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Low-Temperature Magnetic and Electromagnetic Studies of Y–Ba–Cu–O : Ag_x Ceramic Superconductor

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Magnetization and magnetic relaxation measurements have been studied at 4.2 K on $YBa_2Cu_3O_{7-\delta}:Ag_x$ ($x=0$ to 1.2) superconducting samples prepared by a coprecipritation technique. Remanent magnetization and hysteresis loops were found to be increased on Ag addition. Transport current density (J_c) was enhanced with the addition of Ag concentration. Flux creep rate was also increased with increase in Ag addition resulting in stronger pinning potential. Microwave-induced d.c. voltage measurements show a reduction of weak links with increase in Ag concentration. The enhancement of transport J_c is attributed to the stronger Josephson current paths due to the reduction of the total number of weak links after Ag addition.

KEY WORDS: Magnetic hysteresis; remanent magnetization; flux creep; pinning potentials; weak links.

Since the discovery of high T_c oxide superconductors, continuous effort has been exerted toward improving the transport critical current density (J_c). The maximum magnetic $J_c(J_{mc})$ of $YBa_2Cu_3O_{7-\delta}$ has been recorded at zero magnetic field for single crystals. Several papers have been focussed on the enhancement of J_c by silver addition to the oxide superconductors [1–4]. Many other groups have established that silver improves mechanical properties [5] and environmental stability [6] and provides a clean interface at Y–Ba–Cu–O grains [7]. However, a few papers have mentioned the flux creep and microwave properties in Ya–Ba–Cu–O : Ag ceramics with varying silver concentrations.

In this letter, we report experimental evidence of flux creep and microwave-induced d.c. voltage properties of sintered Y–Ba–Cu–O : Ag_x (x up to 1.2) ceramics. The magnetization curves down to 4.2 K have been analyzed in terms of standard flux creep models. Microwave-induced d.c. voltage studies clearly show a reduction of the total number of weak links between the superconducting grains after Ag addition.

$YBa_2Cu_3O_{7-\delta}:Ag_x$ (x up to 1.2) samples were prepared by a coprecipitation process using AR grade chemicals along with Ag_2O powder. Details of the novel preparation technique have been discussed elsewhere [2]. X-ray diffraction patterns of the samples show orthorhombic structure. X-ray-induced Auger analysis of these samples reveal traces of Ag_2O in the material.

The samples were cut into the required cylindrical size so that the longitudinal direction became parallel to the c-axis. Magnetic measurements were carried out using a PAR (model 150A) vibrating sample magnetometer in the temperature range 4.2–77 K. The magnetic field was applied parallel to the c axis, so that the induced shielding currents circulate within the ab plane. For magnetic relaxation (flux creep) measurements, an external field was applied and subsequently removed at a rapid sweep rate (≈500–10 kOe/min). As the maximum magnetic field was increased, the remanent magnetization was recorded as a function of time immediately after the external

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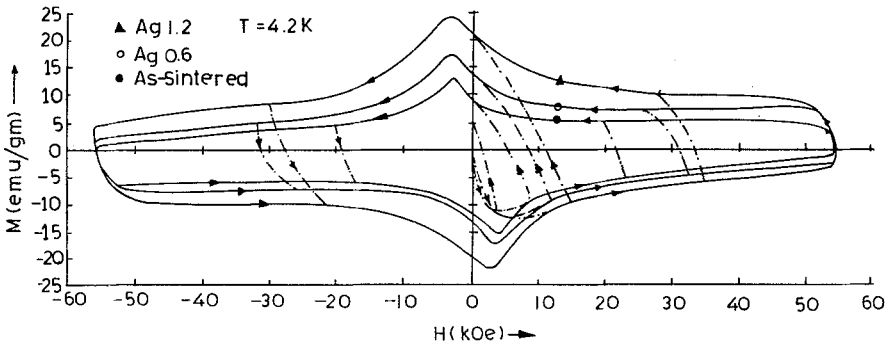

Fig. 1. Magnetization curves for as-sintered and Ag-added Y–Ba–Cu–O samples at 4.2 K. Different saturation magnetic field (H_{max}) values are shown in the loops.

field became zero. Transport critical current was measured using a standard four-probe technique in liquid helium bath.

For the measurement of the inverse a.c. Josephson effect, samples were mounted inside an X-band waveguide and irradiated with highly stabilized phase-locked microwave radiation. The induced d.c. voltage was measured with a high-impedance nanovoltmeter. Correction for thermal emf was also applied for each measurement. Details of the cryostat design for microwave measurements have been discussed elsewhere [8].

