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Solid-phase synthesis and properties of a large-grain high-temperature superconductor based on thulium and neodymium

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Due to relative simplicity and availability of the necessary equipment, the solid-phase synthesis is the most popular way to produce new superconducting materials. This paper describes the solid-phase synthesis with the formation of sufficiently large superconducting granules. Bulk superconducting samples with the 1-2-3 structure based on thulium and neodymium with the grain size of up to 0.1 mm have been synthesized. An increase in the crystallite grain size leads to a significant increase in the trapped magnetic flux.

Keywords: granular superconductor, magnetic hysteresis, magnetization, pinning, RE-123, solid state synthesis.

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Increasing the critical current density in high-temperature superconductors (HTSCs) is the goal of many studies [1,2]. In most experimental works, the effect of composition and structure modifications on the HTSC critical parameters and pinning is studied on polycrystalline materials [3–6] obtained by solid-phase synthesis. The popularity of solid-phase synthesis is associated with the ease of modifying the samplesćomposition and properties. Solid-phase synthesis of compounds having the 1-2-3 structure provides polycrystals with the granule size of about $1-10\,\mu$m [4,5]. The polycrystalline structure promotes additional phenomena affecting the magnetic and transport properties of superconductors [7,8]. The possibility of using HTSC single crystals for exploratory research is limited because their synthesis is a complex resource— and time-consuming process. The search for new methods for simplifying the synthesis of HTSC crystals and large granules is an urgent task. One of the newest methods being developed for producing HTSC crystals is growth on several seed crystals [9]. The method of growing the superconducting phase at the boundaries of non-superconducting phases was used for polycrystalline HTSCs [10,11]. In the above-mentioned studies, the superconducting phase was grown in the contacting boundaries of related materials. A similar procedure is also used to create a high-quality junction of two crystals RE–Ba–Cu–O [12] (RE is a rare earth element). This paper proposes an evolution of the method of growth on seed crystals aimed at increasing the granule size in the solid phase synthesis. We assumed that the granule growth may be optimized by using RE–Ba–Cu–O materials with different peritectic temperatures and by choosing an appropriate synthesis temperature.

We chose for the synthesis $NdBa_2Cu_3O_{7-d}$ with the peritectic temperature of 1068°C and $TmBa_2Cu_3O_{7-d}$ with the peritectic temperature of 980°C. At the first stage, polycrystalline $NdBa_2Cu_3O_{7-d}$ (Nd123) and $TmBa_2Cu_3O_{7-d}$ (Tm123) were obtained from Nd_2O_3, Tm_2O_3, $BaCO_3$ and CuO powders via the conventional ceramic technology for the solid-phase synthesis. The synthesis included three stages of grinding, pressing and annealing at temperatures below the peritectic temperature of the compounds to be synthesized. At the second stage, the synthesized materials were mixed in the proportion of 20% of $NdBa_2Cu_3O_{7-d}$ and 80% of $TmBa_2Cu_3O_{7-d}$ (NdTm123). The component concentrations were chosen so that the refractory phase volume was slightly less than the percolation threshold, which provides a space for the growth of granules. The mixture was pressed, and the obtained pellets were annealed at 980°C for an hour. Such an annealing leads to the formation of a liquid phase (peritectic decomposition of $TmBa_2Cu_3O_{7-d}$) and growth of crystallites from this liquid phase on seeds having a higher peritectic temperature. After annealing, the furnace with samples was cooled at a rate of 0.5°C/min.

