Material development of Pb(Zr,Ti)O₃/Pb(Zr,Ti)O₃ for biomedical ultrasound monitoring

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1. Introduction

Phased array technology can enhance image quality and provide real-time adjustments to optimize imaging for specific anatomical targets. In previous study, An array transducer made by Pb(Zr,Ti)O₃(PZT)/PZT sol-gel composite material manufactured without dicing and was it demonstrated imaging performance with relatively high SNR.¹⁾ Phased-array technology involves the use of multiple transducer elements arranged in a specific pattern. Thus, each transducer element size becomes smaller than A-mode ultrasonic transducer for industrial application and higher dielectric constant is desired for electrical impedance matching.

Sol-gel composites are composed of two materials, sol-gel solution and piezoelectric powder, and each phase affects the overall performance of the composite. From the past studies, the use of PZT powders with high dielectric constant showed lower piezoelectric constant d_{33} than that of medium dielectric constant.²⁻⁴ It was suspected that the electrical field was not applied efficiently to powder phase with high dielectric constant during poling process at room temperature.

In this study, PZT/PZT samples with high dielectric constant powders were poled at various temperatures, as well as the corresponding size of the top electrode area and piezoelectric constant and performance of the transducer were investigated.

2. Sample fabrication

Two PZT powders with different dielectric constants were used as raw materials as shown in **Table I**. It is noted that there is no significant difference in other properties such as particle size, electromechanical coupling coefficient k between the two materials except for dielectric constants.

Table I:	Property	of PZT	powders
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	Property				
Powder	\mathcal{E}_r	k ₃₃ [%]	d ₃₃ [pm/V]		
HIZIRCO A	5500	70	660		
HIZIRCO L	1800	70	400		

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The two PZT powders were mixed with PZT solution, gel sol solution and then grinding balls added and mixed completely were and homogeneously and then the mixture was sprayed onto titanium alloy substrates of 3 mm thickness and 5 mm edge length. Heat treatment is then carried out and the substrate is subjected to drying and annealing processes at 150 °C and 650 °C. Spray coating and thermal treatments were repeated until the thickness of the piezoelectric film reached 50 µm. Poling process at various temperatures was carried out after the films were fabricated to realize the piezoelectricity of the films. Optical images of the ultrasonic transducers made of the two materials are shown in Fig. 1. No significant differences were observed except for the difference in film color due to the piezoelectric powders. The thickness of both samples were about 50 µm.



Fig. 1 Optical images of the samples.

3. Results and discussions

The piezoelectric constant measurements results are shown in **Table** II. A/PZT poled at 350°C showed the highest d_{33} whereas L/PZT has no significance difference. At optimized conditions, A/PZT showed the higher values.

Table II Measured d₃₃ per piezoelectric film

Sample	<i>d</i> ₃₃ (pC/N)
A/PZT RT	32.5
A/PZT 100°C	26.8
A/PZT 200°C	46.8
A/PZT 300°C	50.1
A/PZT 350°C	58.4
A/PZT 350°C (2)	55.2
A/PZT 400°C	51.7
L/PZT RT	41.7
L/PZT 200°C	42.0

Figures 2 and 3 show the ultrasonic response with 6 mm top electrode poled at RT for A/PZT and L/PZT, respectively. The reflected echoes from the bottom of the substrate are clearly visible, indicating that both samples have broadband characteristics and high signal-to-noise ratios. Figure 4 shows the sensitivity dependency on top electrode. Sensitivity was calculated by true gain to achieve 200 mV_{p·p} for the 1^{st} reflected -1 was multiplied echo then for understanding facility. It is noted that PZT/PZT with 2 and 4mm diameter, the sensitivity showed the highest values for both materials, even though L/PZT waveform were deformed.



Fig. 2 Ultrasonic responses of A/PZT.



Fig. 3 Ultrasonic responses of L/PZT.



Fig. 4 Top electrode dependence of signal strength

Table III shows the relative dielectric constant of the four sizes of upper electrodes fabricated on each piezoelectric film. The relative dielectric constant of A/PZT is higher than that of L/PZT according to the Table I. The sol-gel composite shows much lower dielectric constant values than raw materials due to porosity existence. It is noted that the dielectric constant increases by poling temperature increases. 2 mm top electrode has many measurement errors due to the Au film thinness by shadow effect. Top electrode size optimization should conduct again after top electrode fabrication process was improved.

Tal	ble	Ш	Re	lative	diel	lectric	constant
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Sample	Relative dielectric constant				
2 milpro	8mm	6mm	4mm	2mm	
A/PZT RT	944	899	899	412	
A/PZT 100°C	1022	1039	989	763	
A/PZT 200°C	1000	979	989	N/A	
A/PZT 300°C	989	1039	989	639	
A/PZT 350°C	1225	1238	1123	824	
A/PZT 350°C (2)	1225	1178	1123	813	
A/PZT 400°C	1371	1438	1438	N/A	
L/PZT RT	584	593	494	N/A	
L/PZT 200°C	584	639	584	N/A	

4. Conclusions

Two types of PZT powders with different dielectric constant values, A and L, were used to fabricate PZT/PZT sol-gel composite samples, A/PZT and L/PZT for powder phase dielectric constant effect evaluation. A/PZT sample poled at 350 °Cshowed highest d_{33} A/PZT poled at 350 °C could be suitable for phased array transducer application for medical imaging because of its high dielectric constant.

References

- M. Tanabe, K. Sato, T. Uda and M. Makiko, Jpn. J. Appl. Phys. 62, SJ1034 (2023).
- Y. Kiyota, H. Makino, K. Nakatsuma and M. Kobayashi, 39th Symp. Ultrasonic Electronics, 2018, 2P1-11.
- Y. Kiyota, K. Nakatsuma, and M. Kobayashi, 38th Symp. Ultrasonic Electronics, 2017, 3P1-6.
- M. Kobayashi, T. Inoue and M. Sawada, Proc. 33rd Symp. Ultrasonic Electronics, 2012, p.209.