2P1-11

Piezoelectric Powder Permittivity Effect of $Pb(Zr, Ti)O_3/Pb(Zr, Ti)O_3$

PZT/PZT における圧電粉体誘電率の影響

Yuto Kiyota[1‡], Hiroto Makino[1] , Kei Nakatsuma[1] , Makiko Kobayashi[1]
([1]Kumamoto Univ.)
清田　湧斗 [1‡]，牧野　博翔 [1] 中妻　啓 [1] ，小林　牧子 [1] ([1] 熊本大学)

1. Introduction

These days, piezoelectric materials were used for various devices. Piezoelectric sol-gel composite materials were developed to fabricate thick film and used as ultrasonic transducer for non-destructive testing (NDT). Piezoelectric sol-gel composite films are fabricated from sol-gel solution and piezoelectric powder.

In previous study, about sol-gel phase, it was confirmed that high dielectric constant sol-gel phase material could improve performance of sol-gel composite.[1-2)] About piezoelectric powder phase, pulse-echo signal strength of lower dielectric constant powder phase was higher than that of higher dielectric constant powder phase.[3] But, dielectric constant of powders were quite differently and only 2 kind of PZT powder were compared in the study. It can’t be concluded that the lower dielectric constant, the better performance of sol-gel composite. Also, there is some possibility that the result affected by individual difference depending on manual spray coating. In another study, three types of PZT powder having different dielectric constant were compared. Pulse-echo signal strength of lowest dielectric constant powder phase was lowest. And, pulse-echo signal strength of highest dielectric constant powder phase was lower. [4] However, film quality of lowest dielectric constant powder phase was low. Difference of particle size could cause the low film quality. Therefore, it is necessary to research the effect of piezoelectric powder phase permittivity on condition that particle size is same.

In this paper, $Pb(Zr, Ti)O_3/Pb(Zr, Ti)O_3$ (PZT/PZT) sol-gel composite films were fabricated by automatic spray method. And, dielectric constant effects of powder phase were investigated with three kinds of sol-gel composite materials made by same PZT sol-gel and different PZT powders, very similar properties except dielectric constant and Curie temperature.

2. Fabrication process of PZT/PZT films

In this study, PZT/PZT thin films were fabricated onto titanium substrates by sol-gel spray coating method. The area of the titanium substrate was 3cm × 3cm square. And, the titanium substrate thickness was 3mm.

First of all, PZT sol-gel solution and PZT powder were mixed by ball milling machine for a day. After mix, the mixture was sprayed onto titanium substrate with automatic spray coating machine. Automatic spray was performed two times per 1cycle. Drying at room temperature was performed for 5minutes. During spray coating and drying at room temperature, the space was kept humidity at 30% or lower. Drying at 150℃ was performed on a hot plate for 5minutes. After that, firing at 650 ℃ was performed in furnace for 5minutes. Spray and drying, firing process was repeated until target thickness. In this study, target thickness was 50µm.

Corona poling was performed at room temperature after reaching target thickness. During poling process, poling space was kept humidity at 30% or lower. Silver top electrodes were fabricated by a conductive pen. Ultrasonic responses were measured at each electrode size for performance comparison purpose between three kinds of sol-gel composite materials.

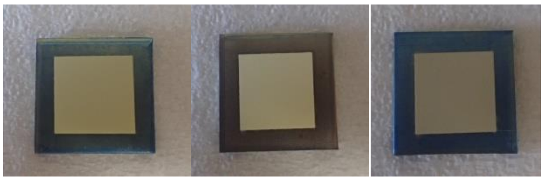
In this study, three types of PZT powder, HIZIRCO A(A), HIZIRCO L(L), HIZIRCO MPT(MPT) were used for PZT/PZT film. Particle size of PZT powder was same. **Table I** shows property of each powder. As shown in Table 1, dielectric constant of each PZT powder is different even though electromechanical coupling coefficient k_{33} is the same. Piezoelectric constant d_{33} is varied almost according to relative dielectric constant, so that it was expected that piezoelectric constant $_{g33}$ becomes almost the same.

