

Piezoelectric properties of PZT/YTZ composite

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The various mole percents of YTZ spheroids (0.3mm diameters) were mixed with PZT; $\text{Pb}(\text{Zr}_{0.53}\text{Ti}_{0.47})\text{O}_3$ to investigate their effects on the piezoelectric properties. The system is the kind of diphasic composite where the YTZ spheroids does not react with the matrix PZT. The spheroids affected the decreasing of relative permittivity and k_{33} slightly as well, depending on the amount of the spheroids up to 30 mol%. The strain under the load of 5 kgf was also discussed.

1. INTRODUCTION

The composites of PZT have been theoretically studied to guide the dielectric constant and the 3-3 composite materials consisting of PZT with a coral skelton were developed introducing a concept of connectivity by Newnham et. al¹⁾. In early study, Kitayama et.al fabricated the 0-3 composite using PVDF²⁾ and polymers such as silicon rubber and plastic spheres³⁾ were mostly used for piezoelectric composites. Porous materials of PZT relating to the 3-3 composite were studied using capsule-free O_2 -HIP by one of authors⁴⁾. In porous PZT ceramics, the connectivity was improved by the capsule free O_2 -HIP leading to a larger permittivity and a smaller depolarizing factor. The polymer surrounded the PZT with the 0-3 structure in which the g-constant can be increased by decreasing the dielectric constant due to no-connectivity of PZT.

The present study relates to make a new composite using the spheroidal YTZ ball with PZT, which maintains a connectivity of PZT in diphasic structure. The dielectric properties were evaluated and the depolarizing factor was calculated for YTZ in PZT as pores in order to apply the analysis to the present study. Furthermore, Poisson's ratio was also discussed as well as strain for one of the composites.

2. EXPERIMENT

The $\text{Pb}(\text{Zr}_{0.53}\text{Ti}_{0.47})\text{O}_3$ powders (here-after, PZT; Fuji Titanium Co.,Ltd.) were mixed with 2, 5, 10, 20 and 30 mol% of YTZ ball (0.3mm Φ) for 24 hrs. After mixing, the PZT powders adhered to the YTZ balls by PVA solvent and the complexes are pressed at 196 MPa followed by an isostatic press at 300 MPa. The green pellets were sintered at 1230°C for 2 hrs in Pb-atmosphere.

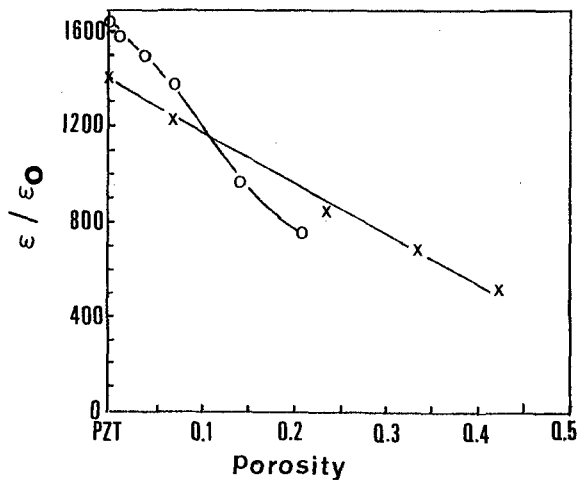


Fig. 3 Relative permittivity against porosity of Y TZ converted to the PZT.

different from that of the Y TZ/PZT composite. It is suggested that each unit cell in the Y TZ/PZT composite is largely affected by the electric field from the nearest neighbour one at least, and the effects reasonably will become significant by increasing the amount of Y TZ.

The depolarization factor (N_i) was introduced in the porous ferroelectric materials by Okazaki et al⁶). The depolarizing factor is the one of important factors in the connectivity of the porous composites, which equation is indicated as following,

$$\epsilon_{app}^{-1} = \frac{(1-p)(\epsilon_{s1}-1)}{1+N_i(\epsilon_{s1}-1)}$$

where ϵ_{app} is apparent permittivity of the ceramics including pores and ϵ_{s1} corresponds to the relative permittivity without pores, and p is porosity. The pore shapes greatly affect the depolarizing factor⁶). In the present study, the amount of Y TZ spheroid is regarded as a porosity to

the theoretical density of PZT. Figure 4 shows the depolarizing factor against the porosity. The factor increased with porosity although there was not linear in the whole, and the factor increased rapidly from around 0.14 %. This fact suggested that the existing of the rigid Y TZ spheroid has different meaning from a pore in ferroelectric ceramics and herein the effects of the nearest neighbour are significant in the Y TZ/PZT composite. The basic concept for the 3-3 or even the modified 3-3 composite model does not include the mutual effect in the other cells. Therefore, the second, the third or more non-linear terms should be taken into consideration against porosity or other substances in order that the dielectric properties of the composite are evaluated.

