Conformal piezoelectric energy harvesting and storage from motions of the heart, lung, and diaphragm

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Here, we report advanced materials and devices that enable high-efficiency mechanical-to-electrical energy conversion from the natural contractile and relaxation motions of the heart, lung, and diaphragm, demonstrated in several different animal models, each of which has organs with sizes that approach human scales. A cointegrated collection of such energy-harvesting elements with rectifiers and microbatteries provides an entire flexible system, capable of viable integration with the beating heart via medical sutures and operation with efficiencies of ~2%. Additional experiments, computational models, and results in multilayer configurations capture the key behaviors, illuminate essential design aspects, and offer sufficient power outputs for operation of pacemakers, with or without battery assist.

biomedical implants | flexible electronics | transfer printing | wearable electronics | heterogeneous integration

Nearly all classes of active wearable and implantable biomedical devices rely on some form of battery power for operation. Heart rate monitors, pacemakers, implantable cardioverter-defibrillators, and neural stimulators together represent a broad subset of bioelectronic devices that provide continuous diagnostics and therapy in this mode. Although advances in battery technology have led to substantial reductions in overall sizes and increases in storage capacities, operational lifetimes remain limited, rarely exceeding a few days for wearable devices and a few years for implants. Surgical procedures to replace the depleted batteries of implantable devices are thus essential, exposing patients to health risks, heightened morbidity, and even potential mortality. The health burden and costs are substantial, and thus motivate efforts to eliminate batteries altogether, or to extend their lifetimes in a significant way.

Investigations into energy-harvesting strategies to replace batteries demonstrate several unusual ways to extract power from chemical, mechanical, electrical, and thermal processes in the human body (1, 2). Examples include use of glucose oxidation (3), electric potentials of the inner ear (4), mechanical movements of the human body in a manner that does not induce adverse side effects. An ideal device construct would enable power harvesting throughout the macroscale displacement cycles associated with natural motions of an organ, but in a manner that does not induce any significant constraints on those motions. Development of flexible devices based on arrays of piezoelectric ZnO nanowires (14, 15) represents an important step in this direction. Experiments performed with a linear motor to periodically deform the device indicate electrical outputs as large as 1–2 V (open-circuit voltage) and ~100 nA (short-circuit current) (13). Initial in vivo tests on rabbit hearts yielded voltages and currents of ~1 mV and ~1 pA, respectively. The associated electrical power is substantially less than that required for operation of existing classes of implants, such as pacemakers. Some improvement in performance is possible with thin-film geometries, as demonstrated in bending experiments on devices based on barium titanate (16) and lead zirconate titanate (PZT) (11, 12, 17). In vivo evaluations are needed, however, to assess the suitability for realistic use and potential for producing practical levels of power.

Significance

Heart rate monitors, pacemakers, cardioverter-defibrillators, and neural stimulators constitute broad classes of electronic implants that rely on battery power for operation. Means for harvesting power directly from natural processes of the body represent attractive alternatives for these and future types of biomedical devices. Here we demonstrate a complete, flexible, and integrated system that is capable of harvesting and storing energy from the natural contractile and relaxation motions of the heart, lung, and diaphragm at levels that meet requirements for practical applications. Systematic experimental evaluations in large animal models and quantitatively accurate computational models reveal the fundamental modes of operation and establish routes for further improvements.


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The present work reports several advances in the materials science and engineering of biocompatible, flexible, energy harvesters. The key findings include in vivo demonstrations of PZT-based devices (i) with output open-circuit voltages and short-circuit currents that are greater by three and five orders of magnitude, respectively, than previous in vivo results; (ii) monolithically integrated with rectifiers and millimeter-scale batteries for simultaneous power generation and storage, including high-power, multilayer designs; (iii) in biocompatible forms, evaluated through cell cultures and large-scale, live animal models, on various locations/orientations on different internal organs, each approaching thereby minimizing the risks of failure or immune response. A compatible materials isolates them from bodily fluids and tissue, Appendix consists of 12 groups of 10 such structures that are electrically electrodes. See bottom (Ti/Pt, 20 nm/300 nm) and top (Cr/Au, 10 nm/200 nm) (43x600) through constitutive models of piezoelectric behavior. The voltage (43x657) power generation at a level necessary for continuous operation of a cardiac pacemaker; (ii) mechanically and electrically stable device behavior over 20 million cycles of bending/unbending in a moist, hydrogel environment; and (iii) quantitatively accurate analytical models that couple mechanical deformations and piezoelectric effects to predict electrical output as a function of key design parameters and materials properties, including multilayer configurations.

Results and Discussion

Fig. 1A and SI Appendix, Figs. S1 and S2 provide schematic diagrams of a flexible PZT mechanical energy harvester (MEH). An optical microscope image and a photograph appear in Fig. B and C, respectively. The key functional element is a capacitor-type structure that consists of a layer of PZT (500 nm) between bottom (Ti/Pt, 20 nm/300 nm) and top (Cr/Au, 10 nm/200 nm) electrodes. See SI Appendix for details. An MEH module consists of 12 groups of 10 such structures that are electrically connected in parallel. Each of the 12 groups is connected in series to its neighboring group to increase the output voltage (SI Appendix, Fig. S2). Encapsulation of these devices with biocompatible materials isolates them from bodily fluids and tissue, thus minimizing the risks of failure or immune response. A thin spin-cast layer of polyimide (PI) (SI Appendix) serves effectively in this role, as demonstrated in leakage tests performed in PBS (P-5368, pH 7.4; SIGMA) in a way that maintains excellent mechanical flexibility. The computed bending stiffness (per unit width) is 0.22 N·mm for regions co- (43x676) illustrated with a top view (43x676) and electrically stable device behavior over 20 million cycles of bending/unbending in a moist, hydrogel environment; and (iii) quantitatively accurate analytical models that couple mechanical deformations and piezoelectric effects to predict electrical output as a function of key design parameters and materials properties, including multilayer configurations.

As a screen of the biocompatibility and absence of cytotoxicity of the constituent materials, we examined the adherence, growth, and viability of rat smooth muscle cells (SMCs), as a representative tissue cell, on the encapsulated PZT capacitor structures described above. SMCs readily adhere to fibronectin-coated structures, with evident spreading (Fig. 1D), and intact detectable cytoskeletal structures, i.e., Vinculin focal contacts via fluorescence (green in Fig. 1D) and cytoskeletal actin microfilaments (red in Fig. 1E). The scanning electron microscope (SEM) image shows spreading cells in Fig. 1F. No detectable cytotoxicity was observed in a live/dead assay that identifies calcine acetoxymerthoxy derivative (green) for viable cells and ethidium bromide (red) for damaged cells, suggesting that the majority of SMCs are healthy (green in Fig. 1D). In fact, more than 96% of cells were viable after 9 d of culture. Cells grown on device structures showed no differences from those grown on standard tissue culture plates at days 3 and 9 (Fig. 1G).

Measurements with a bending stage reveal dynamic mechanical properties, as shown in Fig. 2A. Fig. 2B illustrates 3D deformations of the PZT ribbons, with distributions of strain obtained by finite element analysis (FEA). The overall shapes match those observed in Fig. 2A. The strain distributions in SI Appendix, Figs. S3 and S4 quantitatively capture the nature of deformations in the various device layers. The results determine the electrical field and electrical displacement in the PZT through constitutive models of piezoelectric behavior. The voltage and current outputs of a representative device before (Fig. 2C) and after (SI Appendix, Fig. S5) rectification illustrate the nature of operation. See SI Appendix for details.

