Supplementary Material

The two aluminum sites in the ^{27}Al MAS NMR spectrum of kaolinite :
Accurate determination of isotropic chemical shifts and quadrupolar
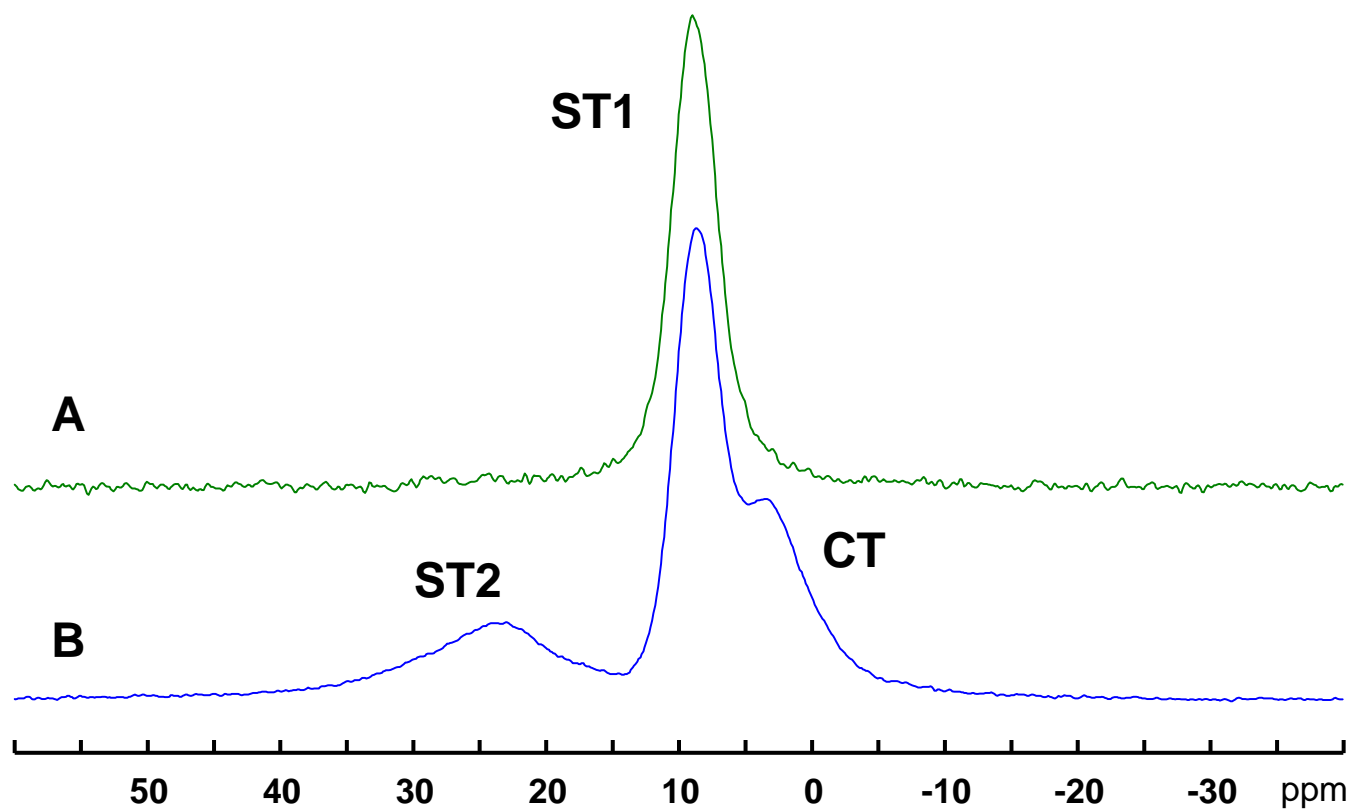interaction parameters

Michaël Paris

Figure SM1. ^{27}Al Rotor-Synchronized acquisition of kaolinite KGa-1b with (A) and without Double-Quantum filtering (B). The former exhibits the ST1 satellite transition only.
