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Theoretical chemistry investigation of H12-M2-(DOBDP)3, a new mixed electron and proton conductivity MOF

Metal-organic frameworks (MOFs) with mixed proton and electron conductivity (MPEC) are promising materials for electrochemical energy systems. So far, a very small number of 2D MOFs have been shown to exhibit both types of conductivity. Two new 3D MOF candidates have recently been investigated in an experiment-theory collaboration [1], having the general formula H_{12}-M_2-$(DOBDP)_3$ with M being Fe^{3+} or Al^{3+}, and H_6-DOBDP is the 2,5-dihydroxy-1,4-benzenediphosphonic acid. Both MOFs follow up to 4 hydration-driven phase transitions, consisting of scaffold breathing and bending, rendering X-ray diffraction (XRD) resolution and other measurements challenging or impossible. Quantum chemistry methods at the Density Function Theory level of approximation with Periodic Boundary Conditions have therefore been carried out, following a recently published methodology [2] (calculation parameters, basis sets, and exchange-correlation functionals), in order to rationalize the behavior and the structural features of the scaffolds for the different hydration level phases. Geometry optimization, band structure, XRD, and density of state calculations allowed 1) to determine the iron centers spin multiplicity, 2) to show the integrity of the scaffolds even under strong distortions, 3) to propose mechanisms driving the transition phases, 4) to rationalize the ion-exchange experimental results, and 5) to better understand the electronic conductivity mechanism.

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