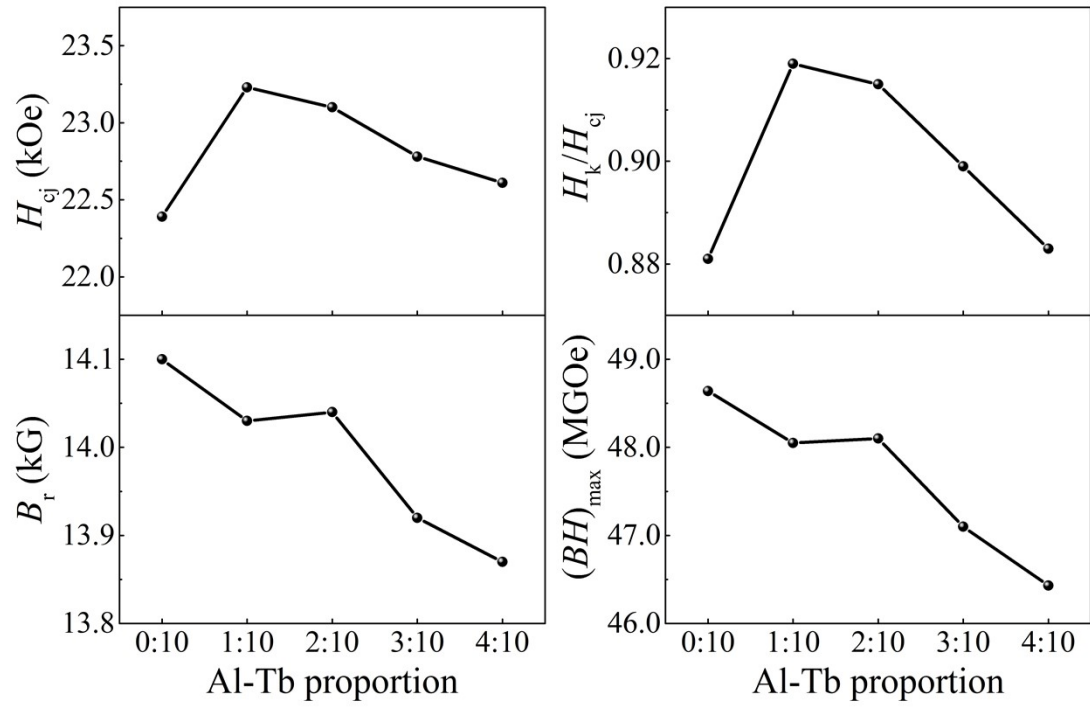


Fig. S6 Variation of magnetic properties of the T-3h and AT-3h magnets with Al-to-Tb ratio

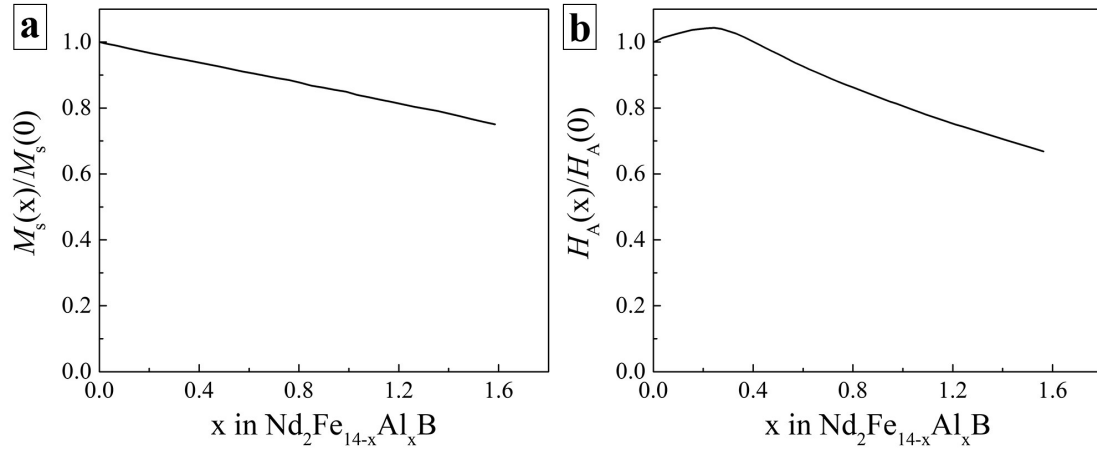


Fig. S7 Variation of the intrinsic magnetic parameters of $\text{Nd}_2\text{Fe}_{14-x}\text{Al}_x\text{B}$ phase with Al content. a:

