SIMULATION OF MAGNETIZATION REVERSAL AND HYSTERETIC LOOPS OF $(Nd,Pr)_2Fe_{14}B$ MAGNETS AT LOW TEMPERATURES

[1]Vladimir P. MENUSHENKOV, [1]Alexander G. SAVCHENKO, [1]Vasily L. STOLYAROV, [1]Mikhail V. ZHELEZNYI, [2]Natalia B. KOLCHUGINA, [2]Nikolay A. DORMIDONTOV, [3]Kateřina SKOTNITCOVÁ, [4]Yurij S. KOSHKID'KO, [4]Jacek ĆWIK

[1]National Research Technological University "MISiS", Moscow, Russian Federation, menushenkov@gmail.com

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

[3]Vysoka Skola Banska-Technical University, Ostrava, Czech Republic, EU katerina.skotnicova@vsb.cz

[4]Institute of Low Temperature and Structure Research, Polish Academy of Sciences, Wroclaw, Poland, EU yurec@mail.ru

https://doi.org/10.37904/metal.2019.953

Abstract

In the present work, the computer simulation of hysteresis loops of $(Nd_{1-x}Pr_x)_2Fe_{14}B$ compounds with different Pr contents was performed for a temperature range of 300-5 K. The simulation results suggest that the alloying of $Nd_2Fe_{14}B$ with Pr (1) leads to reducing the negative effect of the spin-reorientation phase transition on the residual magnetization of the compound $(Nd_{1-x}Pr_x)_2Fe_{14}B$, (2) reduces the slope of the demagnetization curve making the hysteresis loop more rectangular, (3) does not have a noticeable effect on the maximum value of the coercive force, shifting its maximum to lower temperatures as the Pr content increases. Thus, the alloying of $Nd_2Fe_{14}B$-based compositions with Pr can increase the temperature stability of the hysteresis characteristics of permanent magnets and ensures their operation at low temperatures without loss of magnetic properties.

Keywords: Nd-Pr-Fe-B magnets, coercive force, magnetic properties, simulation of hysteretic loops

1. INTRODUCTION

The excellent magnetic properties of the sintered Nd-Fe-B magnets result from the high both magnetization and magnetocrystalline anisotropy of the tetragonal $Nd_2Fe_{14}B$ phase. At room temperature, this phase is characterized by the uniaxial anisotropy; the easy magnetization axis coincides with the *c* axis. Below T_{sr} = 135 K, the preferred magnetization direction starts to deviate from the *c*-axis direction to form an easy-axis cone; at 4 K, the cone angle reaches 30° [1-5]. As a result, below T_{sr}, a deflection appears in the magnetization reversal curves, which leads to a decrease in the maximum energy product and the remanence of magnets [4-5]. The $Pr_2Fe_{14}B$ compound at room temperature has magnetic properties close to those of $Nd_2Fe_{14}B$ and has no spin-reorientation transition [6]. It can be assumed that the partial substitution of Pr for Nd in the $Nd_2Fe_{14}B$ phase can lead to the significant improvement in the hysteresis characteristics of the (Nd,Pr)-Fe-B magnets at low temperatures [7-8]. In this regard, the task of developing a model and algorithm for calculating hysteresis loops of $(Nd_{1-x}Pr_x)_2Fe_{14}B$ compounds with different Pr contents and analyzing changes in their hysteresis and energy characteristics is very promising and relevant.

2. CALCULATION PROCEDURE

In a crystalline ferromagnet, the vector of the total magnetization M_s is spontaneously established along certain crystallographic directions, which are called the easy magnetization axes (EMAs). In magnets with uniaxial hexagonal or tetragonal lattices, anisotropy energy (E_a) is expressed by the formula [7]:

$E_a = -K_1(\cos^2\varphi) - K_2(\cos^4\varphi)$ (1)

where K_1 and K_2 are the first and second anisotropy constants, respectively, and φ is the angle made by the tetragonal axis and vector M_S.

When a magnetic field H is applied, the orientation of vector M_S is also determined by the energy of the magnetic field. As a result, the magnetization vector M_S will be oriented so that the sum of energies $E = E_a + E_H$ is minimal.

In terms of our model, we consider a single crystal in the form of a flat disk located along a certain crystal plane of the lattice. When the magnetic field is applied in this plane, the magnetization vector always lies in it. For simplicity, we assume that the sample magnetization is homogeneous and the sample is single-domain.

