Fully Printed Unobtrusive and Skin-conformable Piezoelectric Energy Harvester

Karem Lozano Montero, Mika-Matti Laurila, Matti Mäntysalo
Tampere University
Korkeakoulunkatu 3
FI-33720, Tampere, Finland
karem.lozanomontero@tuni.fi

***Abstract*— Flexible piezoelectric energy harvesters have the potential to be used as a power supply in low-power wearable electronic systems. However, the complexity of the fabrication process complicates their integration in these types of systems. This study proposes a simple printing-based fabrication process for a lightweight and skin-conformable piezoelectric energy harvester. First, an interdigitated electrode (IDE) structure is inkjet printed using the conductive polymer ink PEDOT:PSS. The piezoelectric polymer (P(VDF-TrFE)) is then bar-coated on top of the electrodes. The fabricated energy harvester shows a maximum power of 2.4 nW and a power density of 0.5 µW/cm^3. This study introduces a promising fabrication process for the development of flexible piezoelectric energy harvesters for self-powered systems.**

Keywords—printed electronics; P(VDF-TrFE); energy harvester; interdigitated; flexible electronics

I. Introduction

The rapid development of novel flexible electronic devices has increased the interest in their integration in wearable applications, such as healthcare devices, sport monitoring devices, and other portable electronics. This approach requires the development of innovative devices which allow the miniaturization and autonomous operation of these systems. In this regard, it is required to investigate new mechanisms to harvest energy from different sources present in the environment to guarantee an optimal operation of flexible electronic devices. Mechanical energy sources are more accessible compared to other energy sources. For example, mechanical energy can be harvested from body movements, such as running, walking, finger movements, and arm bending [1][2]. Therefore, the conversion of mechanical energy to electrical energy has the potential to be used as a working principle in flexible energy harvesters. So far, this type of energy conversion has been exploited through several techniques [3], including electromagnetic induction [4], piezoelectricity [5][6], triboelectricity [7], and magnetostriction [8]. Among these techniques, piezoelectricity is an attractive mechanism owing to its characteristic mechanical-to-electrical energy conversion capability which allows the fabrication of devices with simple structures that are appropriate for miniaturization [9].

In particular, the research of energy harvesting from biomechanical energy has been focused on biomedical applications that require highly flexible, thin, and lightweight devices to allow unobtrusiveness and skin-conformability of the whole system [10][11]. Hence, it is needed to develop flexible piezoelectric energy harvesters based on biocompatible materials. Previous studies have reported flexible piezoelectric energy harvesters based on the copolymer poly(vinylidenefluoride-trifluoroethylene) (P(VDF-TrFE)) [12]–[14]. P(VDF-TrFE) is characterized by a relatively high piezoelectric coefficient, and its flexibility, biocompatibility, and optical transparency which makes it especially compatible material for biomedical applications. Additionally, this material can be solution-processed using additive fabrication methods, such as highly scalable printed electronics technologies [9][15]–[17], which lead to simplified fabrication process and to reduced fabrication costs [18].

This study demonstrates a fully printed flexible piezoelectric energy harvester. The fabricated device is based on an interdigitated electrodes (IDE) structure. The electrodes are deposited by inkjet printing using the conductive polymeric ink poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS), and the piezoelectric material P(VDF-TrFE) is bar-coated on top of the electrodes. The energy harvesting capabilities of the devices are characterized, and the experimental results are presented to validate the potential use of the fabricated device.

II. Materials and Methods

A. Device fabrication

The fabrication process of the fully printed flexible piezoelectric energy harvester is shown in Fig. 1. Previously, we showed a similar approach to fabricate a piezoelectric sensor [19]. A Parylene-C (GALXYL C Galentis) layer was deposited on a temporary glass carrier by chemical vapor deposition (LabTop 3000, ParaTech Coating). The Parylene-C layer was UV/ozone treated for 15 min to improve its wettability. Then, an IDE structure was inkjet-printed (DMP-2801, Fujifilm Dimatix) using PEDOT:PSS ink (Clevios P Jet 700, Heraeus) using the following printing parameters: a drop spacing of 40 µm, cartridge temperature of 38 °C, and stage at room temperature. The IDE printed structure was then annealed at 130 °C for 15 min in a convection oven. This was followed by the deposition of the piezoelectric material P(VDF-TrFE) (FC Ink P, Arkema Piezotech) using an automatic bar-coater (Motorized Film Applicator CX4, MTV Messtechnik). The P(VDF-TrFE) layer was patterned using a polyimide (DuPont Kapton) mask, and the wet thickness of the printed layer was 200 µm. Then, the samples were annealed at 135 °C for 1 hour in a convection oven.

