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Project title: Self-sorting of Metallic Carbon Nanotubes for High Performance Large Area Transparent Electrodes for Solar Cells

Investigators
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Abstract

SWNTs have superb electronic and mechanical properties; however, SWNTs come in different chiralities making them either metallic (MET) or semiconducting (SC). It is desirable to have only MET tubes in transparent electrode applications. We have recently developed a method to selectively separate the SC and MET SWNTs and form self sorted networks of each on different functionalized surfaces. To enable MET-SWNT networks on other substrates like flexible polymers and quartz, and to increase the density of sorted networks past the surface sorting threshold, we developed a transfer printing method. SWNTs were picked up with nearly 100% yield and transferred intact onto both hydrophobic and hydrophilic surfaces including polymeric and quartz substrates as determined by AFM and UV-Vis spectroscopy. A multi-layer stack was also accomplished using this same method with different surface treated substrates. We plan to use this transfer method to prepare high density MET-SWNT networks for transparent electrode application.

We investigated a new method to control the uniformity of carbon nanotube network thin film and its morphology. While carbon nanotube network electrodes show promise, characteristically poor dispersion properties have limited their practicality. We report that addition of small amounts of conjugated polymer to nanotube dispersions enables straightforward fabrication of uniform network electrodes by spin-coating, and simultaneous tuning of parameters such as bundle size and density. After treatment in thionyl chloride, electrodes with sheet resistances as low as 70 ohm/sq with 81% transmittance at 550 nm are obtained; these are among the best reported carbon nanotube based transparent electrodes to date.

Introduction

The application of carbon nanotubes (CNT) to the fabrication of transparent conductive electrodes (TCE) for photovoltaic applications has been investigated in the recent years for their potential to address some of the shortcomings of current Indium-Tin-Oxide (ITO) technologies, namely the high material and fabrication costs and the brittleness of the deposited layers. CNTs have the prospect to meet low fabrication cost targets for the low cost of earth-abundant carbon and for their potential to be dispersed into solution for large area coating. Additionally, they have excellent thermal, mechanical, and electrical properties that make them an ideal candidate for TCEs: they are stable at the high temperatures required to process inorganic devices; they have excellent strength and flexibility; and they have an electrical conductivity three orders of magnitude greater than metals like copper.

Despite their potential, state-of-the-art CNT-based TCEs have still poor performance with a sheet resistance more than ten times higher than ITO electrodes at comparable light transmittance (~80%). The key problem in reducing the sheet resistance of CNT-based layers is that as-synthesized CNTs usually contain a mixture of approximately 67% semiconducting CNTs (sc-CNT) and only 33% metallic CNTs (met-CNT), where the difference in electronic properties depends on the chirality of the nanotubes. This implies that two-thirds of the film contributes to the absorption of light while decreasing the conductance. Moreover, tube-to-tube contact resistance between sc- and met-CNTs is three orders of magnitude higher than met-met junctions and two orders of magnitude higher than sc-sc junctions. In order to maximize both transmittance and conductance and reach the efficiency of the best ITOs, it is hence necessary to develop a technology that uses 100% met-CNTs.

Some concurrent research efforts are investigating the selective synthesis of sc- or met-CNTs or bulk separation methods. However, these technologies still present fundamental obstacles that require further developments. This research is exploring an alternative strategy based on a self-sorting mechanism to be implemented during the deposition process.

In addition to sorting met-CNTs, the technology investigated in this project is ultimately capable to organize the CNT network into partially aligned bundles. Controlling the morphology of the CNT layer allows to further decrease the sheet resistance of CNT-TCEs by minimizing tube-to-tube junctions. Finally, a CNT doping method is also being explored to increase the film conductivity. The conjunction of all these approaches will lead to a CNT electrode with the potential to outperform best available ITOs.

Results

SWNTs have superb electronic and mechanical properties; however, SWNTs come in different chiralities making them either metallic (MET) or semiconducting (SC). It is desirable to have only MET tubes in transparent electrode applications. We have recently developed a method to selectively separate the SC and MET SWNTs and form self sorted networks of each on different functionalized surfaces. To enable MET-SWNT networks on other substrates like flexible polymers and quartz, and to increase the density of sorted networks past the surface sorting threshold, we developed a transfer printing method. SWNTs were picked up with nearly 100% yield and transferred intact onto both hydrophobic and hydrophilic surfaces including polymeric and quartz substrates as determined by AFM and UV-Vis spectroscopy. A multi-layer stack was also accomplished using this same method with different surface treated substrates. We plan to use this transfer method to prepare high density MET-SWNT networks for transparent electrode application.

We investigated a new method to control the uniformity of carbon nanotube network thin film and its morphology. While carbon nanotube network electrodes show promise, characteristically poor dispersion properties have limited their practicality. We report that addition of small amounts of conjugated polymer to nanotube dispersions enables straightforward fabrication of uniform network electrodes by spin-coating, and simultaneous tuning of parameters such as bundle size and density. After treatment in thionyl chloride, electrodes with sheet resistances as low as 70 ohm/sq with 81% transmittance at 550 nm are obtained; these are among the best reported carbon nanotube based transparent electrodes to date.

<u>Publications:</u>

1) S.L. Hellstrom, H.W. Lee, Z. Bao, “Polymer-assisted direct deposition of carbon nanotube bundle networks for high performance transparent electrodes”, **ACS Nano**, accepted.

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