The developed model and the algorithm used for calculating the hysteresis loops (program "Hysteresis") of the ferromagnetic $(Nd_{1-x}Prx)_2Fe_{14}B$-based compound with a uniaxial tetragonal lattice operate with the following parameters: π is the angle made by Z axis (EMA) and vector M_S; α is the angle made by an arbitrary plane and the X axis, a field is applied in this plane, and angles π are counted; and θ is the angle made by direction of field H and Z axis (EMA). The anisotropy energy is measured in units of K_1 and is always measured from its minimum value; in the graphs, it is plotted on logarithmic coordinates.

An example, a screen shot of the "Hysteresis" program is given in **Figure 1**.

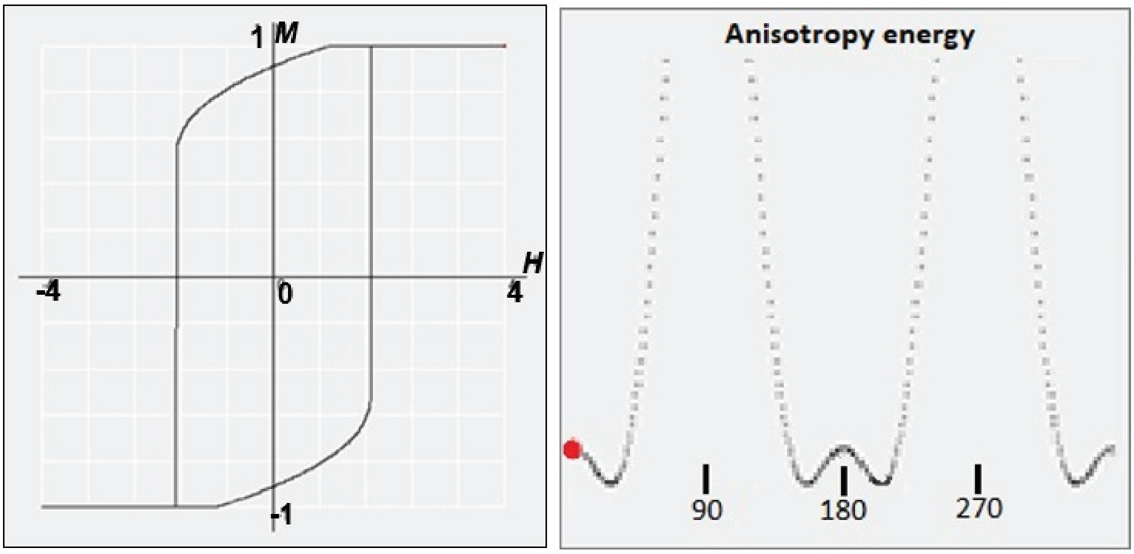


Figure 1 Screen shot of results obtained with the program "Hysteresis": α = 0°; the magnetic field H forms the angle θ = 0° with the Z axis, the field magnitude varies from 0 to 4 at a step of 0.0001. The limiting value (saturation, I_S) reaches magnetization in a sufficiently high field. During magnetization reversal, the sign of magnetization changes at $H = H_c$ (coercive force). Remanence, I_R =0.908 I_S, coercive force H_C =1.681, loop area P = 5.930; anisotropy type - hexagonal; anisotropy constant K < 1; $K_2/abs(K_1)$ = 2.9; Plane orientation α = 0. Magnetic field parameters: angle made by field H and <100> axis and field amplitude 4.

To calculate the values of anisotropy constants of the $(Nd_{1-x}Prx)_2Fe_{14}B$ compounds (with $0 \leq x \leq 1$) at different temperatures, the numerical values of K_1 and K_2, obtained from the data given in **Figure 2** [6], were determined and the following relationships were used:

$K_1(T,x) = (1 - x)\, K_1\,(T)_{Nd_2Fe_{14}B} + K_1\,(T)_{Pr_2Fe_{14}B}$ (2)

$K_2(T,x) = (1 - x)\, K_2\,(T)_{Nd_2Fe_{14}B} + K_2\,(T)_{Pr_2Fe_{14}B}$ (3)

The ratio of the absolute values of the K_1 and K_2 constants and the θ angle were calculated by formulas:

$$\frac{K_2(T,x)}{|K_1(T,x)|}$$

at $K_1(T,x) \leq 0$ $\theta(T,x) = \arcsin\left(\sqrt{-\frac{K_{1(T,x)}}{2\,K_{2(T,x)}}}\right)$ (4)

at $K_1(T,x) > 0$ $\theta(T,x) = 0$.