M_s , b: H_A [7, 40, 65]

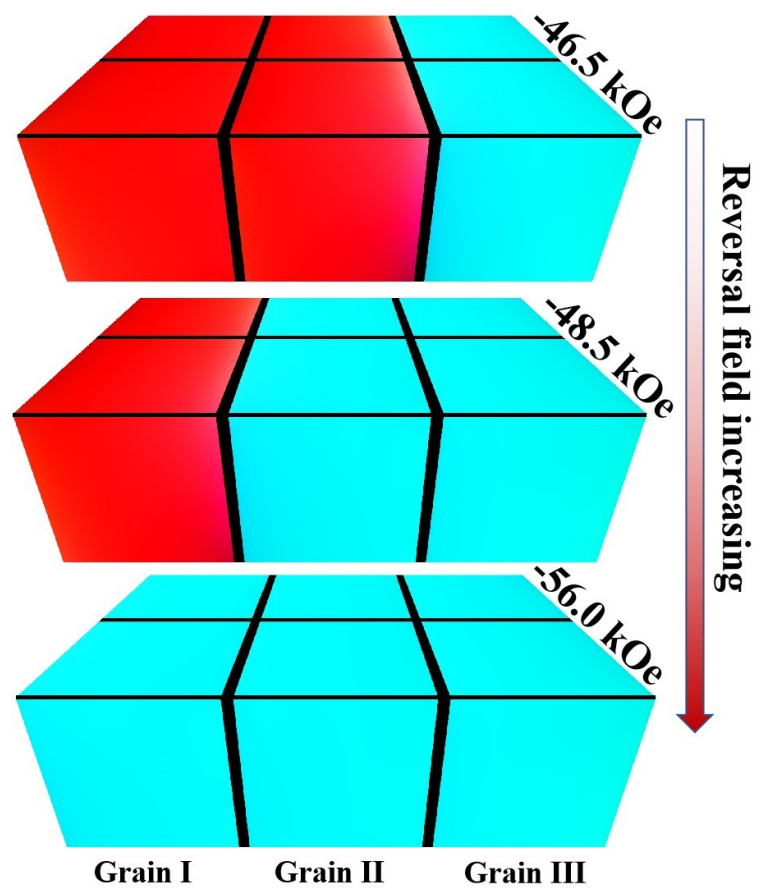


Fig. S8 The magnetization distribution map during the demagnetization process of the without-Al model

