Demonstration of an electret generator using self-assembled electret for energy harvesting without any charging process

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Abstract. Electret generators (EGs) for energy harvesting is a device which ambient vibrations convert into electric energy. They are expected as power supplies for low energy applications such as wireless sensors. In EGs, charges are induced by an electric field of electret, and thus output power is determined by a surface charge density ($\sigma$) of the electret. Various charging processes were proposed so far to realize the electret with high $\sigma$, however, they were one of problems decreasing productivity. The elimination of the processes is extremely desired. In this study, we developed charged film composed of 1,3,5-tris(1-phenyl-1H-benzimidazol-2-yl) benzene (TPBi) without any charging process. A surface potential (SP) exceeded 46 V for ca. 740 nm thick film due to spontaneous orientation of TPBi, and was sufficiently stable under room light illumination in atmosphere. Finally, we demonstrated EGs utilizing TPBi as electret generated an AC current of the order of nano-ampere in environmental condition, indicating the feasibility of EGs using TPBi film in practical atmosphere. The series of results strongly suggested that application of spontaneous orientation of polar molecules was promising for the realization of EGs with high productivity.

1. Introduction
In recent years, energy harvesting which can convert vibrations into electrical energy has attracted much attention as power supplies for low power devices such as wireless sensors for health monitoring, home/building energy management systems (HEMS/BEMS) and livestock industry \cite{1, 2}. The frequency of vibration, e.g. human motion or mechanical vibration, existing in environment is usually below 100 Hz. Thus, vibration-driven electret generators (EGs) which obtain a large power output for low frequency are suitable for implementation \cite{1-5}.

Generally, EGs consist of a capacitor structure, in which an electret and air gap are sandwiched by top and bottom electrodes as shown in Fig. 1(a) \cite{2-5}. Electret is a dielectric material with a quasi-permanent electrical charge or dipole polarization \cite{6}, and thus charges are induced in the electrodes of EGs as shown in Fig. 1(a). When vibration is applied to the electrode, current is generated due to change of the amount of the induced charges. An output power of EGs depends on the surface charge density of electret ($\sigma$). Therefore, various charging processes, for example, corona charging, poling, contact electrification or particle beam irradiation have been proposed in order to make electret with high $\sigma$ \cite{2, 6}. These processes are, however, one of the factors decreasing productivity because of complexity.
In a vacuum evaporated film of tris-(8-hydroxyquinolinato) aluminum (Alq₃) which is a typical molecule for organic light-emitting diode (OLED), our group found that surface potential (SP) of the film linearly increases with the film thickness, reaches 28 V for 560 nm [7, 8]. The σ can be estimated to be 1.4 mC/m² assuming the relative permittivity of 3.2 [9]. Such giant surface potential (GSP) appears because of spontaneous orientation of dipole moment of Alq₃ as shown in Fig. 1 (b). Sugi et al. reported that the SP of Alq₃ was held for a long time (10% loss in 10 years) [10], suggesting that Alq₃ can be regarded as electret. So far, various materials showing GSP have been found [9, 11-14]. Because no charging process is necessary for these charged films, hereafter, they are referred to as self-assembled electret (SAE). By applying SAE to EGs, it is expected that the charging processes can be eliminated, leading to simplification of manufacturing process.

One of problems in utilizing SAE materials including Alq₃ as electret is the stability of SP in environment. In fact, SP declines by light irradiation because generated photocarriers cancel the polarization charges [7-9]. In order to decrease visible light absorption of SAE, in this study, we adopted 1,3,5-tris(1-phenyl-1H-benzimidazol-2-yl) benzene (TPBi) which has a larger optical band gap than Alq₃. We found that SP exceeds 46 V of 739 nm, indicating the formation of charged film without charging process due to orientation polarization. In addition, the SP was stable under room light illumination in atmosphere. By utilizing these advantages of TPBi SAE, the model EGs without any charging process was developed. A current in the order of nano-ampere was generated due to the vibration in environmental condition, suggesting the TPBi SAE is suitable for EGs used in atmosphere.

