

1 Effect of layer bending on montmorillonite hydration and
2 structure from molecular simulation

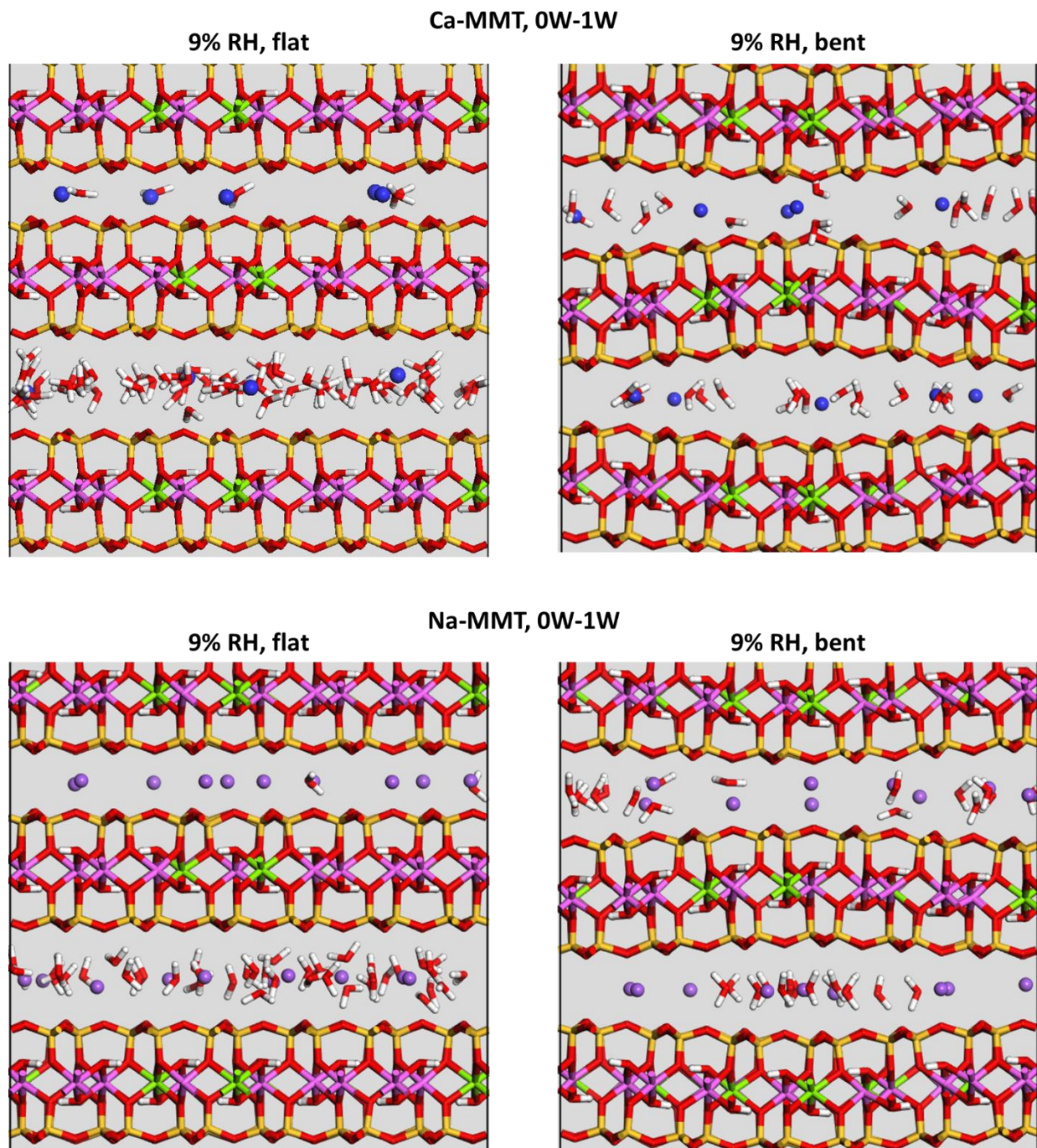
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9 Supplementary Information

