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Theoretical modeling of carbon nanostructure for energy storage and nanoelectronics applications.

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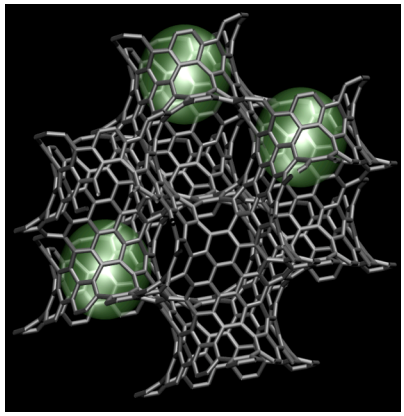
Abstract – Computational methods are used to study applications of carbon nanostructures as electrical energy storage media (supercapacitors) and nanoelectronic devices (including 3D nanomeshes and heterojunctions). Supercapacitors are studied using a heuristic theoretical model based on density functional theory that takes pore curvature into account. Electronics applications in carbon nanomeshes are studied using a quantum transport approach within Landauer-Buttiker theory and Green's functions formalism. We emphasize the role of modeling in providing a precise understanding of processes responsible for a given functionality, and how modeling can be used to enhance desired properties and suppress unwanted ones.

Theoretical methods have evolved to a point where the properties of materials can be successfully predicted based solely on their atomic structure. As such, they provide a unique tool, able to help identifying the origins of the properties of a given structure and uncovering principles that can be used to tailor structure for target applications. Here we show two distinct uses of computational modeling for applications involving carbon-based nanostructures.



Unit cell of a typical carbon nanonetwork, representing a junction between multiple carbon nanotubes. This system possess interesting transport properties and can be tailored into a useful device. It is also a model system to study nanoporous carbon for electric energy storage applications.

The first application presented is concerned with capacitive electrical energy storage (i.e. not involving chemical reaction). Supercapacitors based on nanoporous carbon materials, commonly called electric double-layer capacitors (EDLCs), are emerging as a novel type of energy-storage device with the potential to substitute batteries in applications that require high power densities. The EDLC model has been used to characterize the energy storage of supercapacitors for decades. In particular, I will present a heuristic model that avoids the shortcomings of the EDLC and that takes pore curvature into account as a replacement for the EDLC model. [1-2] The density functional theory based model explains experimental observations for a range of pore sizes: from the micropore regime (< 2 nm), mesopore regime (2–50 nm), and macropore regime (> 50 nm); and diverse carbon materials and electrolytes. The model allows the properties of a supercapacitor to be correlated with pore size, specific surface area, Debye length, electrolyte concentration, dielectric constant, and solute ion size, and lead to a optimization pathway of carbon supercapacitors properties through experiments.

In the second application discussed in this talk, I will present an overview of our work devoted to electronic transport in carbon nanomeshes and networks. [3-5] I will first show how atomistic model can be built, based solely on carbon nanotubes as elementary building blocks and a combination of point and space group symmetries. A few specific cases will be presented in detail, highlighting the intricate mechanisms involved in the current distribution in the network. The effect of the presence of defects will also be highlighted, revealing that somewhat contrary to common wisdom, a sufficiently high density of topological defects can in fact induce functionality. [5] Finally, I will present the theoretical branching mechanism that can be used to devise experimental methods to create cabon nanonetworks. In particular, the importance of using hetero-doping during carbon nanostructure growth will be highlighed [6-7].

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