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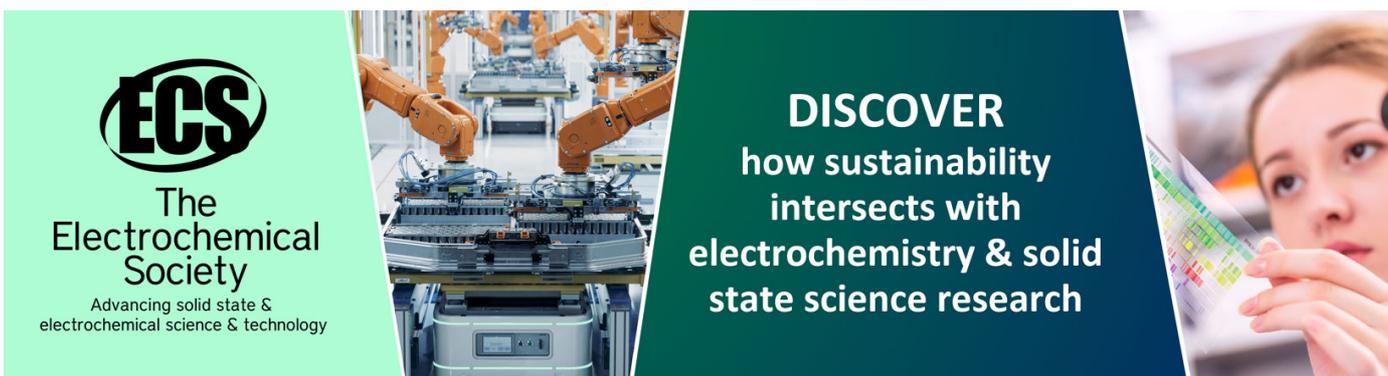
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An electromechanical model of ferroelectret for energy harvesting

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Abstract

A ferroelectret is a cellular polymer foam that is able to convert compressive and bending forces into electrical signals, which can be used for both sensing and energy harvesting. In the past several research groups have proposed theoretical models that relate the output voltage of a ferroelectret to its mechanical deformation. This is particularly useful for sensing applications where the signal-to-noise ratio is important. However, for energy harvesting applications, a theoretical model needs to include both the voltage across a resistive load and the duration of the electrical signal as energy is an integral of power over time. In this work, we propose a theoretical model that explains the behavior of a ferroelectret when used as an energy harvester. This model can be used to predict the energy output of a ferroelectret by knowing its parameters, and therefore optimize the harvester design for specific energy harvesting application.

Keywords: energy harvesting, ferroelectret, electromechanical model

(Some figures may appear in colour only in the online journal)

1. Introduction

A ferroelectret is a thin and flexible porous polymer that can store positive and negative charges in its internal voids after charging. It is then able to convert compressive and bending forces into electrical signals that can be used for both sensing and energy harvesting [1–10]. Our previous study has demonstrated that the output energy from porous polypropylene (PP) ferroelectret is sufficient to power a low-power wireless sensor chipset [10]. When a ferroelectret is used in energy harvesting applications, its output pulses can be used to charge an energy storage device, such as a capacitor, to store the energy that generated from the mechanical deformation. This is similar in principle to piezoelectric energy harvesting using piezo ceramics [11, 12]. However, ferroelectret materials are flexible and therefore more attractive for wearable applications.

When a ferroelectret is used as the sensing material in a sensor, the magnitude of the output voltage and the signal-to-noise ratio are the key design parameters [8, 13]. Previous studies [3, 14–16] have proposed a cellular layer model to predict the voltage output when mechanically compressing

the ferroelectret. This model shows that the piezoelectric charge coefficient d_{33} of a ferroelectret is inversely proportional to its Young's modulus, Y . As the voltage V of an electrical signal from a ferroelectret is directly proportional to its piezoelectric voltage coefficient g_{33} therefore d_{33} , V is inversely proportional to its Young's modulus. Using this model, by knowing the d_{33} and Young's modulus of a ferroelectret, the amplitude of its output voltage can be predicted, which is particularly useful for sensing applications. However, predicting output voltage only is not adequate for energy harvesting applications. Power and energy of the output signal should also be considered.

