

Manipulating compositional stoichiometry in high-temperature superconductors to enhance critical currents under high magnetic fields

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The influence of the deviation in the target chemical composition from the stoichiometric ratios of yttrium-barium-copper oxide (YBCO) $Y:Ba:Cu = 1:2:3$ on the properties of coated conductors fabricated through pulsed laser deposition was investigated. An optimal target composition was empirically established, facilitating both a reduction in anisotropy and an enhancement of the critical current at liquid helium temperatures and under intense magnetic fields.

Keywords: YBCO, coated conductor, pinning centers, anisotropy, target.

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1. Introduction

The technology of the second generation high-temperature superconductors (HTS-2 or coated conductors) consists of epitaxial synthesis of a chemical compound belonging to the $(RE)Ba_2Cu_3O_{7-x}$ (REBCO, where RE is a rare earth element) family on textured buffer layers covering a flexible metallic tape substrate. This technology has the potential to revolutionize many industries, including electric motors and generators for aviation and space, energy storage devices, medical imaging devices, among others. The prospect of employing HTS-2 within the magnetic systems of compact thermonuclear fusion (TF) devices is particularly significant. The essential relevance for TF is the capability to produce high magnetic field ~ 20 T and higher, which subsequently results in the reduction of the operational temperature of the coated conductors to near liquid helium temperatures. Previous studies [1,2] have demonstrated that the maximal critical current (I_c) at this temperature level can be achieved through the utilization of yttrium-barium-copper oxide (YBCO). Further improvement of the current-carrying capacity of YBCO can be achieved by introducing artificial pinning centers.

It is well-established that, in contrast to the oxygen [3,4], deviations in the stoichiometric indices of the Y, Ba and Cu target from the nominal values leads to a significant change in the coated conductor characteristics. An excess of Ba is considered undesirable, since the resulting phases are unstable and negatively affect the performance properties [5]. The effect of Cu-excess remains insufficiently examined, and the available results are contradictory. It has

been confirmed that Cu-excess contributes to the generation of relatively large ($> 1 \mu m$) CuO particles on the surface of the HTS layer, which are nearly absent within the bulk of $REBa_2Cu_3O_{7-x}$ layer. Consequently, these particles cannot act as pinning centers. Nevertheless, a positive correlation has been reported between the appearance of CuO and an enhancement in I_c [5]. A moderate Y-excess leads to the formation of finely dispersed of Y_2O_3 particles, which act as effective pinning centers. This is a rather attractive way to increase the critical current, since it is relatively easy to scale up [2]. In addition, by sequentially sputtering multiple targets, it is possible to induce the precipitation of non-superconducting phase Y_2BaCuO_5 (Y211) particles within the HTS layer, which also contributes to an increase in the critical current [6].

In this paper, we present the preliminary findings from investigations aimed at identifying the optimal deviations from the target stoichiometry to enhance the current-carrying capacity of coated conductors in high magnetic fields.

2. Experiment

The 4-mm-wide coated conductors were fabricated on the experimental production line at the Kurchatov Institute [7,8]. The HTS layer was synthesized via pulsed laser deposition (PLD) on a steel substrate with YSZ (yttrium-stabilized zirconium oxide) and CeO_2 buffer layers. The targets were produced at JSC VNIINM through oxalate co-precipitation from a mixture of Y, Ba, and Cu nitric acid solutions,

microwave drying, pyrolysis, pressing, and high-temperature treatment. The deviation from stoichiometry was created in several ways. In the first target, the initial solutions for co-precipitation were taken with a deficiency of barium so that the gross formula of the mixture corresponded to $Y:Ba:Cu = 1:1.8:3$. In the second target, an additional incorporation of Y_2O_3 was implemented enhance the Y-excess, resulting in a molar fraction of Y_2O_3 equating to 8%. Finally, in the third target, the ceramic powder of YBCO of stoichiometric composition was mixed with additionally prepared powder of Y_2BaCuO_5 (Y-211), ensuring that the molar fraction of Y-211 also constituted 8%.

The critical current was determined using the criterion of $1 \mu V/cm$, achieved in the current-voltage characteristics (CVC) during standard transport measurements using the four-terminal method. The critical current angular dependences $I_c(\theta)$ were recorded in the so-called maximum Lorentz force configuration: the magnetic field is always orthogonal to the transport current. The angle θ was quantified relative to the tape plane. The angular dependences were studied under conditions of liquid nitrogen. The critical current was also measured in liquid helium (4.2 K) with the external magnetic field oriented normally to the tape. The critical current at intermediate temperatures (20 and 40 K) was estimated using the magnetization loop width measured on a PPMS vibration magnetometer. The proportionality coefficient between the loop width and the critical current was determined using transport measurements at 4.2 K.

