Effect of Piezoelectric Powder Phase Permittivity on Pb(Zr, Ti)O₃/Pb(Zr, Ti)O₃ Thin Films

圧電粉体相の誘電率が PZT/PZT 薄膜に与える影響 Yuto Kiyota^{1‡}, Kei Nakatsuma¹, Makiko Kobayashi¹ (¹Kumamoto Univ.) 清田 湧斗^{1‡}, 中妻 啓¹, 小林 牧子¹(¹熊本大)

1. Introduction

Piezoelectric sol-gel composite films has been developed mainly for non-destructive testing (NDT) ultrasonic transducer applications. NDT by ultrasonic transducer is one of the major NDT methods and has advantage such as economical sub-surface inspection capability. Sol-gel composite have several advantages; the backing material is not necessary, it could be used for curved surface NDT, etc.

In previous study, it was observed that performance of sol-gel composite could be improved by using high dielectric constant sol-gel phase material.¹⁻²⁾ On the other hand, about piezoelectric powder phase, pulse-echo signal strength of higher dielectric constant powder phase was lower than that of lower dielectric constant powder phase.³⁾ However, only two kinds of Pb(Zr,Ti)O₃ (PZT) powders were compared and other parameters, such as electromechanical coupling coefficient, were also different so that it might affect the performance. In addition, since the samples were manufactured by hand spray, individual difference caused by manual spray coating was a factor of the result as well. Therefore, it is necessary to investigate the effect of piezoelectric powder phase permittivity in more detail.

In this paper, PZT/PZT sol-gel composite films were fabricated by automatic spray coating method and try eliminating the individual difference effect and to investigate powder phase effect more accurately. Three types of PZT powders, HIZIRCO A, HIZIRCO L, and HIZIRCO MPT, different relative permittivity and same electromechanical coupling coefficient, were used for more accurate comparison. **Table I** shows property of each powder.

Table I: Property of PZT powder

Powder	Property			
	$\boldsymbol{\varepsilon}_r$	k ₃₃ [%]	d ₃₃ [pm/V]	Q _m
HIZIRCO A	5500	70	660	55
HIZIRCO L	1800	70	400	75
HIZIRCO MPT	1300	70	290	2000

2. Fabrication process of PZT/PZT films

PZT/PZT films were fabricated onto titanium substrates by sol-gel spray method. First, PZT powder and sol-gel solution were mixed for a day. After that, PZT/PZT films were formed onto titanium substrates by spray coating. Thickness of the titanium substrate was 3mm and the area of the titanium substrate was 3cm × 3cm square. An Automatic spray coating machine was used for spray coating. Thermal process, drying at room temperature, drying at 150°C by a hot plate, firing at 650°C by an electrical furnace for 5min each, was performed. Spray coating and thermal process were repeated 4-5 times. After reaching target thickness, Corona poling was performed at room temperature. Aluminum top electrodes were fabricated by vacuum deposition method. The area of top electrode is 1cm diameter.

3. Experimental Results

Figs. 1(a)-(c) shows optical images of PZT/PZT sol-gel composite films onto titanium substrate using HIZIRCO A, HIZIRCO L, and HIZIRCO MPT, respectively. The film color difference was mainly caused by powder color difference. Thicknesses of each PZT/PZT films were 39.5, 38.6, and 38.5 µm, respectively, measured by an Eddy current thickness meter. The thickness became more constant than that by manual spray. Capacitance of PZT/PZT film used HIZIRCO A, HIZIRCO L, HIZIRCO MPT was 19.12nF. 8.48nF, and 9.65 nF, respectively. Permittivity of PZT/PZT films was calculated by following equation;

$$\varepsilon = \frac{Cd}{A} \tag{1}$$

where *C* is capacitance, *d* is thickness and *A* is area of top electrode. Relative dielectric constant of PZT/PZT film used HIZIRCO L, HIZIRCO A, HIZIRCO MPT was 1084, 474, and 531. Piezoelectric constant d_{33} of PZT/PZT film using HIZIRCO A, HIZIRCO L, HIZIRCO MPT was 45.1pC/N, 51.5 pC/N, and 23.0 pC/N, respectively. d_{33} of PZT/PZT using HIZIRCO A was lower than that using HIZRCO L and it would be caused by insufficient poling of powder phase.



Fig.1 Optical images of PZT/PZT film using (a) HIZIRCO A, (b)HIZIRCO L, (c)HIZIRCO MPT

Next, PZT/PZT ultrasonic response was measured in pulse-echo mode. Figs.2-4 shows pulse-echo measurement result of HIZIRCO A, L, and MPT PZT/PZT film, respectively. Clear multiple reflected echoes were confirmed in all the results. Since top electrode size and pulser/receiver setting was exactly same for all the samples, electrical impedance mismatch of PZT/PZT using HIZIRCO A was larger than other samples. As a result, dead zone length of Fig. 2 is longer than others. Signal amplitude of PZT/PZT using HIZIRCO MPT was much lower than others. There were clacks on the surface only for the PZT/PZT using HIZIRCO MPT by microscope observation. When the spray system was not totally adjusted, film quality was significantly deteriorated.



Fig.2 Ultrasonic response of HIZIRCO A PZT/PZT film



Fig.3 Ultrasonic response of HIZIRCO L PZT/PZTfilm



Fig.4 Ultrasonic response of HIZIRCO MPT PZT/PZT film

4. Conclusions

In this paper, PZT/PZT sol-gel composite films were fabricated by automatic spray coating using three types of PZT powder with different dielectric constant in order to investigate influence of powder phase permittivity more accurately. Film thickness became more constant than manual spray. d_{33} and amplitude of ultrasonic response on HIZILCO L PZT/PZT film was highest, however, HIZIRCO A PZT/PZT had electrical impedance mismatch and HIZIRCO MPT PZT/PZT had poor film quality problem. Further investigation is required to make conclusions.

References

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