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Evaluation of Ferroelectric Domain Size by Impedance Response in $Pb[(Zn_{1/3}Nb_{2/3})_{0.91}Ti_{0.09}]O_3$ Single Crystal

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Domain size in $Pb[(Zn_{1/3}Nb_{2/3})_{0.91}Ti_{0.09}]O_3$ (PZNT91/09) single-crystal plates was analyzed by measuring the impedance responses of k_{31} fundamental vibration and their overtones. Increasing the poling field (E), the split impedance responses of odd number overtones became single response, as a result, the single-crystal plate possessed giant k_{31} over 80%. The homogeneity of the domain size distribution was estimated by the E to realize the single response of the overtones. The frequency of 410 kHz to obtain the 11^{th} overtone on k_{31} mode corresponded to 1.1 mm on domain size distribution. The poling field dependence of the impedance responses of the fundamental vibration on k_{31} mode was fitted in well with the impedance response analysis of the overtones on k_{31} mode.

Key words: Domain size, Impedance response, Overtone, $Pb[(Zn_{1/3}Nb_{2/3})_{0.91}Ti_{0.09}]O_3$ single crystal, Giant k_{31}

1. INTRODUCTION

The evaluation of ferroelectric domain size in relaxor single crystals has been carried out by TEM, AFM, PFM (piezoresponse force microscopy) and optical microscope observations [1,2]. However, the size of domains evaluated was mainly in the range from nano to micron. Furthermore, it is difficult to estimate the relationships between the size of nano or micro domain and the piezoelectric properties, especially the electromechanical coupling factors such as k_{31} and k_{33}. We found recently that (100) $Pb[(Zn_{1/3}Nb_{2/3})_{0.91}Ti_{0.09}]O_3$ (PZNT91/09) single-crystal plates poled in [100] direction had giant k_{31} over 86% by controlling the domain strictures in the plates [3-5]. On the process to obtain giant k_{31}, we measured frequency responses of impedance regarding the fundamental vibration and their overtones on k_{31} mode at various poling fields.

In this study, we report that the impedance response analysis on vibration modes of single-crystal plates is a useful tool to evaluate the domain size and the distribution.

2. EXPERIMENTAL POCEDURE

(100) PZNT91/09 single-crystal plates with dimensions of 13^Lx4.0^Wx0.37^T mm were prepared to cut from crystal-bulk grown by a solution Bridgman method [6]. Gold electrodes for the following DC poling and electrical measurement were fabricated on the (100) planes by conventional sputtering. Poling was conducted at 40°C for 10 min while varying the poling field (E) from 0 to 1400 V/mm. After each poling, the dielectric and piezoelectric properties were measured at room temperature using an LCR meter (HP4263A), an impedance/gain-phase analyzer (HP4194A) and a d_{33} meter (Academia Sinica: ZJ-3D). Furthermore, the frequency responses of impedance between 0~500 kHz (k_{31} fundamental vibration and their overtones) and between 30~90 kHz (k_{31} fundamental vibration) were measured.

3. RESULTS AND DISCUSSION

3.1 Poling field dependence of dielectric and ferroelectric properties

Figures 1 (a)~(d) show the poling field dependence of dielectric constant (ε_r), k_{31}, piezoelectric d_{31} and d_{33} constants, while two PZNT91/09 single-crystal plates were DC poled. In the poling process, one plate sample was realized giant k_{31} over 80% and the other was k_{31}=60%. The coercive field for poling to obtain giant k_{31} was 800 V/mm, because of the k_{31} and d_{31} behavior toward E (Figs. 1 (b) and (c)). It is found that the domain structures in the thickness were almost same among two samples (Figs. 1 (a) and (d)); on the other hand, the domain structures in the plate were different (Figs. 1 (b) and (c)).

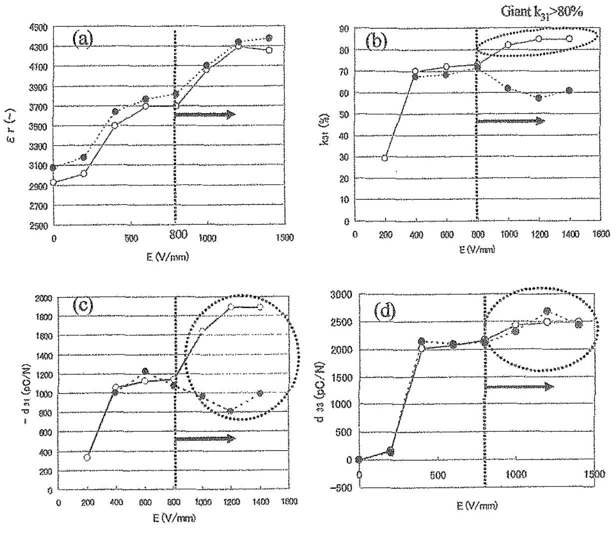

Fig. 1 Poling field (E) dependence of ε_r, k_{31}, d_{31} and d_{33} in the process to obtain (○) and not to obtain (●) giant k_{31} over 80% in PZNT91/09 single-crystal plates.

