Transformation of Gibbsite to Boehmite in Caustic Aqueous Solution at Hydrothermal Conditions

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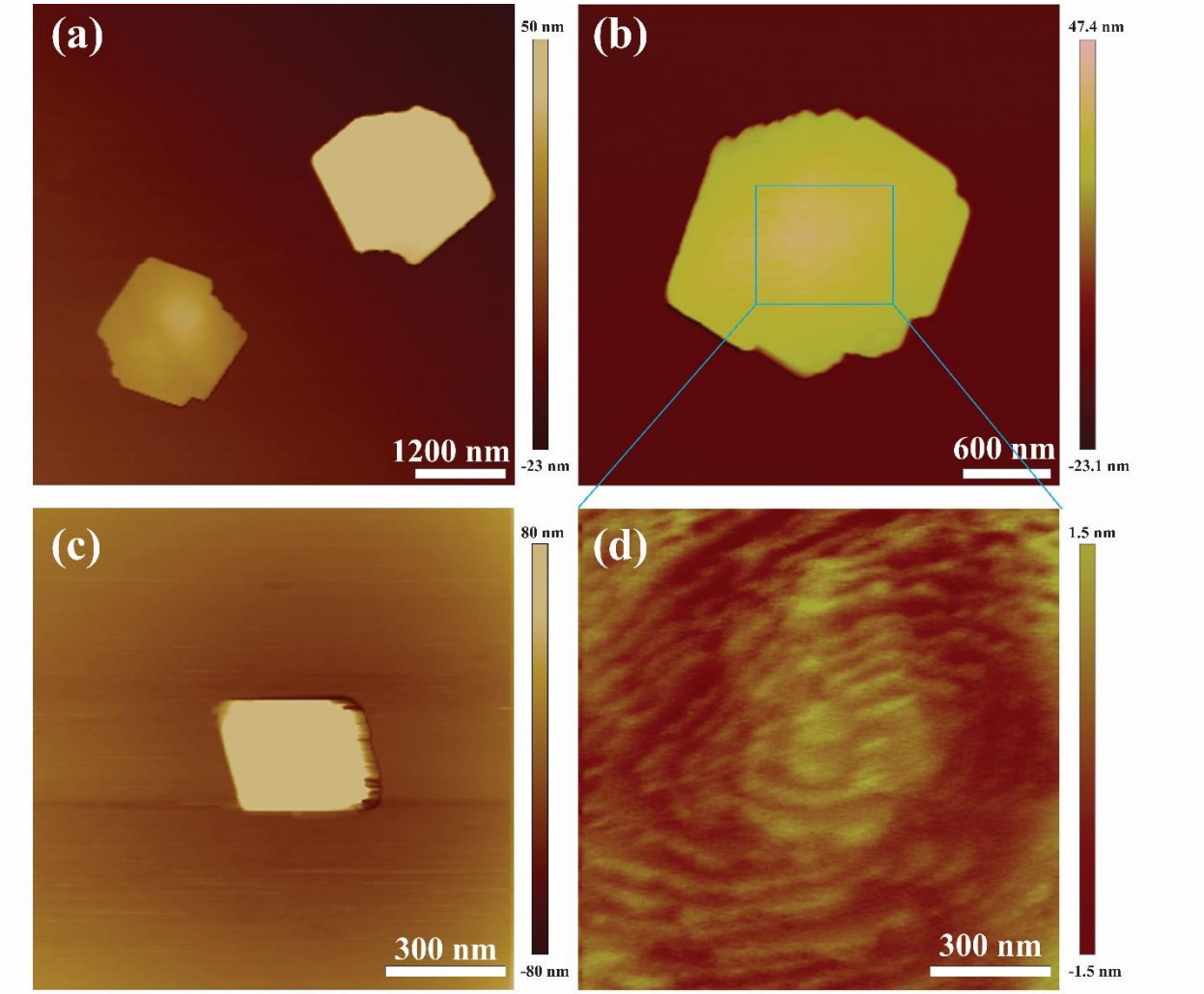

Figure S1. AFM images of boehmite produced at 100 °C, 10 days (a), 120 °C, 2 days (b), and 200 °C, 2 days (c) showing (a, b) hexagonal particle shapes and (c) rhombic particle shapes. (d) AFM image of boehmite produced at 120 °C, 2 days revealing spiral growth hillocks on the basal (010) surface. The concentration of precursor gibbsite and NaOH is 0.256 M and 0.2 M, respectively.

