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Sol-gel composite Film Measurement by Scanning Nonlinear Dielectric Microscopy

走査型非線形誘電率顕微鏡によるゾルゲル複合膜の測定

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1. Introduction

In industrial facilities such as power plants and chemical plants, it is important to prevent accidents from occurring. This is because a single accident can have a large impact on society, so continuous monitoring during operation is required. Therefore, non-destructive inspection using ultrasonic transducers is effective. This is a method for detecting sub-surface defects in objects. However, since the backing material and couplant of ultrasonic transducers cannot be used at high temperatures, it is necessary to stop the operation of the object to be measured at high temperatures for inspection.(1) Therefore, in order to develop an ultrasonic transducer that can be used at high temperatures, we developed an ultrasonic probe using the sol-gel spray method.(2) In a previous study, we measured the ultrasonic response of PT/PZT by polarizing it with a corona discharge of negative polarity, and found that when the polarity of the P/R and the poling voltage were matched, depoling was suppressed and high-temperature durability was improved.(3) However, because of the chaotic nature of corona discharges, the poling may not be uniform. In this study, the poling distribution of unpoled PT/PZT and PT/PZT poled by negative corona discharges was measured using a Scanning Nonlinear Dielectric Microscopy (SNDM). (4)

2. Sample Fabrication

PT/PZT sol-gel composite was fabricated by mixing PT powder and PZT solution and coated on titanium substrate by automatic spraying system. The dimensions of the titanium substrate were 30mm*30mm*3mm, which was selected because of its high heat resistance and low heat capacity. After spraying, the film was dried and fired in the sprayer at room temperature, in a drying furnace at 150°C, and in a firing furnace at 650°C for 5 minutes each. This process was repeated until the sample thickness reached 50um. After that, the film was poled by corona discharge with negative polarity. First, PT/PZT was heated in a furnace at 400°C for 5 minutes, and then poled at 40 kV for 5 minutes. The optical image of the fabricated PT/PZT sol-gel composite film is shown in **Fig. 1.**

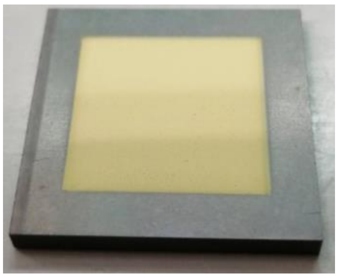

Fig. 1 Optical image of PT/PZT film on 3mm thick titanium substrate.

3. SNDM

SNDM is a microscope to visualize electrical anisotropy such as poling and electric dipole moment in a material. When an electric field E with an angular frequency ω_p is applied to a piezoelectric film, the capacitance Cp(t) changes alternately with time due to the electric field. The electric flux density D and electric field E of a dielectric are given by Equation (1).

$$\boldsymbol{D} = \boldsymbol{P}_r + \epsilon_{33}\boldsymbol{E}_3 + \frac{1}{2}\epsilon_{333}\boldsymbol{E}_3^2 + \frac{1}{6}\epsilon_{3333}\boldsymbol{E}_3^3 + \cdots \quad (1)$$

ε_{333} has the property of changing polarity according to the direction of residual poling P_r and is called the nonlinear dielectric constant. The ratio of the capacitance$\triangle C_p$ that changes when an electric field of $E_3\cos(\omega t)$ is applied externally to a ferroelectric material to the capacitance C_{p0} at zero applied field is given by Equation(2).

$$\frac{\Delta C_p(t)}{C_{p0}} = \frac{\epsilon_{333}}{\epsilon_{33}} E_3 \cos\omega_p t + \frac{\epsilon_{3333}}{4\epsilon_{33}} E_3^2 \cos 2\omega_p t \quad (2)$$

From Equation (2), the capacitance change caused by the third-order nonlinear dielectric constant varies at the same frequency as the applied electric field, and its amplitude is proportional to the applied electric field. By extracting the component that synchronized with the applied electric field through the LC resonance circuit, the nonlinear dielectric constant can be measured. (5) The configuration of

SNDM is shown in **Fig. 2**.