3.2 Frequency responses to 500 kHz

Figures 2 (a)~(f) show the frequency responses of impedance in the process to obtain giant k_{31} at various poling fields.

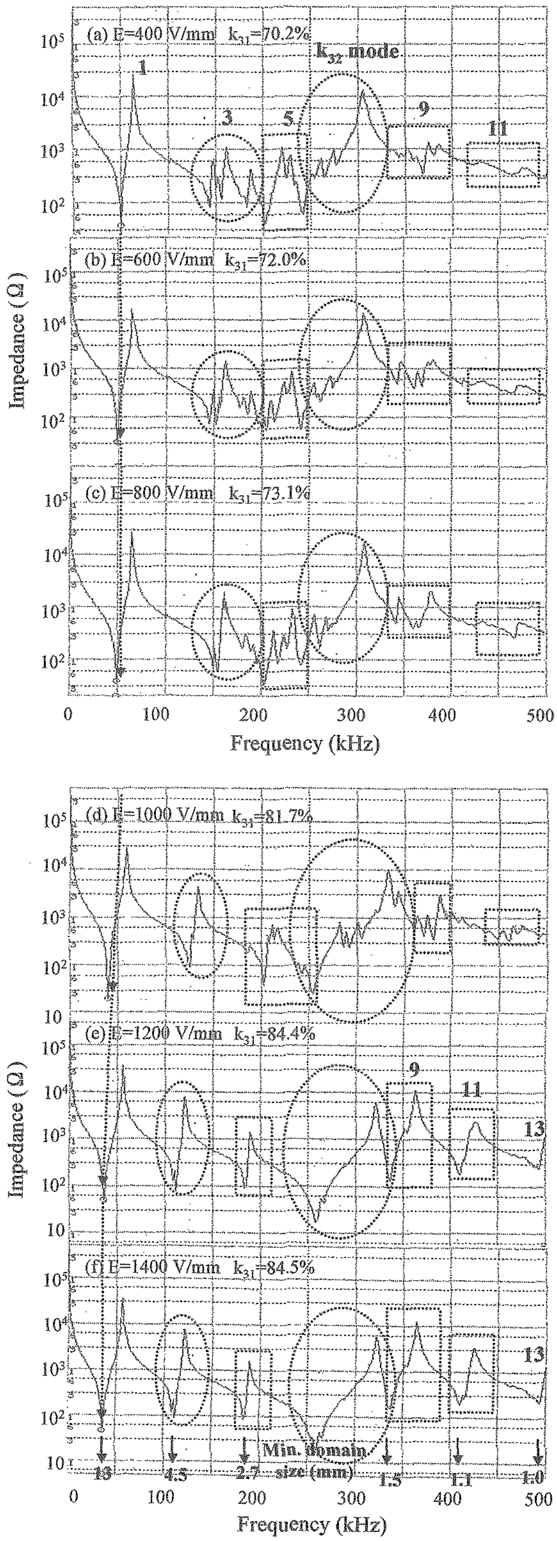

Fig. 2 Frequency responses (0~500 kHz) of impedance in the process to obtain giant k_{31} at various poling fields (E); No. 1: fundamental vibration and Nos. 3, 5, 9, 11, 13: 3^{rd}, 5^{th}, 9^{th}, 11^{th}, 13^{th} overtones on k_{31} mode.

The number in the figures indicates the fundamental vibration (No. 1) and their odd number overtones (Nos. 3~13). Although the 3^{rd} overtone response at E=400 V/mm (No. 3 in Fig. 2 (a)) split into three parts, the parts became single response at E=1000 V/mm (Fig. 2 (d)) and the k_{31} reached to 80% simultaneously. The 5^{th}, 9^{th}, 11^{th} and 13^{th} overtones consisted of single response at E=1200 kV/mm (Fig. 2(e)). The responses near 300 kHz shown in ellipses are k_{32} fundamental vibration, which corresponds to the width (4.0^W mm) direction. The k_{32} fundamental vibration also became single response at E=1200 V/mm (Fig. 2(e)). Those mean that the single-crystal plate poled at E≧1200 V/mm possesses single vibration mode along the width (4.0^W mm) as well as the length (13^L mm).

