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First Principles Simulations of Hydrogen Storage via Spillover in MOF-5 DONALD SIEGEL, University of Michigan — Metal organic frameworks (MOF) have attracted considerable attention as hydrogen storage materials due to their high surface areas and ability to adsorb large quantities of hydrogen by weight (~ 12 wt. % at 100bar). However, as a consequence of the weak H_2 -MOF bonding interaction (~ 5 kJ/mol H_2), this uptake occurs only at cryogenic temperatures; at room temperature, gravimetric capacities do not exceed ~ 0.5 wt. %. As an ideal storage system would operate under ambient conditions, renewed interest in these materials has been sparked by recent experiments demonstrating RT uptake of ~ 4 wt. % via the so-called “spillover” mechanism [JACS **128**, 8136 (2006)]. In contrast to the conventional mechanism of MOF-based storage, where *molecular* H_2 bonds directly to the MOF, spillover employs a hydrogen dissociation catalyst to generate *atomic* hydrogen (H), presumably resulting in stronger H-MOF bonding. Recent computational studies of spillover have reported conflicting results regarding the nature of this interaction. In an effort to resolve these ambiguities and clarify the thermodynamics of MOF-based spillover, DFT calculations are used to evaluate binding energies for several H adsorption configurations on MOF-5. Importantly, our calculations avoid the cluster approximation to the MOF geometry—a source of significant uncertainty in previous studies—and account for finite-temperature contributions to the free energy of adsorption.

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