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Extending DFT to Long Time-Scales: Using the Density Functional Tight Binding Approach for Materials Under Extreme Temperatures and Pressures[1]

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We report here on density functional tight binding (DFTB) simulations of covalently bonded materials over a pressure range of 10 – 2,000 GPa and a temperature range of 300 – 30,000 K using both standard and new interaction potentials we have created for these conditions. Density Functional Theory (DFT) has been shown to accurately reproduce the high pressure-temperature chemistry, phase boundaries, and EOS of many materials. DFT-MD simulations, though, scale poorly with computational effort and thus are generally limited to nanometer system sizes and picosecond time-scales. In contrast, chemical kinetic effects and phase changes can span up to nanosecond timescales and significantly longer length scales. The DFTB method holds promise as a high throughput simulation capability by providing orders of magnitude increase in computational efficiency while retaining most of the accuracy of Kohn-Sham DFT. We show that DFTB interaction potentials can be created by (a) fitting the DFTB repulsive energy to measured and computed compression data, and (b) using an extended basis set that includes *d*-orbital interactions, as needed. Our new potential for carbon yields accurate material properties for diamond, graphite, the BC8 phase, and simple cubic carbon, as well as for the shock Hugoniot of diamond compressed up to the conducting liquid. We also discuss simulations of the long-time scale reactivity of H_2O_2 under detonation conditions, and shock compression of astrophysical ice mixtures and the subsequent synthesis of pre-biotic materials. Our results provide a straightforward method by which DFTB can be made to provide equation of state and long-time scale chemical kinetic data at a similar accuracy to standard quantum codes. Our approach could be extended to any number of materials related to geology and planetary science, including silicon, SiO_2, and hydrocarbon systems.

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