Supporting Information

Amorphous High Surface Area Aluminum Hydroxide-Bicarbonates for Highly-Efficient Methyl Orange Removal from Water

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Table S1. Results of elemental analysis (wt%) of aluminum hydroxide-bicarbonates

Compound	pH of synthetic solution *m*	Total BET surface area *n* [m^2 g^{-1}]	Al	C	H
I-NH_3	4.8	38	31.2	1.08	3.14
I-NH_3	7.8	165	31.3	1.01	3.04
I-NH_3	9.6	6.7	31.8	1.22	3.33
$[Al_{13}O_4(\mu\text{-}OH)_{24}(H_2O)_6(OH)_6](HCO_3)$			32.1	1.10	3.96

Amounts of N in **I-NH_3** were negligible (< 0.2wt%). See ref. 11 for the elemental analysis of **I-NaOH** prepared under different pH.

Table S2. MO adsorption on various adsorbents at r.t. (or 303 K)

Compound	[a]*K* [g mg^{-1} min]	[b]q_m [mg g^{-1}]	Ref
I-NH_3	0.037	154	This work
activated carbon	$1.6\text{-}9.6 \times 10^{-5}$	217	S1
chitosan/Al_2O_3/ magnetite	0.002-0.02	417	S2
MOF-235	$7.7\text{-}9.1 \times 10^{-5}$	477	S3
MIL-101	9.01×10^{-4}	114	S4
banana peel	-	21	S5
orange peel	-	21	S5

[a]Equilibrium rate constant calculated by the pseudo-second order model. [b]Maximum adsorption capacity calculated by the Langmuir model. [S1] S. Chen, J. Zhang, C. Zhang, Q. Yue, Y. Li, C. Li, Desalination 252 (2010) 149–156. [S2] B. Tanhaei, A. Ayati, M. Lahtinen, M. Sillanpää, Chem. Eng. J. 259 (2015) 1-10. [S3] E. Haque, J. W. Jun, S. H. Jhung, J. Hazardous. Mater. 185 (2011) 507-511. [S4] E. Haque, J. E. Lee, I. T. Jang, Y. K. Hwang, J. –S. Chang, J. Jegal, S. H. Jhung, J. Hazardous. Mater. 181 (2010) 535-542. [5] G. Annadurai, R. –S. Juang, D. –J. Lee, J. Hazardous. Mater. B92 (2002) 263-274.