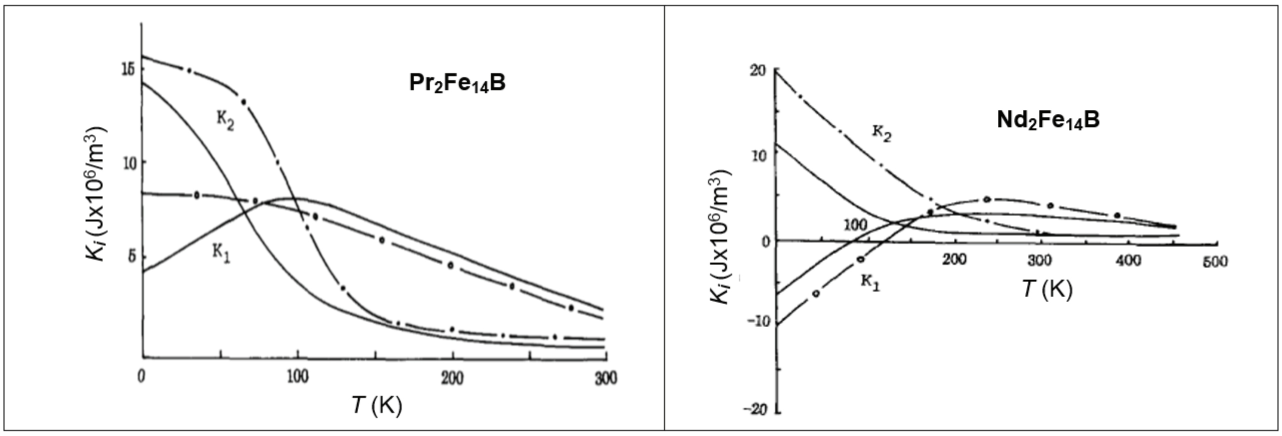


Figure 2 Temperature dependences of the magnetocrystalline anisotropy constants K_1 and K_2 of the (a) $Pr_2Fe_{14}B$ and (b) $Nd_2Fe_{14}B$(b) compounds [6]

3. RESULTS AND DISCUSSION

Figure 3 shows the temperature dependence of magnetocrystalline anisotropy constants K_1 and K_2 [6], the ratio of the absolute values of anisotropy constants K_1/K_2, and temperature dependence of the angle θ made by the magnetization axis and the *c* axis of the compounds $(Nd_{1-x}Pr_x)_2Fe_{14}B$ compounds with $x = 0$ (a), $x = 0.5$ (b) and $x = 0.75$ (c) calculated by the Eqs. (2-4)**.**

