Description of Additional Supplementary Files

File name: Supplementary Movie 1

Description: 5 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 40 MPa and 300 K for a 1×2×1 nanocell of MIL-53(Al). The subcells are color coded according to the volume. The closed-pore phase (V < 900 Å³), unstable region (900 Å³ ≤ V < 1400 Å³), and large-pore phase (V ≥ 1400 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 2

Description: 5 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 40 MPa and 300 K for a 2×2×2 nanocell of MIL-53(Al). The subcells are color coded according to the volume. The closed-pore phase (V < 900 Å³), unstable region (900 Å³ ≤ V < 1400 Å³), and large-pore phase (V ≥ 1400 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 3

Description: 5 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 40 MPa and 300 K for a 4×2×4 nanocell of MIL-53(Al). The subcells are color coded according to the volume. The closed-pore phase (V < 900 Å³), unstable region (900 Å³ ≤ V < 1400 Å³), and large-pore phase (V ≥ 1400 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 4

Description: 5 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 40 MPa and 300 K for a 6×2×6 nanocell of MIL-53(Al). The subcells are color coded according to the volume. The closed-pore phase (V < 900 Å³), unstable region (900 Å³ ≤ V < 1400 Å³), and large-pore phase (V ≥ 1400 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 5

Description: 5 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 40 MPa and 300 K for a 8×2×8 mesocell of MIL-53(Al). The subcells are color coded according to the volume. The closed-pore phase (V < 900 Å³), unstable region (900 Å³ ≤ V < 1400 Å³), and large-pore phase (V ≥ 1400 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 6

Description: 25 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 180 MPa and 300 K for a 8×2×8 mesocell of DMOF-1(Zn). The subcells are color coded according to the volume. The closed-pore phase (V < 1450 Å³), unstable region (1450 Å³ ≤ V < 2300 Å³), and large-pore phase (V ≥ 2300 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 7

Description: 5 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 80 MPa and 100 K for a 8×2×8 mesocell of MIL-53(Al)-F. The subcells are color coded according to the volume. The closed-pore phase (V < 1000 Å³), unstable region (1000 Å³ ≤ V < 1250 Å³), and large-pore phase (V ≥ 1250 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 8

Description: 5 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 60 MPa and 300 K for a 8×2×8 mesocell of MIL-53(Al)-F. The subcells are color coded according to the volume. The closed-pore phase (V < 1000 Å³), unstable region (1000 Å³ ≤ V < 1250 Å³), and large-pore phase (V ≥ 1250 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 9

Description: 5 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 60 MPa and 500 K for a 8×2×8 mesocell of MIL-53(Al)-F. The subcells are color coded according to the volume. The closed-pore phase (V < 1000 Å³), unstable region (1000 Å³ ≤ V < 1250 Å³), and large-pore phase (V ≥ 1250 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 10

Description: 15 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 80 MPa and 300 K for an empty 8×2×8 mesocell of CoBDP. The subcells are color coded according to the volume. The closed-pore phase (V < 1200 Å³), unstable region (1200 Å³ ≤ V < 2350 Å³), and largepore phase (V ≥ 2350 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 11

Description: 30 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 80 MPa and 300 K for a 8×2×8 mesocell of CoBDP with two methane molecules per unit cell. The subcells are color coded according to the volume. The closed-pore phase (V < 1200 Å³), unstable region (1200Å³ ≤ V < 2350 Å³), and large-pore phase (V ≥ 2350 Å³) are indicated by circles, crosses, and squares, respectively.

File name: Supplementary Movie 12

Description: 15 ps (N, P, $\boldsymbol{\sigma}_a = \mathbf{0}$, T) molecular dynamics simulation at 80 MPa and 300 K for a 8×2×8 mesocell of CoBDP with four methane molecules per unit cell. The subcells are color coded according to the volume. The closed-pore phase (V < 1200 Å³), unstable region (1200 Å³ ≤ V < 2350 Å³), and large-pore phase (V ≥ 2350 Å³) are indicated by circles, crosses, and squares, respectively.