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Resistance Effect of $LiNbO_3$-Based Sol-Gel Composites on Poling Temperature

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1. Introduction

In recent years, non-destructive testing (NDT) has been in increasing demand in the industrial field [1-2] and is used as a means for early detection of defects.

Conventionally used ultrasonic transducers require a couplant and a backing material, so it is difficult to measure at a high temperature. However, the sol-gel composite ultrasonic transducers do not require them and can be used at higher temperatures. Therefore, it is expected to be used for NDT in high-temperature environments.

$LiNbO_3$ (LN) is a piezoelectric material with a very high Curie temperature of 1200 ℃, and has the advantage of not containing lead. However, it has the disadvantage of being very difficult to poling. In previous studies, sol-gel composite ultrasonic transducers of LN/Pb(Zr,Ti)O_3 (PZT), LN/$Bi_4Ti_3O_{12}$ (BiT), LN/TiO_2(TO)+Sr, and LN/Aluminum Oxide (AO) have been developed[3-5]. Among them, LN/TO+Sr and LN/AO were reported to be poled at 200 °C and 400 °C, respectively. It is thought that the cause was poling due to the high resistance of TO+Sr and AO. But the specific characteristics have not been clarified. The purpose of this study was to investigate the cause of low-temperature poling by measuring the electrical characteristics of LN/TO+Sr and LN/AO.

2. Sample Fabrication

LN/TO+Sr and LN/AO were fabricated by an automatic spraying device. The sol-gel solution TO+Sr and AO are mixed with LN piezoelectric powder by a ball milling machine, respectively. The sol-gel composite solution was adjusted to the appropriate viscosity and sprayed on Inconel substrate with 3 mm thick, 30 mm length, 30 mm width by automatic spraying device. In the case of LN/TO+Sr, dry at 150 °C and anneal at 400 °C for 5 min. This process was repeated 6 times to reach a film thickness of 50 µm. In LN/AO, the annealing temperature was changed to 650 °C and the same process was repeated 9 times to reach a film thickness of 50 µm.

These samples were poled by negative corona discharge. LN/TO+Sr was heated in an electric furnace at 200 °C for 10 min and then poled for 5 min. Similarly, LN/AO was heated at 400 °C and poled for 10 min. When both samples were poled, the output voltage of the power supply was about 45 kV. The distance between the tip of the needle and the film was 30 mm for LN/TO+Sr and 20 mm for LN/AO. The piezoelectric constants d_{33} of the LN/TO+Sr and LN/AO after poling were -2.4 pC/N and -1.2 pC/N, respectively.

The top electrode was made using the sputtering method. This method eliminates individual differences in electrodes. In this case, platinum was used to improve high-temperature durability. Sputtering was performed at 1.3 atm and 20 mA current for 25 min while filled with argon gas. The diameter of the top electrode was 10 mm. The optical image of the fabricated LN/AO sample is shown in **Fig. 1**.

The ultrasonic response was measured to confirm that the fabricated samples were working properly. It was measured by the pulse echo mode of the pulsar/receiver. **Fig. 2** shows the ultrasonic response of LN/AO at 700 °C.

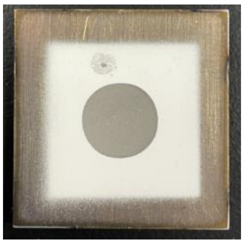

Fig. 1 Optical image of LN/AO film with Pt top electrode made by sputtering method on 3 mm thick Inconel substrate

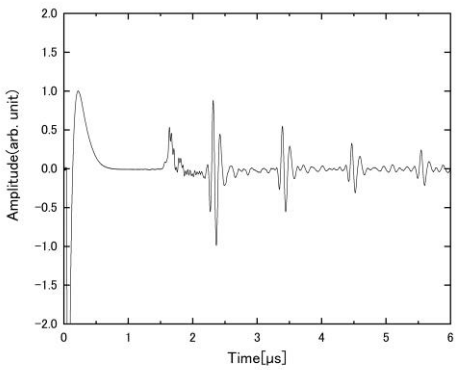

Fig. 2 Ultrasonic response of LN/AO at 700 ℃

3. Experimental results

The resistance, capacitance, and dielectric loss were measured using an LCZ meter. Samples were heated by the furnace and measured until substrate temperatures increased from room temperature (RT) to 700 °C. Measurements were conducted by LabVIEW's automatic measurement program. BiT/BiT was also used for comparison because of its high resistance at RT. The results of the resistance and capacitance measurements at LN/TO+Sr, LN/AO, and BiT/BiT are shown in **Fig. 3** and **4**, respectively.

