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Joining of $Y_1Ba_2Cu_3O_{7-x}$ and Pb(Zr, Ti)O_3, and their Interfaces

Sunao Sugihara* & Hiroshi Fujitani

Department of Materials Science and Ceramic Technology, Shonan Institute of Technology, 1–1–25, Tsujido Nishikaigan, Fujisawa 251, Japan

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Abstract

The joining of the superconducting oxide, YBCO, with the piezoelectric ceramic, Pb(Zr,Ti)O_3, was studied using a thick film of YBCO. The electrical resistivity of the composite was evaluated from liquid nitrogen temperature to room temperature; the resistivity was 0·1 Ω cm at 80 K without a clear critical temperature. Furthermore, Pb(Zr, Ti)O_3 was sandwiched by YBCO electrodes to measure the piezoelectricity.

A slurry of YBCO was pasted on a sintered Pb(Zr,Ti)O_3 pellet followed by sintering for 5, 60 and 420 min at 1193 K. The YBCO film thicknesses were 5, 17 and 35 μm as determined by scanning electron microscope (SEM). An electron probe microanalyzer (EPMA) revealed diffusion of Ba into Pb(Zr,Ti)O_3. The hetero-joining was achieved, although the YBCO film was porous as investigated by SEM. A higher density film can be obtained by optimizing the sintering conditions for the composite.

1 Introduction

In considering an electrode for ceramics, the wettability of the metal for the ceramic is of importance. A glass frit is commonly employed for joining a metal to a ceramic. Sugihara *et al.* have widely studied the wettability and bondability of metal/ceramics and ceramics/ceramics for electronic ceramics in particular.[1,2]

The joining of a superconductor to a semiconductor has been theoretically studied in terms of boundary effects by P. G. De Gennes;[3] superconducting electrons migrate towards the semiconductor in the proximity effect. There have been several studies of the proximity effect in superconductors;[4] a Y–Ba–Cu–O based multilayer for optoelectronic devices was investigated on the sapphire and Al_2O_3 as as IR detector.[5] The YBCO films were hundreds of nm thick and employed for the counterelectrode and base.

On a more macroscopical scale, Srivastava *et al.* reported that $Y_1Ba_2Cu_3O_{7-x}$ (hereafter YBCO) adhered to $BaTiO_3$ and to Pb(Zr,Ti)O_3 (hereafter PZT); electrical properties of the devices were characterized.[6] Furthermore, Moya *et al.* formed YBCO composites on Ce–TZP, Y–TZP and spinel substrates.[7] The interfaces between YBCO and other ceramics are very important for devices or composites when one uses YBCO as an electrode.

The first purpose of the present work is to join YBCO thick film to piezoelectric ceramics, then to find the appropriate thickness and the best contact of the film to give satisfaction for both piezoelectricity and superconductivity. The second aim is associated with the use of YBCO electrode for a field effect transistor at the temperature even if the temperature is not below the transition.

2 Experimental

2.1 Fabrication of PZT

The PZT powder with composition of Pb $(Zr_{0·53}, Ti_{0·47})O_3$ (Fuji Titanium Ind. Co. Ltd, Japan) was pressed at 180 MPa followed by sintering for 2 h at 1503 K. The sintering was carried out in an atmosphere of lead or lead oxide in the crucible. The size of the sintered pellets was 15 mm in diameter and 0·8–1·0 mm thickness. The sintered density was 95–96% of the theoretical value.

2.2 Slurry of YBCO

The YBCO powder (Seimi Chemical Co. Ltd, Japan) calcined at 873 K was milled with the solvent and ZrO_2 balls (3 mm in diameter) for 24 h. The slurry was painted on the PZT fabricated above, then sintered at 1193 K in an oxygen atmosphere (0·5 litre/min) for each sintering time; 5, 60, 420 min. The samples were cooled down to

*To whom correspondence should be addressed.

room temperature at the rate of about 1·5 K/min. The surface of the thick films was examined by X-ray diffraction analysis (XRD).

2.3 Interfaces between YBCO and PZT

The morphological analysis of the interfaces was carried out by a scanning electron microscope, and the element distributions through the interfaces were qualitatively investigated by mapping with an electron probe micro analyzer.

