Molecular Dynamics Simulation and Cryo Electron Microscopy Investigation of AOT Surfactant Structure at the Hydrated Mica Surface

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Supplementary Material

Comparison of random initial configuration and initial AOT bilayer at the mica surface S2
Micrograph of a mica/ice interface S3
Micrograph from 16 CMC Na-AOT at the mica surface S4
Snapshots from MD simulations of 64 AOT systems at 300 K S5
One-dimensional atomic density profiles from MD simulations S6
Cation-oxygen radial distribution functions and running coordination numbers S9
Snapshots from MD simulations at 300 K and 350 K S10
MD trajectories of AOT in the *xy* plane above the mica surface S12

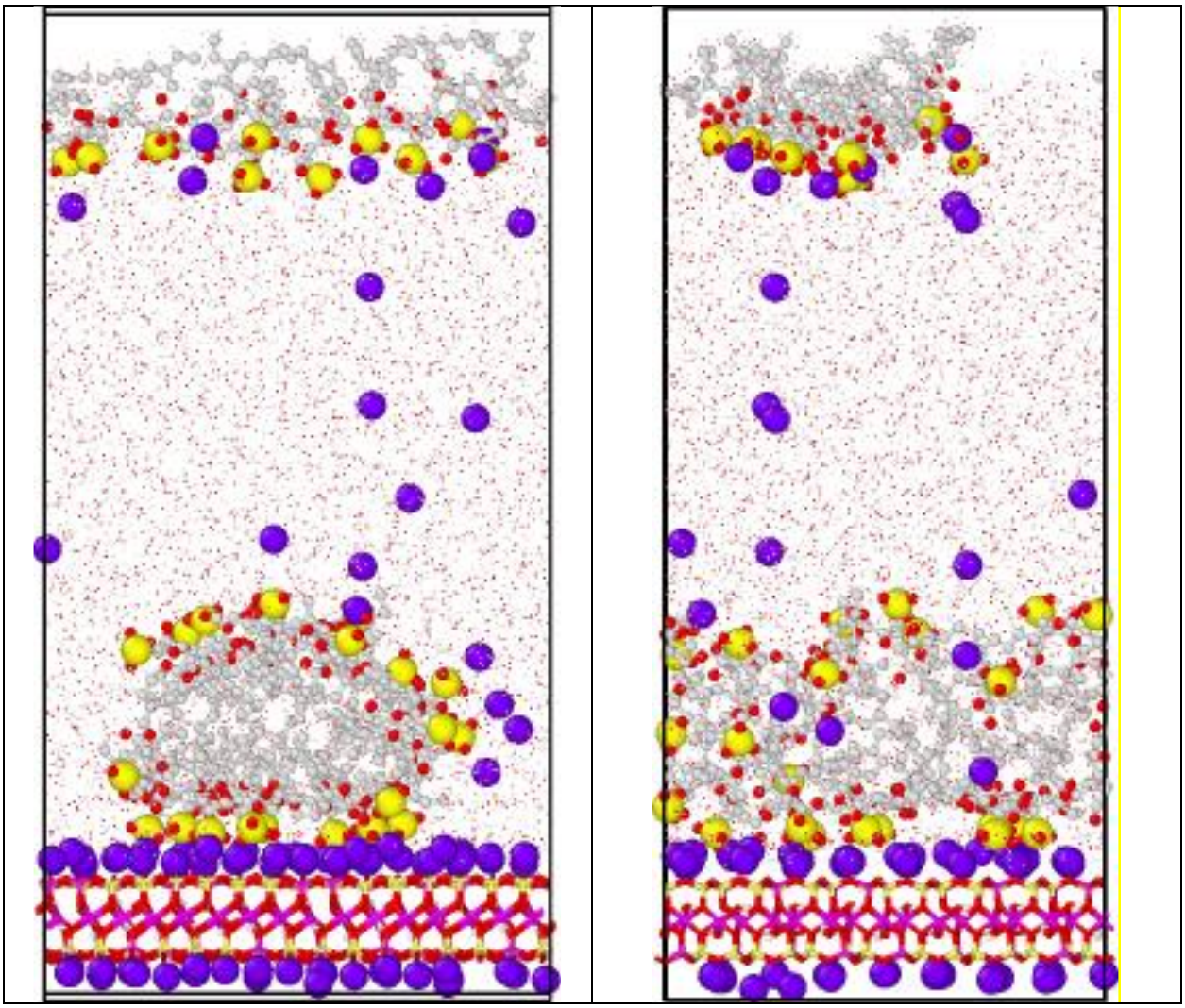


Figure S1. Snapshots from MD simulations from 32 AOT systems at 300 K with K^+ counterions. Results are shown for random initial configuration of AOT anions (left) and initial AOT bilayer at the mica surface (right). The simulation cell boundaries are shown as black lines. The mica layer is shown as sticks (red = O, dark yellow = Si, magenta = Al), AOT anions as spheres (red = O, gray = C, yellow = S), K^+ as large purple spheres, and water atoms as gray dots (red = O, gray = H). Hydrogen atoms in the mica layer and AOT anions are omitted for clarity.

