Ferroelectric Dipole Electrets Prepared from Soft and Hard PZT Ceramics in Electrostatic Vibration Energy Harvesters

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Abstract. Aiming at longer stability of surface potential, we propose a ferroelectric dipole electret (FDE) prepared from hard ferroelectric material. We compared output power of electrostatic vibration energy harvester and surface potential stability between FDEs prepared from soft and hard PZT ceramics, as well as a CYTOP polymer electret. The hard FDE showed a seven-fold increase in output power over the soft FDE and nine-fold increase over the CYTOP polymer electret. The hard FDE also showed longer stability of surface potential than that of the soft FDE, whereas the stability of the hard FDE was not yet comparable to that of CYTOP polymer electret. A FDE prepared from harder PZT ceramic (with higher coercive electric field and Curie temperature) may provide more stability in surface potential.

1. Introduction

A vibration energy harvester (VEH) is considered to be viable power sources for operating sensor nodes in autonomous wireless sensor networks because of its high power density and inexhaustibility in operational environment [1–3]. In actual operation environment, VEHs are expected to generate power from low-level mechanical vibration (typical frequency and acceleration are below 200 Hz and 9.8 m/s^2, respectively) [2]. An electrostatic vibration energy harvester (E-VEH) using a charged dielectric called "electret" is being recognized as a promising solution to satisfy the aforementioned demand because of its lower operating frequency and wider frequency bandwidth, compared to piezoelectric or electromagnetic one [4–10]. However, its output power still remains lower than that obtained from other VEHs. Increase in surface charge density of an electret is one of the key factors to enhance output power in the E-VEH. Nevertheless, surface charge density of the electret so far developed is limited by the relative permittivity of its dielectric material, such as polymers [11] or silicon-based inorganics [12, 13]. Ferroelectrics could be highly promising because their relative permittivity and the resulting surface charge density exceed 1000 and 100 mC/m^2, respectively.

Recently, we developed an electret consisting of a polarized lead zirconate titanate (PZT) ceramic, and named it ferroelectric dipole electret (FDE) [14, 15]. The E-VEH using the FDE showed an eight-fold increase in output power over a conventional polymer electret; on the other hand, the FDE exhibited poor stability in surface potential [15]. In previous studies, we chose a so-called "soft" PZT to form a FDE for two reasons: high value of remnant polarization (P_r) and easy poling condition [low coercive electric field (E_c) and low Curie temperature (T_c)]. In general, soft PZT can be obtained by donor doping, for example, by substitution of La^{3+} for Pb^{2+} and/or Nb^{5+} for Ti^{4+}/Zr^{4+}. The donor doping will enhance the domain wall mobility by reducing the concentration of oxygen vacancies

playing a role as pinning center for the domain wall motion by forming defect dipole [16–18]. Thus, soft PZT has high P_r, low E_c and T_c. These ferroelectric properties may result in easy depoling and thus the previously-reported poor stability in surface potential. Alternatively, "hard" PZT is generally tuned by acceptor doping, for example, by substitution of K^+ for Pb^{2+} and/or Fe^{3+} for Ti^{4+}/Zr^{4+}, resulting in intentionally increasing oxygen vacancies and thus high E_c and T_c [16, 18]. These properties of hard PZT may improve the stability in surface potential of the FDE.

In this study, we compare output power of E-VEHs and surface potential stability between FDEs prepared from soft and hard PZT ceramics, as well as a CYTOP polymer electret.

2. Fabrication

We employed unpoled 1-mm thick PZT ceramics with no electrodes [Fuji Ceramics Corp., Japan, No.C-6 (soft) and No.C-2 (hard)]. As detachable electrodes to form the FDEs, we attached the copper foil tapes with conductive adhesive (Hitachi Maxell, Japan, SLIONTEC No.8701) on both surfaces of the PZT ceramics [14]. Poling treatment was conducted in silicone oil bath at room temperature for the soft PZT ceramic and at 120 °C for the hard PZT ceramic, respectively. Applied poling electric field and treatment time are 2 kV/mm and 1 hour, respectively. The hard PZT ceramic was cooled down to room temperature for an hour while keep applying the poling electric field. After the poling treatment, we wiped off the silicone oil remained on both surfaces of the PZT ceramics using KimWipe sheets. Then, poled PZT ceramics served as the FDEs by peeling off the copper foil tapes.

For comparison, we also prepared a conventional electret consisting of CYTOP polymer (Asahi Glass, Japan, CTL-809M). An 8.2-μm thick CYTOP polymer was formed by spin-coating on a 1-mm thick copper rigid plate and then charged by using point-to-grid corona discharging method. The detailed set-up of the point-to-grid corona discharging is reported in the reference [10, 11]. Applied needle voltage and grid voltage were set to -2.3 kV and -600 V, respectively. All electrets have the same surface area (length: 20 mm, width: 20 mm).

