Supporting Information for

Vibrational Mode Analysis of Hydrogen-Bonded Organic Frameworks (HOFs): Synchrotron Infrared Studies.

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Additional DFT images:

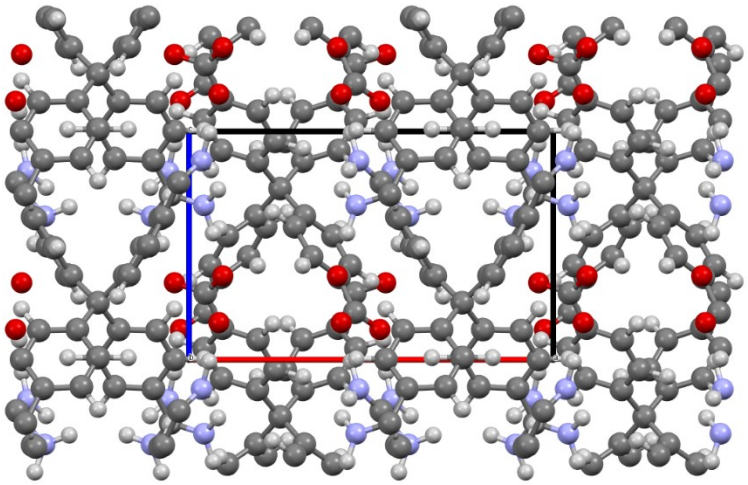

Figure S1: p-DFT optimized 2x2x2 supercell of **2** viewing down the crystallographic b-axis.

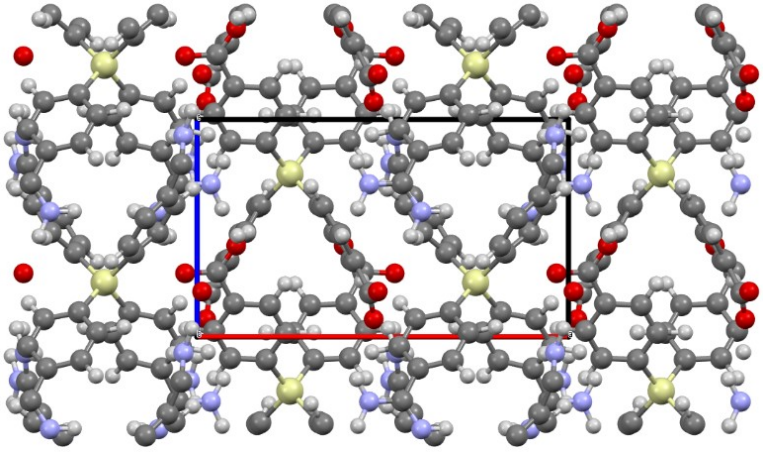

Figure S2: p-DFT optimized 2x2x2 supercell of **3** viewing down the crystallographic b-axis.