2. Methods
TPBi was purchased from Lumtec (sublimed, >99.5% purity). SP of TPBi was measured by the Kelvin Probe (KP) (KP technology, model: UHVKP020) in vacuum and atmosphere. TPBi was deposited on an ITO substrate in an incremental manner, with KP measurement at each step of deposition. The thickness of TPBi film was monitored by a quartz microbalance. The rate of deposition was 0.5 Å/s until 200 nm and 2.0 Å/s from 200 nm to 739 nm. To eliminate the effect of light, the vacuum gauge in the chamber was turned off and all viewing ports were covered by aluminum foil. A flashlight with red filter was used only when we moved the sample by transfer rod in the chamber. For the measurements under illumination, room light was irradiated to the side of the film through a viewing port.

We fabricated a model EG with a structure of Au (bottom electrode) / TPBi (500 nm) / air gap (1.1 mm) / Au (top electrode). The device performances were evaluated in room light and the atmosphere. Both top and bottom electrodes were deposited by vacuum evaporation on a silicon wafer covered with
SiO$_2$. The top electrode was vibrated by a shaker with 30 Hz (frequency) and 1 mm$_{p-p}$ (amplitude), and the output voltage through the external resistance (1 MΩ) was observed by oscilloscope (Tektronix, model: TDS 2001C).

3. Results and Discussion

Figure 2(a) showed that the film thickness dependence of the SP during the deposition of TPBi on the ITO substrate in the dark condition. The SP linearly increased with the film thickness and reached 46.5 V at the 739 nm. This result clearly suggested that a charged film was formed without any charging processes due to the orientation polarization of TPBi molecules. From Fig. 2(a), the electric field of TPBi film was 72.7 mV/nm. Considering the relative permittivity of TPBi is 2.87, $\sigma$ was estimated to be 1.8 mC/m$^2$. Kashiwagi et al. reported a high surface charge density of more than 2.0 mC/m$^2$ by using CYTOP which is one of the amorphous perfluorinated polymer, the $\sigma$ of TPBi-based SAE would be comparable to that of the conventional electret materials.

Figure 2(b) showed time dependence of SP under room light illumination in atmosphere. This measurement was performed after the intense white LED irradiation for 1 hour in vacuum; the SP was decreased to 35.6 V. As shown in Fig. 2(c), the emission from the white LED contained some contribution of UV light (the tail part at 250-400nm in Fig. 2(c)). Although TPBi has a large optical band gap (3.5 eV, 354nm), the photocarriers due to the UV light canceled the polarization charges. In atmosphere under room light illumination including weak UV light from a fluorescent lamp, the SP showed a very slow decay up to 100 min (2.6 % decay). After ca. 2700 min, the SP was gradually

![Figure 2](image-url)

Figure 2. (a) The development of the SP upon the deposition of TPBi in dark in vacuum. (b) The SP of TPBi SAE as a function of time in environmental condition. (c) Emission spectrum of white LED light.
decreased to 31.9 V (6.3 % decay). These results suggested that the possible reduction of SP during the fabrication process of the EGs is negligible. If the light illumination to the electret layer is suppressed (it is practically feasible assumption), much longer life time can be expected. In this way, the favorable stability of SP in TPBi film was achieved, resulting from the wider optical gap of TPBi than Alq3. If we use SAE materials with larger optical band gap than that of TPBi, the stability can be further improved.

By taking advantage of TPBi film, the model EG without any charging process was prepared. Figure 3 indicated the output current of EG with TPBi SAE as a function of time. This measurement was performed after the device was kept in atmosphere under room light illumination for ca. 14 h. An AC current of 0.9 nA was clearly observed. In addition, the frequency of the current was same as the vibration frequency of the shaker. These results clearly suggested that TPBi based EGs operated well even in atmosphere. We believe that application of SAE to electret enables the simplification of fabrication process, leading to the realization of EGs with high productivity.

4. Conclusion
By using KP method, we observed spontaneous buildup of GSP upon the vacuum deposition of TPBi in dark condition. The SP of TPBi film reached 46.5 V at the 739 nm, and $\sigma$ was estimated to be 1.8 mC/m² which is comparable to that of conventional polymer-based electret. In addition, the SP of the film had favorable stability in atmosphere under light illumination. We succeeded in fabricating the charged film prepared without any charging process due to spontaneous orientation of TPBi molecules.

By utilizing these advantages of the film, we fabricated the model EGs including the TPBi SAE. An AC current of 0.9 nA was obtained by vibration in atmosphere under light illumination, demonstrating that the EGs without charging process work well by introducing SAE even in atmosphere. These results show that SAE materials are promising for the realization of EGs with high productivity.

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