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Integrated Ultrasonic Transducers above 500°C

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***Abstract*— New piezoelectric composite materials, manganese doped sodium bismuth titanate (NBT+Mn)/lead zirconate titanate (PZT) and lithium niobate (LN)/PZT have been developed for ultrasonic transducer trials at operation temperature more than 500°C. Three kinds of piezoelectric powders, bismuth titanate (BIT), NBT+Mn, and LN, were dispersed into PZT sol-gel, and ~60μm thick BIT/PZT, NBT+Mn/PZT, and LN/PZT films were deposited onto 26mm thick titanium (Ti) substrates by sol-gel spray tecnhique, respectively. Reflection echoes from Ti substrates were observed for each sample at various temperatures, from room temperature to above 500°C, and the signal amplitude change was monitored. LN/PZT films demonstrated the highest temperature stability among three materials. Using LN/PZT, broadband ultrasonic signals centered at 5.0MHz with 25dB signal to noise ratio were recorded at 700°C without depoling.**

Keywords-high temperature; ultrasonic transducer; lithium niobate; sodium bismuth titanate; bismuth titanate; sol-gel composite

I. INTRODUCTION

Ultrasonic methods are one of the suitable candidates for many applications at elevated temperatures, such as combustion control for diesel engines [1], condition monitoring of airborne gas turbine engines [2], process monitoring of metal manufacturing [3], etc, because they offer non-invasive and non-destructive real-time monitoring. For such monitoring piezoelectric ultrasonic transducers (UTs) are often used due to their simplicity and cost-effectiveness [4].

However, it is challenging to develop such UTs which can endure high temperature (HT). Firstly, the operation temperature range of piezoelectric UT is limited by its Curie temperature. Normally it is operable up to one half of the Curie temperature. The piezoelectricity reduces above such temperature due to depoling [5]. There are high Curie temperature piezoelectric single crystals such as $LiNbO_3$ (LN) and the Curie temperature of LN is 1210°C. However LN single crystals have weak thermal shock resistance. It is not easy to use them at high temperatures together with fast temperature changes. In addition, it is difficult to find long lasting ultrasonic couplant and backing materials which provide broad frequency bandwidth in this temperature range.

Sol-gel spray technique has been studied to fabricate HTUTs [6]. HTUTs were integrated onto the flat and curved surfaces of many substrates under study directly without ultrasonic couplant. Broadband frequency characteristic was obtained without backing materials due to the porosity existed in the piezocomposite film. The porosity also enhanced thermal shock resistance of the HTUT. Several sol-gel composite materials were developed such as lead zirconate titanate (PZT)/PZT [7], lithium tantalate/PZT [8], bismuth titanate (BIT)/PZT [9], etc. BIT/PZT showed ultrasonic performance up to 500°C, although the operation above 500°C has not been examined yet. Furthermore it is desired to obtain a HTUT which has higher piezoelectricity than BIT/PZT at HT.

In this investigation, two new sol-gel composite materials, manganese doped sodium bismuth titanate $Na_{0.5}Bi_{4.5}Ti_4O_{15}$ + $MnCO_3$ 0.1wt% (NBT+Mn)/PZT and LN/PZT will be used for HTUT trials. NBT+Mn is bismuth layer structure and chosen as one of piezoelectric powder phase because it had high Curie temperature of 655°C [10]. It was confirmed empirically that it had lower coercive field and higher resistance than BIT and it was expected that it could be electrically poled easier and show more stable performance at HT than BIT. LN was chosen as another piezoelectric powder phase by the Curie temperature of 1210°C, even though it seems that piezoelectric ceramic sample of LN had not yet been reported [11]. PZT was chosen as sol-gel phase due to its high dielectric constant. BIT/PZT is also investigated here as a reference material. Thick films of BIT/PZT, NBT+Mn/PZT, and LN/PZT were integrated onto titanium (Ti) rod by sol-gel spray technique, and ultrasonic performance above 500°C will be studied.

II. FABRICATION

The fabrication process is based on a sol-gel spray technique developed previously [6, 9]. First, each piezoelectric powders, BIT, NBT+Mn, and LN, were dispersed into PZT sol-gel and ball milled respectively, until appropriate viscosity for spray coating was obtained. Then each sol-gel composite was sprayed directly onto a 26mm thick titanium (Ti) rod with 25mm diameter. Ti was selected as the substrate material due to high temperature resistibility and low acoustic attenuation. After each spray coating, thermal treatments such as drying, firing and annealing were carried out. The film thickness reached around 60μm after five-layers coating. The films were electrically poled using corona discharge. Finally top electrodes were fabricated onto piezoelectric ceramic composite using silver paste. After that Ti foils were used to cover the silver paste top electrode as shown in Fig.1 to prevent oxidation at

HT. Samples with BIT/PZT, NBT+Mn/PZT, and LN/PZT films integrated onto Ti rods are shown in Figs. 1 (a), (b) and (c), respectively.

