High performance of polycrystalline piezoelectric ceramic-based magneto-mechano-electric energy generators

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ABSTRACT

The cantilever-structured magneto-mechano-electric (MME) energy generator with smaller volume exhibits excellent magnetic energy conversion performance than electro-magnetic current induction type energy harvesters. An anisotropic single crystal is an ideal piezoelectric constituent of a high-performance MME generator owing to its superior electromechanical properties and high electromechanical energy conversion efficiency. However, the complicated synthesis and high cost of piezoelectric single crystals limit the wide deployment of the MME generator. Alternatively, the implementation of the polycrystalline ceramic piezoelectric can largely reduce the device fabrication cost. In this work, the PMN-PZT single crystal fiber composites (SFC) were replaced by high-performance polycrystalline ceramic (MnO₂-doped 25PMN-PZT) fiber composites (PCFC) for MME generator. The magneto-electric response and harvested electrical power of SFC- and PCFC-based MME generators were measured. Interestingly, the electrical output power of the PCFC-based MME generator at 10 Oe magnetic field tuned at 60 Hz was found to be ~90% of that of its SFC-based counterpart, in addition to the superior thermal stability. The presented MME energy generator has excellent potential for applications as low cost and efficient power source for wireless sensor networks deployable to the internet of things (IoTs), low power consumption electronic devices, etc., by harvesting the stray magnetic energy.

1. Introduction

The modern era of the Internet of Things (IoTs) needs uninterrupted communications between many objects such as sensors, wireless data transmission devices, security systems, etc [1,2]. In order to provide uninterrupted communication, the electronic circuits need a continuous onboard power supply. However, the foremost challenge in the implementation of the IoTs is the appropriate power source. Though the batteries are a logical solution, they suffer from the issues such as limited lifetime, maintenance, etc. Various autonomous powering paradigms using energy harvesting technology are the prime considerations for the solution [3–9]. Among the wasted energy sources, the ubiquitous stray magnetic field around the current-carrying cables is one of the most promising sources [10,11]. However, the conventional approach to harvest this low amplitude magnetic field is based on the Faraday’s law of induction, which has very low efficiency and a device is bulky [12]. In recent years, the Magneto-Mechano-Electric (MME) energy generator has shown excellent potential for low amplitude magnetic field to electrical energy conversion. [12–17] The typical MME generator is a cantilever structure comprising a magneto-electric (ME) composite structure of piezoelectric and magnetostrictive layers and permanent magnets as proof mass [11,12]. The basic factors contributing to the output electrical performance are magneto-electric (ME) effect, magnetic flexural torque from the magnetic proof mass and their synergetic interaction under externally applied magnetic field [15].

Significant improvements in the output performance of the MME generators have been achieved in recent years by exploiting the various constituents of the device, such as employing low-loss anisotropic piezoelectric single crystal, textured giant magnetostrictive materials, the introduction of magnetic field concentrator, optimization of device dimensions, optimization of the interfacial adhesive layer, etc [13–15,18,19]. Especially, the material properties of the piezoelectric layer in MME generator plays a crucial role in its energy harvesting performance. For example, Annupreddy et al. reported that the MME generator with anisotropic piezoelectric single crystal fiber...
composite (SFC) having low dielectric and mechanical losses shows higher ME coupling and MME generator performance [18]. However, the growth and fabrication of the piezoelectric single crystals used to fabricate SFCs take a very long time and have high costs, which increases the overall device cost of MME generators though it offers excellent energy harvesting performance [20,21]. In addition, the rhombohedral to tetragonal transition temperatures of PMN-PT (Pb(Mg1/3Nb2/3)O3-PbTiO3) or PZN-PT (Pb(Zn1/3 Nb2/3)O3-PbTiO3)-based piezoelectric single crystals are typically lower than 100°C, which also limit operational temperature of the device. Therefore, instead of piezoelectric single crystals, employing appropriately designed polycrystalline piezoelectric ceramics could minimize the device cost with minimal compromise in output performance. Polycrystalline ceramics possess several advantages over single-crystal piezoelectric materials, such as ease of fabrication and tunability of composition and properties. In contrast, the single crystals need to cut along certain crystallographic directions.