Experimental behaviors can be captured with an analytical model. Compression of a device with length L leads to buckling with an out-of-plane displacement w = A[1 + cos(2πx/L)]/2 as shown in SI Appendix, Fig. S6A, where the origin of coordinate x1 is at the center of the device, and the amplitude A is related to the compression ΔL between the two ends by A = (2/π)√L·ΔL. The PZT layers, together with the top and bottom electrodes, bend with the buckled substrate. The membrane strain in the PZT ribbons is given analytically by

\[ E_m = 4\pi E_T \frac{\Delta L}{L} \]

(see SI Appendix for details), where \( E_T \) and \( E_p \) are the bending stiffness (per unit width) of the PI with and without the PZT and electrodes, respectively, and h is the distance from the center of the PZT layers to the neutral mechanical plane of the cross-section as shown in SI Appendix, Fig. S6B. The bending strain in the PZT ribbons has opposite signs above and below the

![Fig. 1. Flexible MEH based on thin ribbons of PZT, and tests of biocompatibility using rat smooth muscle cells. (A) Exploded-view schematic illustration with a top view (Inset). (B) Optical microscope image of PZT ribbons printed onto a thin film of PI. (C) Photograph of a flexible PZT MEH with cable for external connection. ACF, anisotropic conductive film. (D) Live/ dead viability assay showing live cells (green) with intact nucleus (blue) and one dead cell (red) as indicated by the arrow (Inset). This image corresponds to day 9 of the experiment. (E) Fluorescent image showing Vinculin focal contact points (green), actin filaments (red), and nuclei (blue). (F) SEM image of cells on PZT ribbons encapsulated by a layer of PI. (G) Lactate dehydrogenase assay shows no indications of toxicity for cells on a membrane of PZT at day 9. (*P value < 0.05 between the two groups.) Error bar is calculated standard error.]
midpoint through the thickness of the PZT, and therefore does not contribute to the voltage or current output. For thin PZT layers, the bending strain is much smaller than the membrane strain; the total strain is therefore dominated by the membrane strain. See SI Appendix for details.

PZT is transversely isotropic with the polarization direction $x_3$ normal to the surface. The elastic, piezoelectric, and dielectric constants are denoted by $c_{ij}$, $d_{ij}$, and $k_{ij}$ respectively. For planestrain deformation ($e_{22} = 0$) the strain $e_{33}$ and the electrical field $E_3$ along the polarization direction $x_3$ satisfy the constitutive relations $0 = c_{11}e_{11} + e_{33}E_3$ and $D_3 = e_{31}E_1 + e_{33}E_3 + k_{33}E_3$, where the electrical displacement $D_3$ along the polarization direction is a constant to be determined. For measurements of current, the top and bottom electrodes connect to an ammeter, which has negligible electrical resistance. As a result, voltage between the top and bottom electrodes is zero, and $D_3 = \varepsilon_m$, where the effective piezoelectric constant is $\varepsilon = e_{31} = \frac{k_{33}}{e_{33}}e_{33}$ and the membrane strain $\varepsilon_m$ is given in Eq. 1. For each group of devices in series, the charge induced by $\varepsilon_m$ gives the current $I = -A_{PZT}\varepsilon_m$ (SI Appendix, Fig. S6c), i.e.,

$$I = (-\varepsilon)A_{PZT}\frac{d\varepsilon_m}{dt},$$

where $A_{PZT}$ is total area of the PZT ribbons in each group. In experiments, the compression $\Delta L$ between the two ends of the device is a periodic function of time $t$, given by

$$\Delta L = \begin{cases} \frac{\Delta L_{max}}{4} \left(1 - \cos \left(\frac{\pi t}{T_1}\right)\right)^2, & 0 \leq t < T_1 \\ \frac{\Delta L_{max}}{4} \left(1 - \cos \left(\frac{\pi(t - 2T_1 - T_2)}{T_1}\right)\right)^2, & T_1 \leq t < T_1 + T_2 \\ \frac{\Delta L_{max}}{4} \left(1 - \cos \left(\frac{\pi(t - 2T_1 + 2T_2)}{T_1}\right)\right)^2, & T_1 + T_2 \leq t < (T_1 + T_2) \end{cases}$$

in the first period, where $\Delta L_{max}$ is the maximum compression, and $T = 2(T_1 + T_2)$ is the period. Fig. 2C shows $\Delta V$ vs. $t$ for $T_1 = 0.8$ s, $T_2 = 1.3$ s, and $\Delta L_{max} = 1.5, 3, 5$, and 10 mm as in the experiments. Fig. 2C shows that the current obtained from Eq. 2 for these four values of $\Delta L_{max}$ agrees reasonably well with the experiments, where $L = 2.5$ cm, $E_{PZT}/E_{PZT,\text{comp}} = 0.45$, $h = 24.7$ μm, and $A_{PZT} = 2.24$ mm$^2$ as in the experiments (see SI Appendix for details), and $\varepsilon = 10\text{Coulomb/m}^2$, which is on the same order of magnitude as the literature values (18). The peak current ranges from 0.06 to 0.15 μA for $\Delta L_{max}$ from 1.5 to 10 mm, respectively.

In measurements of potential, the voltage drop across each group is $V/N$, where $V$ is the total voltage for $N$ groups of devices in series (SI Appendix, Fig. S6c). The electrical displacement is $D_3 = \varepsilon_m + \frac{K}{(Nt_{PZT})}$, where $K = e_{33} + (e_{31}/e_{33})(e_{33})$ is the effective dielectric constant and $t_{PZT}$ is the thickness of PZT ribs. The current $I = -A_{PZT}D_3$ is related to the voltage $V$ and the resistance $R$ of the voltmeter by $I = VR$, which gives $V = \frac{R}{R_k} - A_{PZT}D_3$, i.e.,

$$dV/dt = \frac{N_{PZT}}{A_{PZT}R_k}V = -\frac{N_{t_{PZT}}}{K}d\varepsilon_m/dt$$

For the initial condition $V(t = 0) = 0$, the voltage is given by

$$V = \frac{(-\varepsilon)N_{t_{PZT}}}{K} \frac{\tau_{PZT}}{c_{PZT}} \int \frac{d\varepsilon_m}{dt} \frac{\tau_{PZT}}{c_{PZT}} \, dt$$

For $R = 60 \times 10^6 ohms$ in the experiment and $K = 4 \times 10^{-8} C/V m$ (18), Fig. 2C shows the voltage $V$ vs. time $t$ obtained from Eq. 5, which agrees well with the experiments, including both the shape and peak value. The peak voltage can reach values as large as 3.7 V for the maximum compression $\Delta L_{max} = 10$ mm. (The differences between the measured and predicted behavior of the output current in Fig. 2C arise because, according to Eqs. 1 and 2, the current is directly proportional to the rate of compression $d\Delta L/dt$, and is therefore sensitive to $d\Delta L/dt$ when the compression reverses direction. The assumed displacement profile does not follow precisely the one in the experiment. By contrast, the voltage is relatively insensitive to $d\Delta L/dt$ because Eqs. 1 and 5 involve the integration of $d\Delta L/dt$.)

Fig. 2D shows that the currents and voltages increase with frequency, even with the same load amplitudes, consistent with predicted dependence (Eqs. 2, 4, and 5) on the strain rate $d\varepsilon_m/dt$. This behavior can be understood by recognizing that increasing the frequency with the same load amplitude increases the amount of work performed by the external force; as a result, the...
harvester produces enhanced output energy through increases in current and voltage.

This energy can be captured directly by use of a chip-scale rechargeable battery (EnertChips CBCi012; Cymbet Corporation) and a Schottky bridge rectifier (MB12S; Micro Commercial Components) co-integrated on the same flexible substrate with the MEHs. Characterization can be performed using the setups shown in Fig. 2A. Fig. 2E and F show the device and the output voltage of the battery measured in such a case; the results reveal stepwise increases in the stored energy with each cycle of bending and unbinding (Fig. 2A). As the process continues, the voltage of the battery saturates at a value (~3.8 V) characteristic of the battery specification.

