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Ultrafast and efficient photothermal conversion for sunlight-driven thermal-electric system

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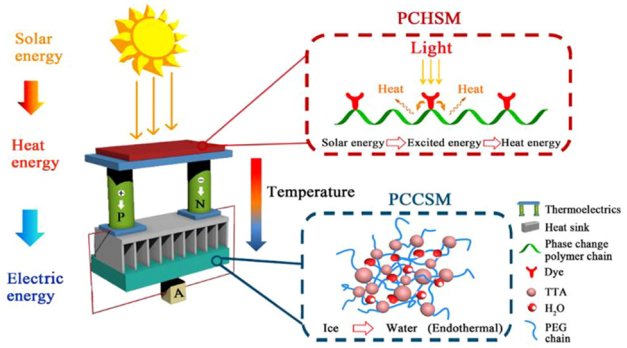
HIGHLIGHTS

- A new sunlight-driven thermoelectric system was constructed.
- Temperature difference was controlled based on the phase-change temperature control characteristics.
- The photothermal conversion mechanism of hot-side material PCHSM was investigated.
- The cold-side material PCCSM can render the cold side with stable low temperature.

GRAPHICAL ABSTRACT



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ABSTRACT

Thermoelectric battery is effective in converting thermoelectricity based on Seebeck effect. As an all-solid-state energy conversion model, this battery is highly significant in improving the utilization of solar energy using a clean and noiseless process. However, efficient photothermal conversion and temperature difference control are the key challenges in enhancing solar-thermal-electric conversion. Herein, we constructed a novel sunlight-driven thermoelectric system with temperature difference control characteristic. In which the light-driven phase change heat storage material (PCHSM) was used as the hot side, and the phase change cool storage material (PCCSM) as the cold side. Ultrafast photothermal conversion of the PCHSM was achieved through the non-radiative decay of the excited state, which was revealed by means of femtosecond transient absorption technique. Solar radiation was efficiently utilized as shown by the excellent visible light absorption, ultrafast photothermal transformation, and phase change heat storage characteristics of the PCHSM. The PCCSM with high phase change cool storage performance could provide enough cold energy to maintain the cold-side temperature. Temperature gradient was controlled using the cold-side and hot-side materials based on the phase-change temperature control characteristics.

1. Introduction

Energy is the cornerstone of human survival and development, environment is the storage place of materials and energy for human existence. Fossil energy is currently the most important source of energy globally, but its exhaustion crisis and resulting serious environmental pollution have become major challenges [1–3]. Delightedly, solar energy can be an effective solution to the energy shortage and

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environmental pollution, because it is universal, abundant, and clean [4–9].

Transforming solar radiation to electric energy is important to utilize solar energy. This process mainly includes photovoltaic [10–13] and solar thermoelectric generator (STEG) [14–16]. Based on the Seebeck effect, STEG utilizes the temperature difference to convert thermal energy to electric energy [17–21]. The thermoelectric generation device involves an all-solid-state energy conversion without chemical reaction or fluid medium, resulting in the absence of noise, wear, and medium leakage during power generation [22–24]. However, thermoelectric power generation depends on stable temperature difference to convert the thermoelectricity [25–27]. The energy density of the solar radiation reaching the earth's surface is low, and fluctuated by the conditions of season, climate, time and latitude, solar radiation is thin, discontinuous and unstable [28,29]. Moreover, the direct thermal effect of visible light (43% of solar radiation) is very low [30–33]. So it is difficult to use solar energy as heat source directly in solar thermoelectricity. Therefore, constructing a highly efficient sunlight-driven STEG system remains a great challenge. Phase change material (PCM) is a kind of energy storage material, which has advantages of high heat storage and strong temperature control capacity [34–37]. In recent years, our group and many researchers has devoted to high efficient photothermal conversion phase change materials (PCMs) [38–44]. Carbon fiber, Ti_4O_7, dye, expanded graphite, graphene foam and graphene oxide were all utilized as photothermal conversion agent and obtained relatively high efficiency. Dye can be combined with materials by chemical bond, which has no effect on the mechanical properties of materials. So in this work, we choose dye as photothermal agent for PCMs.

Herein, we constructed a new sunlight-driven thermoelectric system (Scheme 1). Given the ultrafast photothermal conversion of the non-radiative decay of the excited state of a dye, solar-thermal energy conversion and thermal energy storage were effectively achieved using the hot-side material through visible-light-driven reversible phase transition (Scheme 1, phase-change heat storage material (PCHSM)). Meanwhile, the cold-side material displayed high phase change cool storage and form-stable properties (Scheme 1, phase-change cool storage material PCCSM)). Based on the phase change temperature control characteristics, the hot-side and cold-side materials provided a relatively stable temperature difference for the thermoelectric device to realize thermal-electric energy conversion.