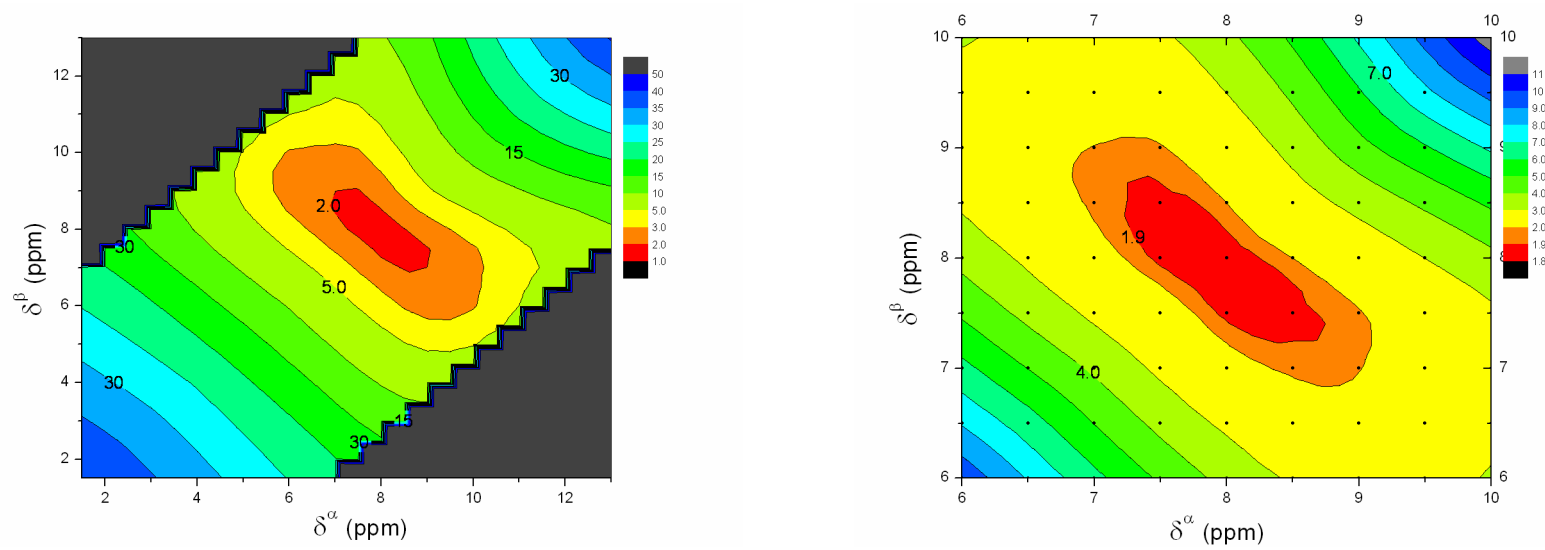
Figure SM2. Contour map of the lowest r.m.s.d. obtained from the fit of the ^{27}Al MAS spectrum as a function of δ^{α} and δ^{β} (left). This is the color version of Figure 3. Enlargement of the 6-10 ppm region (right).

Figure SM3. Lowest r.m.s.d. contour maps of the $(C_Q^\alpha, \eta_Q^\alpha)$, $(C_Q^\beta, \eta_Q^\beta)$, (C_Q^α, C_Q^β) and $(\eta_Q^\alpha, \eta_Q^\beta)$ projections of the $(C_Q^\alpha, \eta_Q^\alpha, C_Q^\beta, \eta_Q^\beta)$ space for δ^α=8.0 ppm and δ^β=7.5 ppm. These are the color versions of Figures 4 and 5.