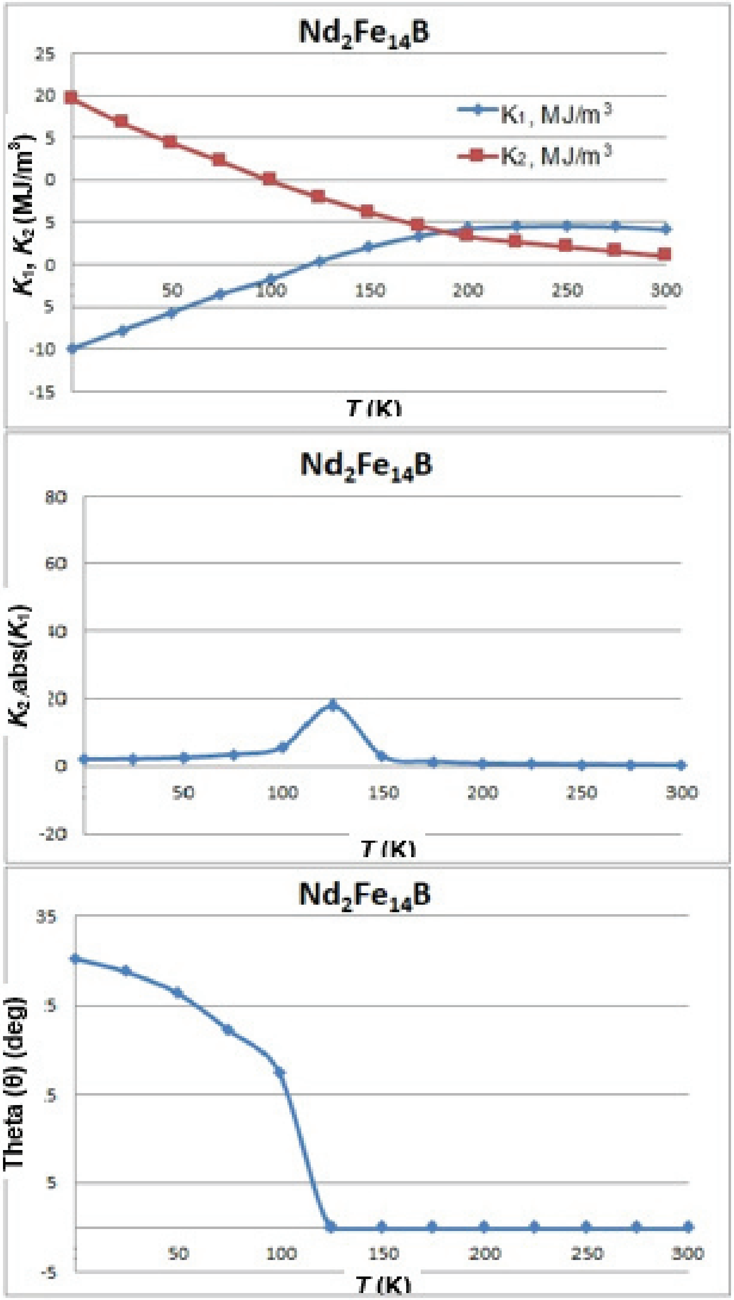
Figure 4 presents the temperature dependences of the residual magnetization of the $(Nd_{1-x}Pr_x)_2Fe_{14}B$ compounds with $x = 0, 0.25, 0.5, 0.75, 0.9$, and 1. It can be seen from the data given in **Figure 4** that, as the temperature decreases from 300 K to the temperature of the spin-reorientation temperature (T_{sr} = 135 K), the value of *Ir*/*Is* remains unchanged and equal to 1 for all compositions $(Nd_{1-x}Pr_x)_2Fe_{14}B$. This means that the magnetiztion vector after saturation remains parallel to the E_A as the magnetizing field decreases to zero.

As is seen from **Figure 4**, as the temperature decreases from 300 K to the spin-reorientation phase transition temperature (T_{sr} = 135 K), the I_r/I_s value remains constant and equal to 1 for all compositions $(Nd_{1-x}Pr_x)_2Fe_{14}B$. This means that the magnetization vector after saturation remains parallel to the E_A with decreasing magnetizing field to zero. An analysis of the I_r/I_s (T) dependences in **Figure 4** and the shape of the hysteresis loops of the compounds $(Nd_{1-x}Pr_x)_2Fe_{14}B$ shows that, as praseodymium atoms substitute for neodymium atoms, in the temperature range from 135 K to 4 K the decrease in I_r/I_s value gradually slows down and at x = 0.75, 0.9, 1, the value of Ir/Is remains constant and equal to 1.

Figure 5 presents the temperature dependences of the relative magnitude of the coercive force of the compound $(Nd_{1-x}Pr_x)_2Fe_{14}B$ with x = 0, 0.25, 0.5, 0.75, 0.9, and 1.

As can be seen from **Figure 5**, the relative magnitude of the coercive force of the compound $Nd_2Fe_{14}B$ reaches the maximum value H_c/H_a (≡ $2K_1/I_s$) = 8.3 at the temperature of the spin-reorientation phase transition (T_{sr} =

135 K). As the Pr content in the $(Nd_{0,75}Pr_{0,25})_2Fe_{14}B$ and $(Nd_{0.5}Pr_{0.5})_2Fe_{14}B$ compounds increases, the maximum specific coercive force remains unchanged, but the maximum itself shifts to lower temperatures 90 and 25 K, respectively. A further increase in the Pr content in the compounds, $(Nd_{0.25}Pr_{0.75})_2Fe_{14}B$, $(Nd_{0.1}Pr_{0.9})_2Fe_{14}B$ and $Pr_2Fe_{14}B$, leads to the disappearance of the maximum in the temperature dependences $H_c/H_a(T)$ and, as the temperature decreases from 135 K to 4 K, a gradual decrease in the value of H_c/H_a to 4, 2.5, and 2 takes place, respectively, i.e. the influence of the constant K_2 is gradually waning.