From the E to realize the single response of the overtones, we can estimate the homogeneity of the domain size distribution. The frequency constant of k_{31} mode (fc_{31}) is defined as fc_{31}=fr x L, where fr and L are the resonant frequency and the length of vibration body. As the size of the vibration body corresponds to the domain size, we can recognize the domain homogeneity by the E to obtain single response. For example, the frequencies of the fundamental vibration, 3^{rd}, 5^{th} 9^{th} and 11^{th} overtones with single response are equivalent to the homogeneity of domain size in the plate such as 13, 4.5, 2.7, 1.5, 1.1 and 1.0 mm, because the fc_{31} of giant k_{31} plate is 520 Hz-m. For example, the relationships between the 3^{rd} overtone and domain size (~4.5 mm) in the plate are shown in Fig. 3. Therefore, the homogeneity of the domain size was estimated by the impedance response analysis on the vibration modes.

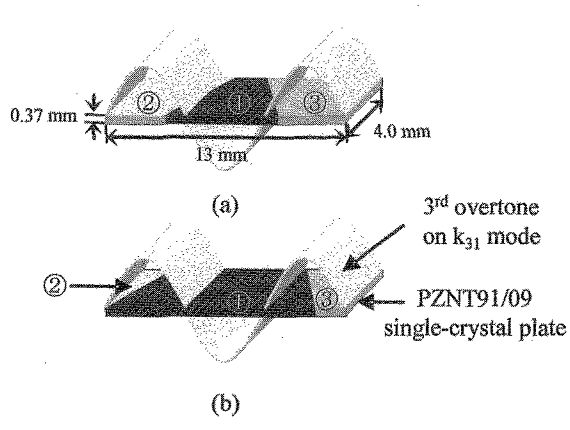

Fig. 3 Schematic pictures to describe relationships between 3^{rd} overtone splitting and domain size at (a) E=400 V/mm in Fig. 2(a) and (b) E=600~800 V/mm in Figs. 2(b) and (c); ① domain increases in size and ② and ③ domains decreases in size with increasing E.

Figures 4 (a)~(f) show the frequency responses of impedance in the process not to obtain giant k_{31} at various poling fields. The number in the figure indicates the frequencies of the vibration modes (k_{31} fundamental and their overtones) to be estimated by the data in Figs. 2 (a) ~ (f). The overtones of k_{31} mode split into many parts with other spurious vibrations, as a result, k_{31} became around 60%. It was thought that multi-domain was generated in the single-crystal plate.

