Supporting information for "Molecular dynamics study on ions and water confined in the nanometer channel of Friedel's salt: structure, dynamics and interfacial interaction"

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1. ClayFF Force field

In molecular dynamics simulations, the total energy in the simulation is determined by the interaction terms between atoms in the system. In general, total energy includes Coulomb interactions, van der Waals forces, and bonding interactions. The formula is as follows:

$$U = \sum_{i,j}\left\{\frac{q_i q_j}{4\pi\varepsilon_0 r} + 4\varepsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r_{ij}}\right)^2 + \left(\frac{\sigma_{ij}}{r_{ij}}\right)^6\right]\right\} \quad (1)$$

Where r_{ij} is the distance between the atom i and the atom j, q_i and q_j are partial charges centered on these atoms, σ and ε are parameters of the Lennard-Jones interaction potential, and ε_0 is the dielectric permittivity of vacuum (equal to 8.85×10^{-12}F/m). These parameters and partial charges are listed in the Table 1. The Lennard-Jones parameter, a combination of interactions of different ions, can be obtained as follows:

$$\sigma = \frac{\sigma_{ii} + \sigma_{jj}}{2}$$

$$\varepsilon_{ij} = \sqrt{\varepsilon_{ii}\varepsilon_{jj}} \quad (2)$$

All long-range electrostatic interactions in the formula are calculated by the Ewald summation method.

Table S1 Force Field Parameters of Related Atoms in ClayFF Force Field：

species	**symbol**	**Charge (e)**	D_0 **(kcal/mol)**	R_0 **(Å)**
Water hydrogen	Hw	0.41		
Hydroxyl hydrogen	Ho	0.425		
Water oxygen	Ow	-0.82	0.1554	3.5532
Hydroxyl oxygen	Oh	-0.875	0.1554	3.5532
Octahedral aluminum	Al	1.575	$5.56\text{x}10^{-6}$	4.7929
sodium ion	Na	1	0.1	2.6378
chloride ion	Cl	-1	0.1	4.9388
Octahedral Calcium	Ca	1.05	$2.10\text{x}10^{-5}$	6.2409

2. Local structure of ions

The local structure and chemical environment of ions can be understood by investigating the coordination neighbors of ions while the adsorption mechanism can be explained by comparing the coordination number in different environments, i.e. in the vicinity of the interface or in the solution. Table S2 shows the coordination number of sodium ions. It shows that the coordination number of sodium ions is 5.76, including the coordination of 5.71 water molecules and 0.05 chloride ions. It indicates that the local environment of sodium ions in the pore solution mainly forms a hydration shell with the surrounding water molecules. It is worth noting that the

coordination of the structural chloride ions occurs around the sodium ions in the solution. It suggests that the structural chloride ion in the Friedel's salt surface tends to dissolve into the solution. Compared with the sodium ions in the solution, the coordination number of water molecules around the sodium ions in the vicinity of the interface decreases while the coordination number of the chloride ions increases to 0.33. In this case, this indicates that the adsorption capacity of the interface is limited due to the structural chloride ion attraction effect. Sodium ions and chloride ions could form ion clusters to enhance the adsorption. The coordination number of chloride ion in solution is 7.5, which is mainly demonstrated that chloride ion attracts water molecules and form a layer of hydration film. Differently, the chloride ion at the interface has a more complex chemical environment. As listed in Table S3, the coordination number of water molecules accounts for 5.61 in a 7.48 coordination sites of total chloride ions. Meanwhile, the coordination number of the structure calcium ions and hydroxyl groups increases to 1.85, indicating that the adsorption of chloride ions mainly depends on the Ca^{2+} attraction in Friedel's salt surface structure and hydrogen bonding network with hydroxyl groups.

Table S2. The coordination number of sodium ions.

	Na	
	Surface	**pore**
-O_w	4.91	5.71
-O_h	0	0

-Cl_w	0.05	0.02
-Cl_s	0.28	0.03
Total	5.24	5.76

Table S3. The coordination number of chloride ions.

	Cl	
	Surface	**Pore**
-O_w	5.61	7.47
-Na	0.02	0.03
-Ca	0.62	0
-H_o	1.23	0
Total	7.48	7.50