



The Functionalization of Carbon Nanosheets

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ABSTRACT

Carbon nanosheets are a novel two-dimensional nanostructure made up of 2 - 20 graphene atomic planes oriented with their in-plane axis perpendicular to the growth substrate. Previous efforts in developing nanosheet technology have focused on the characterization of the system and their development as an electron source due to the high atomic enhancement factor (β) and low turn on field. Further investigation of nanosheets as high surface area electrodes revealed poor wetting by polymeric material and extreme hydrophobic behavior.

Because nanosheet technology has promise as a high surface area electrode material, this thesis research has focused on three areas of interest: the enhancement of nanosheets through chemical modification, the incorporation of the nanosheets into a polymeric composite and the delivery of a proof of concept measurement. We have successfully introduced defects into the graphene lattice of the nanosheet system via an acid treatment. Inspection of these defects by x-ray absorption near-edge spectroscopy (XANES) shows the introduction of two features in the spectra assigned to C=O π^* and C-O σ^* transitions. Thermal desorption spectroscopy (TDS) was used to identify the oxygen containing groups created during the functionalization as carboxylic and hydroxyl functional groups. These groups were identified through the combination of carboxylic, hydroxyl, anhydride and lactone peaks in the CO₂, CO and H₂O TDS spectra and the presence of not only carboxylic and hydroxyl signature peaks in the spectra. Deconvolution of the TDS spectra using 1st and 2nd order Polanyi-Wigner equations enables the calculation of desorption energy values for individual features and for the estimation of the number of atoms desorbing from the surface during a particular event. Identification of the exact nature of the functional groups was attempted through high resolution x-ray photoelectron spectroscopy (XPS) of the C(1s) and O(1s) peaks. Though the pairing of sub-peaks with specific functionalities of the system was not possible due to the complexities of the spectra, the trends observed in the data support the data gathered via the XANES and TDS experiments.

Also, a procedure for the classification of defect density and exact functionality was outlined. Deconvolution of the TDS spectra using 1st and 2nd order Polanyi-Wigner equations enabled the calculation of desorption energy values for individual features and for the estimation of the number of atoms desorbing from the surface during a particular event. This information along with the changing sub-peak areas from dedicated and calibrated XPS system would allow for not only a more accurate estimation of defect density, but also for the identification of sub-peaks in the C(1s) and O(1s) spectra.

Finally, photoluminescence measurements of poly[2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylene vinylene] (MEH-PPV) and MEH-PPV/nanosheet systems showed a quenching of three orders of magnitude for the MEH-PPV/nanosheet system suggesting that nanosheets are a viable option for excitation separation in organic photovoltaics.