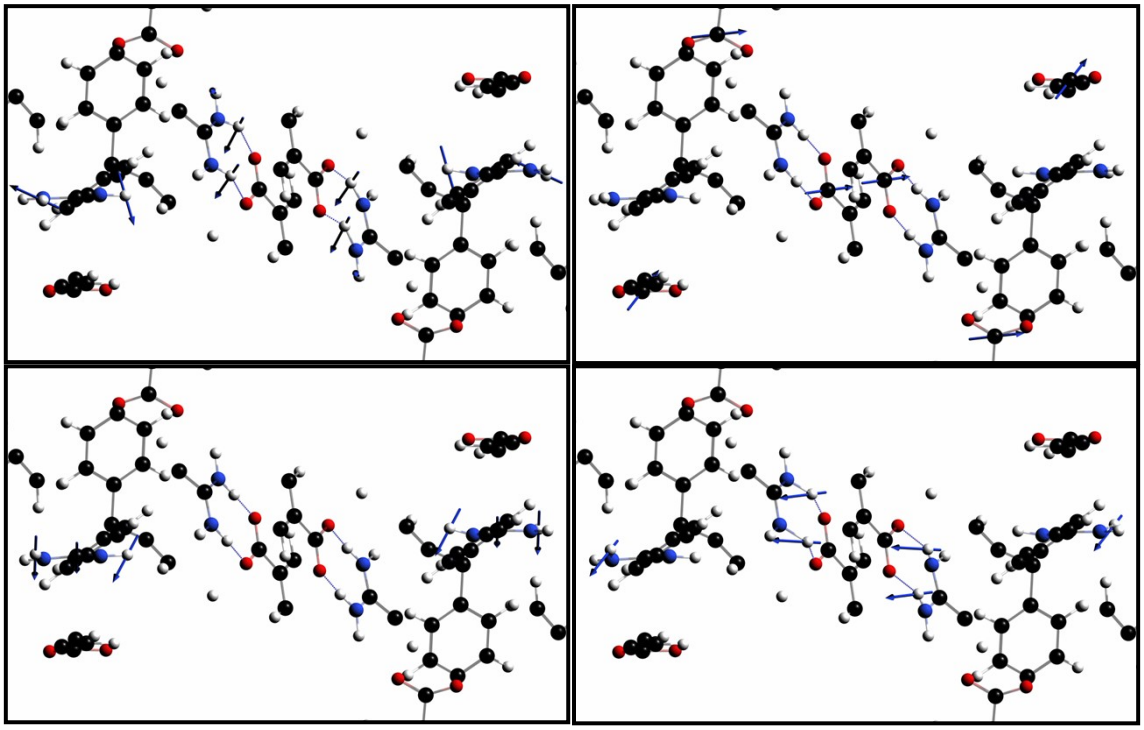

Figure S3: p-DFT eigenvectors (B3LYP-D3/6-311G(d)) for the 4:2 tetraamidinium:terephthalate (**1**) framework; **a:** 1030, **b:** 1349, **c:** 1691, and **d:** 2410 cm^{-1} vibrational modes.