The analytical framework can be modified to account for the rectifier via its resistance \( R_{\text{rectifier}} \), as illustrated in SI Appendix, Fig. S6D. Here, the resistance in the current measurement is \( R_{\text{rectifier}} \) instead of 0, whereas the resistance \( R \) in the voltage measurement is replaced by \( R + R_{\text{rectifier}} \). The voltage in Eq. 5 is replaced by its absolute value. See SI Appendix for details. The results (SI Appendix, Fig. S5) agree well with experiments. The amplitudes of the battery specification.

Additional in vitro characterization illustrates that these MEH devices can be wrapped onto various curved objects such as balloons, fingers, and wrists, as seen in SI Appendix, Fig. S7. Control experiments (SI Appendix, Fig. S8) reveal that reversing the electrical connections to measurement equipment reverses the signal polarity, as expected. The devices exhibit excellent fatigue properties. Bending and releasing more than 20 million times while in direct contact with a transparent layer of gelatin (Knox), to mimic a moist environment (SI Appendix, Fig. S9 A and B), induce no noticeable degradation in the properties, as in SI Appendix, Fig. S9 C and D.

In vivo testing involved affixing the devices to epicardial sites on the right ventricle (RV), left ventricle (LV) base, and free wall of bovine and ovine hearts as shown in Fig. 3A and Movies S1 and S2. The anchoring scheme used sutures at three points, to maintain focal contact, although without rigid attachment so as to minimize any alteration or constraint on cardiac motion. Similar suturing techniques have been previously used to suture 3D patches onto the LV in a rat model (20). Furthermore, alternative suturing techniques, including running locked stitch and five-point lacerations can further reinforce the attachments. No detectable change in cardiac conduction or epicardial motion occurs following this simple procedure for fixing the device onto the epicardium. The PZT MEH maintains conformal contact, without delamination from the heart during the entire cycle of cardiac motion from contraction to relaxation. Evaluation of various mounting sites identified optimal locations for power extraction (Fig. 3 A and B). Analysis of voltage outputs from devices placed on the RV, LV base, and free wall appear in Fig. 3 C, D, and E, respectively. The RV yields the best results, even though the LV has more muscular fibers (9–11 mm thick) than the RV (3.5–5 mm thick) (21). The differences in shape and other functional characteristics are, however, most important. The RV chamber is box- or wedge-shaped in form, with a concave free wall, which is thinner than the LV and is attached to the convex interventricular septum (22). The LV is roughly cylindrical in shape and has a thick wall structure with three spiraling layers of muscle to enable contractions with a twisting or torsional motion (23, 24). The RV ejects blood primarily by shortening its free

![Fig. 3. In vivo evaluation of the optimal placement and orientation of PZT MEHs on the heart, and assessment of voltage output by varying the heart rate via dobutamine infusion and use of a temporary pacemaker. (A) Photograph of PZT MEHs on the RV, LV, and free wall of a bovine heart. (B) PZT MEH co-integrated with a rectifier and rechargeable battery, mounted on the RV of a bovine heart, shown during expansion (Left) and relaxation (Right). Open circuit voltage as a function of time for PZT MEHs on bovine (green) and ovine (blue) hearts, mounted on (C) RV, (D) LV, and (E) free wall at an orientation of 0° relative to the apex of the heart. Here, the heart rate is 80 beats per min. (F) Photograph of PZT MEHs oriented at different angles. (G) Measurements of maximum values of the peak open circuit voltages produced by these devices indicate peak output at 45°. Error bar is calculated standard error. (H) Maximum peak voltages of a PZT MEH on the RV of a bovine heart at 0° for various heart rates (80–120 beats per min) controlled by a temporary pacemaker. (I) Maximum peak voltage for various dosages of dobutamine infusion, for the case of a device on the RV (green points) and LV (pink points) of a bovine heart at 0°. Error bar is calculated standard error. Voltage as a function of time for PZT MEHs on the LV (J) and RV (K) of a bovine heart, with a base and maximum dose of dobutamine.](https://www.pnas.org/cgi/doi/10.1073/pnas.1317233111)
Wall, whereas ejection from the LV primarily involves a reduction in its diameter or circumference, associated with wall thickening (25). The contraction of the RV by shortening the free wall likely results in enhanced overall wall motion and hence increased bending of the MEH than the twisting contraction of the LV.

Orientation of the device is also important, as illustrated by the data in Fig. 3 F and G. Bending in the longitudinal direction ($x_1$ direction in SI Appendix, Fig. S3) relative to the orientation of the PZT fibers provides the highest efficiency (see SI Appendix for details). To examine the dependence on orientation on the heart, measurements involve the longitudinal direction of PZT fibers along the 0°, 45°, and 90° directions with respect to the apex of the heart. The 0° and 45° directions produce greater voltages than those at 90° (Fig. 3G). This behavior, for 0° and 45°, is expected because the myocardial tissue of the heart is anisotropic and contracts in a generally circumferential direction (26) with cardiac fibers aligned in a continuous manner from +60° on the endocardium to −60° on the epicardium (27); 90° is clearly out of this range. Theoretically, as shown in Eq. 5, the voltage is proportional to the strain rate, or equivalently, the strain amplitude for a given period. FEA confirms that the strain by bending along the $x_1$ direction is larger than that along $x_2$, as shown in SI Appendix, Figs. S3 and S4. Aligning the $x_1$ direction with the strongest bending direction optimizes the harvested energy.

The size of the heart, beat rate, and force of contraction are additional factors that affect voltage output. In the studies reported here (Figs. 3–5), the bovine heart is nearly twice the mass of the ovine model. All devices deployed on the former yield significantly higher voltage outputs than those in the ovine model, for any site of implantation on the heart (SI Appendix, Figs. S10 and S11). Modulating the heart rate through electrical and chemical stimulation reveals the dependence on frequency. Fig. 3H demonstrates that increasing the bovine heart rate with a temporary pacemaker (Medtronic 5388) increases the voltage in direct proportion to heart rate (see SI Appendix, Figs. S10 and S11 and Movie S3), consistent with modeling results, assuming that the amplitude does not vary significantly with frequency. Similarly, increasing the force and degree of contractility of the myocardium via dobutamine infusion, i.e., the inotropic state (15 μg kg⁻¹ min⁻¹), increases both the voltage and the frequency of the output (SI Appendix, Fig. S12). Such a response occurs for both the LV and RV, as shown in Fig. 3 I–K. The lung represents another internal organ of interest for mechanical energy harvesting. Results for mounting on the bovine (Fig. 4A and Movie S4) and ovine lung of the bovine are consistent with expectation. In particular, both models respiratory movement can be converted to voltage (Fig. 4 B and D). Unlike observations with the heart, data collected from the lung show no strong interspecies differences (Fig. 4 C and E).

Fig. 4. In vivo evaluation of PZT MEHs on the lung and diaphragm. (A) Photograph of a PZT MEH cointegrated with a microbattery and rectifier, mounted on the bovine lung. Voltage as a function of time for such devices on the bovine (B) and ovine (C) lung. D and E show plots corresponding to the regions indicated by the red dashed boxes in B and C, respectively. (F) Photograph of a PZT MEH on the bovine diaphragm. Voltage as a function of time for such a device on the bovine (G) and ovine (H) diaphragm.