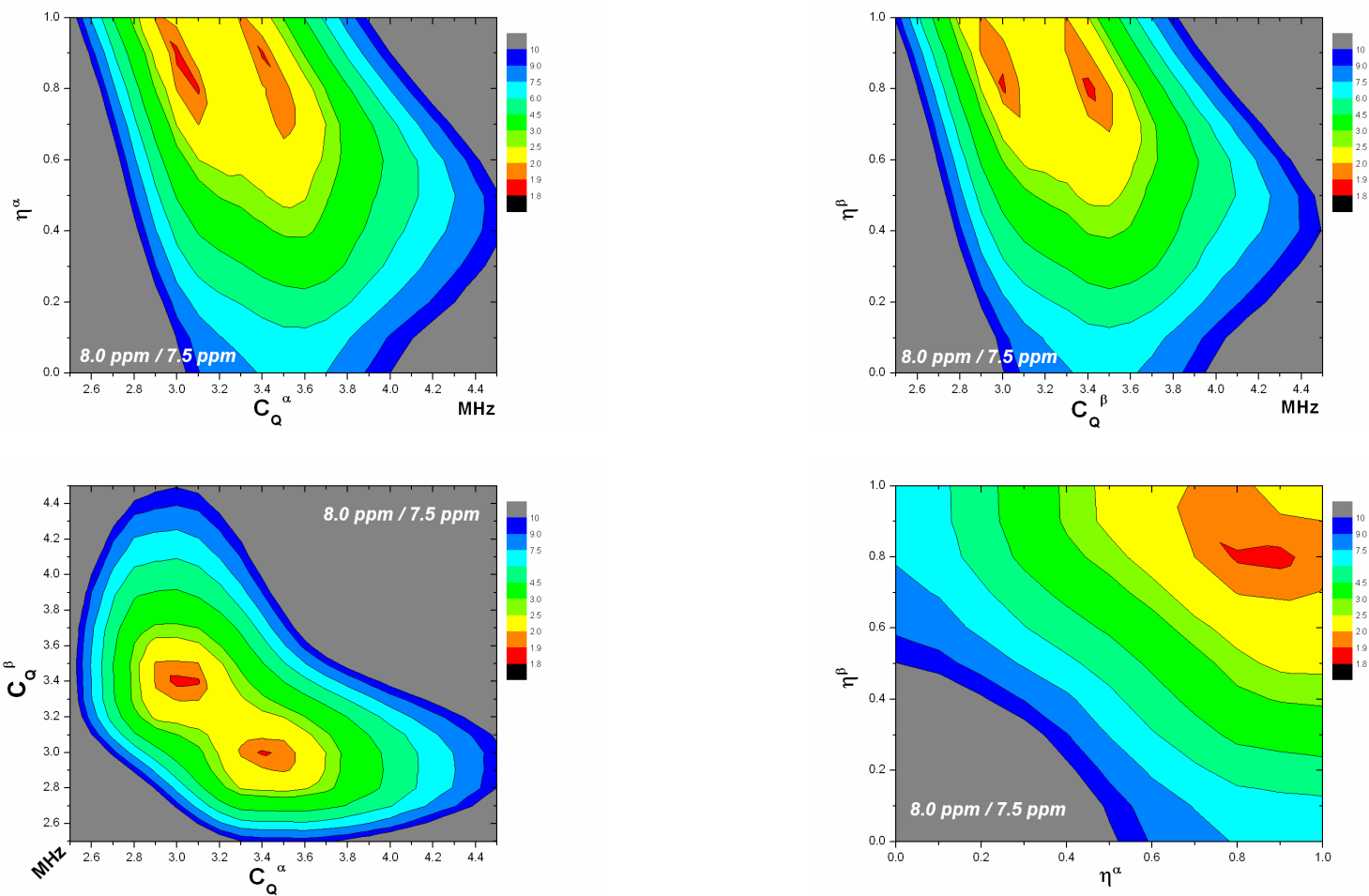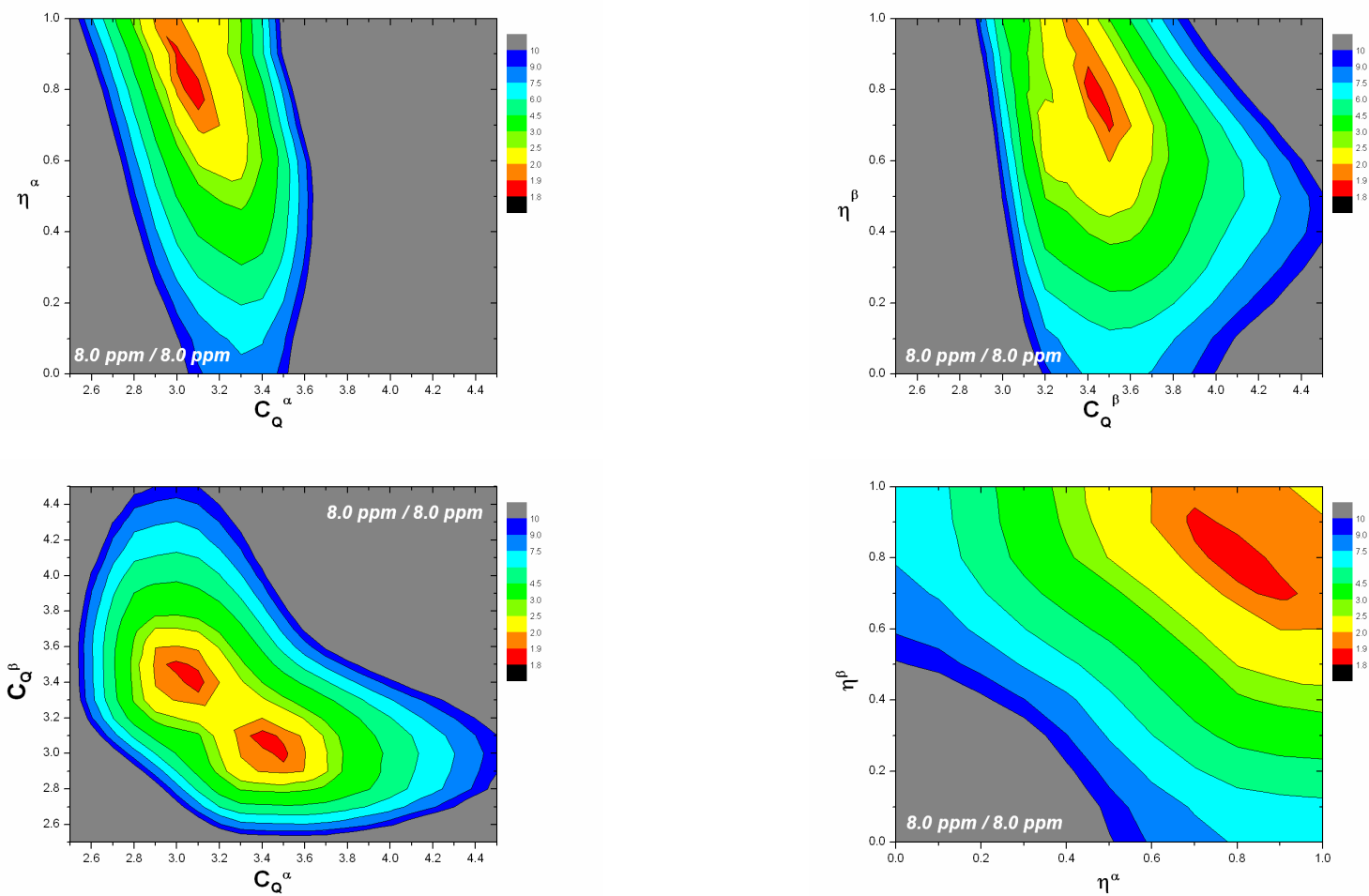
Figure SM4. Lowest r.m.s.d. contour maps of the $(C_Q^\alpha, \eta_Q^\alpha)$, $(C_Q^\beta, \eta_Q^\beta)$, (C_Q^α, C_Q^β) and $(\eta_Q^\alpha, \eta_Q^\beta)$ projections of the $(C_Q^\alpha, \eta_Q^\alpha, C_Q^\beta, \eta_Q^\beta)$ space for δ^α=8.0 ppm and δ^β=8.0 ppm.

