



Communication

Electricity generation from water droplets via capillary infiltrating

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ABSTRACT

Electricity generation from the interaction between water and carbon materials is a new approach in the pursuit of high-efficiency energy conversion. Here, we report that a water droplet dropping onto a piece of porous carbon film can spontaneously generate electricity under ambient conditions. Twelve 5- μL water droplets can generate a voltage up to 5.2 V and illuminate a liquid crystal display. The voltage can be controlled by modulating the direction of the droplet infiltration, the zeta potential of the porous carbon, and the ion concentration. The results demonstrate that a hydrophilic porous carbon film with water droplets may function as a cost-effective electricity generator for harvesting energy from natural resources.

1. Introduction

Harvesting energy from the environment offers great promise in the application of self-powered devices and systems [1]. In the past few decades, intensive studies in this field have inspired an explosive growth of environmental energy utilization approaches [2–13]. Ideally, highly effective energy conversion processes should be environmentally friendly and spontaneous while not requiring any external energy input, such as mechanical movement, concentration difference or temperature variation.

Electricity generation from the interaction between water flow and a carbon nanomaterial is a new approach in the pursuit of high-efficiency energy conversion and represents an interesting topic for both basic research and practical application viewpoints [14]. In 2001, Král and Shapiro first theoretically predicted that metallic carbon nanotubes (CNTs) in a flowing ionic liquid could generate a net potential difference and an associated electric current [15]. Since then, numerous researchers have reported voltage generation phenomena in which the detailed results differ with the carbon nanomaterials [16–25], the ion content of the flowing water, and the device configuration [26–28]. However, the reported voltage is typically in the range of several of microvolts to tens of millivolts, which is not adequate for modern devices. For example, the generated voltage is $\sim 9\ \mu\text{V}$ –30 mV and $\sim 8\ \text{mV}$

for ionic solution flows outside [16–18] of and inside of CNT [19], respectively. While, the generated voltage is $\sim 50\ \mu\text{V}$ to 85 mV for water or ionic solution flows over graphene [20–25]. Furthermore, as previously reported, external pressure or force is necessary to drive the fluid flow, i.e., the energy harvesting process consumes energy.

Recently, we have demonstrated that natural evaporation can drive the capillary water flow within porous carbon sheet, thus can produce significant voltage up to 1.0 V with lifetime over hundreds of hours under ambient conditions [41,42]. To deeply understand to underlying mechanism, we systematically studied electricity generation through the transformation of water droplets into water flow via the capillary force produced by a hydrophilic porous carbon film (PCF). A water droplet with volume of 1 μL can reliably generate a voltage of up to 0.8 V during the spontaneous filtration process of the water in the PCF. On the basis of the dependence of the induced voltage on the direction of the droplet infiltration, zeta potential of the porous carbon and ion concentrations, the phenomenon may be attributed to the electrokinetic effect [29–32]. In addition, a self-powered sensor for in situ detection of water-droplet delivery and positioning on a hydrophilic surface was also demonstrated. These results demonstrate a novel means to generate electricity from a natural process under ambient conditions, and the developed approach has potential applications in self-powered devices and systems.

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2. Experiment

2.1. Materials

Multi-walled carbon nanotubes (MWCNTs) were purchased from XF Nano, Nanjing, China. Sulfuric acid (H_2SO_4), nitric acid (HNO_3), sodium hydroxide (NaOH), ethanol, ethyl cellulose, acetic acid, glutaric dialdehyde (25 wt% aqueous solution), terpineol and toluene were purchased from Sinopharm. 1H,1H,2H,2H-Perfluorodecyltriethoxysilane (PFDTs) was purchased from Sigma-Aldrich. PEI (MW \approx 600) was purchased from Aladdin Industrial Corporation. Alumina strips were purchased from EMS Corp., Kunshan, China.

2.2. Synthesis of soluble CNT ink

3 g of MWCNTs were dispersed in 130 mL of mixed HNO_3 and H_2SO_4 ($\text{HNO}_3/\text{H}_2\text{SO}_4 = 1/3$ v/v) solution by sonication for 10 min. The mixture was then refluxed at 90 °C in an oil bath for 2.5 h under constant magnetic stirring. Afterwards, the mixture was cooled to room temperature. The mixture was subsequently washed with distilled water until the supernatant was neutral and was then re-dispersed in deionized water to form a 20 mg mL⁻¹ MWCNT ink.

2.3. Synthesis of carbon slurry

Toluene carbon black powder was first prepared by a methylbenzene flame synthesis method. One gram of toluene carbon black, 1 g of ethyl cellulose and 3 mL of terpineol were subsequently mixed in 50 mL of ethanol. The resulting mixture was probe-sonicated (Kesheng Sonics Vibra Cell, 550 F) for 20 min. Finally, the homogeneous slurry was evaporated to a total volume of 10 mL in an oil bath at 60 °C under magnetic stirring.

2.4. Fabrication of the device

Alumina strips were sequentially ultra-sonicated in acetone, alcohol and DI water and then dried at 70 °C in an oven. Next, CNT ink was printed into the alumina strip with required patterns using a floating knife coater. The carbon slurry was next blade-coated across the electrodes. After the electrodes were sintered at 350 °C for 150 min and cooled to room temperature, the obtained porous carbon film (PCF) was treated with air plasma (pressure 100 Pa, RF power 130 W) for 60 s in a plasma cleaner (Mingcheng, PDC-MG). The device was subsequently wired and encapsulated with epoxy in the exposed electrodes regions.

