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Simple fabrication of heterojunction solar cells by utilizing carbon materials films

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ABSTRACT

We present a simple fabrication process of heterojunction solar cells from single-walled carbon nanotubes (SWCNTs) and *n*-type silicon wafer. SWCNTs were synthesized from alcohol-catalytic chemical vapor deposition and were made into a film with transmittance of 70% by vacuum filtration. The mean diameter of the nanotube was about 1.1 nm. The patterned silicon substrate was prepared by a simple wet-etching process with hydrofuric acid utilizing a hand-made physical mask. The nanotube film was transferred onto the patterned silicon substrate. Power-conversion efficiency value of 4.3% with fill factor of 0.67 can be achieved.
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1. Introduction

Carbon materials have been widely studied for either electronic or optical devices regarding their properties [1–4]. They have also been promising materials for solar energy conversion known as photovoltaic devices [5–10]. Solar cells can be fabricated into many aspects, which result in different power of conversions and fill factors. Although there have been many researches focusing on improvement of efficiency of solar cells performance, silicon is still vital to achieve high efficiency [11–16]. The combinations of silicon with other types of carbon materials are intensively studied such as fullerene [17,18], carbon nanotube [6,7,11–13], graphene [8,9], etc. Considering band gap energy of each component is significant, and carbon materials with certain energy gap are sometime required to incorporate with silicon. Carbon nanotube is, therefore, a choice of selectable energy gap with highly air stable and exciting efficiency. Selective structure of carbon nanotubes can be performed via minimizing catalyst agglomeration [19,20] or post-treatment process [21] in order to control nanotube mean diameter as indirect selective properties, or to achieve a certain structure of nanotubes. While nanotubes are used as hole collector and conductive layer [11,14], simple method for silicon substrate patterning needs to be considered to achieve low cost process. It is then easy to study capability of solar cell fabrication.

In this study, we present a simple fabrication process of heterojunction solar cells from single-walled carbon nanotubes (SWCNTs) and *n*-type silicon wafer. SWCNTs were synthesized from alcohol-catalytic chemical vapor deposition, and nanotubes were made into a film with transmittance of 70% by vacuum filtration. The mean diameter of nanotubes was about 1.1 nm. The patterned silicon substrate was prepared by a simple wet-etching process with hydrofuric acid (HF) with hand-made physical mask. The nanotube film was transferred onto the patterned silicon substrate.

2. Experiment (or materials and methods)

2.1. Synthesis of single-walled carbon nanotubes (SWCNTs)

SWCNTs were synthesized at 800 °C by alcohol-catalytic chemical vapor deposition (CVD) process [22]. The cobalt acetate (Co) and iron nitrate (Fe) used as binary catalyst were prepared by impregnation method with zeolite particles. The concentration of 2.5 wt.% each of Co and Fe were dissolved in 40 g of ethanol along with 1 g of zeolite particles. The solution was then mild sonicated for 90 min before leaving it dried at 80 °C. Dried catalytic zeolite was ground, and catalytic powder of Co/Fe binary catalyst was stored at 80 °C for use in nanotube synthesis.

A quartz boat containing the catalytic powder was placed in the CVD furnace and heated to 800 °C under Ar/H_2 (3%H_2) atmosphere to reduce catalysts for 30 min. Once the growth temperature was reached, the Ar/H_2 supply was terminated. Ethanol vapor was then flowed into the CVD

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chamber with a flow rate of 450 sccm for 10 min, prior to cooling down to room temperature.

2.2. Preparation of nanotube film

SWCNT film was prepared based on the process reported in [23]. As-grown SWCNTs on zeolite particles were dispersed in deionized water containing 1% of sodium dodecylbenzenesulfonate (SDBS) surfactant. Dispersed SWCNTs were mild sonicated for 90 min and ultra-sonicated for 30 min. The nanotube suspension was then centrifuged for 15 min at the speed of 85,000 rpm (327,000*g*). Eventually, the supernatant was extracted. In order to make the nanotube film, the supernatant was filtered by vacuum filtration process and rinsed again with deionized water. The supernatant was also used for further optical measurements. The nanotube film was finally obtained by dissolving filtration membrane with acetone and rinsed again with isopropyl alcohol.

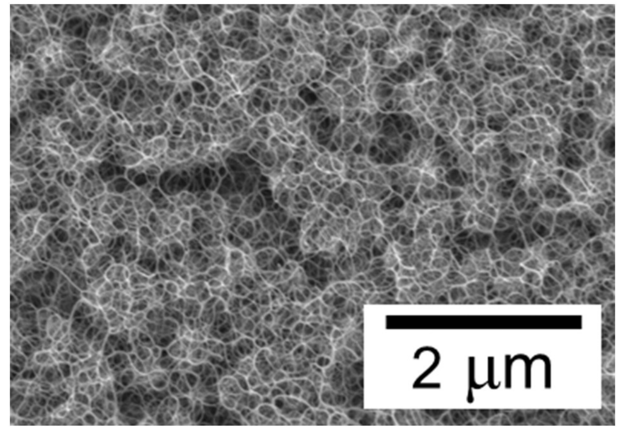

Fig. 2. SEM micrograph of as-grown SWCNTs synthesized from alcohol-catalytic CVD (ACCVD) method on zeolite particles.

