Supporting Information

Boehmite and Gibbsite Nanoplates for the Synthesis of Advanced Alumina Products

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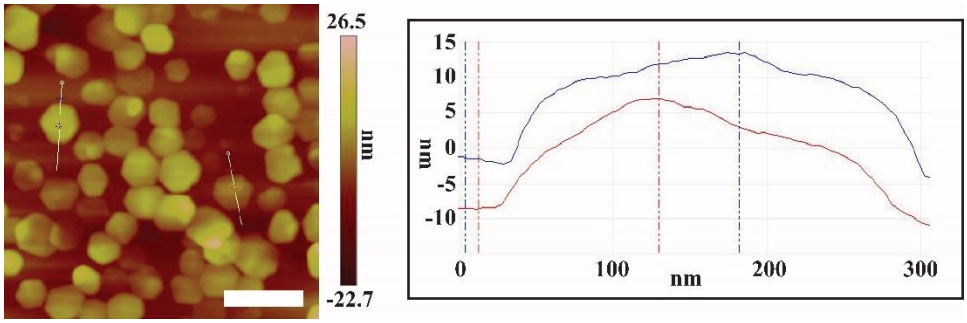

Figure S1. Topographic AFM image and corresponding cross-section for the gibbsite nanoplates. The insert scale bar is 500 nm.

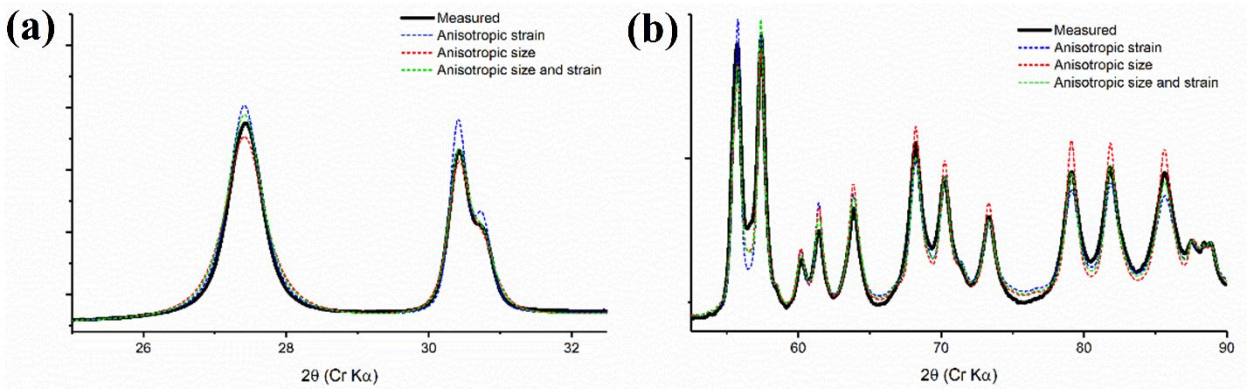

Figure S2. Gibbsite XRD pattern compared with simulated patterns incorporating anisotropic crystallite size and strain broadening.