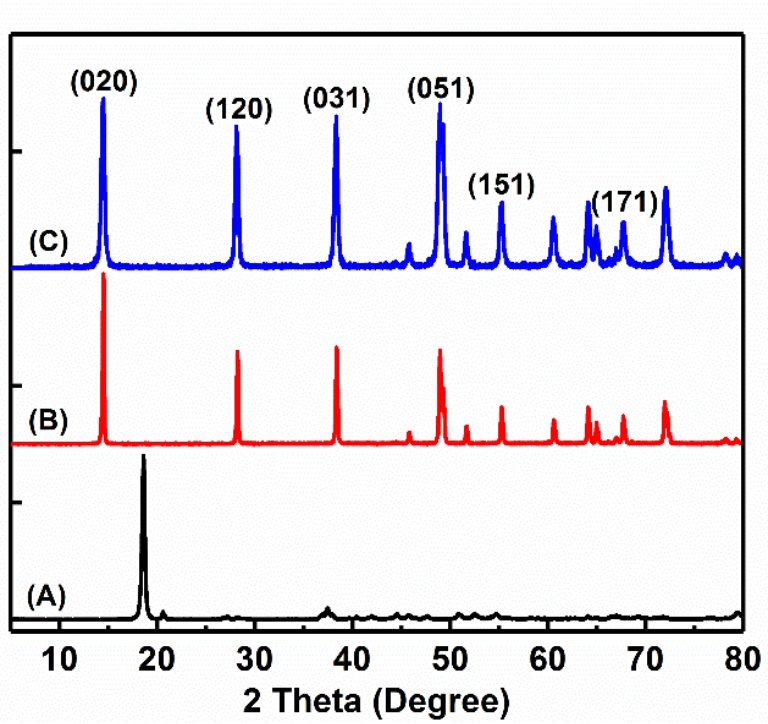

Figure S2. XRD patterns of product boehmite produced at (A) 0.01 M; (B) 0.1 M; and (C) 0.2 M NaOH. The temperature and reaction time for all reactions were 120 ^{o}C and 3 days, respectively. The concentration of the precursor gibbsite was 0.256 M.

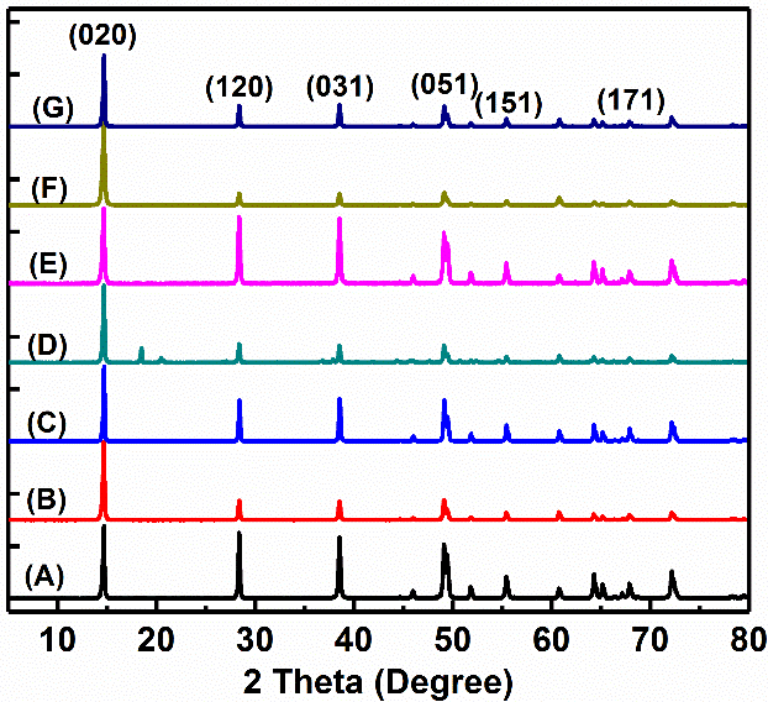

Figure S3. XRD patterns of product boehmite produced at (A) 0.064 M Gibbsite, 0.1 M NaOH, 120 ^{o}C; (B) 0.25 M Gibbsite, 0.1 M NaOH, 100 ^{o}C; (C) 0.25 M Gibbsite, 0.1 M NaOH, 120 ^{o}C; (D) 0.641 M Gibbsite, 0.2 M NaOH, 100 ^{o}C; (E) 0.641 M Gibbsite, 0.2 M NaOH, 120 ^{o}C; (F) 0.641 M Gibbsite, 0.5 M NaOH, 100 ^{o}C; and (G) 0.641 M Gibbsite, 0.5 M NaOH, 120 ^{o}C. The reaction time was 10 days and 7 days for 100 ^{o}C and 120 ^{o}C.