3. Experimental

We use non-contacting electrostatic voltmeter (Trek Inc., Model 347) to measure the surface potential of electrets. Effective surface charge density σ is calculated from $\varepsilon_r \varepsilon_0 V_s / t$, where ε_r is the relative permittivity of an electret material, ε_0 the permittivity of vacuum, V_s the measured surface potential, and t the thickness of each electret. In this study, the negatively-charged surface is defined as top surface and the opposite is bottom surface.

Figure 1 shows our experimental configuration for evaluating vertical-type E-VEH [7–9, 14, 15]. An electret was put on a lower electrode attached to a shaker controlling vertical vibration, and set to face up a negatively-charged top surface against an upper electrode mounted on a translational stage equipped with a micrometer. Then initial air gap (center of the vibration) between the upper electrode and an electret was adjusted using the micrometer. A goniometer installed under the lower electrode was also used to keep the electret parallel to the upper electrode. Consequential change in the air gap distance under vertical vibration is accompanied by change in the capacitance between the upper electrode and the lower electrode, and generates alternate current. Output voltage V_{out} was calculated from $(r_1+r_0)V_{osc}/r_0$, where r_1 is the variable load resistance (3.3–99 MΩ), r_0 the fixed load resistance (0.5 MΩ), and V_{osc} the voltage measured by the oscilloscope having the internal impedance of 10 MΩ. Output power P_{out} is calculated as follows.

$$P_{out} = \frac{1}{T} \int_0^T \frac{V_{out}^2(t)}{R} dt , \tag{1}$$

where T is the measuring time period (0.5 second) and R is the total load resistance (r_1+r_0).
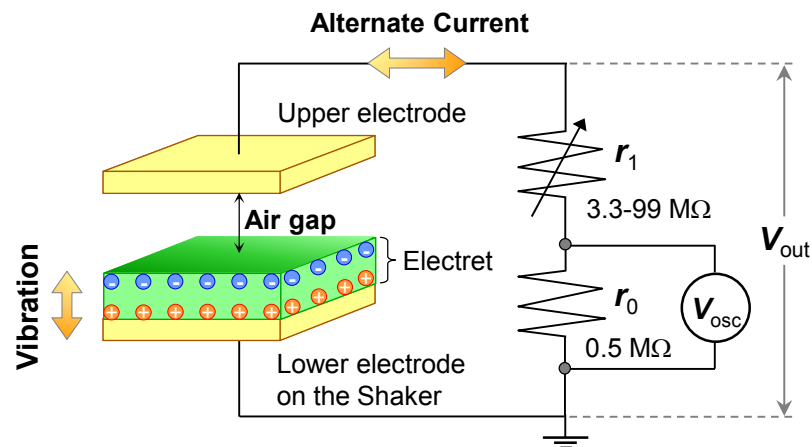
Figure 1. Schematic of the experimental configuration for output power evaluation of vertical-type electrostatic vibration energy harvester.

4. Results and discussion

Table 1 summarizes the measured surface potential and the effective surface charge density of each electret. For each FDE, top and bottom surfaces are charged with similar magnitudes but opposite signs. In contrast, only top surface is effectively charged in the CYTOP polymer electret (CPE). The difference between these electrets is that the FDE utilizes a dipole-orientation by poling, whereas the CPE does implanted charges by corona discharging. The σ of the hard FDE was twice higher than that of the soft FDE.

Table 1. Relative permittivity (ε_r), thickness (t), measured surface potential (V_s) on top and bottom surface, and effective surface charge density (σ) on the top surface of hard and soft ferroelectric dipole electret (FDE) and CYTOP polymer electret (CPE). All electrets have the same surface area (length: 20 mm, width: 20 mm).

	ε_r	t (mm)	top V_s (V)	bottom V_s (V)	σ (mC/m^2)
Hard FDE	1536	1.0	-731	+713	-10
Soft FDE	2281	1.0	-241	+231	-4.9
CPE	2.1	0.082	-580	+32	-1.3

