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An experimental investigation of piezoelectric P(VDF-TrFE) thick film on flexible substrate as energy harvester

Chow Khoon Keat[1,2*], Kok Swee Leong[2], Lau Kok Tee[3]

[1] Department of Electrical Engineering, Politeknik Ungku Omar, Jalan Raja Musa Mahadi, 31400, Ipoh, Perak, Malaysia
[2] Faculty of Electronics and Computer Engineering, Univerisiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia.
[3]Faculty of Manufacturing Engineering, Universiti Teknikal Melaysia Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia.

E-mail: *markchowmy@yahoo.co.uk

Abstract. This paper proposes an experimental inves tigation of energy harvester using poly(vinylidene fluoride-trifluoroethylene) or P(VDF-TrFE) thick-film on flexible substrate by using print screen and rod method. Polyester film being used as the substrate where a sandwiched layer of electrode-piezopolymer-electrode thick film is deposited on. The thick- film is then annealed at 100°C and polarized at 100 V for the film with a thickness of about 18µm, being inspected under EDX, FESEM and XRD. The fabricated energy harvester piezoelectric is able to generate a maximum output power of 4.36 µW at an externa l electrical load of 1 kΩ with a maximum peak-to-peak of about 3.0V when an impact free-fall force of 0.2N was applied on the thick-film.

1. Introduction

In 21st century, energy harvesting applications have become a great interest in wearable technology and flexible electronics. Most of these applications need low power consumption in the range of mW or µW level of power source and currently there are using conventional batteries as their power source. Therefore, many researchers have been conducted for energy harvesting as self-power source of wearable technology and flexible electronics application using piezoelectric material such as piezoceramics and piezopolymers. The piezoceramic materials such as barium titanate ($BaTiO_3$), lead zirconatetitanate (PZT), sodium potassium niobate (KNN) and piezopolymers materials are not suitable for wearable technology because they need to be cured at higher temperature and on rigid substrate. Therefore piezopolymer such as polyvinylidenefluoride or P(VDF) and poly(vinylidene fluoride-trifluoroethelene) or P(VDF-TrFE) are discovered for the purposes of wearable technology. [1-3].

The piezopolymer materials have many advantages as compared to piezoceramic materials such as PZT and its become popular after the discovery of piezoelectricity in polyvinylidene by Kawai in the early 1964 [4]. One major advantages of polymer piezoelectric materials is that it is easily fabricated at lower temperatures compared to ceramic piezoelectric such as KNN and $BaTiO_3$. This property make them attractive for piezoelectric applications on flexible substrate, although there are few known fact that piezopolymers have much lower piezoelectric coefficients compared to piezoceramic. The other advantages are PVDF and PVDF-trifluoroethylene (PVDF-TrFE) piezopolymers with lower piezoelectricity compared to piezoceramics material such as PZT but it can conform to complex structural surfaces due to its flexibility nature [5].

The main disadvantage of the PVDF piezopolymer materials is mechanical stretching, which is not suitable for conventional micro-fabrication processes [6]. As a solution, P(VDF-TrFE) materials is chosen where it consists of crystalline structure and therefore the piezoelectric properties depend on the molecular proportion x of vinyledene fluoride in P(VDFxTrFE1−x). The presence of TrFE element in the copolymer of the PVDF-TrFE film introduces significant features to the PVDF homo-polymer. One of the advantages of PVDF-TrFE is that it increases the tendency to crystallize in the polar β- phase without the requirement of mechanical stretching to transform the non polar α-phase to the polar β-phase as in the case of PVDF, when $0.6 < x < 0.85$ [7].Many studies has been carried out on PVDF- TrFE copolymers, and the copolymer at composition near 75/25 mol.% exhibits the highest ferroelectric responses for energy harvesting application [8].

This paper reports the experimental investigation of energy harvester by using poly(vinylidene fluoride-trifluoroethylene) or P(VDF-TrFE) as thick-film on flexible substrate, the result of screen-printing techniques with rod method for fabrication of flexible thick film by using Field-emission Scanning Electron Microscope and the voltage response to free-fall force on P(VDF- TrFE) thick film was studied for micro energy harvester application.