(a)

Figure 3 (a) Temperature dependences of the magnetocrystalline anisotropy constants K_1 and K_2 [6], the ratio of absolute values of anisotropy constants K_1/K_2, and the temperature dependence of the θ angle made by the easy magnetization axis and c axis of the $(Nd_{1-x}Pr_x)_2Fe_{14}B$ compound with $x = 0$.

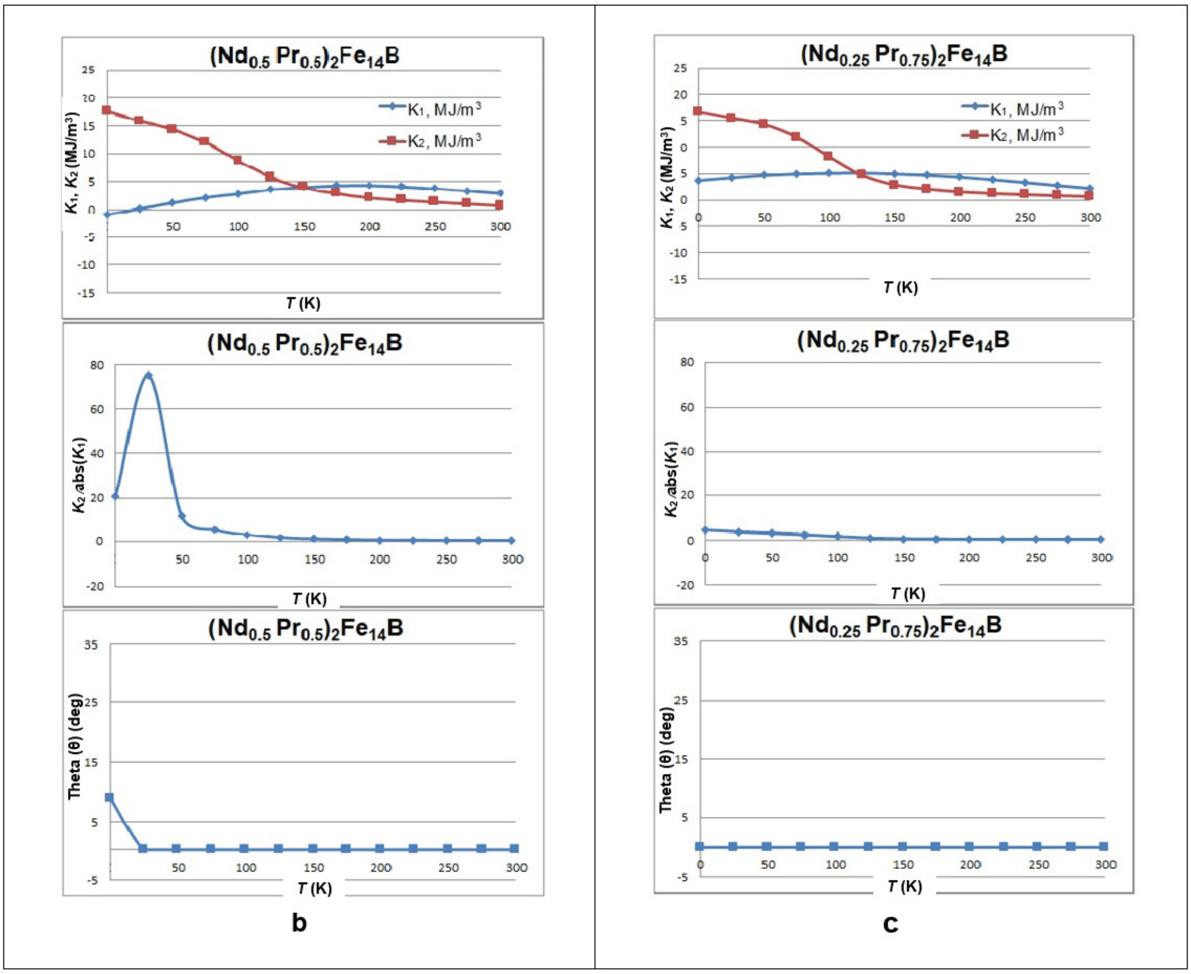


Figure 3 (b, c) Temperature dependences of the magnetocrystalline anisotropy constants K_1 and K_2 [6], the ratio of absolute values of anisotropy constants K_1/K_2, and the temperature dependence of the θ angle made by the easy magnetization axis and c axis of the $(Nd_{1-x}Pr_x)_2Fe_{14}B$ compound with x = 0 (a), x = 0.5 (b) and x = 0.75 (c)

