Reliability of vibration energy harvesters of metal-based PZT thin films

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Abstract. This paper describes the reliability of piezoelectric vibration energy harvesters (PVEHs) of Pb(Zr,Ti)O$_3$ (PZT) thin films on metal foil cantilevers. The PZT thin films were directly deposited onto the Pt-coated stainless-steel (SS430) cantilevers by rf-magnetron sputtering, and we observed their aging behavior of power generation characteristics under the resonance vibration condition for three days. During the aging measurement, there was neither fatigue failure nor degradation of dielectric properties in our PVEHs (length: 13 mm, width: 5.0 mm, thickness: 104 µm) even under a large excitation acceleration of 25 m/s$^2$. However, we observed clear degradation of the generated electric voltage depending on excitation acceleration. The decay rate of the output voltage was 5% from the start of the measurement at 25 m/s$^2$. The transverse piezoelectric coefficient ($e_{31,f}$) also degraded with almost the same decay rate as that of the output voltage; this indicates that the degradation of output voltage was mainly caused by that of piezoelectric properties. From the decay curves, the output powers are estimated to degrade 7% at 15 m/s$^2$ and 36% at 25 m/s$^2$ if we continue to excite the PVEHs for 30 years.

1. Introduction
In the research of piezoelectric micro-electromechanical systems (MEMS), piezoelectric vibration energy harvesters (PVEHs) using Pb(Zr,Ti)O$_3$ (PZT) thin films have been widely investigated because of their high piezoelectric properties [1–6]. Having good compatibility with conventional semiconductor microfabrication techniques, Si substrates are used for most of the piezoelectric MEMS devices including PVEHs. However, brittle Si is not the best material for PVEHs which incur continuous large vibration. One of the solutions for this problem is to use flexible materials such as organic or metal materials as a base substrate [7–10]. Organic materials have excellent flexibility more than metal, however, their melting points are usually less than 200°C; this indicates that it is impossible to deposit the PZT thin films directly on organic substrates using conventional deposition methods because of their high deposition temperature more than 500°C.

On the other hand, the PZT thin films can be deposited directly on metal substrates since melting points of metals are higher than deposition temperature of piezoelectric thin films, indicating that we can use the same fabrication process with Si-MEMS devices. In recent years, PVEHs of metal cantilevers have been actively studied because they can generate the larger displacement at the resonance without break down unlike brittle Si cantilevers [11–13]. However, there have been no detailed study on the long-term reliability for the power generation characteristics of the PVEHs of PZT thin films on metal cantilevers.
In this study, we fabricated the PZT thin-film PVEHs of stainless-steel (SS430) foil cantilevers using rf-magnetron sputtering. To clarify the relationship between aging behavior of the output power or the piezoelectric properties and excitation magnitudes, we measured the output voltage of our PVEHs for three days (4320 minutes) at the various vibration conditions. Based on the short-range aging characteristics, we discuss the long-term reliability of the PZT thin films on SS430 substrates.

2. Device fabrication

Figure 1 shows a schematic illustration of the PVEH of PZT thin films deposited on SS430 foil cantilever. After deposition of Pt/Ti bottom electrodes, 4.0-µm-thick PZT thin films were then directly deposited onto the Pt-coated SS430 substrate (length: 20 mm, width: 5.0 mm, thickness: 100 µm) by rf-magnetron sputtering [14]. The PZT thin films on SS430 substrate were annealed in atmosphere at 600°C for 1.5 hours. Subsequently, the Pt top electrode was prepared through a shadow mask. We fixed one end in the longitudinal direction of the PZT/SS430 substrate by a clamping jig, and the PVEH of PZT/SS430 unimorph cantilever (length: 13 mm, width: 5.0 mm, thickness: 104 µm) was fabricated.

From X-ray diffraction (XRD) measurement, PZT thin films on Pt-coated SS430 substrate have the perovskite structure with a random orientation (figure 2). We also evaluated the dielectric properties of the PZT thin films using an LCR meter (NF ZM2353), and the relative dielectric constant ($\varepsilon_r$) and the dielectric loss (tan $\delta$) were measured to be 425 and 0.018, respectively. The transverse piezoelectric properties of the PZT thin films were assessed from the direct piezoelectric effect for the PVEHs. After the application of a negative unipolar electric voltage on the top electrode as a poling treatment (1000 Hz, $\sim$20 V), we applied periodical displacement to the tip of the cantilever. The generated electric voltage between the top and bottom electrodes was measured using a lock-in amplifier (SIGNAL RECOVERY 7265). From the relationship between the measured electric voltage and the applied displacement, the transverse piezoelectric coefficient $e_{31,f}$ was calculated using equation (1), [15]

$$e_{31,f} = -\frac{4l^3C_p}{3wh_s(1-\nu_s)x_1-x_0(2l-x_1-x_0)}\frac{V}{\delta}$$

(1)

where $h_s$ and $\nu_s$ are the thickness and Poisson’s ratio of a base substrate; $w$ and $x_0$ ($x_1$) are the width and the extremities of the top electrode; $l$, $\delta$, $V$, and $C_p$ are the length of the cantilever, applied displacement to the cantilever, output electric voltage, and capacitance, respectively. When the frequency of applied displacement was 30 Hz, the transverse piezoelectric coefficient $e_{31,f}$ was calculated to be $\sim$3.5 C/m$^2$ from equation (1).
3. Power generation reliability of PVEHs

