Study of reversible changes of the coercive force of Nd-Fe-B-based alloys subjected to cyclic heat treatments

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Abstract. Results of the investigation of the reversibility of the magnetic properties of two Nd(Dy)-Fe(Co)-B alloys during cyclic treatments at temperatures of 1050-700°C and subsequent rapid cooling and annealing at 500-600°C are reported. It is shown that, after a significant decrease in the coercive force for magnetization ($H_c$), which is caused by heating to high temperatures, its recovery begins after 5-min aging at 500°C; the complete restoration of $H_c$ takes place after 1-2 h exposure.

1. Introduction

The Nd-Fe-B system alloys are widely used owing to the combination of high magnetic properties and fairly low costs of the alloy components. However, the mechanism of formation of structural components, which allow one to reach the high coercive force magnitudes, is ambiguously interpreted and remains unclear. In particular, the causes for the substantial changes of the coercive force in the course of heat treatments under different conditions are unknown.

It is known that, after realization of complete manufacturing cycle, a short-time heating of magnets to a high temperature leads to the abrupt degradation of magnetic properties, in particular, a decrease in the coercive force. Low-temperature annealings allow one to restore the magnetic properties. An analysis of literature data shows that the phenomenon of reversibility of the coercive force magnitude during cyclic heat treatments is typical of almost all kinds of industrial magnets (ticonal, magnico-type, SmCo₅, Sm-Co-Cu-Fe-Zr) [1-3]. Taking into account the substantial differences in structural transformations, origin of the high-coercivity state, and magnetization-reversal mechanisms, and as well as the fact that the coercive force is the structure-sensitive properties, it is of importance to study in detail the phenomenon of the coercive force reversibility for the Nd-Fe-B alloys in order to understand the regularities of formation of magnetic properties of these magnets.

The present study is aimed at the effect of temperature and time of annealing on the process of reversible restoration of the coercive force magnitude of Nd-Fe-B alloys after its decrease during heating to high temperatures.

2. Experimental

Table 1 shows the compositions of studied alloys.

Metallographic studies of the alloys were performed using a CarlZeissAxio Lab.A1 optical microscope. The magnetic properties of magnets were measured on a MN-50 hysteresisgraph; the magnetization reversal curve and a protocol of properties were recorded automatically. The error of
measurements is ± 2%. The chemical composition of alloys was determined by X-ray fluorescent analysis.

### Table 1 Chemical composition of alloys A and B (wt %).

|   |   |   |   |   |   |   |   |   |   |
|---|---|---|---|---|---|---|---|---|---|
|   | Nd | Pr | Dy | Tb | Co | Ti | Al | Cu | B  |
| A | 28.8 | 1.0 | 1.7 | 2.3 | 5.7 | 0.7 | 0.3 | 0.1 | 1.2 |
| B | 23.1 | 3.6 | 5.6 | -  | -  | -  | 0.5 | 0.2 | 1.0 |
|   |   |   |   |   |   |   |   |   | Balance |

Samples of magnets were prepared by traditional powder metallurgy technique, which includes milling of alloy in isopropyl alcohol (to an average particle size of 3 µm), subsequent pressing in a transverse magnetic field of to 15 kOe, sintering at T=1100°C, and low-temperature treatment with 1-h holding at T=500°C.

Samples of alloy A were heated to a temperature of 1050°C, held for 1 h, and subsequently cooled to room temperature at a rate of 20°C/min. These samples were subjected to annealing in a temperature range of 500 – 600°C with different holdings at these temperatures. Alloy B was used to study the reversibility of magnetic properties after heat treatments at temperatures of 700 – 1000°C.

Subsequently, the heating to high temperatures, which leads to the decrease in the coercive force, is referred as “deteriorating”; the heat treatments resulting in the increase in coercive force are the “restoring”.

### 3. Results and discussion

Table 2 show changes of the magnetic properties of alloy A samples after “deteriorating” during heating to 1050°C and “restoring” at 500°C for 5, 30, 60, and 120 min. It should be noted that, in terms of the present study, the coercive force for magnetization ($H_c$) is the most interesting property since just this characteristic determines the physics of magnetization reversal process and the nature of its reversibility.

