Supporting Information for

Ultrastable Thorium Metal-Organic Frameworks for Efficient Iodine Adsorption

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Tables and Figures

Table S1. Crystallographic Data for **Th-SINAP-8@I_2/cyclohexane** and **Th-SINAP-8@I_2 vapour**.

Code	**Th-SINAP-8 @I_2/cyclohexane**	**Th-SINAP-8 @I_2 vapor**
CCDC number	1966326	1966327
Formula	$C_{12}H_6O_{15.33}Th$	$C_9H_{4.5}O_{12.75}Th_{0.75}$
Mass	627.54	490.66
Habit	octahedral	octahedral
Space Group	*Fm*-3*m*	*Fm*-3*m*
a (Å)	24.9934(4)	24.9545(11)
b (Å)	24.9934(4)	24.9545(11)
c (Å)	24.9934(4)	24.9545(11)
V (Å^3)	15612.6(7)	15540(2)
Z	24	32
T (K)	139	139
λ (Å)	0.71073	0.71073
Max. 2θ (°)	54.896	54.99
ρ_{calcd} (g cm^{-3})	1.602	1.678
μ (mm^{-1})	5.786	5.822
GoF on F^2	1.087	1.061
R_1, wR_2 [I > 2σ(I)]	0.0613, 0.1643	0.0859, 0.2347
R_1, wR_2 (all data)	0.0714, 0.1749	0.1033, 0.2540
$(\Delta\rho)_{max}$, $(\Delta\rho)_{min}$/e (Å^{-3})	3.96, -1.81	2.90, -1.87

Table S2. Selected bond distances [Å] for **Th-SINAP-7** and **Th-SINAP-8**.

Assignment	Bond Distances	**Th-SINAP-7**	Bond Distances	**Th-SINAP-8**
Th−O^{2-}/OH^-	Th(1)-O(2) ×4	2.48(2)	Th(1)-O(2) ×4	2.44(3)
	Th(1)-O(2A) ×4	2.29(2)	Th(1)-O(2A) ×4	2.28(4)
Th−O_{CO-}	Th(1)-O(1) ×4	2.482(11)	Th(1)-O(1) ×4	2.457(15)
Th−O_w	Th(1)-O(3)	2.67(2)	Th(1)-O(3)	2.66(4)
	Th(1)-O(3A)	2.68(4)		

Table S3 Kinetic parameters of the pseudo-second-order model[a] for iodine adsorption on **Th-SINAP-7** and **Th-SINAP-8**.

	c_0 (mg·kg^{-1})	M/V (mg·g^{-1})	q_0 (mg·g^{-1})	Removal (%)	Second-order kinetic model			
					q_e (mg·g^{-1})	h (mg·g^{-1}·h^{-1})	k (g·mg^{-1}·h^{-1})	R^2
Th-SINAP-7	200	2.5	80	86.84	91.231	71.5786	0.0086	0.998
Th-SINAP-8	200	2.5	80	99.99	100.00	178.4275	0.0178	0.982

[a]$t/q_t = 1/h + t/q_e$ (where q_t, q_e are the amounts of adsorbate at certain time t or at equilibrium time, h is the initial adsorption rate, $h = kq_e^2$, and k is the rate constant).

Table S4. Fitting results of the sorption isotherms according to the Langmuir[a] and Freundlich[b] equations

Sample	Langmuir			Freundlich		
	Q_m (mg/g)	K_L (L/mg)	R^2	k_F (L^n/mol^{n-1}g)	n	R^2
Th-SINAP-7	93.49	0.110	0.65	32.66	0.174	0.98
Th-SINAP-8	232.77	0.127	0.90	76.88	0.176	0.94

[a]The linear equation of the Langmuir isotherm model is expressed as followed:

$$\frac{C_e}{q_e} = \frac{1}{q_m k_L} + \frac{C_e}{q_m}$$

Where q_m is the maximum sorption capacity corresponding to complete monolayer coverage (mg/g) and k_L is a constant indirectly related to sorption capacity and energy of sorption (L/mg), which characterizes the affinity of the adsorbate with the adsorbent. The linearized plot was obtained when we plotted C_e/q_e against C_e and q_m and k_L could be calculated from the slope and intercept.

[b]The linear equation of Freundlich isotherm model can be expressed by:

$$\ln q_e = \ln k_F + \frac{1}{n} \ln C_e$$

Where k_F and n are the Freundlich constants related to the sorption capacity and the sorption intensity, respectively. The linear plot was obtained by plotting $\ln q_e$ against $\ln C_e$, and the values of k_F and n were calculated from the slope and intercept of the straight line.