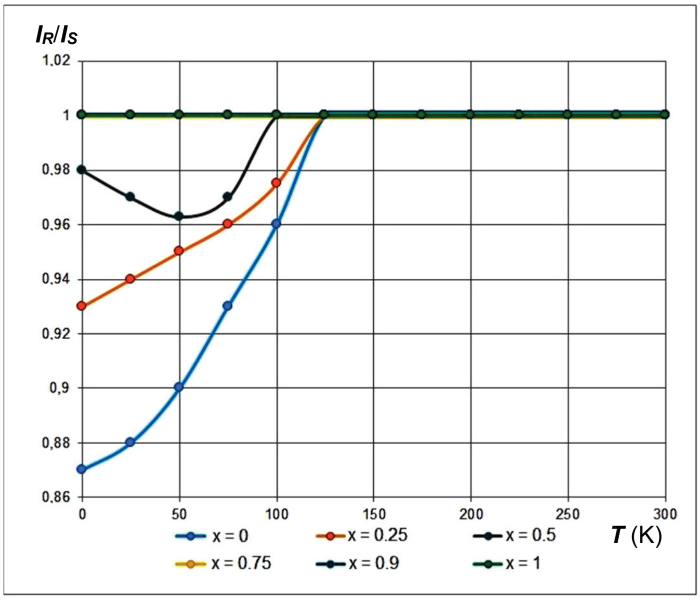


Figure 4 Temperature dependences of the specific residual magnetization of the $(Nd_{1-x}Pr_x)_2Fe_{14}B$ compound with x = 0, 0.25, 0.5, 0.75, 0.9 and 1

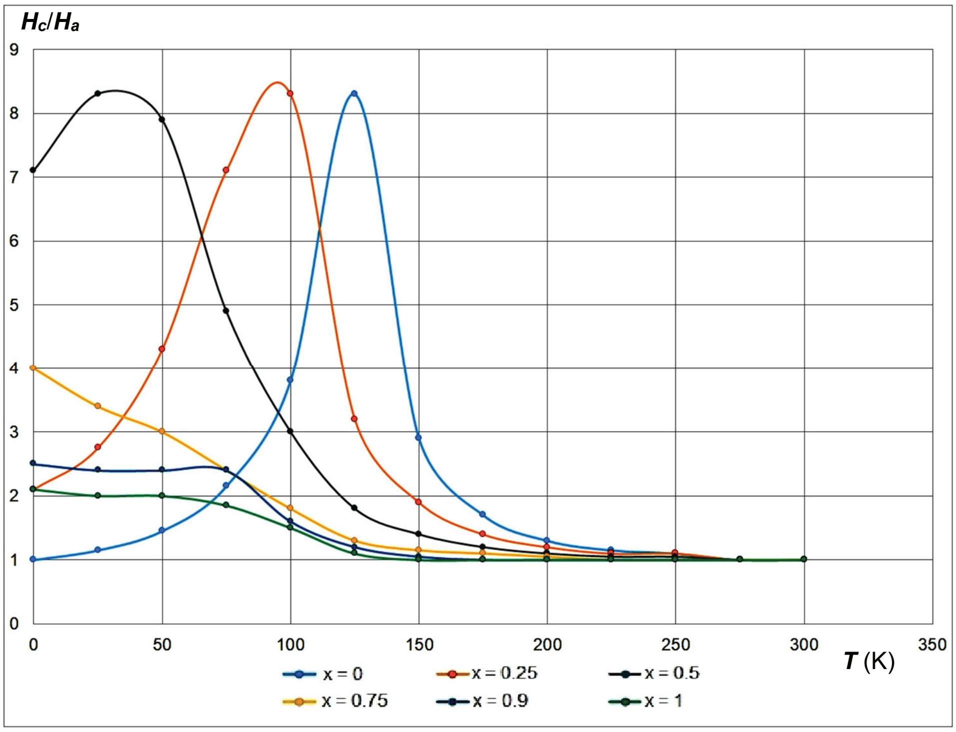


Figure 5 Temperature dependence of the coercive force of $(Nd_{1-x}Pr_x)_2Fe_{14}B$ with x = 0, 0.25, 0.5, 0.75, 0.9, 1