2. Experiment

2.1 Materials

In this work, Poly(vinylidene fluoride-trifluoroethylene) or P(VDF-TrFE) is prepared with a molecular weight of 350,000 gmol^{-1}[75:25mol%] manufactured by Kureha, Japan.

2.2 Fabrication

P(VDF-TrFE) (75:25mol %) powder were dissolved in *N,N*-dimethylformamide (DMF, Sigma-Aldrich Co.), weight in percentage of 15wt% [9-11]. In order to ensure good adhesion in between active layer or P(VDF-TrFE) was sandwiched between top electrode and bottom electrode on the flexible substrate (Melinex with thickness of 75 μm)by using screen printing and rod method. The powder was dissolved in DMF and mechanically stirred at 100 °C for 1 hour. The solution was then immersed in an ultrasonic bath for 20min to ensure that the solution was fully dissolved.

Prior to the P(VDF-TrFE) deposition, first Ag electrode layer were screen-printed using screen stencil onto the blank Melinex substrate. The second Ag electrode layer was applied after the infra-red light treatment. The thick film was dried under infra-red light for 15 min with a temperature of 60˚C in order to remove the residual solvent. Then, the fabricated thick film were annealed in an oven for 4h at 80°C to increase crystallinity [12]. The thick film was subsequently annealed in an ambient environment oven at 100˚C for 1 hour to obtain a smooth and crystalline surface with reduced porosity.

The P(VDF-TrFE) thick film was electrical polled using high voltage polarizer at 100 V for 20 min at 100°C (near to the Curie temperature of P(VDF-TrFE). This polarization is to align the domain dipole according to the polarity of the high voltage. The dimension of the fabricated energy harvested and electrode layers is shown in table 1 with step of process parameters are summarised in table 2.The fabricated device of the sandwiched P(VDF-TrFE) thick film is illustrated in figure 1 which is being used for the experimental testing in next step.

Table 1. Dimension of fabricated sensor device

Design	**Parameter,** L x W (mm)
P(VDF-TRFE) layer	2.5 x 2.0
Top Electrode	3.4 x 1.0
Bottom Electrode	3.4 x 0.8

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Table 2. Summary of paste properties and printed layer process parameters

Name	Materials	Printing methods	Curing conditions
Top and bottom electrode layer	Silver Conductor Paste	Print screen	Infa Red 80°C,30min
P(VDF-TrFE) layer	Poly(vinyli-dene fluoride trifluoroethylene	Rod Method	Infa Red 80°C, 30 min, at plate annealing 100°C
Melinex layer	Polyester film	-	-

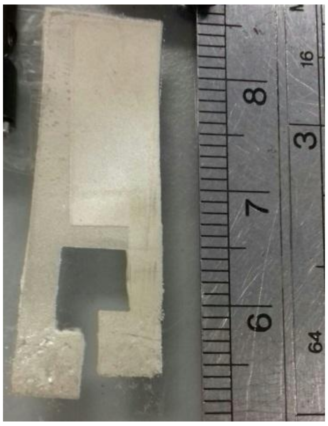

Figure 1. Photograph of a fabricated flexible piezoelectric P(VDF-TrFE) energy harvester

2.3 Experimental setup

A free-fall drop impact test was carried out on the fabricated energy harvester by dropping a known mass of plasticine with varying weights from 0.03 N to 1N (refrains from falling out of the target by using a plastic cylinder as shown in figure 1) at a fixed height of 20cm. Figure shows instantaneous of electrical voltage output voltage from the P(VDF-TrFE) thick film when the plasticine is released to be free-fall onto the P(VDF-TrFE) thick film.