2.4 Electrical properties

Electrical resistivity was measured with a four-probe method from the temperature of liquid nitrogen to room temperature. YBCO-sandwiched PZT(SIS) was investigated for the frequency dependence of the composite response by impedance analyzer (YHP 4192A).

3 Results and Discussion

3.1 Interfaces of YBCO/PZT and YBCO surfaces

Figs 1(a–c) show the SEM photographs at the interfaces of the YBCO/PZT system sintered for 420, 60 and 5 min at 1193 K, respectively. The bonding of YBCO grains is seen, although there seems to be porosity in the YBCO thick film sintered for 420 min. The thickness of the film was about 17 μm. The system maintained for 60 min had a very porous structure with a film thickness of about 33 μm; the 5 min sintered system showed a denser morphology. The SEM photographs of the surfaces of the YBCO/PZT system are shown in Fig. 2 (a–c) maintained for 420, 60 and 5 min in sintering, respectively. As shown in Fig. 2(a), well-developed bonding of the YBCO occurred with porous microstructure on the 17 μm film sin-

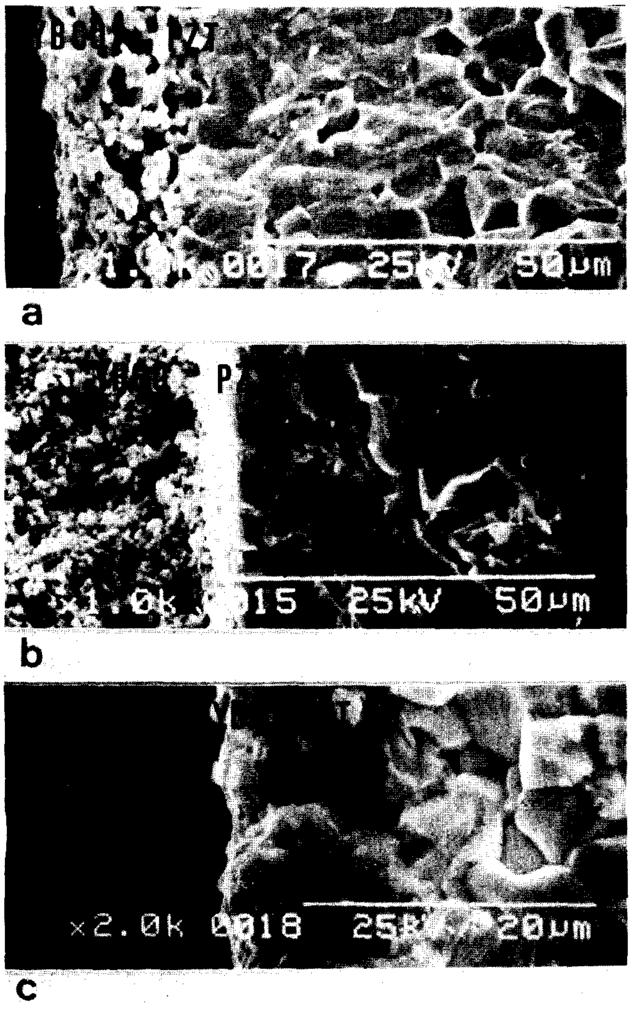

Fig. 1. SEM photographs at the interfaces of YBCO/PZT systems for each sintering time: (a) 420; (b) 60 and (c) 5 min.