It is known that acceptor (such as Mn²⁺, ³⁺, Fe²⁺, ³⁺) doping into the piezoelectric ceramic creates anion (oxygen) vacancies in the lattice, and the ceramics are termed as “hard-type ceramics”. These hard-type ceramics exhibit lower dielectric loss, larger mechanical quality factor owing to excellent energy conversion at resonant condition [20,22]. However, the piezoelectric voltage constant and piezoelectric charge constant of hard-type ceramics are smaller, which are also important factors in higher electromechanical conversion efficiency [23]. The piezoelectric voltage constant and piezoelectric charge constant of hard-type ceramics can be enhanced by donor doping. By combinatorial approach, i.e. an appropriate amount of acceptor and donor doping, one can achieve the piezoelectric ceramic with optimized energy conversion efficiency [24]. Although the piezoelectric properties of polycrystalline ceramics are lower than its single-crystal counterparts; the mechanical quality factor, which is related to electromechanical loss and dielectric loss is superior to typical PMN-PT (Pb(Mg1/3Nb2/3)O3-PbTiO3) or PZN-PT (Pb(Mg1/3Nb2/3)O3-PbTiO3)-based piezoelectric single crystals [25,26]. It is believed that a high-performance MME generator with higher thermal stability can also be utilized if the piezoelectric ceramics, having low loss with high piezoelectric properties and transition temperature if employed into the MME generator structure.

In this context, we investigated the energy harvesting performance of the MME generators composed of fiber composites based on polycrystalline 25PMN-PZT piezoelectric ceramics. The polycrystalline ceramic fiber composites (PCFC) were first fabricated using three different types of polycrystalline 25PMN-PZT ceramics, namely hard, semi-hard and soft. The MME generators were then fabricated by laminating PCFCs with a magnetostrictive Ni sheet using adhesive epoxy. The semi-hard PCFCs showed excellent piezoelectric properties, and low dielectric loss result in a superior figure of merits. The generated electrical output RMS power of the MME generator based on semi-hard PCFC was nearly 90% of well-known SFC-based MME generators. Additionally, the proposed MME generators were subjected to thermal stability tests to ensure their performance stability at adverse conditions for practical applications. The generated output power was demonstrated to drive 60 LEDs and an Internet of Things (IoT) wireless multisensor network module consisting of temperature, humidity, acceleration sensors, and a Bluetooth transmitter.

2. Experiment details

The MME generator specimen were fabricated using three different types of PCFC, e.g. Soft-type (25PMN-35PZ-40PT), Semi-hard-type (25PMN-35PZ-40PT + 0.5 wt% MnO₂) and Hard-type (25PMN-35PZ-40PT + 1.0 wt% MnO₂), polycrystalline ceramics (Ceracom Co., Ltd., Korea) as a piezoelectric layer, and Ni metal sheet as a magnetostrictive layer. The electric and piezoelectric properties of the employed polycrystalline piezoelectric ceramics have been listed in Table 1. A high purity Ni layer (99.5%, Alfa Aesar, Ward Hill, MA) was cut into trapezoidal shapes of 60 mm (L) × 20 mm (W) × 0.25 mm (t). The polycrystalline 25PMN-PZT was machined into a rectangular cuboid shape [20 mm (L) × 13 mm (W) × 0.15 mm (t)]. The obtained shapes were partitioned into fibers having dimensions of 20 mm (L) × 0.5 mm (W) × 0.15 mm (t) fiber forms. For the electrical connections, planar Au electrodes were deposited on both sides of the fibers and subsequently mounted on the Ni layer using the adhesive epoxy and laminated using polyimide film for environmental protection, as schematically shown in Figure 1(a). After the fabrication of the MME generators, the samples were poled under a 3 kV mm⁻¹ electric field at 80°C for 30 min. As the proof mass, four neodymium permanent magnets (NdFeB) having

