VSB TECHNICAL UNIVERSITY OF OSTRAVA | FACULTY OF MATERIALS SCIENCE AND TECHNOLOGY | REGIONAL MATERIALS SCIENCE AND TECHNOLOGY CENTRE

(Pr, Ho)-Fe-B MAGNETS FOR LOW-TEMPERATURE APPLICATIONS

N.B. Kolchugina,[1] A.A. Lukin,[2] G.S. Burkhanov,[1] K. Skotnicová,[3] **O. Životský,[4]** M. Kursa,[3] N.A. Dormidontov,[1] P.A. Prokofev,[2] Y.S. Koshkid'ko,[1,5] T. Čegan,[3] T.P. Kaminskaya,[6] B.A. Ginzburg[6]

[1] *Baikov Institute of Metallurgy and Materials Science, Russian Academy of Sciences, Moscow, Russian Federation, nkolchugina@imet.ac.ru*

[2] *JSC SPETSMAGNIT, Moscow, Russian Federation, pav3387@yandex.ru*

[3] *VŠB – Technical University of Ostrava, FMST, Regional Materials Science and Technology Centre, Ostrava, Czech Republic, katerina.skotnicova@vsb.cz*

[4] *VŠB – Technical University of Ostrava, Department of Physics, Ostrava, Czech Republic, ondrej.zivotsky@vsb.cz*

[5] *Institute of Low Temperature and Structure Research, Polish Academy of Sciences, Wroclaw, Poland, y.koshkidko@intibs.ru*

[6] *Physical Department, Moscow State University, Moscow, Russia, ba.ginzburg@physics.msu.ru*

ABSTRACT

We have investigated the effect of HoH_2 hydride addition on the hysteresis loop parameters of sintered Pr-Fe-Ti-Al-Cu-B magnets. The magnets were prepared by traditional powder metallurgy technology, and 3 wt.% HoH_2 was added to the powder at the fine-milling stage. The magnets exhibited a monotonic increase in all hysteretic parameters with decreasing temperature down to 4.2 K. The coercive force and maximum energy product at 295 K (4.2 K) were 1344 (5402) kA/m and 221 (336) kJ/m^3, respectively. The structure of the magnets was studied in detail by scanning electron microscopy and energy dispersive X-ray spectroscopy, which demonstrated the formation of the so-called "core-shell" structure, which is assumed to favor the marked improvement in the hysteretic properties of the samples analyzed. The surface domain structure was measured in the directions perpendicular and parallel to the magnet texture using magnetic force microscopy. The data obtained indicated fine labyrinth-like and strip domain patterns in the directions perpendicular and parallel to the magnet texture, with an average domain width of 1.2–1.8 μm.