Most of the previous energy harvesting researches on ferroelectret were focusing on the energy output generated by machinery vibration [4–6], and the vibration was usually at the resonant frequency (more than 100 Hz) of the ferroelectret. In such applications, since the vibrational input excitation is continuous, the output electrical signals can be treated as continuous instead of being individual pulses. Thus the term 'power' was used to describe the amount of the harvested energy [4–6] because energy is simply proportional to power in this case. The cellular layer model [3, 14–16] is still able to

predict the harvester's output from vibration because power can be calculated using voltage V over resistive load R . However, if the energy harvesting application has a low frequency or discontinuous mechanical input with variable amplitude, such as from footstep [10], the output electrical signals are discontinuous pulses. Energy generated by each pulse is a more accurate measurement of the harvester's output than its output power. In this case, the duration t of an electrical pulse is as important as its voltage V since energy is an integral of power over time. The cellular layer model is not adequate as it does not take duration of the electrical signal t into consideration. Therefore, in this work we propose a novel model that includes both the voltage V and the duration t of an electrical signal generated by a ferroelectret, and relates them to the d_{33} as well as Young's modulus. This model allows one to calculate the voltage, power output and energy output of energy harvesters based on cellular polymers. It is useful for describing the performance of ferroelectret energy harvesters and for optimizing their design.

To support our proposed theoretical model we have selected two types of ferroelectrets with different d_{33} and Young's modulus. By applying compressive forces on these materials, their generated electrical signals are measured and used to charge a capacitor. The ferroelectrets we studied are cellular PP and sandwiched porous polytetrafluoroethylene (PTFE) films. PP ferroelectret is one of the most researched [7–9] and was one of the first commercialized ferroelectrets [3, 9, 17]. It usually has d_{33} ranging from 200 to 300 pC N⁻¹ [2, 7, 18], with Young's modulus on the order of 1 MPa [15, 19, 20]. The other ferroelectret in this work is a porous PTFE film sandwiched between two layers of fluorinated ethylene propylene (FEP) films, creating a FEP/PTFE/FEP structure [16, 21]. Its functional void structure that can store charge is in the porous PTFE layer. However, PTFE has open-porous structure so the charge cannot be permanently trapped in the film due to the possible breakdown-induced conductivity during charging, resulting in a poor ferroelectret charging ability and stability [21, 22]. Therefore, the FEP films are used as electrically blocking dielectric layers to seal the pores and confine the charges in the PTFE. The FEP/PTFE/FEP ferroelectret is reported to have high d_{33} after charging, ranging from 300 to 800 pC N⁻¹ [23–25]. It is reported to have an estimated Young's modulus of 0.2–0.3 MPa in the thickness direction [26, 27]. Since these PP and FEP/PTFE/FEP ferroelectrets have large variations in d_{33} and Young's modulus, their generated electrical signals can be compared and used to demonstrate how these parameters are related to the harvested energy using the theoretical model we propose.

2. Experimental details

The PP ferroelectret films were purchased from Emfit Ltd. These commercial samples have been corona charged by the manufacturer. They were sheets in the size of 230 mm × 210 mm, with thickness of 70 μm. These films were further cut into test samples of 60 mm × 70 mm. Ag

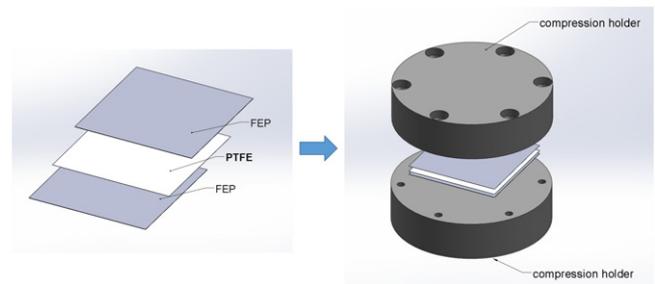


Figure 1. Schematic diagram of compressing two FEP films and one PTFE film between two aluminum disks before the fuse treatment.

electrodes were printed on both sides of the samples using a screen printer (Dek 248, Dek Printing Machines Ltd), using a Ag ink (silver Fabinks TC C40001, Smart Fabric Inks Ltd). The electrode had an area size of 50 mm × 60 mm. The samples after electrode printing were cured in an oven at a temperature of 50 °C for 10 min.

