



Effect of single-chirality single-walled carbon nanotubes in dye sensitized solar cells photoanodes

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ABSTRACT

Dye-sensitized solar cells (DSSCs) with photoanodes composed of chirality selected single-wall carbon nanotubes (SWNTs) have been fabricated and tested for first time. Single chirality SWCNTs separation (93% purity) have been achieved by modifying standard size exclusion gel chromatography. Chirality selection has allowed for “tuning” of the energy barrier at the TiO₂/SWNT/FTO interface, electronic conductivity enhancement, and reduced SWCNTs-ruthenium dye competition for light absorption resulting in a 81% energy conversion efficiency improvement compared to mixed chirality cells. Unfortunately, energy conversion efficiency has been limited by cells’ low shunt resistance. Additionally, SWCNTs electron transfer properties have been exploited to prepare mixed and chirality specific CNTs based DSSCs’ counter electrodes.

1. Introduction

Since their discovery in 1991, carbon nanotubes (CNTs) have attracted considerable attention for the development of electronic devices [1,2]. However, CNTs grown by standard methods (i.e. arc discharge, PECVD, catalytic CVD, etc.) are comprised of a mixture of metallic and semiconducting tubes. Furthermore, semiconducting tubes possess different chirality-dependent optoelectronic properties (i.e. energy gap, work function, tube-tube junction resistivity, etc. [3]) making CNTs’ structure control crucial for integrated circuits (ICs) applications. Although application of single chirality CNTs in optical and electronic devices has been tested with positive results [4], improving selectivity and yield of chirality separation techniques remains a major goal.

DSSCs represent an attractive alternative to traditional solar cells (Si, CdTe, CIGS, etc.) because of their cost-effectiveness and ease of manufacture due to the employment of non-toxic materials [5]. DSSCs generate current from the absorption of photons by a dye molecule (usually a ruthenium bi-pyridine complex) which injects electrons into the conduction band of a wide band gap semiconductor, usually TiO₂. The dye is then regenerated by an electrolytic solution containing an iodide/triiodide (I⁻/I₃⁻) redox couple which has been reduced at the counter electrode of the cell (usually platinum). The competition between the transport of the electrons through the TiO₂ porous network and their recombination with the electrolyte’s redox couple

at the photoanode/electrolyte interface, defines the electron collection efficiency of the device and its limitations. The disordered network of TiO₂ nanoparticles contains numerous grain boundaries which ultimately result in slow electron transport and increased recombination. Approaches to improve electron transport have included the incorporation of low dimensional nanostructured metal oxide materials such as fibers [6] and flakes. Carbon materials, especially graphene sheets and CNTs, have been used to increase the conductivity of DSSCs’ photoanodes either as composites with TiO₂ [7,8] or as thin film deposited on FTO substrates [9]. In both cases, their catalytic properties for the reaction $I_3^- + 2e^- \rightarrow 3I^-$ constitute the ultimate limitation to the energy conversion efficiency [10]. DSSCs represent a unique device for testing at once different properties stemming from CNTs chirality separation. In fact, DSSCs’ light generation, charge separation, and carrier collection mechanisms allow to compare optoelectronics characteristics such as:

- Electrical conductivity. Bundles of single chirality tubes are expected to show less intertube resistance than mixed-chirality CNTs networks. Furthermore metallic tubes possess low values of bulk resistivity favoring the electron transport through the photoanode [11].
- Work function. Depending on the electronic structure of a particular CNT (semiconducting or metallic), when it is put into contact with the TiO₂ network, it will form a Schottky-type barrier at the TiO₂/

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CNT/FTO interface. The height of the barrier depends on the work function (Fermi level position) of the tubes. TiO_2 is considered an n-type semiconductor, however when in contact with a carbon nanotube, electrons from the TiO_2 will move towards the CNT surface. If the CNT has a lower Fermi level than the TiO_2 network, the CNT will act as an electron sink resulting in the TiO_2 behaving similar to a p-type semiconductor as the holes in its valence band travel to the surface [12].

- Optical band gap. Single chirality absorption spectra in the Uv-visible range depend on tubes diameter. Moreover the open circuit voltage of the DSSCs is influenced by the CNTs band gap, [8,9].
- Catalytic properties. They affect the electron transfer from the TiO_2 photoanode to the electrolyte (leakage current).

The present study introduces a “single-pass” chromatographic procedure for producing a sufficient amount of chirality separated tubes to be employed as photoanode material for DSSCs. Five different samples have been investigated: a mixed chiralities (30 chiralities) sample, two samples containing mixtures of two different chiralities ((6,4)/(6,5) and (6,4)/(7,3)), and two single chirality samples ((7,3) and (6,5)). While this study does not reveal the best DSSCs performances it reports a trend of increasing device efficiency with chiral selectivity and an improvement over the mixed chirality film (with a 81% improvement for the 7,3 chirality). Furthermore, it has been determined that certain chiralities (i.e (6,4)) are not compatible for use in a DSSC electrode.