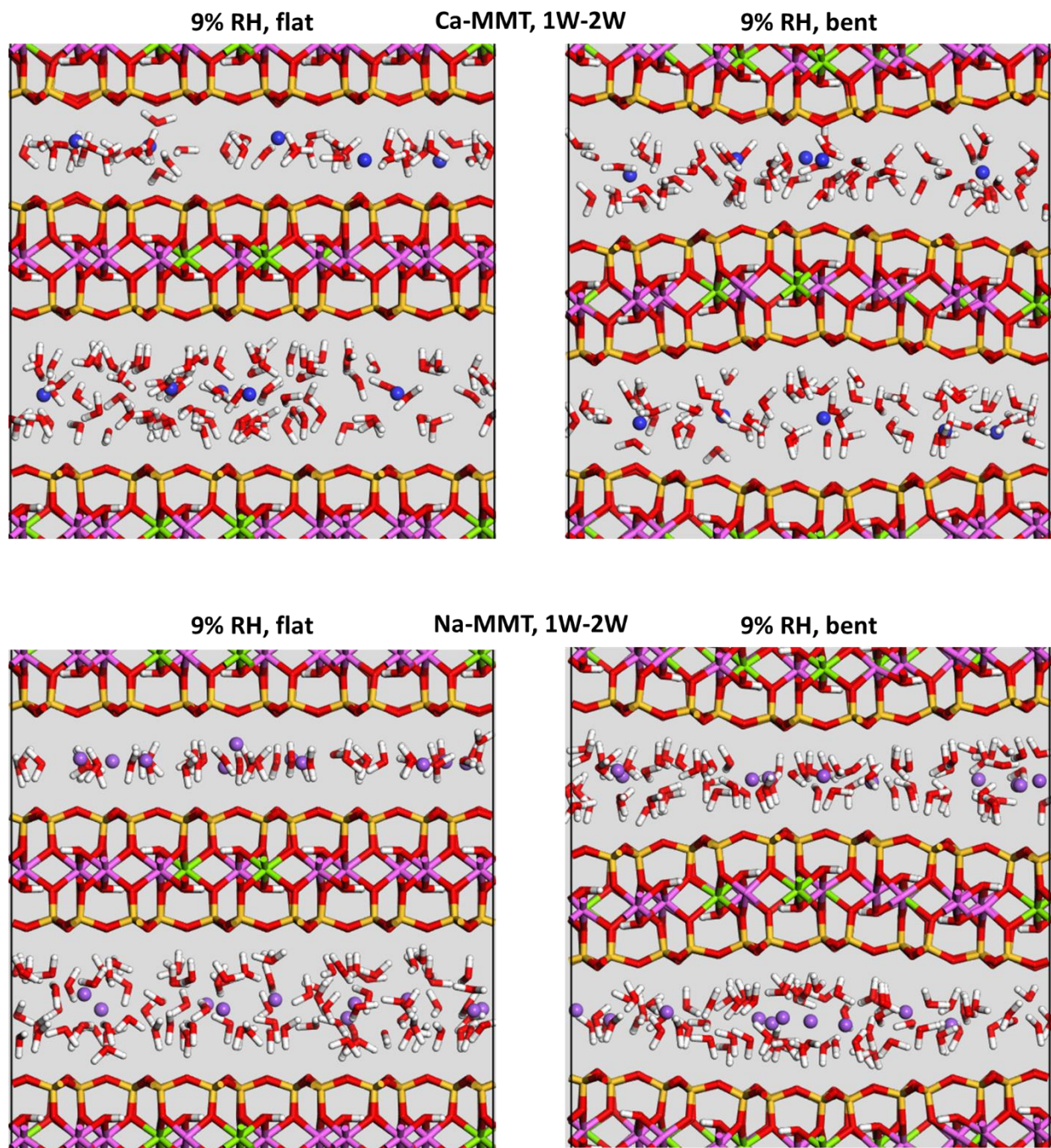


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11 **Figure S1.** Equilibrium snapshots (yz plane) from GCMC simulations in the 0W-1W hydration

12 state at 9% RH. Atoms are colored as follows: red (O), white (H), yellow (Si), magenta (Al),

13 green (Mg), purple (Na), blue (Ca). Periodic boundaries are shown as black lines.

