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Improving density functional tight binding predictions of free energy surfaces for peptide condensation reactions in solution¹ MATTHEW KROONBLAWD, NIR GOLDMAN, Lawrence Livermore Natl Lab — First principles molecular dynamics using highly accurate density functional theory (DFT) is a common tool for predicting chemistry, but the accessible time and space scales are often orders of magnitude beyond the resolution of experiments. Semi-empirical methods such as density functional tight binding (DFTB) offer up to a thousand-fold reduction in required CPU hours and can approach experimental scales. However, standard DFTB parameter sets lack good transferability and calibration for a particular system is usually necessary. Force matching the pairwise repulsive energy term in DFTB to short DFT trajectories can improve the former's accuracy for chemistry that is fast relative to DFT simulation times (<10 ps), but the effects on slow chemistry and the free energy surface are not well-known. We present a force matching approach to increase the accuracy of DFTB predictions for free energy surfaces. Accelerated sampling techniques are combined with path collective variables to generate the reference DFT data set and validate fitted DFTB potentials without a priori knowledge of transition states. Accuracy of force-matched DFTB free energy surfaces is assessed for slow peptide-forming reactions by direct comparison to DFT results for particular paths. Extensions to model prebiotic chemistry under shock conditions are discussed.

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