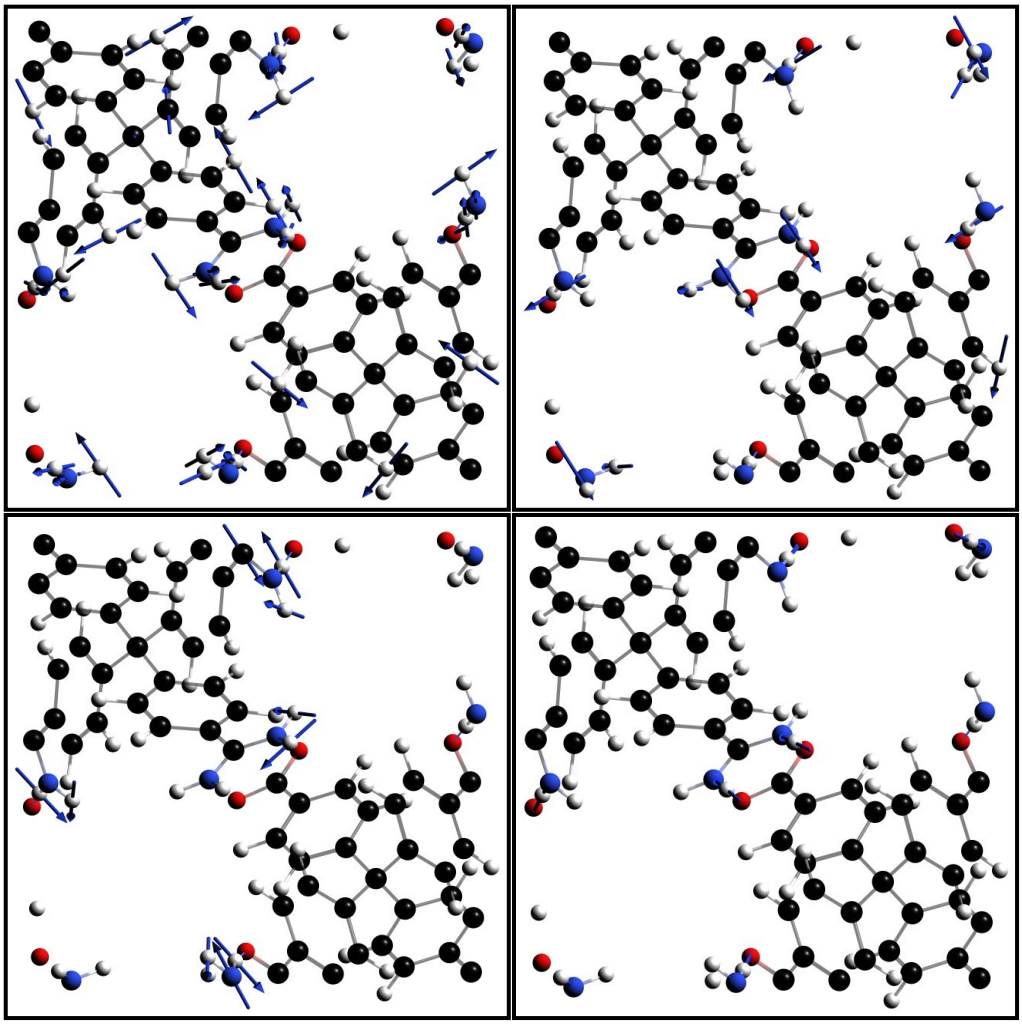

Figure S4: p-DFT eigenvectors (B3LYP-D3/6-311G(d)) for the 4:4 tetraamidinium:tetracarboxy (**2**) framework; **a:** 489, **b:** 1147, **c:** 1682, and **d:** 2479 cm^{-1} vibrational modes.

