Supporting Information

Effect of Steric Hindrance on the Interfacial Connection of MOF-on-MOF Architecture

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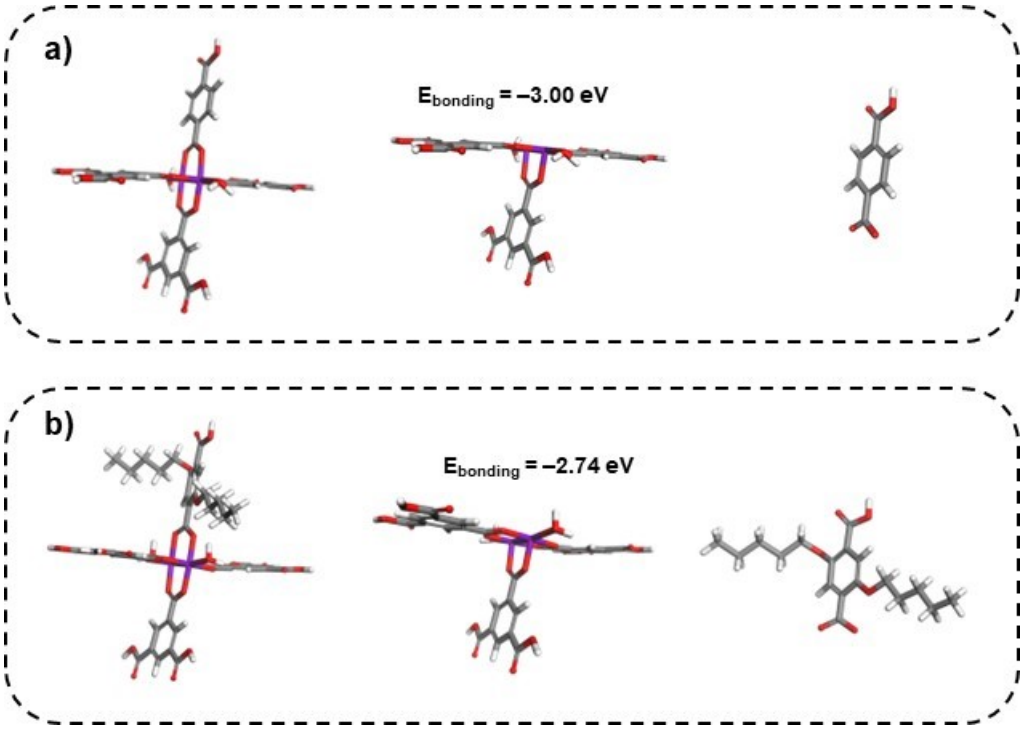

Figure S1. Bonding energy calculation cluster models. (a) HKUST@IR1, and (b) HKUST@IR5.