Figure SM5. Lowest r.m.s.d. contour maps of the $(C_Q^{\alpha}, \eta_Q^{\alpha})$, $(C_Q^{\beta}, \eta_Q^{\beta})$, $(C_Q^{\alpha}, C_Q^{\beta})$ and $(\eta_Q^{\alpha}, \eta_Q^{\beta})$ projections of the $(C_Q^{\alpha}, \eta_Q^{\alpha}, C_Q^{\beta}, \eta_Q^{\beta})$ space for δ^{α}=7.5 ppm and δ^{β}=8.5 ppm.
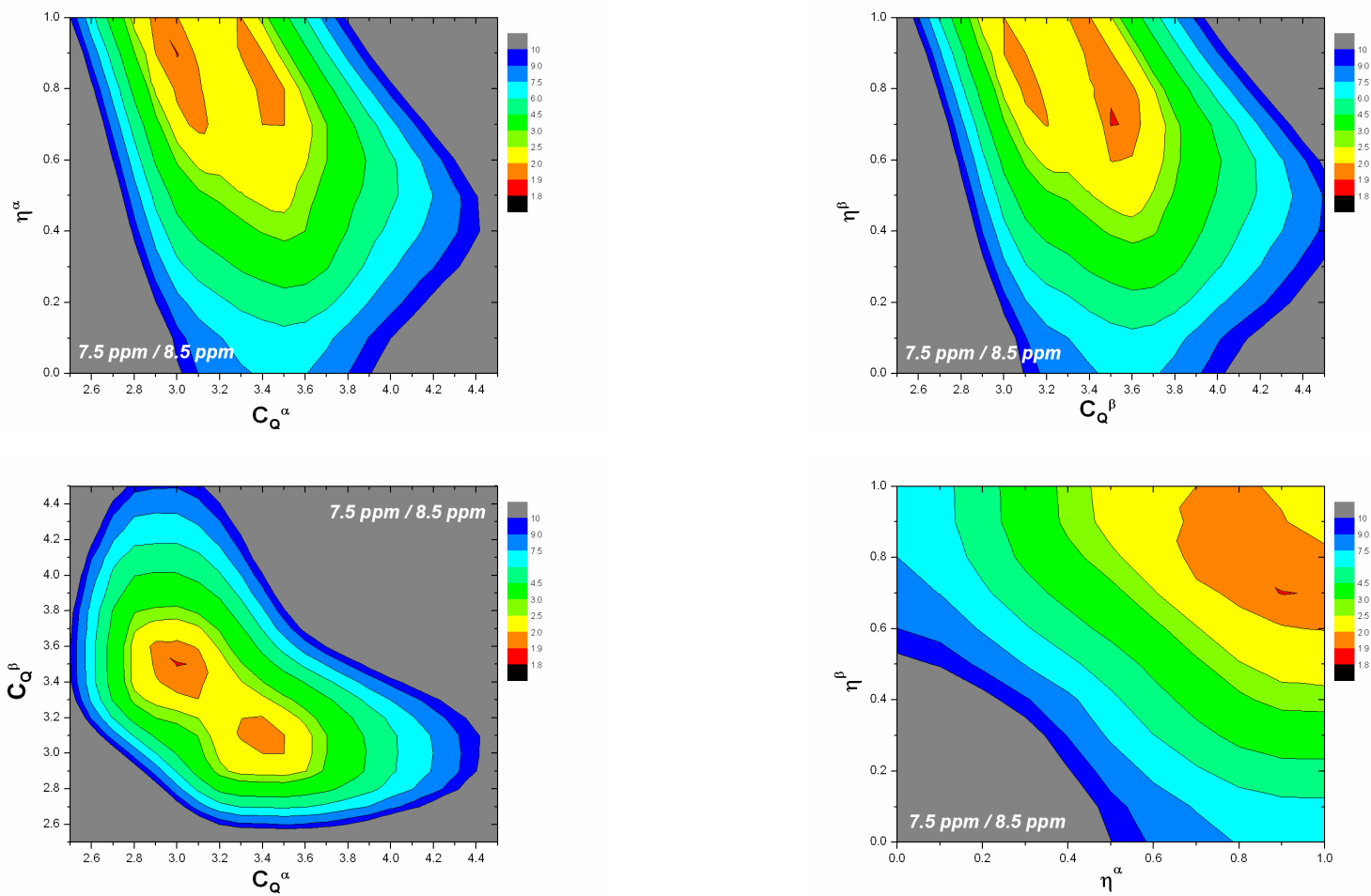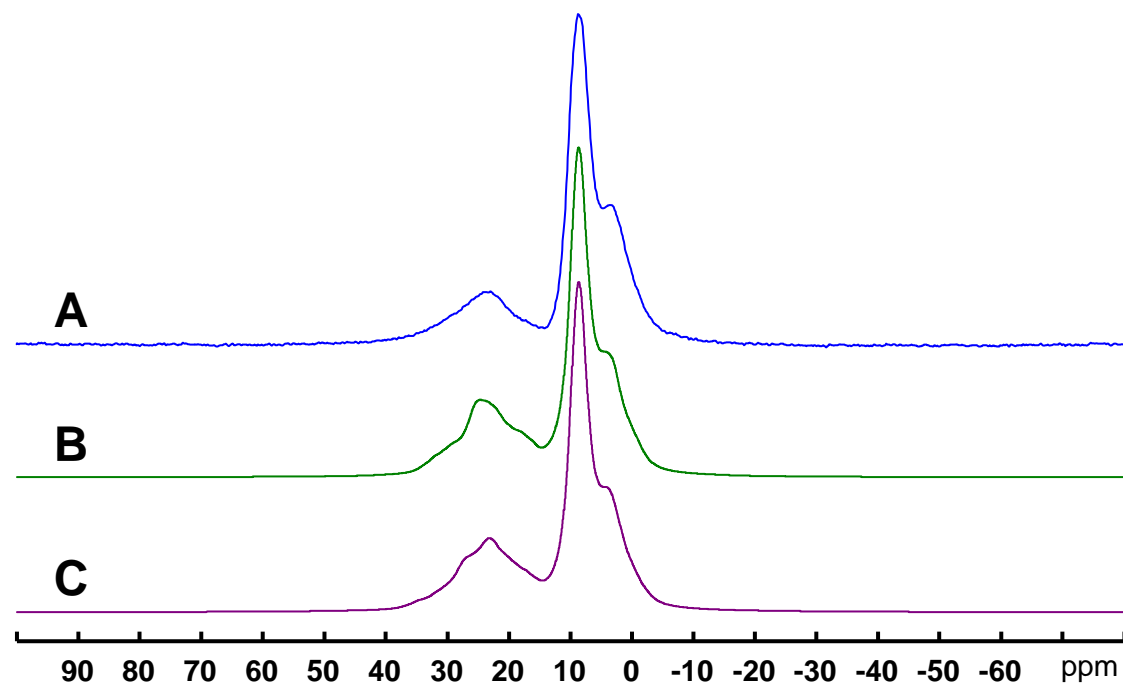
Figure SM6. ^{27}Al Rotor-Synchronized acquisition spectrum (11.75 T) of kaolinite KGa-1b (A). Simulated spectra using the parameter values from the 1-site model (B), from the 2-site model (δ^α=8.0 ppm, δ^β=7.5 ppm) (C).