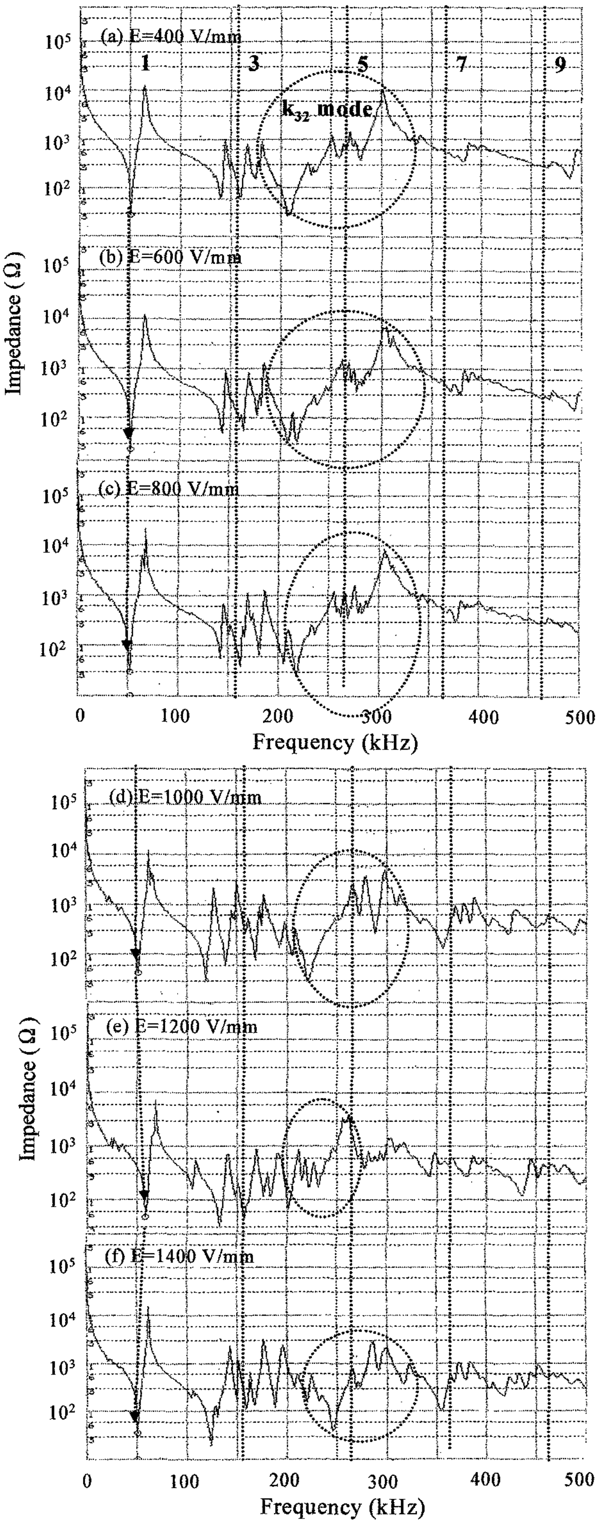

Fig. 4 Frequency responses (0~500 kHz) of impedance in the process not to obtain giant k_{31} at various poling fields (E); No. 1: estimated fundamental vibration and Nos. 3, 5, 7, 9: 3rd, 5th, 7th, 9th overtones on k_{31} mode.

3.3 Frequency responses on k_{31} fundamental vibration

Figures 5 (a)~(f) show the frequency responses of the k_{31} fundamental vibration in the process to obtain giant k_{31} at various poling fields. Since the impedance responses and the phases near anti-resonant frequency were split, the single-crystal plates became multi-domain during E of 400 to 1000 V/mm (Figs 5(a)~(d)). Otherwise, there were no spurious vibration in the fundamental k_{31} responses at E≧1200 V/mm (Figs. 5(e) and (f)). Therefore, single domain was achieved in the plates.

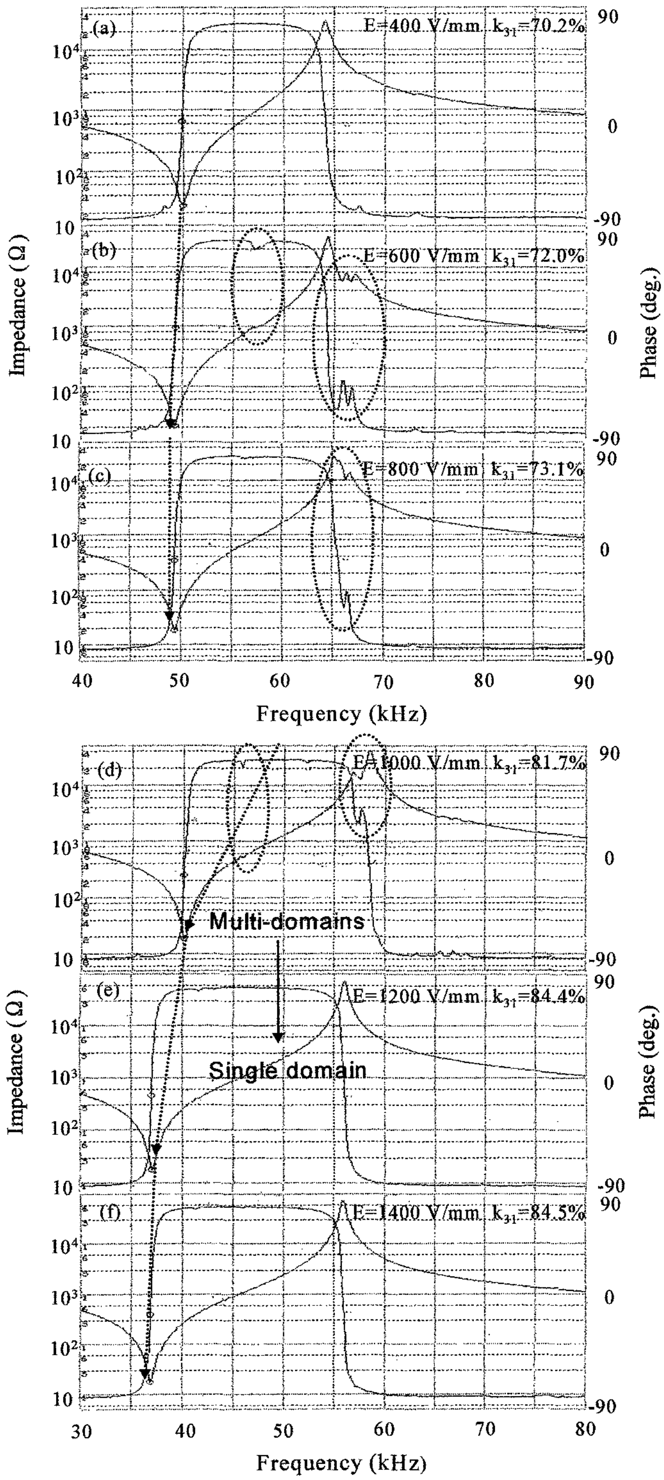

Fig. 5 Frequency responses (30~90 kHz) of impedance and phase of fundamental vibration on k_{31} mode in the process to obtain giant k_{31} at various poling fields (E).