Table I: Property of PZT powder

Powder	Property		
	ε_r	k_{33}[%]	d_{33} [pm/V]
HIZIRCO A	5500	70	660
HIZIRCO L	1800	70	400
HIZIRCO MPT	1300	70	290

3. Experimental Results

Fig.1 shows optical images of PZT/PZT sol-gel composite films. Thicknesses of A/PZT, L/PZT, MPT/PZT film was 53µm, 51µm, 52µm. Piezoelectric constant d_{33} of A/PZT, L/PZT, MPT/PZT film was 33.5, 43.9, and 22.3 pC/N, respectively. d_{33} of A/PZT was lower than L/PZT despite highest d_{33} of HIZIRCO A powder. It would mean that poling degree of PZT powder A was not high due to high permittivity. d_{33} of MPT/PZT was lowest in three types of PZT/PZT films. It results from lowest d_{33} value of HIZIRCO MPT powder.



(a) (b) (c)

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

Ultrasonic response was measured for each sample in pulse-echo mode. **Fig.2** shows typical measurement result obtained by L/PZT film. Similar ultrasonic responses were obtained from all the samples. FFT was executed for the samples with 3mm diameter top electrode. 1st reflected echo From FFT result, center frequency of A/PZT, L/PZT, MPT/PZT were 11.2MHz, 15.1MHz, 12.2MHz and 6dB bandwidth of A/PZT, L/PZT, MPT/PZT were 10.9, 18.9, and 11.6MHz, respectively. All the samples has broadband characteristic. However, center frequency of L/PZT was somehow higher than those of A/PZT and MPT/PZT. Since top electrode was made by hand painting of silver paste to change top electrode size easily and top electrode thickness difference might affected frequency characteristic. Further investigation is required.

Sensitivity of PZT/PZT films were derived by using the value of signal amplitude as following;

$$\text{Sensitivity} = -(20\log_{10}\frac{V_1}{V_2} + \text{ gain })(\text{dB}) \quad (1)$$

where V_1(V) is reference amplitude ($0.1V_{p-p}$in this experiment), V_2 (V) is the signal amplitude of the 2nd reflected echo, gain is gain of pulser/receiver. As a result, the sensitivity of A/PZT, L/PZT, and MPT/PZT was 13.3dB, 30.4dB, and 29.3dB, respectively. The sensitivity measurement, the samples had optimized top electrode area. The sensitivity of A/PZT was lowest among the samples and it corresponded to the piezoelectric constant d_{33} value. MPT/PZT showed comparable sensitivity with L/PZT, because of comparable piezoelectric constant g_{33} value. Capacitance measurements will be carried out for g_{33} calculation.

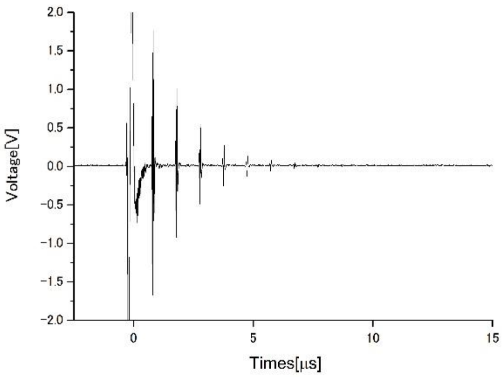


Fig.2 Typical ultrasonic response in pulse-echo mode obtained by L/PZT sample.

4. Conclusions

In this paper, three types of PZT powders with different dielectric constant values, A , L, MPT, were used to fabricate PZT/PZT sol-gel composite samples, A/PZT, L/PZT, MPT/PZT for powder phase dielectric constant effect evaluation. d_{33} and sensitivity of L/PZT sample was highest, whereas those of A/PZT sample was lowest. From the result, it is thought that poling degree of sol-gel composite would be low in case powder phase had too high permittivity, though there would be saturation as well.

References

1. K. Kimoto, M. Matsumoto, T. Kaneko, and M. Kobayashi: Jpn. J.Appl. Phys. **55** (2016) 07KB04
2. M. Kobayashi: Acoustical Society of Japan. **71** (2015) 253.
3. M. Kobayashi, T. Inoue and M. Sawada: Proc. Symp. Ultrason. Electronics (2012) 209.
4. Y. Kiyota, K. Nakatsuma and M. Kobayashi: Symp. Ultrason. Electronics, **38** (2017) 3P1-6