Structure and magnetic properties of Nd-(Fe,Co)-B hard magnetic powders with and without Cu doping

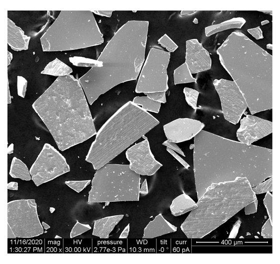
V.Yu. Samardak[1*], A.V. Ognev, Kharitonov V.N., Belov A.A., Shichalin O.O., Papynov E.K., Samardak A.S
[1]Institute of High Technologies and Advanced Materials,
Far Eastern Federal University, Vladivostok 690922, Russia
*e-mail: vadimsamardak@gmail.com

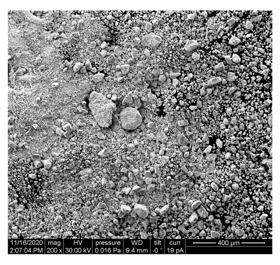
MOTIVATION

The development of modern technology requires strong and compact permanent magnets, which are used in electric drives and generators. Research has been focused on new methods for synthesizing and forming the optimal microstructure since the discovery of Nd2Fe14B hard magnetic alloys. Nd-Fe-B magnets require magnetic powders for their synthesis. Their production is the most important technological task that requires an understanding of the processes of structural transformation during grinding and the development of methods for stabilizing magnetic properties. The aim of this work is to investigate the influence of the Cu additive on the structure and magnetic properties of powders, which was obtained from the melt-spun of ribbons of the Nd-(Fe,Co)-B alloy.

EXPERIMENTAL

Nd-(Fe,Co)-B powders	XRD	Phases volume (%)	Parameter a (Å)	Parameter c(Å)	CSR (nm)	Deformation (%)
Initial powder	2-14-1 α-Fe	99 ~1	8.789 2.847	12.197 -	26 -	0.1 -
Powder after milling (600 rpm, 6 h)	2-14-1 α-Fe	99 ~1	8.790 2.847	12.199 -	32 -	0.3 -
Powder after milling with 1 wt% Cu (700 rpm, 6 h)	2-14-1 α-Fe	99 ~1	8.790 2.847	12.199 -	30 -	0.4 -





Hard magnetic powders were obtained by ball milling of the commercial rapidly quenched Nd-(Fe,Co)-B alloy (product No. 04821610, Jovi International, China) using a Tencan XQM-0.4A planetary ball mill (China), regime: 700 rpm in one 6-hour cycle in an inert gas atmosphere (argon) and acetone. Powder samples with Cu incorporation were synthesized by adding 1 wt% of Cu powder in the initial alloy before ball milling. Furthermore, powders without additives and with 1 wt% Cu incorporation were annealed at 750 °C for 10 minutes for thermal stability investigation. Such heat treatment is traditionally used for the compaction of nanocrystalline powders via the hot-pressing technique.

MORPHOLOGY

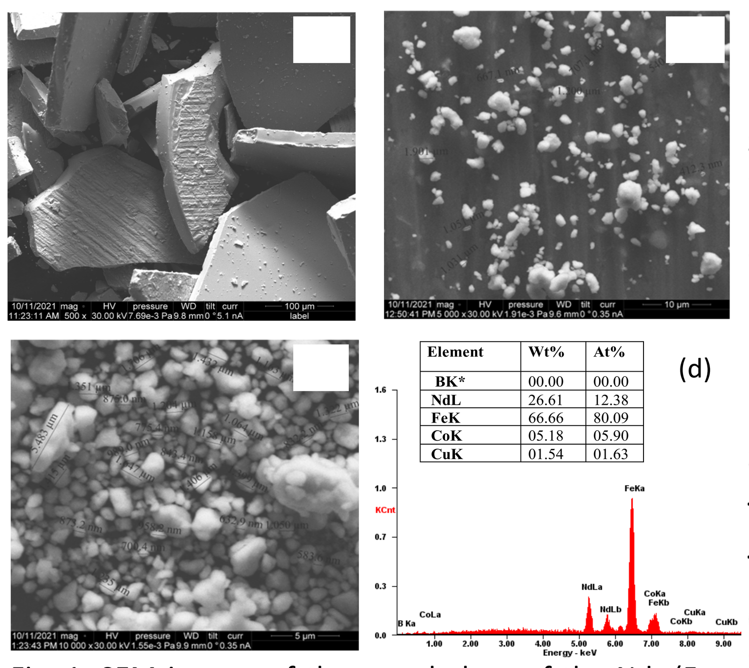


Fig. 1. SEM images of the morphology of the Nd- (Fe, Co) -B alloy particles in the initial state (a), after being milled without copper (b) and morphology (c) and results of the EDX analysis (d) of the Nd- (Fe, Co) -B alloy with the 1 wt% Cu additive