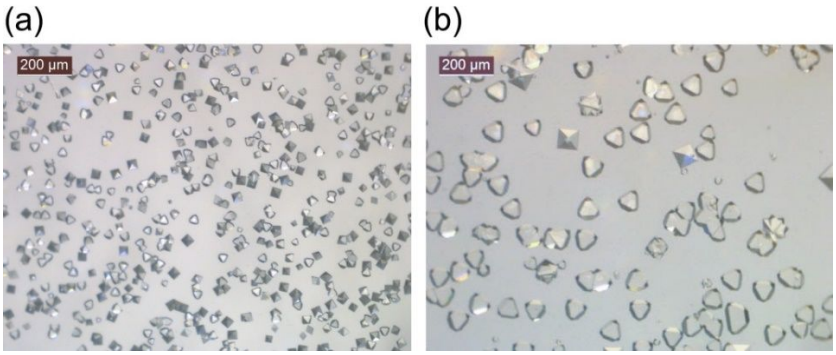

Figure S1. Micrographs of single crystals of (a) **Th-SINAP-7** and (b) **Th-SINAP-8**.

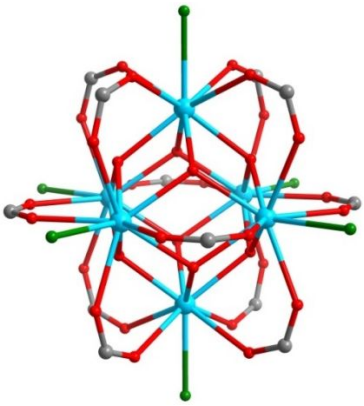

Figure S2. The $Th_6(\mu_3\text{-}O)_4(\mu_3\text{-}OH)_4(NDC)_6(H_2O)_6$ SBU in **Th-SINAP-7** and **Th-SINAP-8**. O atoms from μ_3-OH/-O and NDC are shown in red, O atoms from water are shown in green, Th atoms are shown in sky blue and C atoms are shown in gray.

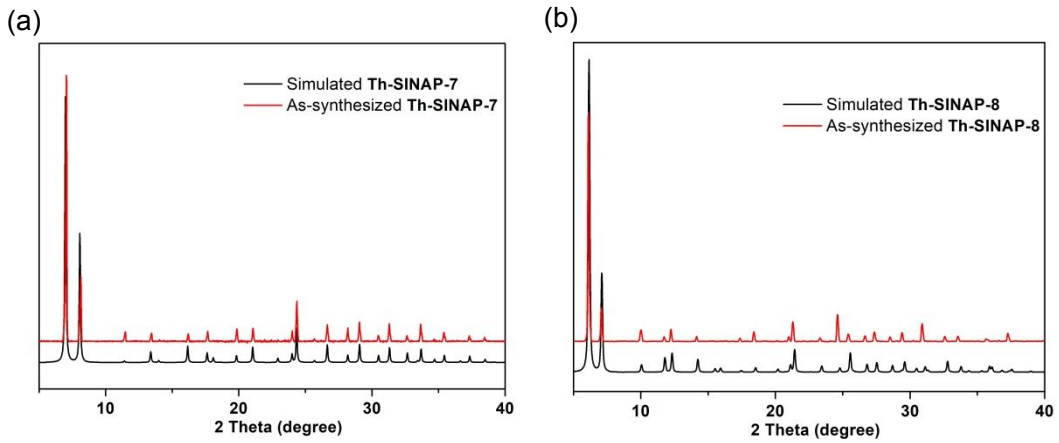

Figure S3. PXRD patterns of as-synthesized (a) **Th-SINAP-7** and (b) **Th-SINAP-8**.