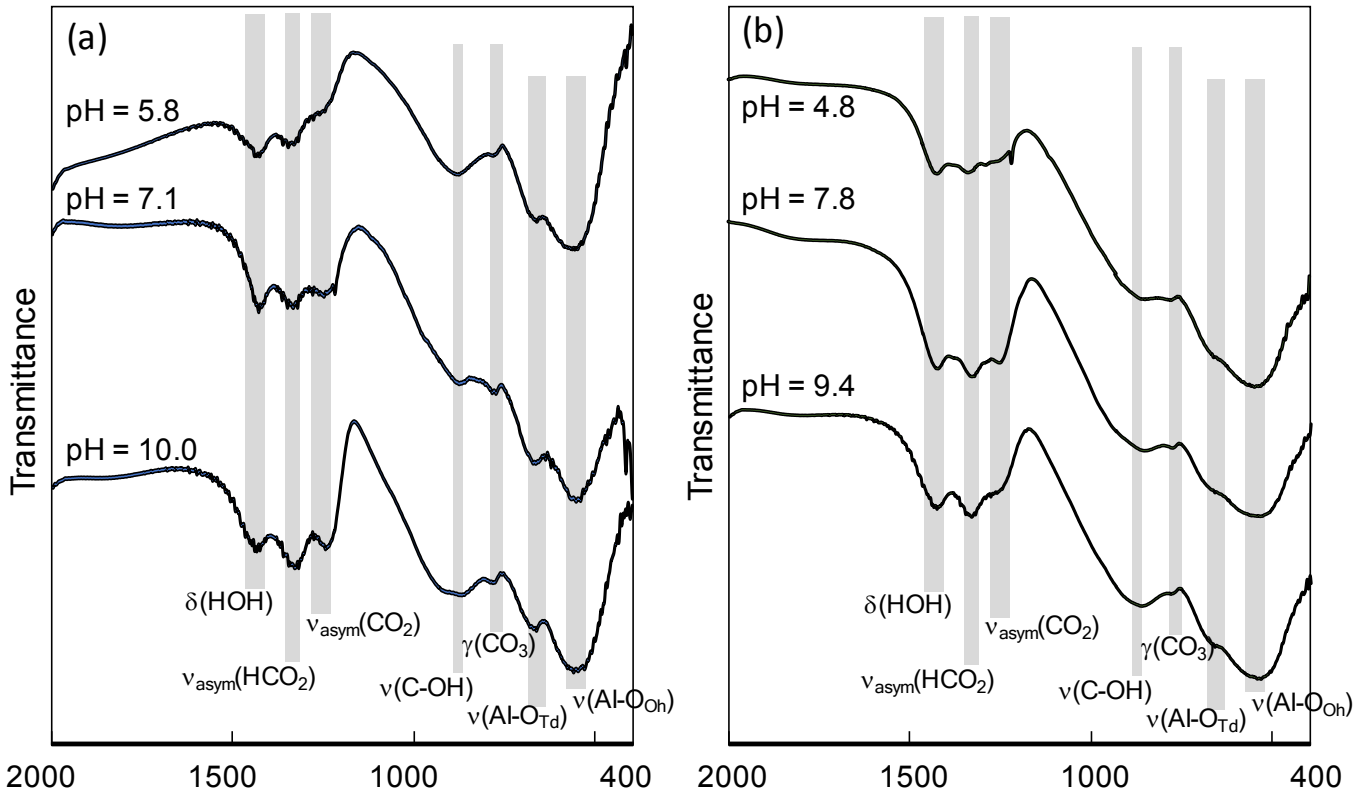

Figure S1. IR spectra of (a) **I-NaOH** and (b) **I-NH_3** synthesized at various pH levels.