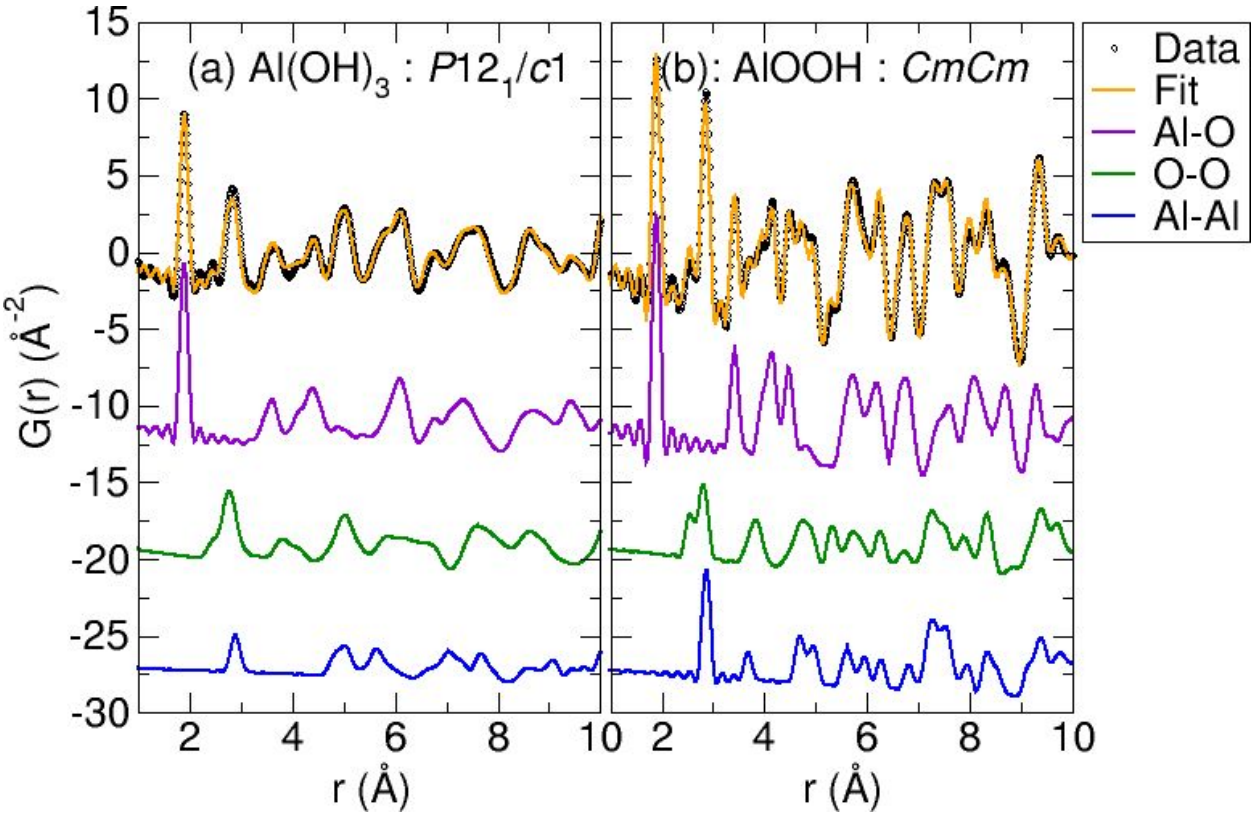

Figure S3. First 10 Å of 100 Å X-ray PDF fits for (a) gibbsite, $Al(OH)_3$ and (b) boehmite, AlOOH nanoplate data. Data are shown as black circles and fits are shown as orange lines. The calculated Al-O, O-O, and Al-Al partial PDFs composing the respective models are shown below the data sets and fits, as purple, green and blue lines, respectively.

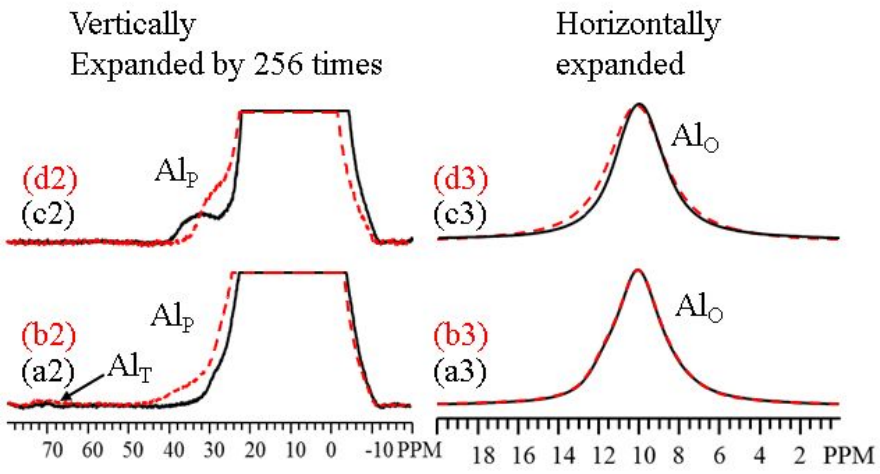

Figure S4. Comparison of the ^{27}Al MAS NMR spectra, a duplicate of the results in Figure 1 but plotted with the spectra of the dehydrated samples (dotted red traces) superimposed on the corresponding as-synthesized samples (solid black traces) to highlight the effect of the dehydration. Only the expanded regions are shown. Labels are the same as those in Figure 1 of the text.