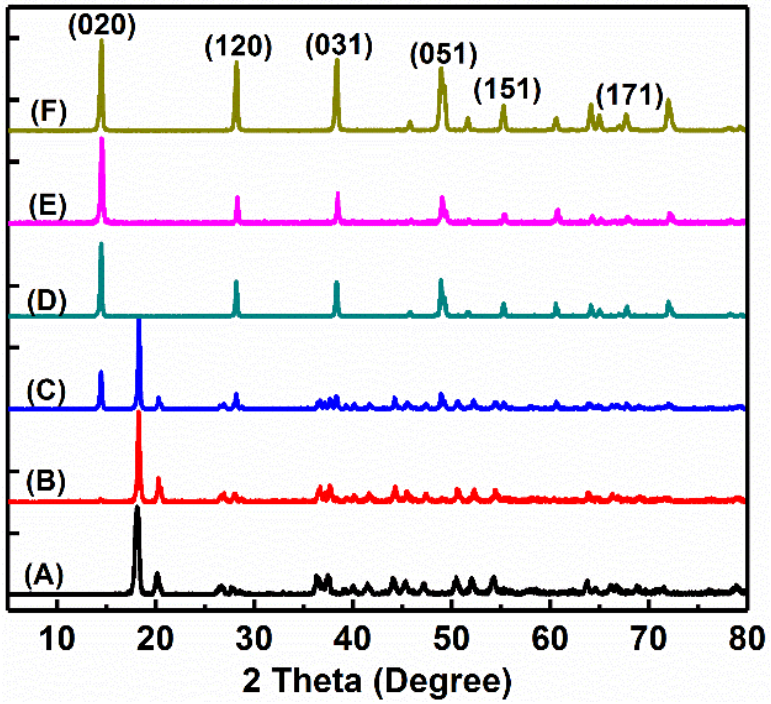

Figure S4. XRD patterns of samples synthesized at different hydrothermal treatment times: (A) 6 h; (B) 24 h; (C) 30 h; (D) 36 h; (E) 42 h; and (F) 48 h. The concentration of gibbsite and NaOH was 0.256 M and 0.2 M, respectively. The temperature was 120 ^{o}C.

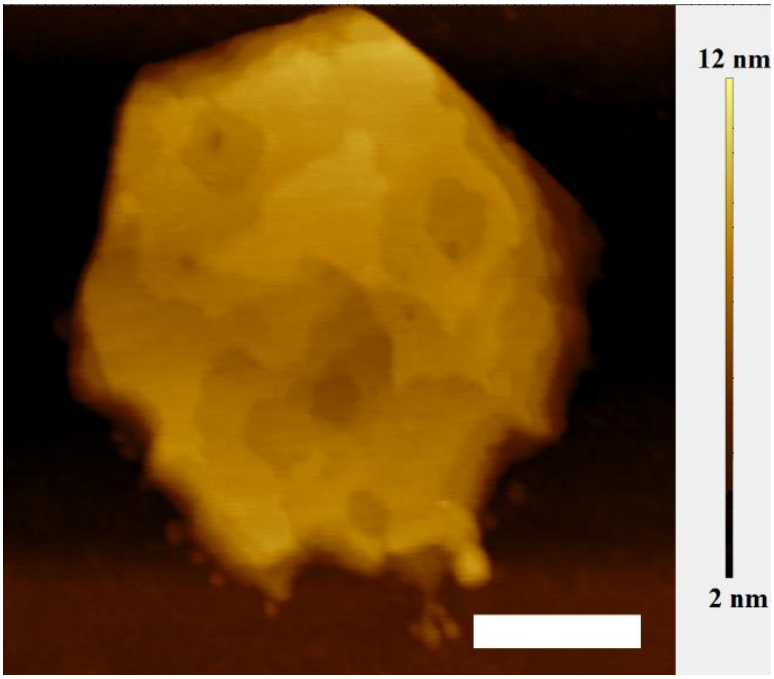

Figure S5. AFM image of sample collected at 6h hydrothermal treatment time. The concentration of gibbsite and NaOH was 0.256 M and 0.2 M, respectively. The temperature was 120 ^{o}C. The insert scale bar is 100 nm.