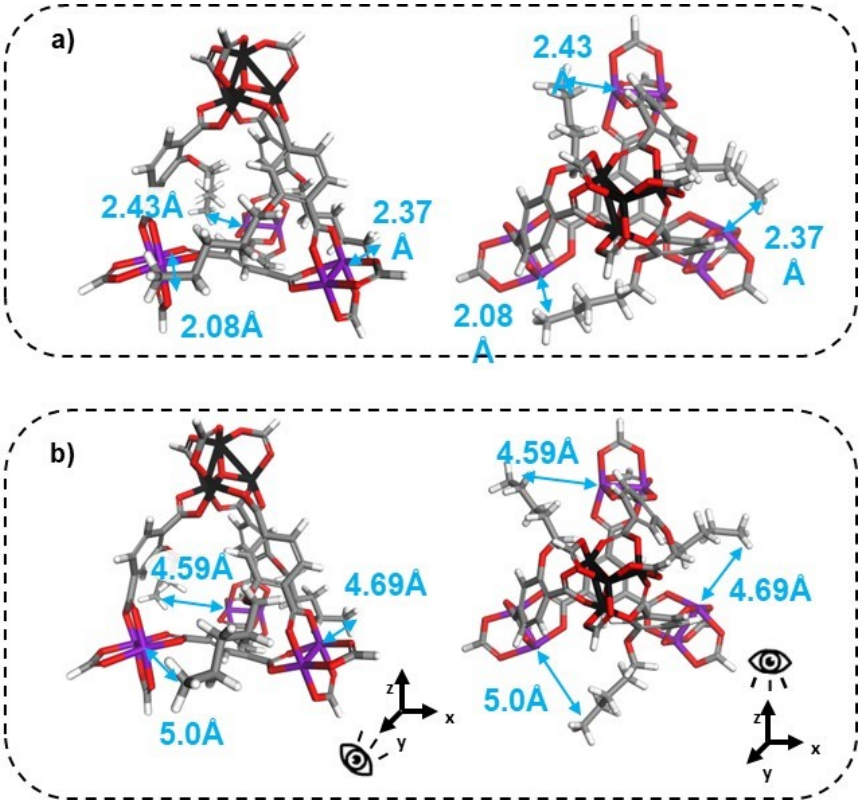

Figure S2. Interaction energy calculation cluster models. (a) Hindered structure extracted from final snapshot of MD simulation of HKUST@IR5 and (b) unhindered structure obtained by moving Pe groups far away with constrained bond lengths and angles.

Table S1. Interaction energy calculation results in Figure S2

	Energy (eV)
Figure S2a	-994.31
Figure S2b	-992.68
Interaction Energy	-1.63
Interaction Energy / number of pentoxy	-0.54

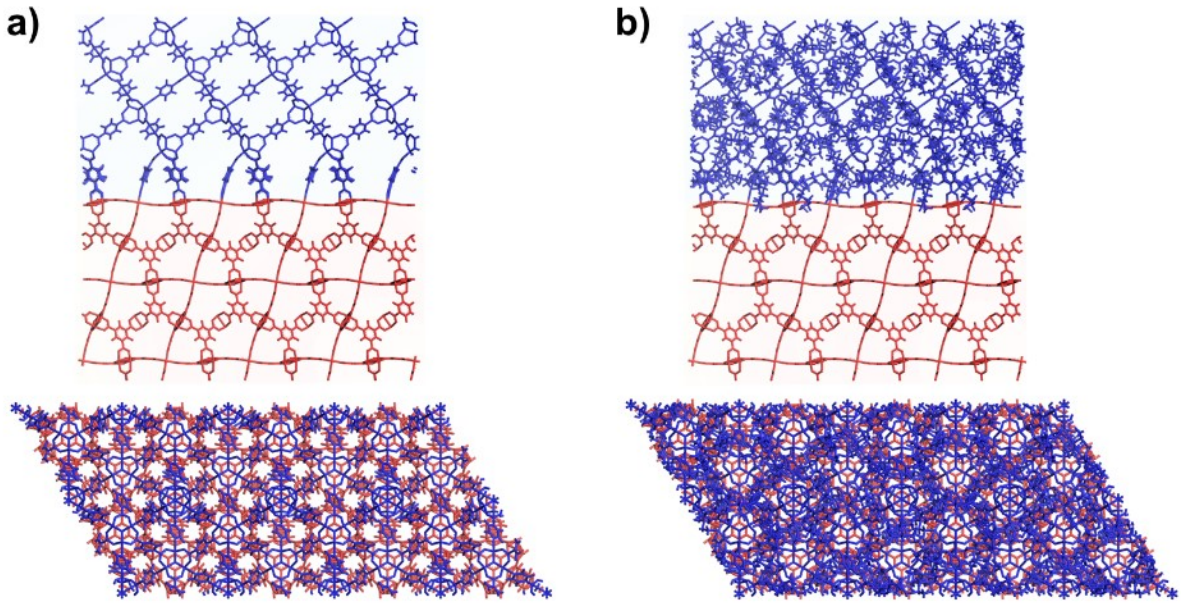

Figure S3. Computational structural models for the side view (top) and top view (bottom) of (a) HKUST@IR1 and (d) HKUST@IR5 MOF-on-MOFs. Blue: IRMOF shell; red: HKUST core.