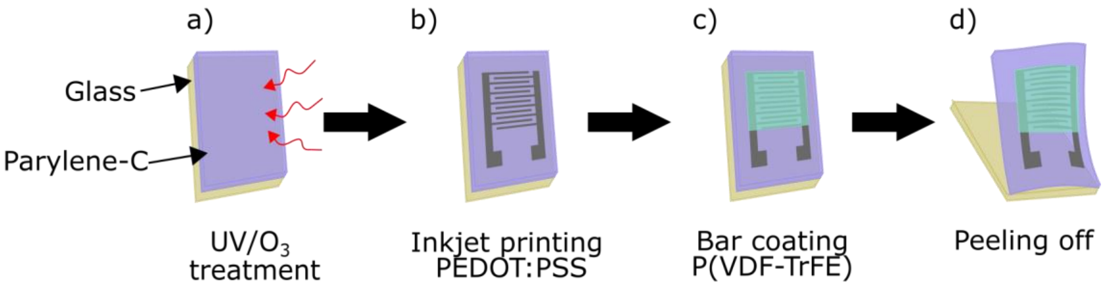

Fig. 1. Schematic of the fabrication process. The substrate is attached to a glass carrier. Before the deposition of the electrodes, the Parylene-C layer is UV/O_3 treated. The piezoelectric material is then bar-coated. Finally, the sensor is peeled off from the carrier.

B. Poling and piezoelectric characterization

The poling process of the piezoelectric material was done simultaneously with polarization electric field hysteresis loop (PE-loop) characterization, which was performed with the ferroelectric characterization tool (aixACCT TF2000, aixACCT Systems GmbH) coupled with a high-voltage amplifier (610C, TREK). The PE-loop was measured by sweeping the electric field up to 100 V/µm. The effective electrode area was calculated based on the IDE dimensions using the method described previously in [19] and this value was used to determine the level of polarization.

C. Experimental setup

Fig. 2 shows the experimental setup used to measure the energy harvesting performance of the fabricated sample. To perform these measurements, the sample was peeled off from the glass carrier. Then, it was laminated to a polyethyleneterephalate (PET) foil (Melinex ST506, DuPont) with a thickness of 125 µm using a tattoo paper adhesive (Silhouette, USA). The sample attached to the PET foil was then placed on a metal holder with a 20 mm distance between the supports (see inset in Fig. 2) such that the harvester was facing downwards and the interdigitated electrodes were in parallel with the bending axis. The bending was induced from the top of the structure using a metal bar. An oscillatory dynamic force of 0.5 N was applied to the sensor using a piezometer (PiezoMeter PM300, Piezotest), and a static force of 0.5 N to press the sample between the sample holder and the metal bar. To measure the output power, various load resistors were connected in parallel with the sample while monitoring the voltage drop over the load resistor with a combination of an in-house built unity gain amplifier (input impedance of approximately 30 GΩ) and a digital oscilloscope (DSOX2002A, Keysight). The recorded voltage signal was analyzed using a Matlab script by fitting a sinusoidal waveform to it and using the fitted sinusoidal to determine the voltage amplitude.

D. Finite element model for bending radius estimation

Finite element modelling software (COMSOL) was used to estimate the deflection of the sensor during the bending test. The relevant material parameters for PET, Parylene-C and P(VDF-TrFE) are given in TABLE I, while the compliance matrix of P(VDF-TrFE) was taken from [20]. To avoid high aspect ratio layers and to reduce the computation time, the PEDOT:PSS electrodes were modelled as a zero-thickness material layer, and its effect on the bending was thereby neglected (reasonable assumption due to low electrode thickness of ~25 nm). A high-resolution mesh was generated for the P(VDF-TrFE) – Parylene-C interface and swept through the respective material layers, while a lower resolution mesh was used for the PET layer to further reduce computation time.