Figure 1 (a) shows SEM images of the initial powder of Nd-(Fe,Co)-B alloy. The powder particles are plate-shaped with sharp edges. The thickness of the particles is about 20 µm with an average in-plane size of about 370 µm. Figures (b) and © show SEM images of the powder of the Nd-(Fe,Co)-B alloy after milling. These powders contain agglomerates, as well as individual particles of different sizes and irregular shapes. The average grain size is 2 µm. Figure (d) shows the data of the EDX results for the powder alloy Nd-(Fe,Co)-B with the 1 wt% Cu additive. The content of Nd, Fe, and Co does not change for all samples. The composition of individual particles of the Nd-(Fe,Co)-B powder alloy differs significantly from the average composition of the large area.

XRD ANALYSIS

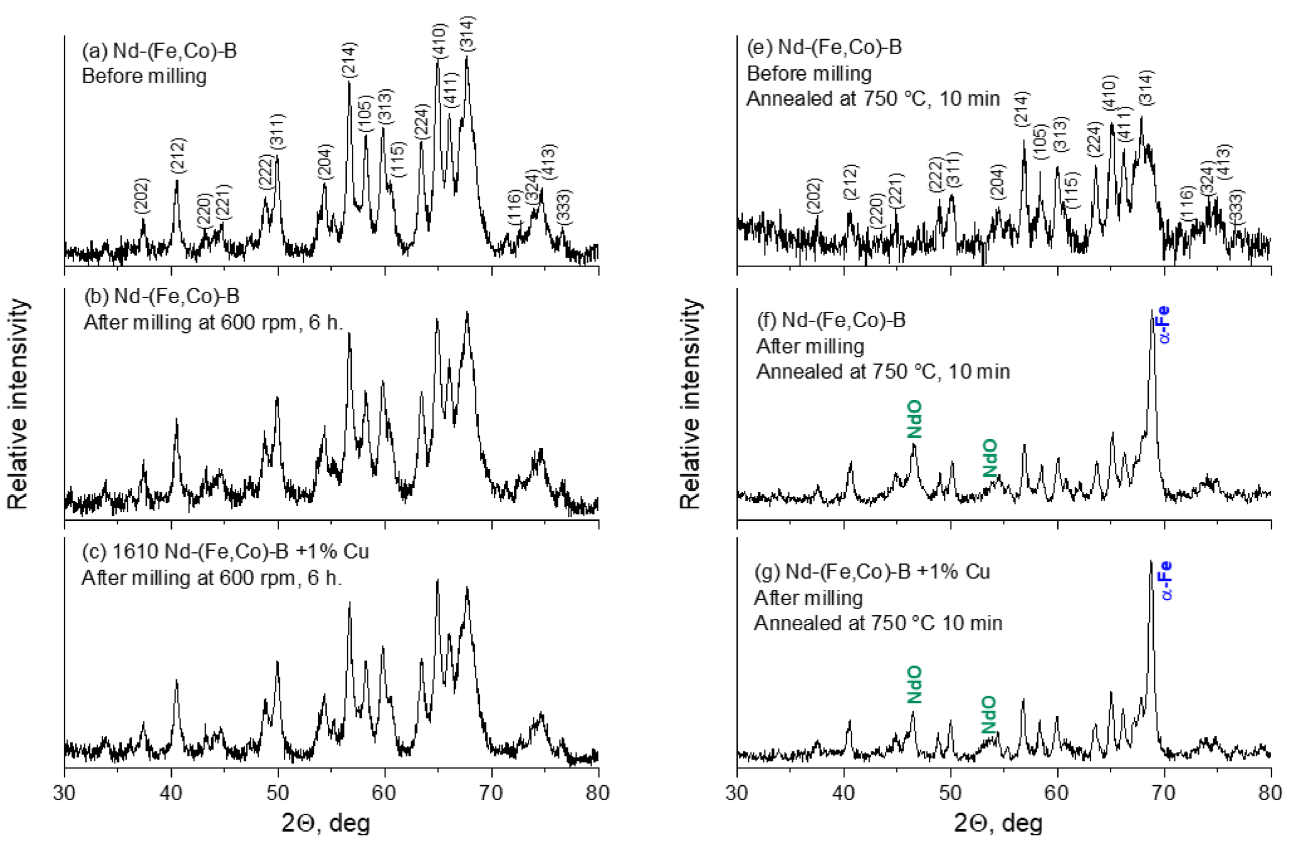


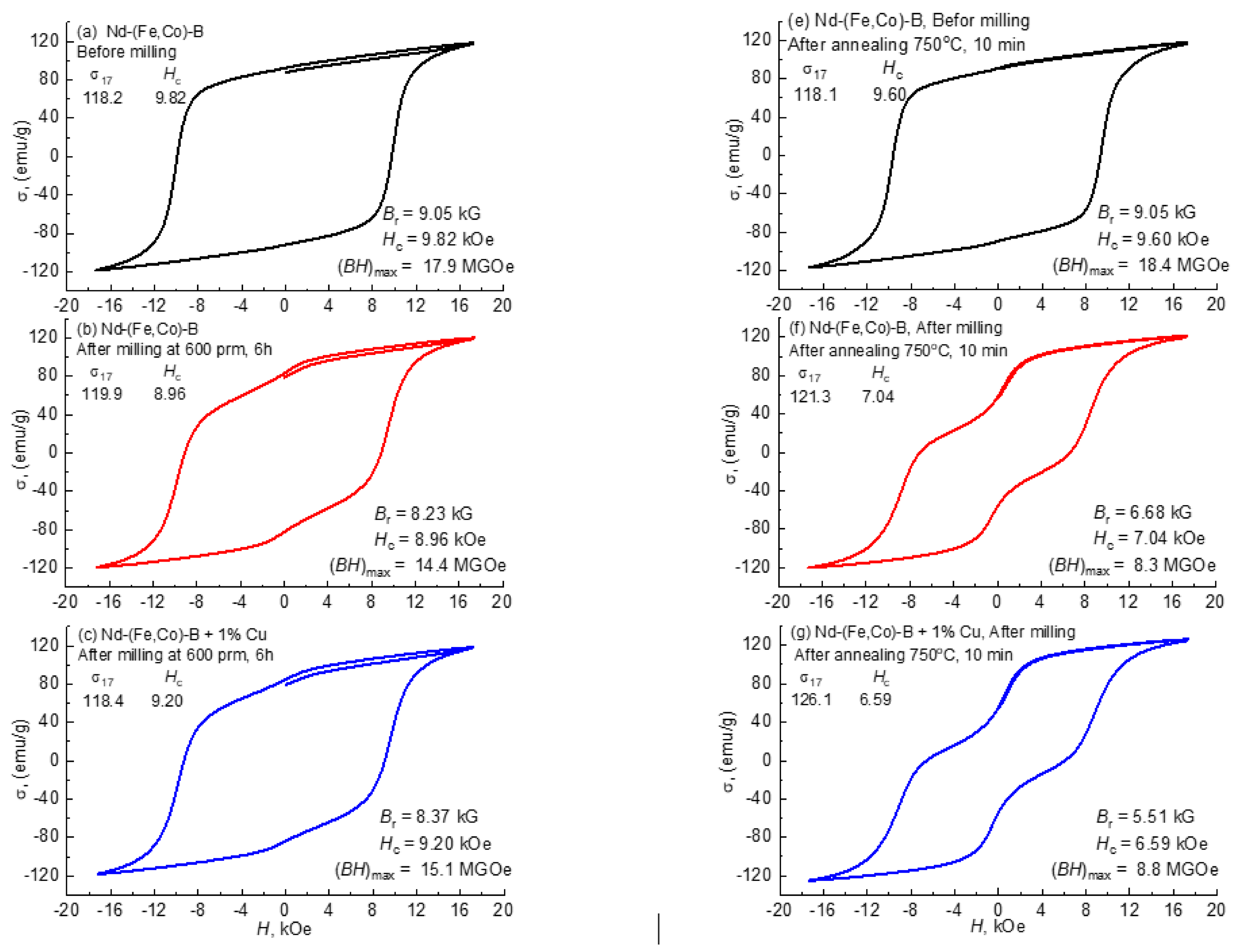
Fig. 2. X-ray diffraction patterns of the original powder of the Nd-(Fe,Co)-B alloy and its crushed form, as well as after alloying with 1 wt.% Cu

The XRD of the initial powder of Nd-(Fe,Co)-B alloy and its milled form, as well as after doping with 1 wt% of Cu, are shown in Figure 1. The composition of the powder phase and the lattice parameters of the phases are presented in Table 1. The main phase of the powders is the tetragonal phase Nd2Fe14B (2-14-1) with the spatial symmetry group P42/mnm. Miller indices of the phase are shown in Figure 1a. The lines of the (2-14-1) phase are very broad, which allows for an estimate of the coherent scattering regions (CSRs) size of about 30 nm. The grain deformation of the initial powder is 0.1%, and after milling it increases several times.

The XRD diffractograms of the initial powder of the Nd alloy (Fe, Co) -B before and after annealing qualitatively coincide. However, the XRD diffractograms of milled powders before and after annealing are significantly different. Intensive lines of NdO oxide and α-phase Fe-Co phases appear in XRD diffractograms for milled powders after annealing. This explains the decrease in Hc for the powders.