The synthesized samples were studied by scanning electron microscopy with a Hitachi TM4000Plus microscope. Micrographs of synthesized NdTm123 (Fig. 1, *a*) demonstrate individual granules about 0.1 mm in linear size surrounded by clusters of small granules about $3\,\mu$m in size. Dark areas in the micrograph represent the material pores. The granulesḿean size is $11 \pm 1\,\mu$m. The largest granules had the sizes of up to $135\,\mu$m, cross-section area of up to $1.4 \cdot 10^{-8}$ m^2, and volume of up to $1.3 \cdot 10^{-12}$ m^3. Fig. 1, *b* presents a histogram of the granule size distribution. The inset to this figure shows what fractions of the total sample volume are occupied by granules of different sizes. The obtained granule size distribution is almost lognormal (see solid line in Fig. 1, *b*). The number of granules 0.1 mm in size and larger is only 0.4%. However, the volume fraction of those large granules reaches 36% (see the inset to Fig. 1, *b*).

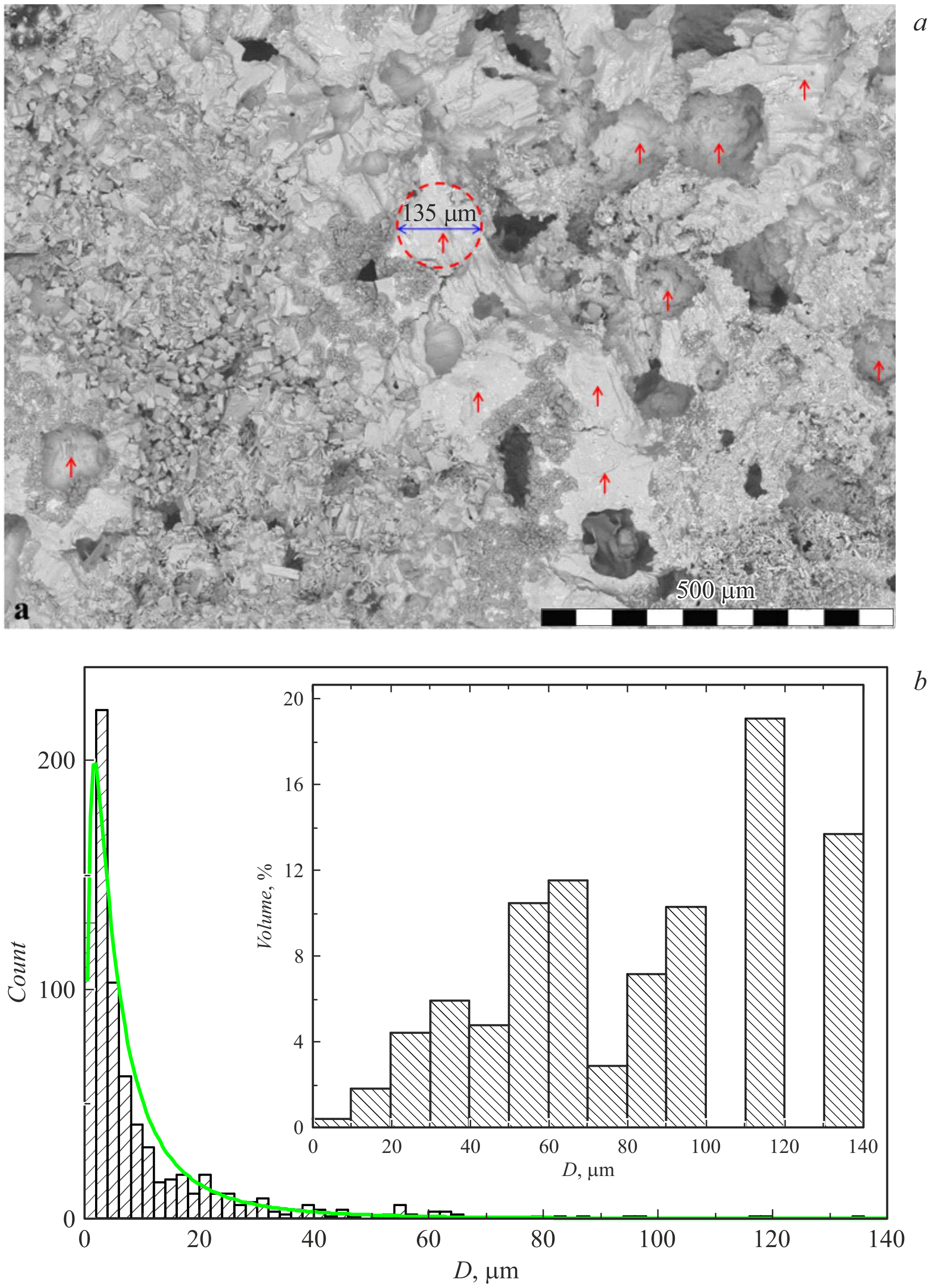

Figure 1. *a* — micrograph of the NdTm123 sample. Arrows indicate granules 0.1ḿm in size and larger. In the image, the size of one of the largest granules is indicated. *b* — the granule size distribution. The solid line represents the lognormal distribution. The inset demonstrates the relative volume occupied by different-size granules.

The samplesḿagnetization was measured with a Quantum Design PPMS-9T vibration magnetometer. Critical temperature T_c determined from the beginning of the magnetization decrease with decreasing temperature was 88 K for Nd123, 92 K for Tm123 and 92 K for NdTm123 (see the inset to Fig. 2, it b). Magnetization hysteresis loops for samples Nd123 (curve *1*), Tm123 (curve *2*) and NdTm123 (curve *3*) measured at 4.2 K (Fig. 2 , *a*) and 80 K (Fig. 2, *b*) exhibit paramagnetic and superconducting contributions, as is characteristic of RE–Ba–Cu– O [5]. The obtained loops are considerably different. First of all, there is observed a significant magnetization width ΔM for the NdTm123 sample hysteresis loop, which exceeds ΔM for the Nd123 and Tm123 samples by 2–4 times for the loops at 4.2 K. The ΔM value for samples Nd123 and Tm123 at 80 K quickly decreases with increasing external