Fig. 5. Performance of a PZT MEH evaluated with the chest open and closed and scaling of power output in multilayer stacked designs. Photograph of a PZT MEH without and with battery, rectifier, and pacemaker connection on a bovine heart when the chest is (A) open and (B) closed. Voltage as a function of time for a PZT MEH on the bovine RV with the chest open (C) and closed (D). (E) Schematic illustration of a multilayer stack of five independent PZT MEHs connected in series. A circuit schematic appears in the upper right-hand corner. (F) Photograph of such a stacked configuration, peeld apart at the left edge to show the separate layers. Each device appears just before stacking (Inset). (G) Schematic illustration of the theoretical shape for buckling of a stack of PZT MEHs with spin-cast layers of silicone elastomer (thickness = 10 μm) in-between and under compression. (H) Time-averaged power density as a function of the bending load displacement for stacks consisting of one (pink curve), three (green curve) and five (blue curve) PZT MEHs, connected in series. E, experimental data; T, theory.
output of a PZT MEH depends on its design. With layouts described previously, the time-averaged power density (electrical power output per unit area of PZT) corresponds to 0.12 and 0.18 $\mu$W/cm$^2$ for mounting on the RV at 0° and 45° as seen in SI Appendix, Fig. S14 A and B, respectively. Stacking multiple PZT MEH sheets (Fig. 5 E and F and SI Appendix, Fig. S15) increases the power output. Fig. 5G shows a stack of $n_{\text{MEH}}$ PZT MEHs thin, spin coated of silicone layer in between as adheres and strain isolating layers. The silicone, which is much more compliant (Young’s modulus 60 KPa) than the PI (Young’s modulus 2.5 GPa), does not significantly alter the modes of deformation of the PZT MEHs (SI Appendix, Table 1). As a result, for a stack of $n_{\text{MEH}}$ PZT MEHs, Eq. 5 can be applied simply by $N$ using $N_{\text{MEH}}$ with $n_{\text{MEH}}$. For $\Delta L_{\text{max}} = 10$ mm, in multilayer stacks with $n_{\text{MEH}} = 3$ and 5, in vitro experiments show peak voltages of 5.8 and 8.1 V (SI Appendix, Fig. S16), respectively, consistent with Eq. 5. Both values are higher than that (3.7 V) for a single-layer device. In vivo demonstrations on the bovine heart are consistent with these results (SI Appendix, Fig. S17). The time-averaged power density increases with $n_{\text{MEH}}$ and can reach as large as 1.2 $\mu$W/cm$^2$ (Fig. S1H) for $n_{\text{MEH}} = 5$, which is sufficient to operate a cardiac pacemaker (1) (see SI Appendix for details).

Conclusions

The reported results provide evidence that piezoelectric MEHs can yield significant electrical power from motions of internal organs, up to and exceeding levels relevant for practical use in implants. Theoretical models establish design rules and provide predictive capabilities for efficiencies associated with various single and multilayer layouts. Practical applications demand long-term reliable operation in the closed body cavity. The extensive cycling tests and initial measurements of biocompatibility summarized here represent starting points for the sort of qualifications that are required. In addition to uses on internal organs, the same types of systems can be implemented in skin-mounted configurations for health/wellness monitors or nonbiomedical devices. The potential to eliminate batteries or, at least, the need to replace them frequently represents a source of motivation for continued work in these and related directions.

Materials and Methods

Detailed fabrication steps for the devices, designs for the data acquisition systems, and related hardware all appear in SI Appendix. In vitro biocompatibility study and in vivo implantation of the device on animal models are also described in the SI Appendix. All animal procedures and experiments were approved by the Institutional Animal Care and Use Committee at the University of Arizona.

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Conformal Piezoelectric Energy Harvesting and Storage From Motions of the Heart, Lung and Diaphragm

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1. SI Materials and Methods

1.1. Fabrication of Lead Zirconate Titanate (PZT) Ribbons Array and Transferprinting Them on PI

PZT ribbons embedded in capacitor type structures with top and bottom electrodes were fabricated as following. The top electrode was formed by deposition of Au/Cr (200 nm/10 nm) with an electron beam evaporator on the surface of a multilayer stack of Pb(Zr0.52Ti0.48)O3/Pt/Ti/SiO2 (500 nm/300 nm/20 nm/600 nm; INOSTEK) on a silicon wafer. Coating the wafer with photoresist (PR, AZ5214E) followed by patterning by photolithography defines the top electrode areas (50 µm x 2 mm) for each PZT ribbon. Au and Cr layers were etched with gold etchant (TFA, Transene Company Inc., USA) and CR-7 chrome etchant (OM Group, USA), respectively. PZT ribbons with thickness of 500 nm and an area of 100 µm x 2.02 mm were created by wet chemical etching with HNO3 (nitric acid): BHF (buffered hydrogen fluoride): H2O (DI water) = 4.51:4.55:90.95 through a hard-baked mask of PR (AZ4620, Clariant) (See Fig. S1A). The hard baking involved 80 °C for 5 minutes, 110 °C for 30 minutes and then 80°C for 5 minutes (See Fig. S1B). The bottom Pt/Ti electrode with area of 140 µm x 2.02 mm was patterned by wet chemical etching with HCl (hydrochloric acid) : HNO3 : DI water = 3:1:4 at 95°C through a hard baked mask of AZ4620. The PZT layers were protected by photolithographically patterned PR during partially removal of the sacrificial layer, SiO2 with dilute HF (hydrofluoric acid) (DI water: 49% HF = 1:3). Hard baked photoresist mask was completely removed in an acetone bath for 3 hours after etching the SiO2.

A PDMS stamp for transfer was fabricated by casting a mixture of PDMS (Sylgard 184, Dow Corning; 10:1 ratio of prepolymer to curing agent) in a plastic petri dish, and curing at
room temperature for 24 hours. Next, the stamp was conformally contacted on the top of the ribbons (See Fig. S1C). The devices were retrieved by peeling the stamp away from the Si wafer and then transfer printed on a film of PI (75 μm, DuPont, USA). This film was formed by spin-coating a layer of poly(pyromellitic dianhydride-co-4,4’-oxydianiline) amic acid solution, to a thickness of 1.2 μm (See Supplementary Fig. S1D). The printed PZT ribbons on the PI were spin-coated with another layer of PI for encapsulation and hard baked at 250 °C in a vacuum oven.

To open contact holes for the top and bottom electrodes, the device on PI was patterned with photoresist (PR, AZ 4620), and developed with diluted AZ®400K developer (AZ Electronic Materials, USA) (deionized water (DI) : 400 K developer = 1:2). The PI was etched in reactive ion etching (RIE, March) to open contact holes. Connection lines were obtained by the deposition of Au/Cr (200 nm/10 nm) using electron beam evaporation. The interconnection lines were spin-coated with another layer of PI for encapsulation and hard baked at 250 °C in a vacuum oven.

1.2. Poling of the PZT thin film and device analysis via mechanical testing

The PZT thin films sandwiched between Ti/Pt (20 nm/300 nm) bottom electrode and Cr/Au (10 nm/200 nm) were poled with an electric field of 100 kV/cm at 150°C for 2 hours. A semiconductor parameter analyzer (4155C, Agilent) was used to measure the open voltage and short current values of mechanical energy harvester. Figure S15 shows a mechanical bending stage for testing the PZT MEH. The system includes a high precision linear stage (ATS100-150; Aerotech, Inc.; USA), equipped with precision ground ball screw, noncontact rotary encoder with 1000 line/rev, and brushless servomotor to achieve a motion with an accuracy of ±0.5 μm.
and a bidirectional repeatability of ±0.3 µm over 150 mm stage motion range. The stage can achieve velocities up to 100 mm/sec with maximum side load of 100 N in horizontal configuration. The stage motion was controlled with a Soloist single axis PWM digital controller (SOLOISTCP20; Aerotech, Inc.; USA) and USB interface. Additionally, two U-Form Vise grip (TH240k, Grip Engineering Thümler GmbH; Germany) were attached to the stage. Pyramid shape jaws (TH240k-BP, Grip Engineering Thümler GmbH; Germany) were attached to the vices to achieve maximum tensile gripping force of 2.5 kN of the specimen during the test. A LabVIEW (National Instruments Corporation; USA) based program was designed to control the stage to perform the test cycle (Fig. S15).

