1P1-5

Fabrication of $CaBi_2Ta_2O_9/Bi_4Ti_3O_{12}$ ultrasonic transducers by automatic spray method

自動スプレー法による $CaBi_2Ta_2O_9/Bi_4Ti_3O_{12}$ 超音波トランスデューサの作製

Ayako Inano[1†],Shohei Nozawa[1], Takumi Hara[1], Kei Nakatsuma[1],Makiko Kobayashi[1]([1]Kumamoto Univ)
稲野　絢子 [1†],野澤　勝平 [1],原　拓未 [1],中妻　啓 [1],小林　牧子 [1] ,([1] 熊本大)

1. Introduction

Nondestructive inspection (NDT) is used to detect cracks and corrosion inside objects and is used in various industrial fields. Especially in plants such as thermal power plants, accidents can cause serious damage, and maintenance of equipment and structures is very important. Therefore, NDT in the operating state is required. New thermal power plants developed to reduce carbon dioxide emissions require an NDT that operates at temperatures up to 700°C. Sol-gel composite transducers do not require coplanar or backing materials, and can be applied to curved surfaces, so they are durable at high temperatures. Traditional sol-gel composite ultrasonic transducers contain $Pb(Zr,Ti)O_3$ (PZT) sol-gel phase because of high dielectric constant and poling facility.[3,5)] However, lead component in PZT sol-gel phase could evaporate at high temperatures and might cause a negative impact on the environment. Therefore, it was necessary to develop lead-free sol-gel composite. $CaBi_2Ta_2O_9$(CBTa)/$Bi_4Ti_3O_{12}$(BiT) had been developed in previous study. CBTa was selected as a ferroelectric powder phase material because it has sufficiently high Curie temperature such as 923°C. BiT was selected because it is lead-free material and has a relatively high Curie temperature of 675°C. As a result, CBTa/BiT exhibited high temperature durability up to 800°C, though.[1)] However, CBTa/BiT ultrasonic transducers had been fabricated by manual spray coating, and it made difficult to achieve reproducibility and mass production. In this study, automated spray coating system was applied for CBTa/BiT and ultrasonic performance was investigated.

2. Sample fabrication

CBTa/BiT sol-gel composite were manufactured by automatic spray coating system. First, BiT sol-gel solution was self-manufactured. CBTa/BiT sol-gel composite was prepared by mixing CBTa powders and BiT sol-gel solution by a ball mill machine for about 1 day. Next, the thoroughly mixed material was coated onto a titanium substrate by an automatic spray coating machine. The dimensions of titanium substrate were 30mm × 30mm × 3mm. Titanium was chosen as substrate material due to low thermal capacitance and high temperature durability. After spray coating, drying at 80°C and 150°C by a hot plate and an oven, and firing at 650°C by a furnace were processed for 5 min each. Those spray coating process and thermal processes were repeated until film thickness reached 50μm. Thereafter, a platinum paste top electrode with a diameter of 1cm was formed on the CBTa/BiT films by stencil printing. Thermal processes such as drying at 150°C and firing at 700°C for 2h each were operated for top electrode curing. Poling process was performed by corona discharge. After heating a sample at 950°C for 10 min, high electrical field was applied until temperature decreased to room temperature. CBTa/BiT fabricated on a titanium substrate is shown in **Fig. 1**. Crack-free film was successfully manufactured by automatic spray coating system.

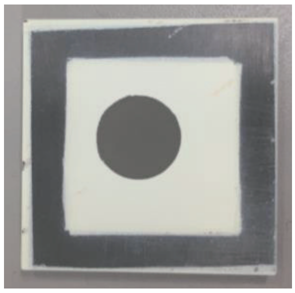

Fig. 1 Optical image of CBTa/BiT film on 3mm thick titanium substrate.

3. Experimental results

The thermal cycle test was performed between room temperature and 700°C for CBTa/BiT sol-gel composite film on titanium substrate. The sample was set into a furnace, platinum electrical cables were connected to a platinum top electrode and titanium substrate . The temperature was changed every 100°C up to 700°C. After 5min holding time at each temperature, ultrasonic waveform in pulse-echo mode was recorded by a digital oscilloscope. This thermal cycle test was conducted for 4 cycles. **Figs. 2** and **F 3** show ultrasonic responses at 700℃ in the 1st and 4th cycles, respectively. From **Fig. 3**, clear multiple echoes were still observed at 700°C. There is no waveform change between **Figs 2 and 3.** Signal to noise ratio (SNR) was improved because of sufficient top electrode curing.

