Supporting Information

Mechanism of Wettability Alteration of the Calcite \{10\bar{1}4\} Surface

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Figure S1. Pressure, temperature, total energy, and root mean square deviation in the NPT MD simulation of (deionized) water on the calcite \{10\bar{1}4\} surface.

Figure S2. Arrangement of water molecules on the calcite \{1014\} surface (average structure from the last 5 ps of the MD simulation). H is represented in white and O in red.
**Figure S3.** Side view of the calcite slab used in the simulations (2 × 3 supercell, four layers of Ca$^{2+}$ and CO$_3^{2-}$ ions; vacuum region of 15 Å thickness). C is represented in gray, O in red, and Ca in green.

**Figure S4.** Top and side views of the favorable binding geometries of one Cl$^-$ ion and one (1W) or two (2W) water molecules (numbered) on the calcite {1014} surface. Dashed lines mark hydrogen bonds. H is represented in white, C in gray, O in red, Cl in gold, and Ca in green.
Figure S5. Top views of the less favorable binding geometries for one (1W), two (2W), or three (3W) water molecules (numbered) on the calcite \{10\bar{1}4\} surface. Dashed lines mark hydrogen bonds. H is represented in white, C in gray, O in red, and Ca in green.

Figure S6. Pressure, temperature, total energy, and root mean square deviation in the NPT MD simulation of NaCl/water on the calcite \{10\bar{1}4\} surface.
Figure S7. Pressure, temperature, total energy, and root mean square deviation in the NPT MD simulation of MgCl$_2$/water on the calcite \{10\overline{1}4\} surface.

Figure S8. Pressure, temperature, total energy, and root mean square deviation in the NPT MD simulation of CaCl$_2$/water on the calcite \{10\overline{1}4\} surface.
Figure S9. Pressure, temperature, total energy, and root mean square deviation in the NPT MD simulation of Na₂SO₄/water on the calcite \{10\textbar 4\} surface.

Figure S10. Pressure, temperature, total energy, and root mean square deviation in the NPT MD simulation of smart water on the calcite \{10\textbar 4\} surface.
**Figure S11.** Pressure, temperature, total energy, and root mean square deviation in the NPT MD simulation of sea water on the calcite \{10\overline{1}4\} surface.

**Figure S12.** Representative structures of hydrated divalent salt ions (Mg\(^{2+}\)\cdot6\text{H}_2\text{O}, \text{Ca}^{2+}\cdot7\text{H}_2\text{O}; average structure from the last 5 ps of the MD simulation). H is represented in white, O in red, Ca in green, and Mg in magenta.
Figure S13. Effect of the charge transfer reported in Table 3 on the density of states.