1.3. *In Vitro* Biocompatibility Assessment of the Flexible PZT MEH

Aortic smooth muscle cells (SMC) were harvested from albino Sprague-Dawley rat (with IACUC approval protocol from the University of Arizona) and subsequently cultured in Dulbecco’s Modified Eagle Medium (DMEM) supplemented with 10% (v/v) fetal calf serum, 2% (v/v) of 0.2 M glutamine, 1% (v/v) of antibiotic/antimycotic solution. All cell culture supplements and medium were either purchased at Invitrogen (Grand Island, NY, USA) or BioWhittaker (East Rutherford, NJ, USA). SMCs were then sub-cultivated and cultured in an incubator at 37 °C, 5% CO₂, and 95% relative humidity to passages 2-5 before subjecting to ribbonping (trypsin-EDTA), cell counting, and seeding on the sterilized fibronectin (BD-Biosciences, San Jose, CA, USA) coated PZT MEH.

To prepare for biocompatibility studies, PZT MEH ribbons were cut into 1 cm² coupons to fit in a 24 well culture plate. These coupons were then cleaned using Basic Harrick Plasma (Ithaca, NY, USA) for 10 minutes at 1 torr to increase surface hydrophobicity. Subsequently, coupons were sterilized under UV light for 30 minutes then coated with 150 µL of 1 mg/ml fibronectin.
for another 15 minutes. Any excess fibronectin was removed and allowed to dry in the culture hood for another 15 minutes prior to $5 \times 10^4$ SMCs cultivation for 1, 3, and 9 days at 37 °C, 5% CO$_2$, and 95% relative humidity. Medium was changed every 24 hours.

At the given latter endpoints, SMCs were stained using actin cytoskeleton/focal adhesion staining kit (Millipore, MA, USA). Cells were fixed with 4% paraformaldehyde for 15 minutes, then washed and permeated the membrane with 0.05% Triton X for 5 minutes. Cells were washed and blocked with 1% protein standard (fractionated bovine serum albumin) in PBS at pH7.4 and subsequently stained with anti-vinculin for 1 hour at room temperature. Cells were washed and stained with fluorescein isothiocyanate conjugated mouse anti-immunoglobulin G (mIgG-FITC) to label vinculin and tetramethyl rhodamine isothicyanate (TRITC) conjugated Phalloidin to selectively label F-actin. After washing off all the excess stains, cells were then mounted in vector shield with DAPI and imaged using the Nikon C1Si Laser Scanning Confocal Fluorescence Microscope. SMCs on the PZT MEH exhibit normal morphology where intact cytoskeletal fibers, nucleus, and focal adhesion points are present, suggesting that the surface of PZT MEH is a suitable environment for cell growth.

Scanning electron microscopy (SEM, FEI Inspec S, Thermo, Rockford, IL, USA) was also utilized to physically demonstrate the adherent of cells on PZT MEH ribbons. Briefly, SMCs after 9 days of culture were fixed in 5% Glutaraldehyde in PBS at pH7.4 (100% fixator) then subjected to a graded series of water and ethanol (100% fixator $\rightarrow$ 3:1 $\rightarrow$ 1:1 $\rightarrow$ 1:3 $\rightarrow$ 100% Distilled water $\rightarrow$ 3:1 $\rightarrow$ 1:1 $\rightarrow$ 1:3 $\rightarrow$ 100% Ethanol. Samples were soaked for 5 min at each step. Finally, Samples were freeze-fried using critical point drying (CPD, EMS #3100, Hatfield, PA, USA). A more detailed CPD protocol can be found in (1). Subsequently, samples were sputter-coated with gold at about 5-8 nm thick and imaged at 30 kV with aperture spot size of 3.
To evaluate the biocompatibility of the PZT MEH structures, the viability and cytotoxicity of SMC were determined after 9 days of cultivation by utilizing two color fluorescence LIVE/DEAD viability (Invitrogen) assay and Lactate Dehydrogenase (LDH) assay (Thermo, IL, USA), respectively. For LIVE/DEAD assay, SMCs grown on PZT MEH ribbon after 9 days were prepared and stained according to manufacture protocol with the exception that samples were mounted in Flouroshield containing DAPI (Sigma Aldrich, St. Louis, MO, USA). Briefly, the culture medium was aspirated from each of the wells then rinsed three times with 1x PBS, and a working solution (consisting of 5 mL 1x PBS, 10 µL of 2mM EthD-1, and 2.5 µL of 4mM Calcein AM) was added to cover each of the samples. The submerged samples were incubated for 30 minutes at 37°C. After the incubation period, the working solution was removed, and the samples were rinsed once with 1x PBS, then mounted and immediately imaged with the Nikon C1Si fluorescence microscope. For the LDH cytotoxicity assay, 50 µL medium from cells grown on fiber surfaces at the latter given endpoints (ie. 1, 3, and 9 days) were mixed with 50 µL reaction mixture (prepared according to the manufacture recipe) in a 96 wells plate for 30 minutes at room temperature. Stop solution (50 µL) was added and the plate was read at 490/680nm. Mean percent of healthy cells was reported with standard error of mean. All statistical analysis was performed using Microsoft Excel 2010. TTEST was analyzed using one-tailed distribution and two-sample unequal variance type. Statistical significance of differences between the means was determined using a student’s t-test (p = 0.05).

1.4. Implantation of the PZT MEH: an In vivo Study

All animals received humane care and were handled in accordance with IACUC approval protocol at the University of Arizona Animal Care Center. Male Corriente bovine (n=4, 90-140
kg) and domestic ovine (n=1, 45 kg) were used. Animals fasted for 12 hours without food and 8 hours without water prior to left thoracotomy. Animals were operated off-pump, without cardiopulmonary bypass and survived for two hours post-surgery.

Briefly, animal were restrained with Telazol (2.2 mg/kg Intramuscular (IM) or 4 mg/kg Intravenous (IV)), weighed, and anesthetized with 3-5% Isoflurane induction administered via the facemask. An oral endotracheal tube was then placed. Butorphanol (0.01-0.02 mg/kg) and Xylazine (0.1-0.2 mg/kg) were administered via intramuscular (IM) injection into the hamstring muscle to relieve pain. Animal were clipped, shaved, and prepped for the surgery. Glycypyrrolate was administered at 0.002-0.005 mg/kg Intravenous (IV) when bradycardia is present. Animal was then intubated and anesthesia was further maintained at 0.5-3% Isoflurane and at room temperature throughout the study. An orogastric tube was placed to decompress the rumen. A triple lumen catheter (7-8F) was inserted percutaneously into the right jugular vein for IV line for drip infusion of lactated ringers to maintain hydration and for delivery of drugs. An arterial pressure catheter was placed in the carotid artery and ECG was connected to monitor the animal.