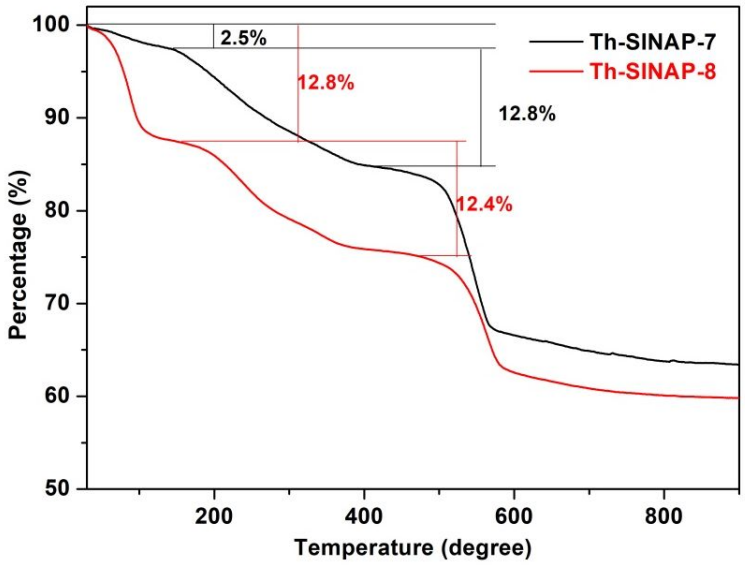

Figure S4. TGA curves of **Th-SINAP-7** and **Th-SINAP-8**.

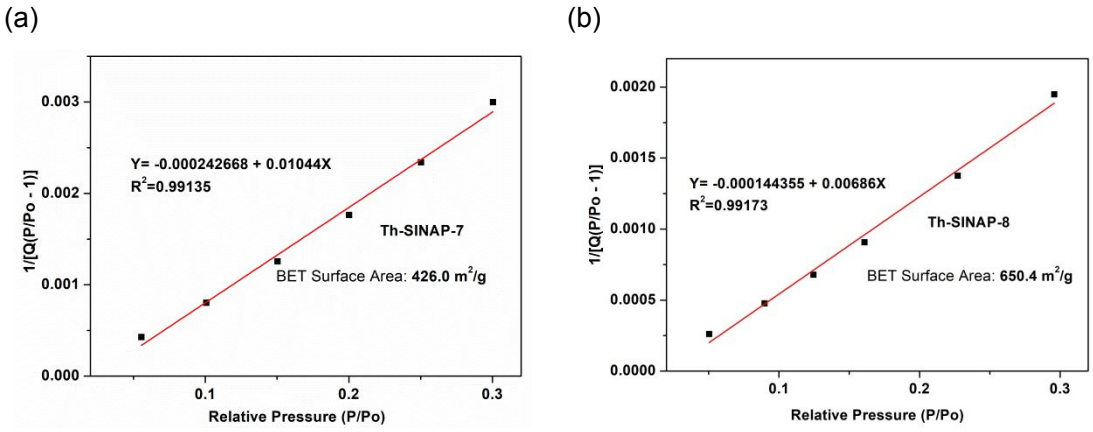

Figure S5. BET surface area plots for (a) **Th-SINAP-7** and (b) **Th-SINAP-8**.