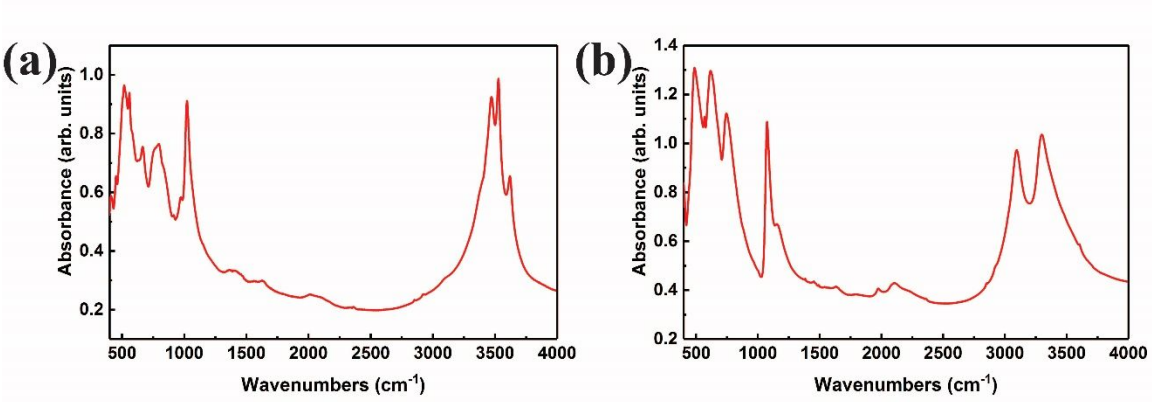

Figure S5. FTIR spectrum of (a) gibbsite and (b) boehmite.

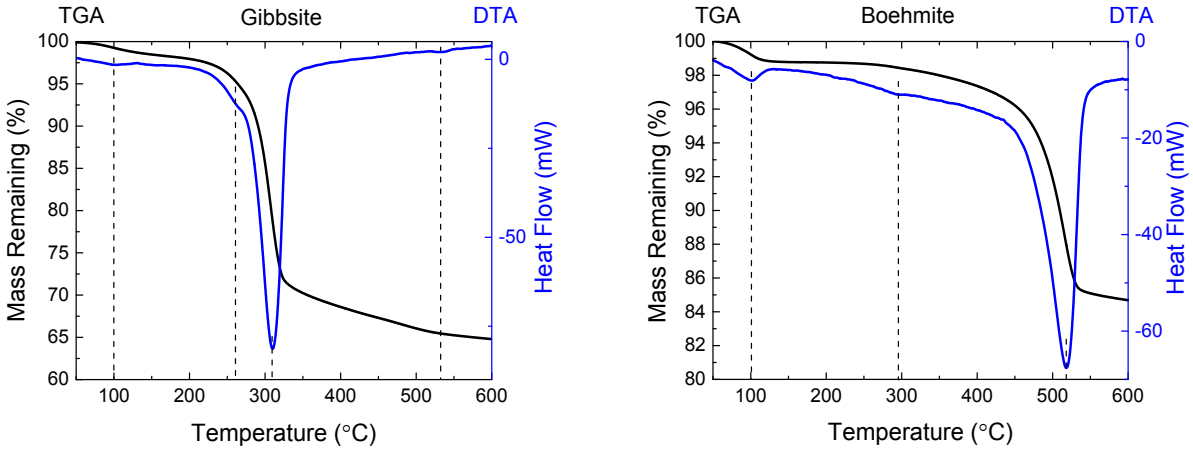

Figure S6. Mass loss (black line) and heat flow (blue line) curves for *left:* gibbsite and *right:* boehmite. Endotherm minima are accentuated with dashed lines.

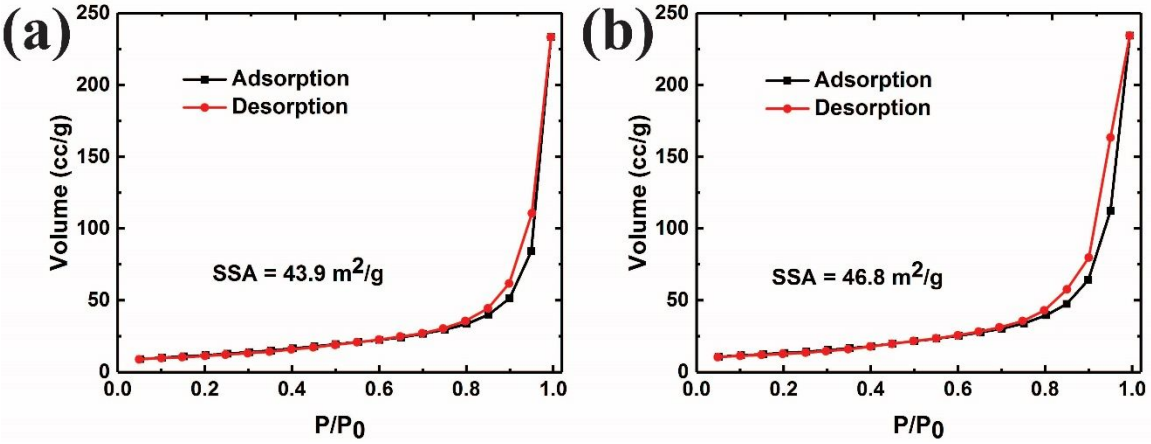

Figure S7. Nitrogen isotherms for (a) gibbsite and (b) boehmite.

