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Origins of Lithium-Carbon Binding in Carbon-based Lithium-ion Battery Anodes BRANDON WOOD, Lawrence Livermore National Laboratory, YUANYUE LIU, Rice University, MORRIS WANG, Lawrence Livermore National Laboratory, BORIS YAKOBSON, Rice University — Many key performance characteristics of carbon-based lithium-ion battery anodes are determined by the strength of binding between lithium (Li) and sp^2 carbon (C). Using extensive density functional theory calculations, we investigate the detailed interaction of Li with a wide variety of sp^2 C substrates, including pristine, defective, and strained graphene; planar C clusters; nanotubes; C edges; and multilayer stacks. We find that in almost all cases, the Li-C binding energy scales is determined largely by the work required to fill unoccupied carbon states, suggesting that intrinsic quantum capacitance is important for predicting Li capacity. This allows the binding energy and capacity to be estimated based solely on the electronic structure of the substrate. It also provides a connection to carbon-based supercapacitors, and underscores the role of electronic structure in interfacial electrochemical systems. Implications for improving the effective capacity of carbon-based anodes will be discussed. This work was performed under the auspices of the U.S. DOE by LLNL under Contract DE-AC52-07NA27344.

Brandon Wood
Lawrence Livermore National Laboratory

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