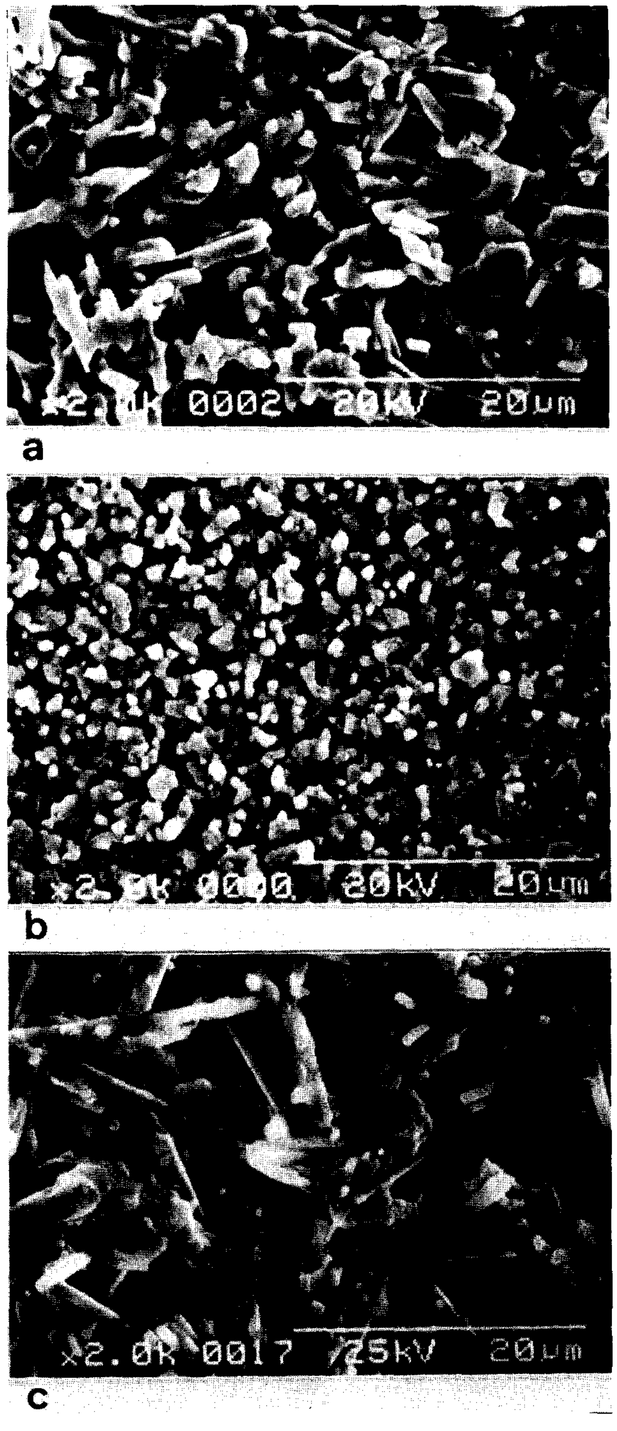

Fig. 2. SEM photographs on the surfaces in YBCO/PZT systems for each sintering time: (a) 420; (b) 60 and (c) 5 min.

tered for 420 min. The inter-particle bonding did not develop in the film which had a thickness of about 33 μm after 60 min sintering (Fig. 2(b)); this seemed to be more porous with a different microstructure from those of the 5 and the 420 min sinterings. It is not clear at this moment why the 60 min sintering shows a pronounced difference in microstructure. In the 5 min sintering, the film was thinner (5 μm) and more dense (Fig. 2(c)); the thinner film seems to be better from the view-point of morphology.

3.2 Diffusion of barium from YBCO to PZT

Y_2BaCuO_5 and $BaCuO_2$, as well as YBCO were found by XRD analysis in samples sintered for 420 min at 1193 K, and Y_2BaCuO_5 of green colour and non-superconducting character was detected together with YBCO peaks in the film sintered for 5 min. Figs 3(a and b) show the EPMA line analyses indicating a Ba diffusion distance of 35 and 5 μm into the PZT from the interfaces, respectively. The length of Ba diffusion is illustrated in Fig. 4.

3.3 Electrical resistivity and piezoelectric properties

The resistivities were measured at temperatures from that of liquid N_2 to 296 K. They indicated semiconductive temperature dependences. The

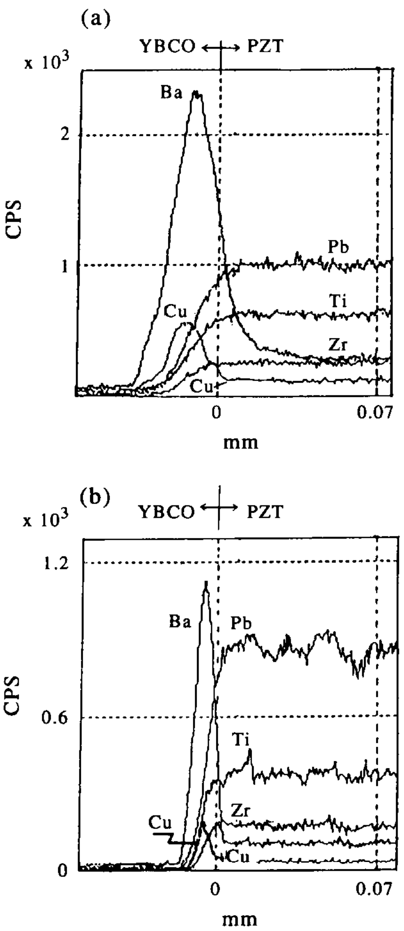

Fig. 3. EPMA line analysis at the interfaces between YBCO and PZT in each sintering time: (a) 420; (b) 60 and (c) 5 min.