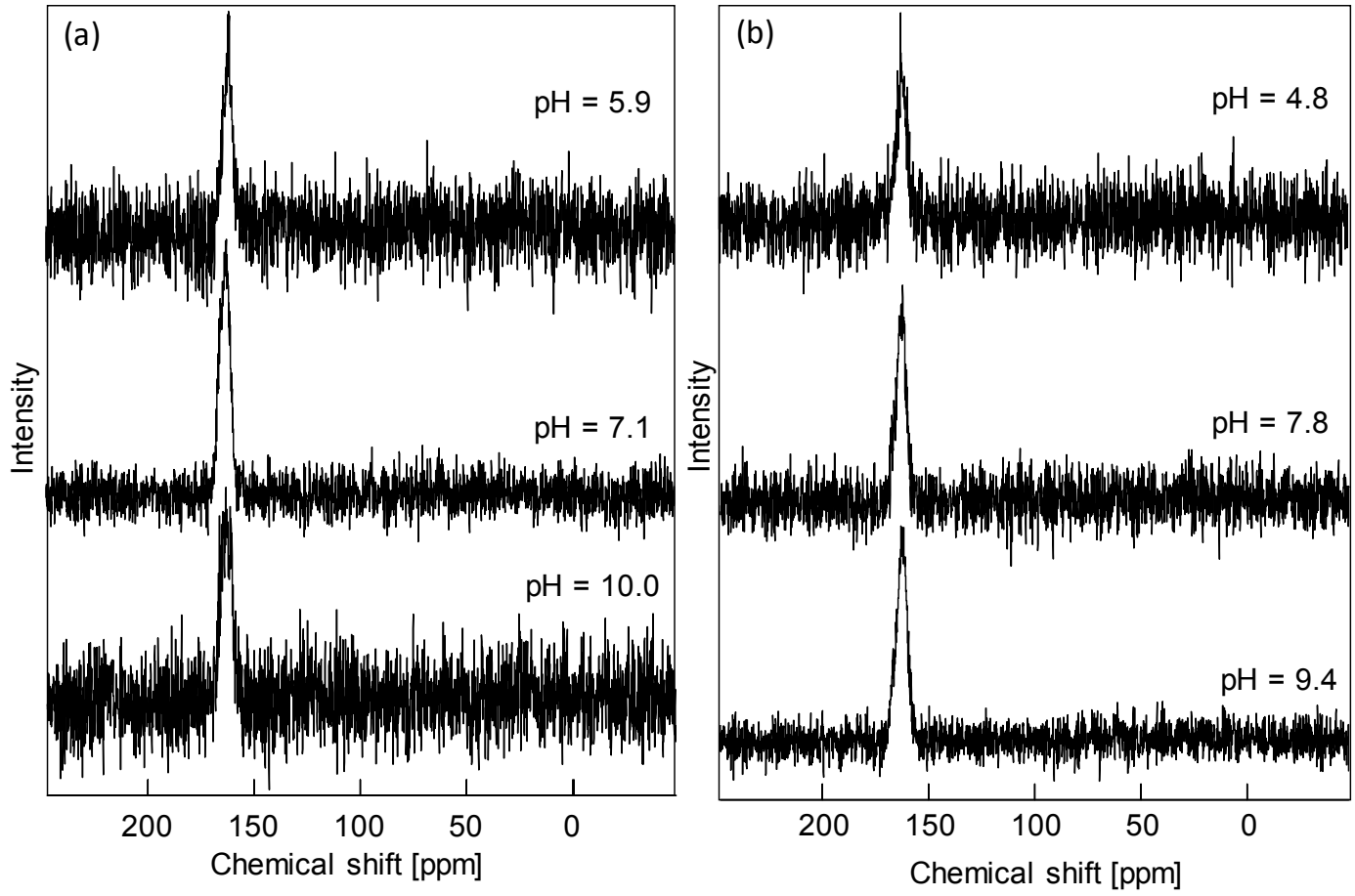

Figure S2. ^{13}C CPMAS NMR spectra of (a) **I-NaOH** and (b) **I-NH_3** synthesized at various pH levels.

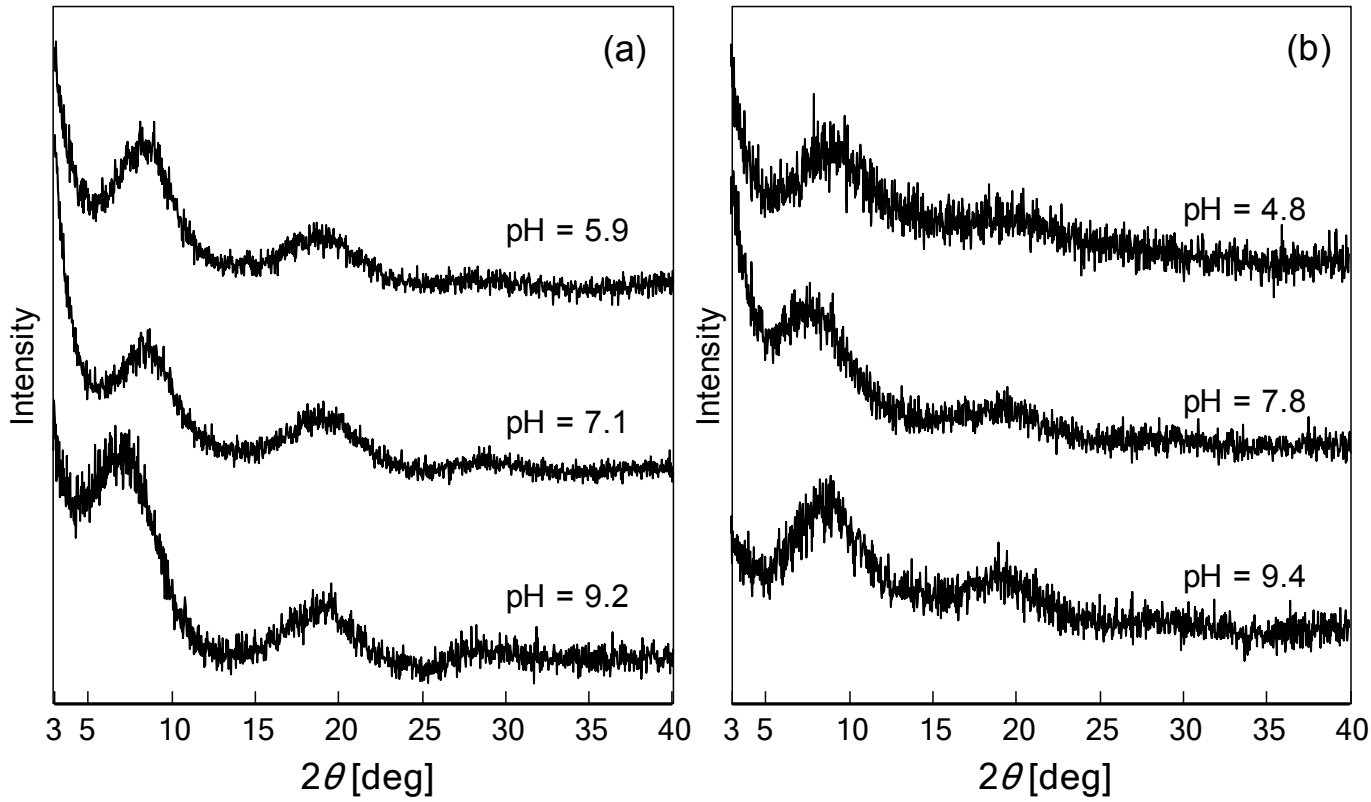

Figure S3. PXRD patterns of (a) **I-NaOH** and (b) **I-NH_3** synthesized at various pH levels.