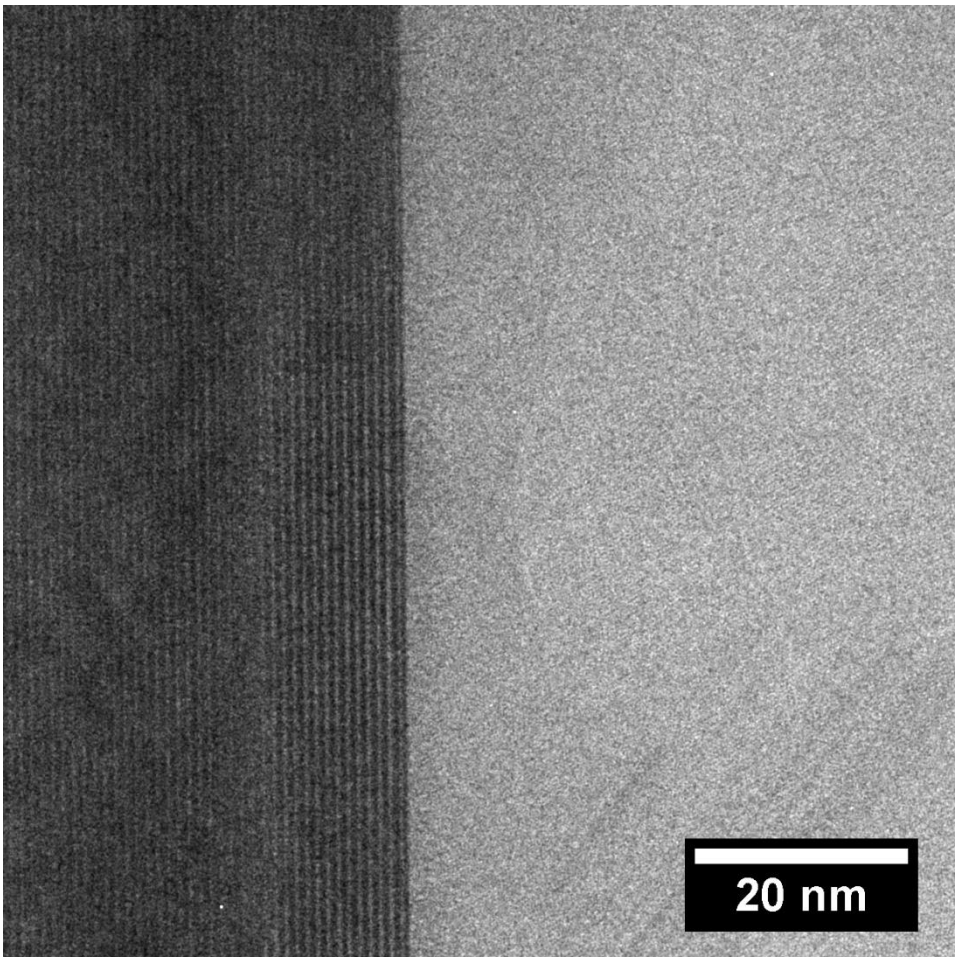


Figure S2. Micrograph of a mica/ice interface at higher resolution showing the mica basal plane spacing of 1.0 nm. Acquired on a FEI Titan operated at 300 kV with a K2 direct electron detector.

