High performance ultrasonic transducers made by LiNbO₃/Bi₄Ti₃O₁₂

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Abstract— LiNbO3 (LN)/ Bi4Ti3O12 (BiT) ultrasonic transducers by sol-gel spray technique on 3mm thick titanium substrates were fabricated and conducted the maximum temperature test in order to carry out sensitivity comparison between LN/BiT and BiT/BiT. As a result, sensitivity of LN/BiT was higher than that of BiT/BiT for all temperatures in addition to higher temperature durability. Furthermore, in order to investigate long-term thermal durability, LN/BiT ultrasonic transducers by automatic spray coating system were fabricated on 3mm thick Inconel substrates and conducted thermal cycle test and long-term test at 700°C. Throughout the thermal cycle test, sensitivity was very stable. Long-term test of LN/BiT showed sufficient thermal durability under 700°C for 72h. Therefore, LN/BiT ultrasonic transducers showed the potential for non-destructive inspection of new thermal power stations at 700 °C

Keywords— nondestructive testing; high performance; ultrasonic transducer; thermal durability; high temperature

I. INTRODUCTION

Non-destructive testing (NDT) widely used for industrial fields to detect early stage small defects before fatal failure. Various NDT methods have been developed and ultrasound NDT is one of the common methods because it is possible to measure thinning of the structure and to detect defects/cracks inside the structure, which is difficult by external image diagnosis. Sol-gel composite ultrasonic transducers have been developed for on-line NDT monitoring and there are several advantages [1-5]. First, backing material is unnecessary because there are small pores inside the sol-gel composite film and it reduces ringing effect. Second, since the sol-gel composite film has small pores as described above, it has high thermal shock durability. Third, since the oxide layer is formed by the heating process between the sol-gel composite and the substrate, the coupling agent is unnecessary since it has good acoustic coupling with the substrate. Nondestructive inspection during high-temperature operation has been desired to assure safety and reduce economical loss during shut-down. In addition, new thermal power plants require operating temperatures up to 700°C, so high temperature durable ultrasonic transducers are required to monitor new system during test operation. Sol-gel composite

ultrasonic transducers are suitable for high temperature ultrasonic transducer applications and LiNbO₃(LN) based solgel composite should have high performance at high temperatures because LN has a high Curie temperature around 1200 °C.

In the past studies, high temperature properties of sol-gel composites, $Bi_4Ti_3O_{12}(BiT)$ piezoelectric powder and several types of lead-free sol-gel solution, were investigated and BiT sol-gel solution demonstrated high potential for lead-free high temperature ultrasonic transducer application, though BiT/BiT showed severe deterioration above 600°C. There are several reports that LN/BiT showed ultrasonic performance up to 1000 °C [6-7], though there was a doubt of insufficient poling degree.

In this study, maximum temperature test was conducted for LN/BiT ultrasonic transducers by manual spray coating on a titanium substrate in order to sensitivity comparison between LN/BiT and BiT/BiT In addition, in order to improve the sensibility and reproducibility, LN/BiT ultrasonic transducers by automatic spray coating on Inconel substrate were fabricated and conducted thermal cycle test and long-term test for 72 hours at 700°C.

II. SAMPLE FABRICATION

LN/BiT samples were manufactured by sol-gel spray technique [8-11]. LN/BiT sol-gel composite was prepared by mixing LN piezoelectric powders and BiT sol-gel solution by a ball mill machine for about 1 day. The thoroughly mixed solution was coated onto a 3 mm thick titanium substrate by spray method. The spray-coated sample was dried at 150 ° C and calcined at 650 °C for 5 minutes each. These processes were repeated to produce a LN/BiT sample having a film thickness of 50 μ m. Accordingly, an electrode having a diameter of 1 cm was formed on the sample by using platinum paste, and the electrode was subjected to poling treatment. Poling was performed by applying a corona discharge to a sample from 900°C to room temperature conditions. The optical image of LN / BiT on titanium substrate sample is shown in Fig. 1.

However, long-term testing could not be performed with titanium substrate because it was deformed by high temperatures. Therefore, LN/BiT was coated on 3mm thick Inconel substrate by automatic spray method. Drying and Program Digest 2019 IEEE IUS Glasgow, Scotland, October 6-9, 2019

calcining were conducted in the same way as LN/BiT coated on a titanium substrate and produced a LN/BiT sample with thickness of 50 μ m. Identically, an electrode having a diameter of 1 cm was formed on the sample by using platinum paste, and the electrode was subjected to poling treatment. Poling was performed by applying a corona discharge to a sample from 900 ° C to room temperature conditions. The optical image of LN/BiT on Inconel substrate sample is shown in Fig. 2.