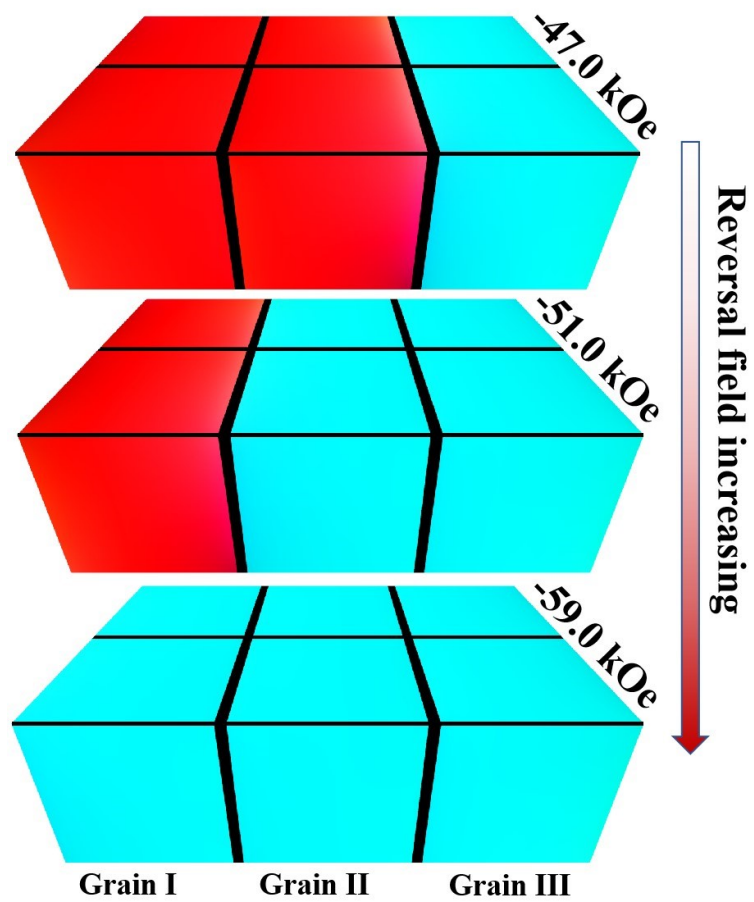


Fig. S9 The magnetization distribution map during the demagnetization process of the low-Al model

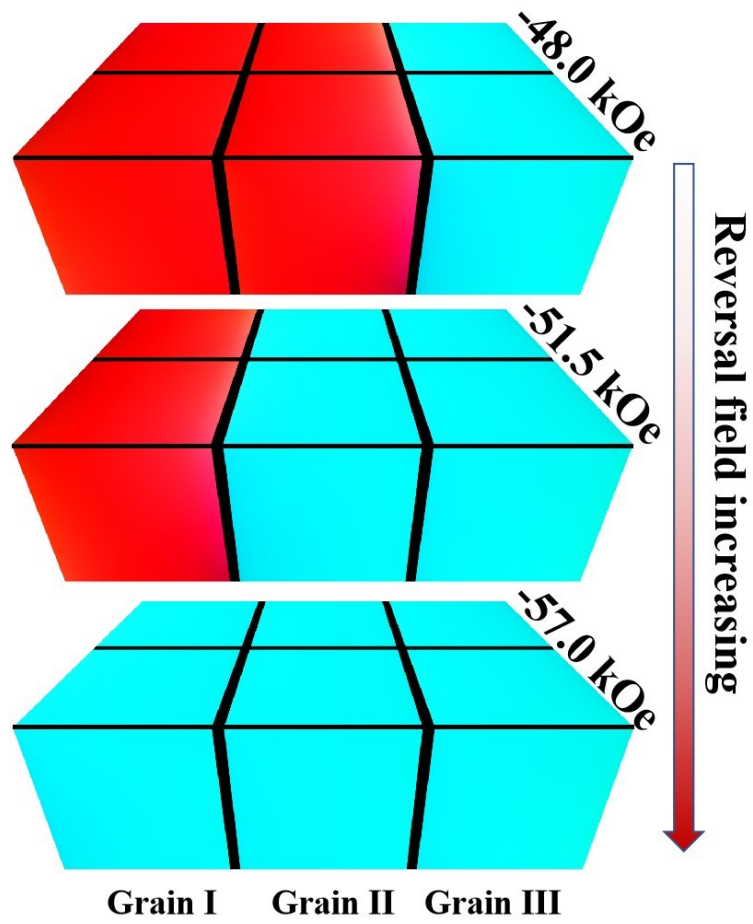


Fig. S10 The magnetization distribution map during the demagnetization process of the medium-Al model

