Investigation of AC poling of sol-gel composites

Ryota Ono[1†], Takeshi Hamada[1], and Makiko Kobayashi[1*]
([1]Kumamoto Univ.)

1. Introduction

Non-invasive diagnostic techniques using ultrasound are widely used in the medical and industrial fields. However, conventional ultrasonic transducers require a backing material to reduce excess vibration and a couplant to match acoustic impedance, making the probes large and unusable at high temperatures. Ultrasonic transducers using sol-gel composites don't require baking materials and coulplant, thus achieving flexibility and heat resistance[1)].

Recently, ultrasonic phased arrays using the sol-gel composite method have been developed[2)]. In improving the performance of phased arrays, a large number of elements must exhibit equivalent ultrasonic performance. DC poling, in which a DC voltage is applied to a sample in an oil-bath, is used for common bulk piezoelectric ceramics. Although the oil-bath works to suppress dielectric breakdown, corona discharge poling is used for sol-gel composites because of the risk of oil contamination. However, corona discharge poling has a problem with polarization uniformity. There are reports that AC poling improves piezoelectric properties compared DC poling[3,4)]. In addition, since the voltage is time-varying., it may not cause dielectric breakdown, and is attracted attention as a new polarization method for sol-gel composites.

Since the effectiveness of AC poling for $Pb(Zr,Ti)O_3$(PZT)/PZT, the most common sol-gel composite, has already been confirmed, in this study, AC poling was performed on $Bi_4Ti_3O_{12}$(BiT)/PZT, which shows excellent performance at 200~500℃[5)].

2. Sample Fabrication

First, BiT powders and PZT sol-gel solution were mixed by bail milling machine for 24 hours to adjust to the appropriate viscosity. The mixed sol-gel composite solution was sprayed onto a 3mm thick titanium substrate by an automatic spray system. Then, the piezoelectric film dried at 150 ℃ for 5minutes and annealed at 200℃ for 5minutes. The process from spraying to annealing was repeated 5 times to achieve a film thickness of 25μm. After deposition, platinum top electrode with 4mm diameter was deposited by sputtering. The atmosphere during electrode fabrication was argon gas, and sputtering was performed at 1.3Pa for 25minutes. The completed BiT/PZT optical image is shown **Fig. 1**.

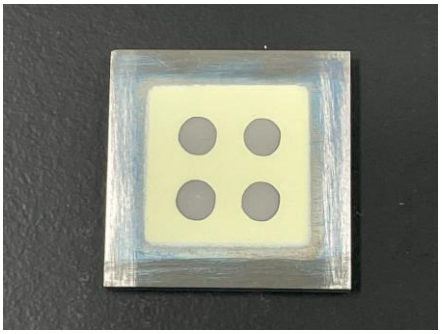

Fig. 1 Optical image of the completed BiT/PZT sample

3. Experimental method

After sample preparation, polarization is performed. AC poling was performed using an AC/DC stabilized power supply(PCR1000WEA, KIKUSUI ELECTRONICS CORPORATION) The AC voltage supplied from the power supply is applied to the piezoelectric film through platinum electrode. A sample with corona discharge poling was also prepared for comparison. Corona discharge poling was performed on the piezoelectric film before fabricating a top electrode at a DC voltage of approximately 40kV for 5minutes with the humidity during polarization set at less than 20%.After corona discharge poling, the platinum electrode was fabricated by sputtering. For judgement of polarization capability and comparison of AC and corona discharge poling, piezoelectric constant d_{33} measurements and ultrasonic measurements using the pulse echo method were performed on the polarized sample.

4. Experimental results

First, polarization was performed at applied voltage of 200V/rms, application time of 1min, frequency of 1Hz, and end phase angel of 280°. The end phase angle affects the direction of polarization, which in this experiment is positive. As a result, polarization was successful without dielectric breakdown.