Fig. 1. LN/BiT 50µm thick on titanium substrate by manual spray coating.



Fig. 2. LN/BiT 50µm thick on Inconel substrate by automatic spray coating.

III. ULTRASONIC MEASUREMENT AT HIGH TEMPERATURE

Ultrasonic measurements in pulse echo mode were recorded from room temperature till 900 °C. Fig. 3 shows sensitivity comparison results between LN/BiT and BiT/BiT. The sensitivity was calculated as following;

Sensitivity = - $(20\log_{10}V_1/V_2 + \text{gain of P/R})$ [dB] (1)

where V_1 is the ideal amplitude, that was 0.1 [V_{p-p}] in this experiment, V_2 is the amplitude [V_{p-p}] of the second reflected echo from the bottom surface of the substrate.



Fig. 3. Sensitivity comparison between LN/BiT and BiT/BiT

Sensitivity of LN/BiT was higher than BiT/BiT for all temperatures in addition to higher temperature durability. Signal to noise ratio of LN/BiT was sufficiently high enough for thickness measurement even at 900 °C.

In order to improve the sensibility and reproducibility, LN/BiT ultrasonic transducers by automatic spray coating on Inconel substrates were fabricated and conducted thermal cycle test between room temperature and 700 °C. Long-term test for 72 hours at 700 °C was also executed.

First, three thermal cycles of LN/BiT were performed. The ultrasonic response was measured at each temperature while increasing the temperature from room temperature by 100°C. After 5min holding time at each temperature, ultrasonic response was recorded by a digital oscilloscope up to 700°C. The ultrasonic response of LN/BiT at RT in the first cycle is shown in Fig. 4. The ultrasonic response of LN/BiT at 700°C in the third cycle is shown in Fig. 5. Thermal cycle test results of LN/BiT are shown in Fig. 6. From Fig. 3 and Fig. 6, LN/BiT sample fabricated on the Inconel substrate shows higher sensitivity than the one fabricated on the titanium substrate by ~8 dB at 700 °C. Thermal stability and SNR of LN/BiT on the Inconel substrate by automatic spray coating was also superior to those of LN/BiT on the titanium substrate by manual spray coating.



Fig. 4. Ultrasonic response of LN/BiT on 3mm thick Inconel substrate at RT in the first cycle



Fig. 5. Ultrasonic response of LN/BiT on 3mm thick Inconel substrate at 700°C in the third cycle



Fig. 6. Temperature dependence of sensitivity of LN/BiT on 3mm thick Inconel substrate.

Next, a long-term test of LN/BiT was performed at 700 °C for 72 hours. The measurement was performed by connecting the electrode and the ground in reverse. The ultrasonic response that heating starts at 700°C is shown in Fig. 7. Ultrasonic response continued heating for 72 hours is shown in Fig. 8. Time dependence of sensitivity of LN/BiT at 700°C for 72 hours is shown in Fig. 9. Those results also supported excellent stability of LN/PZT at 700°C.



Fig. 7. Ultrasonic response of LN/BiT at 700°C after 0h.



Fig. 8. Ultrasonic response of LN/BiT at 700°C after 72h



Fig. 9. Time dependence of sensitivity of LN/BiT at 700 °C for 72 hours

IV. CONCLUSIONS

In this research, high temperature performance of LN/BiT composite was investigated for ultrasonic transducer applications with high operation temperature at 700°C. LN/BiT sol-gel composites were fabricated on 3mm thick titanium and Inconel substrate by spray coating method. conducted the maximum temperature test in order to sensitivity comparison between LN/BiT and BiT/BiT. Sensitivity of LN/BiT was higher than BiT/BiT for all temperatures in addition to higher temperature durability. Throughout the thermal cycle test, sensitivity was very stable and SNR of LN/BiT on Inconel substrate by automatic spray coating was equivalent or superior to SNR of LN/BiT on a titanium substrate by manual spray coating. In addition, longterm test was conducted, and no significant difference was confirmed after 72hours heating. Therefore, LN/BiT ultrasonic transducers showed the potential for nondestructive inspection of new thermal power stations at 700 °C.

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