The sample synthesized utilizing the barium-deficient target ($YB_{1.8}CO$) is designated as „387-1“, and has a HTS layer thickness of $1.4 \mu m$ and a critical current in the self-field (at 77.4 K) of 96 A. The sample made from the target with the addition of yttrium oxide ($YB_{1.8}CO + Y_2O_3$) is „466-4“, the thickness is $1.6 \mu m$, and the critical current is 133 A. The target with the addition of the Y211 phase (YBCO + Y211) corresponds to the sample „282“, with a thickness of $1.6 \mu m$ and a critical current of 91 A.

3. Results and discussion

Figure 1, *a* illustrates the critical current angular dependences in an external magnetic field of 0.5 T. All angular dependences are well described within the framework of the anisotropic pinning model [9,10]. Note that for sample 387-1, a pronounced asymmetry in the angular dependence $I_c(\theta)$, is observed, the explanation of which is given in [10]. The most favorable orientation of the field from the point of view of maximizing the critical current is in the plane of the tape ($\theta = 0^\circ$). In this geometric arrangement, the critical current decreases in the sequence 466-1 \rightarrow 282 \rightarrow 387-1. Excluding the minor asymmetry of sample 387-1, the least favorable orientation is the normal direction of the field, wherein the sequence of decreasing critical current reverses to 387-1 \rightarrow 282 \rightarrow 466-1.

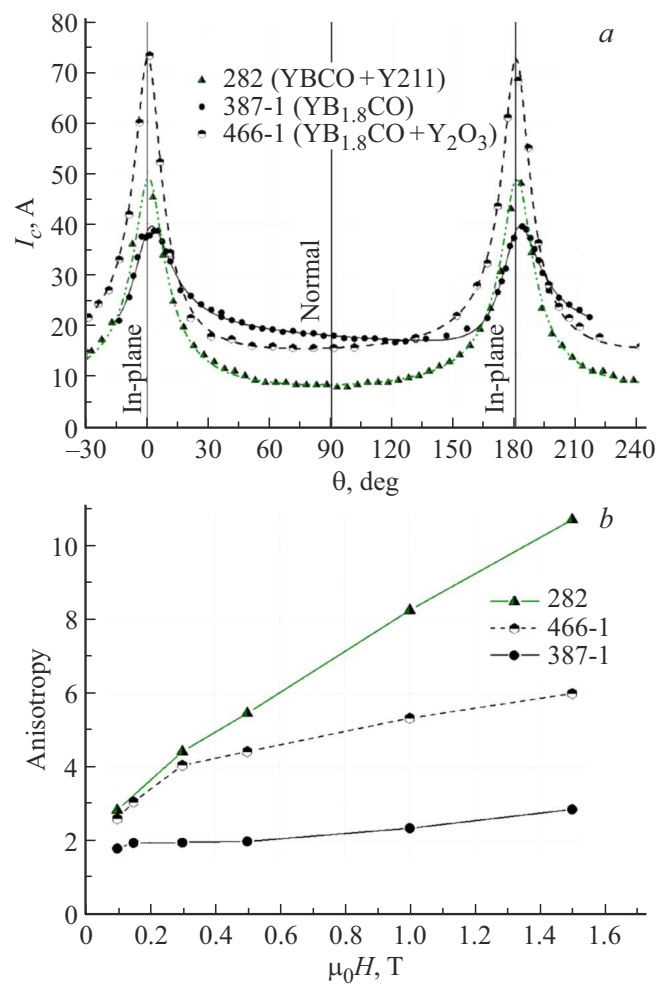


Figure 1. *a* — angular dependence of the critical current $I_c(\theta)$ at 77.4 K in an external field $\mu_0 H = 0.5$ T; *b* — field dependence of anisotropy $I_c(\theta = 0^\circ)/I_c(\theta = 90^\circ)$ at 77.4 K.

The degree of anisotropy can be estimated by the ratio $I_c(\theta = 0^\circ)/I_c(\theta = 90^\circ)$. The anisotropy differs significantly for the studied samples (Figure 1, *b*) and increases with increasing field. The lowest anisotropy (< 4) is observed in sample 387-1, which was synthesized utilizing the barium-deficient target.

