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Growth of faceted 123 crystals in superconductive YBCO/Ag composites fabricated by infiltration-growth method

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Abstract

Growth process and structures of faceted 123 ($YBa_2Cu_3O_{7-X}$) crystals fabricated by conventional unidirectional solidification and infiltration-growth method were studied experimentally and numerically to clarify the formation mechanism of the solidification structures of YBCO/Ag composites. Sample size was almost unchanged during the infiltration-growth process, since molten Ba–Cu–O with a small amount of Y and Ag was just infiltrated into pores in pre-sintered porous primary phase: 211 (Y_2BaCuO_5) with Ag and then it was cooled to grow 123 crystal. Solidified samples had uniform distribution of primary phase and Ag particles in peritectic 123 crystals. The critical transition condition of macrostructures from columnar to equiaxed structures, and the freezing front temperature of 123 crystal during solidification were clarified. The formation processes of macro/microstructure of faceted 123 crystals were simulated, and the results compared with the experimental ones.

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Keywords: Superconductor; Crystal growth; Simulation; YBCO; REBCO; Peritectic; Facet

1. Introduction

$YBa_2Cu_3O_{7-X}$ (YBCO) superconducting oxides are expected to be used in many applications such as magnetic levitation, high-field magnets, magnetic shields, motors and generators, because of their high critical current density (J_c) at 77 K and a high magnetic field. Unidirectionally solidified YBCO oxides have high J_c, since they have columnar or single 123-crystal structures with normal particles which work as flux pinning centers [1–3]. It is known that YBCO/Ag composites has better mechanical properties due to the improving effects of Ag in YBCO-oxides. The infiltration-growth process [4] is effective way to improve the shape consistency, since in this method, molten Ba–Cu–O with a small amount of Y and Ag was just infiltrated into pores in pre-sintered porous primary 211 (Y_2BaCuO_5) phase with Ag, the shrinkage of specimens during the process of melting and solidification is diminished. However, the structure formation mechanism [5–8] of the above process is not still well known. In this paper, the formation process of macro/microstructures [9–12] of faceted 123 crystals in YBCO/Ag composites fabricated by the infiltration-growth process were analytically and numerically simulated, and the results compared with the experimental ones.

2. Experimental

The specimen for the infiltration-growth process is composed of two parts: Ba–Cu–O (Y:Ba:Cu = 0:3:5) powder pressed into a plate, and a pre-sintered (1323 K, 3600 s) porous Y211 (Y_2BaCuO_5) preform. Ba–Cu–O powder contains a small amount of Y (0.3–0.6%Y) to avoid violent melting reaction of porous Y211. Initial average radius of Y211 powder was 0.8 μm. The dimensions of the preform

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Table 1
Sample compositions fabricated by the infiltration-growth process

Ag (%)	Y:Ba:Cu (at.%)	Before growth L:211 (at.%)	After growth 123:211 (at.%)
0	34.0:29.9:36.1	29:71	46:54:00
8.4	35.1:28.7:36.2	27:72	43:57:00

were 30 mm × 10 mm × 3 mm. Ag was added to both Ba–Cu–O powder (10–12 mass%Ag) and pre-sintered porous perform (7–10 mass%Ag). As temperature rises, Ba–Cu–O powder melts and the molten oxide gradually penetrated into pre-sintered porous Y211 perform ($T \geqq T_p$: peritectic temperature), and then cooled below the peritectic temperature to grow 123 crystal under a temperature gradient (15–30 K/cm). Some specimens were quenched during the solidification to reveal the interface of 123 crystal. The atmosphere during solidification was air. Typical compositions of samples with or without Ag after the infiltration-growth process were shown in Table 1.

3. Results and discussion

3.1. Macro/microstructures of 123 crystals in YBCO/Ag samples fabricated by infiltration-growth

Fig. 1 shows an example of the macrostructure and the shape of YBCO/Ag specimen solidified by the infiltration-growth method. Initial shape is indicated by the outer solid line, and the sample shows almost no difference in the shape during the process, while the samples fabricated by the conventional unidirectional solidification method showed large contraction after the process. Thus, the infiltration-growth process gives big advantage in shape-consistency. This sample shows columnar 123-crystal structure. In 123 crystals, 211-particles and Ag-particles were uniformly distributed in this sample, while Ag-free zone in the central region of 123-cell formed in the specimen fabricated by conventional unidirectional solidification ($f_{211\mathrm{inL}} = 0.43$).