TABLE I. DEVICE MATERIALS PARAMETERS

Material	Poisson's ratio	Young's modulus (Pa)	Density (kg/m^3)
PET	0.37	$2 \cdot 10^9$	1380
Parylene	0.40	$2.758 \cdot 10^9$	1289
P(VDF-TrFE)	[20]	[20]	1676

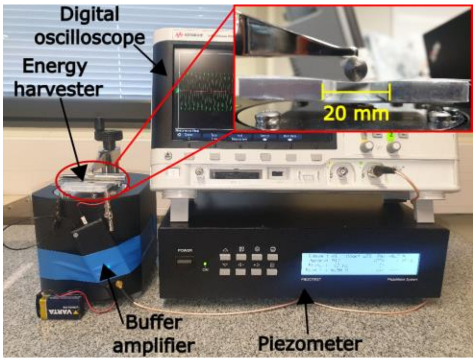

Fig. 2. The experimental setup for the electrical characterization of the sample. The inset image shows the sample placed on the metal holder.

III. RESULTS AND DISCUSSION

A. Sample characterization

The fabricated sample is shown in Fig. 3. The surface profile of the P(VDF-TrFE) layer is shown in Fig. 4. The overall thickness of the device is approximately 21 µm. The remanent polarization (P_r) of 6.4 µC/cm^2 was measured from the PE hysteresis loop shown in Fig. 5, which was obtained for a poling voltage of 100 V/µm. This parameter determines the polarization when the applied electric field is zero. The coercive field (E_c) was approximately 61 V/µm, which is determined by the intersection of the loop with the x-axis. These results compare well with previously reported literature values [14][21].

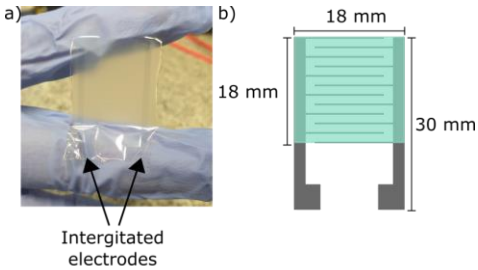

Fig. 3. a) Picture of the flexible piezoelectric energy harvester after peeling off the sample from the glass carrier. b) Energy harvester dimensions.

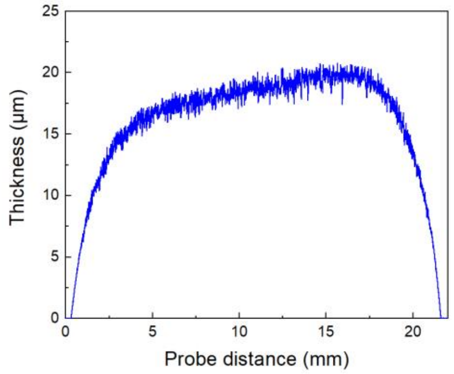

Fig. 4. Surface profile picture of the P(VDF-TrFE) layer.

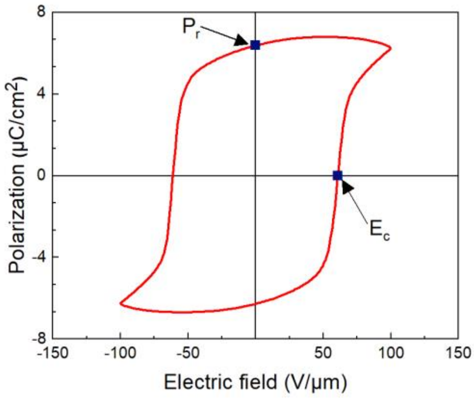

Fig. 5. PE hysteresis loop of the characterized sample.

B. Energy harvesting

As the energy harvesting performance of the device is related to the material deformation during the bending, the displacement of the sample was estimated using COMSOL software. A maximum deflection of ~3 mm was obtained at the middle of the sensing area corresponding to a bending radius of ~ 16 mm. The simulation results are shown in Fig. 6.

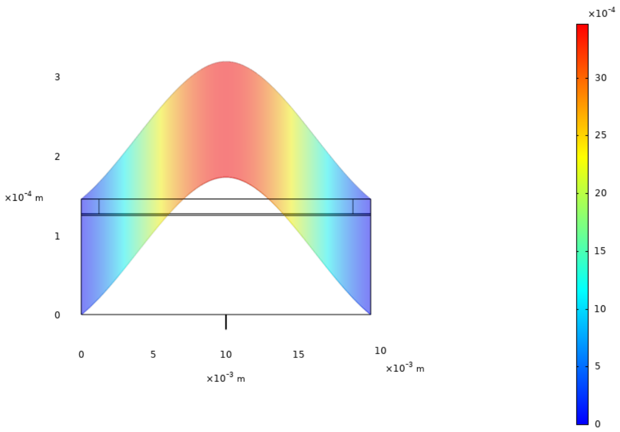

Fig. 6. Simulation of the sensor displacement.