The fabrication of the FEP/PTFE/FEP ferroelectret followed the method described by Zhukov *et al* [16]. It was fabricated by stacking a PTFE film with thickness of 63 μm and porosity of 91% (Goodfellow Cambridge Ltd) between two layers of 12.5 μm thick FEP films (Lohmann Technologies Ltd). These sandwiched films were then placed between two aluminum disks with diameter of 80 mm. Bolts were used to tighten the discs to apply a compressive pressure to the stacked films (figure 1). The compressed films were put in an oven and heated up to 280 °C, then left dwelling at this temperature for half an hour before cooling down. Since the FEP films started to melt at 280 °C, this process enabled the PTFE film to be completely sealed between two FEP layers, resulting a fused sandwich system. The thickness of these fabricated FEP/PTFE/FEP ferroelectrets were measured using a micrometer to be 68 μm.

After the fabrication, a needle to plane corona charge process with a corona voltage of -25 kV and charging time of 60 s was used to charge the FEP/PTFE/FEP. Ag electrodes were printed on both sides of the fabricated FEP/PTFE/FEP sample using the Dek 248 screen printer. The Ag electrodes had an area of 30 cm² on each side of the sample. After screen printing the samples were cured in an oven at a temperature of 50 °C for 10 min.

A precision source/measure unit (B2902A, Keysight Technologies UK Ltd) was used to quasi-statically determine the d_{33} of the ferroelectret samples, and a piezometer system (PM300, Piezotest Pte. Ltd) were used to determine dynamically. The energy output of the ferroelectrets under compressive forces were investigated using an Instron electrodynamic instrument (EletroPuls E1000, Instron Ltd), which can quantify the applied forces and frequency. Trapezoidal function of compressive forces were applied on the sample at a maximum force of 800 N for a duration of 0.5 s, as shown in figure 2. The capacitance of the ferroelectret when compressed was measured using a LCR meter (Wayne Kerr 4300, Wayne Kerr Electronics Ltd). The generated electrical signals were recorded using a digital oscilloscope (TDS2014, Tektronix Ltd).

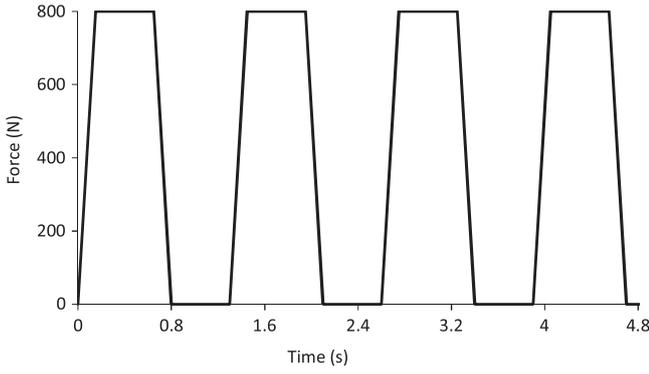


Figure 2. Trapezoidal function of the applied compressive forces.

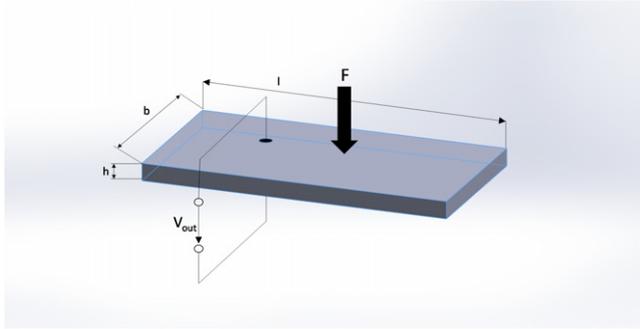


Figure 3. A capacitor model of ferroelectret.