The frequency dependence of impedance for the 5 mol% Y TZ/PZT composite, for in-

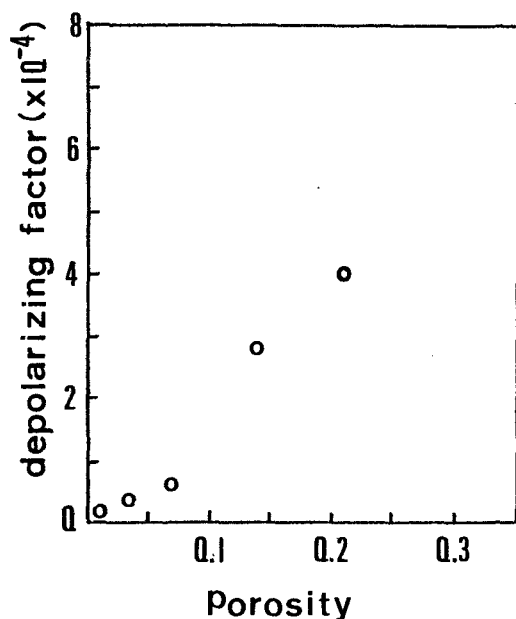


Fig. 4 Depolarizing factor against porosity of Y TZ converted to the PZT.

The composites were analyzed by XRD and SEM. The dielectric constant were measured by LCR meter at 1 kHz and the impedance analyzer were employed for the piezoelectric properties at 50 to 1 MHz as well. The strain was also measured with the 350 Ω -gauge of 1 mm in length under 5 kgf load.

3. RESULTS AND DISCUSSION

The YTZ spheroids of 5 mol% in the PZT, for instances, were closely packed as their

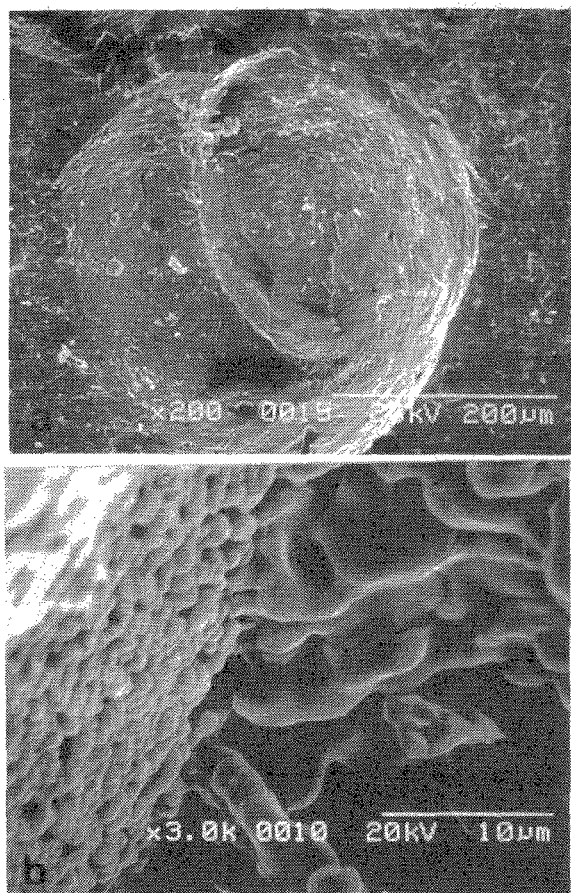


Fig.1 SEM photographs of PZT/YTZ composite;
a; YTZ ball in PZT
b; Interfaces between PZT and YTZ.

shapes were (Fig. 1-a). When more than 30 mol% of the spheroids were added, the gaps were liable to be generated although the reasons were not clear at this moment. Figure 1-b shows the SEM photograph of the boundaries between PZT and the part of the circumference of the spheroidal YTZ. The PZT grains with 10 μm approximately closely grew up and the area of the PZT in the composite was shown in Fig.2. The diphasic patterns in XRD were not clear, which presented the typical PZT patterns and the lattice constant did not differ from that of PZT without YTZ. The spheroid of YTZ did not seem to react with PZT to maintain their shapes.