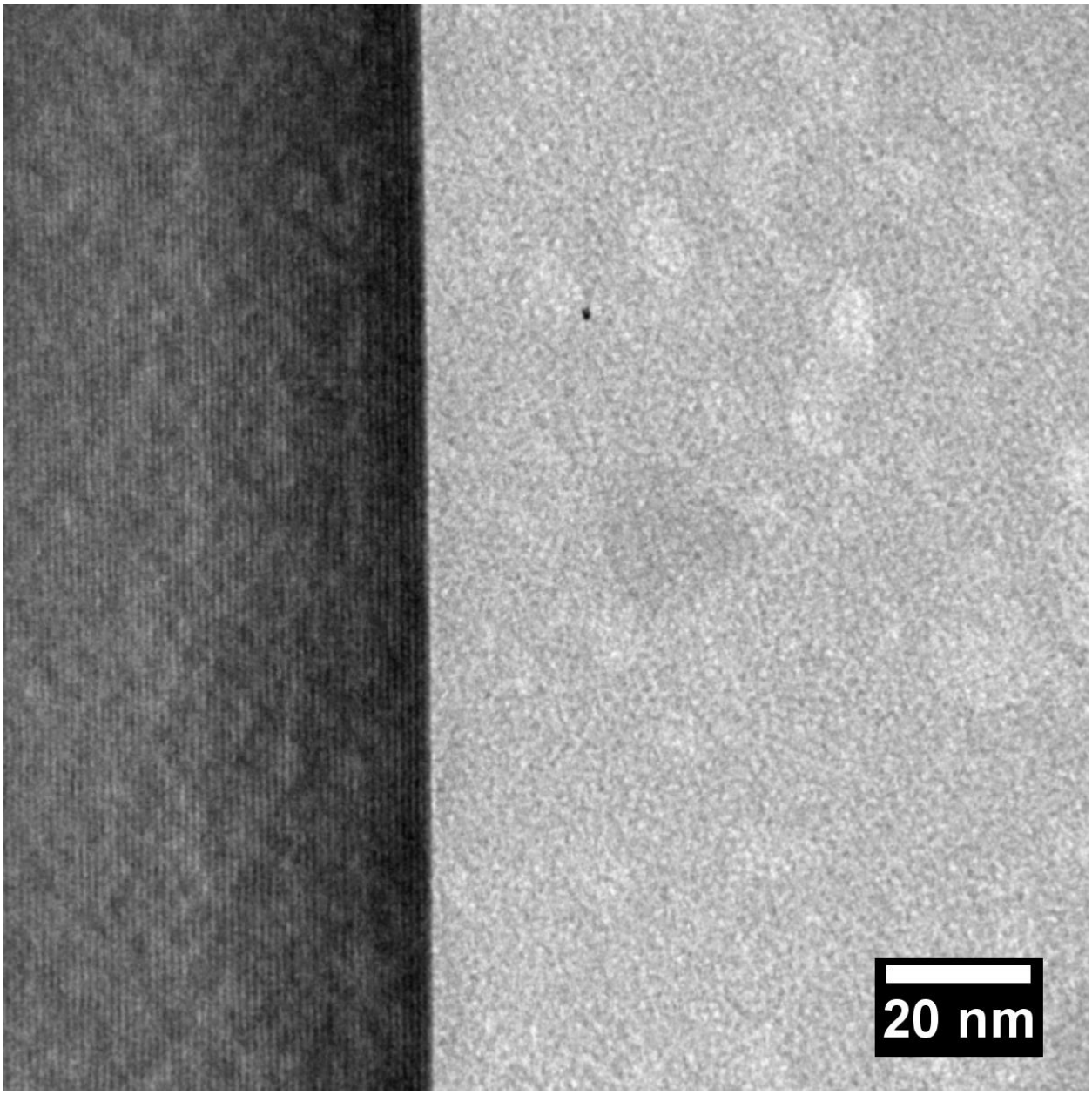


Figure S3. Micrograph from 16 CMC Na-AOT with no discrete micelle structure at the mica interface. The contrast in the solution has a characteristic length of ~2 nm suggesting the solution is a dense collection of micelles formed from the high concentration of AOT.

