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$PbTiO_3$/PZT Sol-Gel Composite for Ultrasonic Transducer around 300°C

$PbTiO_3$/PZT ゾルゲル複合体の 300 度用超音波トランスデューサ応用に関する研究

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1. Introduction

In industrial facilities, regular monitoring of the equipments and structures at high temperature is desired for safety assurance. Especially for nuclear power plants, on-line monitoring during operation becomes critical, since the accident results in huge influence for long term. The temperature of cooling pipe is around 300°C and various piezoelectric materials that can be used as high temperature ultrasonic transducers have been tested, however, above 300°C is challenging conditions [1,2].

High temperature ultrasonic transducers made by sol-gel composites have advantages such as thermal shock resistance, good signal-to-noise ratio (SNR), and backing and couplant unnecessary. [3] In Previous study, it was suspected that the composite consisting of sol-gel solution with high dielectric constant and piezoelectric powder with low dielectric constant could have superior sensitivity. [4] Therefore $PbTiO_3$ (PT) was considered as good piezoelectric powder canditate because of high Curie temperature around 500°C and low dielectric constant, even though it has not been popular material becuase poling was difficult due to its high coercive field.[5] In this study, PT/PZT sol-gel composite was developed and the properties were compared with PZT/PZT and $Bi_4Ti_3O_{12}$ (BiT)/PZT to evaluate ultrasonic transducer peformance.

2. Fabrication of ultrasonic transducers

Ultrasonic transducers made by PT/PZT, PZT/PZT and BiT/PZT sol-gel composite films were manufactured. The mixture of PZT sol-gel solution and each piezoelectric powder, PT, PZT and BiT were ball milled for more than two days. Then the mixtures were sprayed onto few mm thick metal substrates. It should be mentioned that PT/PZT films were fabricated onto titanium substrates with elevated temperature durability for thermal cycle test, whereas PZT/PZT and BiT/PZT were fabricated onto steel substrates, which were commonly used for industrial structures. After spray coating, drying process at 150°C, and firing process at 650°C were operated. Those spray coating process and thermal process were repeated in order to obtain ~50μm thickness. After corona poling, top electrodes were fabricated onto piezoelectric films.
The poling temperature of PZT/PZT and BiT/PZT films was room temperature, whereas different poling temperatures, room temperature and 200°C, were tested for PT/PZT. The dimension of PT/PZT film was ~20mm × ~20mm and the diameter of silver top electrode was ~3mm, respectively.

3. Experimental results

3.1. Performance comparison

First, PT/PZT, PZT/PZT and BiT/PZT ultrasonic transducers onto metal substrates were compared at room temperature. The capacitance of each film was measured in order to calculate dielectric constant. As a result, the relative dielectric constant of PT/PZT poled at 200°C, PZT/PZT and BiT/PZT were ~130, ~220 and ~105, respectively. Piezoelectric constants d_{33} of PT/PZT and PZT/PZT were measured by ZJ-3EN Piezo d33 meter supplied by Institute of Acoustic, Chinese Academy of Science and the values are ~40pC/N and ~70pC/N, respectively. For sensor performance evaluation, another type of piezoelectric constant g_{33}, which is derived by dielectric constant/d_{33}, should be also considered and those values were ~35mV ·N and ~36mV·N, respectively. The values of g_{33} between PT/PZT and PZT/PZT were very close and it indicated that the sensor performance of PT/PZT could be comparable with PZT/PZT.

To evaluate the transducer capabilities, those transducers made by PT/PZT poled at room temperature and 200°C, PZT/PZT, and BiT/PZT were placed onto the hot plate and heated from room temperature to 360°C. Every 30°C temperature rise, the ultrasonic response in pulse-echo mode was monitored and recorded. **Fig. 1** shows 360°C ultrasonic response of PT/PZT poled at 200°C. Clear multiple echoes from bottom surface of substrate were observed with reasonable signal strength and high SNR. **Fig. 2** shows 1st thermal cycle results. Sensitivity was

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calculated from Pulser/Receiver gain to obtain 4Vp-p for 3rd reflected echoes. PT/PZT_200 PT/PZT_Room indicated PT/PZT film poled at 200°C and room temperature, respectively. As mentioned before, PZT/PZT and BiT/PZT were poled at room temperature only. PT/PZT_Room showed a lower sensitivity than BiT/PZT and PZT/PZT at any temperature and it resulted from high coercive field of PT. On contrary, PT/PZT_200 demonstrated higher sensitivity than PZT/PZT and BiT/PZT around 360°C, and comparable sensitivity with PZT/PZT even at room temperature as well. This result was agreed with expectation from high enough Curie temperature and low dielectric constant of PT and g_{33} measurement results. It means that PT/PZT could be good piezoelectric material candidate for ultrasonic transducer application, not only at 300°C but also from room temperature.