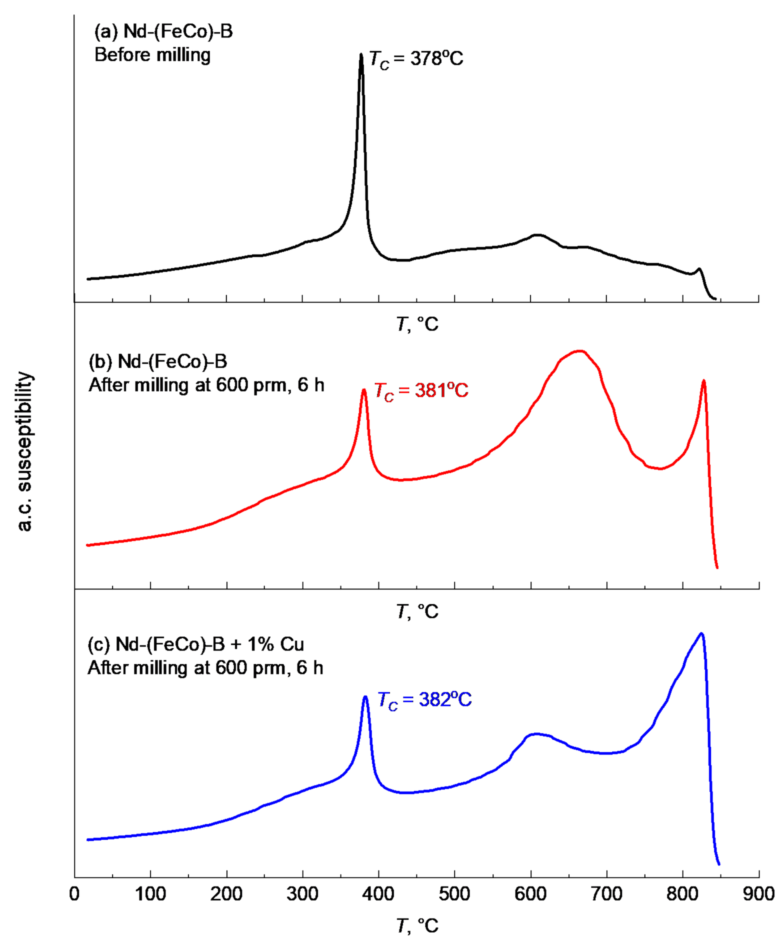
MAGNETIC PROPERTIES

Figure 3 shows the magnetic hysteresis loops for Nd-(Fe,Co)-B alloy powders measured by a vibrating magnetometer. Each figure shows the values of the maximum specific magnetization σ17 measured in a magnetizing field with a strength of 17 kOe, the coercive force Hc, the residual induction Br, and the maximum energy product (BH)max that was calculated assuming that the alloy density ρ is 7.87 g/cm3. To get an idea of the influence of high annealing temperatures on the powder coercivity of the Nd-(Fe,Co) -B alloy and the same milled powders without and with the 1 wt% Cu additive, these powders were annealed at 750°C for 10 minutes.



Figures 3 e-g shows hysteresis loops of annealed powders. The magnetic characteristics of the initial powders did not change significantly after annealing (Fig. 2a, e). The coercivity decreased considerably for milled powders, indicating significant powder contamination after milling and increased contributions of surface effects. The coercivity and energy product are higher for samples with 1 wt.% Cu additive than for those without Cu. This trend persists even after annealing. The improvement in magnetic characteristics for Nd-(FeCo)-B alloy powders with the 1 wt% Cu additive is probably due to the segregation of Cu in the intergranular phase region and the stabilization of the magnetically stiff phase.

TEMPERATURE DEPENDENCE



The Tc peaks are observed approximately 70°C above the Curie temperature of the Nd2Fe14B ternary phase (Tc = 312°C).

After the phase transition of Nd2(Fe, Co)14B to the paramagnetic state, the ac susceptibility does not return to zero.

The contribution of the α-phase to the susceptibility increases from the initial powder to the ground powders

CONCLUSIONS

- It has been established that the initial rapidly quenched Nd-(Fe,Co)-B alloy powder has a composition of approximately 28Nd, 66Fe, 5Co, 1B (wt.%)
- The main phase of the alloy is the structure Nd2Fe14B
- The high coercivity of the powder up to 9.2 kOe is due to the presence of nanocrystalline grains about 30 nm in size inside the powder particles with an average size of about 370 µm
- The particle size of the powder is reduced to an average size of approximately 2 µm after grinding
- The coercive force of crushed powders slightly decreases, but remains at a fairly high level (8.96 kOe and 9.20 kOe)
- Adding 1 wt. % Cu leads to an increase in the coefficient of coercive force compared to the powder without Cu. For annealed samples, we observe the opposite picture - alloyed powders show degraded magnetic properties.

ACKNOWLEDGEMENTS

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