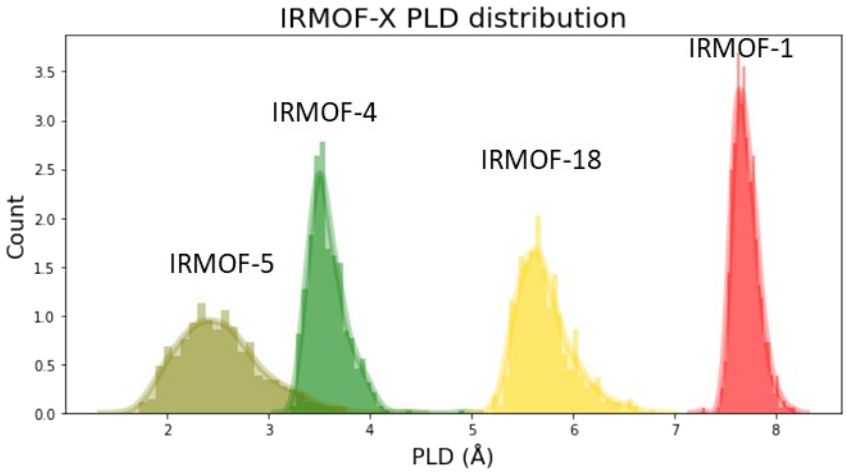

Figure S4. Pore Limiting Diameter (PLD) distribution plots for IRMOF-X (X=1, 4, 5, and18)

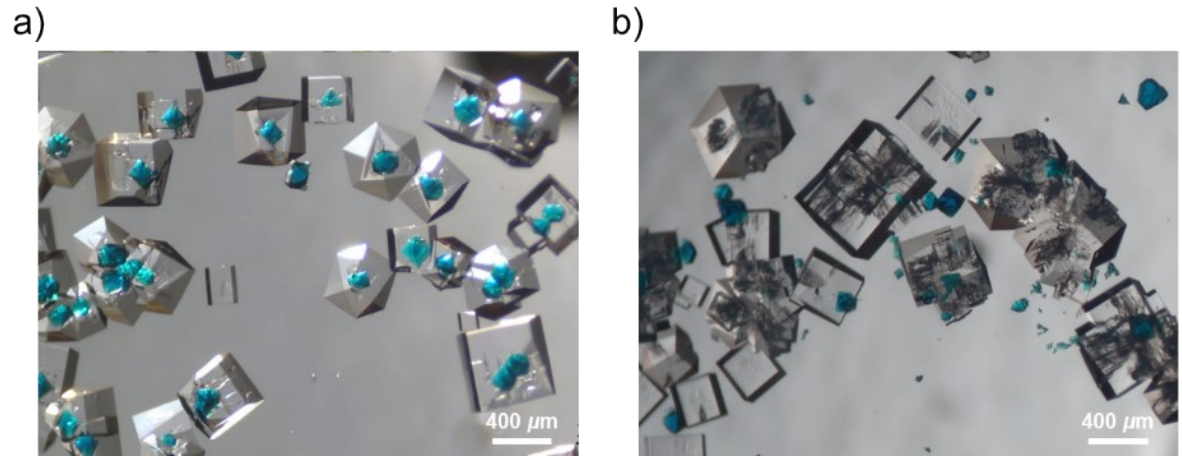

Figure S5. Optical microscope image of (a) HKUST@IR1 and (b) HKUST@IR5 synthesised by reference condition.[S1]