Prior to surgery, Ketamine (3-4 mg/kg) and Midazolm (0.25 mg/kg) were inducted intravenously to relax muscle. Pancuronium Bromide (0.04-0.1 mg/kg, IV) were used to block myoneural junctions. Fentanyl (loading dose of 5 µg/kg and dripping dose of 5 to 10 µg/kg/hr), Lidocaine (loading dose of 2 mg/kg and dripping dose of 2 mg/kg/hr), and Ketamine (loading dose of 2 mg/kg and dripping dose of 2 mg/kg/hr) were infused intravenously in addition to inhalant throughout the operation. Left thoracotomy was performed and cauterized with the electrosurgico knife (Conmed Excalibur Plus, Utica, NY, USA). The fifth left rib was snipped and a retractor was used to expose solid tissues like the heart, lung, and diaphragm. Amiodarone
(loading dose of 50 mg IV bolus and dripping dose of 0.3 to 0.9 mg/kg/hr) was administered to control arrhythmias prior to the manipulation of the heart. PZT MEH was then sutured to solid tissues using 2-0 bladed polypropylene (Ethicon, San Angelo, TX, USA). A cable (ACF; Anisotropic conductive film, 3M Co.) was connected to a voltmeter storage monitor systems. To evaluate the responsiveness of the PZT MEH, temporary pacemaker (Medtronic 5388, Minneapolis, MN, USA) and Dobutamine (5-15 µg·kg⁻¹·min⁻¹, IV) was used to fluctuate the heart rate. After all initial readings, animal were survived for 2 hours to allow the PZT MEH to recharge the battery. Incision sites were closed using the standard technique. At the termination of the surgery, animal were humanely euthanized with 30 ml of Beuthanasia while still under general anesthesia. The PZT MEH was then removed for detailed examination and reading.

2. Theory

2.1. Mechanics analysis

For the out-of-plane displacement \( w = A \left[ 1 + \cos \left( \frac{2 \pi x}{L} \right) \right] / 2 \) shown in Fig. S6A for plane-strain analysis (\( \varepsilon_{22} = 0 \)), the bending energy in PI is related to the curvature \( w'' \) by \( \left( \frac{EI_{PI}}{2} \right) \left( w'' \right)^2 ds \), where the integration is over the PI length, and \( \frac{EI_{PI}}{2} = \left( \frac{E_{PI} t_{PI}^3}{12} \right) \), \( E_{PI} \) and \( t_{PI} \) are the plane-strain bending stiffness, plane-strain modulus, and thickness of PI, respectively. The membrane energy can be obtained following the same approach of Song et al. (2). Minimization of the total energy (sum of bending and membrane energies) gives the amplitude \( A \) as

\[
A = \frac{2}{\pi} \sqrt{L \cdot \Delta L - \frac{\pi^2 t_{\text{kapton}}^2}{3}} \approx \frac{2}{\pi} \sqrt{L \cdot \Delta L},
\]  
(S1)
where the last approximation holds when the compression of PI $\Delta L$ is much larger than its critical value $\pi^2 t_{pl}^2/(3L)$ to initiate buckling. For a 75 $\mu$m-thick and 2.5 cm-long PI, $\pi^2 t_{pl}^2/(3L) \approx 0.74$ $\mu$m is negligible as compared to compression $\Delta L = 1.5 \sim 10$ mm in the experiments.

The bending moment $M$ in PI is related to the curvature $w''$ by $M = \overline{EI} w''$. For the part of PI covered by the PZT ribbons (Fig. S6B), the local curvature is reduced to $M/\overline{EI}_{comp}$ due to the additional bending stiffness of PZT ribbons, where

$$\overline{EI}_{comp} = \sum_{i=1}^{n} E_i t_i \left[ t_i^2/3 + \left( \sum_{j=1}^{i} t_j - y_{\text{neutral}} \right) \left( \sum_{j=1}^{i} t_j - y_{\text{neutral}} - t_i \right) \right]$$

is the effective bending stiffness of multi-layer structure (Fig. S6B) with PI as the 1st layer ($i=1$) and the summation over all $n$ layers, $E_i$ and $t_i$ are the plane-strain modulus and thickness of the $i^{th}$ layer, respectively, and $y_{\text{neutral}} = \left[ \sum_{i=1}^{n} E_i t_i \left( 2 \sum_{j=1}^{i} t_j - t_i \right) \right]/\left( 2 \sum_{i=1}^{n} E_i t_i \right)$ is the distance from the neutral mechanical plane to the bottom of 1st (PI) layer. The membrane strain in PZT is the axial strain at the center of PZT ribbons, and is given by

$$\varepsilon_m = (\overline{EI}_{pl}/\overline{EI}_{comp}) w'' h,$$

(S2)

where $h$ is the distance from the center of each PZT ribbon to the neutral mechanical plane of the cross section as shown in Fig. S6B. For the length of PZT ribbons much smaller than that of the PI, $w''$ is evaluated at the center $x_1=0$ of PZT ribbons as $w'' = -4\pi \sqrt{\Delta L/L}$, which gives the membrane strain as shown in Eq. (1) in the main text. For the structure shown in Fig. S6B, $\overline{E}_1 = 2.83$ GPa and $t_1 = 75 \mu$m for PI, $\overline{E}_2 = 2.83$ GPa and $t_2 = 1.2 \mu$m for the PI layer, $\overline{E}_3 = 129$ GPa and $t_3 = 20$ nm for the Ti layer, $\overline{E}_4 = 196$ GPa and $t_4 = 0.3 \mu$m for the Pt layer,
\( E_5 = 69.2 \text{ GPa} \) and \( t_5 = 0.5 \mu \text{m} \) for the PZT layer, \( E_6 = 292 \text{ GPa} \) and \( t_6 = 10 \text{ nm} \) for the Cr layer, \( E_7 = 96.7 \text{ MPa} \) and \( t_7 = 0.2 \mu \text{m} \) for the Au layer, and \( E_8 = 2.83 \text{ GPa} \) and \( t_8 = 1.2 \mu \text{m} \) for the PI layer; these give \( \frac{EI_{ PF}}{EI_{ comp}} = 0.45 \), \( y_{neutral} = 52.0 \mu \text{m} \) and \( h = (t_1 + t_2 + t_3 + t_4 + t_5/2) - y_{neutral} = 24.7 \mu \text{m} \).

2.2. Piezoelectric analysis

The constitutive model of piezoelectric materials gives the relations among the stress \( \sigma_{ij} \), strain \( \varepsilon_{ij} \), electrical field \( E_i \) and electrical displacement \( D_i \) as

\[
\begin{bmatrix}
\sigma_{11} \\
\sigma_{22} \\
\sigma_{33} \\
\sigma_{12} \\
\sigma_{23} \\
\sigma_{31}
\end{bmatrix} = \begin{bmatrix}
c_{11} & c_{12} & c_{13} & 0 & 0 & 0 \\
c_{12} & c_{11} & c_{13} & 0 & 0 & 0 \\
c_{13} & c_{13} & c_{33} & 0 & 0 & 0 \\
0 & 0 & 0 & c_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & c_{44} & 0 \\
0 & 0 & 0 & 0 & 0 & (c_{11} - c_{12})/2
\end{bmatrix} \begin{bmatrix}
\varepsilon_{11} \\
\varepsilon_{22} \\
\varepsilon_{33} \\
2\varepsilon_{12} \\
2\varepsilon_{23} \\
2\varepsilon_{31}
\end{bmatrix} = \begin{bmatrix}
0 & 0 & e_{31} \\
0 & 0 & e_{31} \\
0 & 0 & e_{33} \\
2\varepsilon_{12} & 0 & 0 \\
2\varepsilon_{23} & 0 & 0 \\
2\varepsilon_{31} & 0 & 0
\end{bmatrix} \begin{bmatrix}
E_1 \\
E_2 \\
E_3
\end{bmatrix}, \quad (S3)
\]

\[
\begin{bmatrix}
D_1 \\
D_2 \\
D_3
\end{bmatrix} = \begin{bmatrix}
0 & 0 & 0 & e_{15} & 0 & 0 \\
0 & 0 & 0 & e_{15} & 0 & 0 \\
e_{31} & e_{31} & e_{33} & 0 & 0 & 0
\end{bmatrix} \begin{bmatrix}
\varepsilon_{11} \\
\varepsilon_{22} \\
\varepsilon_{33} \\
2\varepsilon_{12} \\
2\varepsilon_{23} \\
2\varepsilon_{31}
\end{bmatrix} + \begin{bmatrix}
k_{11} & 0 & 0 \\
k_{22} & 0 & 0 \\
k_{33} & 0 & 0
\end{bmatrix} \begin{bmatrix}
E_1 \\
E_2 \\
E_3
\end{bmatrix}, \quad (S4)
\]