| Table 1: Material properties of the employed soft, semi-hard, hard type polycrystalline piezoelectric ceramics compared to its single crystal counterpart. |
| Samples/ Parameters | Soft type | Semi-Hard type | Hard type |
| --- | --- | --- | --- |
| εr | (at 1 kHz and RT) | 4465 | 1611 | 1137 |
| tan δ | 0.05 | 0.006 | 0.015 |
| k33 | 0.72 | 0.73 | 0.68 |
| k31 | 0.38 | 0.40 | 0.32 |
| d33 (pC/N) | 800 | 600 | 350 |
| Qm | 100 | 450 | 800 |
| Ec (kV/cm) | 6.2 | 7.5 | 12 |
| Tg (°C) | 150 | 220 | 300 |
| g33 (10⁻³ VmN⁻¹) | 20 | 42 | 35 |
| FOM | d33²g33/tan δ | 320 | 4200 | 813 |

Experiments
dimensions 10 mm × 5 mm × 1 mm (3 g) were attached at the tip of the MME generator to induce flexural torque. For comparison, MME generator with 25PMN-PZT single crystal (sample info) was also fabricated and characterized. The geometry and dimensions were identical to the MME generator with ceramics.

The temperature-dependent dielectric measurements in a wide temperature range of room temperature (RT) to 350°C of the polycrystalline ceramics were performed using an impedance analyzer (4284A, Agilent Technologies) coupled with a heat chamber. The ferroelectric hysteresis loops of the ceramics were performed using a ferroelectric evaluation system (TF Analyzer 2000, aixACCT GmbH, Germany) at 1 Hz carrier frequency. The magnetostrictive coefficient measurement of the ME composites (without proof mass attached) was performed using the setup comprising a lock-in amplifier (SR865A; Stanford Research Systems, CA, USA), a bipolar power amplifier (HSA 4011, NF Corporation, Japan), a digital Tesla meter (Model 425, Lake Shore Cryotronics), a DC electromagnet, and an AC Helmholtz coil. The electrical output performance of the MME generators was evaluated using the computer-assisted automated setup comprising a Helmholtz coil, a digital multimeter (2700, Keithley, OH, USA) with a multichannel resistance control relay comprising multiple load resistors from 1 kΩ to 1 MΩ, and an oscilloscope (WaveSurfer 64Xs-A, Teledyne LeCroy, NY, USA).

3. Results and discussion

The structure of the fabricated polycrystalline ceramic-based MME generator has been shown in Figure 1(a). The three different types of PCFC, i.e. soft, semi-hard, and hard-type, and 25PMN-PZT single crystal fiber composite (SFC), were employed as the piezoelectric layer of the MME generator. The temperature-dependent dielectric properties of the polycrystalline piezoelectric ceramics have been evaluated in a wide temperature range of RT to 350°C and shown in Figure 1(b,c). The soft-type ceramic exhibits the highest value of the dielectric constant ($\varepsilon_r \approx 4465$ at RT), whereas the hard-type ceramic shows the lower dielectric constant ($\varepsilon_r \approx 1137$ at RT) and dielectric loss. The semi-hard-type piezoelectric ceramic exhibits the dielectric constant values ($\varepsilon_r = 1611$ at RT) in between the corresponding values of soft and hard-type ceramic. Also, the dielectric loss of the semi-hard-type ceramic is in the range of hard-type ceramics ($\approx 0.006$). The polarization vs. electric field hysteresis loops of the ceramics were measured at 1 Hz carrier frequency, as shown in Figure 2(a). The obtained data plot shows a typical behavior of the ferroelectric 25PMN-PZT ceramics, as the soft-type ceramic shows the lowest coercive electric field ($E_c$) and highest saturation polarization ($P_{max}$). The material properties of the three types of piezoelectric 25PMN-PZT polycrystalline ceramics compared with the single crystal have been listed in Table 1. The
Figure 2. (a) Polarization vs. Electric field hysteresis loops of the soft, semi-hard and hard type PCFC at 1 Hz. (b) ME voltage coefficient (α_{ME}) vs. applied DC magnetic field of the soft, semi-hard, hard type PCFC- and SFC-based MME composites measured at 1 Oe (1 kHz) AC magnetic field.

