# Supplementary Information

**Table 1 Nonbond Parameters for ClayFF Force Field**

| species | Symbol | Charge (e) | $D_0$ (kcal/mol) | $R_0$ (Å) |
|---|---|---|---|---|
| Water hydrogen | h* | 0.4100 | | |
| Hydroxyl hydrogen | ho | 0.4250 | | |
| Water oxygen | o* | -0.8200 | 0.1554 | 3.5532 |
| Hydroxyl oxygen | oh | -0.9500 | 0.1554 | 3.5532 |
| Tetrahedral silicon | st | 2.1000 | 1.8405*10^-6 | 3.7064 |
| Hydroxide calcium | cah | 1.0500 | 5.0298*10^-6 | 6.2428 |

**Table 2 Bond Parameters for ClayFF Force Field**

**a. bond-stretch of water and hydroxyl groups**

| species i | species j | $k_1$ (Kcal/mol/Å$^2$) | $r_0$ (Å) |
|---|---|---|---|
| h* | o* | 554.1349 | 1.0000 |
| ho | oh | 554.1349 | 1.0000 |

**b. angle bend of water**

| species i | species j | species k | $k_2$ (Kcal/mol/$\theta^2$) | $\theta_0$ |
|---|---|---|---|---|
| h* | o* | h* | 45.7696 | 109.47 |

Large numbers of experimental structures for simple oxides, hydroxides, and oxyhydroxides were employed to derive the optimal values for these parameters. The interaction parameters between the different atoms are obtained according to the arithmetic mean rule for the distance parameter, $R_0$, and the geometric mean rule for the energy parameter, $D_0$:

$$R_{o,ij} = \frac{1}{2}(R_{0,i} + R_{0,j})$$

$$D_{0,ij} = \sqrt{D_{0,i} D_{0,j}}$$

The short-range interactions in ClayFF force field are described by van der Waals energy represented via Lennard-Jones potential:

$$E_{VDW} = \sum_{i \neq j} D_{0,ij} \left[ \left( \frac{R_{0,ij}}{r_{ij}} \right)^{12} - 2 \left( \frac{R_{0,ij}}{r_{ij}} \right)^{6} \right]$$

The Coulombic potential is represented by:

$$E_{Coul} = \frac{e^2}{4\pi\epsilon_0} \sum_{i \neq j} \frac{q_i q_j}{r_{ij}}$$

where the partial charges $q_i$ and $q_j$ can be referred to Table 1, $e$ is the charge of the electron, and $\epsilon_0$ is the dielectric permittivity of vacuum (8.85419 *$10^{-12}$ F/m).

The bond stretch can angle bend in water molecules and hydroxyl groups can be described by simple harmonic potentials:

$$E_{bond\ stretch\ ij} = k_1(r_{ij} - r_0)^2$$

$$E_{angle\ bend\ ijk} = k_2(\theta_{ijk} - \theta_0)^2$$

where $k_1$ and $k_2$ are radial and angular stiffness, respectively. Parameters $r_0$ and $\theta_0$ represent the equilibrium hydroxyl bond length and H-O-H angle in water molecules, respectively. (see Table 2)