The plane-strain condition \( \varepsilon_{22} = 0 \) of PZT ribbons, together with \( \sigma_{33} = 0 \) from the traction free on the top surface of the structure, gives \( D_3 = \tilde{\varepsilon}e_{11} + \tilde{k}E_3 \), where \( \tilde{\varepsilon} = e_{31} - (c_{13}/c_{33})e_{33} \) and \( \tilde{k} = k_{33} + (e_{33}^2/c_{33}) \) are the effective piezoelectric constants. The electrical displacement can be further obtained as
\[ D_3 = \bar{\varepsilon} \varepsilon_m + \frac{\bar{K}V}{N_{\text{PZT}}} \]  

(S5)

from the charge equation \( dD_3/dx_3 = 0 \) and the relation \( E_3 = -\partial \phi/\partial x_3 \) between the electrical field and electrical potential, together with the boundary condition that the voltage difference between the bottom and top of PZT is \( V/N \), where \( V \) is total voltage between the two ends of the \( N \) groups of PZT ribbons in series, and \( t_{\text{PZT}} \) is the thickness of PZT ribbons. Eq. (S5) shows that the electrical displacement is linear with the membrane strain of PZT ribbons, and is independent of the bending strain. Therefore the bending strain does not contribute to the voltage and current output of the MEH given in the following.

2.3. Current

The voltage \( V \) across the two ends of the \( N \) groups of PZT ribbons in series is zero after the PZT ribbons are connected to an ampere meter (Fig. S6C). The electrical displacement in Eq. (S5) then becomes \( D_3 = \bar{\varepsilon} \varepsilon_m \), where \( \varepsilon_m \) is given in Eq. (1) in the main text. Its rate gives the current \( I = -A_{\text{PZT}} \dot{D}_3 \), or equivalently Eq. (2) in the main text, where \( A_{\text{PZT}} = m \left( w_{\text{PZT},1}l_{\text{PZT},1} + w_{\text{PZT},2}l_{\text{PZT},2} \right) \) is total area of PZT ribbons in each group; \( m = 10 \) is the number of PZT ribbons in each group, \( w_{\text{PZT},1} = \text{100\,\mu m} \), \( w_{\text{PZT},2} = \text{140\,\mu m} \), \( l_{\text{PZT},1} = \text{2 mm} \) and \( l_{\text{PZT},2} = \text{160\,\mu m} \) are the widths and lengths of the two rectangular parts of each PZT ribbon, respectively (Fig. S6B).
2.4. Voltage

For voltage measurement, the voltage $V$ in Eq. (S5) across the two ends of the $N$ groups of PZT ribbons in series is no longer zero after the PZT ribbons are connected to a voltmeter (Fig. S6C). The rate of the displacement in Eq. (S5) gives the current $I = -A_{PZT}\left\{\bar{\epsilon}\dot{\epsilon}_m + \left[\frac{\bar{E}}{N_i}\right]V\right\}$, which, together with the Ohm’s law gives Eq. (4) in the main text.

2.5. Rectifier

For current measurements with the rectifier (Fig. S6D), the resistance across the PZT groups is no longer zero because of the resistance of the rectifier $R_{rectifier}$. Instead it is the same as the voltage measurement shown Fig. S6C except that the resistance of the voltmeter $R$ is replaced by $R_{rectifier}$. After this replacement Eq. (5) still holds, but is taken by its absolute value because of the rectifier (3). The current is then obtained from Ohm’s law as

$$I = \frac{1}{R_{rectifier}} \left(\bar{\epsilon}\right)\frac{N_{PZT}}{k} e^{-\frac{N_{PZT}}{R_{rectifier}k}} \left[\int_0^t \dot{\epsilon}_m e^{-\frac{N_{PZT}}{R_{rectifier}k}t} dt\right].$$  \hspace{1cm} (S6)

For the voltage measurement with the rectifier (Fig. S6D), the resistance $R$ is replaced by $R + R_{rectifier}$. After this replacement Eq. (5) still holds for the voltmeter and rectifier in series, and the voltage on the voltmeter is then obtained by multiplying the factor $R'\left(R + R_{rectifier}\right)$ of the absolute value of Eq. (5) as

$$V = \frac{R}{R + R_{rectifier}} \left(\bar{\epsilon}\right)\frac{N_{PZT}}{k} e^{-\frac{N_{PZT}}{R + R_{rectifier}k}} \left[\int_0^t \dot{\epsilon}_m e^{-\frac{N_{PZT}}{R + R_{rectifier}k}t} dt\right].$$  \hspace{1cm} (S7)
2.6. Direction of bending

Figs. S3 and S4 show the strain in PZT bent along \( x_1 \) and \( x_2 \) directions, respectively, for the axial compression of \( \Delta L = 1.5, 3, 5 \) and 10 mm. For the same axial compression \( \Delta L \), the strain in PZT bent along \( x_1 \) direction is larger than that along \( x_2 \) direction, and is therefore more effective for energy harvesting.

2.7. The time-averaged power density

The electrical energy output is obtained by integrating the ratio of the square of the voltage \( V \) and the resistance \( R \) over total time \( t_{\text{total}} \) as \( W = \int_0^{t_{\text{end}}} V^2/R \, dt \) (4). The time-averaged power density is given by \( P = W/(t_{\text{total}}N_{A_{\text{PZT}}}) \).

2.8. FEA for the deformation of the stacks

Finite element analysis (FEA) is used to study the deformation of a single PZT MEH and the stacks of 3 and 5 PZT MEHs with spin-casted silicone layer of 10 \( \mu \)m thickness in between. The solid elements in the ABAQUS finite element program (5) are used to mesh all materials, including the PZT ribbons. Fig. S18 shows that the amplitudes of the PZT MEHs in the stacks are very close to that of the single PZT MEH, with the difference less than 1\%, which clearly suggests that the soft silicone layers do not make significant contribution to the deformation of the PZT MEHs.
References


SI Figure Legends

**Figure S1.** Schematic illustration of procedures for fabricating a PZT MEH on a polyimide (PI) substrate. Schematic illustration of (A) array of ribbons of PZT in a capacitor structure on a SiO₂/Si wafer (left). A cross section of one element in this array appears on the right. (B) Pattern of photoresist on the array (left), and cross section of one element during undercut etching with dilute HF solution (right). (C) Retrieving the array with a PDMS stamp to leave them adhered to the surface of the stamp. (D) Result after transfer printing onto a flexible film of PI.

**Figure S2.** Fabrication steps to fabricate PZT ribbons. Optical microscope image of printed ribbons of PZT on a film of PI (A) before bending, (B) after bending with the radius of 20 mm curvature and (C) with the view of top and bottom connection pads. (D) The cross sectional illustration of a ribbon of PZT with top and bottom electrodes, on a PI substrate with a thin overcoat of PI for encapsulation. (E) Schematic illustration of PZT ribbons grouped and connected in series.