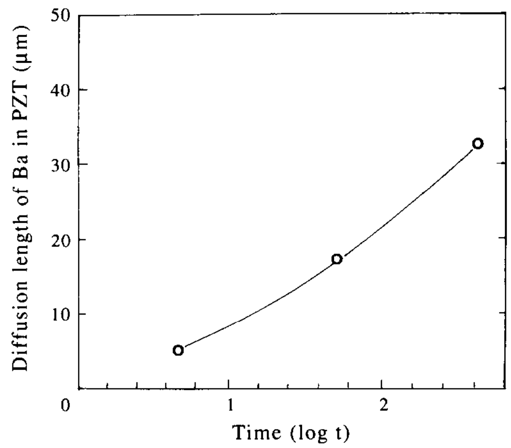

Fig. 4. Change of Ba diffusion length in PZT with sintering time at 1193 K.

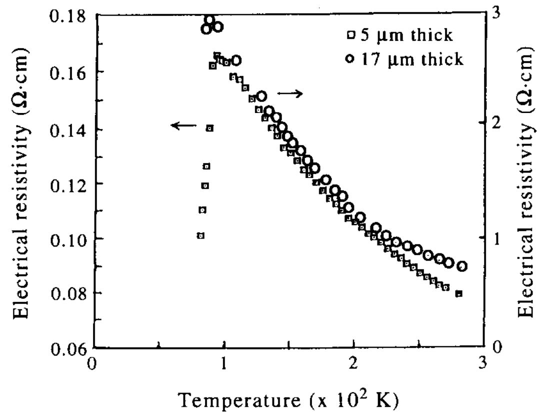

Fig. 5. Electrical resistivity changes with temperature.

critical temperature of the 5 μm YBCO film was not clear although 0·1 Ω cm was shown at a temperature of 80 K, and the resistivity at 296 K was 0·08 Ω cm. The $BaCuO_2$ produced in the 60 min sintering is a non-superconductive material. More Ba and Y were detected on the surface of the YBCO than in the bulk, due to a large amount of CO_3^{2-} ions; these elements and the ions concentrated on the surface are estimated to reduce the superconductivity.[8]

The composite piezoelectricity with electrodes of 5 μm thickness YBCO is shown in Fig. 6.

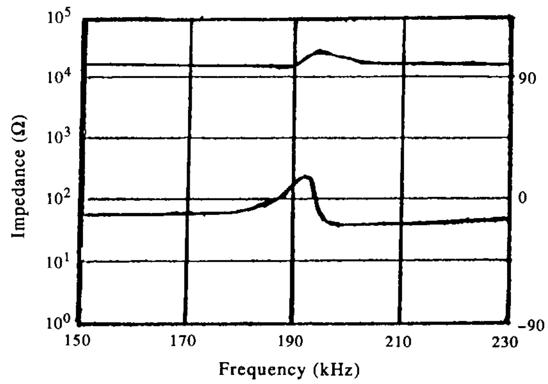

Fig. 6. Impedance analyses with frequency: (a) starting material; (b) Ba-doped PZT (electrodes were Ag).

Absorption of energy by PZT was low at a frequency around 190 kHz as indicated by impedance analysis suggesting poor piezoelectricity. The poor film density and Ba diffusion may cause the poor piezoelectricity in the YBCO/PZT/YBCO system. It is suggested that the density of the YBCO thick film is not high enough for the material to act as electrode for a device.

4 Conclusions

(1) YBCO thick films (5–33 μm) adhered onto PZT. Investigation of the interfaces revealed diffusion of Ba from YBCO to PZT; the length of the diffusion zone depended on the sintering time at 1193 K.
(2) YBCO thick film of 5 μm indicated semiconductive properties rather than superconductivity with no clear indication of a critical temperature.
(3) The thick film should be denser to function as successful electrodes for the composites consisting of superconductor/PZT/superconductor; also a film thickness of 5 μm or less can be suggested as well as a short sintering time.

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