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Corona discharge polarity influence on $Pb(Zr,Ti)O_3/TiO_2$

コロナ放電の極性の違いによる $Pb(Zr,Ti)O_3/TiO_2$ への影響

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1. Introduction

High temperature ultrasonic transducers using have been investigated in the field of non-destructive testing (NDT) for safety assuarance. It is difficult to apply commercial ultrasonic transducer in this application, mainly due to lack of high temperature durability caused by backing material and couplant. Sol-gel composite ultrasonic transducers have been developed to elminate backing material and couplant problems.[1-5)] Sol-gel composites based on $Pb(Zr,Ti)O_3$(PZT) powder and PZT sol-gel solution PZT/PZT were shown to have improved high-temperature properties due to negative corona polarization. However, PZT as a sol-gel solution requires a high sintering temperature of 650°C. Since it is difficult to sinter at 650°C, a low-temperature sintering solution is desired for the actual production of PZT.

In this study, the sol-gel solution was changed from PZT to TiO_2 and the polarization was positive and negative, respectively. The sol-gel solution of TiO_2 can be sintered at a low temperature of 400°C. If the electric field concentration is greater due to the change to TiO_2, the samples polarized by negative corona discharge may have better high temperature durability. Therefore, samples positively and negatively polarized samples are prepared and compared.

2. Sample fabrication

PZT/TiO_2 sol-gel composite was made by sol-gel spray technique. First, PZT powders and TiO_2 sol-gel solution were prepared. TiO_2 sol-gel solutions was self-manufactured. PZT powders are commercially available. The mixtures of PZT powders and TiO_2 sol-gel solution were ball milled. Then, the mixtures were sprayed onto titanium substrate by automatic spray machine. The dimensions of titanium substrate were 30mm × 30mm × 3mm. This substrate was chosen due to low thermal capacitance and high temperature durability. After spray coating, the sample was dried at 150°C for 5 min. After drying, the sample was fired at 400°C for 5 min as well. Those spray coating process and thermal process were repeated until film thickness reached 50µm. After film fabrication, poling was operated by positive corona discharge and negative corona discharge at room temperature. The output voltage of the positive power supply was 34.0 kV. And the output voltage of the negative power supply was 35.0 kV. Optical image of PZT/TiO_2 film onto titanium substrate is shown in **Fig. 1**. Film thickness of positively polarized PZT/TiO_2 was measured by a micrometer and the value were 54µm. Film thickness of negatively polarized PZT/TiO_2 was 53µm. Piezoelectric constant d_{33} was measured by ZJ-3B piezo d33 meter and the value was 9.8pC/N and -8.3pC/N. After these processes, high temperature silver paste was fabricated on the film as a top electrode. The top electrode diameter was about 1cm. For drying silver paste, thermal process at around 100°C were carried out for 2 h.

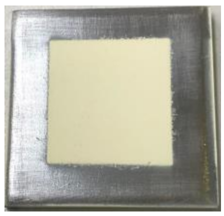

Fig. 1 Optical image of PZT/TiO_2 film

3. Experimental results

Ultrasonic responses of the PZT/TiO_2 samples in pulse-echo mode were recorded from room temperature to 380°C. It should be noted that this temperature is not the sample temperature, but the temperature of the furnace indicator, so the actual temperature should be much lower. Platinum wires were used as electrical cables and electrical connection between electrodes and wires were established by a ceramic weight. Samples were set inside an electrical furnace and measurement data was recorded by a digital oscilloscope. Firstly, the ultrasonic response was measured at room temperature to check the piezoelectricity. Results are shown in **Fig. 2** and **Fig. 3**. Clear multiple echoes from the bottom surface of the titanium substrate were obtained and it was confirmed that poling was successful.