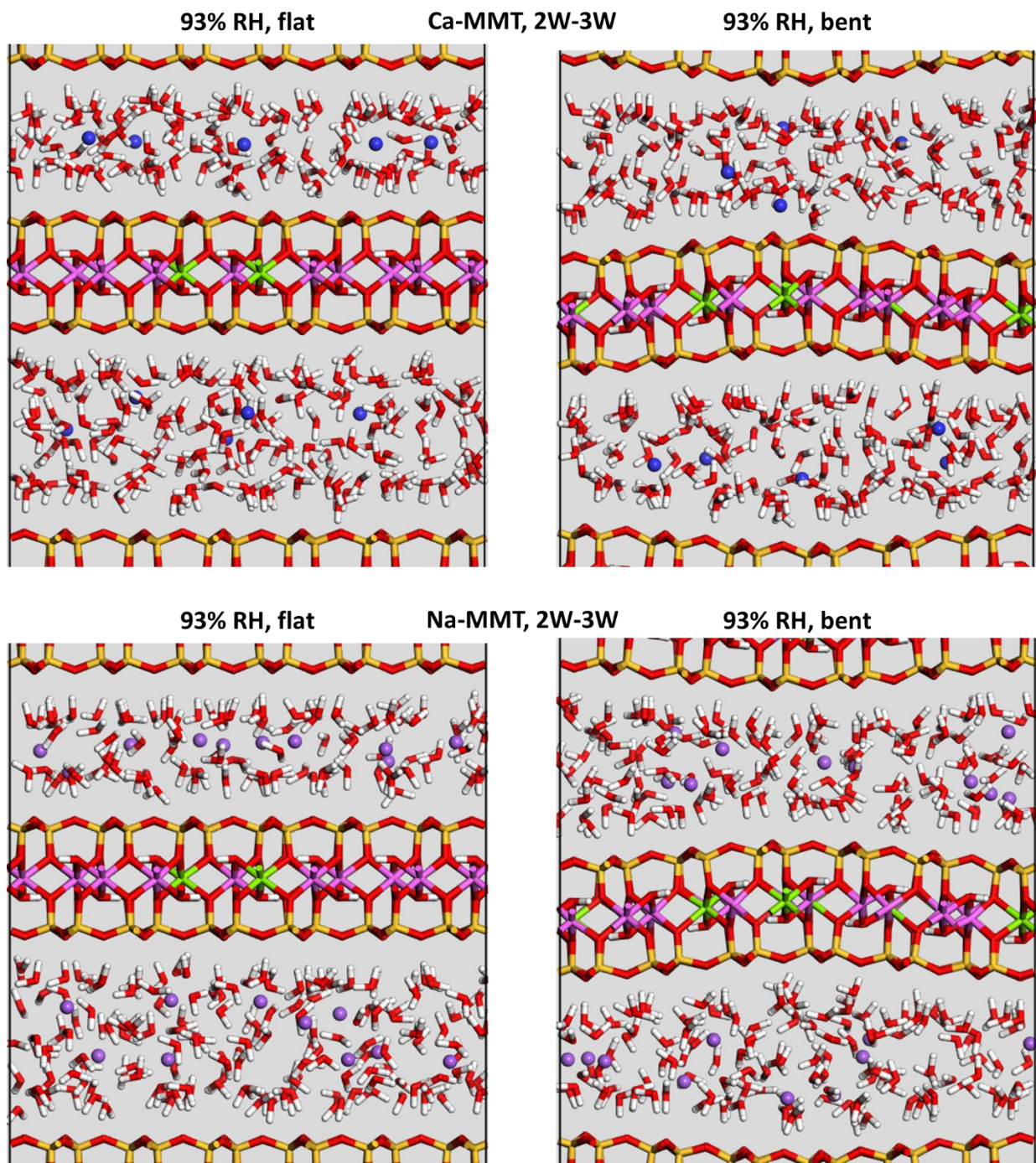


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15 **Figure S2.** Equilibrium snapshots (yz plane) from GCMC simulations in the 1W-2W hydration