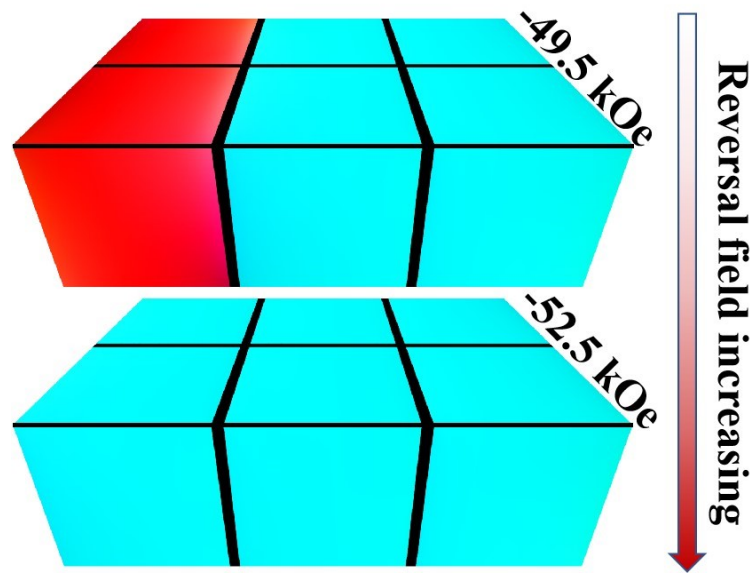


Fig. S11 The magnetization distribution map during the demagnetization process of the high-Al model

Table S1 Energy of Fe atom in Nd₂Fe₁₄B substituted by Al atom

Site	Total Energy (eV/molecular)	Formation Energy (eV/atom)
16k ₁	-543.13	-0.0241
16k ₂	-545.39	-0.0573
8j ₁	-546.27	-0.0702
8j ₂	-546.28	-0.0704
4c	-542.22	-0.0107
4e	-544.00	-0.0369

Table S2 Energy of Nd atom in Nd₂Fe₁₄B substituted by Tb atom

Site	Total Energy (eV/molecular)	Formation Energy (eV/atom)
Nd ₂ Fe ₁₄ B	-549.97	-0.0575
4f	-548.07	-0.0621
4g	-546.99	-0.0463

Table S3 Energy of Nd atom in Nd₂(Fe,Al)₁₄B substituted by Tb atom

Site	Total Energy (eV/molecular)	Formation Energy (eV/atom)
4f	-544.50	-0.0767
4g	-544.03	-0.0698

Table S4 the intrinsic magnetic parameters of different grains in the without-Al model [7, 40, 65]

	Grain I			Grain II			Grain III	
	Core	Shell	Surface	Core	Shell	Surface	Grain	Surface
K (MJ/m ³)	5.06	4.92	3.99	4.5	4.78	3.87	4.5	3.65
M_s (MA/m)	0.99	1.07	0.96	1.28	1.14	1.03	1.28	1.15
$\mu_0 H_A$ (T)	10.2	9.2	8.3	7.0	8.4	7.5	7.0	6.3

Table S5 the intrinsic magnetic parameters of different grains in the low-Al model [7, 40, 65]

	Grain I			Grain II			Grain III	
	Core	Shell	Surface	Core	Shell	Surface	Grain	Surface
K (MJ/m ³)	4.68	5.11	4.14	4.61	4.9	3.97	4.52	3.66
M_s (MA/m)	1.27	1.06	0.95	1.27	1.13	1.02	1.27	1.15
$\mu_0 H_A$ (T)	7.4	9.6	8.7	7.3	8.7	7.8	7.1	6.4

Table S6 the intrinsic magnetic parameters of different grains in the medium-Al model [7, 40, 65]

	Grain I			Grain II			Grain III	
	Core	Shell	Surface	Core	Shell	Surface	Grain	Surface
K (MJ/m ³)	4.54	4.97	4.02	4.68	4.97	4.02	4.59	3.72
M_s (MA/m)	1.26	1.05	0.95	1.27	1.13	1.02	1.27	1.15
$\mu_0 H_A$ (T)	7.2	9.5	8.5	7.4	8.8	7.9	7.2	6.5

Table S7 the intrinsic magnetic parameters of different grains in the high-Al model [7, 40, 65]

	Grain I			Grain II			Grain III	
	Core	Shell	Surface	Core	Shell	Surface	Grain	Surface
K (MJ/m ³)	4.15	4.53	3.67	4.41	4.68	3.79	4.68	3.79
M_s (MA/m)	1.24	1.04	0.93	1.25	1.12	1.01	1.14	1.27
$\mu_0 H_A$ (T)	6.7	8.7	7.9	7.1	8.4	7.5	8.2	6.0