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Co-pelletization of a zirconium-based metal-organic framework (UiO-66) with polymer nanofibers for improved useable capacity in hydrogen storage

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Highlights

• Polymer nanofibers incorporated as layers in UiO-66 MOF by co-pelletization.

• Co-pelletization created hierarchical porosity in initially highly microporous MOF

• Microstructure of composites correlates with useable H₂ storage capacity.

• UiO-66/nanofiber monoliths improved low H₂ useable capacity in UiO-66.

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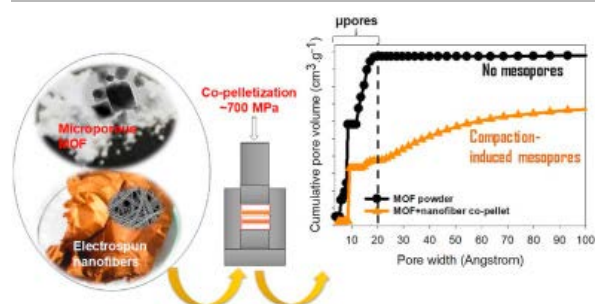
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Abstract

We report on a concept of co-pelletization using mechanically robust hydroxylated UiO-66 to develop a metal-organic framework (MOF) monolith that contains 5 wt% electrospun polymer nanofibers, and consists of an architecture with alternating layers of MOF and nanofiber mats. The polymers of choice were the microporous Polymer of Intrinsic Microporosity (PIM-1)

and non-porous polyacrylonitrile (PAN). Co-pelletized UiO-66/PIM-1 and UiO-66/PAN monoliths retain no less than 85% of the porosity obtained in pristine powder and pelletized UiO-66. The composition of the pore size distribution in co-pelletized UiO-66/PIM-1 and UiO-66/PAN monoliths is significantly different to that of pristine UiO-66 forms, with pristine UiO-66 forms showing 90% of the pore apertures in the micropore region and both UiO-66/nanofiber monoliths showing a composite micro-mesoporous pore size distribution. The co-pelletized UiO-66/nanofiber monoliths obtained improved useable H_2 capacities in comparison to pristine UiO-66 forms, under isothermal pressure swing conditions. The UiO-66/PIM-1 monolith constitutes the highest gravimetric (*and* volumetric) useable capacities at 2.3 wt% (32 g L^{-1}) in comparison to 1.8 wt% (12 g L^{-1}) and 1.9 wt% (29 g L^{-1}) obtainable in pristine UiO-66 powder and UiO-66 pellet, respectively. The co-pelletized UiO-66/PAN monolith, however, shows a significantly reduced surface area by up to 50% less in comparison to pristine UiO-66, but its pore volume only 13% less in comparison to pristine UiO-66. As a result, total gravimetric H_2 capacity of the co-pelletized UiO-66/PAN monolith is 50% less in comparison to that of pristine UiO-66, but crucially the useable volumetric H_2 capacity is 50% higher for the UiO-66/PAN monolith in comparison to pristine UiO-66 powder. The co-pelletization strategy provides a simple method for generating hierarchical porosity into an initially highly microporous MOF without changing the structure of the MOF through complex chemical modifications. The UiO-66/nanofiber monoliths offer improvements to the typically low H_2 useable capacities in highly microporous MOFs, and open new opportunities towards achieving system-level H_2 storage targets.

Graphical abstract



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Keywords

Co-pelletization; Hydrogen useable capacity; UiO-66;
Metal-organic framework; Hierarchical porosity

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