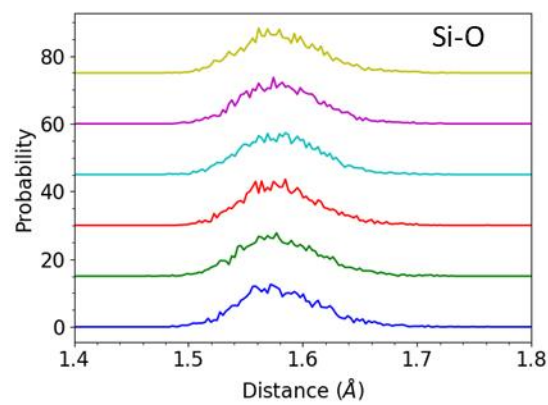
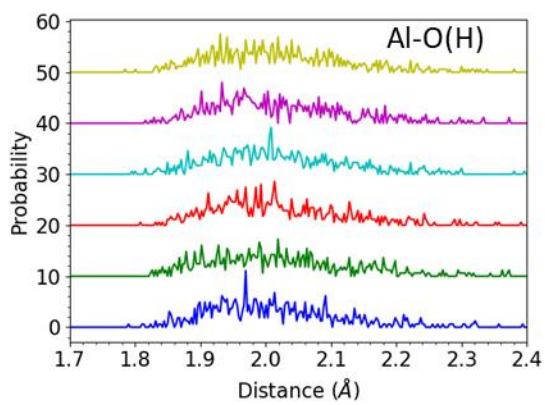
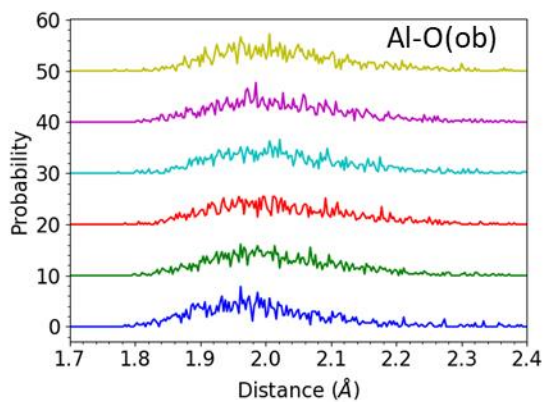
16 state at 9% RH. Atoms are colored as follows: red (O), white (H), yellow (Si), magenta (Al),

17 green (Mg), purple (Na), blue (Ca). Periodic boundaries are shown as black lines.



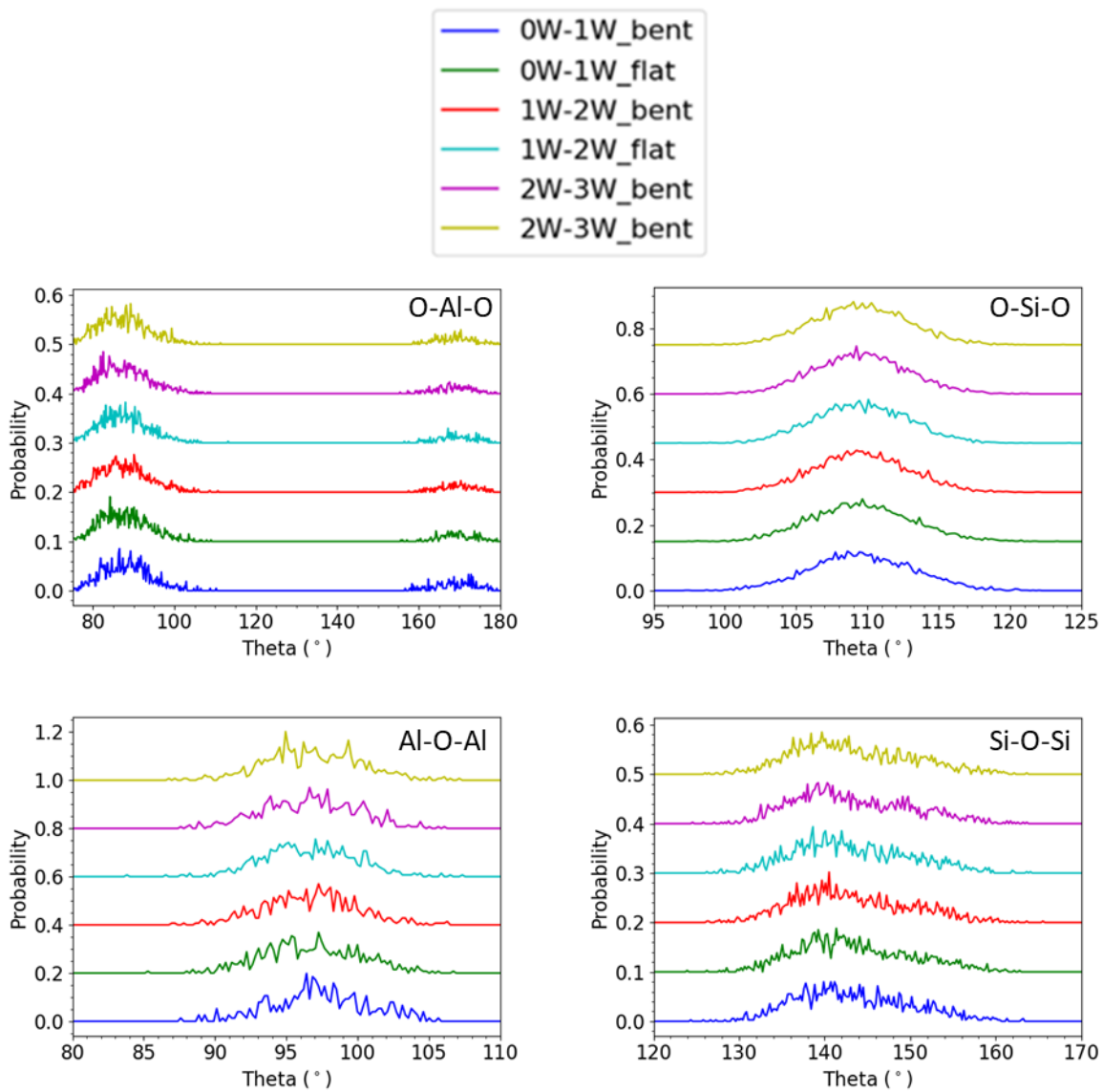
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19 **Figure S3.** Equilibrium snapshots (yz plane) from GCMC simulations in the 2W-3W hydration
 20 state at 93% RH. Atoms are colored as follows: red (O), white (H), yellow (Si), magenta (Al),
 21 green (Mg), purple (Na), blue (Ca). Periodic boundaries are shown as black lines.