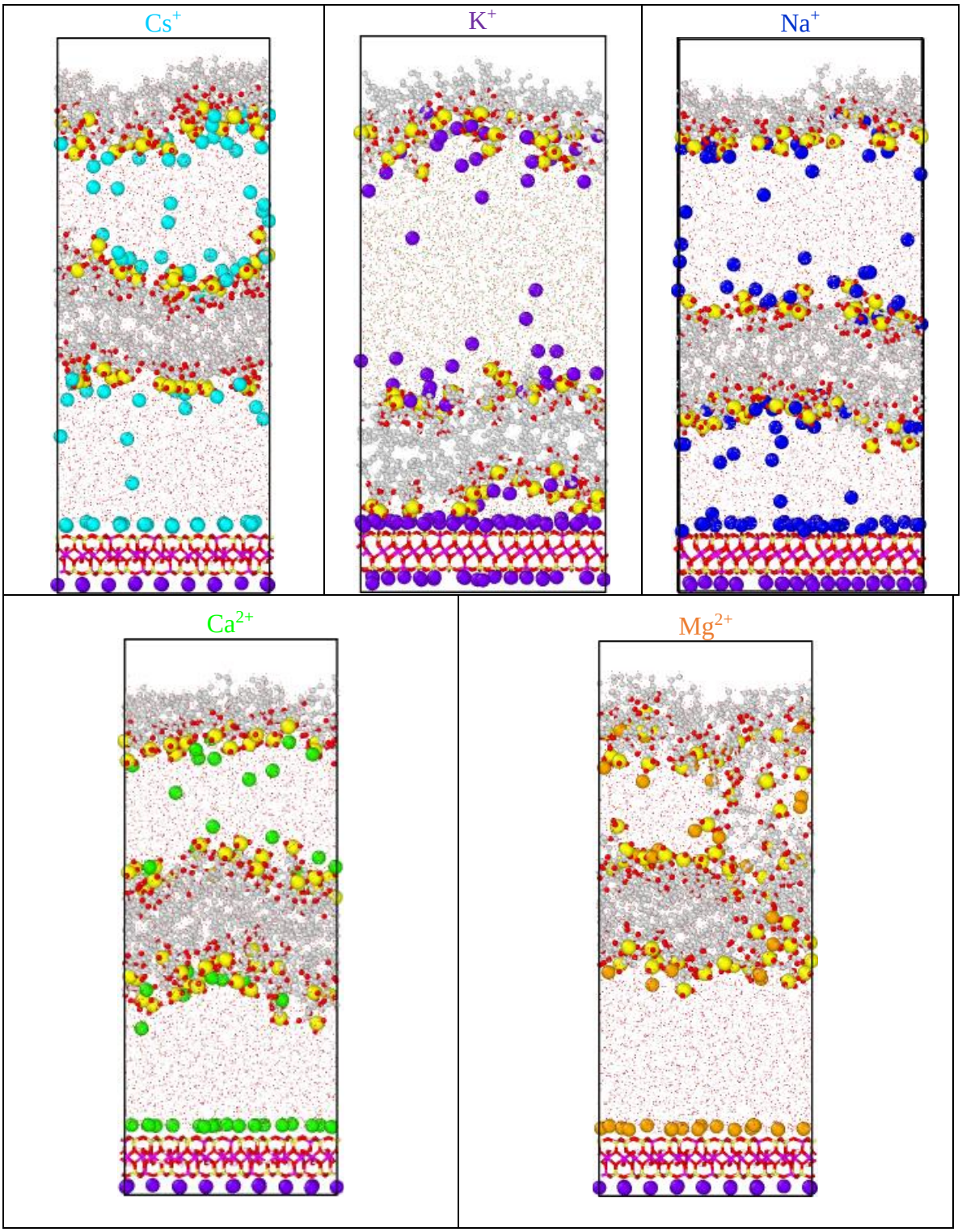


Figure S4. Snapshots from MD simulations of 64 AOT systems at 300 K showing the formation of bilayers at the mica-water interface. Atom rendering and coloring is the same as Figure S1 with aqueous cations shown as large spheres (purple = K^+, light blue = Cs^+, blue = Na^+, green = Ca^{2+}, dark orange = Mg^{2+}).

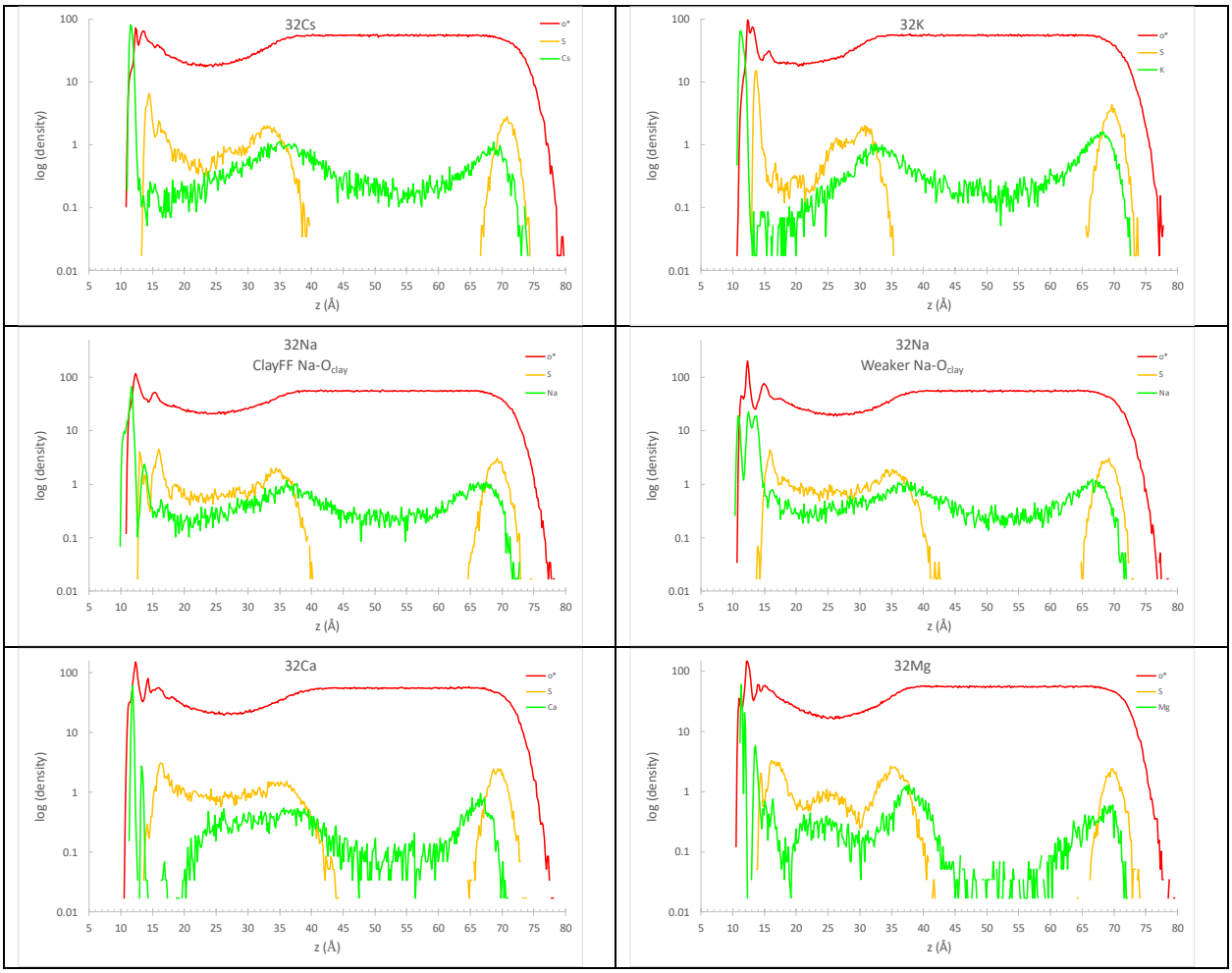


Figure S5. One-dimensional atomic density profiles from MD simulations of 32 AOT systems at 300 K. Profiles are shown for water oxygen atoms (o*), AOT sulfur atoms (S), and counterions (Cs^+, K^+, Na^+, Ca^{2+}, Mg^{2+}). Profiles are shown for Na-AOT with original (ClayFF) Na-O_{clay} Lennard-Jones parameters and modified (weaker) Na-O_{clay} Lennard-Jones parameters.[1]

