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Densification and Validation of Binderless MOF Monoliths Based On Temperature-Pressure Swing Hydrogen Storage and Delivery Conditions

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Transitioning to a sustainable energy source is the first and most crucial step in combating climate change. The critical bottleneck to using hydrogen (H₂), an energy carrier, as a stationary and transportation fuel has been the development of materials capable of providing a viable storage capacity. Metal-organic frameworks are a class of crystalline materials composed of metal ions or clusters coordinated by organic ligands, structuring a highly porous material having a high surface area and porosity. Despite, having significant characteristics for practical implementation as viable materials for H₂ storage, the poor packing of powdered MOF needs to be improved based on volumetric energy capacity. Thus, the densification of MOFs is crucial in reducing the void volume as they give way to enhanced gravimetric and volumetric storage capacities, improved kinetics, stability, and compatibility with storage tank designs. In this paper, we report the engineering of the best-performing high bulk-density monolithic MOFs in terms of their usable capacities, namely UiO-67 and MOF-177, without binders or pressure compaction. After shaping, monolithic MOF-177 and UiO-67 stores up to 49.95 g/L and 47 g/L at 77 K and 100 bar and deliver up to 47.02 g/L and 44 g/L operating in a combined temperature-pressure swing (TPS) delivery conditions between 77 K/100 bar 160 K/5 bar. These findings substantially improve the deliverable capacities of the mechanically robust densified MOFs based on onboard storage applications.

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