4. CONCLUSIONS

The model and algorithm for calculating the hysteresis loops of ferromagnets, which is developed in the framework of this work, allows us to obtain important data on the effect of the crystal lattice type (uniaxial, cubic), parameters of crystallographic anisotropy and temperature dependences of anisotropy constants on the characteristics of hysteresis loops (anisotropy field, residual magnetization, hysteresis loop shape, coercive force) in a wide temperature range. The simulation results indicate that the alloying of the $Nd_2Fe_{14}B$ compound with praseodymium firstly, leads to a decrease in the negative effect of the spin-reorientation phase transition on the residual magnetization of the compound $(Nd_{1-x}Pr_x)_2Fe_{14}B$, secondly, reduces the slope of the demagnetization curve and makes the back of the hysteresis loop more rectangular and, thirdly, does not have a noticeable effect on the maximum value of the coercive force, shifting this maximum to lower temperatures, as the praseodymium content increases. Thus, the obtained results show that the alloying of $Nd_2Fe_{14}B$-based compositions with praseodymium can increase the temperature stability of the hysteresis characteristics of permanent magnets prepared from the alloys and ensure the operation of the magnets at low temperatures without marked losses of magnetic properties.

ACKNOWLEDGEMENTS

This study was supported by the Ministry of Science and Higher Education of the Russian Federation, agreement no. 14.616.21.0093 (unique identification number RFMEFI61618X0093) and the Ministry of Education, Youth, and Sports of the Czech Republic, project no. LTARF18031.

REFERENCES

[1] TOKUHARA, K., OHTSU, Y., ONO, F., YAMADA, O., SAGAWA, M. and MATSUURA, Y. Magnetization and torque measurements on $Nd_2Fe_{14}B$ single crystals. *Solid State Communication*. 1985. vol. 56, no. 4, pp. 333-336.

[2] BARANOV, N.V., SINITSYN, E.V., IGNATYEV, E.A. and ANDREEV, S.V. Magnetization reversal of Nd-Fe-B sintered magnets at low temperatures. *Journal of Magnetism and Magnetic Materials.* 1994. vol. 130, pp. 133-137.

[3] CHRISTODOULOU, C.N., BOLTICH, E.B. and MASSALSKI, T.B. Spin-reorientation effect on magnetization - application to $Nd_2Fe_{14}B$. *Journal of Magnetism and Magnetic Materials.* 1989. vol. 81, pp. 209-220.

[4] DIEZ-JIMENEZ, E., PEREZ-DIAZ, J.L., CANEPA, F. and FERDEGHINI, C. Invariance of the magnetization axis under spin reorientation transitions in polycrystalline magnets of $Nd_2Fe_{14}B$. *Journal of Applied Physics.* 2012. vol. 112, p. 063918.

[5] DIEZ-JIMENEZ, E., PEREZ-DIAZ, J.L., FERDEGHINI, C., CANEPA, F., BERNINI, C., CRISTACHE, C., SANCHEZ-GARCIA-CASARRUBIOS, J., VALIENTE-BLANCO, I., RUIZ-NAVAS, E.M. and MARTINEZ-ROJAS, J.A. Magnetic and morphological characterization of $Nd_2Fe_{14}B$ magnets with different quality grades at low temperature 5-300 K. *Journal of Magnetism and Magnetic Materials*. 2018. vol. 451, pp. 549-553.

[6] JIANG, S., LI, H., XU, J. and GAO, R. Research on the origin of magneto-crystalline anisotropy in $R_2Fe_{14}B$ compounds. *Journal of Magnetism and Magnetic Materials*. 1994. vol. 136, pp. 294-300.

[7] LIU, Z.W. and DAVIES, H.A. Sub-ambient magnetic properties of nanophase Nd/Pr-Fe-B based hard magnetic alloys. *Journal of Alloys and Compounds*. 2006. vol. 424 pp. 255-261.

[8] OTANI, Y., MIYAJIMA, H. and CHIKAZUMI, S. Demagnetization process of sintered $R_{17}Fe_{83-x}B_x$ magnets (R=Nd or Pr). *Journal of Applied Physics.* 1986. vol. 63, no. 8, pp. 3316-3318.