mechanical quality factor (Q_m) represents the degree of underdamping of the resonator, while \( k_{33} \) or \( k_{31} \) represents the ability to convert the applied mechanical energy to electrical energy or vice versa [20]. Although, the piezoelectric coefficient and mechanical quality factor values of the semi-hard type polycrystalline piezoceramic (\( d_{33} = 600 \) and \( Q_m = 450 \)) lie in between the soft (\( d_{33} = 800 \) and \( Q_m = 100 \)) and hard type (\( d_{33} = 350 \) and \( Q_m = 800 \)) ceramics, it exhibits excellent electromechanical coupling factors (\( k_{33} = 0.73, k_{31} = 0.40 \)) as compared to the soft (\( k_{33} = 0.72, k_{31} = 0.38 \)) polycrystalline ceramics. Similarly, the energy harvesting performance of the MME generators at the off-resonance condition can be directly related to the figure of merit (FOM) at off-resonance condition corresponding to the product of the dielectric strain constant (\( d_3 \)) and piezoelectric voltage constant (\( g_p \)) i.e. \( d_3 \times g_p/\tan \delta \) [10,18,27]. The calculated values of the FOM (i.e. \( d_3 \times g_p/\tan \delta \)) for the soft, semi-hard, and hard type of polycrystalline ceramics are \( 320 \times 10^{-12} \), \( 4200 \times 10^{-12} \), and \( 813 \times 10^{-12} \) m²/N, respectively. Therefore, the obtained superior FOM values for the semi-hard polycrystalline ceramic indicate a superior energy harvesting performance at off- and resonance conditions as compared to that of soft and hard type ceramics [28,29].

Since the ME effect is one of the main contributions in the output performance of the MME generator, we have evaluated the magnetoelectric voltage coefficient (\( \alpha_{ME} \)) with respect to the DC magnetic field (−400 Oe to 400 Oe with a superimposed AC magnetic field of 1 Oe) at off-resonance condition (1 kHz) for the PCFC-based ME composite and compared with that of SFC-based ME composite; as shown in Figure 2(b). The obtained \( \alpha_{ME} \) response of the ME composite samples exhibited a typical ME hysteresis loop. Though the SFC-based ME composite exhibited much larger \( \alpha_{ME} \) values (1.3 V/cm.Oe⁻¹) as compared to the PCFC-based ME composites, the semi-hard PCFC-based ME composite exhibited the highest \( \alpha_{ME} \) value (0.39 V/cm.Oe⁻¹) among PCFC-based ME composites.

The output energy harvesting performance of the fabricated MME generators with different PCFCs was evaluated under the influence of an AC magnetic field of 10 Oe at 60 Hz frequency applied using a Helmholtz coil. The corresponding generated AC voltage waveforms (at 1 MΩ impedance) of each type of MME generator were recorded using an oscilloscope and shown in Figure 3(a). The soft and hard type PCFC-based MME generators exhibited peak to peak voltage of ~28 V and ~52 V, respectively. In contrast, both the semi-hard PCFC- and SFC-based MME generators exhibited almost similar peak-to-peak voltages of ~65 V, which contrasts with the \( \alpha_{ME} \) values at off-resonance. It indicates that the output performance of the semi-hard PCFC-based MME generator is more dominant at the resonance condition. The RMS voltage and current (\( V_{RMS} \) and \( I_{RMS} \)) values of the MME generators at 10 Oe with respect to subjected electrical load resistance (\( R_L = 1 \) kΩ to 1 MΩ) were measured, and their corresponding RMS power was calculated, and the plots are shown in Figure 3(b,c), respectively. The RMS power was calculated using the equation \( P_{RMS} = V_{RMS}^2/R_L \). The obtained maximum \( V_{RMS} \) values for the soft, semi-hard, hard PCFC- and SFC-based MME generators were ~10 V, 17 V, 20 V, and 22 V, respectively. Similarly, the calculated \( P_{RMS} \) values of the soft, semi-hard, hard PCFC- and SFC-based MME generators were 0.9 mW, 0.8 mW, 1.75 mW, and 2 mW, respectively. Here, the output \( P_{RMS} \) of the semi-hard PCFC-based MME generator is nearly 90% of that of the single crystal-based MME generator, whereas their soft and hard PCFC-based counterparts yield very low output RMS power.

