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Room-Temperature Poling of $CaBi_4Ti_4O_{15}$ /Pb(Zr,Ti)O_3 Sol-Gel Composite films by Pulse Discharge

パルス放電による $CaBi_4Ti_4O_{15}$ /Pb(Zr,Ti)O_3 ゾルゲル複合体膜の室温分極

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1. Introduction

High-temperature ultrasonic transducers by sol–gel composites have been developed for industrial applications.[1-2] Sol-gel composite material is made by ferroelectric powders and sol-gel solution and composed of three phases, ferroelectric powder phase, dielectric sol-gel phase, and air phase. Sol-gel composites ultrasonic transducers have several merits. First, couplant is unnecessary because sol-gel composite film has good acoustic coupling with a substrate since oxide layer is created between the substrate and the piezoelectric film during film fabrication thermal process. Second, backing material is also unnecessary because tiny pores are manufactured inside sol-gel composite film, which cause ultrasonic loss and reduce the ringing effect. Third, sol-gel composite film has high thermal shock resistance because of tiny pores which contribute no necessity of backing material.

$CaBi_4Ti_4O_{15}$/Pb(Zr,Ti)O_3(CBT/PZT) sol-gel composite is promising piezoelectric material for high temperature ultrasonic transducer applications above 500 °C. In previous study, CBT/PZT was poled by positive DC corona discharge. CBT/PZT sol-gel composites has been developed for NDT above 550 °C and it showed clear multiple echoes at 550 °C.[3] However, efficient poling of CBT/PZT required high temperature in order to obtain high piezoelectricity owing to the high coercive field of the CBT powder phase. Therefore, poling process takes a long time because of the long heating time and cooling time. If CBT/PZT can be poled at room temperature, the manufacturing cost is reduced and it will be suitable for on-site fabrication. In this research, a new poling method using by pulse corona discharge generation at room temperature was developed for CBT/PZT.

2. Fabrication of CBT/PZT films

The fabrication process was based on a sol–gel spray technique developed previously. The CBT/PZT films were fabricated onto titanium substrates by spray coating. First, CBT powders were mixed with self-manufactured PZT sol-gel solution and ball-milled for more than one day until appropriate viscosity for spray coating was achieved. Then it was sprayed onto titanium substrates. The area of the titanium substrate was 9 cm^2, and the thickness was 3 mm. After spray coating, they were heated as follows: drying at 150 °C for 5 min on a hot plate and firing at 650 °C for 5 min by a furnace. In this research, those spray coating process and thermal process were repeated 5 times to obtain a 50-μm-thick CBT/PZT films. After ~50 μm thick films were fabricated on the titanium substrates, poling processes were performed.

3. Poling process

In this study, CBT/PZT films were polarized by two poling methods. First, traditional poling is corona discharge using a high-DC-voltage source at a high temperature using a hot plate. A positive corona discharge was used for poling because a high electrical field could be applied on the piezoelectric film without dielectric breakdown. In this experiment, output voltage of the high-DC-voltage source was +30 kV and the poling time was 15 min, and the sample was heated to 400 °C. Next, the new poling method is corona discharge poling at RT using a self-built high-pulse-voltage source. In this experiment, pulse repetition frequency was 2kHz, the output voltage was +21 kV, and the poling time was 1 min. After corona poling, ~5 mm diameter silver top electrode was fabricated on the CBT/PZT film. Optical images of two samples after fabricating silver top electrodes were shown in **Fig. 1**.

(a) (b)

Fig. 1 CBT/PZT film optical images poled by (a) DC corona at 400 °C, and (b) pulse corona at RT.

4. Experimental results

First, pulse-echo measurement result poled by DC corona at high temperature is shown in **Fig. 2.** The P/R gain was 35 dB. Clear multiple reflected echoes from the bottom surface of the titanium substrate were confirmed. Piezoelectric constant d_{33} of that sample was 6.3 pC/N. Measurement result poled by pulse discharge at RT is shown in **Fig. 3**. The used gain was 50 dB. Clear multiple reflected echoes were also confirmed as well. From this result, CBT/PZT was successfully poled by pulse corona discharge at RT. Piezoelectric constant d_{33} of CBT/PZT poled at RT was 4.2 pC/N. From those results, poling efficiency of CBT/PZT poled by pulse discharge at RT was slightly lower than poled by DC corona at HT though it was improved by 5dB than previous study.[3].