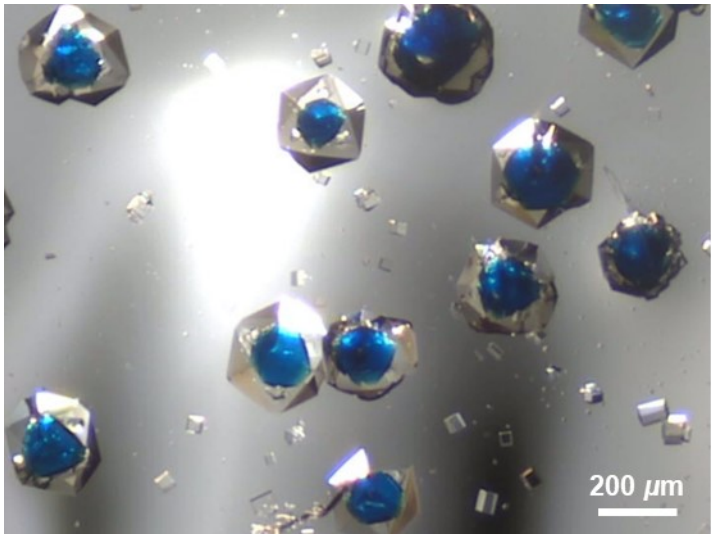

Figure S6. Optical microscope image of HKUST@IR5 synthesised by optimised condition.

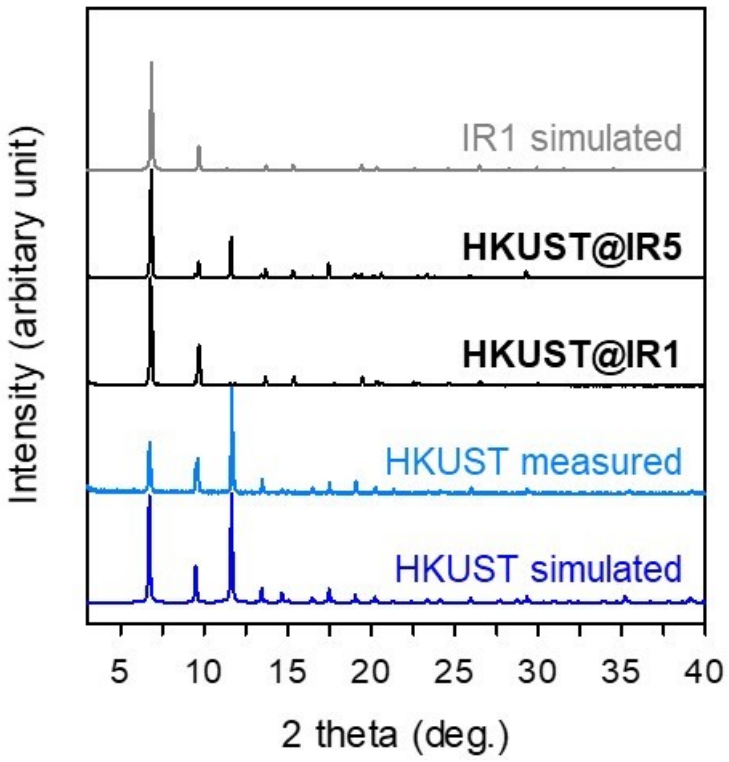

Figure S7. PXRD patterns of HKUST, HKUST@IR1 and HKUST@IR5.

Table S2. Comparison of cell parameters for HKUST, IR1, and IR5 obtained from crushed core-shell crystals with reference data.