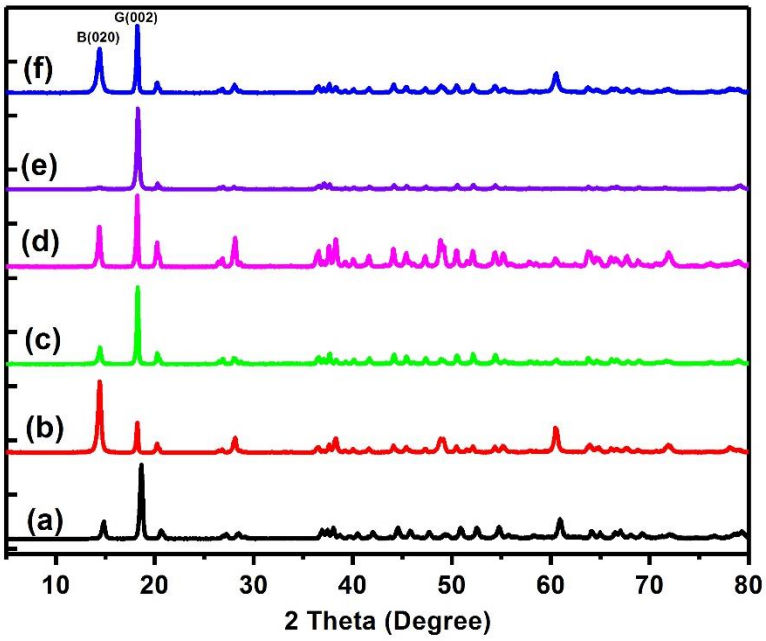

Figure S6. XRD patterns of samples synthesized with different additives: (A) 6 h reaction solution with 0.128 M $NaAlO_2$ solution; (B) 24 h reaction solution with 0.128 M $NaAlO_2$ solution; (C) 6 h reaction solution with 0.128 M NaAc solution; (D) 24 h reaction solution with 0.128 M NaAc solution; (E) 6 h reaction solution with 0.128 M sodium oleate solution; and (F) 24 h reaction solution with 0.128 M sodium oleate solution. The 6 h and 24 h reaction solution was prepared by using the conditions: concentration of gibbsite and NaOH was 0.256 M and 0.2 M, respectively; temperature was 120 ^{o}C, reaction time was 6 h and 24 h, respectively.