3. Results and discussion

3.1. Electromechanical model

To predict the output energy of a ferroelectret, we propose an electromechanical model that treats the ferroelectret as both a capacitor and a spring–mass–damper system. In this model a ferroelectret can be treated as a capacitor with internal spring that provides restoring force when the applied compressive force is released. Figure 3 is a rectangular model of ferroelectret under a compressive force F , where b is the width, l is the length, h is the thickness, Δh is the change of thickness and V_{out} is the output voltage when compressed. When the ferroelectret is treated as a capacitor containing charge [3, 28], its capacitance C will increase as the thickness reduces when a compressive force is applied. Using $d_{33} = \frac{Q}{F}$ and $Q = VC$, the V_{out} of the ferroelectret can be expressed as

$$V_{out} = \frac{Fd_{33}}{C}, \quad (1)$$

where C is the capacitance of the ferroelectret when compressed. Since $C = \epsilon_{33} \frac{bl}{h - \Delta h}$ in figure 3, equation (1) can be derived into

$$V_{out} = \frac{Fd_{33}(h - \Delta h)}{\epsilon_{33}bl}, \quad (2)$$

where ϵ_{33} is the permittivity of the material. Using equation (2), the V_{out} of a ferroelectret can be related to its dimensional parameters (b , l , h), thickness deformation Δh , applied force F and d_{33} .

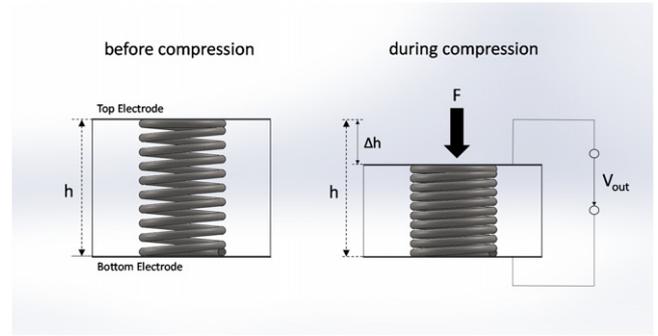


Figure 4. A spring–mass–damper model of a ferroelectret.

Equations (1) and (2) show that the V_{out} of an electrical signal generated by a ferroelectret under compressive force F is directly proportional to its d_{33} . However, since the harvested energy from the generated electrical signal is calculated by $\int \frac{V_{out}^2}{R} dt$ (R is the load resistance), the duration t of the signal remains unknown. To solve this problem we consider the ferroelectret as a spring–mass–damper system, as in figure 4. Using the law of conservation of energy, the energy injected into the system is $\int Fv dt'$, where v is the velocity of compression and t' is the time of the force acting on the system, the resulted kinetic energy in the system is $\frac{1}{2}mv^2$, the resulted elastic energy is $\frac{1}{2}k\Delta h^2$, the resulted electrical energy is $\int \Gamma V_{out} dt$. In this system the damping losses can be neglected due to the small spring constant (Young's modulus) of the system and the frequency of the applied forces is much less than the resonance frequency of the ferroelectret [4, 6], thus

$$\int Fv dt' = \frac{1}{2}mv^2 + \frac{1}{2}k\Delta h^2 + \int \Gamma V_{out} dt, \quad (3)$$

where

$$\int \Gamma V_{out} dt = \frac{1}{2}CV_{out}^2 + \int \frac{V_{out}^2}{R} dt. \quad (4)$$

The generated electrical energy consists of the energy stored in the self-capacitor and the energy delivered to the electrical load. The latter energy is also the harvested electrical energy.