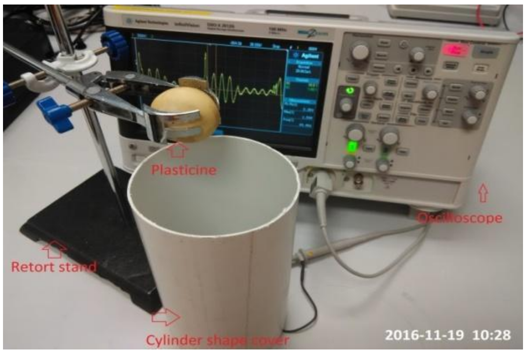

Figure 2. Experiment setup

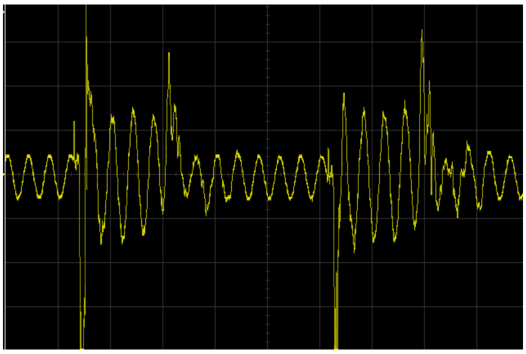

Figure 3. Output AC voltage peak when impact force

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3. Results and discussion

3.1 Microstructure inspection and characterization

The surface microstructures of the deposited layers of P(VDF-TrFE) energy harvester were characterized by Ultra High Resolution Field Emission Scanning Electron Microscopy (FE-SEM, accelerating voltage = 3 kV, Model: Merlin, Carl Zeis AG). Figure 4 (a) and (b) shows FESEM top surface of dried thick film which had layers of the Ag/P(VDF-TrFE)/Ag/Melinex composite structure, while (b) displays the cross-section of P(VDF-TrFE) thick film with thickness about 60µm.

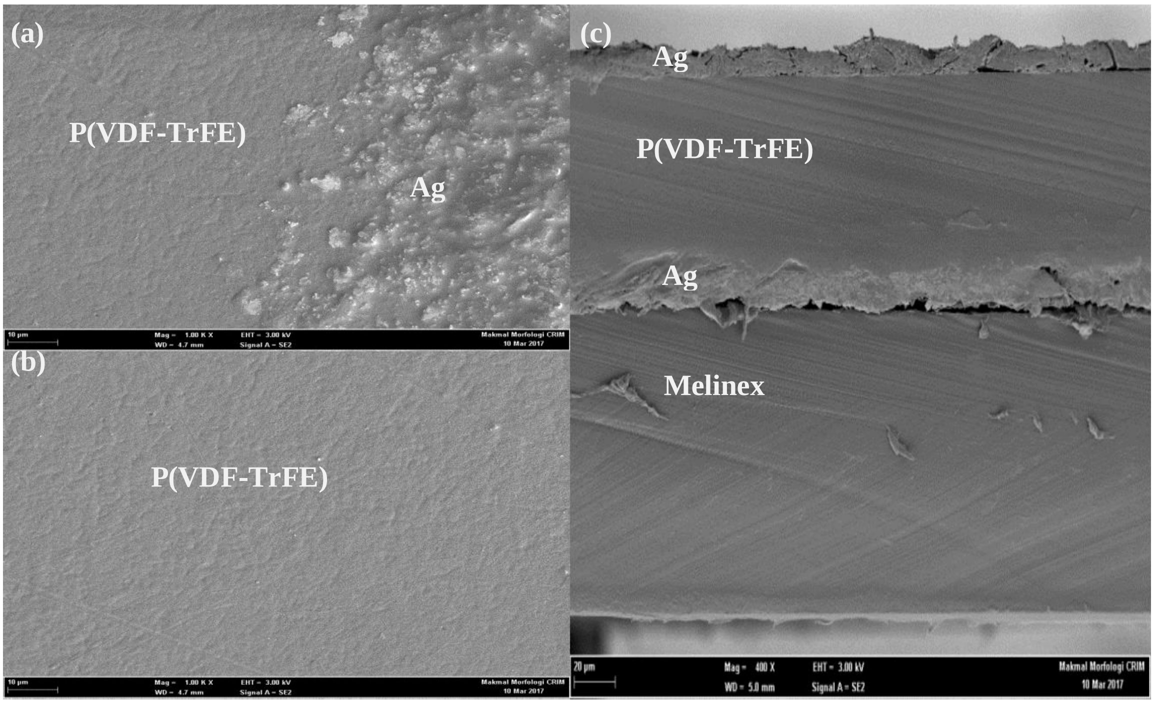

Figure 4. (a)FESEM of the surface layers P(VDF-TrFE)/Ag, (b)FESEM of the surface of a P(VDF-TrFE) thick film dried at 80°C and annealed at 100°C for 15 mins and (c) Cross-section micrograph of electrical harvester structure

3.2 Electrical performance

To verify piezopolymers responses, an impact force test using free-fall plasticine were conducted and the electrical voltage outputs were measured. The peak-to-peak AC voltage versus the free-fall impact force shows a saturation at about 3V when a impact force of 0.2N is applied (see figure 5) A highest output voltage of 3.06V was generated at 0.2N load. When the force exceeding 0.7 N, the average output voltage settled down to 2.70V.