In comparison to the soft and hard type PCFC, the semi-hard PCFC has shown excellent magnetoelectric coupling and output energy harvesting performance.
Figure 3. Represents the output performance of the soft, semi-hard, hard type PCFC and SFC-based MME generators when subjected to an AC magnetic field of 10 Oe at 60 Hz applied using a Helmholtz coil. (a) Comparison of the output AC voltage waveforms with respect to time of four different MME generators. (b & c) Represents the recorded RMS voltage, current and their corresponding calculated power of the MME generators.

The obtained output performance of the MME generators is generally related to the FOM (the product of piezoelectric strain constant i.e. $d_{ij}$ and the piezoelectric voltage constant i.e. $g_{ij}$) [18,30]. However, few recent reports conclude that other factors such as mechanical loss, $\tan \theta$ (inverse of mechanical quality factor) and dielectric loss, $\tan \delta$ (inverse of electric quality factor) play a very important role in the output performance of the MME generator at resonance condition [18,30]. Therefore, minimization of these losses of the piezoelectric layer is necessary to enhance the output performance of the MME generator. The hysteresis behavior of piezoelectric material can induce intensive losses when subjected to AC vibrations. In such state, the induced $\alpha_{\text{ME}}$ at resonance condition is a crucial factor for energy harvesting performance of MME generator and can be represented as,

$$\alpha_{\text{ME}}^* = \alpha_{\text{ME}} \times \frac{1}{(\tan \theta + \tan \delta + A)}$$

where, $A = (C - C_r/C_f$), here $C$ and $C_f$ are the capacitances at a given frequency and at 1 kHz frequency respectively) is a constant for a particular frequency [30]. The employed semi-hard PCFC in the MME generator has the lowest RT dielectric loss at 1 kHz frequency. According to the obtained parameters, as mentioned in Table 1, the calculated multiplying factor $[1/(\tan \theta + \tan \delta + A)]$ at 1 kHz are nearly 16.67, 121.6, 61.8 for soft, semi-hard, and hard type polycrystalline ceramics, respectively. It can be noted from Table 1 that among the polycrystalline ceramics, the semi-hard ceramic exhibited the highest $k_t$ (thus maximum energy conversion), lowest dielectric loss ($\tan \delta$), and tuned mechanical loss ($\tan \theta$). Therefore, the obtained $\alpha_{\text{ME}}$ values and output energy harvesting performance of the semi-hard PCFC-based ME composite and MME generator were higher than the soft and hard type PCFC-based counterparts.

The obtained excellent electrical output performance of the semi-hard PCFC-based MME generator was evaluated for their real-time application deployment. Here also, the constant AC magnetic field of 10 Oe at 60 Hz was generated using a Helmholtz coil around the MME generator. The output AC voltage of the semi-hard PCFC-based MME generator was converted into DC voltage using a conventional full bridge rectifier (FBR). Then, the obtained DC voltage was applied to glow 60 LEDs. The schematic of the circuit and picture of glowing LEDs are shown in Figure 4(a,b) and ‘Supplementary Video 1’. Furthermore, the
Figure 4. (a & c) Schematic diagrams of the circuits employed for driving the 60 LEDs and charging of the 4.7 mF capacitor. (b) Photograph of the 60 LEDs illuminated using the semi-hard ceramic-based MME generator. (c) Schematic of the circuit for capacitor charging measurement. (d) Represents the corresponding recorded charging voltage vs. time plot of the employed 4.7 mF capacitor using four different type of MME generators. (e) Photograph of the demonstration circuit setup comprising semi-hard type PCFC-based MME generator, conventional full-wave rectifier, a 4.7 mF capacitor, and a WSN module.

A comparison of the output electrical energy yield from each type (soft, semi-hard, hard-type PCFC- and SFC-based) of MME generators was performed by employing a FBR and a 4.7 mF capacitor. The output AC voltage from the MME generators was converted into the DC voltage and fed to the capacitor for charging, as shown in Figure 4(c). The corresponding charging voltage values were recorded with respect to charging time, and corresponding stored energy was calculated, as shown in Figure 4(d). The plot of the calculated stored energy of the capacitor has been shown in the inset of Figure 4(d). Although the SFC-based MME generator is the fastest among all, it can be clearly noted here that the semi-hard PCFC-based MME generator charges the capacitor faster than the soft and hard type PCFC-based MME generators owing to its higher output $P_{\text{RMS}}$.