	IR1 (EDUSIF) / this work	HKUST (XAMDUM) / this work	IR5 (EDUTEC) / this work
Crystal system	cubic	cubic	cubic
Space group	*Fm-3m*	*Fm-3m*	*Pm-3m*
Unit cell parameter a = b = c (Å)	25.8320(5) / 25.664(3)	26.3368(12) / 26.288(3)	12.8818(12) / 12.8910(15)
α = β = γ (°)	90 / 90	90 / 90	90 / 90
Z	8	8	1
Cell parameter difference (%)	2.4		1.9

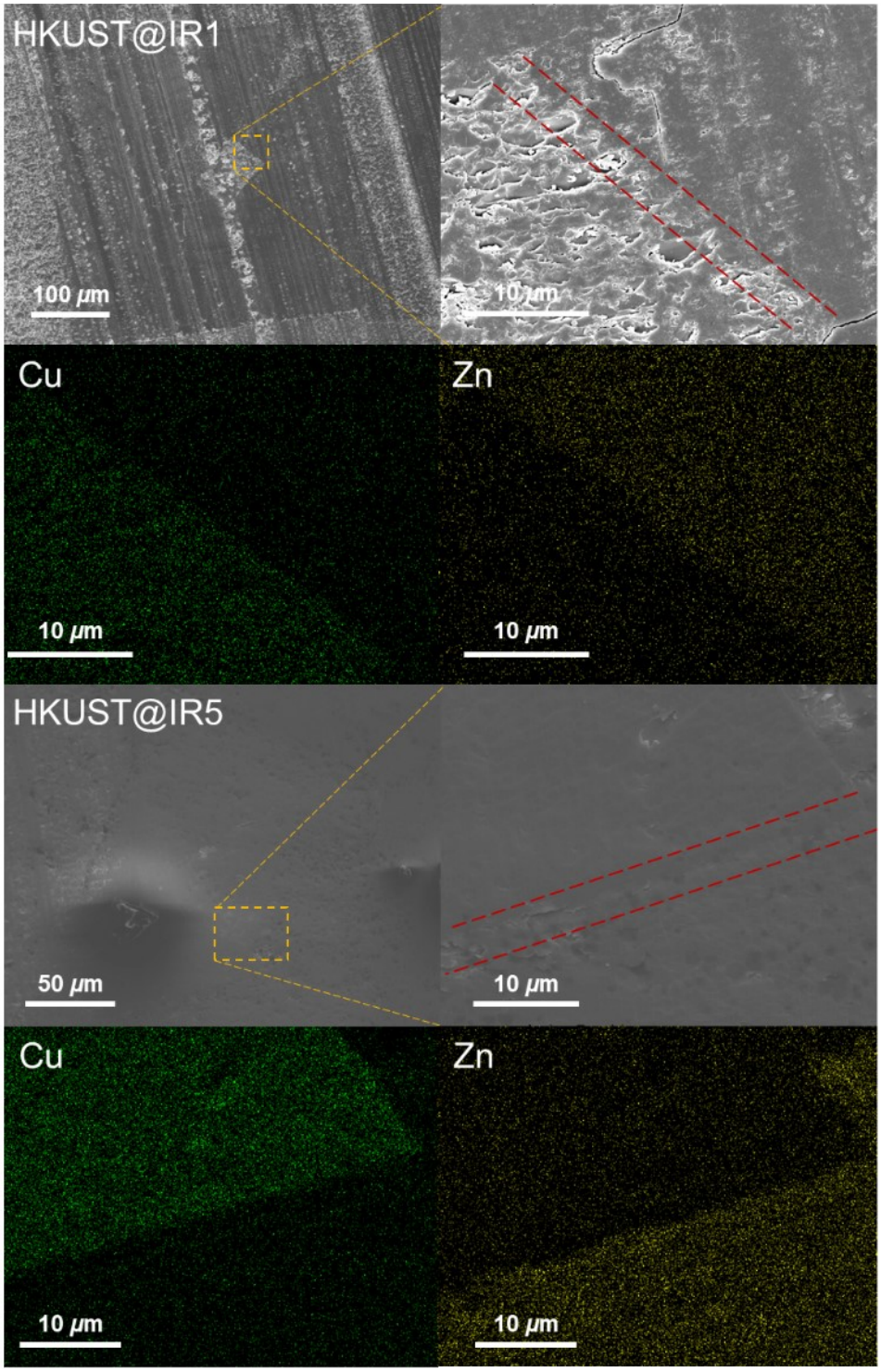

Figure S8. SEM images along with the corresponding EDS elemental maps showing the cross-sectional interface of HKUST@IR1 and HKUST@IR5. Red dotted line indicates the interface of MOF-on-MOF.

