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The Reaction of Carbon Dioxide with Water Clusters: an Ab Initio Metadynamics Study GREGOIRE GALLET, FABIO PIETRUCCI, CECAM EPF Lausanne (CH), WANDA ANDREONI, CECAM and Institut de Théorie des Phénomènes Physique EPF Lausanne (CH) — Simulations are often invoked as aid to understand and optimize carbon capture and sequestration processes. The hydration of carbon dioxide (CO_2) offers an excellent test case for assessing the reliability of computational schemes. We present a density-functional-theory study of the reaction of CO_2 with water clusters. The first step was to validate DFT results in different approximations of the exchange and correlation functional with respect to quantum chemical methods for the structure, binding energies and vibrational frequencies of several isomers. Next, simulations of the reactions leading to the formation of carbonic acid were performed using metadynamics as accelerating procedure. This method allows us both to identify the reaction mechanisms and to obtain an estimate of the free energy barriers via the reconstruction of the free energy profiles. Comparisons were drawn with previous static calculations of the barriers. As reference, a similar calculation in liquid water will be presented.

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