Figure 6 shows the electrical output power of piezopolymers P(VDF-TrFE) thick film when the output terminal of the film is connected to external resistive load varied from 700Ω to 5 kΩ. This was after the impact force was fixed at 0.2N. A maximum output power of 4.36 µW is being measured across an optimum external load of 1kΩ.

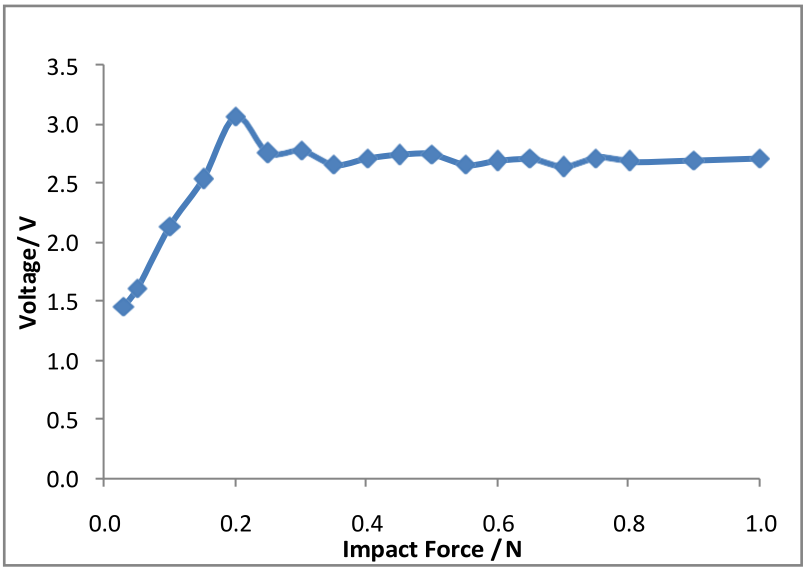

Figure 5. Voltage output vs impact force.

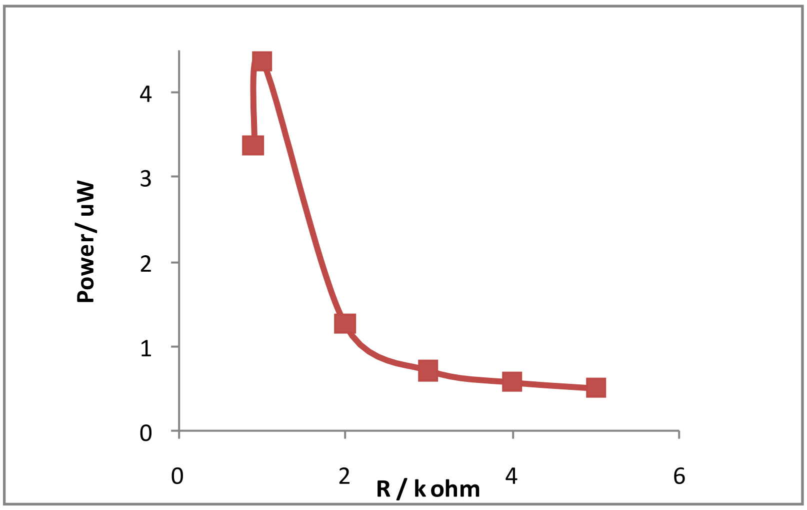

Figure 6. Power vs Resistive load.

4. Conclusion

In this paper, the piezopolymers P(VDF-TrFE) thick film were successfully developed onto flexible substrate as an energy harvester which it provides a promising solution to harvest a consistent electric voltage and power output when exerted a range of free-fall impact force. The output voltage achieved a maximum of about 3V with an impact force of 0.2N which derived a power of 4.36 µW when connected with 1kΩ external resistive load.

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