Furthermore, the elastic constant k in this system can be related to the Young's modulus Y . From Hooke's law $F = k\Delta h$ in a spring-damper-mass system and $Y = \frac{\text{stress}}{\text{strain}} = \frac{F/bl}{\Delta h/h}$, thus

$$k = \frac{Ybl}{h} \quad (5)$$

by substituting equations (5) and (4) into equation (3), it can be derived that

$$\int \frac{V_{out}^2}{R} dt = \int Fv dt' - \frac{1}{2}mv^2 - \frac{1}{2} \frac{Ybl\Delta h^2}{h} - \frac{1}{2}CV_{out}^2. \quad (6)$$

Since the output electrical signal is a sinusoidal pulse or a trapezoidal pulse closed to sinusoidal shape [10], V_{out} can be calculated as $V_{\text{out(rms)}} = \frac{1}{\sqrt{2}}V$, where V is the peak output voltage. It can be derived that

$$\int \frac{V_{\text{out}}^2}{R} dt \approx \frac{V^2}{2R}t. \quad (7)$$

In equation (6), the pulse duration $\int Fv dt'$ can be approximated as $F\Delta h$. The resulting kinetic energy $\frac{1}{2}mv^2$ in the system can be neglected due to its small mass, by substituting equation (7) into equation (6), it can be derived that

$$\frac{V^2}{2R}t = F\Delta h - \frac{Ybl\Delta h^2}{2h} - \frac{CV^2}{2}. \quad (8)$$

From the cellular layer model [3, 14–16] and equation (1), the output voltage V_{out} is inversely proportional to Y , thus V_{out} can be expressed as $V_{\text{out}} = \frac{A}{Y}$, where $A = \frac{F\varepsilon_{33}\sigma(1+(S_2/S_1))}{C(1+\varepsilon_{33}(S_2/S_1))^2}$. A is a simplified value depending on the ferroelectret's total thickness S_1 of solid layers and S_2 of gaseous layers, permittivity ε_{33} , charge density σ , capacitance C and applied force F . Therefore, equation (8) can be derived into

$$\frac{A^2}{2R}t + \frac{CA^2}{2} = \left(F\Delta h - \frac{Ybl\Delta h^2}{2h} \right) \times Y^2. \quad (9)$$

Since $Y = \frac{\text{stress}}{\text{strain}} = \frac{F/bl}{\Delta h/h} = \frac{Fh}{bl\Delta h}$, equation (9) can be derived into

$$t = \frac{RF\Delta h \times Y^2}{A^2} - RC. \quad (10)$$

From equation (10), the duration t of the output electrical signal is directly proportional to square of the Young's modulus Y .

To summarize, in our proposed model, the output energy $E = \frac{V^2}{2R}t$ of a ferroelectret can be predicted using equations (1) or (2) to estimate V , and using equation (10) to estimate t . Therefore, using this model, the amount of the output energy generated from a ferroelectret can be related to its d_{33} and Young's modulus.

3.2. Experimental results

The cross-sectional morphology of PP and FEP/PTFE/FEP ferroelectrets is shown by SEM images in figure 5. Since the PP ferroelectret is fabricated by stretching the original polyolefin material in two perpendicular directions in a continuous biaxial orientation process [29, 30], its voids are mostly lens-shape in the tensile direction as in figure 5. Unlike PP, the PTFE film has an open-porous structure. Thus the FEP/PTFE/FEP shows a fiber structure sealed between two solid layers as in figure 5. This difference in microstructure contributes to the different Young's modulus of the PP and FEP/PTFE/FEP ferroelectrets.

