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High Temperature Properties of $PbTiO_3$ / $Ba_{0.7}Sr_{0.3}TiO_3$

チタン酸鉛/チタン酸バリウムストロンチウムの高温特性

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1. Introduction

Ultrasonic transducers using sol-gel composite method have been investigated in the field of non-destructive testing (NDT). Sol-gel composite material is made by mixture of piezoelectric powder and sol-gel solution[1)]. Sol-gel composite material could be applied for NDT field ultrasonic transducer applications and new sol-gel composites with desirable characteristics have been studied. Based on past investigations, it was found that the dielectric constants of the sol-gel phase and the piezoelectric powder phase influence the sol-gel composite performance and sol-gel composite with $PbTiO_3$ (PT) piezoelectric powder has been developed because PT had relatively low dielectric constant, relatively high piezoelectric constant, and relatively high Curie temperature[2)]. In previous study, three kinds of sol-gel composite materials composed of different dielectric constant sol-gel phase, $Pb(Zr,Ti)O_3$ (PZT), $Bi_4Ti_3O_{12}$ (BiT), and $BaTiO_3$ (BT), and same piezoelectric powder phase, $PbTiO_3$ (PT), were manufactured and their properties were compared quantitatively. As a result, PT/BT, sol-gel composite with the highest dielectric constant sol-gel phase, showed the higher d_{33} value and signal strength. In this study, PT/$Ba_{0.7}Sr_{0.3}TiO_3$(BST), which is a composite material made by PT powder and BST sol-gel solution, will be tested because BST has a higher dielectric constant than BT[3)] and poling facility is expected compared with PT/BT.

2. Sample fabrication

First, PT powders and BST sol-gel solution were prepared. BST sol-gel solution was self-manufactured according to the reference[4)]. The sol-gel precursor was produced by dissolving barium acetate and strontium acetate in water. This solution was added to a mixture of titanium butoxide, acetic acid, and methanol. After mixing, agitation was carried out. Table 1 shows the mass of each reagent to synthesize BST sol-gel solution.

Table 1 BST sol-gel raw materials

Reagent	Mass (g)
Titanium(IV) Butoxide	3.16
Acetic acid	6.90
Methanol	2.59
Barium acetate	1.66
Strontium acetate (0.5H_2O)	0.60
Water	5.18

In order to confirm sol-gel solution synthesize result, X-ray diffraction (XRD) was performed for BST sol-gel derived powder after thermal process. XRD result of BST sol-gel powder is shown in **Fig. 1**. As a result, molar ratio of barium and strontium in this study is 0.67:0.33. The molar ratio of barium was slightly lower than 0.7 because of undesired $BaCO_3$ was also synthesized. However, the molar ratio of $BaCO_3$ was much smaller than BST and it could be ignored in this study.

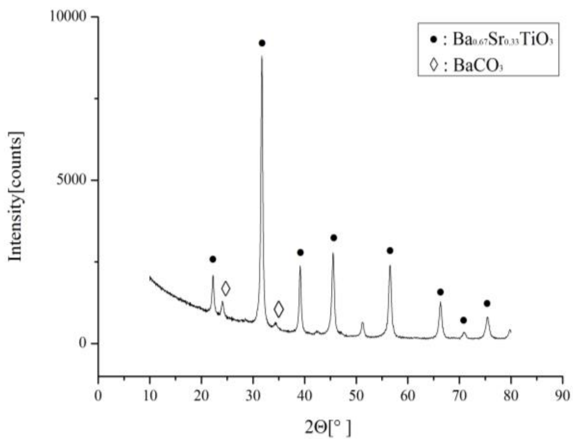

Fig. 1 XRD result of BST sol-gel powder

After preparing BST sol-gel solution, an ultrasonic transducer made by PT/BST sol-gel composite film was fabricated. The mixtures of PT powders and BST sol-gel solution were ball milled for more than a day. Then, the mixtures were sprayed onto titanium substrates. Titanium substrates has dimensions of ~3mm thickness, ~30mm length, ~30mm width. After spray coating, drying process at 150℃, and firing process at 650℃ for 5 minutes each were operated. Those spray coating process and thermal process were repeated until film thickness film reached 70μm. After film fabrication, poling was operated by corona discharge at room temperature. The output voltage of power supply was 23kV. Poling process was operated for 5 minutes.

3. Experimental results

Optical image of PT/BST film onto titanium substrate is shown in **Fig.2**. Film thickness of PT/BST was measured by a micrometer and the values were ~70μm. d_{33} of PT/BST film was measured by ZJ-3B piezo d33 meter and the values was 1.2pC/N. Surface roughness was also very high. These results indicated that poor film quality due to high porosity. Further fabrication process optimization should be carried out.

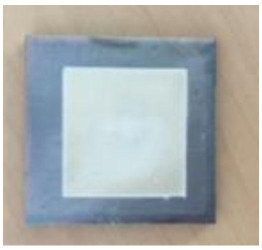

Fig. 2 Optical images of PT/BST film onto titanium substrate.

Ultrasonic responses of the PT/BST film in pulse-echo mode were recorded at room temperature and reflected echoes from the bottom surface of titanium substrate was shown in **Fig. 3**. It should be noted that PT/BST poling was progressed by pulser/receiver machine during measurement. In Fig. 3, signal amplitude and signal-to-noise ratio (SNR) was low because appropriate ratio of PT powder and BST sol-gal has not been established yet as mentioned above. Fig. 4 shows FFT result of PT/BST. Center frequency was ~14MHz. High temperature performance will be investigated.

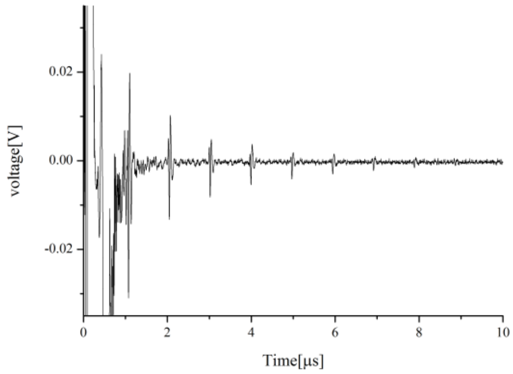

Fig. 3 Ultrasonic response of PT/BST ultrasonic transducer on ~3mm thick titanium substrate.

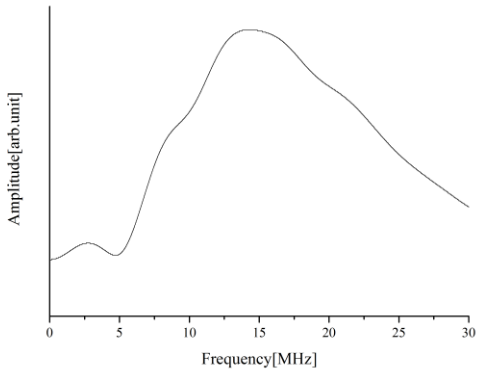

Fig. 4 FFT result of PT/BST

4. Conclusions

PT/BST sol-gel composite film was fabricated onto titanium substrates. PT/BST demonstrated d_{33} value was 1.2pC/N. Clear multiple echoes from 3mm thick titanium substrate was observed. Center frequency of PT/BST was ~14MHz. Further investigation is necessarily for fabrication process optimization and high temperature capability of PT/BST.

References

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