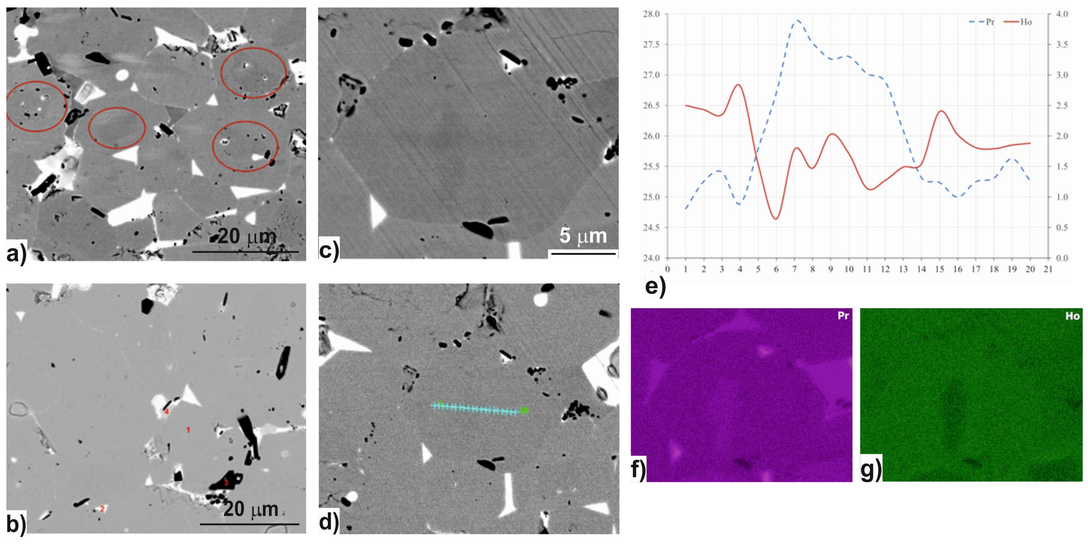
SAMPLE PREPARATION

- **Alloy** containing (wt.%) Pr-33, Ti-0.9, Al-0.3, Cu-0.15, B- 1.3, Fe-balance prepared by induction melting in an argon atmosphere, cast into a water-cooled copper mold, and subjected to hydrogen decrepitation using a bell-type furnace. The hydrogenation process performed during heating to 473 K for 1.5-2 h where it was held for 1 h. The furnace chamber then washed with nitrogen gas, and the alloy subjected to furnace cooling to room temperature (RT).
- **Powders** of the $Pr_2Fe_{14}B$-based alloy and its mixture with 3 wt.% HoH_2 prepared by milling in an isopropyl alcohol medium using a vibratory ball mill.
- **Magnet blanks** prepared by compacting at a force of 300 kg/cm^2 using a hydraulic press with a loading rate no faster than 10 mm/s, texturing magnetic field of 1.6 T applied perpendicular to the pressing force direction. The blanks dried and sintered at 1375 K for 1 h (single-cycle technological operation) and subjected to low-temperature treatment at 775 K for 1 h using vacuum resistance furnaces.

EXPERIMENTAL TECHNIQUES

- **XRD (X-Ray Diffraction)** – Ultima IV (Rugaku, Japan) diffractometer equipped with a D/teX detector and Cu Kα radiation, 2θ range 3° - 100°, evaluation – Rietveld structure refinement method by using the PHAN and PHAN% software
- **SEM (Scanning Electron Microscopy)** – QUANTA 450 FEG equipped with an energy dispersive X-ray (EDX) APOLLO X analyzer
- **Hysteresisgraph** – MH-50, close magnetic circuit, maximal magnetic field 2 MA/m (2.5 T), room temperature bulk magnetic properties of prepared magnets - 30 mm in diameter and 10 mm high
- **VSM (Vibrating-Sample Magnetometer)** – magnetic properties of small piece of magnet (mass about 100 mg) at low temperatures (LT) 4.2 K - 295 K, magnetic field up to 7.2 MA/m (9 T)
- **MFM (Magnetic Force Microscopy)** – Solver Pro EC (NT MDT), observation of magnetic domains in directions parallel and perpendicular to the magnet texture

MICROSTRUCTURE



(a,b) Scanning electron microscopy (SEM) images of the (Pr, Ho)$_2$Fe$_{14}$B-based magnet obtained from two different points on the surface in the back-scattering electron (BSE) mode; (a) circles indicate the cores of grains; (b) digits correspond to **electron microprobe analysis (EMA)** points (table below). (c,d) Amplified SEM images of 2-14-1 phase grain; (e) EMA data along the scanning line highlighted in subplot (d); (f) Pr distribution map; (g) Ho distribution map.

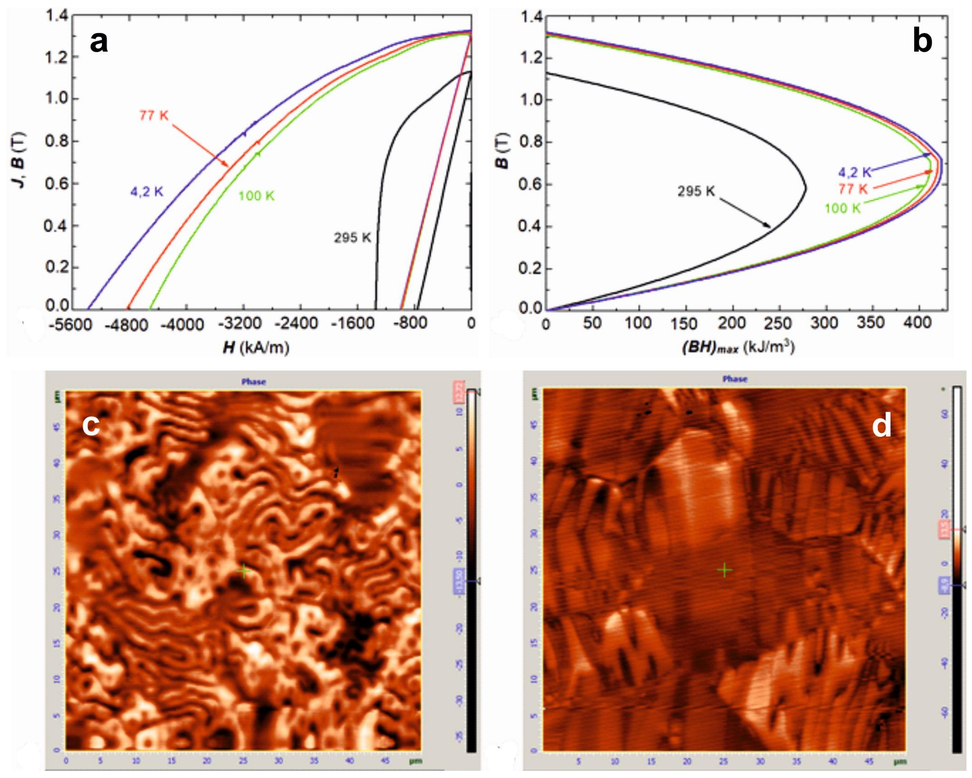
EMA data (wt.%) of the (Pr, Ho)-Fe-B magnet. *Compositions vary.