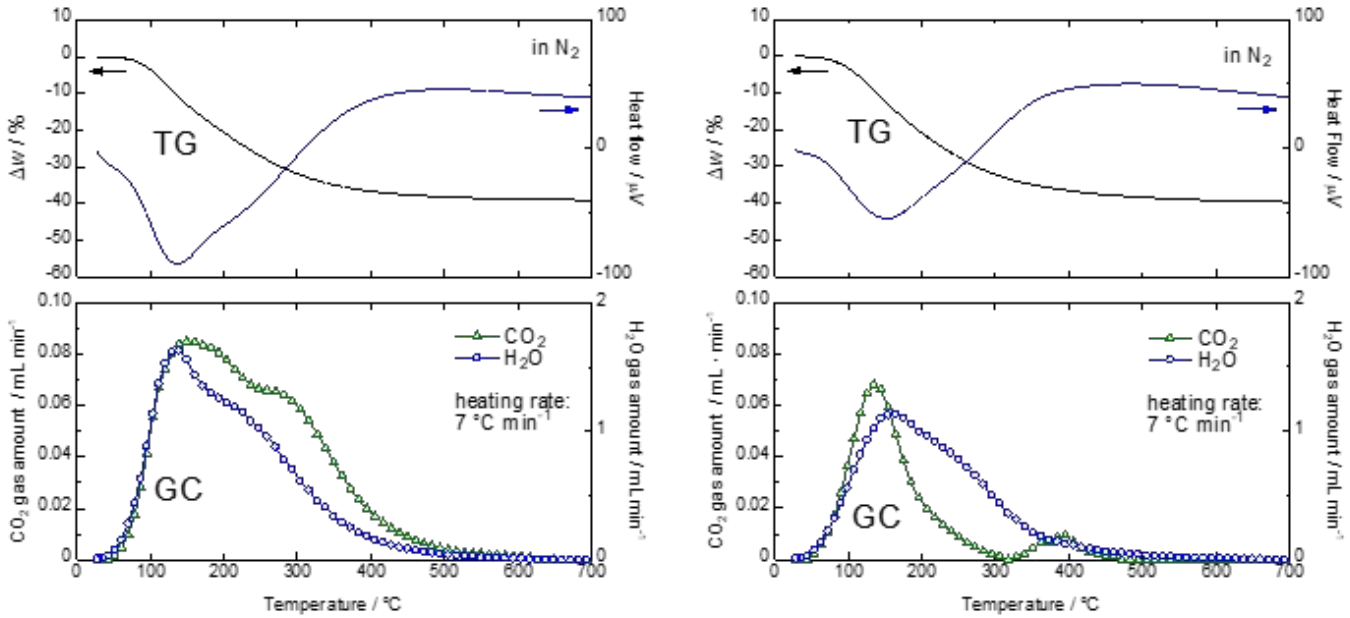

Figure S4. TG-GC data of (left) **I-NaOH-7.8-216** and (right) **I-NH_3-7.8-165**.

