Investigation of the effect of Al plasma treatment on the superconducting properties of YBa$_2$Cu$_3$O$_{7-\delta}$

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Abstract. The effect of the partial substitution of Cu for Al on the structural, compositional, and magnetic properties of polycrystalline compounds YBa$_2$Cu$_3$O$_{7-\delta}$ was studied. All prepared samples turned out to be single-phase with a small fraction of Ba secondary phases. Substitution of more than 2% aluminum causes an increase in spurious phases. DC susceptibility measurements show that the superconducting transition temperature T_C decreases. Hysteresis loops show that magnetic irreversibility decreases with increasing Al content. The critical current density J_C obtained using the Bean formula does not follow the same change. Al compensates for a decrease in J_C and an increase in its content near the solubility limit gives a higher J_C than in an undissolved sample.

1. Introduction

The analysis of impurity effects in high-temperature superconductors (HTSCs) aroused additional interest after confirming that this subject can serve as a critical test for the D-wave symmetry of the order parameter in high-temperature copper oxides [1, 2]. It is well known that oxygen content affects the crystal structure, electron transport, and superconducting properties in YBCO. It is also known that the temperature of the superconducting transition T_C depends both on the concentration of holes in the CuO$_2$ planes and on the relative electric charge of oxygen in these planes [3]. The level of this charge can be controlled by changing the stoichiometry of oxygen in the Cu–O chains, applying pressure, or by ion substitution [4-7].

The study of nonmagnetic cationic substitution in HTSCs in recent years has attracted great interest in connection with the observation of a very high efficiency of increasing T_C in these materials. The most interesting substitution effects are those where impurities occupy CuO$_2$ planes. In this case, impurities, such as Zn^{2+}, create strong elastic scattering simultaneously with the formation of a localized magnetic moment. This is indeed a source of a strong connection between charge and spin dynamics in these systems [8–9].

In this paper, we report on the study of the effect of partial substitution of Cu for Al and double substitution of Al instead of Cu on the structural and magnetic properties of YBa$_2$Cu$_3$O$_{7-\delta}$ compounds.

2. Experimental details

Samples for measurements were prepared by pressing and sintering YBa$_2$Cu$_3$O$_{7-\delta}$ powder with Al nanoparticles (NPs) preliminarily prepared by HTSC. The synthesis of YBa$_2$Cu$_3$O$_{7-\delta}$ composites was described in detail in [10–12]. For the study, samples with 0.5, 1, 2, 4 wt.% Al were used.

The magnetic properties were obtained from measurements of direct current magnetization with zero field cooling (ZFC) and field cooling (FC) M (T), as well as M (H), using the quantum-physical system of material properties (MPMS-XL5) operating in the mode vibrating magnetometer samples. XRD patterns were recorded on a D8 ADVANCE diffractometer (Bruker AXS). The isothermal dependences of the magnetization on the magnetic field were measured at a temperature of 4.2 K. The critical current density of the samples was estimated from the magnetic hysteresis loop using the extended critical state model [13-14]. The grain morphology of the surface of the samples was analyzed by scanning electron microscopy (SEM).

3. Results and discussion

Figure 1 presents X-ray diffraction patterns of the obtained YBa$_2$Cu$_3$O$_{7-\delta}$ + xAl composites (x = 0.5, 4 wt.%) at room temperature.

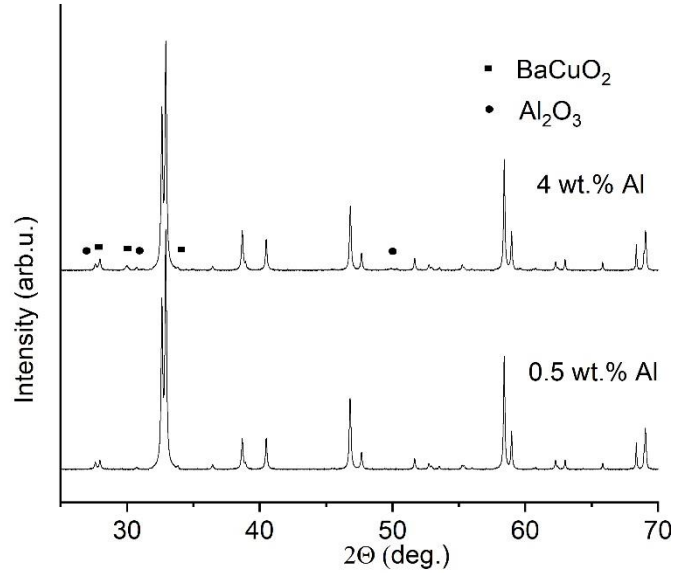**Figure 1.** XRD patterns of $Y_1Ba_2Cu_3O_{7-\delta} + x$Al samples with $x = 0.5, 4$ wt.%.