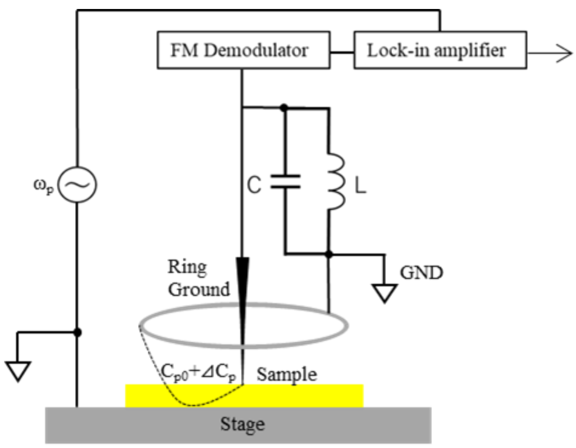

Fig. 2 The configuration of SNDM.

When a voltage of angular frequency ω_p and amplitude V_p is applied to the sample from an external electric field, the capacitance of the sample changes by ΔC_p due to nonlinear effects, which is reflected in the change in the resonant frequency of the LC resonator. This is converted to amplitude and phase by an FM demodulator and lock-in amplifier to visualize the poling distribution.

3. Experimental Results

The SNDM measurements were performed on unpoled and poled PT/PZT. The results are shown in **Figs. 3.** and **4.** The left side of the figure shows the amplitude, and the right side shows the phase. The phase represents the phase of the measured point, which is usually around ±90°, otherwise it is affected by noise. If the polarity of the measurement point in the dielectric is slightly upward, it is recorded as 90°. The amplitude represents the coefficient of the first term in equation (2). The amplitude indicates the volume integral of the nonlinear dielectric constants' vertical component in the measurement area of the probe tip. For non-poled samples, the polarization is distributed in various directions and the volume integral is canceled, as a result, the amplitude of the output becomes almost zero. In constant, for the poled sample, the larger the amplitude, the more polarization is occurring in the ±90° direction, because this system measures the vertical component of the nonlinear dielectric constant. The amplitude of the unpoled sample is low, and more than 90% of the samples are distributed between 0 and 500mV, and the phase is also randomly distributed. The poled samples had 4% of the total distribution between 0 and 500 mV, more than 80% of the total distribution between 2000 and 6000 mV, and the phase was uniformly distributed around 90°.

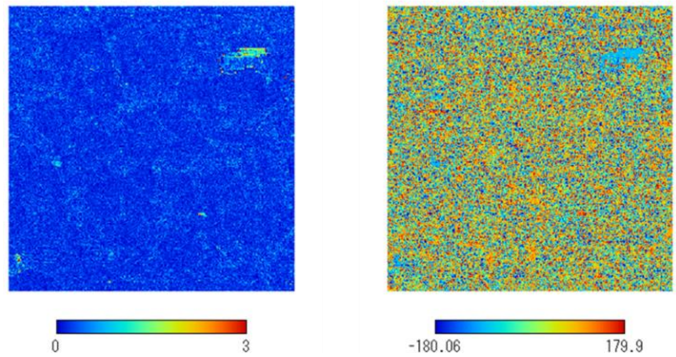

Fig. 3 SNDM of unpoled PT/PZT Left: Amplitude Right: Phase.

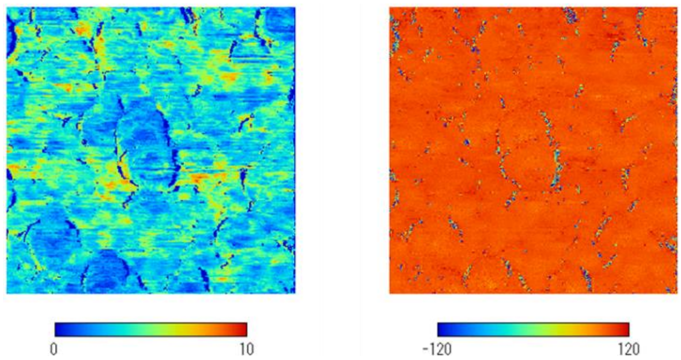

Fig. 4 SNDM of poled PT/PZT Left: Amplitude Right: Phase.

4. Conclusion

The unpoled and poled by negative corona discharge PT/PZT sol-gel composite films were measured by SNDM to evaluate the poling conditions by corona discharge. The results showed that the samples poled by corona discharge were sufficiently poled in the measurement range of 1mm*1mm. In the future, the samples with different poling degrees will be measured by SNDM for future investigation.

References

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