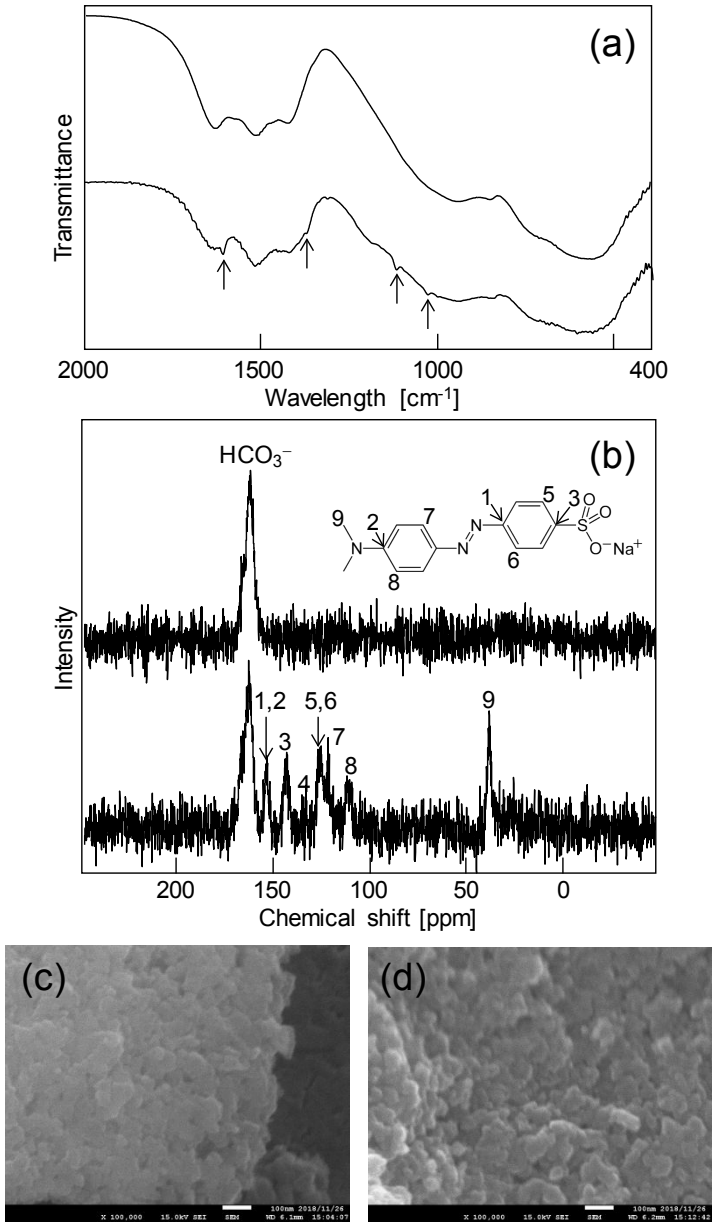

Figure S5. (a) IR and (b) ^{13}C MAS NMR of **I-NH$_3$-8.6-90** (top) before and (bottom) after MO adsorption. Arrows in (a) indicates the signals of MO. SEM images of **I-NH$_3$-8.6-90** (c) before and (d) after MO adsorption. White bars in (c) and (d) indicate 100 nm.

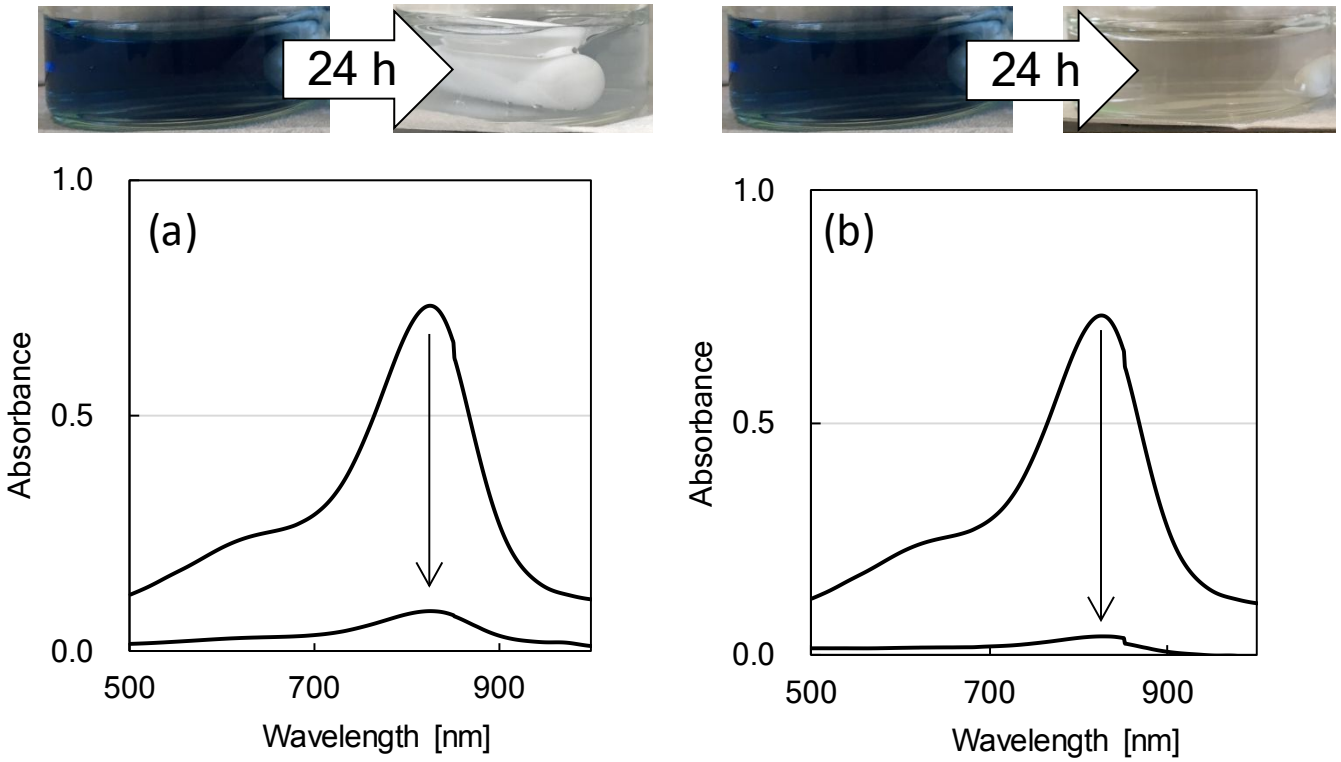

Figure S6. UV-vis spectra and photo images of aqueous solutions of phosphoric acid before (initial phosphate ion concentration 362 mg L^{-1}) and after the addition of 150 mg (1.4×10^{-1} mmol) of (a) **I-NaOH-6.6-61** or (b) **I-NH_3-5.8-67**. The amount of phosphate ions was analyzed by the molybdenum blue method (see the experimental section).