(a) (b) (c)

Figure 1. A ~60 μm thick (a) BIT/PZT (b) NBT+Mn/PZT and (c) LN/PZT film fabricated onto a 26 mm thick Ti rod by sol-gel spray technique.

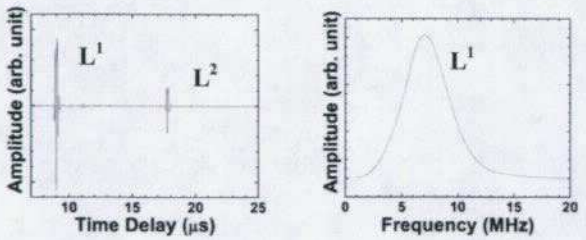
III. CHARACTERIZATION

The capacitances and dissipation factor D of every film were measured by a Hewlett Packard 4192A LF Impedance Analyzer at 1 kHz in order to calculate the relative dielectric constant ε_r. The diameters of the top electrode were different for each piezoelectric ceramic material due to impedance matching requirement.

Ti substrates were heated up by a hot plate up to 500°C and then gas torch was used to reach high temperature. Then ultrasonic signals were measured in pulse-echo mode and recorded every 50°C in most cases. The recording was continued until the ultrasonic signal strength was significantly deteriorated or it reached the maximum temperature (around 750°C) of the experimental setup.

A. BIT/PZT

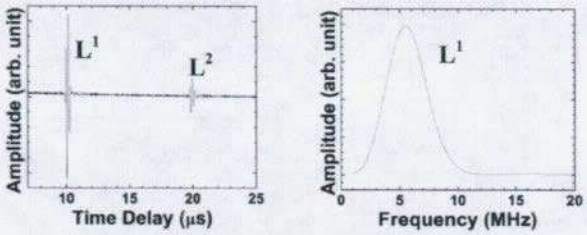
For a 60μm thick PIT/PZT film HTUT deposited on top of a Ti rod as shown in Fig. 1(a) the diameter of the top electrode was 11mm. The measured dissipation factor D and calculated relative dielectric constant ε_r were 0.03 and 60, respectively. Figure 2 shows its ultrasonic performance in (a) time and (b) frequency domain (for L^1 signal) at room temperature. L^1 and L^2 are the 1st and 2nd round-trip echoes in the Ti rod, respectively. In Fig. 2(a), the ultrasonic signal shows a signal-to-noise ratio (SNR) of 32dB. This UT showed the center frequency and 6dB bandwidth of 7.2MHz and 4.4 MHz, respectively at room temperature.



(a) (b)

Figure 2. Ultrasonic performance of a BIT/PZT film HTUT made by sol-gel spray technique onto a 26.2mm thick Ti rod in (a) time and (b) frequency domain at room temperature.

In order to investigate the ultrasonic performance and depoling effect of the film, the sample was thermal cycled twice; heated up to 600°C and cooled down to room temperature and again. During thermal cycling the ultrasonic signals were recorded every 50°C. Figure 3 shows the ultrasonic performance of the BIT/PZT film HTUT as shown in Fig. 1(a) in (a) time and (b) frequency domain (L^1 signal) at 550°C during the 2nd cooling cycle. In Fig. 3(a), the ultrasonic signal shows a SNR of about 30dB. The center frequency and 6dB bandwidth at 550°C were 5.6MHz and 4.0MHz, respectively. The frequency band was shifted to 5.6MHz which is lower than 7.2MHz of room temperature due to the increased ultrasonic attenuation in Ti rod at such temperature.



(a) (b)

Figure 3. Ultrasonic performance of a BIT/PZT film HTUT made by sol-gel spray technique onto a 26.2mm thick Ti rod in (a) time and (b) frequency domain at 550°C.

Figure 4 shows the ultrasonic signal amplitude change during the heating of the 2nd thermal cycle. The signal amplitude was deteriorated rapidly above 450°C, even though below 400°C, the amplitude decreased almost linearly. After the cooling of each thermal cycle the signal strength did not recover to the level before the heating. For the signal amplitude reduction during the 1st cycle and 2nd cycle were 18dB and 16dB, respectively. Such results indicated that BIT/PZT film was continuously depoled above 450°C and it is not suitable for long term use above 500°C. It is noted that a thermal cycle test of a BIT/PZT film deposited onto a 12.7mm thick steel block was also carried out between room temperature and 400°C. After 375 thermal cycles inside a furnace, the ultrasonic performance of this film after each cooling at room temperature showed little change (< 1dB total).