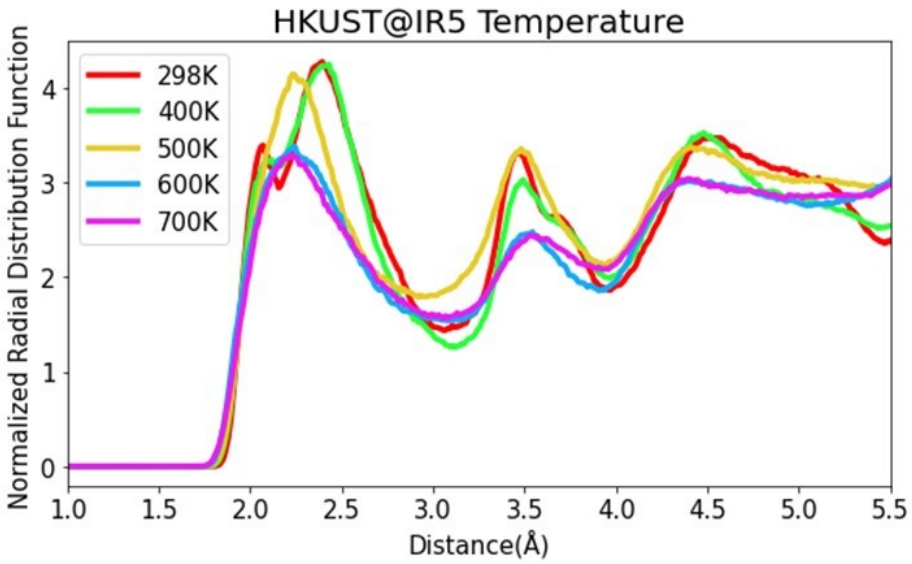

Figure S9. RDF calculation results with respect to the temperature. As the temperature increased, the normalized RDF values were decreased but the position of closest distance were consistent.