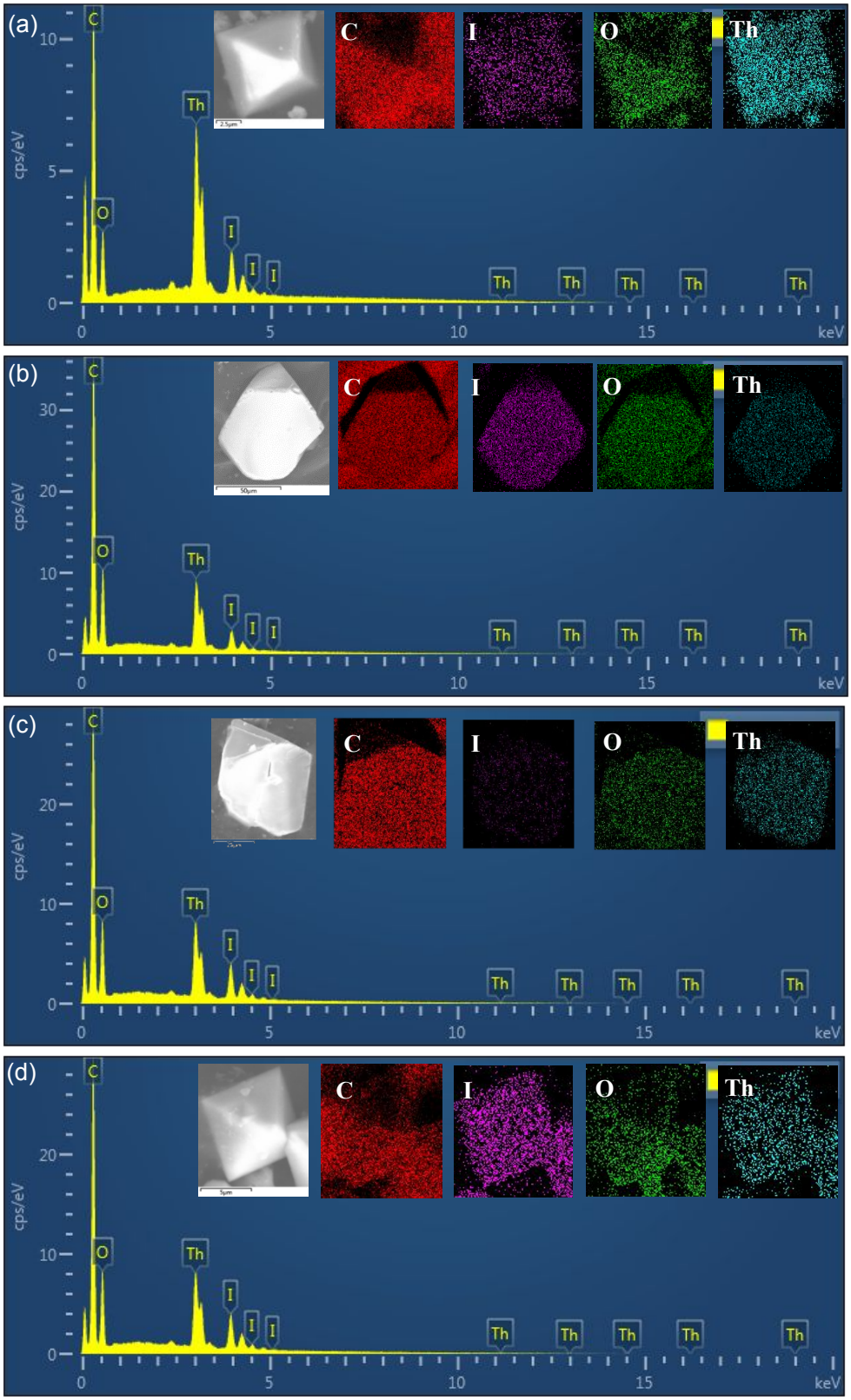

Figure S6. SEM-EDS mapping and EDS spectra profiles of (a) **Th-SINAP-7** after absorption of I_2 from cyclohexane, (b) **Th-SINAP-8** after absorption of I_2 from cyclohexane, (c) **Th-SINAP-7** after absorption of I_2 vapor, and (d) **Th-SINAP-8** after absorption of I_2 vapor.

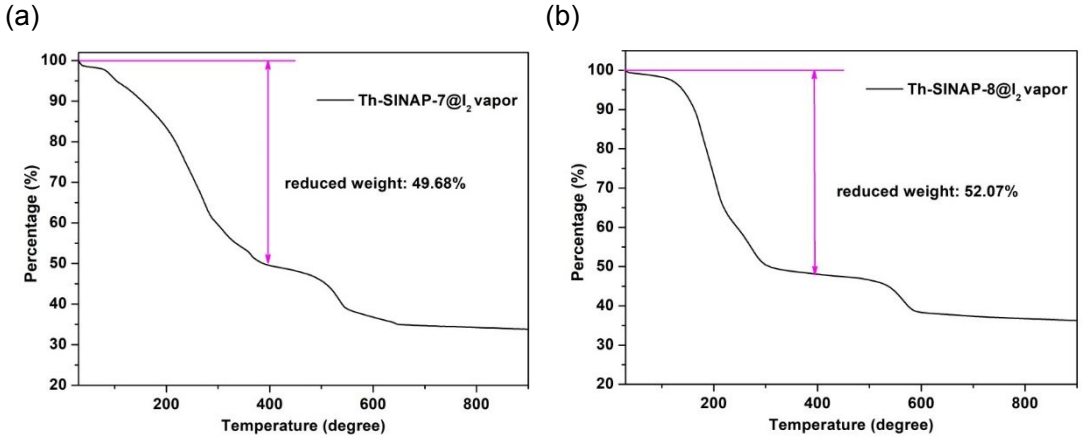

Figure S7. The TGA trace of I_2 vapour adsorbed (a) **Th-SINAP-7** and (b) **Th-SINAP-8**.

