COMPOSITES OF SUPERCONDUCTING $YBa_2Cu_3O_{7-y}$ WITH SILVER

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In order to achieve better fabricability of the otherwise brittle ceramic superconductor $YBa_2Cu_3O_{7-y}$, we have formed composites of this superconductor with metals such as Ag, Cu and Al. It is found that up to 50 wt.% of silver can be added to $YBa_2Cu_3O_{7-y}$ without degradation of its superconducting transition temperature. Such composites, sintered at about 850°C, can be easily rolled in the form of strips. Superconductivity with nearly the same T_c as in the parent compound is retained in the composites even after cold working, thus obviating the need to regenerate superconductivity by heat treatment. The superconducting properties of $YBa_2Cu_3O_{7-y}$, however, degrade when composites are formed with Cu and Al.

1. Introduction

The recent discovery of superconductivity in the 90 K temperature range [1] in a new class of materials represented by the formula $RBa_2Cu_3O_{7-y}$ with R as one of the trivalent rare earths or Y, has generated a tremendous amount of interest and a great deal of activity in these materials as they have vast potential for technological applications. For making superconducting magnets, apart from the high superconducting transition temperature (T_c), one also needs high upper critical magnetic fields, large critical current densities and the ability to draw wires or to make thin strips or tapes out of these materials. Large critical current densities obtained from magnetization data have been reported in single crystals [2,3] and thin films [4] of $YBa_2Cu_3O_{7-y}$ but are still a problem to achieve in polycrystalline samples. Moreover, the inherent brittleness and low impact strength of ceramic superconductors have posed formidable problems in the fabrication of usable shapes out of these materials. In this communication we show that to some extent, this latter problem can be overcome by using ceramic–metal particulate composites. We have investigated the properties of composites formed by $YBa_2Cu_3O_{7-y}$ with powders of Ag, Cu and Al. Among these Cu and Al are found to degrade the superconducting properties while composites with Ag continue to be superconducting with the same transition temperature as the parent materials. These composites can be easily cold-rolled and superconductivity is retained even after cold-rolling. Fabricability is provided by the silver. Both the superconducting and silver matrices are enmeshed with each other. Detailed studies were carried out on $YBa_2Cu_3O_{7-y}$–Ag composite materials results on which are reported here.

2. Experimental

The $YBa_2Cu_3O_{7-y}$ superconductor in the powder form was prepared by the standard ceramic procedure in which stoichiometric amounts of Y_2O_3, $BaCO_3$, and CuO are very intimately mixed and heated at 950°C for several hours. After cooling to room temperature, the product is pulverized and reheated to 950°C in flowing oxygen atmosphere. To make superconductor–metal composites, the ceramic powder was thoroughly mixed with silver metal powder in differing weight proportions ranging from 10 to 50 wt.% silver. Cold-compacted specimens from

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these mixtures were sintered in flowing O_2 atmosphere at 850°C for a few hours. This procedure resulted in densified pellets of $YBa_2Cu_3O_{7-y}$–Ag composites. Care was taken to ensure that the silver does not melt and react with the superconducting material. The samples were characterized from powder X-ray diffraction measurements using Cu Kα radiation and from electron microscopy studies. Superconducting properties of the ceramic samples, composites and the cold-rolled samples were examined using four-probe dc resistivity and low-field magnetic susceptibility measurements.

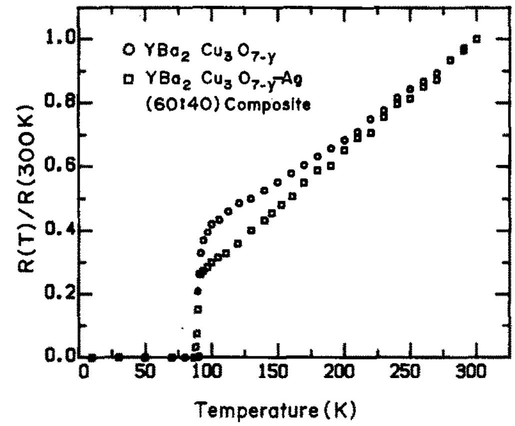

Fig. 1. Normalized resistance versus temperature for a sintered sample of $YBa_2Cu_3O_{7-y}$ and the 60:40 composite.

