

Table S1. Properties and photocatalytic performances of different materials in the partial oxidation of cyclohexane into cyclohexanol (CHAol) and cyclohexanone (CHAone).^[a]

Entry	Sample	$S_{\text{BET}}/\text{m}^2 \text{g}^{-1[\text{b}]}$	$V_p/\text{cm}^3 \text{g}^{-1[\text{c}]}$	$D_p/\text{nm}[\text{c}]$	Yield/ μmol (selectivity/ $\%[\text{d}]$)			Conversion / $\%[\text{e}]$	TON ^[f]
					CHAol	CHAone	CO ₂		
1	P25 TiO ₂	49	-	-	35 (72)	8 (17)	32 (11)	0.26	-
2	TS-1 zeolite	467	0.3 ^[g]	<0.6 ^[h]	2 (40)	3 (60)	n.d.	0.027	0.4
3	SBA-15	1085	1.4	8.1	n.d.	n.d.	n.d.	-	-
4	Ti _{0.02} -SBA	899	1.1	7.1	4 (40)	6 (60)	n.d.	0.054	1
5	Ti _{0.02} -SBA ^[i]				n.d.	n.d.	trace	-	-
6	Fe _{0.2} Ti _{0.02} -SBA	768	1.0	7.1	71 (50)	71 (50)	n.d.	0.77	27
7	Fe _{0.2} Ti _{0.02} -SBA ^[j]				13 (43)	17 (57)	n.d.	0.16	6
8	Fe _{0.2} Ti _{0.02} -SBA ^[k]				1 (25)	3 (75)	n.d.	0.022	0.8
9	Fe _{0.2} Ti _{0.02} -SBA ^[l]				84 (41)	119 (59)	n.d.	1.1	39
10	Fe _{0.2} Ti _{0.02} -SBA ^[m]				67 (50)	66 (50)	n.d.	0.72	26
11	Fe _{0.2} Ti _{0.02} -SBA ^[n]				272 (63)	161 (37)	n.d.	2.3	83
12	Fe _{0.02} Ti _{0.02} -SBA	876	1.0	7.1	38 (49)	40 (51)	n.d.	0.42	9
13	Fe _{0.7} -HUS	42	0.17	<0.8 ^[m]	n.d.	n.d.	n.d.	-	-
14	α -Fe ₂ O ₃ ^[o]	11	-	-	n.d.	n.d.	n.d.	-	-
15	α -FeO(OH) ^[o]	12	-	-	2 (67)	1 (33)	n.d.	0.016	-
16	Fe(acac) ₃	0	-	-	9 (47)	6 (31)	25 (22)	0.10	-
17	Fe _{0.01} -P25	43	-	-	13 (32)	24 (60)	19 (8)	0.22	-
18	Fe _{0.001} -P25	42			27 (62)	12 (28)	27 (10)	0.23	-

^[a] Conditions unless otherwise mentioned: stainless-steel reactor, 75 mL; photocatalyst, 30 mg; cyclohexane, 2 mL; solvent (O₂-saturated acetonitrile), 18 mL; irradiation, solar simulator ($\lambda > 300 \text{ nm}$, 1.5 AM) for 24 h. ^[b] Determined using the BET method. ^[c] Determined using the BJH plot. ^[d] $\{[\text{CHAol}] \text{ or } [\text{CHAone}] \text{ or } 1/6[\text{CO}_2]\} / \{[\text{CHAol}] + [\text{CHAone}] + 1/6[\text{CO}_2]\} \times 100$. ^[e] $\{[\text{CHAol}] + [\text{CHAone}] + 1/6[\text{CO}_2]\} / [\text{cyclohexane added}] \times 100$. ^[f] $\{[\text{CHAol}] + [\text{CHAone}]\} / [\text{Ti grafted}]$. ^[g] Determined using the t-plot. ^[h] Determined from the crystal structure and basal spacing for TS-1 and Fe_{0.7}-HUS, respectively. ^[i] $\lambda > 420 \text{ nm}$. ^[j] $\lambda > 480 \text{ nm}$. ^[k] 60 mg. ^[l] 48 h. ^[m] 60 mg and 48 h under O₂ atmosphere (250 kPa). ^[n] Determined from the basal spacing. ^[o] The particle sizes are 1 μm and 1-2 μm for α -Fe₂O₃ and α -FeO(OH), respectively.