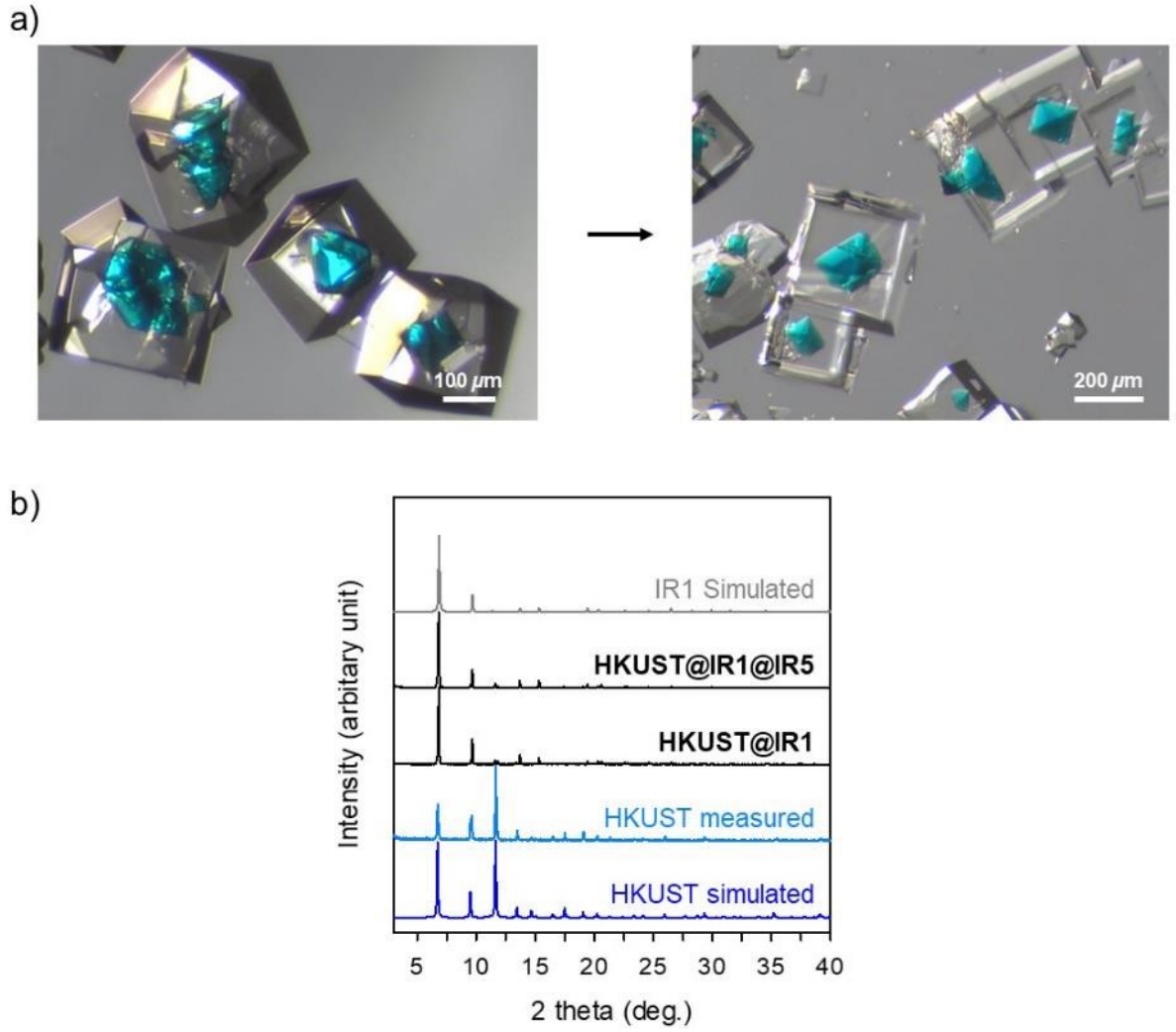

Figure S10. (a) Optical microscope images of HKUST@IR1 (left), HKUST@IR1@IR5 (right), and (b) PXRD patterns (bottom).