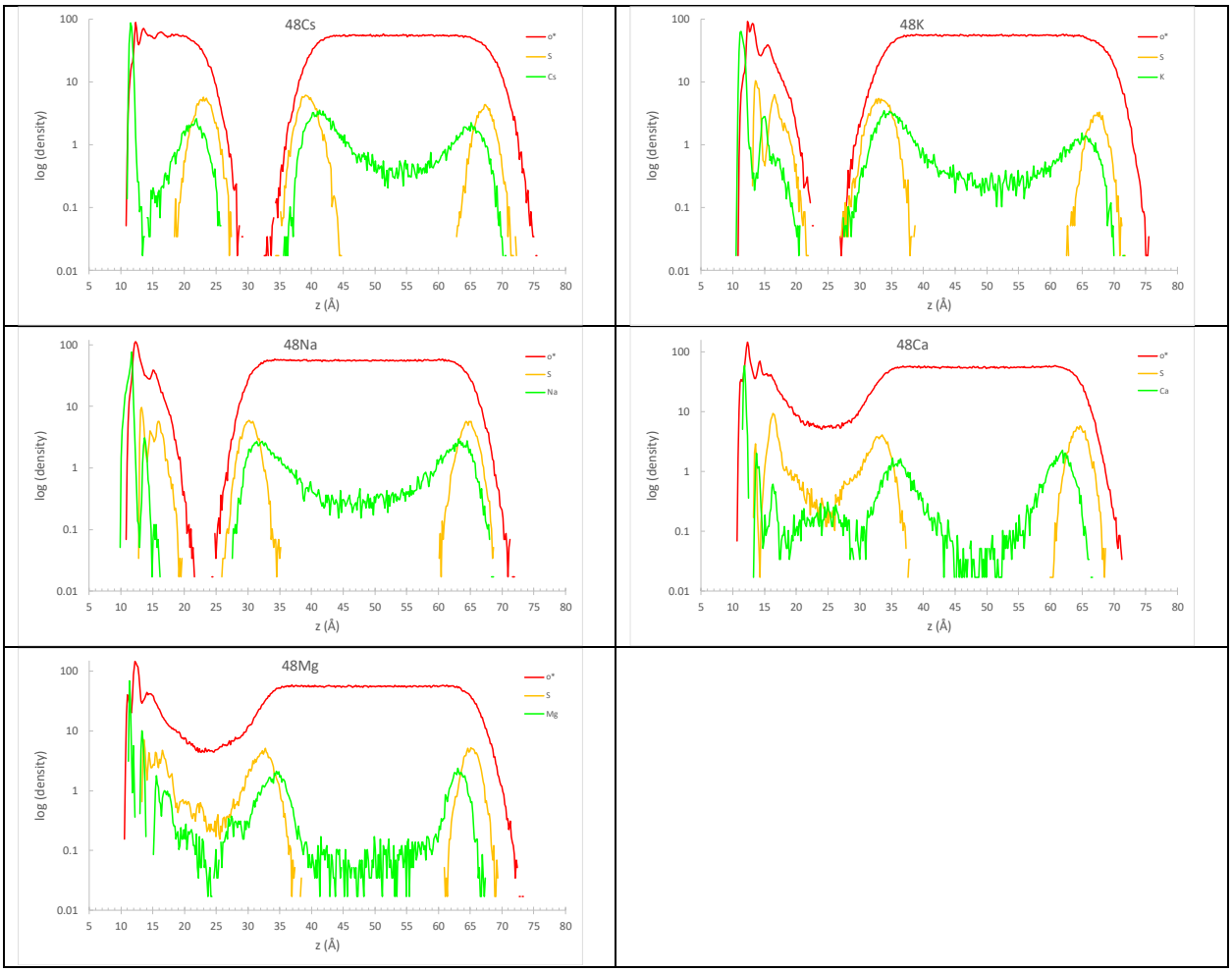


Figure S6. One-dimensional atomic density profiles from MD simulations of 48 AOT systems at 300 K. Profiles are shown for water oxygen atoms (o*), AOT sulfur atoms (S), and counterions (Cs^+, K^+, Na^+, Ca^{2+}, Mg^{2+}).