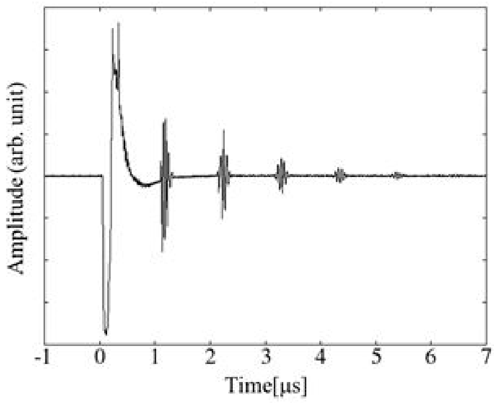

Fig.1 Ultrasonic response of PT/PZT ultrasonic transducer on ~3mm thick Ti substrate at 360°C.

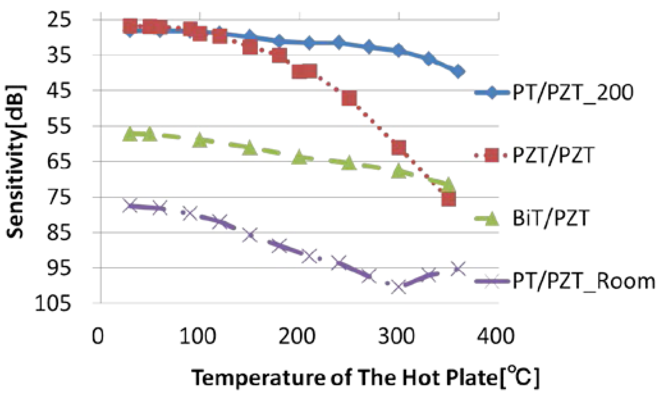

Fig.2 First thermal cycle results of ultrasonic transducers made by PT/PZT poled at 200°C and room temperature, PZT/PZT, and BiT/PZT.

3.2. Thermal cycle result of PT/PZT

Thermal cycles from room temperature to 360°C were operated three times for PT/PZT ultrasonic transducer onto titanium substrate in order to examine the transducer operation stability. The result was shown in **Fig. 3**. During three cycles, the sensitivity of around 360°C was almost same. However, sensitivity at room temperature was continuously decreased. It could caused from silver paste top electrode degradation or continuous depoling of PT/PZT around 360°C. Further research is required to determine the major reason and perform stable thermal cycle test.

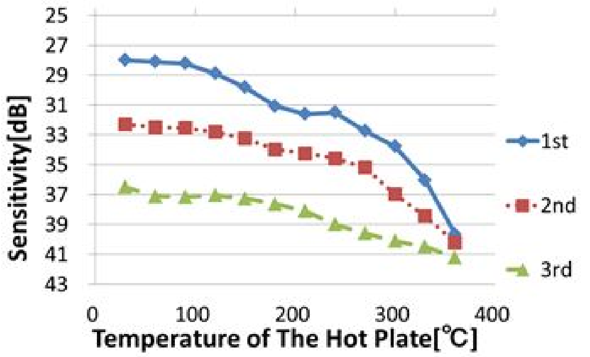

Fig.3 Three thermal cycle test results of PT/PZT ultrasonic transducer onto titanium substrate.

4. Conclusions

PT/PZT sol-gel composite films were fabricated onto titanium substrates and ultrasonic performance was compared with PZT/PZT and BiT/PZT to evaluate as high temperature ultrasonic transducer at 300°C. The transducer made by PT/PZT poled at 200°C showed higher sensitivity than by PZT/PZT and BiT/PZT at 360°C and even at room temperature, PT/PZT ultrasonic transducer had comparable sensitivity with that by PZT/PZT. This result was agreed with past study expectation and measured g_{33} values. Although further research is required for thermal cycling test, PT/PZT could be good piezoelectric material for ultrasonic transducer application below 360°C.

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