3. Results and discussion

First of all, X-ray diffraction patterns of the parent oxide $YBa_2Cu_3O_{7-y}$ and various composite materials were obtained to look for any structural changes on silver addition and to find out whether or not silver has reacted with the superconducting material by entering the lattice. It was observed that the X-ray lines pertaining to the superconducting $YBa_2Cu_3O_{7-y}$ were identical in both the parent material and the composites. The composites also showed characteristic X-ray lines from unreacted silver, the intensities of which increased with increasing silver content. This clearly shows that Ag has not reacted with the superconducting material but is staying as silver metal.

Earlier studies from this laboratory have shown that replacement of Cu by Ag (obtained by substituting part of the CuO by Ag_2O during preparation), in both $La_{1.8}Sr_{0.2}CuO_{4-y}$ and $YBa_2Cu_3O_{7-y}$ reduces their T_c very rapidly [5,6]. Therefore, in order to check that the T_c has not been affected by the silver addition, we carried out resistivity and magnetic susceptibility measurements on the parent ceramic material and the composites. The measurement procedure and the techniques are the same as reported earlier [5,6]. Fig. 1 shows variation of normalized resistance with temperature for the parent $YBa_2Cu_3O_{7-y}$ and for a representative composite material with 60 wt.% $YBa_2Cu_3O_{7-y}$ and 40 wt.% silver (designated as 60:40 sample). The 60:40 composite shows a transition to the superconducting state with a sharp drop in resistance at about 92 K. The zero-resistance state is achieved at 88 K. These temperatures are only slightly lower than the corresponding temperatures of 95 K and 91 K in the starting parent ceramic superconductor $YBa_2Cu_3O_{7-y}$. This clearly demonstrates that a continuous superconducting path is present in the composite samples and that silver has not degraded the T_c very much.

The results of T_c measurements on various other composites are nearly identical to those discussed above for 60:40 sample. All these samples were found to have superconducting transition temperatures only slightly lower than that of the parent $YBa_2Cu_3O_{7-y}$ material. Low-field dc/ac susceptibility measurements [7] also show the onset of the diamagnetic signal in the composites in agreement with the resistivity data. It is noted that the resistivity of the composites (14 μΩ cm for 60:40 composite at 300 K) is considerably smaller than that of the pure $YBa_2Cu_3O_{7-y}$ (470 μΩ cm at 300 K). This is expected because of the good electrical conductivity of silver. Thus from these measurements it is clear that silver powder addition up to 50 wt.% and the heat treatment employed by us does not lead to large suppression of T_c implying that silver does not substitute for any of the elements in this oxide superconductor. Composites with Cu and Al were also tried, but both these elements tend to get oxidised and degrade the superconducting properties.

Experiments on the cold rolling of parent superconducting ceramic and the superconducting–silver composite specimen were carried out. As expected, specimen made from pure ceramic powders, could not take any deformation. However, substantial improvement in rolling characteristics was observed in the case of composite specimen with progressive addition of silver. Samples containing 30 and 40 wt.% silver could undergo 35% and 55% deformation, respectively, without any cracks during rolling. The 50 wt.% silver containing specimen could be elongated to four times its original length. Fig. 2 shows a photograph of a typical 50:50 rolled sample reduced in thickness from 1.2 mm to 250 μm (about 80% deformation). Strips up to 150 μm thickness have been fabricated in this manner. These results have been obtained in a single rolling step. Experiments on intermediate annealing and hot rolling are currently being carried out to make even thinner ribbons. It is important to note that the specimens retain superconductivity during all fabrication steps and hence do not need any further heat treatment for regeneration of superconductivity.

Fig. 3 shows a scanning electron micrograph of a surface parallel to the direction of rolling. This is from a 50:50 sample which has undergone four-fold elongation. Both the silver (white areas) and the oxide (black areas) particles are distributed uniformly to form continuous matrices enmeshed with each other. The corresponding X-ray mapping for silver (Ag Lα) and copper (Cu Kα) of the same area are shown in figs. 4 and 5, respectively. The copper mapping represents the distribution of $YBa_2Cu_3O_{7-y}$ phase. The two mappings can be seen to be complementary to each other. The microstructure along the transverse