On the other hand, there were many spurious vibrations in the process not to obtain giant k_{31} shown in Figs. 6 (a)~(f). These results fitted in well with the ones of the impedance response analysis on vibration modes in the frequency range of 0~500 kHz.

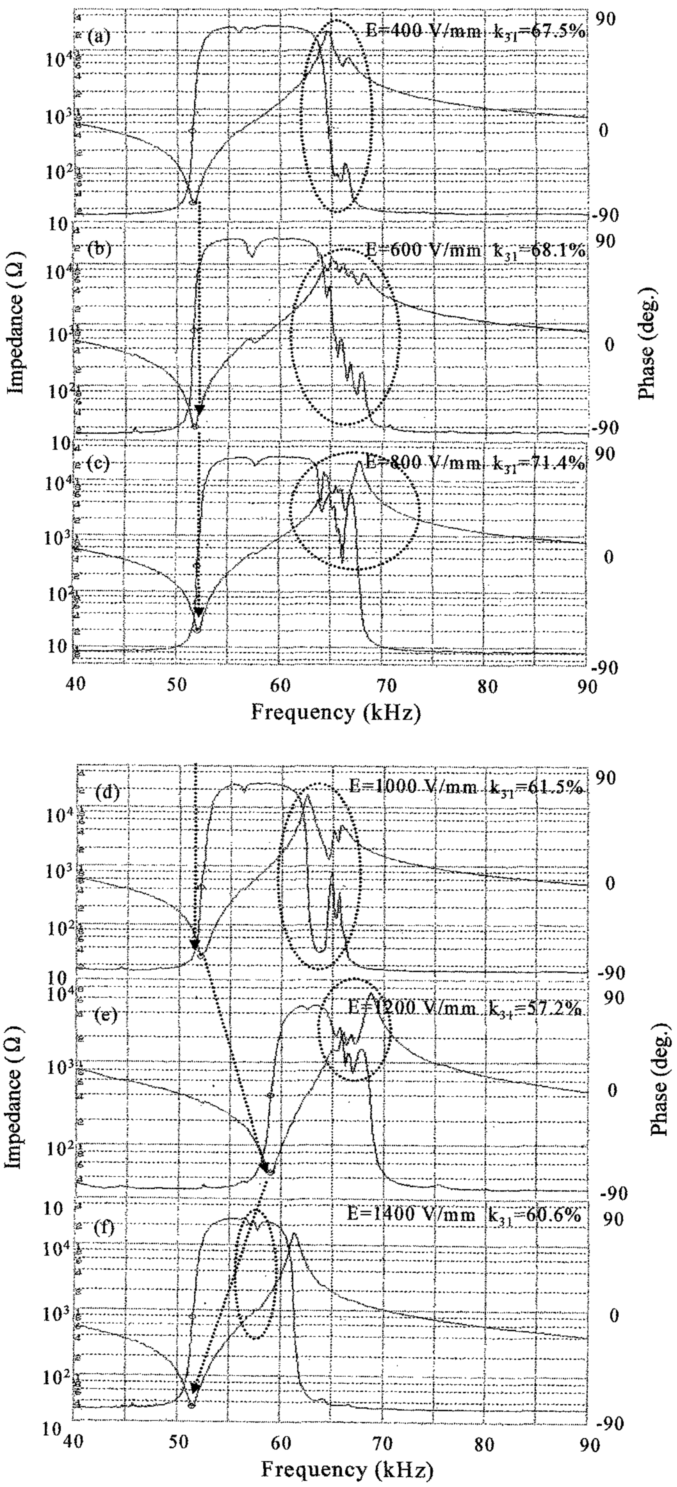

Fig. 6 Frequency responses (40~90 kHz) of impedance and phase of fundamental vibration on k_{31} mode in the process not to obtain giant k_{31} at various poling fields (E).

4. CONCLUSIONS

New method to evaluate ferroelectric domain size was proposed. The method was developed through the measurement on frequency responses of impedance regarding relaxor single-crystal plates with giant k_{31}. It was found that the impedance response analysis on fundamental vibration and their overtones was a useful tool to evaluate the domain size and the distribution.

5. ACKNOWLEDGMENTS

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