Fig. 7a. shows the frequency response of the flexible energy harvester without resistive load. The maximum output voltage of 1.1 V was observed at 60 Hz, and this was selected as the frequency to characterize the output power of the device. The output power of a piezoelectric energy harvester depends on the external load (R_L) that is connected to it. This parameter can be defined as root mean square (RMS) power $P_{RMS}=V_p^2/2R_L$, where V_p is the peak amplitude of the output voltage. Thus, the output voltage was measured as a function of the external load. The measured output voltage and current are shown in Fig. 7b. The effective power of the fabricated samples was then calculated based on these results. Initially, the output voltage increases as a function of the load resistance, but starts to saturate when the load resistance is 1 GΩ; on the other hand, the current decreases when increasing the load resistance reaching a value close to zero at 1 GΩ. In this range, the output voltage increases from 1 mV to 83.6 mV while the output current decreases from 13.6 nA to 0.2 nA. The output power was calculated using these measured values obtaining a maximum value of 2.4 nW (see Fig. 7c) for a load of 100 MΩ, which corresponds to a power density of approximately 0.5 µW/cm^3 (see Fig. 7d). The obtained power output is relatively small when compared to previously reported studies of piezoelectric energy harvesters based on P(VDF-TrFE) [6][22]. This result can be associated with the high resistance of the thin (t ≈ 25 nm) PEDOT:PSS IDE electrodes and the relatively small bending radius of the sample under test.

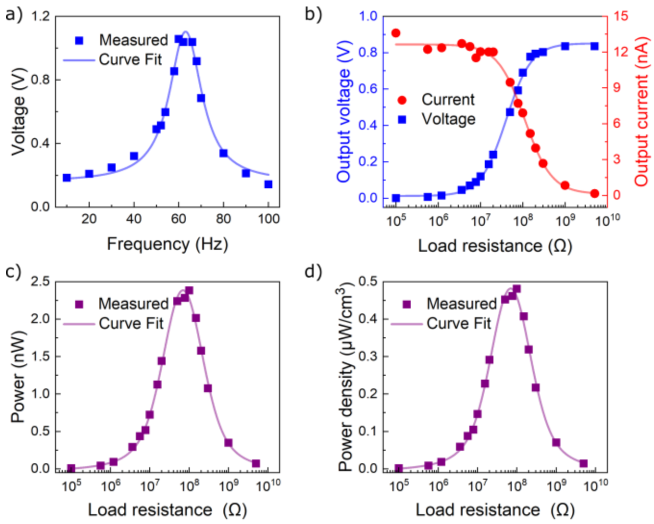

Fig. 7. a) Output voltage variation for the frequency range from 10 Hz to 100 Hz. b) The load voltage and current for external load resistance from 100 kΩ to 5 GΩ. c) Output power and b) power density for external loads from 100 kΩ to 5 GΩ.

IV. Conclusion

This study presented the fabrication and characterization of a fully printed flexible piezoelectric energy harvester based on P(VDF-TrFE) electroactive material. The process developed in this research reduces the fabrication complexity by reducing the number of fabrication steps. The obtained power output can be enhanced by decreasing the internal impedance of the device. The results show the potential use of these types of devices for powering low-power or energy autonomous wearable devices.

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REFERENCES

[1] J. Lim *et al.*, "All-inkjet-printed flexible piezoelectric generator made of solvent evaporation assisted BaTiO3 hybrid material," *Nano Energy*, vol. 41, pp. 337–343, 2017.

[2] M. T. Todaro *et al.*, "Biocompatible, Flexible, and Compliant Energy Harvesters Based on Piezoelectric Thin Films," *IEEE Trans. Nanotechnol.*, vol. 17, no. 2, pp. 220–230, 2018.

[3] D. Wang, G. Yuan, G. Hao, and Y. Wang, "All-inorganic flexible piezoelectric energy harvester enabled by two-dimensional mica," *Nano Energy*, vol. 43, pp. 351–358, 2018.