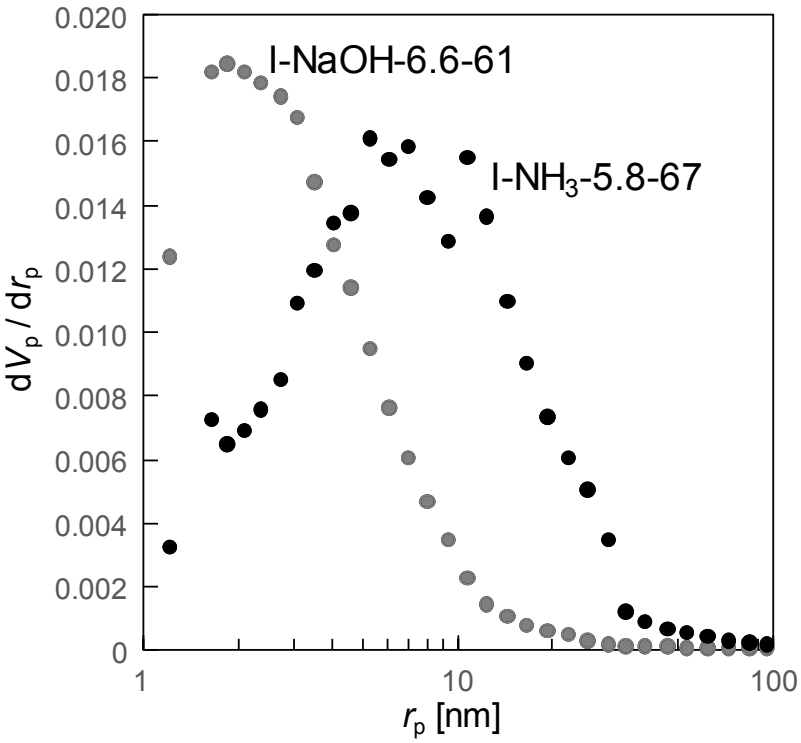

Figure S7. Pore size distributions of **I-NaOH-6.6-61** and **I-NH_3-5.8-67**.