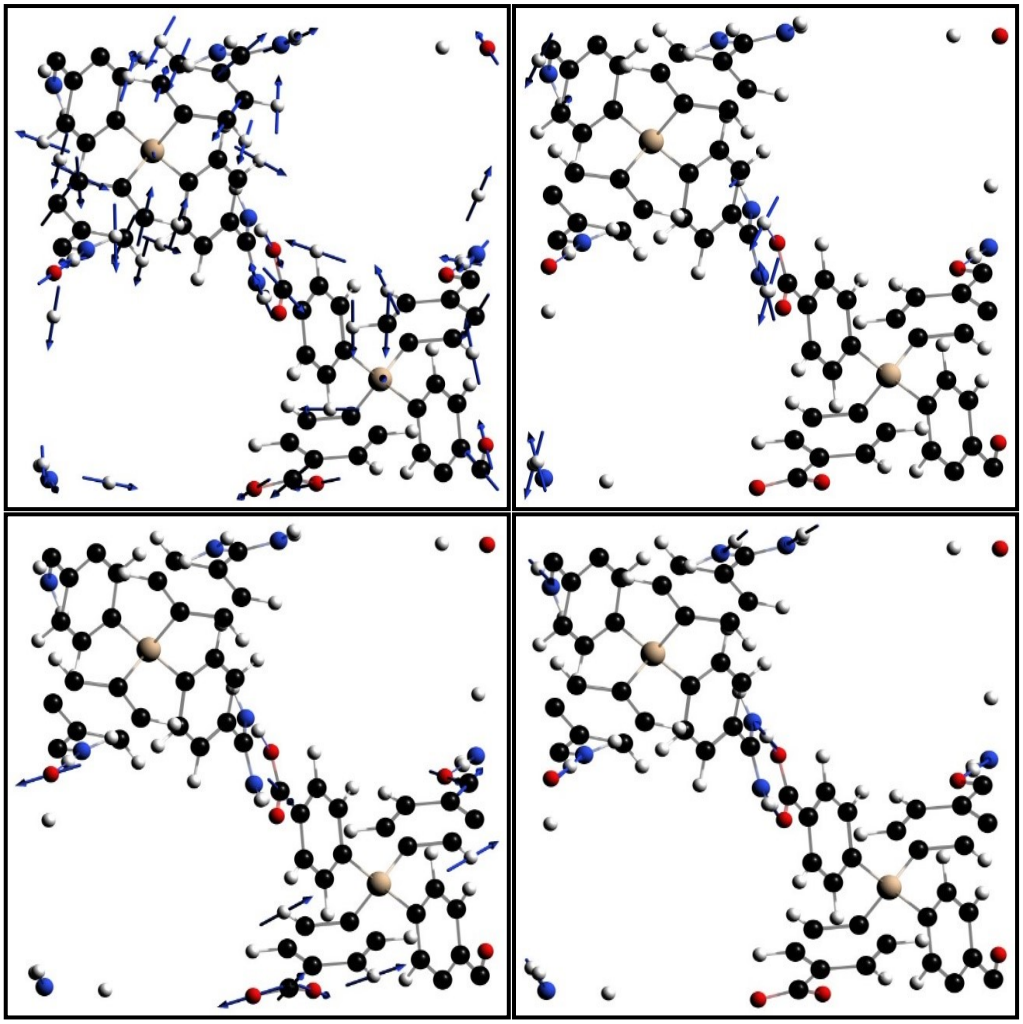

Figure S5: p-DFT eigenvectors (B3LYP-D3/6-311G(d)) for the 4:4 tetra(Si)amidinium: tetra(Si)carboxy (**3**) framework; **a:** 385, **b:** 1147, **c:** 1353, and **d:** 2507 cm^{-1} vibrational modes.

Characterization data for 3:

