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Letter

Preparation of screen-printed YBCO thick films on an alumina substrate under flowing argon

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Abstract

Y–Ba–Cu–O (YBCO) thick films of 10–15 μm thickness were prepared on bare alumina substrates by the screen-printing technique. The films, which were heated at 850–900 °C in argon atmosphere and then exposed to oxygen, showed zero resistance at 79–84 K.

1. Introduction

YBCO thick films of fairly good quality have been successfully prepared on YSZ substrates [1–3]. Alumina is a particularly important substrate because of its low cost, required strength and common industrial use. There have been, therefore, numerous attempts to produce high T_c superconductor (HTS) thick films on this substrate. Few experiments, however, have succeeded in producing HTS films having a T_c higher than 77 K on a bare alumina substrate; associated with this, we cite the review by Bailey *et al.* [4]. The main reason for this is the strong overlayer–substrate interaction [5] and the misfit in expansion coefficient [6]. To overcome this difficulty, the following routes have been taken: (i) use of buffer between HTS films and the substrate; (ii) making the film thicker (100–200 μm) by painting the YBCO paste repeatedly on the substrate [4, 7].

In a previous paper [8] we reported that firing under a flowing rare gas is very useful for the enhancement of the critical current density J_c of films. This technique has recently been applied by Niu *et al.* [9] to the electrophoretic fabrication of HTS films. Here we report the successful preparation of YBCO films of 10–15 μm thickness on a bare alumina substrate through heat-treatments under flowing argon.

2. Experimental procedure

The preparation and characterization methods of YBCO films were similar to those described previously [8]. A brief description is given here. The YBCO powder, the average particle size of which was less than a few micrometres, was mixed thoroughly with an appropriate amount of an organic vehicle (Tanaka-Mattey KK, TMC-10TA) to form a paste. This was printed through a 300 mesh stainless steel screen on to alumina substrates. After being dried in an oven, the films were transferred into the tube furnace and then, as shown in Fig. 1, were fired at the desired firing temperature T_s for a period of time t. At T_s the film was exposed to argon flow (1 l min^{-1}), which was pre-passed at 800 °C, for a time t_1 and then exposed to oxygen flow (0.5 l min^{-1}) for a time t_2. The furnace was then cooled, with post-annealing at 600 °C for 2 h, to room temperature in flowing oxygen.

3. Results and discussion

The films were first fired at $T_s = 940$, 960 and 980 °C for the firing times t ($t_1 + t_2$) = 2, 5 and 10 min under argon flow (*i.e.* $t_2 = 0$). These conditions have been successfully applied to the fabrication of thick films on YSZ substrates [8], but none of these films showed

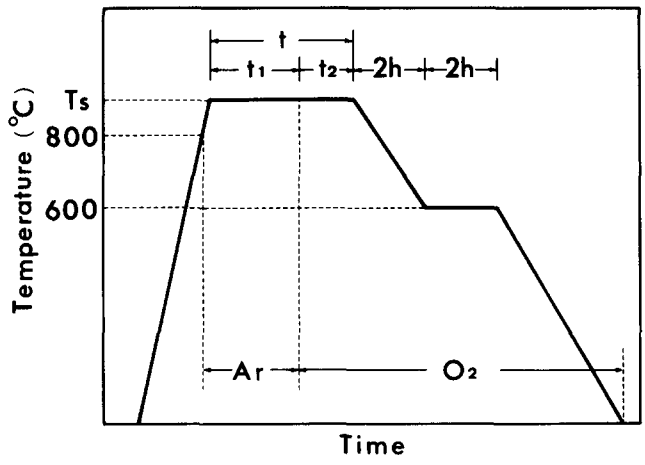

Fig. 1. Schematic of firing schedule for preparation of YBCO thick films.

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zero resistance above 77 K. It was found from scanning electron microscopy (SEM) observation that quite a large number of microcracks, which are probably the result of the discrepancy in expansion coefficient, remained on these films. Moreover, distinct orthorhombic YBCO peaks were found in X-ray diffraction patterns in very few films. Takabatake and Ishikawa [10] have reported that a very small amount of Al doping ($\approx 1\%$) strongly decreased the transition temperature T_c. Li *et al.* [5] have observed the concentration profiles of the cation distribution of a YBCO film/Al_2O_3 substrate sintered at 950 °C for 1 h and have reported that: (i) Al ions diffuse easily into the YBCO film, *e.g.* the concentration of Al cation at a distance of 10 μm from the interface is about 10 wt.%; (ii) the concentration peak values of Y, Ba and Cu cations increase with increasing temperature. The unsuccessful result mentioned above is attributed mainly to these facts, *i.e.* when the film was fired at high temperature (940–980 °C), film poisoning due to the severe interface reaction precluded the formation of the YBCO phase on the Al_2O_3 substrate.