2.3. Solar-cells fabrication

Heterojunction solar cell was fabricated from *n*-type silicon and SWCNT film. Patterned silicon was made by photolithography process. Photoresist was spin-coated on silicon wafer, and heated at 90 °C for 5 min. The wafer was patterned by light passing through physical mask with the window size of 0.3 cm × 0.3 cm for 1 h (Fig. 1), prior to developing in NaOH solution (14%) for 1 min. Hydrofuric acid (HF) with concentration of 20% was used to etch SiO_2 layer by dropping on exposed area. The rest of photoresist was dissolved in acetone, and the etched SiO_2 layer was rinsed with isopropyl alcohol before baking at 120 °C for 5 min. Gold (Au) with the thickness of 40 nm was then evaporated on top of the SiO_2 layer and on the backside of silicon wafer to serve as electrodes. The prepared SWCNT film was finally transferred onto the patterned *n*-type silicon.

3. Results and discussion

The SEM micrograph in Fig. 2 clearly shows viability of as-grown carbon nanotubes on zeolite particles. Single-walled carbon nanotubes are demonstrated by Raman spectroscopy with three different excitation wavelengths (488, 514 and 633 nm) as shown in Fig. 3 [24]. The peak at 1594 cm^{-1} indicates the vibration mode of carbon atom along the axial direction of carbon nanotubes, called tangential mode, or G-band. The splitting of G-band into G^+ and G^- indicate vibration in longitudinal and circumferential directions of carbon atoms. The presence of the so-called radial-breathing (RBM) mode (100–400 cm^{-1}) implies the vibration of carbon atoms in radial direction. The G-band and RBM indicate that single-walled carbon nanotubes were obtained from ACCVD process. According to correlation, $\omega_{RBM} \approx 217.8/d + 15.7$ [25], the Raman shift of RBM mode (ω_{RBM}) is the inverse of nanotube diameter (d). Certain different nanotube diameters would give rise to specific resonance window with three different excitation energies in RBM region as seen in Fig. 3, and nanotube diameter can be evaluated to be approximately 0.8 to 1.6 nm. The G/D ratio measured by excitation wavelengths of 488, 514 and 633 nm are 12, 19 and 9.4, respectively, indicating less imperfection of carbon lattice and low impurity level in the nanotube sample [24].

Fig. 4 shows the optical absorption spectrum of dispersed SWCNTs in water. It reveals the band energy of the first (S_{11}) and second (S_{22}) electronic transitions of semiconducting nanotubes, indicating specific structure of the nanotubes [25]. The S_{11} and S_{22} regions are approximately in the range of 900 to 1500 nm, and 500 to 900 nm, respectively. Specific chiralities are assigned to be (6,5), (7,5), (7,6), (8,6) and (10,3) nanotubes. Based on observable chiralities presented in optical absorbance, nanotube diameter is approximately less than 1 nm [25]. Although the nanotube diameter can be evaluated from the peak positions of S_{11} transition measured from dispersed sample, it does not represent the diameter of the entire sample. TEM measurement was employed for further diameter evaluation (Fig. 5). Fig. 5 shows TEM images of as-grown SWCNT dispersed on TEM microgrid, indicating clean single-walled carbon nanotubes. Diameter distribution obtained from 100 nanotubes was shown in the histrogram from TEM observation. The observed mean diameter is about 1.06 ± 0.24 nm, which corresponds to that obtained from Raman measurement.

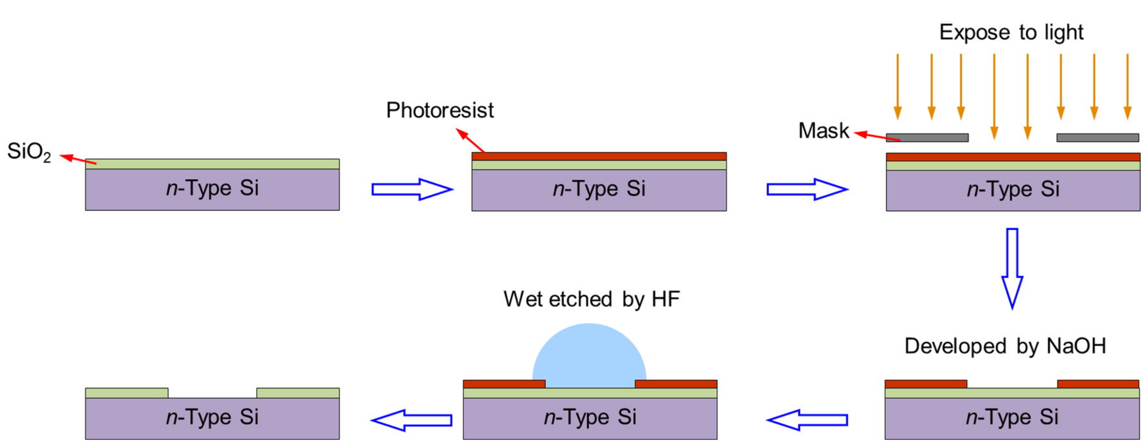

Fig. 1. Schematic of silicon substrate pattering by wet-etching and photolithography processes.

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