An analysis of the diffraction pattern showed that all samples crystallize in the orthorhombic phase with the space group Pmmm. Diffraction lines corresponding to spurious phases appear at $x > 2$; they are easily identified and shown in Fig. 1. Their proportion increases with increasing Al content. Note that the angular position of the peaks at a low doping level $x < 4$ changes significantly with the introduction of aluminum. At a high doping level, the main lines shift toward a higher angle as the Al content increases, which indicates a decrease in the c axis and that some ions of dopants occupy lattice sites. An important change in the intensity ratios between (00l) reflections and other reflections indicates significant areas of oriented samples. All prepared samples show a small fraction of Ba secondary phases at 2Θ between 25° and 31°. The main impurity was identified as $BaCuO_2$; the creation of Cu vacancies is also possible [15]. Doubling and splitting of the main peaks (013) and (103) becomes significant as the Al content increases; doping facilitates the transition from the orthorhombic to tetragonal lattice.

Figure 2 shows SEM photographs of Al doped samples taken at the same magnification, showing uniform grains. As the Al content increases, the grains become much finer, almost spherical in shape and poor in bond. For samples with a low Al content (Figure 2a), we can see the formation of stick-shaped grains with a random orientation and better connectivity between grains.
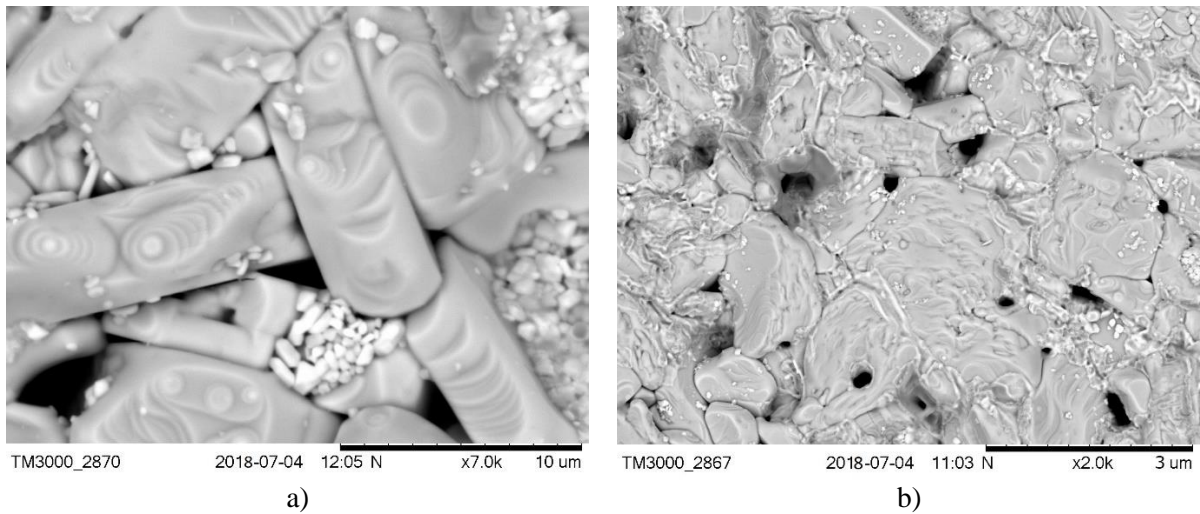

Figure 2. The SEM photographs of samples with: a) 0.5 wt.% Al, b) 4 wt. % Al.

Well-bound and closely packed grains can improve magnetic flux percolation, increase the number of pinning centers of magnetic vortices, and increase the critical current density [16].