The films were next heat-treated by using the longer period and lower temperature processing conditions shown in Table 1. All films fired under these conditions have reached zero resistivity at T_c (end point) = 79–84 K, except the sample fired at $T_s = 900$ °C for $t = 3$ h, and have adhered fairly well to the substrates. This was evaluated by a peel test using adhesive tape. Figure 2 shows the resistance–temperature characteristics and surface SEM images for two typical samples ($T_s = 850$ °C; $t = 1$ h and 10 h). The following features can be seen. (i) Both films showed metallic behaviour from room temperature to T_c (onset). (ii) The grain size increased slightly with increasing firing time t, but its size was still small and the film was porous as a whole. (iii) The microcracks still remained on these films, but their number was reduced compared with those on films heat-treated at higher temperatures. Figure 3 shows the variation in J_c with firing time t for two series of films on alumina fired under the conditions shown in Table 1. The J_c values of films on YSZ fired at 850 °C are also shown in Fig. 3 for reference. The observed

TABLE 1. Firing conditions of screen-printed YBCO thick film

$T_s = 850$ °C			$T_s = 900$ °C		
$t(=t_1+t_2)$[a]	t_1	t_2	$t(=t_1+t_2)$	t_1	t_2
60	40	20	10	5	5
180	120	60	30	20	10
300	240	60	60	40	20
600	540	60	180	120	60

[a]All times in minutes.

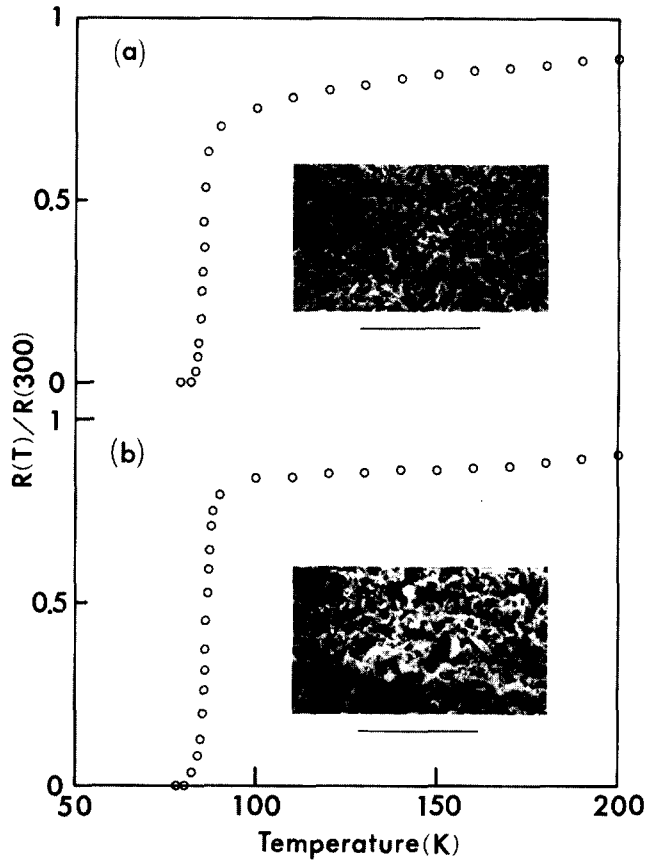

Fig. 2. Temperature dependence of resistance (normalized to room temperature) and surface SEM images (scale bar represents 50 μm) of screen-printed films. The firing conditions are: (a) $T_s = 850$ °C, $t = 1$ h; (b) $T_s = 850$ °C, $t = 10$ h.

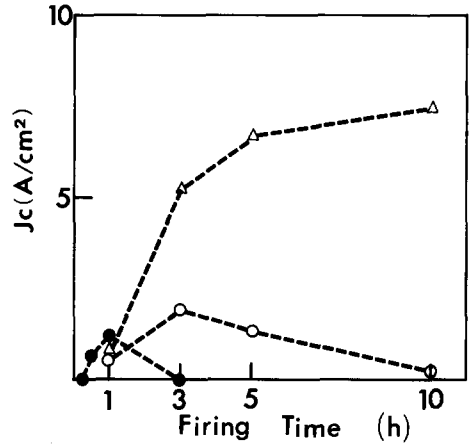

Fig. 3. Critical current density J_c at 77 K in zero magnetic field as a function of firing time t (see text). ○ films on alumina substrate fired at $T_s = 850$ °C; ● films on alumina substrate fired at $T_s = 900$ °C; △ films on YSZ substrate fired at $T_s = 850$ °C.

variations in J_c can be explained as follows. The longer heat treatment enhances the sintering of YBCO, which in turn causes the interface reaction. The J_c has, therefore, a maximum value at an appropriate firing time.

On the contrary, the J_c values of films on YSZ have a tendency to increase with increasing firing time. This may be due to the formation of a $BaZrO_3$ buffer layer [1, 11], and/or to the low chemical resistivity [12], which reduces the film poisoning.

It should be noted here that when the oxygen flow was maintained throughout the heat treatment the films fired at $T_s = 850$ °C for 3 h and $T_s = 900$ °C for $t = 1$ h did not show superconductivity, that is, the resistances of films increase with decreasing temperature. This suggests that the heat treatment under rare gas flow enhances the solid-state sintering kinetics [8, 13], which leads to successful formation of HTS films on alumina substrates.

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