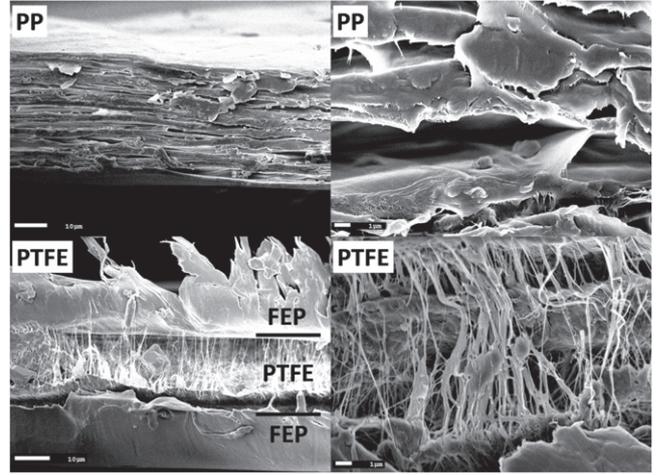


Figure 5. SEM cross-section of PP and FEP/PTFE/FEP ferroelectrets.

Determining the compressive Young's modulus of a thin polymer film is a difficult task. The conventional method of measuring the stress-to-strain ratio cannot be used in this work because the ferroelectret is less than $100 \mu\text{m}$ thick and the thickness of ferroelectret is not uniform, as shown in figure 5. This will cause a large error in the strain measurement. Since the ferroelectret is a quasi-piezoelectric material, an alternative method to obtain its Young's modulus is to measure the dielectric resonance spectra (DRS) [15, 31, 32]. From the spectra a value of anti-resonance frequency can be obtained, which can be used to calculate the Young's modulus. Using the DRS method, previous studies have estimated the Young's modulus of the Emfit's PP ferroelectret used in this work to be on the order of 1 MPa [15, 30]. However, the DRS method cannot be used for the FEP/PTFE/FEP ferroelectret because this Young's modulus calculation only applies to homogeneous samples [15]. The FEP/PTFE/FEP ferroelectret used in this work is a composite film with two types of materials and therefore an estimation proposed by von Seggern and Zhukov [26, 27] has been used. This considers the FEP layers as two solid films and the PTFE layer as one porous film. Using the layered system model the Young's modulus of FEP/PTFE/FEP was estimated to be 0.2 MPa [26].

The d_{33} of the PP and FEP/PTFE/FEP ferroelectrets were experimentally measured using two methods. The 1st method measured the d_{33} quasi-statically. Using the equation $d_{33} = \frac{Q}{F}$, the force $F = mg$ was applied by placing a mass m on the ferroelectret sample's surface (g is the gravity of earth). The charge Q was calculated by measuring the change in the output current I when the force was applied, using the equation $Q = \left| \int I(t) dt \right|$. From this the d_{33} of PP ferroelectret was calculated to be 328 pC N^{-1} , and the FEP/PTFE/FEP ferroelectret 429 pC N^{-1} . The 2nd method used a piezometer to measure the d_{33} dynamically. The piezometer applied a vibrational force of 0.25 N to the sample at 110 Hz, then measured the charge Q to calculate the d_{33} . The d_{33} of PP ferroelectret measured using this method was 295 pC N^{-1} ,

Table 1. Young's modulus and d_{33} of PP and FEP/PTFE/FEP ferroelectrets.

	Y	d_{33}
PP	1 MPa	295–328 pC N ⁻¹
FEP/PTFE/FEP	0.2 MPa	362–429 pC N ⁻¹

and FEP/PTFE/FEP ferroelectret was 362 pC N⁻¹. The quasi-static d_{33} values determined by the 1st method were larger than the dynamic values determined by the 2nd method. This matches the result from the previous studies [15, 33]. The d_{33} values of PP and FEP/PTFE/FEP determined by both methods are in the range reported in the literature [2, 7, 18, 21, 23–25]. These results show that the d_{33} of FEP/PTFE/FEP is between 20% and 30% higher than that of PP.

The Young's modulus and d_{33} of the tested ferroelectrets in this work are summarized in table 1. With these values known, the energy output of a ferroelectret can be predicted using the proposed electromechanical model. Using equation (1) of the model, we calculated the maximum instantaneous output voltage V of a PP ferroelectret sample at applied compressive forces ranging from 100 to 800 N, using the measured d_{33} value of 328 pC N⁻¹, and the capacitances were measured at each applied force. This calculated result was compared with the experimental result, which recorded V_{out} using an oscilloscope, as shown in figure 6. The experimental values fit well with the theoretically calculated values.

