論文の内容の要旨

MEMS-Based Piezoelectric Polymer Electrets for Energy Harvesting

(MEMS 技術を用いた環境発電のための圧電ポリマー・エレクトレット)

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Piezoelectric transducers are widely used in the microelectro-mechanical system (MEMS) transducers such as inertial sensors, actuators, and energy harvesters. Lead zirconate titanate (PZT) thin films prepared with sol-gel or sputtering methods could offer high piezoelectric constants (d_{33}~360 pC/N), whereas the resonant frequencies of the MEMS piezoelectric devices are often very high due to their large Young's moduli. Soft piezoelectric polymers, such as polyvinylidene fluoride (PVDF), have been widely investigated, but their piezoelectric responses are somewhat limited to low values (d_{33}<35 pC/N). Recently, cellular polymer electrets, which contain a large number of micro-scale voids with implanted charges, have attracted much attention due to their low effective Young's moduli, tolerances for large deflection, and high electromechanical sensitivities

In previous studies, porous structures are formed in solid films by thermal expansion or physical foaming methods, which result in random distribution of ellipsoidal voids with different dimensions. Although inflation and stretching processes are often employed to optimize the void heights and thus the porosity of the film, somewhat broad distribution of the void dimensions would result in higher stiffness of the structure and/or nonuniform polarization of the polymer film through dielectric barrier discharges in the cavities with different heights. In addition, previous cellular polymers exhibit relatively poor structural stabilities. Under external forces, high-pressure gases inside the voids could leak to the outside, which leads to a decrease of the film thickness and deterioration of the piezoelectric response. Cellular structures with only a few layers, such as cellular polydimethylsiloxane (PDMS), can be made by thermal molding or bonding polymer substrates with cavities. However, it is not straightforward process to bond many layers firmly without crushing voids at an elevated temperature around the glass transition temperature.

Since most investigations focus on one-face charged single electret, few reports on heterocharges on double-faces charged electret and feasibility of charging multi-layer electrets. For one-face charged single film, either positive or negative charges could be successfully implanted into the electret film, and for some sort of polymer electrets, the negative charge implanted into the electret film is more stable, than the positive one. This fact has already reached a consensus for homocharges property, but still few works have been made to investigate the stability of positive and negative charges (heterocharges) in the dipole unit. The fundamental experiments and theoretical analyze should be carried out to well investigate electret properties for cellular electret.

Based on the above consideration, the objectives of present study are

- To confirm the fundamental properties of heterocharges in single/multi-later electrets and feasibility of soft X-ray uniformly charging multi-layer electrets in series.
- To propose a novel microfabricated high-aspect-ratio and high-density cellular polymer electret based on trench-filled parylene technology.
- To design and establish an in-plane MEMS piezoelectret transducer based on parylene piezoelectret spring to sense low-frequency vibration.
- To propose an improved high-performance piezoelectret with embedded PEDOT electrodes for vibration-driven energy harvester.

Firstly, the theoretical model of homocharges and heterocharges are compared. The relative low external electric field in heterocharges electret might own better stability than homocharges electret, which is confirmed by the experimental data. The nearly equal positive and negative surface potentials in double-faces charged single electret film are observed, which demonstrates the feasibility of soft X-ray charging single electret with heterocharges. On the other hand, two parallel electret films can be charged in series and show the similar surface potentials, which should become the strong evidence to confirm the feasibility of the soft X-ray charging multi-layer electrets.

A novel microfabricated high-aspect-ratio cellular polymer electret based on trench-filled parylene process is proposed, which could solve the long-term stability or reproducibility and charging issues introduced by thermal expansion or physical foaming preparation methods. In order to realize good mechanical property and electret performance, we employ parylene-C as main structural material and also a thin Dix-F layer to improve the electret performance. Both of parylene-C and Dix-F would offer a conformal coating even in deep Si trenches. After releasing the structure, the free-standing cellular parylene structure is obtained.

Based on trench-filled parylene process, the MEMS-based piezoelectret transducer is developed, which enable in-plane oscillation with a low resonant frequency. To realize the macroscopic dipoles inside the high-aspect-ratio cellular parylene structures, charges should be implanted onto the vertical walls of the cellular structures to form electret. Since corona ions cannot be used for charging through the narrow gap due to charge built-up near the opening, we employ the soft X-ray charging, in which in-situ photoionization of air molecules inside the gap using soft X-rays. When the bias voltage is applied across the gap, the abundant positive and negative charges are accelerated and implanted onto the opposite vertical parylene. The uniform artificial dipoles moments could vary along with parylene structural deformation driven by the inertia of a seismic mass.

With respect to the in-plane resonant oscillation, the prototype could provide the acceleration sensitivity from 152 to 197 mV/*g* at the low frequency of 30 Hz, and the piezoelectric charge and voltage sensitivities of 1522 pC/N and 152 V/N could be obtained at 1 *g* acceleration. At the resonant frequency of 149 Hz, the peak to peak voltage of 4.8 V was obtained, corresponding to the sensitivities of as large as 9600 pC/N and 960 V/N, respectively.

As an early power generator, the output power of 78 nW could be obtained at the resonant frequency of 149 Hz with 1.5 *g* acceleration. These results demonstrate the advantages of the cellular electret based on the trench-filled parylene for in-plane low-frequency transducers.

We also propose an improved cellular structure with embedded PEDOT electrodes for higher surface charge density. PEDOT conductive polymer is infused into the parylene spring by the capillary force. In order to electronically isolate positive/negative electrodes yet to mechanically connect parylene springs, short parylene beams with narrow isolation structure are devised to avoid the surface-tension driven PEDOT flow in between the positive and the negative sides. The good robustness of the PEDOT electrodes is confirmed by the oscillation experiment.

Thus, the bias voltage up to 150 V could be directly applied on the each parylene spring, which is expected to increase the power output to several μW.