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Graphene embedded form stable phase change materials for drawing the thermo-electric energy harvesting



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ABSTRACT

We introduce a reusable energy harvesting system that can recover discarded thermal energy by utilizing temperature variation of the environment. The system consists of two different phase change materials (PCMs); poly ethylene glycol (PEG) and 1-tetradecanol (1-TD), and a cell of N and P type semiconductor. Since a large amount of heat energy is absorbed during phase transition, PCMS are generally utilized to maintain isothermal temperature on the opposite sides of the N and P type semiconductor as long as possible. The resulting temperature difference between two sides of semiconductor induces an electric current during the heating and cooling process. It is important for the phase change material (PCM) to possess a good shape stability without significant loss of latent heat. A 3D porous graphene aerogel is selected for the pure phase change material to infiltrate into the aerogel and to increase the shape stability and thermal conductivity of the PCM composite. The thermal conductivity of PEG and 1-TD composites is significantly increased to 0.4268 W/m·K, and 0.3408 W/m·K, respectively. Thermal and electrical analyses are performed to predict the energy harvested by the device. The electric current reaches 10 mA in both heating and cooling processes and an LED lamp was turned on successfully. Additionally, the energy harvesting system is modelled by using the finite element method (FEM) and the numerical prediction is in good agreement with the experimental results.

1. Introduction

Finding a new energy source is our prime mission because of upcoming energy depletion on earth. A renewable energy is certainly required to replace the fossil fuels and reduce the energy crisis. Development of a sustainable energy such as solar energy, wind energy, and heat recovery energy is one of the hot issues in both science and engineering fields. In particular, efficient energy harvesting has attracted a lot of attention in recent years and the thermo-electric energy conversion device which is able to convert the thermal energy to the electric energy has been widely used for energy harvesting [1,2]. Energy storage plays a considerable part in absorbing heat energy for thermo-electric energy conversion. Thermal energy storage (TES), one of the most popular systems for recycling energy, generally employs a phase change material (PCM) as a working material due to its high latent heat, appropriate phase transition temperature range, and high thermal stability [3,4]. The latent heat thermal energy storage (LHTES) is a good example of PCMs applications and the energy storage density can be improved by combining these PCMs with thermo-electric devices

[3,5].

The PCMs can absorb or release a great deal of thermal energy isothermally during the heating and cooling process [6]. Since PCMs have the advantage of high latent heat, they have been applied to the energy field systems including solar cell utilization [7], smart textiles [8], energy storage building materials [9], and wasted heat energy utilization [10]. From the category of phase change materials (PCMs), the solid-liquid PCMs are generally studied in the energy storage system due to the high latent heat under the solid-liquid phase transitions [11,12]. Since solid-liquid PCMs, however, have a leakage problem during the melting and cooling process, the field applications are restricted [13]. To manufacture the form-stable PCMs, some supporting materials are required to improve the dimensional stability of PCMs. In recent decades, various supporting materials such as urea-formaldehyde (UF) [14], polyurethane (PU) [15], polyethylene (PE) [16], and polyaniline (PANI) have been used to retain PCMs with a core-shell composite structure [17]. Since a large portion of the working material has been replaced with the supporting shell material, the overall latent heat of the composite is reduced. As a result, the conventional PCM

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composites have smaller energy storing capacity than the pure PCMs during the phase transition [18,19].

In order to increase the weight fraction of pure PCMs in the final composites, utilizing 3D porous materials as a supporting material has been reported [20]. Compared with conventional supporting materials, the 3D porous materials have larger pore volume than shell materials to be filled up by the pure PCMs [21]. One of the carbon aerogels, graphene aerogel has been investigated as a supporting material to prevent the leakage of PCMs due to its large surface area and complicated internal structure [22]. Compared with other supporting materials, the graphene aerogel has a number of advantages, such as good mechanical properties and thermal stability [23]. The PCM composites stabilized by employing the graphene aerogel can store large thermal energy and then convert the thermal energy to electric current [24]. It is important that the PCMs have high thermal conductivity for the energy conversion efficiency. The thermal conductivity of the supporting materials is controlled by modifying their internal structure including the pore size and porosity [25].

Thermo-electric energy harvesting system which is composed of the shape-stabilized PCMs and semiconductors has shown a great potential in various applications [26,27]. For the energy harvesting system, N and P type semiconductors are required to connect the PCMs for construction of an electrical circuit [28,29]. The N and P type semiconductors including thermo-couples utilize two different effects for energy harvesting; Seebeck effect and Peltier effect [30]. The Seebeck effect induces electrons to move from hot side to cold side of the energy harvesting device by absorbing the heat energy and the Peltier effect results in a temperature difference between two side planes by using the supplied electric power. Recently, Jiang et al., [31] prepared an energy harvesting system to generate the electric current by placing PCMs on the hot side and a heat sink (e.g., ice water) on the cold side of N and P type semiconductor. However, the electric current was obtained only during the cooling stage after removing the heat source, i.e., when releasing the stored energy from the PCM. On the basis of former research, modified the energy harvesting system with two different form stable PCMs placed on the sides of N and P type semiconductor to achieve the thermo-electric energy harvesting during the phase transition process. The environmental temperature was only adjusted to examine the harvested energy and efficiency.