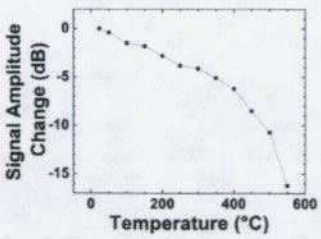
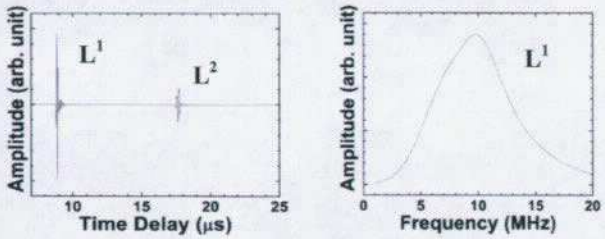


Figure 4. Signal amplitude change versus temperature variation of a BIT/PZT film on top of a Ti rod.

B. NBT+Mn/PZT

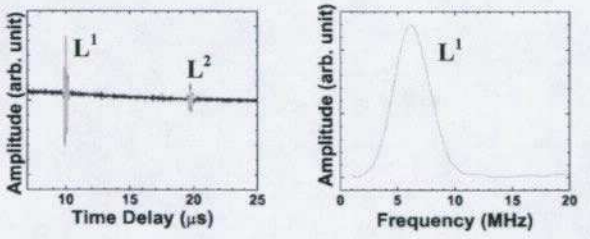
For a 60μm thick NBT+Mn/PZT film HTUT deposited on top of a Ti rod as shown in Fig. 1(b) the diameter of the top electrode was 10mm. The measured dissipation factor D and calculated relative dielectric constant ε_r were 0.02 and 70, respectively and they are similar to those of BIT/PZT film. Figure 5 shows its ultrasonic performance in (a) time and (b) frequency domain (for L^1 signal) at room temperature. In Fig. 5(a), the ultrasonic signal shows a SNR of about 47dB. This result was better than that of the BIT/PZT film. It may be due to higher signal strength of 7dB at room temperature than that of BIT/PZT film. The center frequency and 6dB bandwidth were 9.4 MHz and 6.9 MHz, respectively.



(a) (b)

Figure 5. Ultrasonic performance of a NBT+Mn/PZT film HTUT made by sol-gel spray technique onto a 26.2mm thick Ti rod in (a) time and (b) frequency domain at room temperature.

However, the piezoelectricity of the NBT+Mn/PZT film was more temperature dependent than that of the BIT/PZT film. The ultrasonic signals were monitored up to 525°C during the heating cycle because the signal strength and SNR was dramatically deteriorated. The signal strength was not recovered after cooling to room temperature. The 2nd thermal cycle was not carried out because the signal strength was too weak. Figure 6 shows the ultrasonic performance of the same NBT+Mn/PZT film HTUT as shown in Fig. 1(b) in (a) time and (b) frequency domain (for L^1 signal) at 525°C during the 1st heating cycle. In Fig. 6(a), the ultrasonic signal shows a SNR of about 21dB. The center frequency and 6dB bandwidth at 525°C during the heating process were 6.3MHz and 3.7 MHz, respectively. The center frequency was shifted to 6.3MHz which is lower than 9.4MHz of room temperature because of the increased attenuation in the Ti rod at such temperature.



(a) (b)

Figure 6. Ultrasonic performance of a NBT+Mn/PZT film HTUT made by sol-gel spray technique onto a 26.2mm thick Ti rod in (a) time and (b) frequency domain at 525°C.

Figure 7 shows the ultrasonic signal amplitude change during the first time heating process up to 525°C. It is noted that the signal amplitude was deteriorated rapidly above 450°C, even though below 400°C, the amplitude decreased almost linearly. The signal strength did not recover after the cooling. This tendency was very similar with BIT/PZT. Thus it is expected that NBT+Mn/PZT film was also continuously depoled above 450°C and it is not suitable for long term use above 500°C. The reason of high temperature dependency, which may be caused by low coercive field and incomplete sintering such as oxide vacancy, is under investigation.

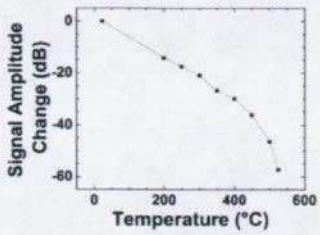
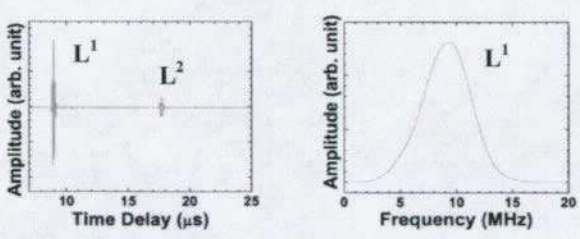


Figure 7. Signal amplitude change versus temperature variation of a NBT+Mn/PZT film on top of a Ti rod.

