Comprehensive Quantum Chemistry Approach for the Evaluation of Binding Energies on Interstellar Ices.

From the Water Dimer to Far-Reaching Surfaces

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The quality of astrochemical models is highly dependent on reliable binding energy (BE) values that consider the morphological and energetic variety of binding sites on the surface of ice-grain mantles. In this work, we present a comprehensive approach for the computation of the binding energy of small interstellar species on amorphous solid water (ASW) surfaces. Our approach is based on the refinement of the binding energy value on two different levels: the accuracy of the employed model chemistry and the ice surface approximation. We propose three tiers of model surfaces. The first model is a set of small water clusters up to the tetramer[1] , which is used to benchmark several density functional theory (DFT) methods to a highly accurate coupled cluster CCSD(T)/CBS reference, in order to choose an appropriate functional to describe the adsorbate-surface interaction. The second ASW model is spanned by a set of 15-20 amorphized 22-water molecule clusters, which allows to build a binding energy distribution, due to the statistical variety of unique binding sites on each cluster. Finally, we selected equilibrium structures of adsorbed molecules on a group of clusters of different sizes, to be used as a starting point to generate a balanced training set for Gaussian Moment Neural Network (GM-NN)[4] potentials, able to reproduce the adsorption site as well as the water hydrogen-bonding network, at an equivalent to coupled cluster curated hybrid-DFT/def2-TZVP level of theory. Using those ad hoc NN-potentials we built a periodic model composed of five ~500-water molecule surfaces and computed highly accurate binding energy distributions. We present the results for astrophysically important CO and HCO species using the described procedure.