A long-standing goal in the field of supramolecular chemistry is the construction of catalysts that more adequately mimic the active sites of enzyme, i.e catalysts whose active sites are (i) confined, (ii) highly functionalized, and (iii) flexible. To this end, our group has introduced metal-organic framework (MOF) materials as scaffolds on which we can deliberately organize complex chemical functionality within confined, 3-dimensional space. MOF materials are porous, crystalline solids with pores of small-molecule dimensions, and whose cavity environments are highly tailorable. While the pores of MOF materials can be decorated with a wide variety of chemical functionality, the ability to uniformly multifunctionalize MOF materials remains a challenge. This presentation will describe strategies we have developed for the construction of uniformly multifunctionalized MOF materials, and our progress towards the synthesis of enzyme-inspired catalytic materials.