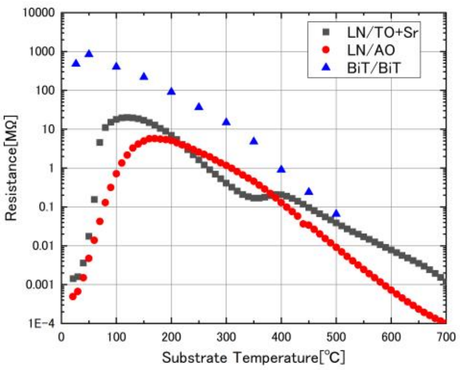

Fig. 3 Resistance shift of LN/TO+Sr, LN/AO, and BiT/BiT with substrate temperature

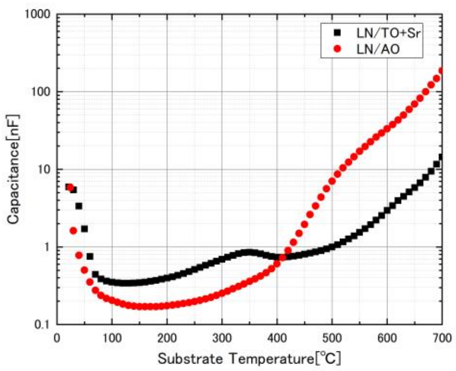

Fig. 4 Capacitance shift of LN/TO+Sr and LN/AO with substrate temperature

Fig. 3 shows that the resistance of LN/TO+Sr and LN/AO increased from 100 °C to 200 °C, where poling is considered to be performed. LN/TO+Sr peaked in resistance at 120 ℃ and was around 20 MΩ. In contrast, the resistance of LN/AO peaked at 170 ℃ and was about 6 MΩ. Therefore, it is considered that the increase in resistance effectively applied an electric field to LN and allowed it poling at low temperatures. However, BiT/BiT has a high resistance of about 478 MΩ at RT, although the poling temperature is 400 °C. It is considered that an electric field was not applied to the piezoelectric powder because the resistance of the sol-gel layer was too high.

Fig. 4 shows that the capacitance of LN/TO+Sr exceeded that of LN/AO from 100 °C to 200 °C. LN/TO+Sr ranged from 340 pF to 400 pF, while LN/AO ranged from 170 pF to 200 pF. The relative permittivity of LN/TO+Sr was 106.4 at RT and 6.1 at 120 °C, while that of LN/AO was 104.7 at RT and 3.1 at 170 °C. The dielectric loss is not significantly different between LN/TO+Sr and LN/AO, suggesting that LN/TO+Sr is poling more effectively due to the difference in relative permittivity. Both samples have high permittivity at RT. However, the dielectric loss is also large, so the real permittivity is considered low. The capacitance of LN/AO increases sharply after 400 °C, while that of LN/TO+Sr shows a similar trend after 500 °C. The capacitance of LN/AO is 186 nF at 700 °C, whereas that of LN/TO+Sr is 14 nF. This indicates that the capacitance of LN/AO is very high in a high-temperature environment.

4. Conclusion

In this study, the cause of the ability of LN/TO+Sr and LN/AO poling at low temperatures was investigated based on their electrical characteristics. Pt electrode made by sputtering method was confirmed to work properly.

Resistance measurements showed that LN/TO+Sr had a peak at 120 °C and a resistance of about 20 MΩ. LN/AO had a peak at 170 °C and a resistance of about 6 MΩ. On the other hand, the resistance of BiT/BiT is about 478 MΩ at RT, but it is not poled at RT. Therefore, it is considered that an electric field was efficiently applied to the LN and poled due to the appropriate range of resistance in LN/TO+Sr and LN/AO.

The capacitance of LN/TO+Sr was larger from 100 °C to 200 °C compared to LN/AO. This means the permittivity is also larger. As a result, LN/TO+Sr is considered to have a larger d_{33} value and ultrasonic response than LN/AO, due to high permittivity.

References

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