Superconductivity and Transport Properties of Tl$_{2-x}$Hg$_x$Ba$_2$Ca$_2$Cu$_3$O$_{10.5}$

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Superconducting samples of Tl$_{2-x}$Hg$_x$Ba$_2$Ca$_2$Cu$_3$O$_{10.5}$ were prepared by solid state reaction of appropriate quantities of HgO, Tl$_2$O$_3$ and the precursor Ba$_2$Ca$_2$Cu$_3$O$_7$. Our best Hg-doped samples have the superconducting onsets at 130 K. Similar to the substitution of Bi ions by Pb ions stabilizing the Bi-based superconducting oxides [1], partial substitution of Tl$^{3+}$ with Hg$^{2+}$ in Tl-2223 produces a stable Tl-2223 phase with highest possible T$_c$.

T$_c$ of Tl$_2$Ba$_2$Ca$_2$Cu$_3$O$_{10.5}$ (Tl-2223) apparently varies from 113 K to 127 K even if the compound has an optimized oxygen concentration [2]. Studies were carried out to stabilize the higher transition phase (127 K). Our investigation indicated that Hg doping stabilizes the highest T$_c$ phase of Tl-2223.

Bulk polycrystalline samples of Tl$_{2-x}$Hg$_x$Ba$_2$Ca$_2$Cu$_3$O$_{10.5}$ were prepared by solid state reaction method. Appropriate amounts of HgO, Tl$_2$O$_3$, and Ba$_2$Ca$_2$Cu$_3$O$_7$ precursor were mixed, ground, and pressed into pellets. All the chemical handling was done in a dry box. The pellets were vacuum sealed into quartz tubes. The quartz ampoules were sintered at 830$^\circ$C to 870$^\circ$C, and finally slow cooled to room temperature.

The structures of the specimens were determined by powder x-ray diffraction. Figure 1 shows the x-ray diffraction pattern with indices of a typical Tl$_{2-x}$Hg$_x$Ba$_2$Ca$_2$Cu$_3$O$_{10.5}$ sample with x = 0.4. A prominent low-angle reflection at 2$\theta$ = 5$^\circ$ was detected. All the major peaks in the diffraction pattern correspond to the Tl$_{2-x}$Hg$_x$Ba$_2$Ca$_2$Cu$_3$O$_{10.5}$ phase [3]. Peaks marked with * are due to impurity phases, such as BaCuO$_2$ and Ca$_2$Cu$_3$O$_4$. The energy dispersive x-ray spectroscopy (EDX) analysis also suggested that incorporated Hg ions substitute for Tl ions in Tl-2223.

The superconducting properties of the samples were characterized by d.c. magnetic susceptibility and transport measurements. The magnetically determined onsets T$_c$ for as-grown Tl$_{1.6}$Hg$_{0.4}$Ba$_2$Ca$_2$Cu$_3$O$_{10.5}$ specimens are 128 K from both the shielding (zero-field-cooled) and the Meissner (field-cooled) measurements.

Figure 2 shows the temperature dependence of the electrical resistivity for samples of Tl$_{2-x}$Hg$_x$Ba$_2$Ca$_2$Cu$_3$O$_{10.5}$ with x = 0, 0.4, and 0.6. For all the samples, the resistivity exhibits a linear temperature dependence in the range 300 to 140 K. At room temperature the Hg-doped samples have a higher resistivity than the undoped specimen (we note that pure Hg-1223, not shown in the figure, has a significantly higher resistivity of $\rho$ = 27 m$\Omega$ cm at 300K [4]). The sample with x = 0 shows behavior typical of undoped Tl-2223: a superconducting onset at 122 K and zero resistance at 116 K. The sample with x = 0.4 shows onset of
Figure 1. X-ray diffraction pattern of Ti1.6Hg0.4Ba2Ca2Cu3O10.5. The reflection (h k l) are indexed as shown. Peaks marked with * are due to impurity phases.

Figure 2. Temperature dependence of the electrical resistivity for Hg-doped Ti-2223 samples with x = 0, x = 0.4, and x = 0.6.

superconductivity at 130 K and zero resistivity at 126 K. While the sample with x = 0.6 displays a superconducting onset at 125 K and zero resistance at 122 K.

As Fig. 2 demonstrates, modest Hg doping increases the superconducting onset temperature of Ti-2223 compounds. For x > 0.6, Tc again decreases [5]. It is most likely that partial substitution of Ti3+ with Hg2+ increases the effective Cu valence and brings the system from an overdoped regime to an optimally doped regime and thus promotes the formation of the 2223 phase. As the Hg concentration increases further (beyond x=0.6), the system goes from the optimally doped regime to an under-doped regime, resulting in the observed decrease of Tc.

We have successfully prepared the Hg-doped Ti2Ba2Ca2Cu3O10 superconductors. EDX analysis indicated that Hg ions partially replaced Ti ions in the system. Our best Hg-doped sample has the onset superconducting transition temperature at 130 K. Partial substitution of Ti3+ with Hg2+ in Ti2Ba2Ca2Cu3O10 produces a stable TI2223 phase with highest possible Tc.

Note added: After completion of this work, we become aware of a related study of Hg-doping in Ti-2223, whose finding agree with ours with x=0.4 [6].

References

2. Thallium-Based High-Temperature Superconductors. ed. A. M. Hermann and J.V. Yakhmi (Marcel Dekker, Inc./New York; Basel; Hong Kong).
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