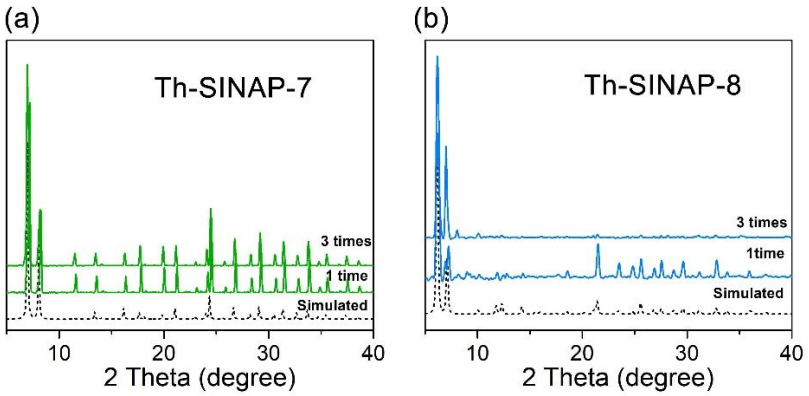

Figure S8. PXRD patterns of (a) **Th-SINAP-7** and (b) **Th-SINAP-8** after one and three cycles of iodine vapor adsorption.

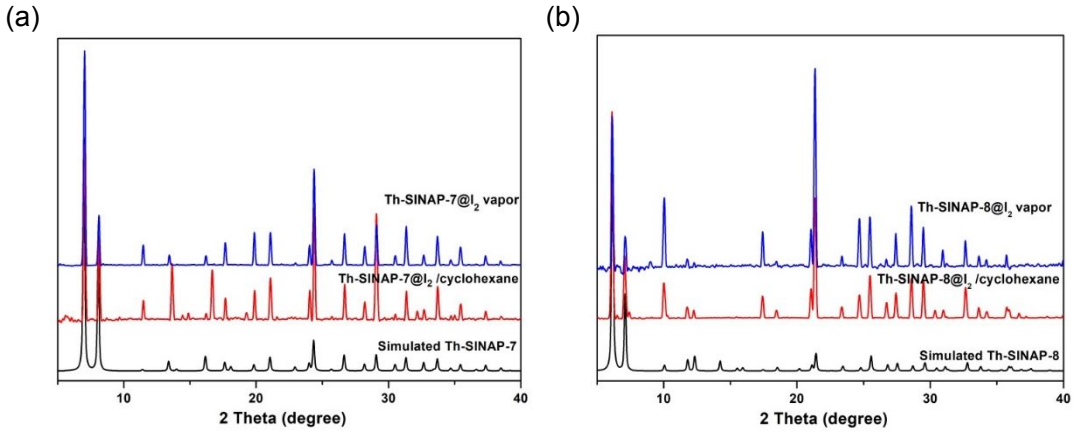

Figure S9. PXRD patterns of (a) **Th-SINAP-7** and (b) **Th-SINAP-8** after absorbing I_2/cyclohexane or I_2 vapor.

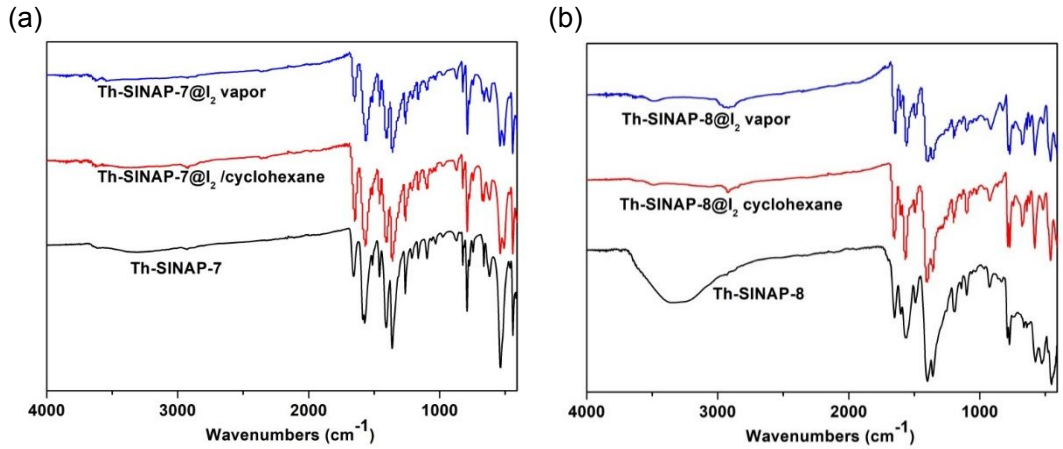

Figure S10. IR spectra of (a) **Th-SINAP-7** and (b) **Th-SINAP-8** after absorbing I_2/cyclohexane or I_2 vapor.