Activation of piezoelectric property of PZT thin films by pulse poling

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Abstract. We have demonstrated the pulse poling technique to activate the piezoelectric property of PZT thin films within 1 second. We have fabricated piezoelectric microcantilevers with Si proof mass using Pb(Zr₀.₅₂,Ti₀.₄₈)O₃ (MPB-PZT) thin films. By applying 1 kHz and 100 V of unipolar triangle pulse voltage, the positive piezoelectric constant $|d_{31}|$ of PZT thin film has been enhanced as high as 95 pm/V, which is larger than dc poled PZT thin films (67 pm/V).

1. Introduction
Microelectromechanical systems (MEMS) devices using lead zirconate titanate (PZT) thin films (PZT-MEMS) are utilized to gyros [1], inkjet heads [1], optical microscanners [2], ultrasonic probes [3] and so on. We are also developing new applications of PZT-MEMS including electrostatic field sensors [4], trigger sensor to activate wireless sensors [5,6] and energy harvesters [7,8].

In order to activate the piezoelectric property of PZT thin films, dc voltage of 10-100 kV/cm is usually applied for at least several minutes as dc poling [9,10]. Such a several minutes of procedure, however, affects productivity at mass production stage. One of the solution for this problem is to use very expensive probe card, which is available only for fixed electrode pattern.

The other possible solution is to reduce poling time by using pulse poling, where pulse voltage is applied [11-13]. Recently, we have found that bipolar pulse poling, where intermittent ac triangle wave larger than coercive field of the PZT thin films is applied, enhances the piezoelectric property of tetragonal composition (Zr/Ti = 30/70) PZT (tetra-PZT) thin films [14]. It is well known that the morphotropic phase boundary (MPB, Zr/Ti = 52/48) composition PZT (MPB-PZT) thin films have the largest piezoelectric constant among all compositions [15]. Thus, MPB-PZT thin films are most important for the practical application. Bipolar pulse poling can be effective for MPB-PZT thin films as well as tetra-PZT thin films. Moreover, unipolar pulse poling, where positive or negative pulse voltages are applied, can be also effective to enhance the piezoelectric property. Then, in the present study, we have investigated the influence of bipolar and unipolar pulse poling on the piezoelectric property of MPB-PZT thin films on MEMS based piezoelectric microcantilevers.
2. Device fabrication

2.1. PZT deposition and MEMS microfabrication

The 1.9-μm-thick PZT thin films were deposited by sol-gel method on thermally oxidized silicon-on-insulator (SOI) wafers with Pt (100 nm)/Ti (10 nm) bottom electrodes [16]. The SOI wafers have 10 μm structural Si and 1 μm buried oxide (BOX). The Pb/Ti/Zr composition of the precursor solution was 120/52/48. After PZT deposition, Pt (100 nm)/Ti (10 nm) top electrodes were deposited as top electrode. The deposited multilayers were fabricated into piezoelectric microcantilevers with Si proof mass as shown in Fig. 1(a,b). We have also fabricated piezoelectric microcantilevers without Si proof mass to estimate inverse piezoelectric constant $d_{31}$ for comparison.

2.2. Pulse poling

After packaging, dc and triangle pulse voltage as illustrated in Fig. 1(c1-c4) was applied. The top and bottom electrodes were connected to signal and ground lines, respectively. Figure 1(a) is a conventional dc poling. When positive voltage is applied, the polarization direction becomes downward as shown in fig. 1(c1). The present conditions for dc poling is +20V and -20V for 5min.

Figure 1(c2) represents bipolar pulse poling reported in our previous study [14]. Intermittent triangle wave of 30-100 V and 1 kHz was applied 10 times at 0.1 sec intervals. Since the last triangle wave is negative voltage, polarization direction is upward. Figures 1(c3) and 1(c4) illustrate unipolar pulse poling, which we newly employed in the present study. Positive or negative intermittent triangle wave of 30-100 V and 1 kHz was applied 10 times. When positive or negative triangle waves are applied, the polarization directions are downward and upward, respectively. Note that the respective coercive voltage of the present tetra- and MPB-PZT thin films are about 20 V and 7V, which means that the dc and pulse poling voltages are larger than the coercive voltage of PZT thin films.

![Figure 1.](image)

**Figure 1.** (a) Schematic of the piezoelectric microcantilevers with Si proof mass. (b) Packaged piezoelectric microcantilevers with proof mass. Waveforms of poling voltage for (c1) conventional DC poling, (c2) bipolar pulse poling, (c3) downward unipolar pulse poling, and (c4) upward unipolar pulse poling.