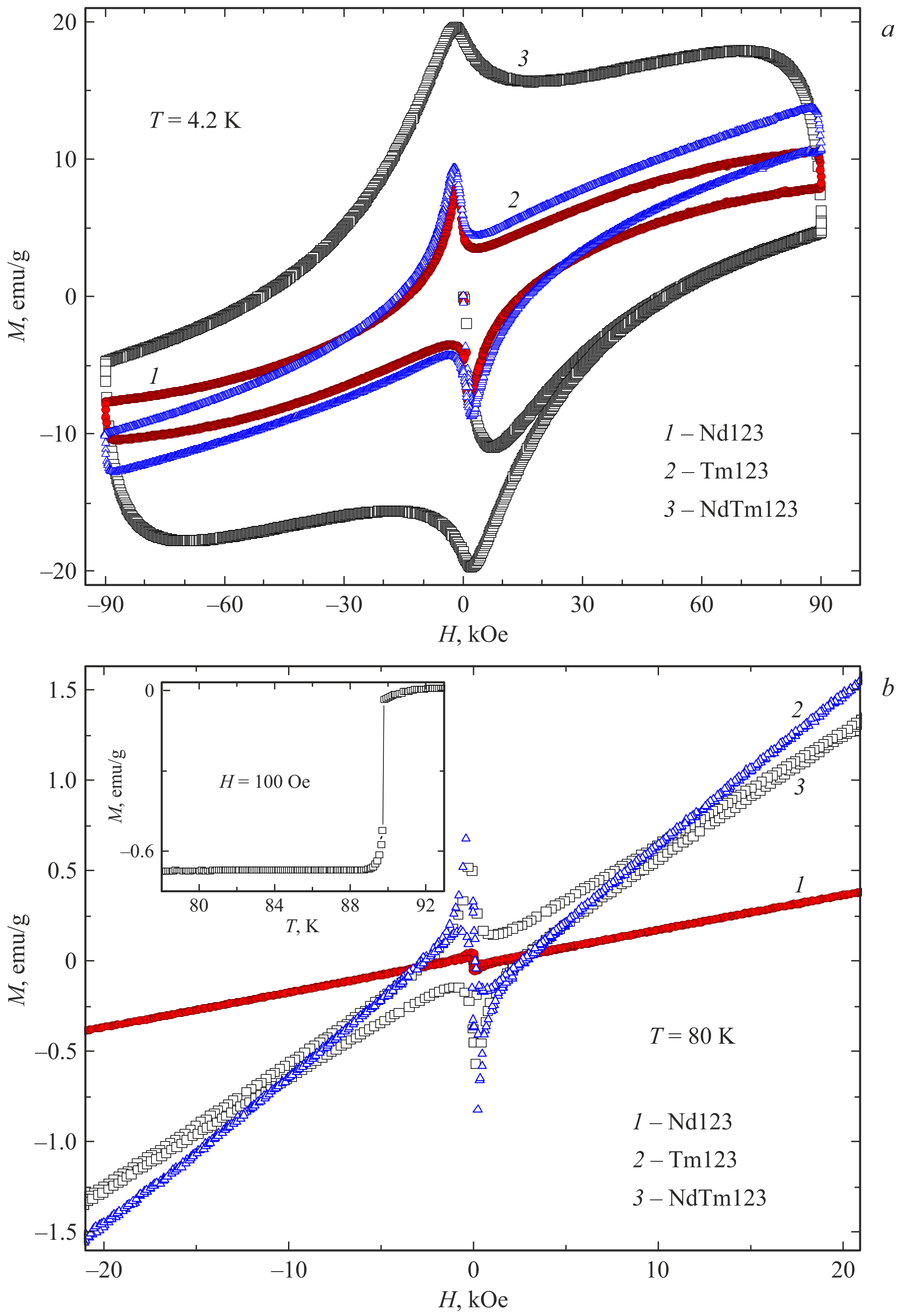

Figure 2. Sample magnetization hysteresis loops at the temperatures of 4.2 (*a*) and 80 K (*b*. The inset presents the temperature dependence of the NdTm123 magnetization in the field of 100 Oe, which was measured after cooling in the zero field.

field H. When the external magnetic field is higher than irreversibility field $H_{irr} \approx 7$ kOe, the field dependences of magnetization for these samples become reversible, and $\Delta M = 0$. For the NdTm123 sample, the ΔM decrease with increasing field is slower, and the irreversibility field is much higher ($H_{irr} \approx 40$ kOe). The NdTm123 trapped magnetic field corresponding to the remanence at $T = 4.2$ K is 0.15 T, while the trapped magnetic flux is $\sim 10^{-7}$ Wb.

Known ΔM values for polycrystalline superconductors fabricated using the conventional ceramic technology are also lower than those for NdTm123 [3,5]. The ΔM value for polycrystalline superconductors depends both on the critical current density and on the average granule size [13]. Partial replacement of the Y positions with rare earth elements does not cause an increase in the critical current density [5,14]. Thus, the main source of high values of ΔM and trapped magnetic flux in NdTm123 is an increase by 3–4 times in the average granule size in the synthesized superconductor, which is caused by growing of large granules on seed grains of $NdBa_2Cu_3O_{7-d}$ from the $TmBa_2Cu_3O_{7-d}$ liquid phase.

In conclusion, note that we have developed a simple method for obtaining superconducting granules larger than $100\,\mu$m. For this purpose, the Tm123/ Nd123 mixture was annealed at a temperature between the peritectic temperatures of the mixture initial components. In the synthesized material 20% $NdBa_2Cu_3O_{7-d}$ + 80% $TmBa_2Cu_3O_{7-d}$, the volume fraction of granules about 0.1 mm in size reached 36%. Due to the presence of large granules, the hysteresis loop of the obtained material magnetization exhibited for polycrystalline superconductors record values of magnetization width ΔM and trapped magnetic flux.

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Conflict of interests

The authors declare that they have no conflict of interests.

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