Atom	x	y	z
Al1	0.2977	0.4943	0.4791
Al2	0.7900	0.3303	0.4766
Si1	0.9917	0.3366	0.0892
Si2	0.5057	0.1652	0.0908
O1	0.0478	0.3517	0.3146
O2	0.1161	0.6622	0.3159
O3	0.0069	0.4976	-0.0068
O4	0.1958	0.2227	0.0168
O5	0.1893	0.7691	-0.0065
O-H1	0.0473	0.9695	0.3250
O-H2	0.9595	0.1670	0.6060
O-H3	0.0332	0.4747	0.6054
O-H4	0.0346	0.8561	0.6118
H1	0.1425	0.0639	0.3277
H2	0.0570	0.1719	0.7375
H3	0.0450	0.5022	0.7357
H4	0.5438	0.3153	0.7334

Table SM1. Fractional atomic coordinates obtained from cell constraint optimization of the kaolinite structure. Lattice parameters : a=5.1535 Å , b=8.941 Å , c=7.3906 Å , α=91.926°, β=105.046° , γ=89.797°. Space group : C1.

Atom	x	y	z
Al1	0.1671	0.5352	-0.0027
Al2	0.3349	0.0284	-0.0028
O1	0.1774	0.2223	-0.1139
O2	0.6685	0.6496	-0.1041
O3	0.5003	0.1345	-0.1058
O4	-0.0220	0.6326	-0.1083
O5	0.2973	0.7218	-0.1064
O6	0.8240	0.1406	-0.1030
H1	0.0752	0.1416	-0.1255
H2	0.5725	0.5476	-0.1037
H3	0.4969	0.1116	-0.2066
H4	-0.0541	0.8184	-0.1128
H5	0.2944	0.7225	-0.2081
H6	0.8080	0.1565	-0.2046

Table SM2. Fractional atomic coordinates obtained from cell constraint optimization of the gibbsite structure. Lattice parameters : a=8.684 Å, b=5.078 Å, c=9.736 Å, β= 94.54°. Space group : P2$_1$/n.

Al1 site		Al2 site	
O-H4 – Al1 – O-H3	97.2°	O-H4 – Al2 – O-H3	97.6°
O-H4 – Al1 – O-H2	98.2°	O-H4 – Al2 – O-H2	96.3°
O-H4 – Al1 – O-H1	78.5°	O-H4 – Al2 – O-H1	78.5°
O-H4 – Al1 – O1	97.6°	O-H4 – Al2 – O2	97.3°
O2 – Al1 – O-H3	95.7°	O1 – Al2 – O-H3	76.3°
O2 – Al1 – O-H2	76.1°	O1 – Al2 – O-H2	96.0°
O2 – Al1 – O-H1	89.7°	O1 – Al2 – O-H1	90.3°
O2 – Al1 – O1	89.0°	O1 – Al2 – O2	89.8°
O-H3 – Al1 – O-H2	97.5°	O-H3 – Al2 – O-H2	96.7°
O-H2 – Al1 – O-H1	95.3°	O-H2 – Al2 – O2	78.0°
O-H1 – Al1 – O1	90.4°	O2 – Al2 – O-H1	91.1°
O1 – Al1 – O-H3	77.9°	O-H1 – Al2 – O-H3	95.5°
O-H4 – Al1 – O2	166.4°	O-H4 – Al2 – O1	166.8°
O-H3 – Al1 – O-H1	166.9°	O-H2 – Al2 – O-H1	167.2°
O-H2 – Al1 – O1	164.0°	O-H3 – Al2 – O2	164.6°

Table SM3. Bond angles for the two octahedral aluminum geometries, Al1 and Al2, derived from the optimized structure of kaolinite given in Table SM1.