3. Test and results.

3.1. Piezoelectric property

The packaged piezoelectric microcantilevers were mounted on a shaker and vibrated to characterize the positive piezoelectric constant $d_{31}$, open circuit voltage (OCV), and ac power generation. The data are summarized as a function of pulse-poling voltage in the following. Figure 2(a) shows absolute values of...
positive piezoelectric constant $d_{31}$ estimated from the output charge $Q$ and acceleration $a$ using the equations,

\begin{align}
  d_{31} &= KQ/3L^2 s_{11}^{\text{Si}} t_{31} (t_{31} + t_{\text{PZT}}) m a \\
  K &= (s_{11}^{\text{Si}})^2 t_{\text{PZT}}^4 + 4 s_{11}^{\text{Si}} s_{11}^{\text{PZT}} t_{31} t_{\text{PZT}}^3 + 6 s_{11}^{\text{Si}} s_{11}^{\text{PZT}}^2 t_{31}^2 t_{\text{PZT}} + 4 s_{11}^{\text{Si}} s_{11}^{\text{PZT}} t_{31} t_{\text{PZT}}^2 + (s_{11}^{\text{PZT}})^2 t_{31}^4 
\end{align}

where $m$, $L$, $s_{11}$, and $t$ are mass, microcantilever length, compliance and thickness of Si and PZT. Although the $|d_{31}|$ of dc poled MPB-PZT thin films in only 67 pm/V, that of unipolar pulse poled MPB-PZT thin films increases with voltage and reaches 95 pm/V at 100 V. Figure 2(b) also shows inverse piezoelectric constant $d_{31}$, estimated from the equations,

\begin{align}
  d_{31} &= K\delta /3ABL^2V \\
  A &= s_{11}^{\text{Si}} s_{11}^{\text{Si}} (s_{11}^{\text{PZT}} t_{\text{PZT}} + s_{11}^{\text{PZT}}) \\
  B &= t_{31} (t_{31} + t_{\text{PZT}})/(s_{11}^{\text{Si}} t_{\text{PZT}} + s_{11}^{\text{Si}} t_{31})
\end{align}

where $\delta$ is tip displacement of the piezoelectric microcantilevers actuated at the voltage $V$. The inverse piezoelectric constant shows similar tendency with that of the positive piezoelectric constant. The inverse $|d_{31}|$ reaches 105 pm/V at the pulse poling voltage of 100 V, which is a little bit larger than positive $|d_{31}|$.

\begin{figure}[h]
  
  \includegraphics[width=\textwidth]{figure2.png}
  
  \caption{Absolute values of (a) positive and (b) negative piezoelectric constant $d_{31}$ as a function of pulse poling voltage.}
\end{figure}

### 3.2. XRD characterization

The increase in $|d_{31}|$ of MPB-PZT thin films with pulse poling voltage can be derived from alignment of polarization direction by disappearance of 180° domain wall and increase in $c$-axis orientation. Then, we have investigated the variation of $c$-axis orientation with unipolar pulse poling by XRD measurement. Figure 3(a) shows XRD patterns around (004)/(400) peaks of MPB-PZT thin films pulse poled at 40-100 V. The peak intensity at $2\theta = 99.1^\circ$ gradually decreased at the unipolar pulse poling voltage of 40-80 V and abruptly decreased at the voltage of 100 V. In addition, the intensity of the shoulder around 97°-98° increased by unipolar pulse poling at 100 V. We have estimated the volume fraction of the $c$-domain ($V_c = V_{(004)}/V_{(004)}/(400)$) by the pseudo-Voigt function fitting.

Figure 3(b) shows the volume fraction of $c$-domain $V_c$ as a function of pulse poling voltage. As shown in fig. 3(b), $V_c$ has increased from 37.4 to 41.6% with the unipolar pulse poling voltage from 40 to 100 V. Based on the results, we concluded that the increase in $|d_{31}|$ is attributed to alignment of polarization direction by disappearance of 180° domain at the pulse poling voltage of 40-80 V and to increase in volume fraction of the $c$-domain at the pulse poling voltage of 100 V.
3.3. Open circuit voltage and power generation

Figures 4(a,b) show the open circuit voltage (OCV) and ac power generation of the MPB-PZT thin films measured under resonant condition of the piezoelectric microcantilevers with proof mass, where ac power generation is characterized under impedance matching with a load resistance. The unipolar pulse poled MPB-PZT thin films have the larger OCV and ac power generation than the dc poled ones. The dielectric constant \(e\) of the MPB-PZT thin films was measured to obtain the figure of merit (FOM) of the OCV \(d_{31}/e\) and the ac power generation \(d_{31}^2/e\). The dielectric constant decrease with the increasing pulse poling voltage as shown in fig. 4(c).

Figure 3. (a)XRD patterns around (004)/(400) peaks of MPB-PZT thin films pulse poled at 40-100 V, (b) volume fraction of \(c\)-domain \(V_c\) as a function of pulse poling voltage.

Figure 4. (a) Open circuit voltage (OCV), (b) ac power generation, (c) dielectric constant, (d) figure of merit of OCV, and (e) figure of merit of ac power generation as a function of pulse poling voltage.

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Figures 2(d,e) show relation between the measured data and The measured data is well proportional to the FOM for the OCV, while ac power generation is not. Since the ac power generation is determined by $V/2R$ at impedance matching condition, FOM for power generation should include the resistivity of PZT thin films as well as dielectric constant.

4. Summary
In conclusion, we have succeeded in activating the positive and inverse piezoelectric constant $d_{31}$ of PZT thin films to 95 and 105 pm/V by applying unipolar triangle pulse voltage as pulse poling. It takes within 1 second, which is much shorter than dc poling leading to much better productivity of piezoelectric MEMS devices.

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