In the present study, we designed and fabricated a reusable energy harvesting device by combining two phase change materials (i.e., PEG and 1-TD) with N and P type semiconductor. Since PEG and 1-TD had different phase transition temperature, a temperature difference between two sides of the semiconductor was generated on both heating and cooling process. In order to prevent the leakage of PCMs, graphene nano-platelet (GNP) embedded graphene aerogel was used as a supporting material. Electric current was measured and an LED lamp was lit by using the output electric energy. In addition, the thermo-electric energy of the device was simulated numerically to provide a meaningful understanding of the energy harvesting system.

2. Experimental

2.1. Materials

Polyethylene glycol (PEG, 6000 Mw) was purchased from Avention[®] corporation, and 1-tetradecanol (1-TD) was supplied by Sigma-Aldrich. To reinforce the mechanical properties of graphene aerogel, GNP (C grade) purchased from XGScience was used. Graphite flakes, sulfuric acid (H₂SO₄), hydrochloric acid (HCl), potassium permanganate (KMnO₄), and hydrogen peroxide (H₂O₂), and hydrazine were also supplied by Sigma-Aldrich. The cell of N and P type semiconductor used in this study had a dimension of $4 \times 4 \times 0.3$ cm³.

2.2. Preparation of GNP embedded graphene aerogel

Graphene oxide (GO) was synthesized by using modified Hummers' method. Firstly, 3.0 g of graphite was added to 12 ml sulfuric acid (H₂SO₄) for pre-oxidation. The pre-oxidized graphite was stirred with 15 g KMnO₄ in an oil bath at 35 °C for 2 h. Then, a small amount of H₂O₂ was poured into a deionized (DI) water diluted mixture to remove an excess of KMnO₄. After washing with 10% HCl solution, the dilution with DI water was repeated to neutralize the graphite oxide solution and graphene oxide powder was obtained via freeze-drying. GNP embedded graphene oxide aerogel was fabricated by using the freezedrving method. The graphene oxide (GO) powder and GNP particles were dispersed in DI water under ultra-sonic excitation with a 2:1 mass ratio. The suspension was poured into a mold with a size of $4 \times 4 \times 0.5$ cm³ and freeze-dried for 2 days. After that, the hydrazine vapor reduction method was employed to prepare the reduced graphene oxide/GNP aerogel. Firstly, the 3D porous graphene oxide (GO) aerogel was placed on the petri dish and hydrazine was added to the dish. The hydrazine solution was transferred to the small vial bottle and then the temperature was maintained at 120 °C. The hydrazine was evaporated to obtain the reduced graphene oxide (GO) aerogel successfully. After 72 h, the sealed system was removed and the color of aerogel became totally black. As the result, the reduced graphene oxide (rGO) aerogel was generated by the hydrazine vapor method.

2.3. Preparation of PCM based energy harvesting system

The PCM composites were fabricated by using the vacuum-infiltration method, which allowed the molten PCM to fill pores of the graphene aerogel. Before the infiltration, PEG and 1-TD were placed on each of petri dish and then put into the vacuum oven which adjusted the temperature at 80 °C to make the liquid state. After vacuuming for about 30 min to remove moisture, the graphene aerogels were immersed in the PEG and 1-TD melts and kept for 6 h in the vacuum state. The PEG and 1-TD composites were successfully fabricated by the solidification method and selected to construct the energy harvesting system. The schematic of energy harvesting mechanism is shown in Fig. 1. The stabilized PCM composites containing the working materials, PEG and 1-TD were placed on each side of the N and P type semiconductor respectively, inducing the electron movement due to temperature difference. As temperature increases, the 1-TD composite first experiences the melting transition while the temperature of PEG composite kept increasing due to its higher melting temperature (T_m) than that of 1-TD. Therefore, the temperature difference between the two sides of the N and P type semiconductor was developed. Electrons in the N type semiconductor are excited from valence band to conduction band and move to the hot side of this semiconductor. Subsequently, the electrons move to the P type semiconductor, thus inducing the electron-to-hole movement in the closed circuit.

2.4. Characterizations

Fourier transform infrared spectroscopy (Varian 660, USA) and Xray photoelectron spectroscopy (AXIS-His, KRATOS, UK) were used to identify the reduction of graphene oxide (rGO). Hydrophilic properties of the rGO were characterized by using a contact angle goniometer (Attension® Theta Lite, Biolin Scientic, Sweden). The morphology of the GNP embedded graphene aerogel and PCM composites was observed using a field-emission scanning electron microscope (Fe-SEM, Merlin compact, ZEISS, Germany) at a 5 kV accelerating voltage. The PEG/ Aerogel composites and 1-TD/Aerogel composites were coated by gold before the FE-SEM characterization. X-ray diffraction (XRD New D8, Bruker, Germany) can detect the intrinsic peaks of PEG and 1-TD with the scanning angle (2 θ) from 10° to 70° at a speed of 3° min⁻¹. To measure the phase transition temperature and latent heat, a differential scanning calorimeter (DSC4000, PerkinElmer, USA) was used in the Download English Version:

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