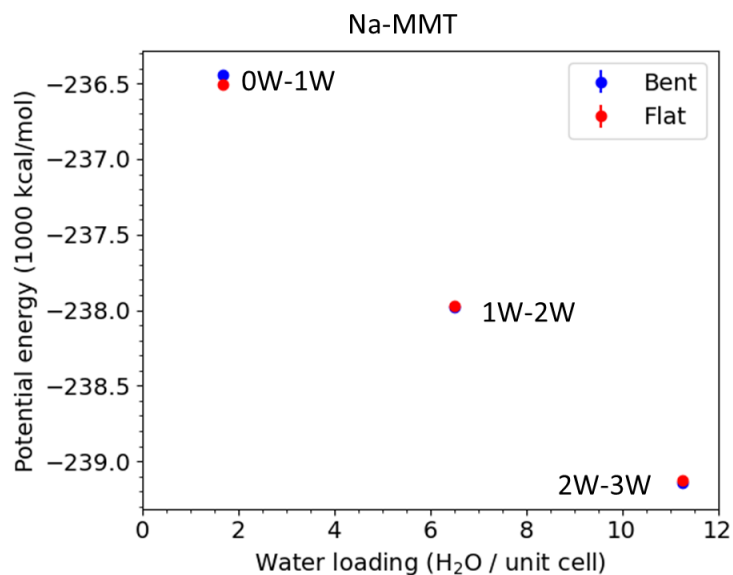
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23 **Figure S4.** Bond distributions in the montmorillonite lattice in each mixed hydration state.

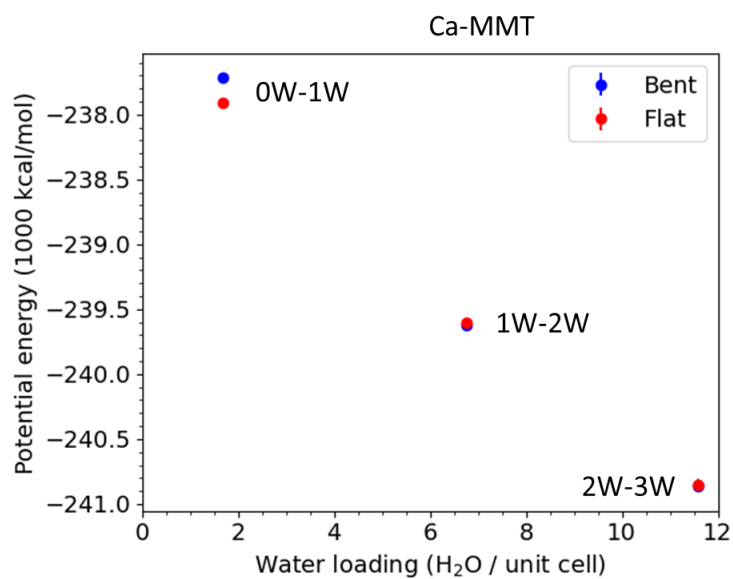


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25 **Figure S5.** Angle distributions in the montmorillonite lattice in each mixed hydration state.



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28 **Figure S6.** Average potential energies from 50 ps MD simulations of Na-MMT and Ca-MMT in
 29 each mixed hydration state. Models with the same layer spacing (*e.g.* 0W-1W) contained
 30 identical number of interlayer water molecules, taken from the maximum loading from
 31 adsorption isotherm of the bent configuration. Atoms in the clay layers were held fixed during
 32 the simulations.