From equations (1) and (2), the capacitor model shows that the output voltage V_{out} of a ferroelectret is directly proportional to its d_{33} . Since the d_{33} of FEP/PTFE/FEP is between 20% and 30% higher than that of PP, its V_{out} should also be higher. This implies a higher maximum instantaneous output power $\frac{V^2}{R}$ from FEP/PTFE/FEP. However, from equation (10) in the spring–mass–damper model, since the Young's modulus of FEP/PTFE/FEP was lower than that of PP, the duration t of its output signal should also be lower. From equation (7), because the output energy $E = \frac{V^2}{2R}t$ of a ferroelectret is a product of power $\frac{V^2}{R}$ and time t , it is not necessarily the case that the output energy of FEP/PTFE/FEP will be higher than PP.

To confirm this theoretical prediction, we applied trapezoidal function of compressive forces (as shown in figure 2) to the ferroelectret samples, and used an oscilloscope to record their generated electrical signals. The oscilloscope has an internal resistance of 1 M Ω which is treated as the load resistance of the ferroelectret energy harvester. The generated signal from the ferroelectrets are shown in figure 7. Due to the limitation in the machinery and set up of the sample holder, the holder's surfaces were not completely flat and level. When compressing a soft and thin ferroelectret sample, it was inevitable that some parts of the sample's surface experienced the compressive force earlier than the other, or even slightly larger force. Thus, even though the input mechanical force is trapezoidal function, the output signal is not perfect trapezoidal shape but fluctuating in the beginning as shown in

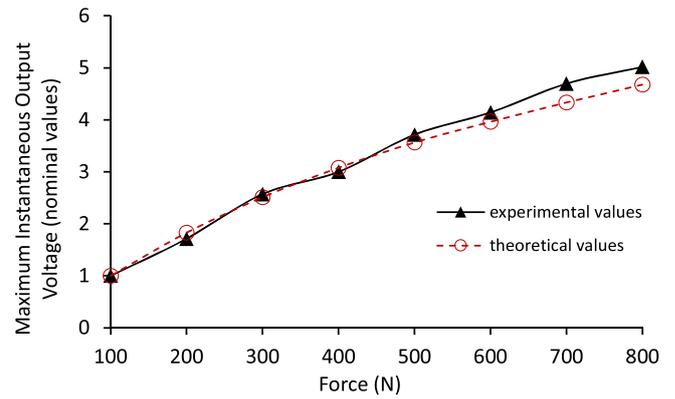
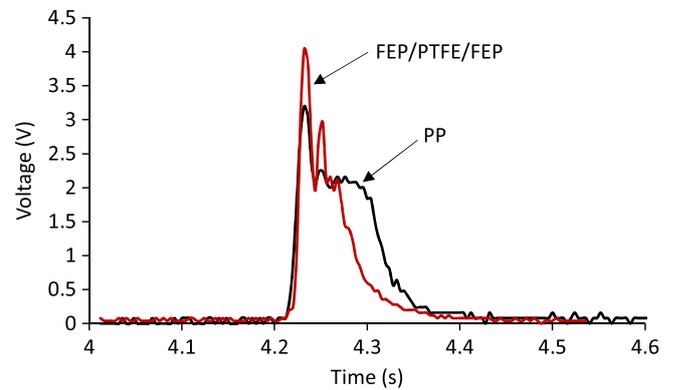
**Figure 6.** Theoretical and experimental values of the V_{out} of a PP ferroelectret at different compressive forces.**Figure 7.** Electrical signals of FEP/PTFE/FEP and PP ferroelectrets at 800 N of compressive force.