C. LN/PZT

For a LN/PZT film HTUT deposited on top of a Ti rod as shown in Fig. 1(c) the diameter of the top electrode was 13mm. The measured dissipation factor D and calculated relative dielectric constant ε_r were 1.8 and 40, respectively. However, the real value of relative dielectric constant should be lower because of a high D value. It may be caused by dielectric constant difference between powder phase and sol-gel phase. Figure 8 shows its ultrasonic performance in (a) time and (b) frequency domain (for L^1 signal) at room temperature. In Fig. 8(a), the ultrasonic signal shows a SNR of about 30dB. The signal strength was lower by 20dB than that of BIT/PZT at room temperature. It is expected because the piezoelectric constant d_{33} of single crystal LN is lower than that of BIT and NBT+Mn, although the piezoelectricity of LN ceramics had not been confirmed. The center frequency and 6dB bandwidth were 9.2MHz and 4.7MHz, respectively.



(a) (b)

Figure 8. Ultrasonic performance of a LN/PZT film HTUT made by sol-gel spray technique onto a 26.2mm thick Ti rod in (a) time and (b) frequency domain at room temperature.

In order to investigate the ultrasonic performance and depoling effect of the film, the sample was thermal cycled twice; heated up to 750°C and cooled down to room temperature and again. 750°C was the maximum temperature allowed of the experimental setup. During thermal cycling the ultrasonic signals were recorded every 50°C. Figure 9 shows the ultrasonic performance of the LN/PZT film HTUT as shown in Fig. 1(c) in (a) time and (b) frequency domain (L^1 signal) at 700°C during the 2nd cooling cycle. In Fig. 9(a), the ultrasonic signal shows a SNR of about 25dB. The center frequency and 6dB bandwidth at 700°C were 5.0MHz and 3.4 MHz, respectively. The center frequency was shifted to 5.0MHz which is lower than those of BIT/PZT and NBT+Mn/PZT samples, because ultrasonic attenuation in Ti rod is higher at 700°C than those at 550°C and 525°C for BIT/PZT and NBT+Mn/PZT films, respectively.

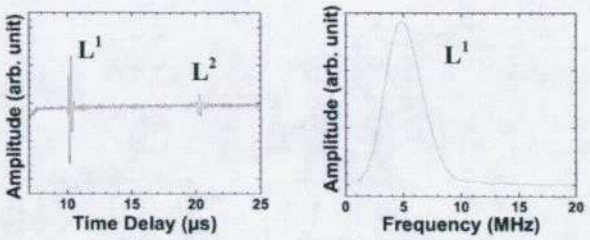


Figure 9. Ultrasonic performance of a LN/PZT film HTUT made by sol-gel spray technique onto a 26.2mm thick Ti rod in (a) time and (b) frequency domain at 700°C.

Figure 10 shows the ultrasonic signal amplitude change during the 2nd thermal cycle. It is noted that the signal amplitude decreased almost linearly through the temperature range from room temperature to 700°C. The signal strength was completely recovered after each cooling cycle. From this result, it is expected that LN/PZT can be suitable for long term use at least up to 700°C. The signal reduction rate would be improved if the thickness of LN/PZT film is higher and the center frequency at room temperature becomes lower, because there will be less attenuation in Ti rod at a lower frequency. Further investigation is needed to determine the maximum operation temperature of LN/PZT film.

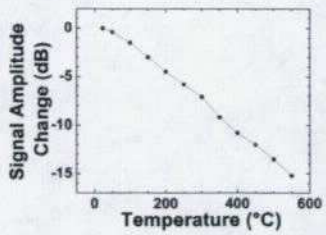


Figure 10. Signal amplitude change versus temperature variation of a LN/PZT film on top of a Ti rod.

IV. Conclusions

Three kinds of ~60μm thick films, BIT/PZT, NBT+Mn/PZT, and LN/PZT were directly integrated onto Ti rod by sol-gel spray technique in order to investigate HTUT application possibility above 500°C. Reflection echoes from the Ti rods were observed from each sample at various temperatures, from room temperature to above 500°C, and the signal amplitude change was monitored every 50°C. Both BIT/PZT and NBT+Mn/PZT films showed severe depoling effect when the temperature is higher than 450°C. It was confirmed that they were not suitable for use above 450°C continuously. It was also observed that the temperature dependency of the NBT+Mn/PZT film is higher than that of the BIT/PZT film. LN/PZT film demonstrated excellent temperature stability. Broadband ultrasonic signal with a 25dB SNR was obtained at 700°C. Negligible depoling effect was observed after two thermal cycles between room temperature and 750°C.

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