Figure 1 shows the magnetization hysteresis loops of various Y–Ba–Cu–O samples with different silver concentrations at 4.2 K. As it is evident from the figure that the sample with the highest Ag concentration, i.e., 12 wt. %, showed a drastically increased magnetic moment in comparison to the as-sintered sample. The systematic increase of ΔM (magnetization difference for increasing and decreasing fields) clearly indicates the creation of additional pinning centers. The calculated values of magnetic J_{mc} at 4.2 K are listed in Table I with the grain size data. Figure 1 shows that a larger grain size leads to an increased magnetic moment due to the larger current loops generated in the superconductor. As has been discussed elsewhere, the addition of silver oxide enhances grain growth, partially orients the crystallites, and creates a favorable environment for better oxygen absorption. The large ΔM is expected for Ag-added samples. The enhanced ΔM for Ag-added samples of larger grain size implies a greater amount of flux pinning or increased J_{mc} at certain applied magnetic fields. However, increase in grain size due to silver addition restricts the enhancement of magnetic J_{mc} which is an intragranular property.

Figure 2 shows the time dependence of relaxation of remanent magnetization on a logarithmic time scale, and Fig. 3 shows the normalized magnetization as a function of time. In these curves, relaxation depends upon the maximum applied magnetic field (H_{max}). The rate increases linearly up to H_{max} and becomes saturated at $H = 15$ kOe for 12 wt.% Ag concentration. The H_{max} changes from an as-sintered sample to silver-added samples (Table I).

It is difficult to determine whether the decay of remanent magnetization is logarithmically time dependent because of its very small relaxation rate, specially at 4.2 K. However, this type of magnetic relaxation behavior in high-T_c superconductors is attributed to thermally activated flux creep, i.e., the depinning process of quantized magnetic flux lines (fluxoids) trapped in pinning potentials due to thermal agitation. The magnetic relaxation based on the

Table I. Transport and Magnetization J_c at 4.2 K, H_{max}, and Pinning Potential U_0 for Y–Ba–Cu–O Superconductor with Varying Ag Concentrations

Sample name	Average grain size (μM)	Transport J_c (A/cm^2) at zero field	J_c (mag) (A/cm^2) at zero field	J_c (mag) (A/cm^2) at 50 kOe	H_{max} (kOe)	U_0 (eV) at 4.2 K
1. As-sintered Y–Ba–Cu–O	2	575	5×10^6	2.5×10^6	8	0.2
2. Y–Ba–Cu–O : $Ag_{0.1}$	5	680	2.5×10^6	1.2×10^6	10	0.22
3. Y–Ba–Ci–O : $Ag_{0.6}$	10	820	1.5×10^6	9×10^5	12	0.25
4. Y–Ba–Cu–O : $Ag_{1.2}$	20–30	1050	8.5×10^5	6×10^5	15	0.28

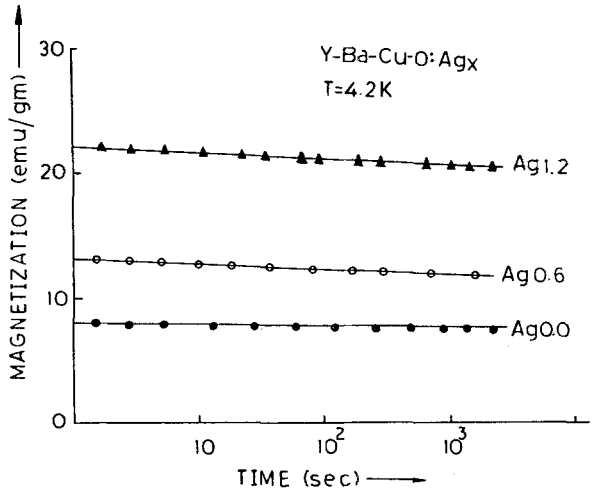

Fig. 2. Decay of the remanent magnetization as a function of time (log scale) for as-sintered and Ag-added U–Ba–Cu–O samples at 4.2 K.