Next, d_{33} and ultrasonic measurements of samples polarized by AC poling with those polarized by corona discharge poling. **Fig. 2** shows the ultrasonic response, **Table I** shows the results of d_{33} and ultrasonic sensitivity measurements of BiT/PZT for each polarization method, and **Table II** shows the frequency analysis results. d_{33} was measured using a d_{33} meter(ZJ-3B , Institute of Acoustics Chinese

Academy of Sciences), and ultrasonic sensitivity was calculated using Equation(1).

$$Sensitivity = -(20\log V_1/V_2 + P/RGain) \quad (1)$$

The reference amplitude V_1 being 0.1V and V_2 being $V_{p\text{-}p}$ of the third reflected wave form the substrate bottom. The d_{33} and ultrasonic sensitivity of AC poling of BiT/PZT are smaller than those of corona discharge poling, but the variations are small. There is no difference in ultrasonic waveforms and frequency characteristics(**Table III**) for each polarization method. The advantage of AC poling in BiT/PZT is smaller than that in PZT/PZT, with d_{33} and sensitivity comparable to those of corona discharge poling, and with greatly reduced variability.

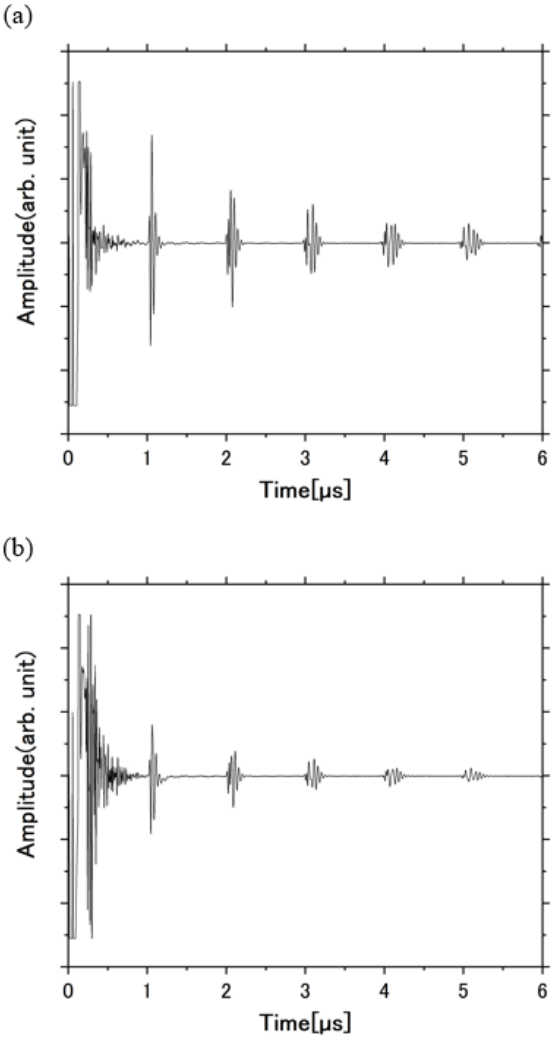

Fig. 2 The ultrasonic response of BiT/PZT samples fabricated on 3mm thick titanium substrate poled by (a) corona discharge, (b) AC

Table I The results of d_{33} and ultrasonic sensitivity of BiT/PZT for each polarization method

Poling method	d_{33}[pC/N]	*Sensitivity*[dB]
Corona	12.0±0.2	−4.1±0.4
AC	3.7±0.1	−25,1±0.2

Table II The results of d_{33} and ultrasonic sensitivity of PZT/PZT for each polarization method

Poling method	d_{33}[pC/N]	*Sensitivity*[dB]
Corona	−36.7 ± 2.9	1.8 ±0.9
AC	−35.1 ±0.8	7.8 ±0.5

Table III The frequency characteristics of BiT/PZT

Poling method	6dB bandwidth			
	f_L [MHz]	f_H [MHz]	f_C [MHz]	BW [MHz]
Corona	16.7	27.8	22.3	11.1
AC	15.2	28.1	21.7	12.9

5. Conclusion

In this study, 25 μm BiT/PZT sol-gel composites were fabricated on 3mm thick titanium substrate and AC poling was performed, Compared to corona discharge poling. As a result, the d_{33} and ultrasonic sensitivity were smaller, but no significant differences in ultrasonic responses and variation between electrodes was small, it can be said that AC poling is a suitable method for phased array applications. The advantage of AC poling over corona discharge poling was greater for PZT/PZT than for BiT/PZT. The polarization difficulty of corona discharge poling is affected by the dielectric constant of the piezoelectric powder and sol-gel solution, the polarization difficulty of PZT/PZT is higher than BiT/PZT. Therefore, it is suggested that AC poling may be effective for materials that were difficult to polarize by corona discharge poling. Future works will include optimization of poling conditions and high-temperature operation tests.

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