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Experimental demonstration of dynamic temperature-dependent behavior of UiO-66 metal-organic framework: Compaction of hydroxylated and dehydroxylated forms of UiO-66 for high-pressure hydrogen storage

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Abstract

High-pressure (700 MPa or ∼100 000 psi) compaction of dehydroxylated and hydroxylated UiO-66 for H2 storage applications is reported. The dehydroxylation reaction was found to occur between 150 and 300 °C. The H2 uptake capacity of powdered hydroxylated UiO-66 reaches 4.6 wt % at 77 K and 100 bar, which is 21% higher than that of dehydroxylated UiO-66 (3.8 wt %). On compaction, the H2 uptake capacity of dehydroxylated UiO-66 pellets reduces by 66% from 3.8 to 1.3 wt %, while for hydroxylated UiO-66 the pellets show only a 9% reduction in capacity from 4.6 to 4.2 wt %. This implies that the H2 uptake capacity of compacted hydroxylated UiO-66 is at least three times higher than that of dehydroxylated UiO-66, and therefore, hydroxylated UiO-66 is more promising for hydrogen storage applications. The H2 uptake capacity is closely related to compaction-induced changes in the porosity of UiO-66. The effect of compaction is greatest in partially dehydroxylated UiO-66 samples that are thermally treated at 200 and 290 °C. These compacted samples exhibit XRD patterns indicative of an amorphous material, low porosity (surface area reduces from between 700 and 1300 m2/g to ca. 200 m2/g and pore volume from between 0.4 and 0.6 cm3/g to 0.1 and 0.15 cm3/g), and very low hydrogen uptake (0.7–0.9 wt % at 77 K and 100 bar). The observed activation-temperature-induced dynamic behavior of UiO-66 is unusual for metal–organic frameworks (MOFs) and has previously only been reported in computational studies. After compaction at 700 MPa, the structural properties and H2 uptake of hydroxylated UiO-66 remain relatively unchanged but are extremely compromised upon compaction of dehydroxylated UiO-66. Therefore, UiO-66 responds in a dynamic manner to changes in activation temperature within the range in which it has hitherto been considered stable.