Figure 2 (a) shows waveforms of output voltage at $R = 30.2$ MΩ, and Figure 2 (b) shows output power as a function of total load resistance. Applied frequency, acceleration, and initial air gap were set to 20 Hz, 4.9 m/s^2, and 0.35 mm, respectively. Maximum output powers are 59 µW for the hard FDE, 9.0 µW for the hard FDE, and 6.9 µW for the CPE. The higher output power of the hard FDE may be attributed to retaining the higher surface charge density even after removing the conductive tapes by the hardening of the domain wall motion due to the defect dipole pinning [17].
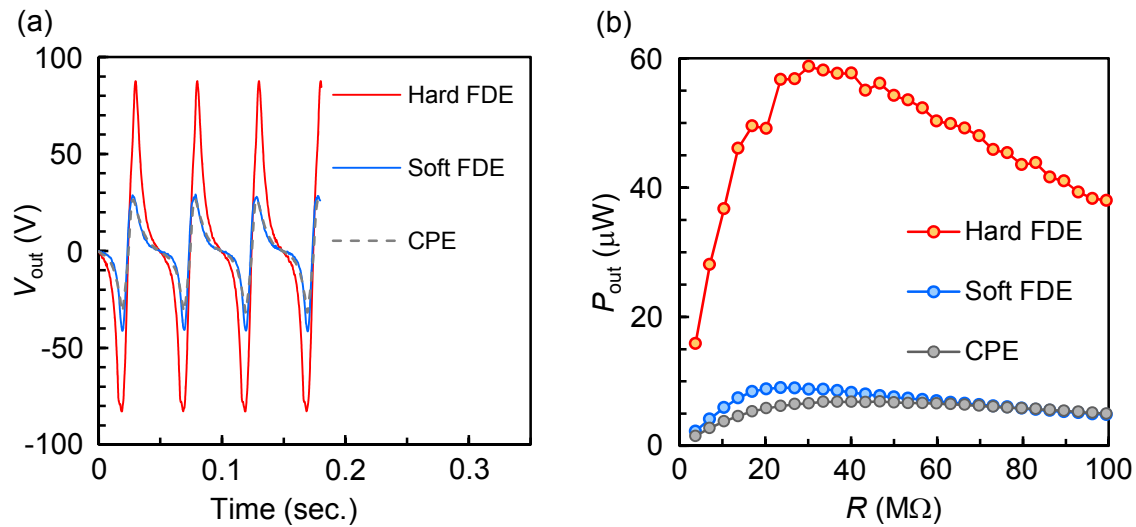
Figure 2. (a) Waveform of output voltage (V_{out}) and **(b)** output power (P_{out}) vs total load resistance (R).

Figure 3 shows surface potential stability over 800 hours. Samples were placed in an experimental room (usually kept 23-25 °C and 50-60 %RH). The surface potential at each measuring point was normalized with the initial value shown in table 1. While the surface potential of the soft FDE started decreasing from one hour later, the surface potential of the hard FDE started decreasing from 30 hours later. The result also may be attributed to the hardening of the domain wall motion due to the defect dipole pinning [17]. The hard FDE showed longer stability in surface potential than that of the soft FDE, whereas the stability of the hard FDE was not yet comparable to that of CYTOP polymer electret. A FDE prepared form harder PZT ceramic (with higher E_c and T_c) may provide further improvement in surface potential stability.
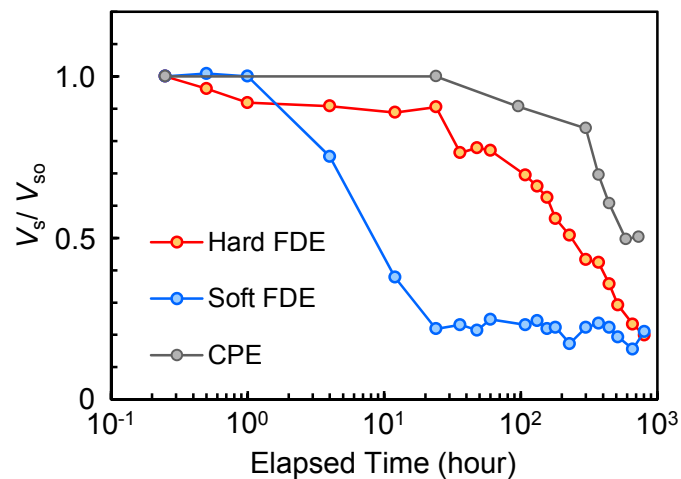

Figure 3. Surface potential stability. Surface potential (V_s) was normalized with the initial value (V_{so}) shown in table 1.

5. Conclusion

We investigated output power of E-VEHs and surface potential stability of hard and soft FDEs, as well as a CPE. The effective surface charge density of the hard FDE reached -10 mC/m^2, which was twice higher than that of the soft FDE. The maximum output power of the hard FDE normalized with surface area of electrets was 15 µW/cm^2, a seven-fold increase over the soft FDE and a nine-fold increase over the CPE. The hard FDE showed longer stability in surface potential than that of the soft FDE. The result may be attributed to the hardening of the domain wall motion due to the defect dipole pinning. However, the stability of the hard FDE was not yet comparable to that of CPE. A FDE prepared from harder PZT ceramics (with higher coercive electric field and Curie temperature) may provide more stability in surface potential.

Acknowledgements

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