3.2. Transition condition of macrostructures of 123 crystals

Macrostructure of 123 crystal such as columnar or equiaxed structure changes depending on solidification conditions [5,6,9–12]. Fig. 2 shows the effects of growth conditions (temperature gradient: G and growth rate: R) on the macrostructures of 123 phase in YBCO/Ag composites. Black marks are faceted columnar crystals, and white marks are equiaxed crystals. Square marks are infiltration-growth samples with $Sv = 0.38$ cm^2 and triangles are conventional samples. The critical transition conditions from columnar to equiaxed structure (the critical G–R relations) are given by the bold solid lines in the figure. Two solid lines (of the same Sv) show the results for $f_s = 0.01$ and 10^{-10}, respectively. We can clearly find crystals in the liquid when they grew up to $f_s = 0.01$ or more. These calculated results show good agreement with corresponding experimental data. On the other hand, circles are columnar crystals (black marks) and equiaxed crystals (white marks) in the infiltration-growth samples of YBCO system (Ag-free), in which case the critical transition condition has larger critical R-values (thin solid lines) for the same size samples ($Sv = 0.38$ cm^2). In case of smaller samples, the critical condition has larger critical R-values and can be given by dotted lines for $Sv = 0.03$ cm^2, for example, in the figure.

Fig. 1. Macrostructure and shape of YBCO/Ag composite fabricated by the infiltration-growth method. ($G = 26$ K/cm, $R = 0.18$ μm/s).

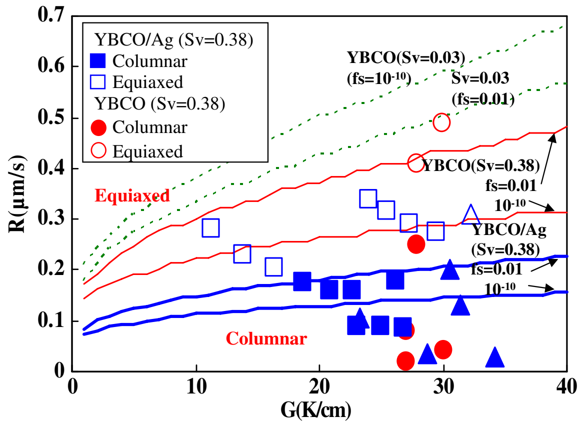

Fig. 2. Effect of growth rate (R) and temperature gradient (G) on the solidification structure of 123 phase of YBCO/Ag composites. Solid lines indicate the calculated transition conditions from columnar to equiaxed crystals. (YBCO/Ag: $Sv = 0.38$(cm^2), $f_s = 0.01$, 10^{-10}, YBCO: $Sv = 0.38$, 0.03 (cm^2), $f_s = 0.01$, 10^{-10}).

The above critical transition conditions of macrostructures from columnar to equiaxed structures were analyzed by the following procedures [9–12]. The relation between growth rate R (μm/s) of faceted 123-cell and undercooling ΔT (K) from the peritectic temperature (T_p) was given by the following equation for YBCO/Ag specimens. Cellular growth rate: R (μm/s) is given by

$$R = A_{kc} \cdot \Delta T_r^2 / \eta(T) \tag{1}$$

where A_{kc} is a constant ($A_{kc} = 1.6 \times 10^{-5}$ for YBCO/Ag, $f_{211\mathrm{inL}} = 0.72$), $\Delta T_r = \Delta T / T_p$, $\Delta T = T_p - T$, T is temperature at the interface, T_p is peritectic temperature of 123 phase, $\eta(T)$ (Pa s) is viscosity of the liquid: $\eta(T) = A_e \cdot \exp(E_g/T)$, A_e and E_g are constants, $A_e = 1.9 \times 10^{-11}$, $E_g = 26000$, and T is temperature (K).

R is larger than the normal growth rate (u) by $2^{1/2}$–$3^{1/2}$ which is obtained from geometric relations of triangular