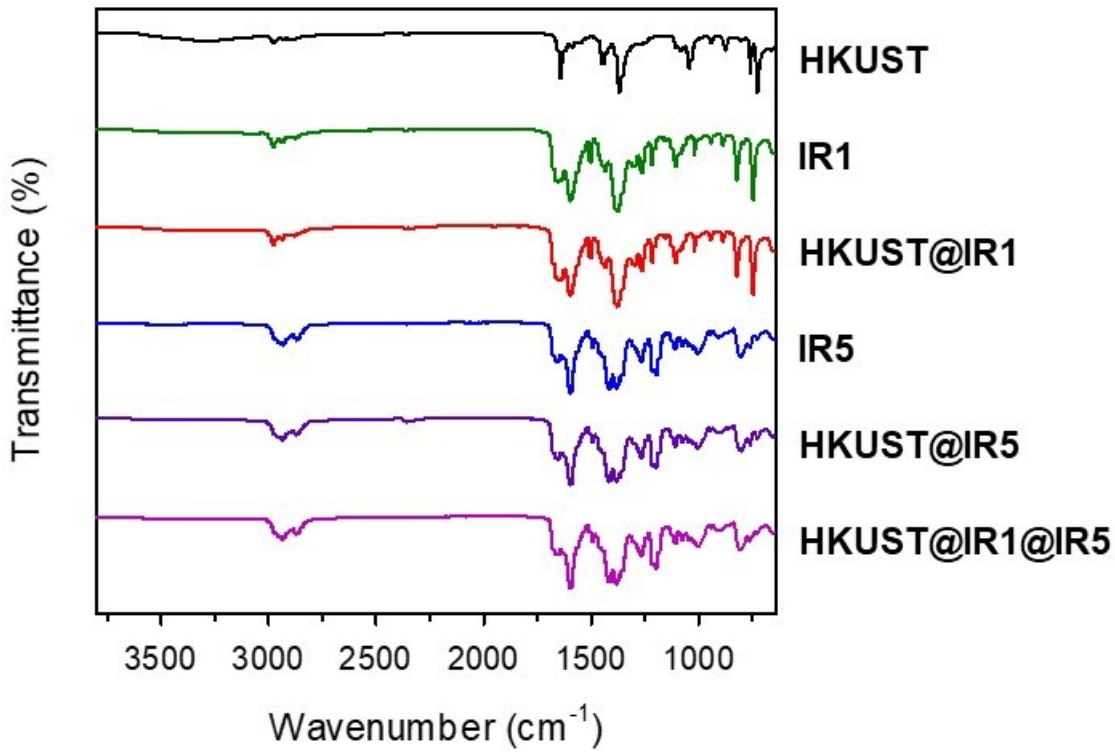

Figure S11. FT-IR spectra of MOFs.

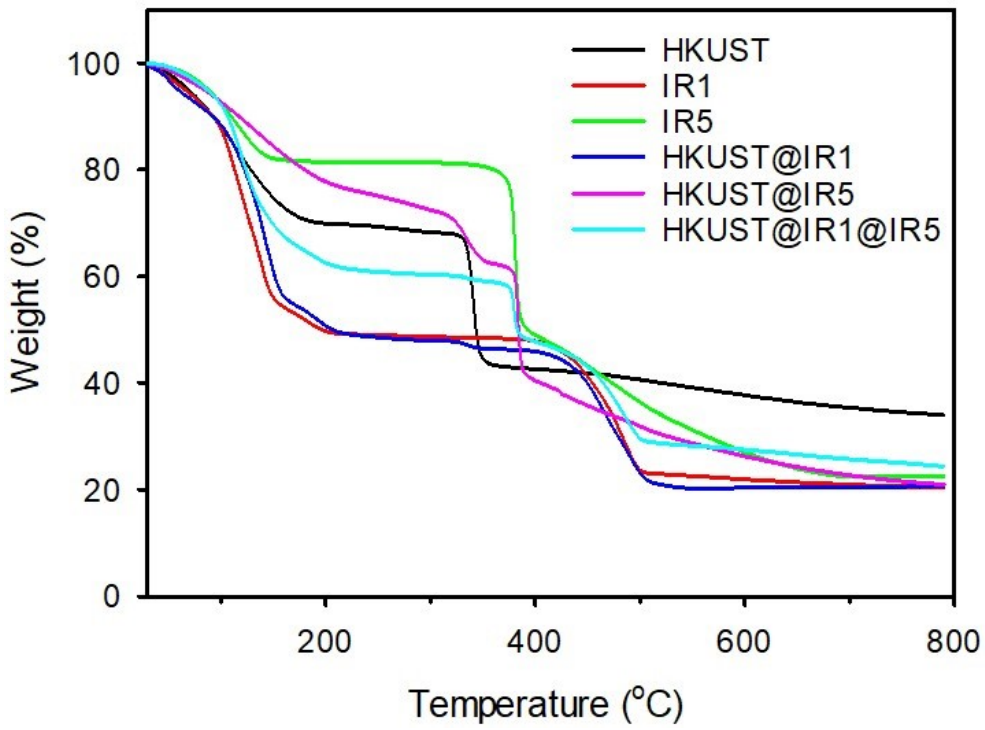

Figure S12. TGA graphs of MOFs.

Supplementary references

S1. O. Kwon, J. Y. Kim, S. Park, J. H. Lee, J. Ha, H. Park, H. R. Moon and J. Kim, *Nat. Commun.*, 2019, **10**, 3620.