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The dependence of the magnetization FC and ZFC on the temperature measured at a constant magnetic field of 50 Oe for samples doped with Al is shown in Fig. 3. The undoped sample has a T_C of ~ 91.85 K, which indicates that the oxygen content is close to optimal. The XRD results of this sample showed that the YBCO phase is the main phase. Samples doped with Al show a significant decrease in their T_C (defined as the temperature of the onset of diamagnetism). This decrease is from about 15 to 20 K and the width of the transition increases to 50%.
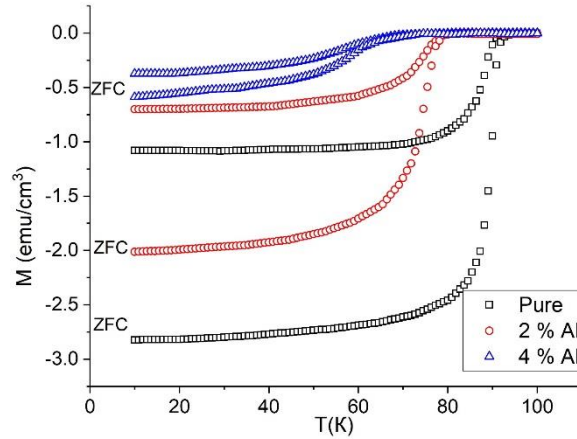

Figure 3. ZFC and FC M(T) curves of $Y_1Ba_2Cu_3O_{7-\delta} + x$Al samples ($x$ = 0.5, 2, 4 wt.%) with an applied DC field of 50 Oe.

This is due to the fact that Al doping in YBCO compresses the inconsistent region at the grain boundary, increasing the electrical resistance. A decrease in T_C is an indication that Al has a moderate effect on the conduction mechanism and magnetic order. The change in the density of charge carriers due to doping [17], the induction of mobile holes by substitution [18], whether or not associated with various structural changes [19], the total oxygen content and disorder in the $Cu - O_2$ planes [20] are various mechanisms responsible for Tc change. The magnetization curves also show that the difference ΔM between the response of the Meissner effect (FC) and the effect of screening streamlines (ZFC) increases with the Al content. ΔM is the trapped flux in the sample and depends on the intergranular and intragranular current for a very low applied field. In other words, both grain size and grain boundary quality affect ΔM. SEM observations show that Al reduces the average grain size. Thus, an increase in ΔM in Al doped samples cannot be a consequence of grain size, but is an indicator of an increase in the quality of grain boundaries.

The identical shapes of the hysteresis loops of the samples at 4.2 K (Figure 4) with a magnetic field of up to 5 T (parallel arrangement) indicate that the magnetization M decreases with increasing Al concentration. A sample showing the best texture derived from XRD analysis has the highest M. For the remaining samples, a decrease in grain size induced by doping corresponds to a decrease in M. At first glance, a decrease in grain size reduces current loops and, therefore, magnetization M, however, the sample, alloyed with 1% Al, does not obey this scheme.
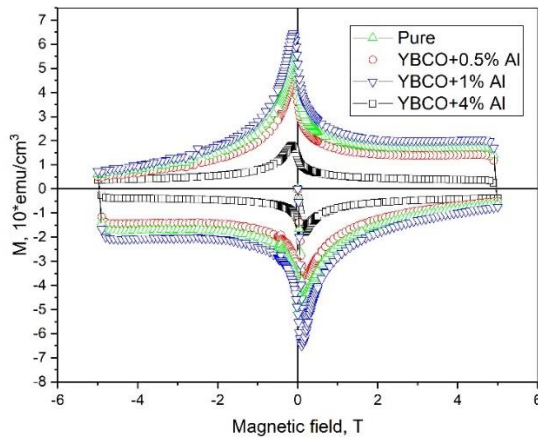
Figure 4. Magnetic hysteresis loops at 4.2K of $Y_1Ba_2Cu_3O_{7-\delta} + x$Al samples ($x = 0.5, 1, 4$ wt.%).
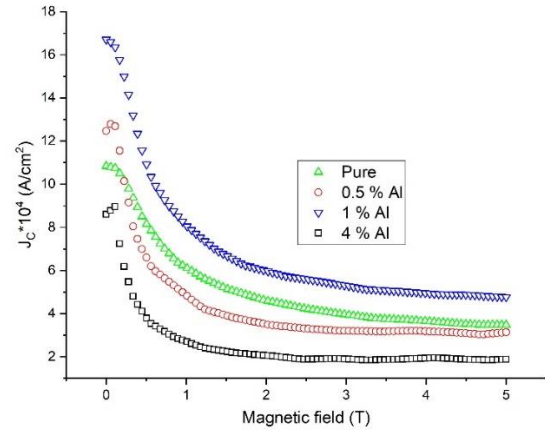

Figure 5. Critical current densities deduced using Bean's model from M(H) curves of Fig. 4 for samples ($x = 0.5, 1, 4$ wt.%).