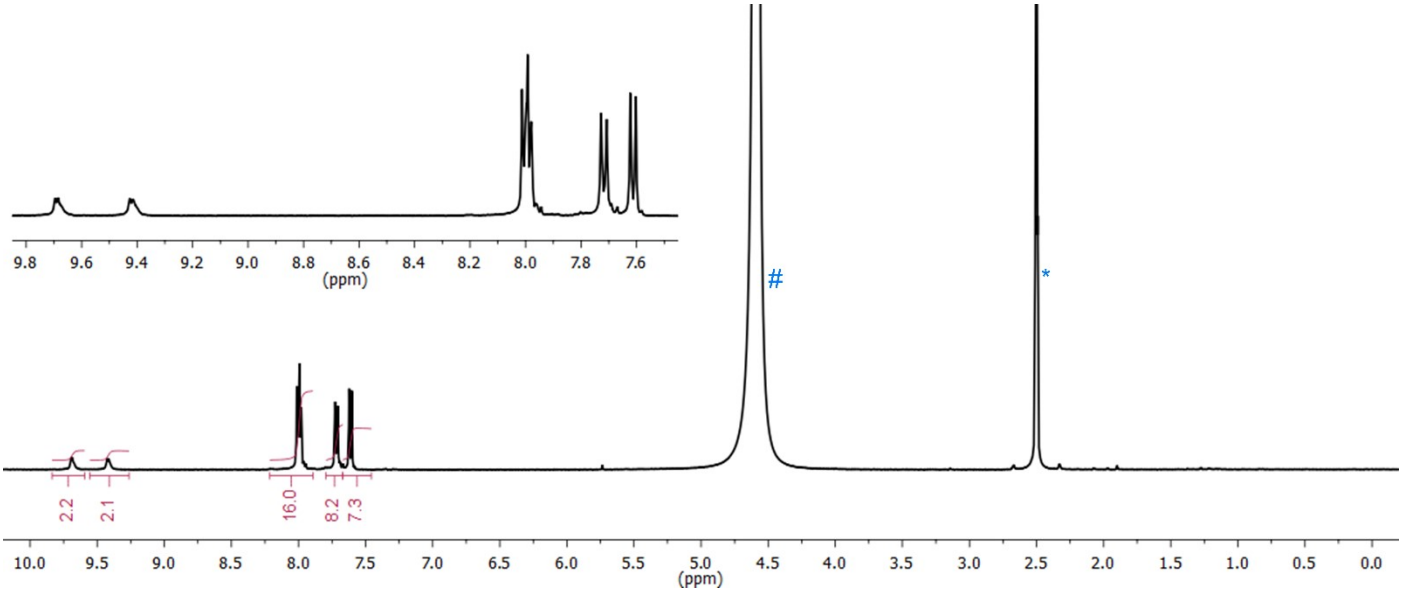

Figure S6. ^{1}H NMR spectrum of **3**; peak marked # is due to water, peak marked * is due to the residual NMR solvent. The integration of the amidinium N–H resonances is lower than expected, which we attribute to H/D exchange (d_6-DMSO containing a drop of 20% DCl in D_2O, 400 MHz, 298 K).

The TGA trace of **3** is shown in Figure S7. A weight loss of 15% is observed, which corresponds with that expected for the loss of ten water molecules, as indicated by elemental analysis.

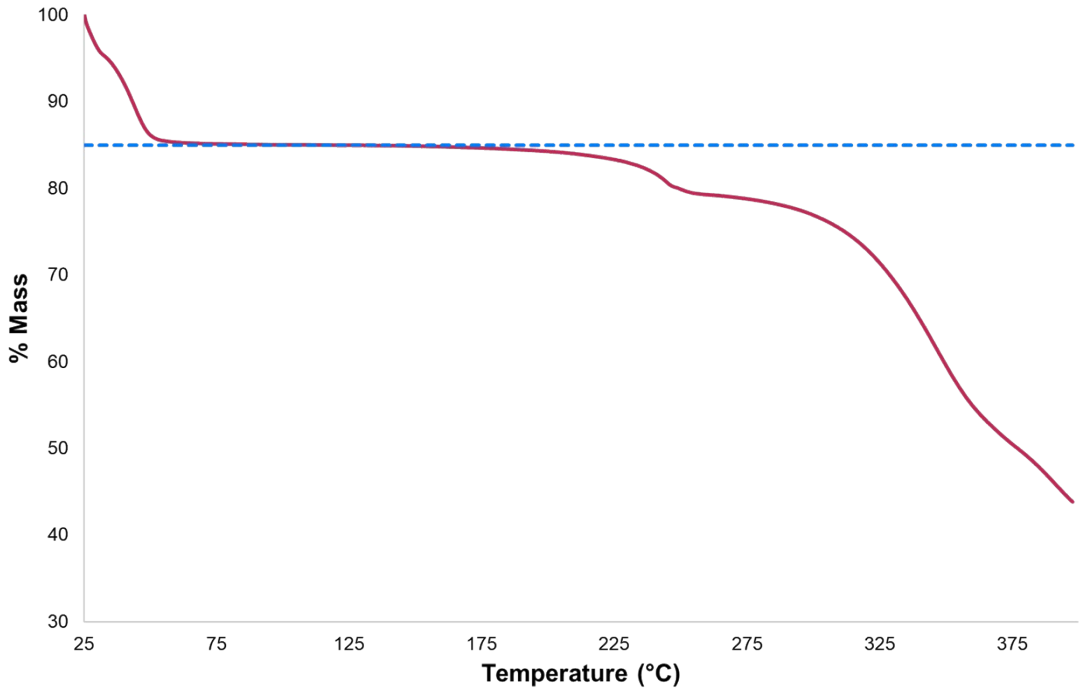

Figure S7. TGA trace of **3**, dotted line indicates 84.95 mass %, corresponding to the loss of ten waters to give anhydrous framework (recorded at 5 °C min^{-1} under N_2).

The PXRD trace of the air-dried framework (Figure S8) is consistent with that calculated based on the single crystal structure indicating the retention of crystallinity and framework structure upon drying.