Figure 2 illustrates the field dependence of the critical current in the perpendicular geometry at temperatures of 4.2, 20, and 40 K. Sample 387-1 demonstrates the highest critical current across all tested temperatures even despite the fact that its thickness ($1.4 \mu m$) is slightly smaller than that of samples 282 and 466-1 ($1.6 \mu m$). It is interesting to compare the obtained results with contemporary advancements documented in publicly available literature. The work [6] collected data on record-breaking values of the volume pinning force achieved up to the year 2024. By employing a non-stoichiometric target it was possible to achieve ~ 0.9 TN/m³ (9 T; 4.2 K) on thin films ($\sim 0.3 \mu m$) grown on single crystals. For comparison purposes, we will assume that the current density is constant across the HTS

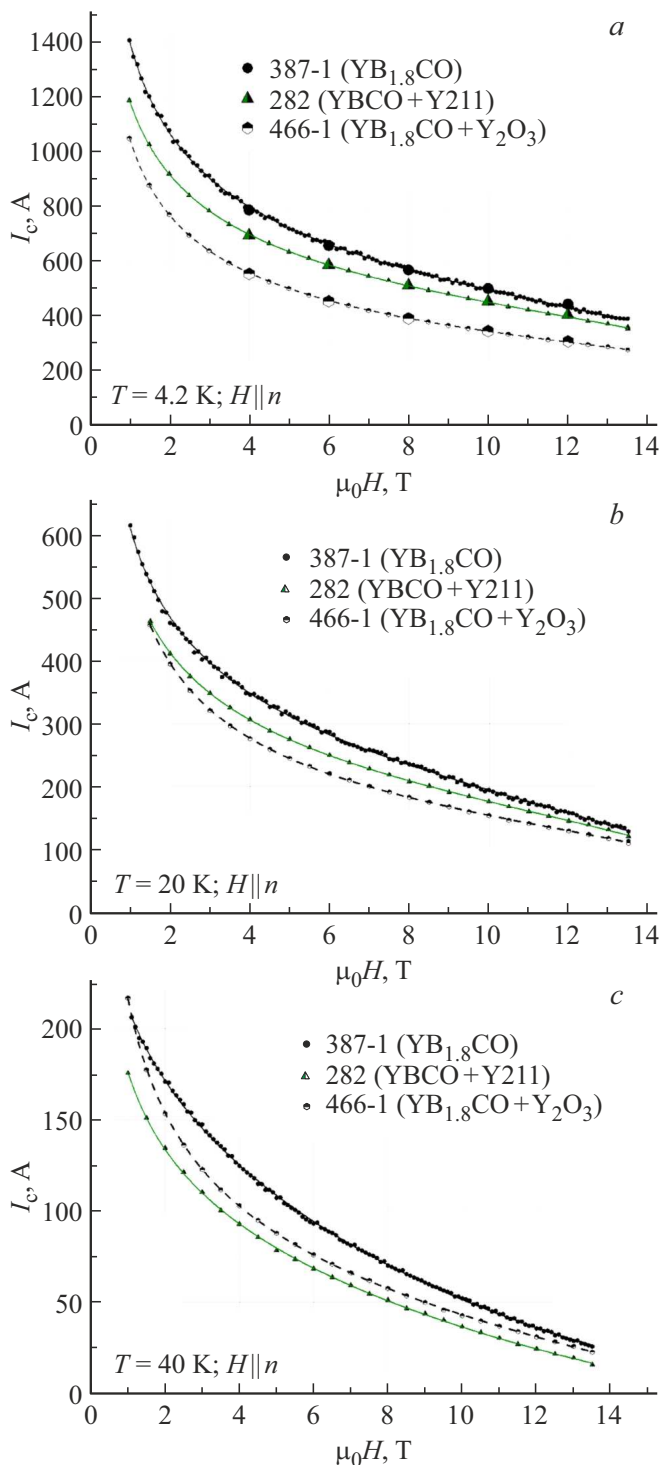


Figure 2. Critical current field dependences at 4.2 (a), 20 (b) and 40 K (c), estimated from the magnetization loop width. Large symbols in panel (a) correspond to values obtained from transport measurements.

layer thickness. For sample 387-1, the calculated value at 9 T and 4.2 K is also $\sim 0.9 \text{ TN/m}^3$, although our HTS layers are much thicker and grown on the buffered metallic substrate. This value also aligns closely with the record

data presented in [2], wherein exceptionally high current densities for superconducting tapes utilized in TF magnetic systems are presented.

4. Conclusion

The results of studies of three samples prepared by pulsed laser deposition on the pilot line of the National Research Center „Kurchatov Institute“ utilizing targets with varying degrees of stoichiometric deviation are presented. The samples produced from a barium-deficient target exhibit the highest current-carrying capacity in strong magnetic fields while simultaneously demonstrating the lowest anisotropy. For this particular sample, critical currents are comparable to the record values encountered in the literature.

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Conflict of interest

The authors declare that they have no conflict of interest.

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