The critical current density was derived from the curves of Fig. 5 using the Bean model. The calculated J_C is shown in Fig. 5. Considering fields above 2 T, where the demagnetizing field and other effects of shape and surface can be ignored, J_C repeats the change in M with Al concentration. With the introduction of 1% Al J_C higher in comparison with an undoped sample. The remaining samples doped with Al show J_C lower than the non-doped sample.

4. Conclusion

The effects of partial substitution of Cu for Al in polycrystalline $YBa_2Cu_3O_{7-\delta}$ were studied. Al reduces T_C and reduces the transition width. The exception is a sample with 1% addition of Al. Substitution, apparently, has a compensating effect, which enhances J_C and leads to values higher than that of a pure sample.

References
[1] Wu X S, Jiang S S, Lin J, Liu J S, Chen W M and Jin X 1998 *Physica C* **309** 25-32
[2] Ushakov A V, Karpov I V, Demin V G, Shaihadinov A A, Demchenko A I, Fedorov L Yu, Bachurina E P and Goncharova E A 2019 *Journal of Materials Science: Materials in Electronics* **30** 16
[3] Karppinen M, Yamauchi H and Inorg J 2000 *Mater.* **2** 589
[4] Palchayev D, Kh Gadzhimagomedov S, Murlieva Z, Kh Rabadanov M and Emirov R 2018 *Journal of Physics: Conference Series* **941** 012076
[5] Inoue K, Miyake Y, Miryala M and Murakami M 2017 *Journal of Physics: Conference Series* **871** 012051
[6] Ushakov A V, Karpov I V, Demin V G, Shaikhadinov A A, Demchenko A I, Bachurina E P, Fedorov L Yu and Goncharova E A 2019 *International Journal of Nanoscience* **18 (5)** 1950010
[7] Lepeshev A A, Ushakov A V and Karpov I V 2017 *Journal of Applied Physic.* **122** 104103
[8] Mendels P et al. 1999 *Europhys. Lett.* **46** 678
[9] Lepeshev A A, Karpov I V, Ushakov A V and Nagibin G E 2017 *Materials Research Express* **4** 056101
[10] Ushakov A, Karpov I, Lepeshev A and Fedorov L 2017 *Technical Phys.* **62** 168
[11] Ushakov A, Karpov I and Lepeshev A 2017 *J. of Superconductivity and Novel Magnetism* **30 (2)** 311–316
[12] Ushakov A, Karpov I and Lepeshev A 2017 *J. of Superconductivity and Novel Magnetism* **30 (12)** 3351–3354
[13] Lepeshev A A, Ushakov A V, Karpov I V, Balaev D A, Krasikov A A, Dubrovskiy A A, Velikanov D A and Petrov M I 2017 *J. of Superconductivity and Novel Magnetism* **30 (4)** 931–936

[14] Lepeshev A A, Patrin G S, Yurkin G Yu, Vasiliev A D, Nemtsev I V, Gokhfeld D M, Balaev A D, Demin V G, Bachurina E P, Karpov I V, Ushakov A V, Fedorov L Yu, Irtyugo L A and Petrov M I 2018 *J. of Superconductivity and Novel Magnetism* **30 (12)** 3841–3845

[15] Raffo L, Caciuffo R, Rinaldi D and Licci F 1995 *Supercond. Sci. Technol.* **8** 409

[16] Karpov I V, Ushakov A V, Lepeshev A A, Fedorov L Yu, Dorozhkina E A, Karpova O N, Shaikhadinov A A and Demin V G 2018 *Technical Physics* **63** 230

[17] Vyas A, Lam C C and Shen L 2000 *Physica C* **935** 341-348

[18] Tallon J L, Bernhard C, Shaked H, Hitterman R L and Jorgensen J D 1995 *Phys. Rev. B* **51** 12911

[19] Tokura Y, Torrance J B, Huang T C and Nazzal A L 1988 *Phys. Rev. B* **38** 7156

[20] Awana V P S, Malik S K and Yelon W B 1996 *Physica C* **262** 272