Grid spacing in the direct space was selected based on a series of convergence tests. Molecular and dissociative adsorption energies of a single water molecule on each of the studied surfaces (Figure S1) as well as vacancy formation energies in the bulk (Figure S2) were examined as functions of the grid spacing. Overall, a clearly converging trend was observed in each case, with the value of grid spacing of ca. 0.20 Å being a reasonable compromise between accuracy and computational demand. Minor fluctuations in the data may be attributed to slight anisotropy in the direct lattice grid that varies from case to case. In some instances with the large grid spacing, the generated direct space grid fails providing sufficient multi-level hierarchy for accurate solution of the Poisson equation, resulting in numerical instabilities.

Figure S1: Molecular (left) and dissociative (right) adsorption energies of water on the studied m-ZrO\textsubscript{2} surfaces as functions of the grid spacing in the direct space: (T11) in red, (011) in green, and (T01) in blue. Solid lines represent fitted exponential decays to highlight the trends.

Figure S2: Formation energies of the three-fold (purple) and four-fold (orange) oxygen vacancies in the m-ZrO\textsubscript{2} bulk as functions of the grid spacing in the direct space. Solid lines represent fitted exponential decays to highlight the trends.