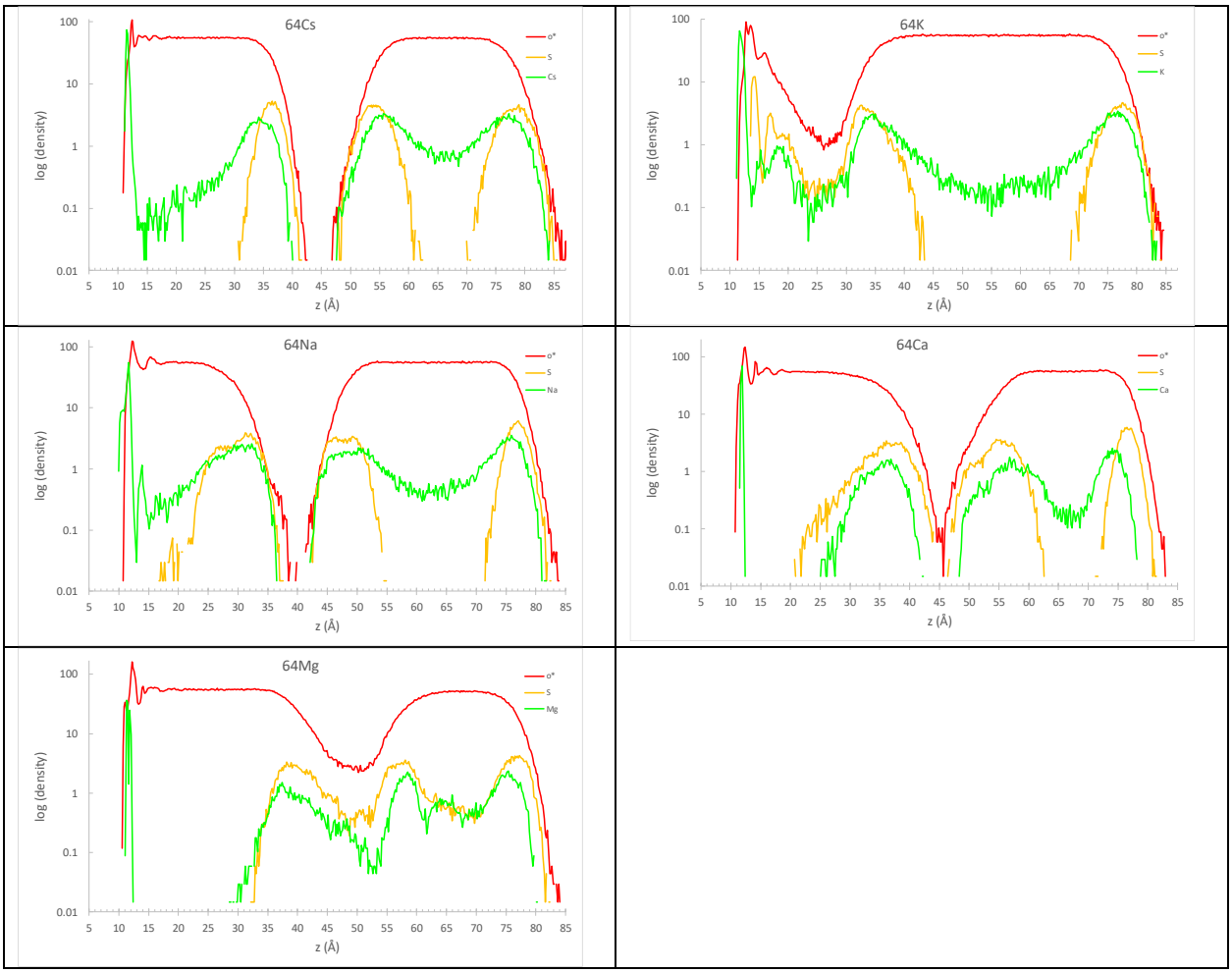


Figure S7. One-dimensional atomic density profiles from MD simulations of 64 AOT systems at 300 K. Profiles are shown for water oxygen atoms (o*), AOT sulfur atoms (S), and counterions (Cs^{+}, K^{+}, Na^{+}, Ca^{2+}, Mg^{2+}).