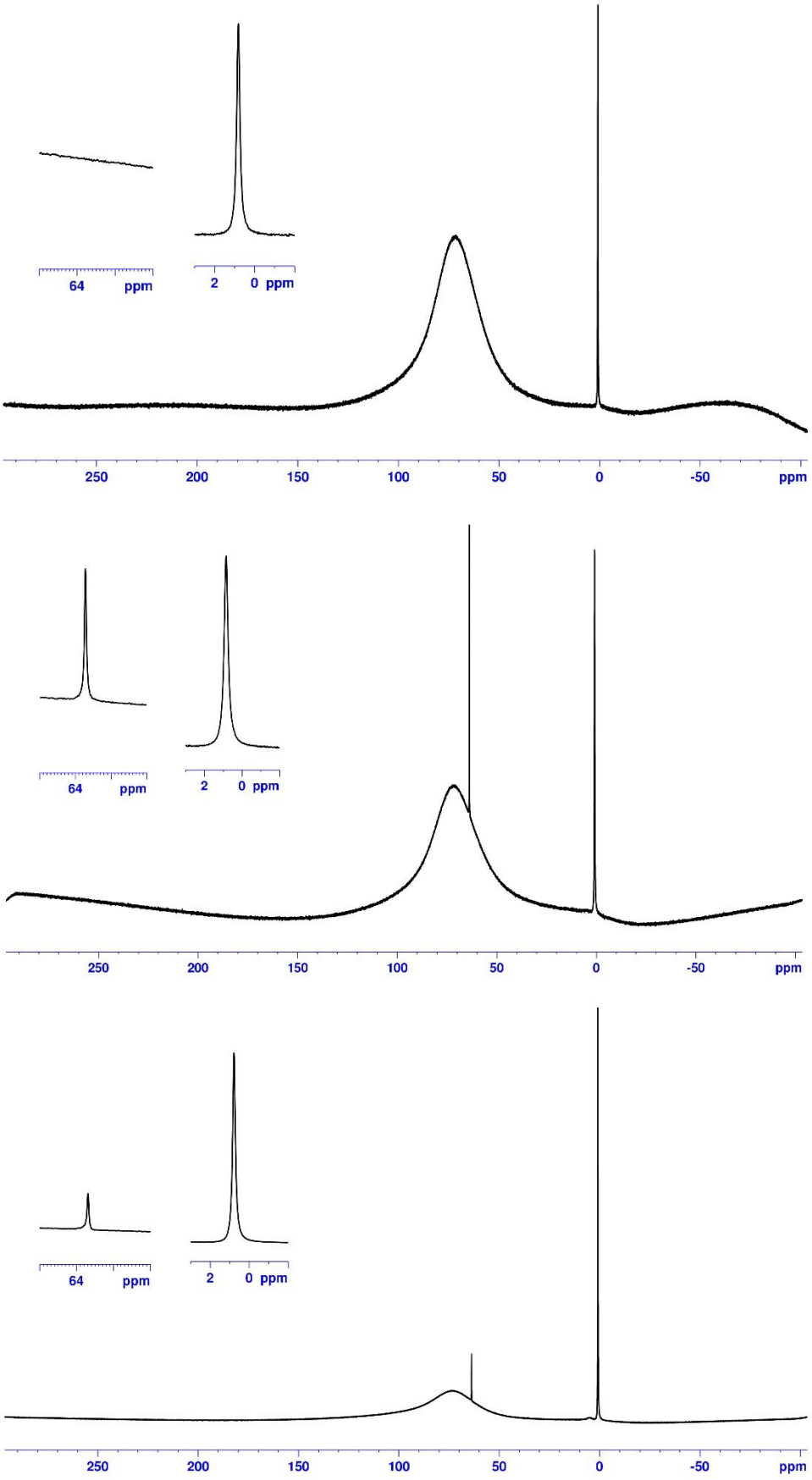

Figure S8. ^{27}Al-NMR spectra of (top) 1 mol L^{-1} $Al(NO_3)_3$aq, **Al_{13}** solution prepared with (middle) NaOH or (bottom) NH_3. Note that $Al(NO_3)_3$aq contains only 6-coordinated aluminum species ($[Al(H_2O)_6]^{3+}$). Broad signals are due to an NMR tube.

