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RAPID COMMUNICATION

A flexible integrated photodetector system driven by on-chip microsupercapacitors



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Abstract

Flexible multi-functional nano/micro-systems with integrated energy units and functional sensing/detecting units on a single chip have gained considerable attention recently due to their optimized maximum functionality within a minimized sized chip and excellent mechanical flexibility and stability. In this work, we present a prototype of a flexible integrated on-chip photodetecting system with a reduced graphene oxide (rGO)-based in-plane microsupercapacitor and a CdS nanowire-based photodetector. For the system, the optimized electrodes in the microsupercapacitor were utilized as the source and drain electrodes of the CdS photodetector simultaneously, configuring a minimized self-powered-like visible light photodetector system without external power source. Driven by the microsupercapacitor (volumetric capacitance = 8.01 F/cm^3 and energy density = 6.204 W h/cm^3), current on/off ratios of 34.50 and 79.81 were obtained for the CdS-based photodetector depending on the number of the microsupercapacitors. The performance of the designed systems exhibited stable photo-current response consistent with the conventional one driven by the external power source, demonstrating the feasibility of the flexible integrated photodetector systems, which are promising for further large-scale and integrated applications. © 2015 Elsevier Ltd. All rights reserved.

Introduction

Compared with traditional single-effect electronics, multifunctional integrated nano/micro-systems have achieved more

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http://dx.doi.org/10.1016/j.nanoen.2015.02.027 2211-2855/© 2015 Elsevier Ltd. All rights reserved. concentration attributed to the multi-functional effect within the smallest volume [1]. Multi-functional integrated nano/ micro-system is a complete system that consists of both energy harvesting and storage units and the functional sensing/detecting or other functional components, aiming at building a self-powered system that operates independently, sustainably and wirelessly by it-self without any external battery, also called the "self-powered nanosystem"[1b]. Taking advantage of highly integration, this nanosystem exhibits the merits of small volume, light weight and multifunctions. During the past 10 years, the self-powered systems built on the energy harvesting and energy storage systems have been widely studied and different kinds of integrated systems have been successfully designed such as the integrated solar cell and lithium battery systems, solar cell and supercapacitor systems, nanogenerator and supercapacitor system etc [2]. Although a few reports about the integrated systems built on the energy storage and sensors have been reported by our group, the traditional stacked energy storage devices in the reported systems limited the space reduction of the whole system [3].

Recently, on-chip micro-energy storage devices are becoming a research hot due to the fast progress and special demanding of microelectronics [1c,4]. Among them, microsupercapacitors (mSCs) with high power density, fast chargedischarge ability, infinite cycle life and intrinsic safety are suitable for the energy storage and power source for microelectronic device and the tuned filter in integration circuit [1c,1d,5]. The shorter ion diffusion path in microsupercapacitor (mSC) results in a higher power density compared with the traditional supercapacitor [6]. Meanwhile, novel mSCs with in-planar interdigital structure would buffer the deformation stress in the flexible devices [7]. Graphene is one of the popular electrode materials utilized in the mSCs for its excellent conductivity, film-forming ability and favorable mechanical property [1d,8]. Till now, several kinds of methods have been explored for manufacturing flexible mSCs, such as laser-scribing method and electrochemical depositing method etc. [1d,1g]. Compared with those methods, the UV photolithography technology combined with plasma etching method seems to be a much simpler and efficient way, which can efficiently couple micro-energy storage device with other nano-devices such as nano photo-detectors on a single chip to form compact integrated on-chip micro/nanosystems [9].

In this work, combining the micro-manufacturing technology of flexible mSC and photodetector, we designed a prototype of a self-driven on-chip photodetector system by integrating an rGO-based in-plane mSC with a CdS nanowire-based photodetector. The optimized electrodes in the mSC device were utilized as the source and drain electrodes of CdS photodetector as well, configuring a self-powered visible light photodetector without external power source. Moreover, the bias voltage of the photodetector can be tuned by scaling up the individual mSC system to a tandem system. The current on/off ratios were obtained by the individual and tandem microdevices respectively, exhibiting a photo-current response consistent with the one driven by the external power source.

Experimental section

Preparation of reduced graphene oxide (rGO) mSC

The fabrication process of the rGO mSC is exhibited in Figure 1. Firstly, PET substrates $(2 \text{ cm} \times 3 \text{ cm})$ were cleaned by acetone, ethanol and distilled water in sequence. To enhance the hydrophility of PET, the top side of the PET substrate was treated by oxygen plasma (750 V, 20 min) in plasma cleaner with 0.6 L/min of air flow. Then, 1 mL of dispersed graphite oxide (GO) solution (1 mg/mL) was dropped onto the surface. GO sheets were synthesized by a modified Hummer method using natural graphite powder [10]. After being dried at room temperature overnight, a uniform GO film was formed on the PET substrate. Subsequently, photoresist was spin-coated on the GO film at 500 rpm for 15 s and 4000 rpm for 30 s. After spinning coating for three times, the substrates were heated at 100 °C for 4 min and the resist was subsequently patterned to obtain the desired surface structures on the GO film using the ultraviolet lithography machine with the mask and developed for 20 s. Then, oxygen plasma treatment (750 V, 80 min) was applied to etch the exposed GO in the plasma cleaner with 0.6 L/min of air flow. After removing the extra resist, patterned GO film was obtained. Hydrazine vapor was used to reduce GO pattern into rGO pattern at room temperature for 24 h. To improve the electrical contact between the square electrodes in the pattern with testing probes, the electrodes were evaporated with 20 nm Au. Furtherly, a clear gel electrolyte composed of 3 g of KOH, 6 g of polyvinyl alcohol (PVA) and 60 mL H₂O prepared at 100 °C under 2 h stirring was spread on the integrated electrodes of the microsupercapacitor. After vaporizing the excess water, an all-solid-state rGO microsupercapacitor was obtained.



Figure 1 Schematic illustration of the fabrication process of the flexible microsupercapacitor made up of 20 fingers integrated on the PET substrate. The fabrication process flow includes spreading GO dispersion on the PET substrate, photolithography of the pattern, air plasma etching of the extra GO, removing the resist, hydrazing reduction of GO, sputtering Au on the electrodes and coating gel electrolyte.

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