[4] D.-A. Wang and K.-H. Chang, "Electromagnetic energy harvesting from flow induced vibration," *Microelectronics J.*, vol. 41, no. 6, pp. 356–364, 2010.

[5] S. Y. Chung *et al.*, "All-solution-processed flexible thin film piezoelectric nanogenerator," *Adv. Mater.*, vol. 24, no. 45, pp. 6022–6027, 2012.

[6] Z. Pi, J. Zhang, C. Wen, Z. Zhang, and D. Wu, "Flexible piezoelectric nanogenerator made of poly(vinylidenefluoride-co-trifluoroethylene) (PVDF-TrFE) thin film," *Nano Energy*, vol. 7, pp. 33–41, 2014.

[7] Z. Wang, L. Lin, S. Niu, and Y. Zi, *Triboelectric Nanogenerators*. 2016.

[8] L. Wang and F. G. Yuan, "Vibration energy harvesting by magnetostrictive material," *Smart Mater. Struct.*, vol. 17, no. 4, p. 45009, 2008.

[9] A. Toprak and O. Tigli, "MEMS Scale PVDF-TrFE-Based Piezoelectric Energy Harvesters," *J. Microelectromechanical Syst.*, vol. 24, no. 6, pp. 1989–1997, 2015.

[10] C. K. Jeong *et al.*, "Self-powered fully-flexible light-emitting system enabled by flexible energy harvester," *Energy Environ. Sci.*, vol. 7, no. 12, pp. 4035–4043, 2014.

[11] M. M. Laurila *et al.*, "A fully printed ultra-thin charge amplifier for on-skin biosignal measurements," *IEEE J. Electron Devices Soc.*, vol. 7, pp. 566–574, 2019.

[12] J.-H. Lee, K. Y. Lee, B. Kumar, N. T. Tien, N.-E. Lee, and S.-W. Kim, "Highly sensitive stretchable transparent piezoelectric nanogenerators," *Energy Environ. Sci.*, vol. 6, no. 1, pp. 169–175, 2013.

[13] Y. Cho *et al.*, "Enhanced energy harvesting based on surface morphology engineering of P(VDF-TrFE) film," *Nano Energy*, vol. 16, pp. 524–532, 2015.

[14] S. S. Chauhan, U. M. Bhatt, P. Gautam, S. Thote, M. M. Joglekar, and S. K. Manhas, "Fabrication and modeling of β-phase PVDF-TrFE based flexible piezoelectric energy harvester," *Sensors Actuators A Phys.*, vol. 304, p. 111879, 2020.

[15] S. Magdassi, *The chemistry of inkjet inks*. World Scientific, 2009.

[16] C. K. McGinn *et al.*, "Formulation, printing, and poling method for piezoelectric films based on PVDF–TrFE," *J. Appl. Phys.*, vol. 128, no. 22, p. 225304, Dec. 2020.

[17] M. . Laurila *et al.*, "Evaluation of printed P(VDF-TrFE) pressure sensor signal quality in arterial pulse wave measurement," *IEEE Sens. J.*, p. 1, 2019.

[18] H. Wu, Y. Huang, F. Xu, Y. Duan, and Z. Yin, "Energy Harvesters for Wearable and Stretchable Electronics: From Flexibility to Stretchability," *Adv. Mater.*, vol. 28, no. 45, pp. 9881–9919, Dec. 2016.

[19] K. Lozano M., M.-M. Laurila, and M. Mäntysalo, "All Printed Flexible Piezoelectric Pressure Sensor with Interdigitated Electrodes," in *2020 IEEE 8th Electronics System-Integration Technology Conference (ESTC)*, 2020, pp. 1–6.

[20] H. Wang, Q. Zhang, L. Cross, and A. Sykes, "Piezoelectric, dielectric, and elastic properties of poly(vinylidene fluoride/trifluoroethylene)," *J. Appl. Phys.*, vol. 74, pp. 3394–3398, Oct. 1993.

[21] T. Furukawa, "Structure and functional properties of ferroelectric polymers," *Adv. Colloid Interface Sci.*, vol. 71–72, pp. 183–208, 1997.

[22] P. Schäffner *et al.*, "Microstructured single-layer electrodes embedded in P(VDF-TrFE) for flexible and self-powered direction-sensitive strain sensors," *Smart Mater. Struct.*, vol. 29, no. 8, p. 85040, 2020.