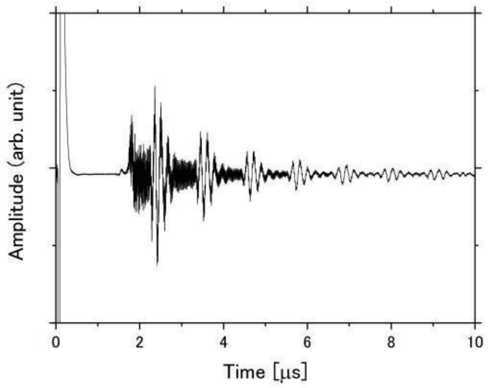

Fig. 2 Ultrasonic response at 700°C in the 1st cycle of CBTa/BiT sample.

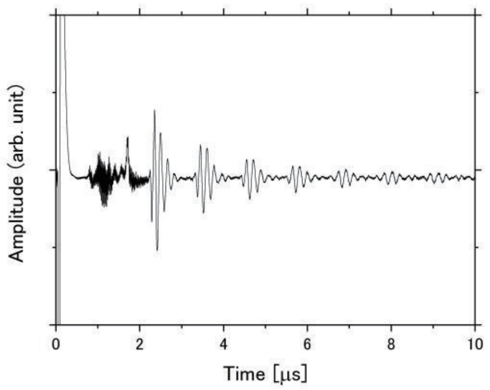

Fig. 3 Ultrasonic response at 700°C in the 3rd cycle of CBTa/BiT sample.

Then, the sensitivity was calculated for the quantitative evaluation purpose. The sensitivity was calculated as following equation;

$$\text{Sensitivity} = -\left(20\log V_1/V_2 + \text{P/R Gain}\right)\ [\text{dB}] \quad (1)$$

where V_1 is the reference amplitude (0.1Vp-p[V] in this study), and V_2 is the signal amplitude [V] of the third echo. The sensitivity of CBTa/BiT ultrasonic transducer at various temperatures is shown in **Fig. 4**. Sensitivity decreased by ~5dB after the first cycle, and it was caused by irreversible depoling of CBTa ferroelectric powder phase and BiT sol-gel phase by high temperatures. In addition, the difference is seen after 600°C in the 2nd and 3rd cycles, even though if sensitivities of 2nd and 3rd should be almost overlapped if piezoelectric material has sufficient temperature durability at 700°C. Electrical power shutdown happened during the 3rd cycle is a suspected reason. Since the 3rd and 4th cycles showed almost identical results, it is expected that CBTa/BiT made by automatic spray coating system could withstand 700°C for long term.

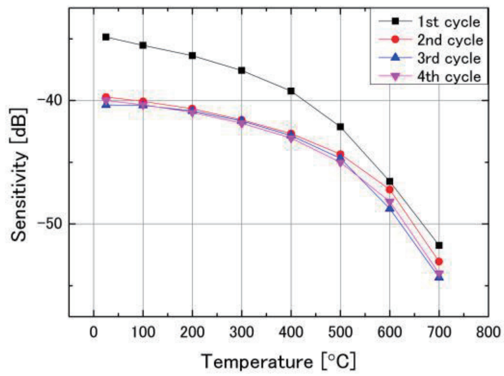

Fig. 4 Sensitivity of CBTa/BiT sample at various temperatures during thermal cycle test.

4. Conclusion

CBTa/BiT sol-gel composite was fabricated on a 3-mm thick titanium substrate by an automatic spray coating machine. High temperature thermal cycle test was carried out between room temperature and 700°C for 4 cycles. In all cycles, clear ultrasonic response and sensitivities between 3rd and 4th cycles were almost the same. Therefore, CBTa/BiT could have sufficient durability for 700°C. It is necessary to carry out long-term measurement at 700°C for final conclusion.

References

1. T. Yamamoto, S. Nozawa, M. Furukawa, H. Nagata, and M. Kobayashi: Proc. USE. (2018) 1P1-10.
2. M. Yugawa,T. Yamamoto, and M. Kobayashi: Proc. USE. (2018) 3P1-4.
3. Y. Inada, T. Inoue, M. Kobayashi, H. Nagata, and T. Takenaka: Jpn. J. Appl. Phys. 53 (2013) 07KB10.
4. T. Yamamoto, M. Kobayashi: Jpn. J. Appl. Phys. 57 (2018) 07LB16.
5. M. Kobayashi and C.-K. Jen: Smart Mater. Struct. 13 (2004) 951.