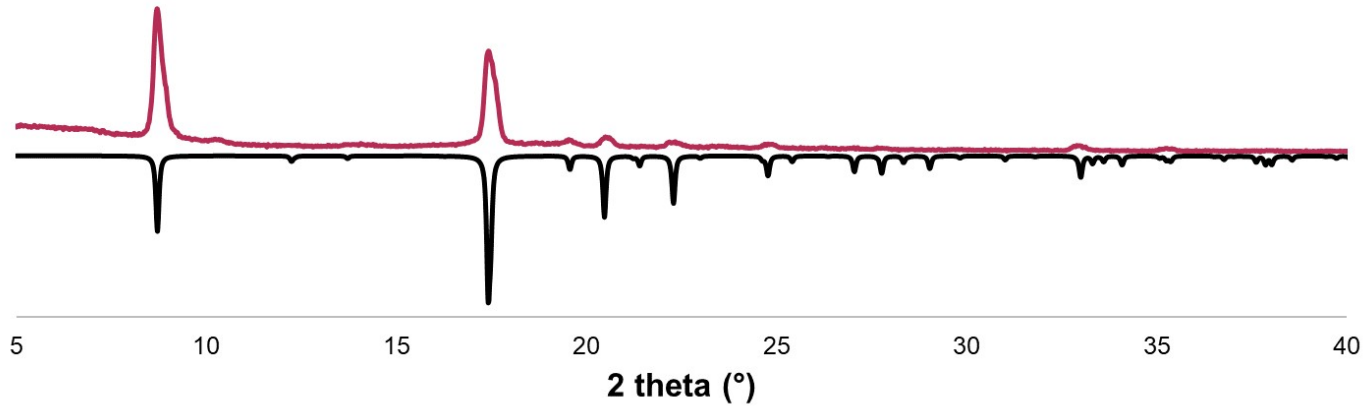

Figure S8. PXRD trace of air-dried **3**; observed trace shown pointing upwards (maroon), trace calculated based on single crystal structure shown pointing downwards (black).

Single crystal x-ray crystallography:

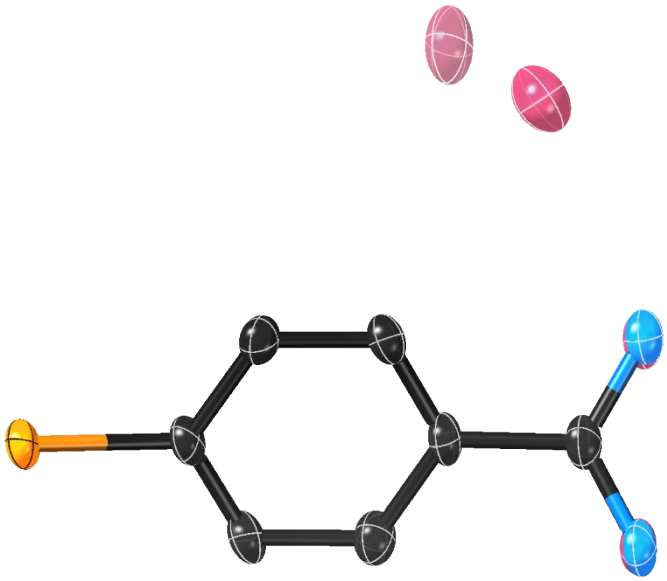

Figure S9. Thermal ellipsoid plot showing the asymmetric unit of **3**; ellipsoids are shown at 50% probability, hydrogen atoms are omitted for clarity.

Table S1. Crystallographic data for **3**.

	3
Radiation type	Cu Kα (λ = 1.54184 Å)
Temperature (K)	150
Formula	$C_{28}H_{32}N_8Si \cdot C_{28}H_{16}O_8Si \cdot 12H_2O$
Formula weight	1233.40
a (Å)	14.33058(3)
b (Å)	14.33058(3)
c (Å)	8.35856(2)
α (°)	90
β (°)	90
γ (°)	90
Unit cell volume (Å^3)	1716.560(8)
Crystal system	tetragonal
Space group	$P4_2/n$
Z	1
Reflections (all)	14784
Reflections (unique)	1688
R_{int}	0.093
R_1 [$I > 2\sigma(I)$]	0.094
wR_2 (all data)	0.233
CCDC number	2151777