3.1. Basic characteristics of power generation

Figure 3 shows a schematic illustration and a photograph of the measurement setup for PVEHs. The PVEH was mounted on the vibration exciter (EMIC 512-A). The acceleration pickup (EMIC 731-B) was attached on the base of the cantilever for monitoring the acceleration amplitude. The top electrode of the PZT thin films was connected to a gold wire using silver paste. The generated electric voltage across an external load resistance was measured using a frequency response analyzer (FRA; NF FRA5095). Average output power \( P \) of the PVEHs was calculated from the root-mean-square (rms) voltage \( V_{\text{rms}} \) in a load resistance \( R \) using

\[
P = \frac{(V_{\text{rms}})^2}{R}
\]

(2)

First, we measured both the resonance frequency and the optimal load resistance which maximize the output power of the PVEHs. Figure 4 shows the frequency response of the output voltage and the cantilever displacement (at 7 mm from the fixed end) of the PVEHs. We swept the excitation frequency, and the output voltage and the cantilever displacement were measured using an FRA and a laser Doppler vibrometer, respectively. As shown in figure 4, clear peaks in both the output voltage and the cantilever displacement appeared at the same frequency of 323.7 Hz at the acceleration of 5 m/s\(^2\). Furthermore, when the acceleration was raised until 25 m/s\(^2\), the resonance frequency increased.

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**Figure 3.** Measurement setup for power generation performance of PVEHs: (a) a schematic illustration and (b) a photograph.

**Figure 4.** Output voltage in an open-circuit state and cantilever displacement as functions of frequency.

**Figure 5.** Output power and output voltage as functions of load resistance.
to 344.4 Hz because of nonlinear resonance of hardening spring effect. Figure 5 shows the output power and output voltage as functions of the load resistance at the resonance frequency, and the optimal load resistance was measured to be approximately 12 kΩ. In the aging measurement, the PVEHs were excited under the conditions of resonance frequency, optimal load resistance, and acceleration obtained in this subsection. Just before the aging measurement, we applied a negative unipolar electric voltage (1000 Hz, −20 V_p-p) on the top electrode for one minute as a poling treatment.

3.2. Aging behavior
Measuring the aging behavior of power generation characteristics, we excited the PVEHs at their resonance frequency measured in the previous subsection 3.1 and adjusted the cantilever displacement to be constant.

During the aging measurement for three days, neither fatigue failure nor degradation of dielectric properties (ε, and tan δ) was observed in our PVEHs under each acceleration of 5, 15 and 25 m/s^2. Figure 6 shows the output voltage in an open-circuit state as a function of vibration time. At the acceleration of 5 m/s^2, the output voltages were almost constant during the three-day measurement. On the other hand, when the acceleration was increased up to 15 and 25 m/s^2, the output voltages monotonically reduced 2.5 mV_{rms} (1.4%) and 11.7 mV_{rms} (4.8%), respectively.

Figure 7 shows the normalized output power as a function of vibration time. Under accelerations of 15 and 25 m/s^2, the output powers reduced 4 and 11% during the measurement period, respectively. The output power at 15 m/s^2 decreased linearly throughout the measurement, whereas the output power at 25 m/s^2 fell rapidly from the times that exceed 200 minutes. The equations in figure 7 represent the decay curves at the accelerations of 15 and 25 m/s^2 calculated using the least square method; x and y mean the vibration time and the normalized output power, respectively. The decay rates of the output powers were 0.93% at 15 m/s^2 and 6.4% at 25 m/s^2 by every decade on a logarithmic time scale. Assuming that the decay curves are appropriate secularly, the output powers of the PVEHs are estimated to decay 7% at 15 m/s^2 and 36% at 25 m/s^2 if we continue to excite the PVEHs for 30 years.

Furthermore, we evaluated the reliability of the PVEH without vibration excitation by measuring the piezoelectric properties. Figure 8 shows the aging characteristics of the transverse piezoelectric coefficient $e_{31,f}$. No clear degradation of the $e_{31,f}$ was observed at the accelerations of 0 and 5 m/s^2. On the other hand, when the accelerations were 15 and 25 m/s^2, the $e_{31,f}$ respectively reduced 1 and 4%, which are almost the same decay rates with those of the output voltages. This indicates that the degradation of power generation performance was mainly caused by that of piezoelectric properties.

From the aging measurement, the PZT thin films on SS430 foil cantilevers are expected to possess good long-term reliability of power generation performance at even large excitation acceleration of 15 m/s^2.

Figure 6. Output voltage in an open-circuit state at the resonance frequency as a function of vibration time.

Figure 7. Normalized maximum output power as a function of vibration time.
4. Conclusions
In this study, we evaluated the reliability of the PZT thin-film PVEHs of SS430 foil cantilevers. During the aging measurement for three days, there was neither fatigue failure nor degradation of dielectric properties in our PVEHs even under a large excitation acceleration of 25 m/s$^2$. However, we observed clear degradation of the output voltage depending on excitation acceleration, and the decay rate of the output voltage was 5% from the start of the measurement at 25 m/s$^2$. The transverse piezoelectric coefficient ($e_{31,f}$) also degraded at almost the same decay rate with that of the output voltage; this indicates that the degradation of the output voltage was mainly caused by that of piezoelectric properties. From the decay curves, the output power at 15 m/s$^2$ is estimated to decay only 7% if we continue to excite the PVEH for 30 years, whereas the output power at 25 m/s$^2$ fell rapidly from the times that exceed 200 minutes. Therefore, the PZT thin films on SS430 foil cantilevers are expected to possess good long-term reliability of power generation performance at even large acceleration of 15 m/s$^2$.

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