2. Experimental

2.1. Materials

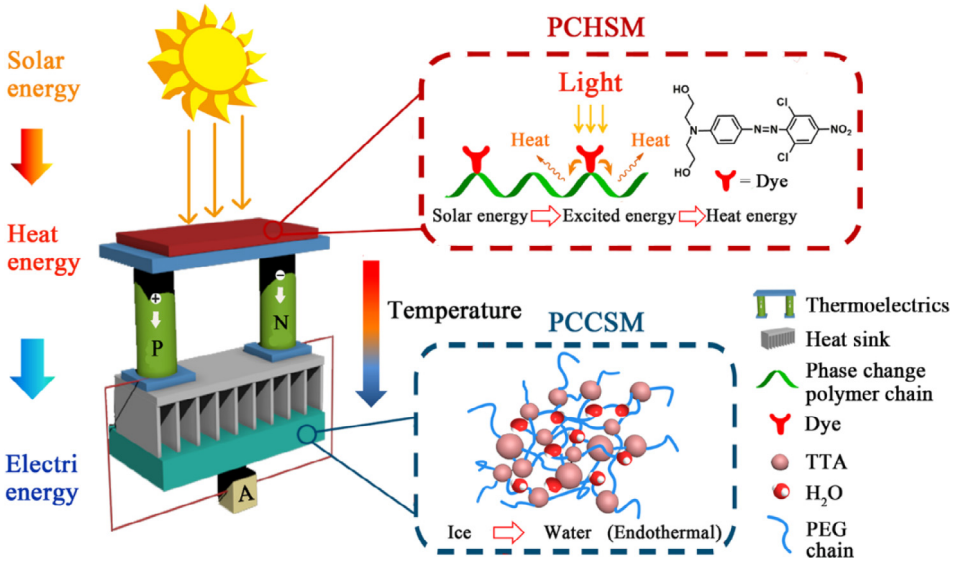
Polyethylene glycol (PEG, Mn = 10,000, Tianjin Guangfu Chemical Reagent Co., Ltd, China) and N,N-Dihydroxyethylaniline (Tianjin Kemiou Chemical Reagent Co. Inc) were dried at 80 °C in vacuum drying oven for 24 h before using. Analytical-grade toluene (Sinopharm Chemical Reagent Beijing Co. Ltd) was dried using a 5 Å molecular sieve prior to use. Toluene diisocyanate (TDI, Sinopharm Chemical Reagent Co., Ltd, China), 2,6-dichloro-4-nitroaniline (Aladdin chemicals Co., Ltd), 1,5-naphthalenedisulfonic acid (Aladdin chemicals Co., Ltd), *tert*-butyl nitrite (Aladdin chemicals Co., Ltd), 27 wt% tris (4-isocyanatophenyl) methane in ethylacetate (TTI, Zhengzhou Alfachem Co., Ltd, China), and dibutyltindilaurate (DBTL, Tianjin Kemiou Chemical Reagent Co. Inc) were used as received. All other reagents employed in this study were of analytical grade.

The thermoelectric device is commercially available, from Peltier Co., Ltd, China. The p and n type thermoelectric legs are made of antimony doped bismuth telluride and selenium doped bismuth telluride, respectively.

2.2. Synthesis of PCHSM

The synthesis of the PCHSM is illustrated in Fig. S1. First, 5 mmol 2,6-dichloro-4-nitroaniline, 5 mmol 1,5-naphthalenedisulfonic acid and 8 mmol *tert*-butyl nitrite were used to prepare the diazonium salt using a reported method [45]. Then, 0.9 g of N,N-dihydroxyethyl aniline was dissolved in hydrochloric acid solution in an ice-water bath, and the diazonium salt aqueous solution was added dropwise. The pH was adjusted to 3, and suction filtration and washing was conducted after 4 h of stirring (Fig. S1a). The dye was dried at 70 °C in vacuum drying oven for 12 h for the subsequent reaction. The appearance of molecular ion peak at m/z $[M+H]^+$ = 399.06 in the mass spectrum (Fig. S2) and the FT-IR spectra of the dye indicate that the structure was according to our expectation (Fig. S3a).

Dried PEG10000 (30 g, 3 mmol) was dissolved in toluene at 60 °C. TDI (1.14 g, 6.5 mmol) and 10 drops of DBTL were injected and stirred at 40 °C for 6 h. Then, the prepared dye in the previous step was added, and the reaction was continued for 9 h at 90 °C (Fig. S1b). The toluene solution containing the PCHSM was evaporated in a vacuum, and the material was dried at 80 °C in a vacuum drying oven for 12 h. The appearance of the N−H and C=O stretching vibrations in the FT-IR



Scheme 1. The train of thought of this work.