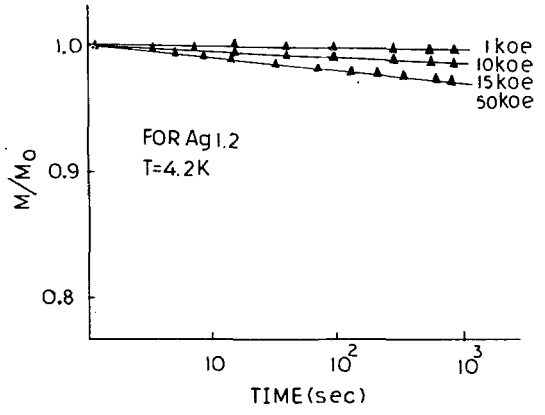

Fig. 3. Relaxation of remanent magnetization normalized to its initial value (Mo) for Y–Ba–Cu–O : $Ag_{1.2}$ sample showing saturation after $H_{max} = 15$ kOe at 4.2 K.

thermally activated model is given by

$$(1/M_0)\, dM/d \ln t = KT/U_0 \qquad (1)$$

where M_0 is the initial magnetization, M is the magnetization after time t, K is Boltzmann's constant, T is the temperature, and U_0 is the pinning potential that a flux bundle feels. Table I shows the pinning potentials for the present set of samples. The pinning potential increases with increase in silver concentration in the sample. Therefore, flux lines are pinned strongly

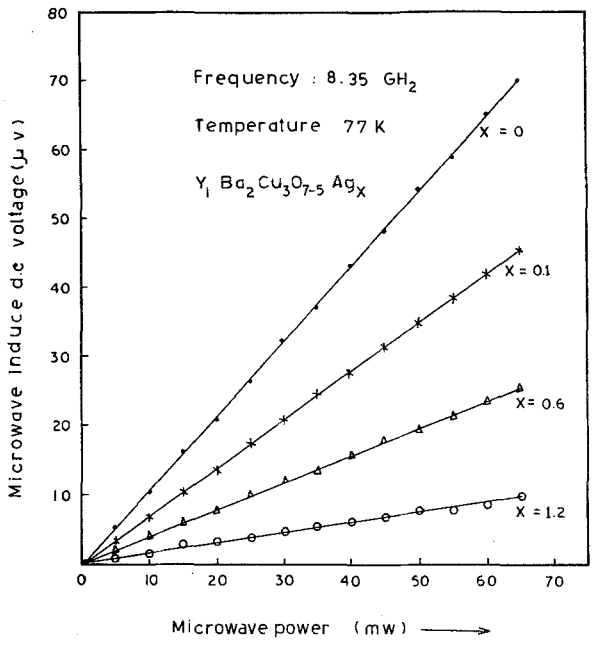

Fig. 4. Microwave-induced d.c. voltage as a function of microwave power for different Ag concentrations in $Y_1Ba_2Cu_3O_{7-\delta}Ag_x$ at 77 K.

against the Lorentz force in Ag-added samples, enhancing the transport current-carrying capacity at higher fields.

It is expected that when $H_{max} < 8$ kOe in as-sintered Y–Ba–Cu–O, relaxation of remanent magnetization takes place such that magnetic flux moves inside as well as outside the sample. However, when $H_{max} > 8$ kOe, magnetic relaxation can only take place by decreasing the magnetic flux in the sample. The flux creep rate presumably increases with increase in H_{max}, and above H_{max}, the flux creep rate is the same because remanent magnetization is the same. It is clear from Table I, that the flux creep rate increases with increase in silver concentration in the Y–Ba–Cu–O samples. In the critical state, a higher J_{mc} also implies a large flux gradient, which drives the flux lines to hop faster.

The inverse a.c. Josephson effect, which can quantitatively demonstrate the presence of weak links, shows less voltage pickup in silver-added samples irradiated inside an X-band waveguide in the frequency range 8–12 GHz and 80 mW of power at 77 K, whereas in as-sintered pellet $\approx 70\ \mu$V of pickup was recorded under similar conditions, indicating the presence of weak links. The enhancement of transport J_c is also due to the reduction of weak links and stronger Josephson current paths after addition of silver.

In summary, we make the following conclusions. Addition of silver in the Y–Ba–Cu–O system leads to

greater magnetization hysteresis at 4.2 K and enhances the transport critical current density. From flux creep measurements, it was found that the pinning potential increases with increase in silver addition, leading to a rise in flux creep rate. A considerable number of weak links between the superconducting grains are reduced with increase in Ag concentration in $Y_1Ba_2Cu_3O_{7-\delta}$ ceramic superconductors.

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