**Figure S3.** Results of FEM analysis for strain mapping of the bare PZT ribbons array. (A) Deformation of a PZT MEH and (B) corresponding strain distributions in the ribbons of PZT. The displacement loads (ΔL) correspond to 0, 1.5, 3, 5 and 10 mm along the x₁ direction, from left to right.

**Figure S4.** Results of FEM analysis for strain mapping of the bare PZT ribbons array. (A) Deformation of a PZT MEH and (B) corresponding strain distributions in the ribbons of PZT. The displacement loads (ΔL) correspond to 0, 1.5, 3, 5 and 10 mm along the x₂ direction, from top to bottom.

**Figure S5.** Experimental and theoretical results for displacement, voltage, and current as a function of time for cyclic bending of a PZT MEH with rectification.

**Figure S6.** Device layouts and information related to variable names and physical parameters used in calculations. Schematic illustration of (A) the theoretical shape for buckling of a PZT MEH under compression, (B) top view of a single PZT ribbon capacitor structure (top), and cross section showing
position of the neutral mechanical plane of the device (bottom). A buckled array of PZT ribbon capacitor structures on a PI substrate without $(C)$ and with $(D)$ rectification.

Figure S7. Images that show the ability of flexible PZT MEHs to conform to various surfaces. Photograph of a PZT MEH $(A)$ on a balloon. $(B)$ A magnified view of the device. Photograph of a PZT MEH wrapped onto the $(C)$ arm and $(D)$ finger.

Figure S8. Responses of a PZT MEH for forward and reverse connections. $(A)$ Forward and $(B)$ reverse connections to the measurement system. $(C)$ Open-circuit voltage and $(D)$ short-circuit current of a PZT MEH under cyclic bending for these two configurations.

Figure S9. Studies of cycle life in a PZT MEH. Photographs of device clamped in a bending stage with a layer of a transparent gelatine (Knox) on top, in $(A)$ flat and $(B)$ bent states. Output voltage as a function of time after a single cycle of bending $(C)$ and twenty million cycles of bending $(D)$.

Figure S10. Output from a PZT MEH mounted on the bovine heart, for different pacing conditions. Voltage as a function of time measured from a PZT MEH on the $(A)$ RV, $(B)$ LV, and $(C)$ free wall of a bovine heart at $0^\circ$ with respect to the apex, at heart rates of 80, 90, 100, 110, and 120 beats/min.

Figure S11. Output from a PZT MEH mounted on the ovine heart, for different pacing conditions. Voltage as a function of time measured from a PZT MEH on the $(A)$ RV, $(B)$ LV, and $(C)$ free wall of a bovine heart at $0^\circ$ with respect to the apex, at heart rates of 110, 120 and 130 beats/min; respectively.

Figure S12. Output from a PZT MEH mounted on the bovine heart for different levels of dobutamine infusion. Voltage as a function of time measured from a PZT MEH on the $(A)$ LV, $(B)$ RV of a bovine heart at $0^\circ$ with respect to the apex with 0, 2, 5, 10 and 15 µg·kg$^{-1}$·min$^{-1}$ of dobutamine.
Figure S13. Performance of a PZT MEH evaluated with the chest open and closed on a pig model.
Photograph of a PZT MEH without battery and rectifier connection on pig heart when the chest is (A) open and (B) closed. Voltage as a function of time for a PZT MEH on the pig LV with the chest open (C) and closed (D).

Figure S14. Effect of orientation of the PZT MEH with respect to the apex of a bovine heart.
Voltage as a function of time measured on the RV of a bovine heart with 80 beats/min (A) at 0° and (B) at 45°.

Figure S15. Image of the experimental setup used for bending and cycling tests. (A) The picture here shows testing of a five layer stack of PZT MEHs, with a LabVIEW control interface. A cross sectional image of the stacked device appears in the top left. (B) A photograph of LabVIEW controllable mechanical bending stage.

Figure S16. Output voltage as a function of time measured from a stacked arrangement of PZT MEHs. Experimental and theoretical results for displacement, voltage as a function of time, for four different bending load displacements. Spin-cast layers of silicone bond the separate PZT MEHs.

Figure S17. In vivo evaluation of stacked PZT MEHs on voltage output. Voltage as a function of time for devices that incorporate (A) 3 and (B) 5 layers of PZT MEHs connected in series and mounted in the RV of a bovine heart (80 beats/min) at 0° (left) and at 45° (right).

Figure S18. The buckle amplitude of a single PZT MEH and that of each PZT MEH in the stacks, incorporating 3 and 5 PZT MEHs with spin-casted silicone layer of 10 µm thickness in between, for compression \( \Delta L = 5 \) mm. The differences between the amplitudes are very small, which suggests that the soft silicone layers do not make significant contribution to the deformation of the PZT MEHs.
Figure S1
A PZT ribbon
B

C 10 ribbons in parallel
D

E 12 groups are connected in series

150 µm

150 µm

150 µm

150 µm

PI : 1.2 µm
PZT : Cr/Au : 20/200 nm
500 nm
Ti/Pt : 20/300 nm
PI : 1.2 µm
PI : 1.2 µm
PI : 75 µm

Figure S2
ΔL = 0
ΔL = 1.5 mm
ΔL = 3.0 mm
ΔL = 5.0 mm
ΔL = 10 mm

Figure S3
Figure S4

ΔL = 0
ΔL = 1.5 mm
ΔL = 3.0 mm
ΔL = 5.0 mm
ΔL = 10 mm
<table>
<thead>
<tr>
<th>Time (sec)</th>
<th>Voltage (V)</th>
<th>Current (µA)</th>
<th>Displacement (mm)</th>
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<tbody>
<tr>
<td></td>
<td>Experiment</td>
<td>Theory</td>
<td></td>
</tr>
<tr>
<td></td>
<td>10 mm</td>
<td>5 mm</td>
<td>3 mm</td>
</tr>
<tr>
<td></td>
<td>1.5 mm</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure S5
Neutral mechanical plane

Figure S6
Figure S8
Figure S9
Figure S12
Figure S14
A

5 stacks of PZT MEHs

1 cm

B
Figure S16
Figure S17
Table 1 The amplitude of the stacks for $\Delta L = 5$ mm.

<table>
<thead>
<tr>
<th>n$^\text{th}$ PZT MEH</th>
<th>Amplitude (mm)</th>
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</thead>
<tbody>
<tr>
<td>single PZT MEH</td>
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<tr>
<td>1</td>
<td>6.41</td>
</tr>
<tr>
<td>3</td>
<td>6.37</td>
</tr>
<tr>
<td>stack of 3 PZT MEHs</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>6.37</td>
</tr>
<tr>
<td>1</td>
<td>6.37</td>
</tr>
<tr>
<td>5</td>
<td>6.35</td>
</tr>
<tr>
<td>4</td>
<td>6.35</td>
</tr>
<tr>
<td>stack of 5 PZT MEHs</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>6.35</td>
</tr>
<tr>
<td>2</td>
<td>6.35</td>
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<td>1</td>
<td>6.35</td>
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</table>
Supporting Information

Dagdeviren et al. 10.1073/pnas.1317233111

Movie S1. Affixing procedure of lead zirconate titanate (PZT) mechanical energy harvester (MEH) on epicardial sites. The anchoring scheme used sutures at three points, to maintain focal contact, though without rigid attachment so as to minimize any alteration or constraint on cardiac motion.

Movie S1
Movie S2. PZT MEH on the left and right ventricles of the bovine heart.
Movie S3. Increasing heart rate with a temporary pacemaker.
Movie S4. PZT MEH on the bovine lung.

Other Supporting Information Files

SI Appendix (PDF)