Phase	Phase/Element	O	Al	Ti	Pr	Nd	Fe	Ho	Cu
	Area_mean	3.1	0.6	1.3	30.1	1.5	57.1	4.6	0.6
(Pr, Ho)$_2$Fe$_{14}$B (2-14-1)	Phase_1_mean	1.6	0.5	0.3	24.5	1.3	64.8	5.2	0.6
(Pr, Ho)$_{rich}$	Phase_2_1*	2.6	0.4	0.2	80.7	2.0	4.3	0.3	8.4
	Phase_2_2	1.9	0.4	0.4	78.3	2.5	4.7	1.9	8.8
	Phase_2_3	2.0	0.4	1.1	71.9	2.2	11.1	1.5	8.4
	Phase_2_4	2.4	0.5	0.3	78.4	2.6	4.5	1.4	8.5
	Phase_2_5	2.2	0.4	0.2	78.7	2.2	6.3	0.7	8.3
	Phase_2_6	2.0	0.4	0.4	72.9	2.6	11.5	1.2	7.9
Ti-based	Phase_3_1	1.5	0.1	76.1	11.9	0.6	7.4	1.1	0.8
	Phase_3_2	1.1	0.2	70.9	10.5	0.4	13.1	1.5	1.2
	Phase_3_3	1.4	0.5	45.1	16.5	0.9	31.8	2.5	0.5
(Pr, Ho)$_2$O$_3$ oxide phases	Phase_4*	16.4	0.1	0.4	71.0	2.6	6.5	2.3	0.3

Variations in the lattice parameters of the main magnetic phase (Pr, Ho)$_2$Fe$_{14}$B following hydrogenation and dehydrogenation in the course of preparation of the magnets.

Powder state	a (nm)	c (nm)	a^2×c (nm^3)
$Pr_2Fe_{14}BH_x$	0.8873(6)	1.2352(14)	0.97261
$Pr_2Fe_{14}BH_x$ + HoH_2	0.8877(6)	1.2354(16)	0.97351
Dehydrogenated $Pr_2Fe_{14}BH_x$	0.8793(8)	1.226(3)	0.94790
Dehydrogenated $Pr_2Fe_{14}BH_x$ + HoH_2	0.8798(7)	1.227(3)	0.95019

MAGNETIC PROPERTIES



(a,b) Magnetic properties of the (Pr, Ho)-Fe-B magnet measured using the VSM: (a) *J*–*H* and *B*–*H* dependences (plotted with allowance for the demagnetizing factor) measured at different temperatures; (b) maximum magnetic energy product $(BH)_{max}$. (c,d) Magnetic force microscopy (MFM) data obtained from the (Pr, Ho)-Fe-B magnet in directions (c) perpendicular (pole surface) and (d) parallel (lateral surface) to the magnetic texture.

(Left) Magnetic parameters of (Pr, Ho)-Fe-B magnet at LT obtained using the VSM. (Right) RT bulk magnetic parameters of Pr-Fe-B and (Pr, Ho)-Fe-B magnets; B_r – remanence of the magnetic flux density; $_jH_c$ – coercivity of the magnetic polarization; $_bH_c$ – coercivity of the *B*–*H* curve; H_k – magnetic field determined at 0.9 × B_r; $(BH)_{max}$ – maximum energy product.

T (K)	B_r (T)	$_jH_c$ (kA/m)	$_bH_c$ (kA/m)	$(BH)_{max}$ (kJ/m^3)
295	1.13	1344	764	221
100	1.31	4527	971	327
77	1.32	4837	987	332
4.2	1.33	5402	1011	336

Sample	B_r (T)	$_jH_c$ (kA/m)	$_bH_c$ (kA/m)	H_k (kA/m)	$(BH)_{max}$ (kJ/m^3)
Pr-Fe-B	1.09	1404	702	335	191
(Pr, Ho)-Fe-B	1.11	1516	806	893	223

ACKNOWLEDGEMENTS

This paper was created within the project LTARF18031 "Development of physico-chemical and engineering foundations for the initiation of innovative resources-economy technology of high-power and high-coercivity (Nd,R)-Fe-B (R = Pr, Tb, Dy, Ho) low-REM permanent magnets" and within the project No. 14.616.21.0093 (the unique identification number RFMEFI61618X0093).