Furthermore, the most important application of the MME generators is as an autonomous onboard power supply for wireless sensor networks (WSN). In this study, the demonstration of the practical application of the semi-hard PCFC-based MME generator as the power source for a multisensor circuit (MIDASCON, Korea), comprising a microprocessor, temperature, humidity, and three-axis gyro sensors, a 2.4 GHz Bluetooth transceiver, and an RF chip antenna were performed. The output AC voltage of the MME generator was converted to DC voltage by the conventional full-wave rectifier and then employed to charge a 4.7 mF capacitor. The voltage across the charged capacitor was applied to the WSN circuit. The corresponding data WSN was received through Bluetooth on a smartphone android application. The picture of the demonstration has been shown in Figure 4(e) and ‘Supplementary Video 2’. In the inset, the magnified picture of the received data has been shown. The output power of the semi-hard PCFC-based MME generator was found to be sufficient enough to drive the WSN module without an external power source or battery. However, the output power generated from the soft and hard-type PCFC-based MME generators was not able to drive the WSN module. The presented demonstration clearly exhibits that the semi-hard type of PCFC-based MME generator has the potential to replace the SFC-based MME generators as an inexpensive onboard power source for WSN circuits.
In order to evaluate the MME generator’s performance for the practical applications, the temperature reliability tests were also performed in the temperature range of –30°C to 70°C. The employed four types of MME generators were placed in a temperature oven, and the temperature was swept from –30°C to 70°C, and corresponding impedance vs. frequency plots was measured (Figure 5(a-d)). The corresponding shifts in the resonance and anti-resonance frequencies ($f_r$ and $f_a$) of the MME generators in the subjected temperature range have been shown in Figure 5(e-h). The PCFC-based MME generators exhibit typical behavior, as soft-type PCFC-based MME generator exhibits the highest shift in $f_r$ and $f_a$ whereas the hard-type PCFC-based MME generator exhibits the lowest shift. The shifts in $f_r$ and $f_a$ in the semi-hard PCFC-based MME generator were nearly 1.5 Hz and 1.75 Hz, respectively, which is in less range than that of SFC-based MME generators (1.8 Hz and 1.7 Hz, respectively) attributed to their higher transition temperature. Therefore, it indicates that the semi-hard e PCFC-based MME generator has excellent potential to replace its single crystal-based counterparts for applications of autonomous power source for IoT-based sensor modules.

4. Conclusion

In this study, the three different types of polycrystalline ceramic fiber composites (PCFC), i.e. soft, semi-hard and hard type, are employed as the piezoelectric layer in the MME generator, and their output performance was evaluated in comparison to the single-crystal fiber composite (SFC) based MME generator. The output energy harvesting performance ($P_{RMS} = 1.75$) of the proposed semi-hard PCFC-based MME generator was nearly 90% of that of its single crystal counterpart. Among all the employed polycrystalline ceramics in the MME generators, the semi-hard ceramic exhibited the highest $k_p$ (thus maximum energy conversion), lowest dielectric loss ($\tan \delta$), and tuned mechanical loss ($\tan \theta$). It eventually explains the obtained highest output electrical performance of the semi-hard polycrystalline-based MME generators. Furthermore, it provides an inexpensive onboard power source for multi-functional WSN modules. Additionally, a successful demonstration for the application of the proposed MME generator as the power source to 60 LEDs and a WSN module comprising a Bluetooth transceiver was shown. For the practical applications, the temperature reliability of the proposed MME generators in the temperature range of –30°C to 70°C was investigated and confirmed that higher thermal stability of PCFC-based MME generators. Therefore, the optimized polycrystalline ceramic material as a piezoelectric layer in the MME generator exhibits an excellent potential for its applications as an autonomous onboard power supply for IoT-based sensor circuits.

Disclosure statement

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