### Table 2 Changes of the magnetic properties of alloy A during heat treatment at 1050°C + 500°C.

| Properties | Starting state | Temperature and time of annealing |
|------------|----------------|----------------------------------|
|            | 1050°C 1 h | 500°C 5 min | 500°C 30 min | 500°C 1 h | 500°C 2 h |
| $H_c$, kA/m | 1384 | 948 | 1011 | 1097 | 1262 | 1516 |
| $B_r$, kA/m | 860 | 837 | 826 | 840 | 875 | 790 |
| $B_r$, T | 1,11 | 1,11 | 1,09 | 1,11 | 1,17 | 1,09 |
| $(BH)_{max}$, kJ/m³ | 237 | 239 | 225 | 233 | 256 | 211 |

It follows from Table 2 that the coercive force of alloy A not only completely restores after 2-h annealing at 500°C but also increases as compared to the value obtained after traditional heat treatment. Table 3 shows changes in the magnetic properties of alloy A after "deteriorating" during heating to 1050°C and "restoring" at 600°C for 5, 30, 60, and 120 min. As is seen from Table 3, the "restoring" of properties during holding at 600°C is incomplete, and after 2-h annealing the degradation the properties takes place.

It should be noted that the coercive force for magnetization demonstrates the more substantial reversible changes. Variations of the other properties are not so significant, and no marked regularities are observed.

It is of interest to monitor the reversibility of the coercive force at different temperatures of "deteriorating" annealing. Table 4 shows changes of the magnetic properties of alloy B subjected to "deteriorating" heating to 700-1000°C for 1 h and subsequent "restoring" 1-h annealing at 500°C.
Table 3 Changes of the magnetic properties of alloy A during heat treatment at 1050°C + 600°C.

| Properties | Starting state | Temperature and time of annealing |
|------------|----------------|----------------------------------|
|            |                | 1050°C 600°C                      |
|            |                | 1 h 5 min 30 min 1 h 2 h          |
| \(H_c\), kA/m | 1384           | 981 1047 1148 1146 1149          |
| \(B_b\), kA/m | 860            | 849 777 827 840 654              |
| \(B_r\), T   | 1,11           | 1,10 1,09 1,15 1,18 1,03         |
| (BH)\(_{\text{max}}\), kJ/m³ | 237            | 233 217 239 254 157              |

Table 4 Changes of the magnetic properties of alloy B in the course of "deteriorating" annealing at 700-1000°C and "restoring" 1-h annealing at 500°C.

| Properties | Starting state | Temperatures of "deteriorating" and "restoring" annealings |
|------------|----------------|-----------------------------------------------------------|
|            |                | 700°C 500°C 800°C 500°C 900°C 500°C 1000°C 500°C          |
| \(H_c\), kA/m | 1755           | 1328 1760 1367 1764 1343 1722 1247 1689                  |
| \(B_b\), kA/m | 890            | 903 881 922 896 917 905 953 891                           |
| \(B_r\), T   | 1,22           | 1,23 1,22 1,22 1,22 1,22 1,23 1,26 1,25                 |
| (BH)\(_{\text{max}}\), kJ/m³ | 275            | 272 263 284 270 283 277 304 273                          |

It is of interest to note that, whatever the "deteriorating" annealing temperature, the reversible change of the coercive force is the same in value; with allowance for the measurement error, the change is 30%. According to metallographic and X-ray diffraction data, no changes in the phase and structural states of the alloys were found.

4. Conclusions
The independence of the reversible change of the coercive force on the heating temperature allows us to conclude that the reversible changes of the coercive force are not related to changes of the phase composition of magnet and processes occurred at grain boundaries. In the case of grain-boundary processes, the coercive force should be dependent on the "deteriorating" annealing temperature. It is likely that the phenomenon of "deteriorating" and "restoring" changes of magnetic properties of the alloys is related to the atomic ordering of the main magnetic phase since the reversibility assumes the short-range diffusion.

5. References
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