2. Experimental

2.1. Samples preparation

Raw HiPCO (High Pressure Carbon Monoxide) CNTs (Nanointegris, U.S.) have been treated using the organic-aqueous phase separation procedure outlined in Tvrđy et al. [4]. A CNT suspension has been created at 1 mg/mL in an aqueous solution of 2% wt. sodium dodecyl sulphate (SDS, 99%, Sigma Aldrich). The mixture has been put first in an ultrasonic bath for 5 min to break up the large CNTs agglomerates before being placed under a tip sonicator (Sonifire 450D, Branson, U.S.) with a 0.5 in. flat tip at 20 W/cm² for 20 h. Before sonication, the tip has been set 1 cm away from the bottom of the beaker and placed in a temperature-controlled bath held at 10 °C. Immediately following tip ultra-sonication the sample has been ultra-centrifuged at 197,000 G for 15 min at 10 °C. The top 90% of the supernatant has been removed from each ultracentrifuge tube and directly poured into the gel column described in the following section. Filtration and chirality separation have been done in two phases by using a gel chromatographic method outlined by Liu et al. [13]. The original method has been modified to a single pass to increase the yield and concentration of purified SWNTs for PV applications.

2.2. Size exclusion gel Chromatography separation of SWCNTs

The general chirality selection procedure reported in Ref. [13] is a two-pass process relying on the interaction between SDS-SWCNT micelles and hydrogels (i.e. an allyl dextrane-N,N-methylene bisacrylamide cross linked co-polymer) that results in the chirality separation of SWCNTs [14]. Sephacryl S-200 HR (GE Healthcare, U.S.) has been used as chromatographic stationary phase in the present study.

The main drawback of such a two-steps chromatographic process is the decrease in CNT concentration after each pass. Obtaining as much as possible of the enriched-chirality CNTs in one pass is mandatory for electronic devices fabrication. Our study proposes a modification of the original procedure to maximize the collection of enriched single-chirality in a single pass. Similar to the original procedure, a number of syringes has been vertically arranged in series and plugged with glass wool (see Fig. 1). The number of syringes arranged depends on the

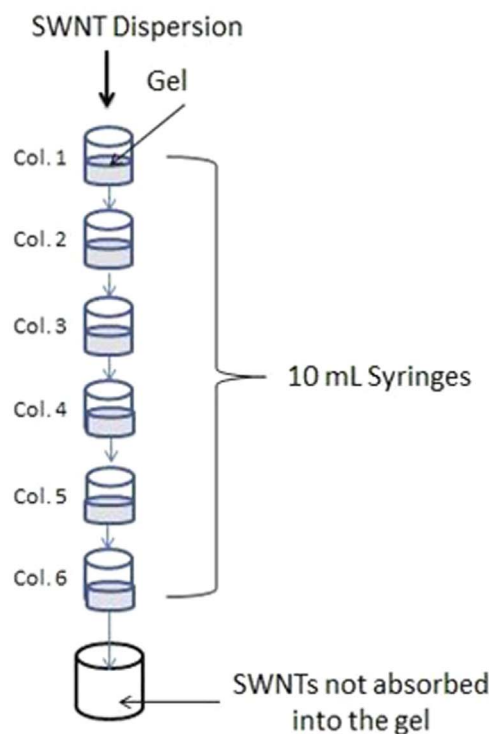


Fig. 1. Schematics of the chromatographic separation process (top), and actual modified apparatus (bottom). (colour on line)

specific chirality to be isolated. 1–2 mL of gel have been injected into each syringe and equilibrated with 10 mLs of 2 wt% of SDS solution. Unbound tubes have been eluted with a 1.8 wt% [15] SDS solution. The chiralities targeted in this study are (6,5) and (7,3). It has been found that in order to separate only those chiralities in a single pass, different ratios of gel/SWNTs are to be used according to the different affinity of specific chiralities for the allyl-dextran gel. In fact, smaller diameter tubes ((7,3) diameter=0.696 nm [16]) strongly interact with the gel requiring a lower degree of gel “overloading” [17]. Thus, to separate enriched (7,3) 11 mLs of SWNT/SDS solution have been injected into the top column with 1.4 mL of gel in each syringe. The (7,3) chirality came out in the first three columns; however the most pure sample has turned out to be in the first. On the other hand, by injecting 17 mLs of

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