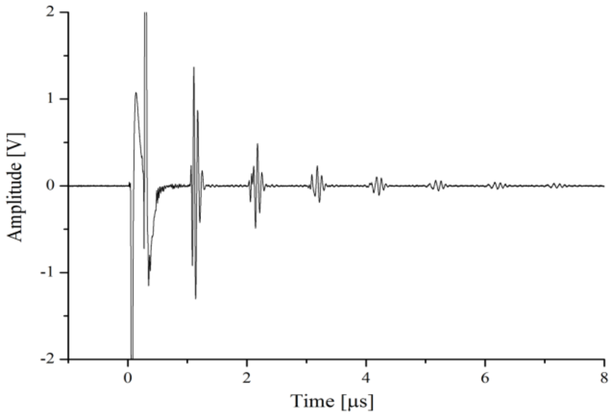

Fig. 2 Ultrasonic response of CBT/PZT poled by DC corona at high temperature onto ~3-mm-thick titanium substrate.

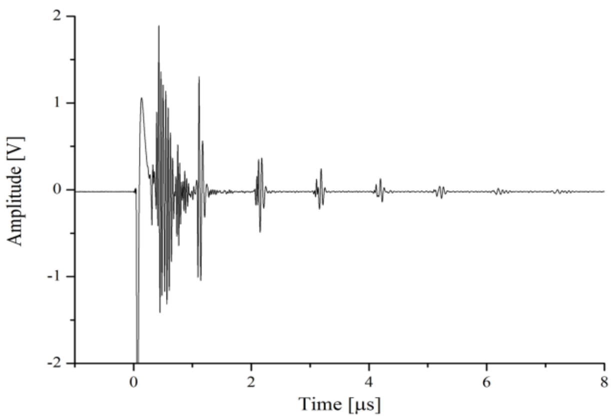

Fig. 3 Ultrasonic response of CBT/PZT poled by pulse discharge at RT onto ~3-mm-thick titanium substrate.

Next, CBT/PZT poled by pulse corona was tested by a furnace at 500 °C for 2 h in order to investigate thermal durability. **Fig. 4** show the ultrasonic response of CBT/PZT transducer at 500 °C after 2 h. Signal amplitued at 500 °C was lower than that at RT though the same gain was used and clear multiple reflected echoes were confirmed. Temperature stability was much improved than previous study.[3] So, It was found that CBT phase has been sufficiently poled. New pulse voltage source machine has been developed for further improvement.

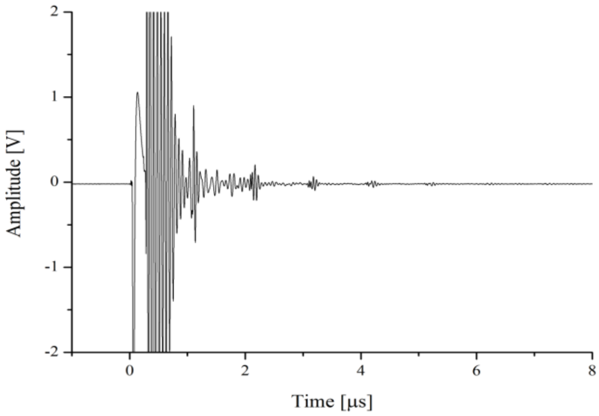

Fig. 4 Ultrasonic response of CBT/PZT transducer in a furnace at 500 °C after 2 h.

5. Conclusions

In this experiment, the poling of CBT/PZT was carried out by new poling method for pulse discharge at RT. Although The sample showed lower piezoelectric properties and ultrasonic response than those of the sample poled by DC corona at high-temperature, multiple reflected echoes and sufficient thermal durability was confirmed. Therefore, it could be concluded that pulse discharge poling of CBT/PZT at RT was demonstrated successfully.

References

1. M. Kobayashi and C.-K. Jen: Ultrasonic transducers: materials design and application, Ed. by K. Nakamura and S. Ueha, Woodhead Publishing (2012) 408.
2. K. Kimoto, M. Matsumoto, T. Kaneko, and M. Kobayashi: Jpn. J. Appl. Phys. 55 (2016) 07KB04.
3. Y. Inada, T. Inoue, M. Kobayashi, H. Nagata, and T. Takenaka: Jpn. J. Appl. Phys. **53** (2014) 07KB10.