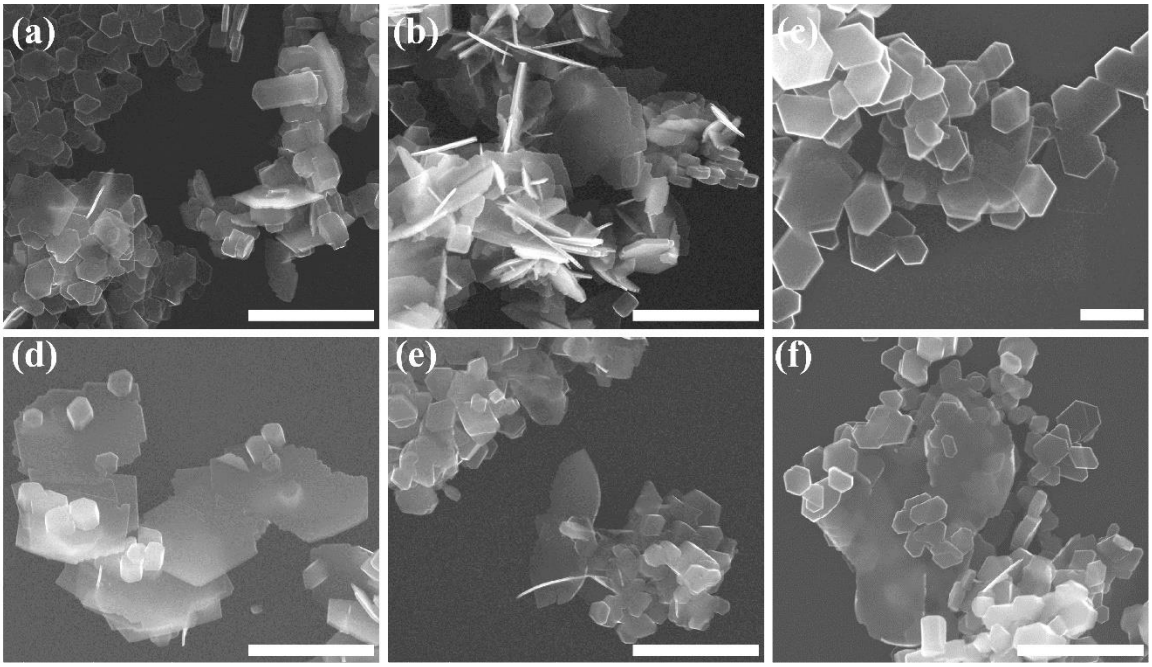

Figure S7. SEM images of samples synthesized with different additives: (A) 6 h reaction solution with 0.128 M $NaAlO_2$ solution; (B) 24 h reaction solution with 0.128 M $NaAlO_2$ solution; (C) 6 h reaction solution with 0.128 M NaAc solution; (D) 24 h reaction solution with 0.128 M NaAc solution; (E) 6 h reaction solution with 0.128 M sodium oleate solution; and (F) 24 h reaction solution with 0.128 M sodium oleate solution. The 6 h and 24 h reaction solution was prepared by using the conditions: concentration of gibbsite and NaOH was 0.256 M and 0.2 M, respectively; temperature was 120 ^{o}C, reaction time was 6 h and 24 h, respectively. The scale bar is 2 μm.

Table S1. Fitting Results of **Figure 10**.

Sample name	Site name	Chemical shift (ppm)	CQ (MHz)	η	Lb (ppm)	Area ratio (%)
Precursor: gibbsite	Octa-site	11.3	2.6	1.0	2.7	99.51
	Penta-site	41.7	6.7	0.6	6.4	0.40
	Tetra-site	-	-	-	-	-
		74.8	4.8	0.5	8.0	0.09
6 h sample: 100% gibbsite	Octa-site	11.2	2.6	1.0	2.7	98.42
	Penta-site	42.8	7.0	0.3	9.7	0.98
	Tetra-site	61.9	4.4	1.0	10.5	0.11
		73.3	4.9	0.8	12.0	0.50
24 h sample: 97% gibbsite and 3% boehmite	Octa-site	11.2	2.6	1.0	2.7	98.66
	Penta-site	42.7	6.8	0.6	8.0	0.94
	Tetra-site	62.0	4.4	1.0	10.3	0.06
		74.1	4.8	0.5	10.3	0.33
30 h sample, 70% gibbsite and 30% boehmite	Octa-site	11.1	2.6	1.0	2.7	99.3
	Penta-site	41.7	6.7	0.6	6.4	0.36
	Tetra-site	62.0	4.3	1.0	9.3	0.05
		73.2	4.8	0.5	10.3	0.25
36 h sample: 100% boehmite	Octa-site	11.1	2.6	1.0	2.7	99.61
	Penta-site	42.2	6.7	0.5	6.4	0.19
	Tetra-site	62.0	4.4	1.0	10.7	0.03
		73.0	4.9	0.5	11.1	0.17
42 h sample: 100% boehmite	Octa-site	10.7	2.6	1.0	3.9	99.61
	Penta-site	41.5	6.7	0.5	5.4	0.18
	Tetra-site	62.0	4.3	1.0	9.5	0.03
		72.9	4.8	0.5	10.0	0.17

Table S2. Overview of boehmite synthesis conditions and resulting products with secondary Al(III) precursor and/or organic additives.

Entry	$[Al^{3+}]$ (M, based on gibbsite)	[NaOH] (M)	Temp. (^oC)	Time (h)	Additive	Temp. (^oC)	Time (h)	Product	
								Boehmite	Gibbsite
1	0.256	0.2	120	6	$NaAlO_2$ (aq) 0.128M	120	48	19.4%	80.6%
2	0.256	0.2	120	24	$NaAlO_2$ (aq) 0.128M	120	48	81.3%	18.7%
3	0.256	0.2	120	6	NaAc (aq) 0.128M	120	48	24.5%	75.5%
4	0.256	0.01	120	24	NaAc (aq) 0.128M	120	48	44.2%	55.8%
5	0.256	0.1	120	6	Sodium oleate (aq) 0.128M	120	48	1.3%	98.7%
6	0.256	0.2	120	24	Sodium oleate (aq) 0.128M	120	48	56.3%	43.7%