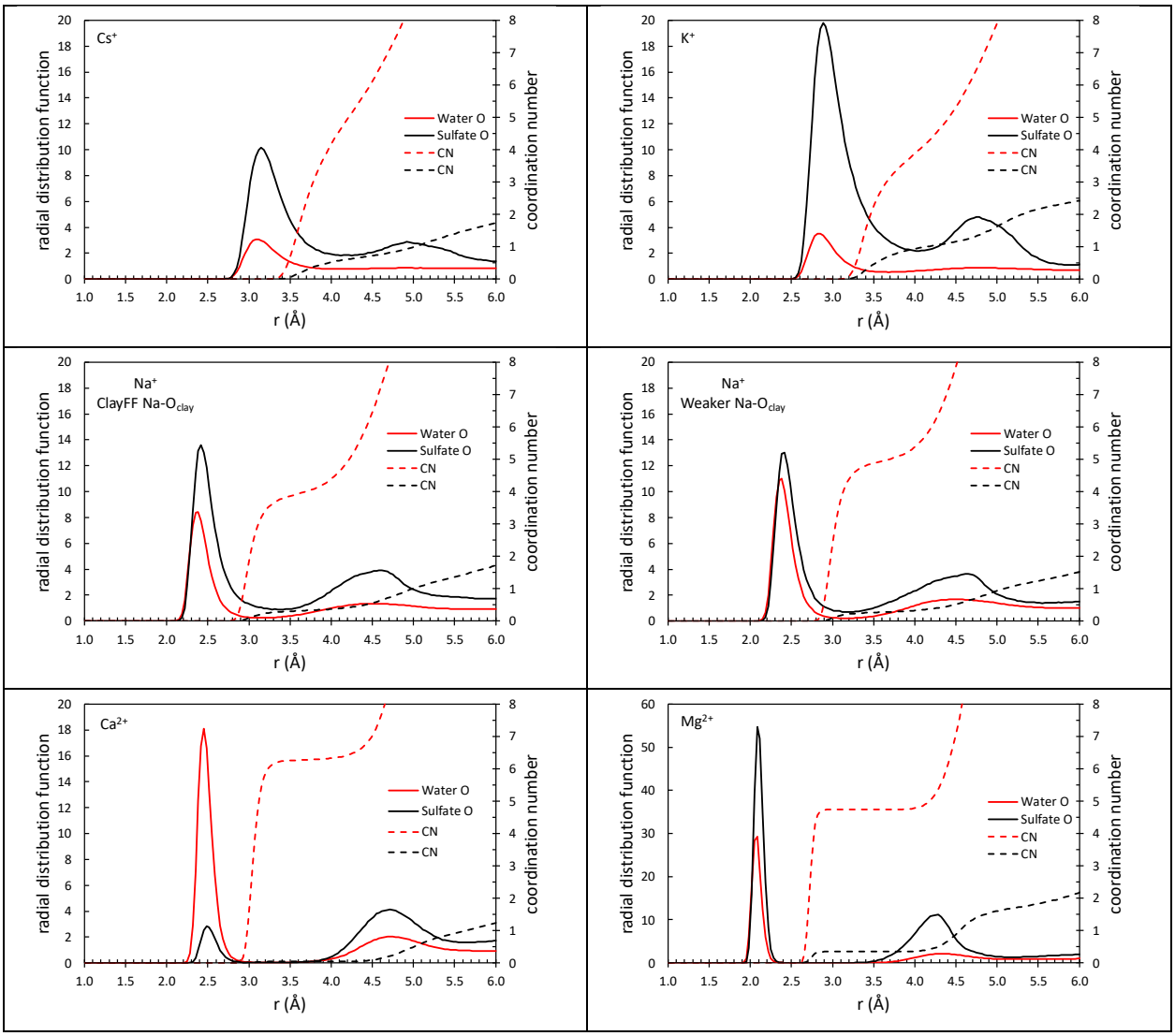


Figure S8. Cation-oxygen radial distribution functions and running coordination numbers from MD simulations of 32 AOT systems at 300 K. Plots are shown for Na-AOT with original (ClayFF) Na-O_{clay} Lennard-Jones parameters and modified (weaker) Na-O_{clay} Lennard-Jones parameters.[1]