figure 7. From figure 7, the maximum V_{out} of the FEP/PTFE/FEP under 800 N of compressive force was 4.1 V, whilst at the same conditions the maximum V_{out} of the PP was 3.2 V. The V_{out} of the FEP/PTFE/FEP was 28.1% higher. This result is close to the theoretical calculation from equation (1), where it predicts that the V_{out} of FEP/PTFE/FEP is 30.8% (d_{33} from 1st method) or 22.7% (d_{33} from 2nd method) higher than that of PP. Figure 7 also shows that the duration t of the generated signal from the FEP/PTFE/FEP was about 0.136 s, whilst the PP was about 0.148 s. The t of the FEP/PTFE/FEP is 8.11% less than the PP. This matches the prediction from equation (10) since FEP/PTFE/FEP has a smaller Young's modulus than PP. Using equation (7), the output energy of FEP/PTFE/FEP was calculated to be 0.393 μ J, and PP is 0.454 μ J. Therefore, despite that the instantaneous power of FEP/PTFE/FEP being higher than PP, its output energy is lower.

The observed characteristics of the generated signals from the FEP/PTFE/FEP and PP ferroelectrets are further supported by the results obtained when using these materials to charge a capacitor. As shown in figure 8, a 2.2 μ F capacitor was charged using a FEP/PTFE/FEP sample and a PP sample respectively, under the same trapezoidal compressive forces. It shows that the PP ferroelectret charged the capacitor at a higher rate at the beginning because it generated more

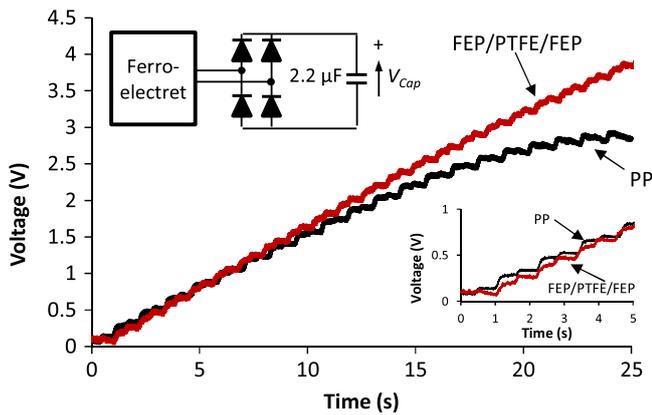


Figure 8. Capacitor voltage of FEP/PTFE/FEP and PP ferroelectrets under the same trapezoidal function of compressive forces, and rectification into a 2.2 μF capacitor.

energy in each step, due to its longer duration t in the generated pulse. However, as the peak voltage of its pulse was around 3 V as shown in figure 7, the capacitor voltage saturated below 3 V. The FEP/PTFE/FEP ferroelectret, on the other hand, had a peak pulse voltage over 4 V, and despite initially charging at a slower rate due to less energy output, it can charge a higher capacitor voltage.

4. Conclusions

In conclusion, we proposed an electromechanical model that can explain the behavior of a ferroelectret when used as an energy harvester. This model treats a ferroelectret as both a capacitor and a spring–mass–damper system. It can relate both the voltage and duration of the output electrical signal from a ferroelectret to its Young's modulus and d_{33} . Therefore, this model can be used to predict the output energy of a ferroelectret by knowing its parameters.

The model has been validated by experimentally comparing the output energy of PP and FEP/PTFE/FEP, which are two types of ferroelectrets with different Young's modulus and d_{33} . The experimental results agree with the theoretical predictions from the proposed model. It shows that a ferroelectret with large d_{33} will generate a high output voltage and thus a high output power. This is favorable for sensing application. However, it is not necessarily advantageous for energy harvesting application since d_{33} is inversely proportional to the ferroelectret's Young's modulus. A ferroelectret with large d_{33} , thus with small Young's modulus will result in a small duration of the output pulse, thus possibly a small output energy as energy is an integral of power over time.

This proposed electromechanical model can be used to optimize the selection of ferroelectret according to the electronic design and application when used as an energy harvester. It is also useful for the design optimization of new ferroelectret materials.

Acknowledgments

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