Title: Clusterization, Electrophoretic Deposition, and Photoelectrochemical Properties of Fullerene-Functionalized Carbon Nanotube Composites

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Abstract: We have successfully developed a new methodology for the selforganization of C_60 molecules on the sidewall of carbon nanotubes for use in photoelectrochemical devices. Novel nanocarbon composites of fullerene (e.g., C_60) and highly soluble, chemically functionalized single-walled carbon nanotubes (f-SWNT) have been prepared by the rapid injection of a poor solvent (e.g., acetonitrile) into a mixed solution of C_60 and f-SWNT in o-dichlorobenzene. Measurements by using scanning electron microscopy of cast samples revealed that the composites are categorized into three groups; i) f-SWNT bundles covered with layers of C_60 molecules, ii) round, large C_60 clusters (sizes of 500-1000 nm) containing f-SWNT, and iii) typical, round C_60 clusters (sizes of 150-250 nm). The electrophoretic deposition of the composites onto a nanostructured SnO_2 electrode yielded the hierarchical film with a gradient composition depending on the difference in the mobilities of C_60 and f-SWNT during the electrophoretic process. The composite film exhibited an incident photon-to-photocurrent efficiency as high as 18% at \( \lambda = 400 \) nm under an applied potential of 0.05 V vs. SCE. The photocurrent generation efficiency is the highest value among carbon nanotube-based photoelectrochemical devices in which carbon nanotubes are deposited electrophoretically, electrostatically or covalently onto semiconducting electrodes. The highly aligned structure of C_60 molecules on f-SWNT can rationalize the efficient photocurrent generation. The results obtained here will provide valuable information on the design of carbon nanotube-based molecular devices.