

Description of Additional Supplementary Files

File Name: Supplementary Data 1

Description: Simulated structures of op and cp phases of DUT-48, -46, -49, -50, and -151.

File Name: Supplementary Data 2

Description: Rietveld-refined structures of CD4-filled DUT-48, -49, and -50.

File Name: Supplementary Movie 1

Description: Cartoon of the pore filling mechanism upon methane adsorption in DUT-49op at 111 K.