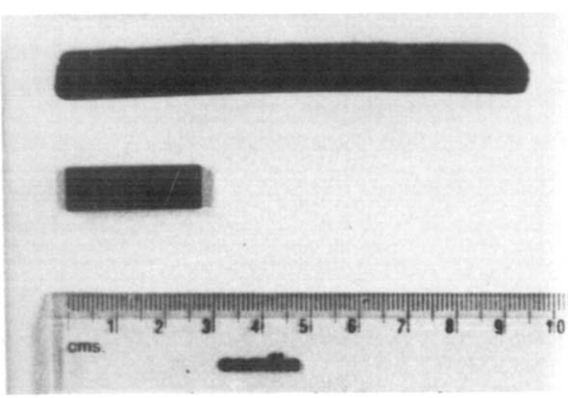

Fig. 2. Photograph of $YBa_2Cu_3O_{7-y}$–Ag 50:50 composite before and after rolling showing the elongation achieved.

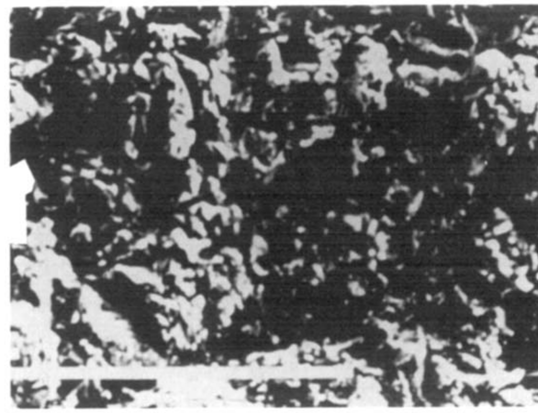

Fig. 3. Scanning electron micrograph of 50:50 rolled strip (×600). Bar = 100 μm.

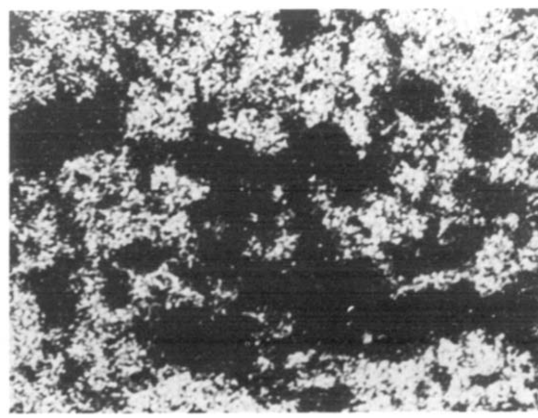

Fig. 4. X-ray mapping of silver (Ag Lα) for the area shown in the micrograph.

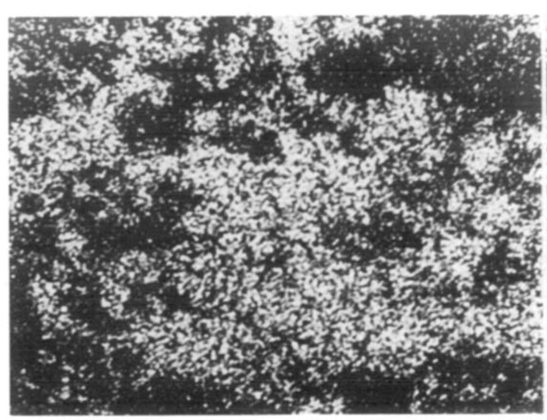

Fig. 5. X-ray mapping of copper (Cu Kα) for the area shown in the micrograph.

section of the rolled specimen was found to be identical to the structure shown in fig. 3. It was also observed that the distribution of silver and oxide particles was the same in as-sintered and the rolled samples. These measurements clearly corroborate the results of the X-ray diffraction studies which showed that the silver particles had not reacted with the oxide. Both $YBa_2Cu_3O_{7-y}$ and silver are dispersed uniformly with no segregation occurring even after the rolling operation. This was further confirmed by taking X-ray beam scans with EPMA.

In conclusion, ceramic–metal composites of superconducting $YBa_2Cu_3O_{7-y}$ with silver metal powder have been investigated. The addition of silver up to 50 wt.% does not degrade superconducting properties. The silver powder provides a continuous matrix enmeshed with the superconducting oxide powder. The silver continuum imparts the fabricability to the material while the ceramic oxide provides superconducting properties to the aggregate. The composite can undergo considerable deformation while retaining the T_c of the parent superconducting oxide material thus enabling the fabrication of thin ribbons.

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