Thereafter, PZT/TiO_2 samples were heated to 380°C. The ultrasonic response was measured every 50 °C until 200 °C. The ultrasonic response was then measured every 20°C. The electric furnace was held for 5 minutes to keep the temperature. Clear multiple echoes were still observed in both samples at 380°C. However, as mentioned before, the actual temperature of the substrate cannot be accurately determined because substrate temperature measurement was not operated in this time.

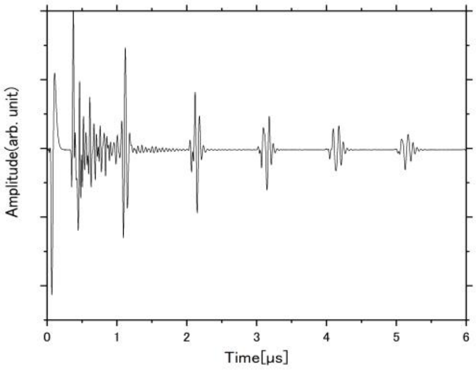

Fig. 2 Ultrasonic response of PZT/TiO_2 film poling by positive corona discharged at room temperature.

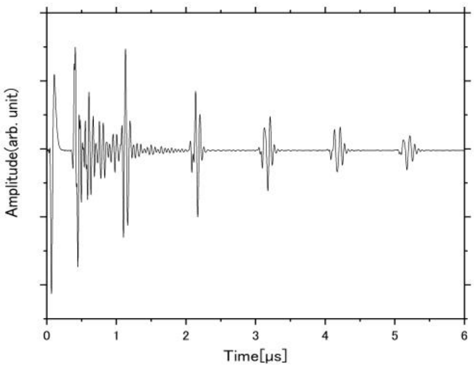

Fig. 3 Ultrasonic response of PZT/TiO_2 film poling by negative corona discharged at room temperature.

As the temperature rises, the piezoelectricity weakens. In order to determine the temperature effect quantitatively, sensitivity was calculated as following equation;

$$\text{Sensitivity} = -(20\log_{10}\frac{V_1}{V_2} + \text{Gain of P/R}) \quad (1)$$

where V_1 is the reference amplitude, which is 0.8 $V_{p\text{-}p}$ in this experiment, V_2 is the $V_{p\text{-}p}$ of the third reflect echo from the bottom surface of the titanium substrate. P/R means pulser/receiver so that this equation calculates true required gain of pulser/receiver in order to achieve 0.8V. -1 is multiplied to assist intrinsic understanding. The temperature dependence of PZT/TiO_2 sensitivity is shown in **Fig. 4**. The positively and negatively polarized PZT/TiO_2 and negatively polarized PZT/TiO_2, respectively, show almost the same temperature dependence. This may be due to the small field concentration of the TiO_2 sol-gel solution.

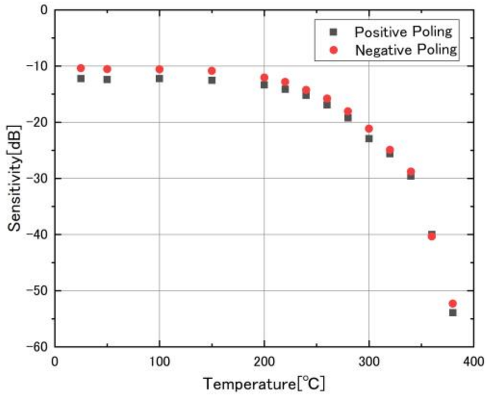

Fig. 4 Sensitivity of PZT/TiO_2 film

4. Conclusion

54µm and 53µm thick PZT/TiO_2 sol-gel composites were fabricated on a 3mm thick titanium substrate to compare negative corona discharge and positive corona discharge. The piezoelectric constant d_{33} was 9.8pC/N and -8.3pC/N. Pulse-echo mode ultrasonic measurement was carried out from room temperature to 380°C. This study shows that positively and negatively polarized PZT/TiO_2 have almost the same temperature dependence. Further research is required for comparison in thermal cycle test.

References

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