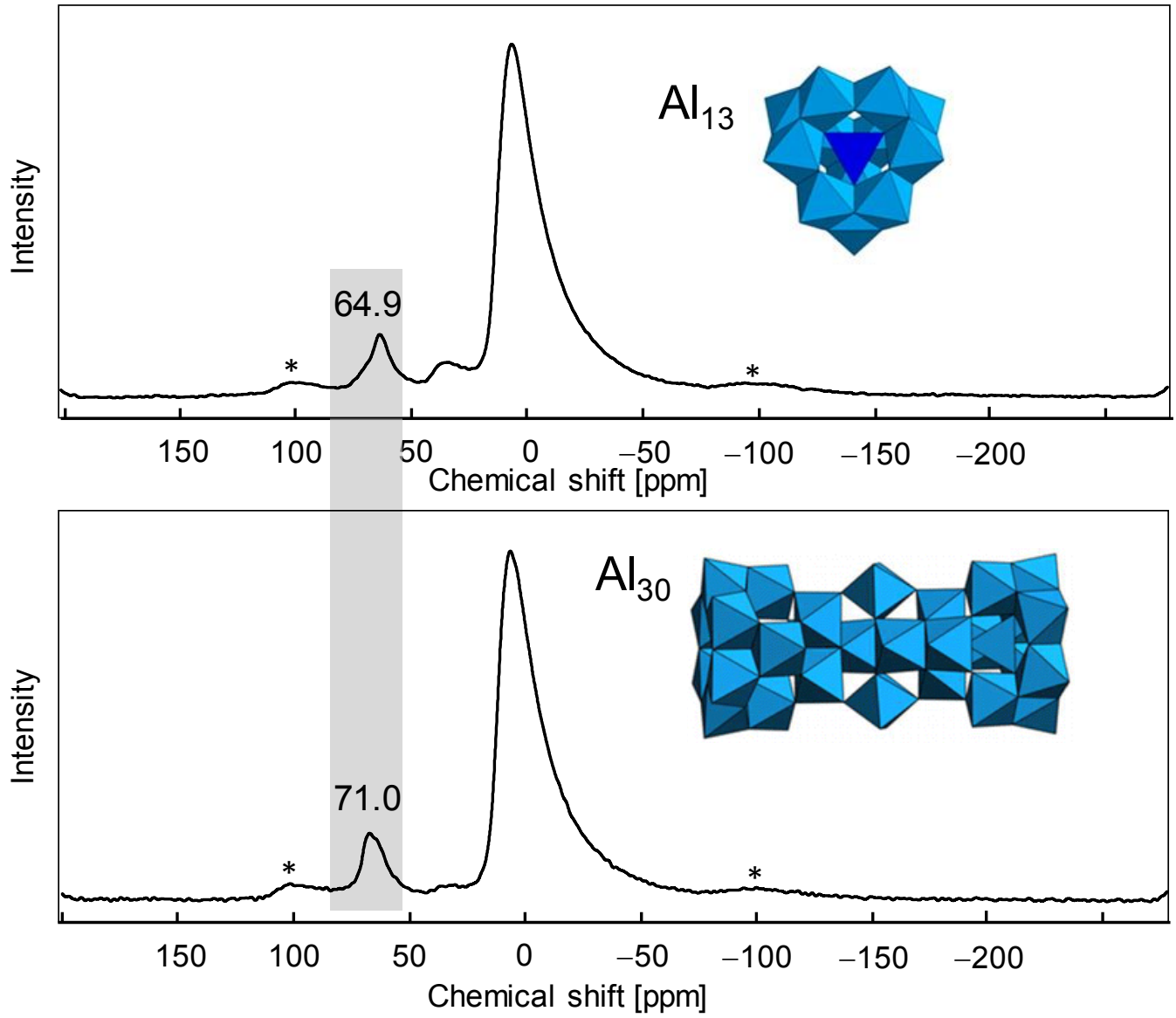

Figure S9. ^{27}Al MAS NMR spectra (^{27}Al = 104.27 MHz, MAS = 10 kHz) of aluminum hydroxide-bicarbonates synthesized with (top) **Al_{13}** or (bottom) **Al_{30}**. Asterisks denote spinning side bands.

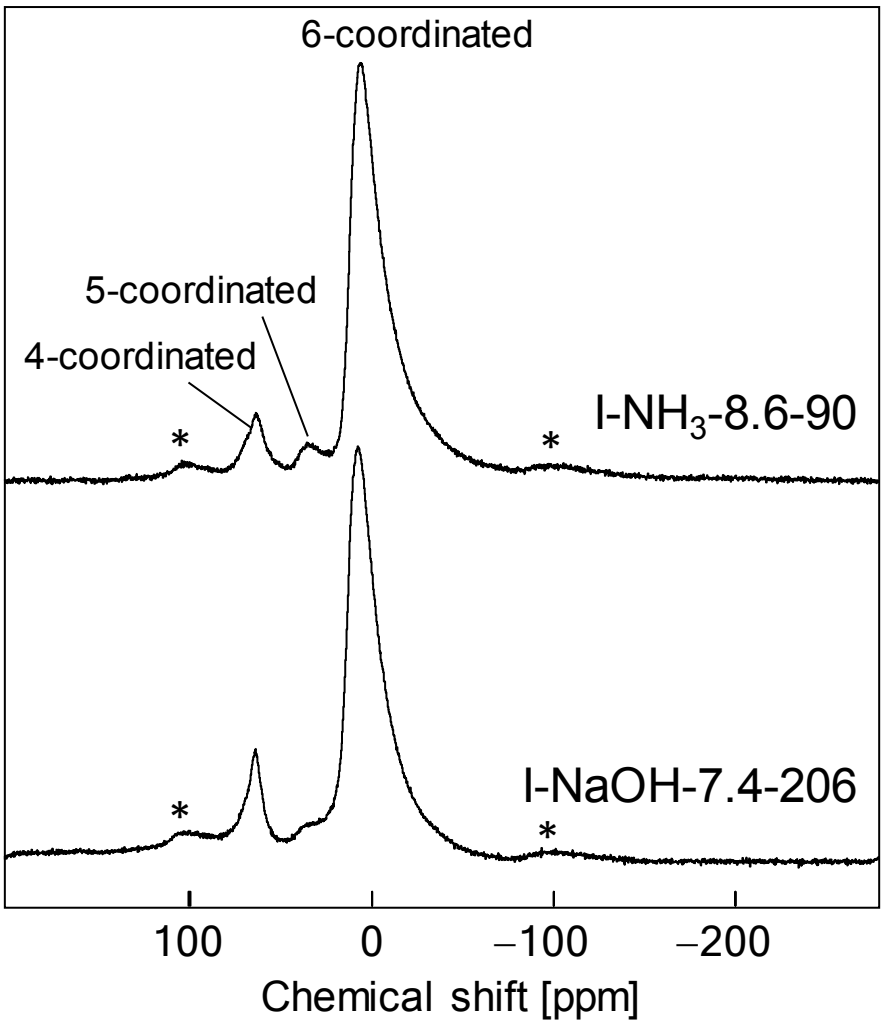

Figure S10. ^{27}Al MAS NMR spectra (^{27}Al = 104.27 MHz, MAS = 10 kHz) of **I-NH$_3$-8.6-90** and **I-NaOH-7.4-206**. Asterisks denote spinning side bands.