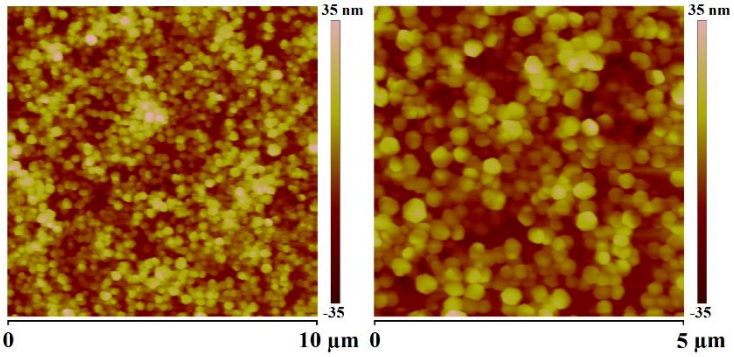

Figure S8. AFM image of gibbsite nanoplates on Si substrates to show these nanoplates aggregated along the [001] direction.

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Table S1 Crystal structure of gibbsite nanoplates, $Al(OH)_3$, as determined from refinement of X-ray PDF data between 1 and 100 Å. Refined values are given with estimated standard deviation from refinement in parentheses. Parameters with a '*' were constrained to be equivalent.

$Al(OH)_3$	S. G. $P12_1/c1$	a =8.6645(7) Å, b = 5.0594(4) Å, c = 12.5281(19) Å, β=129.443(7)°				
$l1$ = 30(4) nm, $l2$ = 6(1) nm, delta1 = 1.457(11) Å^{-1}, R_{wp} = 18.62%						
Atom	**Wyck.**	***x***	***y***	***z***	**Occ.**	**B_{eq} (Å²)**
Al1	4e	0.1718(5)	0.0200(7)	0.0050(3)	1	0.49(2)
Al2	4e	0.3320(4)	0.5231(5)	-0.0001(2)	1	0.33(1)
O1	4e	0.0779(6)	0.1471(9)	0.3991(4)	1	0.63(1)*
O2	4e	0.0893(7)	0.1298(9)	0.1081(5)	1	0.63(1)*
O3	4e	0.2867(5)	0.7049(8)	0.1087(5)	1	0.63(1)*
O4	4e	0.3943(7)	0.1308(9)	0.3958(4)	1	0.63(1)*
O5	4e	0.4094(5)	0.2197(7)	0.1118(4)	1	0.63(1)*
O6	4e	0.7648(6)	0.1577(8)	0.1004(5)	1	0.63(1)*

Table S2 Crystal structure of boehmite nanoplates, AlOOH, as determined from refinement of X-ray PDF data between 1 and 100 Å. Refined values are given with estimated standard deviation from refinement in parentheses.

AlOOH	S. G. *CmCm*	a = 2.86248(9) Å, b = 12.19646(38) Å, c = 3.68675(10) Å				
$l1$ = 50(2) nm, $l2$ = 20(3) nm, delta1 = 1.604(8) Å^{-1}, R_{wp} = 14.18 %						
Atom	**Wyck.**	***x***	***y***	***z***	**Occ.**	**B_{eq} (Å²)**
Al	4c	0	0.68119(3)	¼	1	0.261(26)
O1	4c	0	0.29128(4)	¼	1	0.354(62)
O2	4c	0	0.08242 (4)	¼	1	0.492(21)

Table S3. Calculated Al−O and Al−Al distances (Å) from AIMD simulations of boehmite and gibbsite at 298.15 K. The thermal disorder parameters are presented as the uncertainty on the last digit, shown in parentheses.

Phase	Site	Al−O	Al−O	Al−O	Al−O	Al−O	Al−O	Al−Al	Al−Al	Al−Al
Boehmite	1	1.902 (5)	1.911 (6)	1.919 (2)×2	1.986 (4)×2			2.914 (2)×2	2.923 (20)×2	2.922 (14)×2
Gibbsite	1	1.931 (3)	1.945 (3)	1.945 (3)	1.918 (3)	1.839 (3)	1.936 (2)	2.934 (3)	2.955 (3)	2.886 (3)
Gibbsite	2	1.950 (3)	1.940 (3)	1.890 (2)	1.904 (3)	1.880 (2)	1.972 (3)	2.955 (3)	2.885 (3)	2.869 (3)

Reference:

1. Peskleway, C. D.; Henderson, G. S.; Wicks, F. J. Dissolution of gibbsite: Direct observations using fluid cell atomic force microscopy. *Am. Mineral.* **2003,** *88*, 18-26.

2. Prange, M. P.; Zhang, X.; Bowden, M. E.; Shen, Z.; Ilton, E. S.; Kerisit, S. N. Predicting Surface Energies and Particle Morphologies of Boehmite (γ-AlOOH) from Density Functional Theory. *J. Phys. Chem. C* **2018,** *122*, 10400-10412.