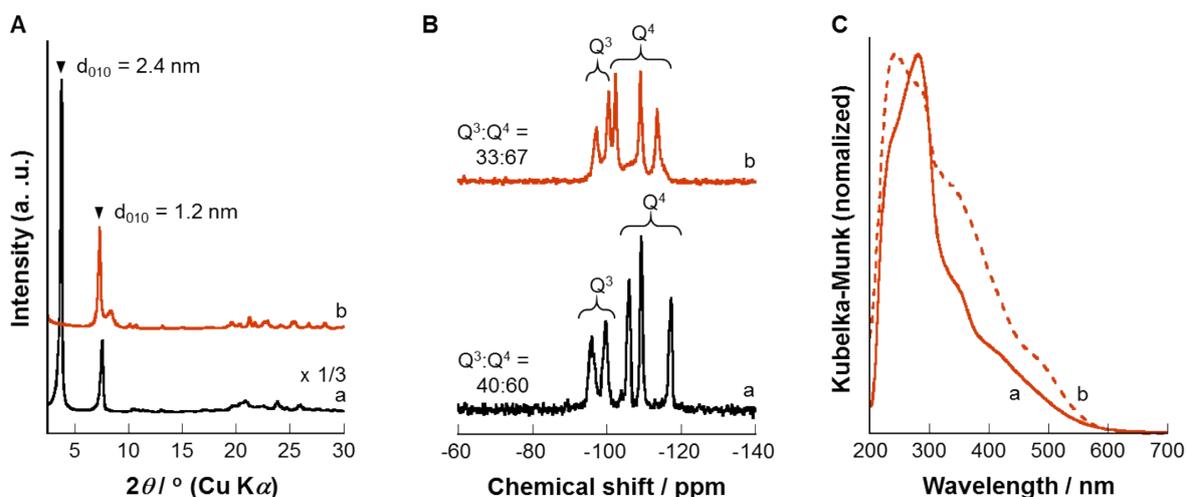


Fig. S1. (A) XRD patterns and (B) ^{29}Si MAS NMR spectra of (a) the hexadecyltrimethylammonium cation-exchanged form of HUS-2 and (b) $\text{Fe}_{0.7}\text{-HUS}$. (C) UV-vis spectra of (a) $\text{Fe}_{0.7}\text{-HUS}$ and (b) $\text{Fe}_{0.2}\text{Ti}_{0.02}\text{-SBA}$. The change in the basal spacing and decrease in the Q^3/Q^4 ratios indicate the grafting of $\text{Fe}(\text{acac})_3$ onto the interlayer surface of HUS-2. Like $\text{Fe}_{0.2}\text{Ti}_{0.02}\text{-SBA}$, $\text{Fe}_{0.7}\text{-HUS}$ shows absorption bands assignable to isolated tetrahedrally coordinated Fe (< 300 nm), Fe oxide clusters (300~400 nm) and Fe oxide nanoparticles (> 400 nm).

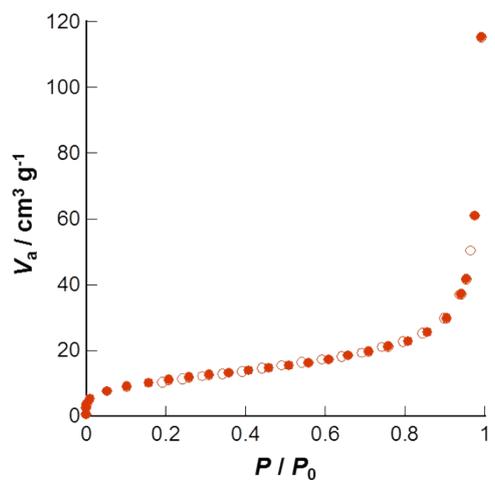


Fig. S2. N₂ adsorption/desorption isotherms of Fe_{0.7}-HUS.