2.5. PFDTs modification

PFDTs was diluted with ethanol (1:10 v/v), and 1 wt% acetic acid was added to the solution and well shaken before use. The desired PCF regions were modified by being dip-coated in pre-diluted PFDTs ethanol solution at 70 °C and then heated for 30 min at 100 °C.

2.6. PEI modification

A 0.1 wt% PEI-600 aqueous solution was carefully dripped onto half of the PCF region and then heated to 70 °C for 30 min; after being washed with water and ethanol, the PEI-treated PCF area was then dipped in 0.1% glutaric dialdehyde aqueous solution heated to 70 °C in a water bath. Finally, the PEI-modified PCF was thoroughly washed with 70 °C DI water to remove residual reactants.

2.7. Characterization

The morphology, structure and functional groups of the PCFs were characterized by scanning electron microscopy (SEM, FEI Nova Nano450) and FTIR spectroscopy coupled with infrared microscopy

(FTIR, Bruker Vertex 70). Contact angles were measured using a contact angle meter (Kino SL200B). The current-voltage characteristics of the device were measured using a Keithley 2400 source measurement unit. The V_{oc} and I_{sc} as a function of time were recorded using a Keithley 2000 multimeter and a low-noise current preamplifier (SRS model SR570), respectively. The environmental temperature and humidity were simultaneously recorded by a Center 310 RS-232 humidity and temperature meter.

3. Results and discussion

A schematic of the device for measuring the droplet-induced voltage is shown in Fig. 1a. First, two carbon nanotube (CNT) electrodes were coated onto the ends of a well-cleaned Al_2O_3 ceramic plate (details in Experimental Section). Next, carbon slurry composed of toluene carbon black, terpineol, ethyl cellulose and ethanol was coated using a blade such that the slurry crossed the CNT electrodes. Afterwards, the device was sintered at 350 °C for 150 min to obtain a porous carbon film (PCF). The PCF was then treated with air plasma for 60 s to ensure superhydrophilic. A scanning electron microscope (SEM) image (Fig. 1b) shows that the PCF has a thickness of \sim 5 μm and consists of interconnected carbon nanoparticles with a mean diameter of tens of nanometers, and with most of the pore sizes ranging from several to tens of nanometers (Fig. S1). After the device was wired and encapsulated by epoxy in the exposed electrode regions, selected regions of the device were modified with 1H,1H,2H,2H-perfluorodecyltriethoxysilane (PFDTs) to make it superhydrophobic. The right inset of Fig. 1a shows a photograph of a typical device with two ends modified by PFDTs. The Fourier transform infrared (FTIR) spectrum of PFDTs@PCF (red line in Fig. 1c) shows obvious additional absorption peaks at 1256 cm⁻¹ and 1155 cm⁻¹ compared to the spectrum of pristine PCF (black line); these new peaks correspond to C–F and C–O–Si bonds, respectively [33]. The pristine PCF and PFDTs@PCF exhibit different degrees of wettability, with water contact angles of \sim 0° and \sim 152° (insets of Fig. 1c), respectively.

The prepared device shows robust ohmic contact, with a resistance of \sim 23.6 M Ω (Fig. S2). When deionized water (DI water, conductivity of \sim 1.6 \times 10⁻⁴ S m⁻¹, 1 μL) was dropped onto the PFDTs@PCF/PCF interface, water at the bottom of the droplet infiltrated into the porous carbon film, driven by capillary force. Because of the superhydrophobicity of the PFDTs@PCF region, water inside the PCF infiltrated only toward the hydrophilic region; infiltration in the opposite direction was disfavored. During this process, surprisingly, a sustainable voltage up to \sim 0.3 V was generated between the two CNT electrodes (Fig. 1d). The voltage remained nearly unchanged during the entire process and decreased quickly to zero when the droplet was completely vaporized. Such a phenomenon was highly reproducible. When five water droplets with the same size were dropped at the interface in sequence, similar open-circuit voltage signals were detected for each droplet (Fig. 1d). The short-circuit current was demonstrated to exhibit the same response (Fig. S3). On the contrary, water droplets moving on surface of graphene or aligned single-walled nanotubes only generated pulsed-electric signals [34–36].

The duration of the voltage was observed to be dependent on the size of the water droplets, as shown in Fig. 1e. The voltages induced by the water droplets with volumes of 1 μL , 2 μL , 5 μL and 10 μL were observed to persist for periods of \sim 73.7 s, \sim 195 s, \sim 610 s and \sim 1177 s, respectively, under ambient conditions, and nearly identical corresponding voltages (\sim 0.31 V, \sim 0.31 V, \sim 0.33 V, \sim 0.33 V and \sim 0.33 V, respectively) were generated (Fig. 1e). The whole phenomenon appears to be due to the PCF functioning as a unique electricity generator.

To explore the behavior of the water-droplet-induced voltage, we used a device with dimensions of \sim 50 \times 7 mm² with two ends and the central section modified with PFDTs. In the measurements, the right end was taken as positive and the left end was taken as negative

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