The change of relative permittivity was illustrated against the converted porosity to PZT in each molar ratio of YTZ in Fig.3, and the porous PZT⁴⁾ for comparison as well. The relationship between the converted porosity and the permittivity seemed to agree with the theory of 3-0/3-3 composite model ($r=c/a=0.5\sim 0.7$)⁵⁾. Furthermore, the decreasing tendency in the permittivity of the porous PZT was

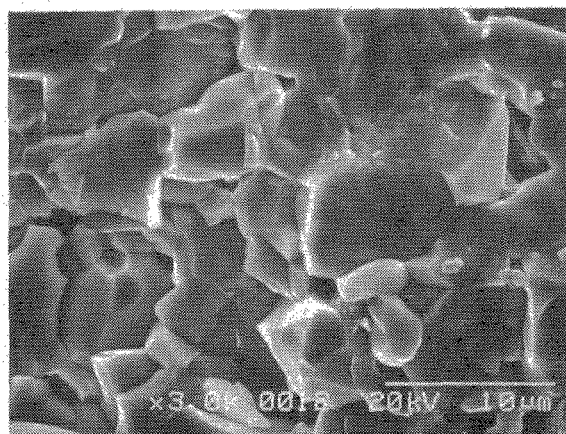


Fig.2 SEM photograph in the area of PZT in the composite;

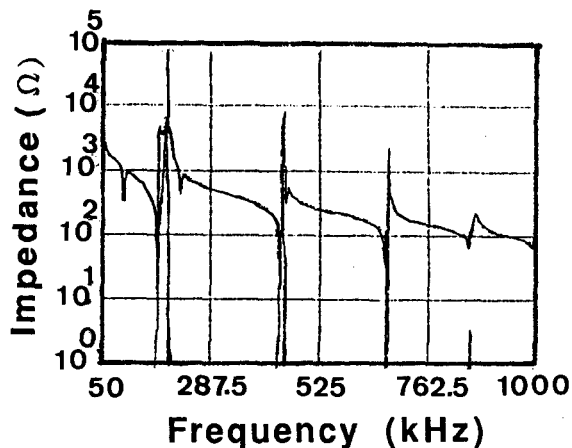


Fig.5 Frequency dependence of impedance for 5 mol% YTZ/PZT.

stances, is shown in Fig.5. The frequency patterns of each composite are similar to those of PZT without YTZ, but the peaks at higher order of the resonant and anti-resonant frequencies disappeared with increasing the amount of YTZ. The values of k_{33} changed 0.157 to 0.486 for PZT only and 30 mol% YTZ/PZT, respectively.

According to the calculation by the third overtone frequency, the Poisson's ratio slightly changed 0.25 to 0.27 with increasing the amount of YTZ, while 0.3 in ZrO_2 ⁷⁾. In the another mechanical properties, the strains were measured at the 5 kgf using the stress-strain gauge. The strain of the 5 mol% YTZ/PZT was $14.6 \mu m$ while $13 \mu m$ in the PZT only, resulting in soft material by adding the YTZ to PZT. Whether the optimum amount of YTZ is

existing should be studied in terms of the dielectric properties.

4. CONCLUSION

The composite of YTZ spheroid (0 ~ 30 mol%) with PZT was fabricated to study the effect on the dielectric properties of PZT resulting in decreasing relative permittivity. The YTZ did not chemically react with PZT, and depolarizing factor of the composite was discussed with the analogy to the dielectric analysis for the pores in PZT, and particularly the consideration of the non-linear term in the composite is proposed as well. Furthermore, electro-mechanically soft PZT was formed by adding YTZ.

REFERENCES

1. R.E. Newnham, D.P. Skinner and L.E. Cross, Mater. Res. Bull., 13(1978) 525.
2. T. Kitayama and S. Sugawara, CPM72-17, 1972, pp.1.
3. T.R. Shrout, W.A. Schulze and J.V. Biggers, Mater. Res. Bull., 14(1979) 1553.
4. S. Sugihara, T. Hayashi and K. Okazaki, Proceedings of Mater. Res. Soc. Symp., Vol. 251 (1992) 127.
5. H. Banno, Proceedings of the ISAF '94, 1994, pp. 186.
6. K. Okazaki and H. Igarashi, Ceramic Microstructure'76 (Westview Press, Boulder, Colorado) pp. 564.
7. T. Sata, "Fine Ceramic Engineering", P.206, Asakura shoten, Tokyo, 1990 (in Japanese).