

Abstract Submitted
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Binding and Diffusion of Lithium in Graphite: Quantum Monte-Carlo benchmarks and validation of van der Waals density functional methods¹ PAUL KENT, PANCHAPAKESAN GANESH, MINA YOON, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge 37831, JEONGNIM KIM, FERNANDO REBOREDO, Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge 37831 — Benchmark diffusion quantum monte-carlo (DMC) studies of the adsorption and diffusion of atomic lithium in graphite are compared with density functional theory (DFT) calculations using several van der Waals methods. The charge transfer is captured adequately with conventional local density functionals. At fixed geometries, these yield surprisingly accurate energetics. In unconstrained geometries, van der Waals corrections are required to correctly reproduce graphite and lithium binding. We find that the empirical method of Grimme et al. only gives correct diffusion barriers when the Li polarizability is reduced to nearly zero, consistent with the charge transfer in the solid-state environment. The Tkatchenko-Scheffler scheme captures the polarizability reduction, yielding accurate results at low computational cost. The self-consistent vdW-DF2 functional yields the best overall results but at increased cost. Slight differences in barrier heights remain with all the DFT approaches compared to the DMC. These results establish a hierarchy of modeling approaches for the lithium-carbon system.

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