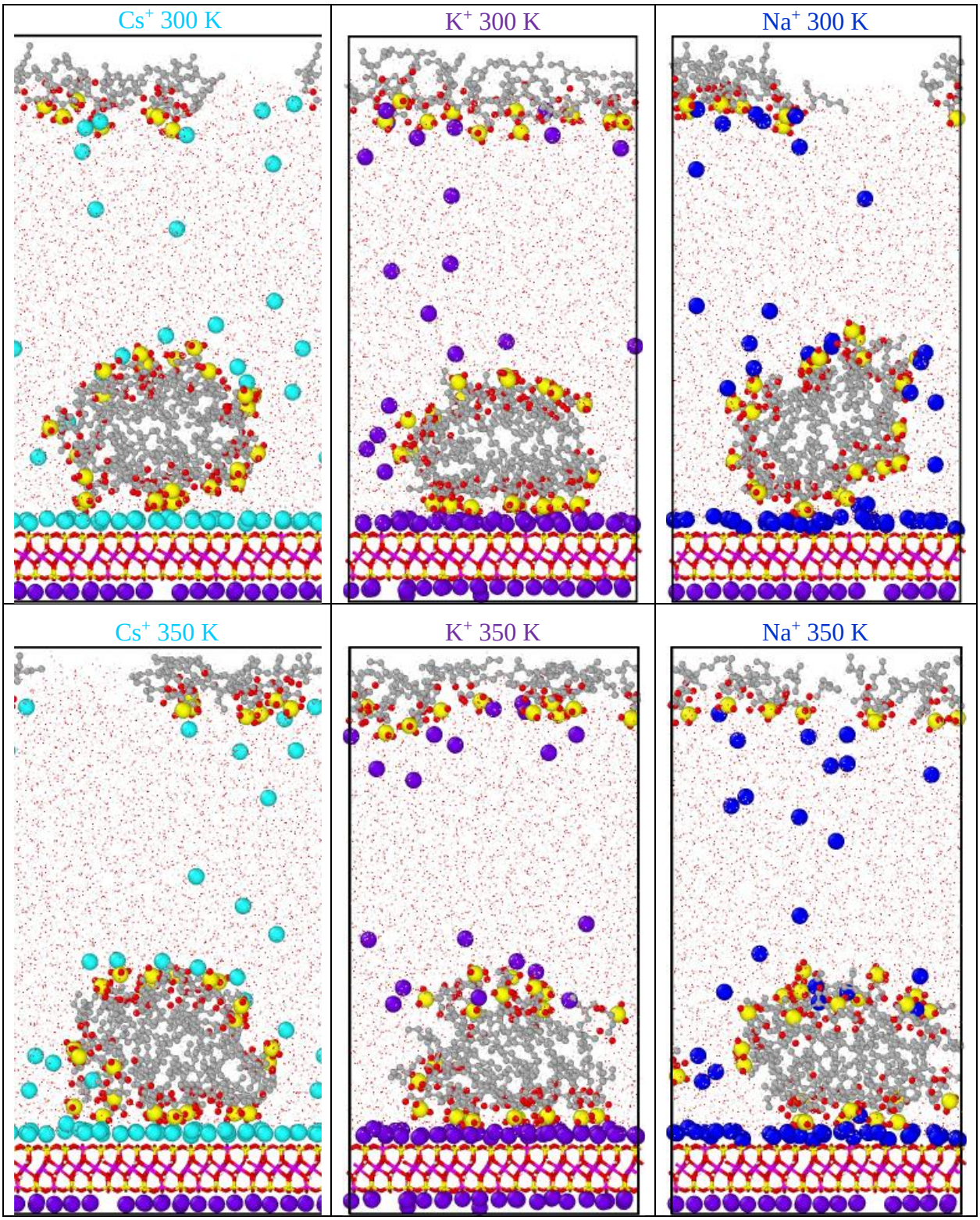


Figure S9. Snapshots from MD simulations of 32 AOT systems with monovalent counterions at 300 K and 350 K. Atom rendering and coloring is the same as Figure S1 with aqueous cations shown as large spheres (purple = K^+, light blue = Cs^+, blue = Na^+).

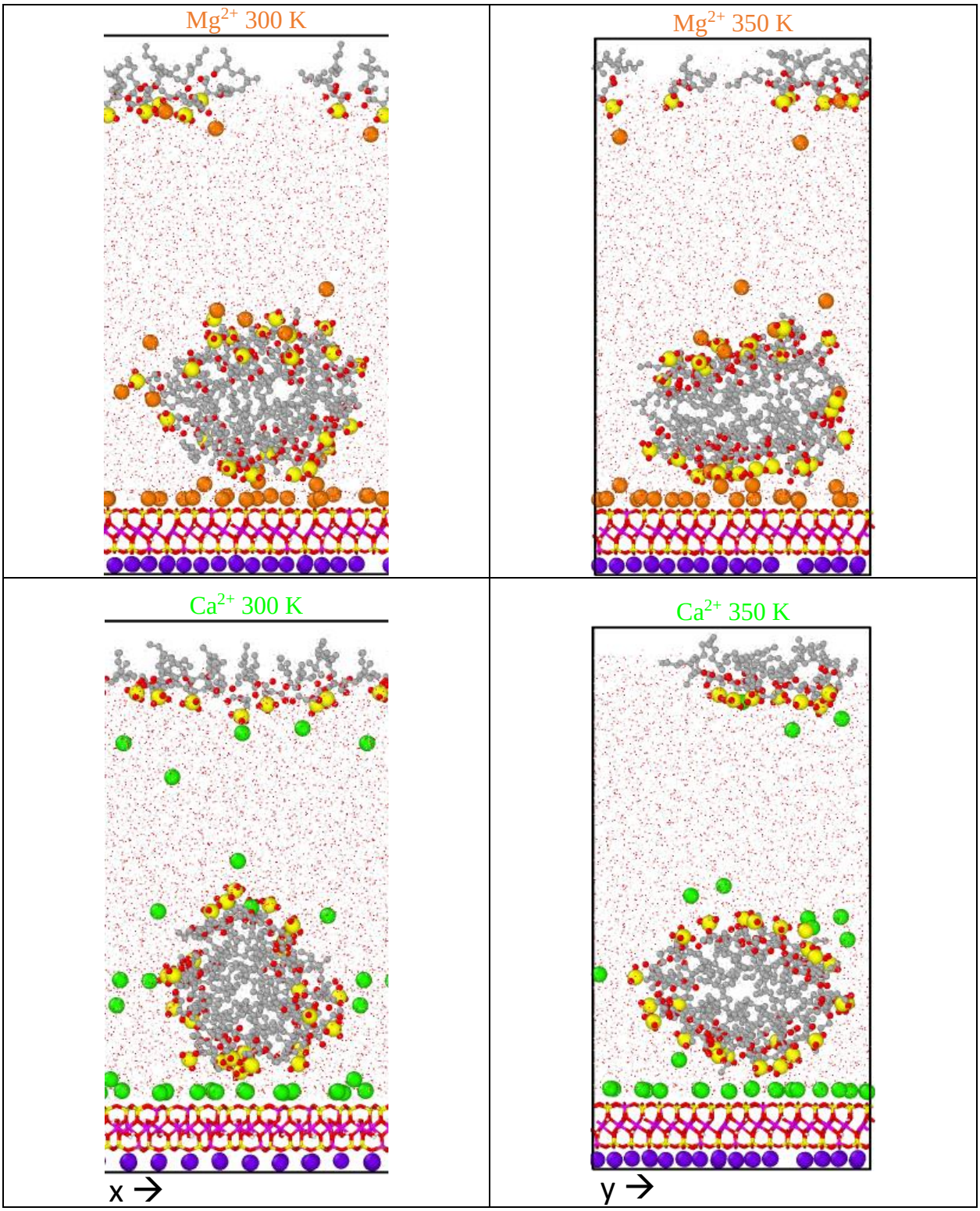


Figure S10. Snapshots from MD simulations of 32 AOT systems with divalent counterions at 300 K and 350 K. Atom rendering and coloring is the same as Figure S1 with aqueous cations shown as large spheres (purple = K^+, green = Ca^{2+}, dark orange = Mg^{2+}). Note the change in micelle orientation for Ca-AOT: parallel to the *y*-axis at 300 K but parallel to the *x*-axis at 350 K.

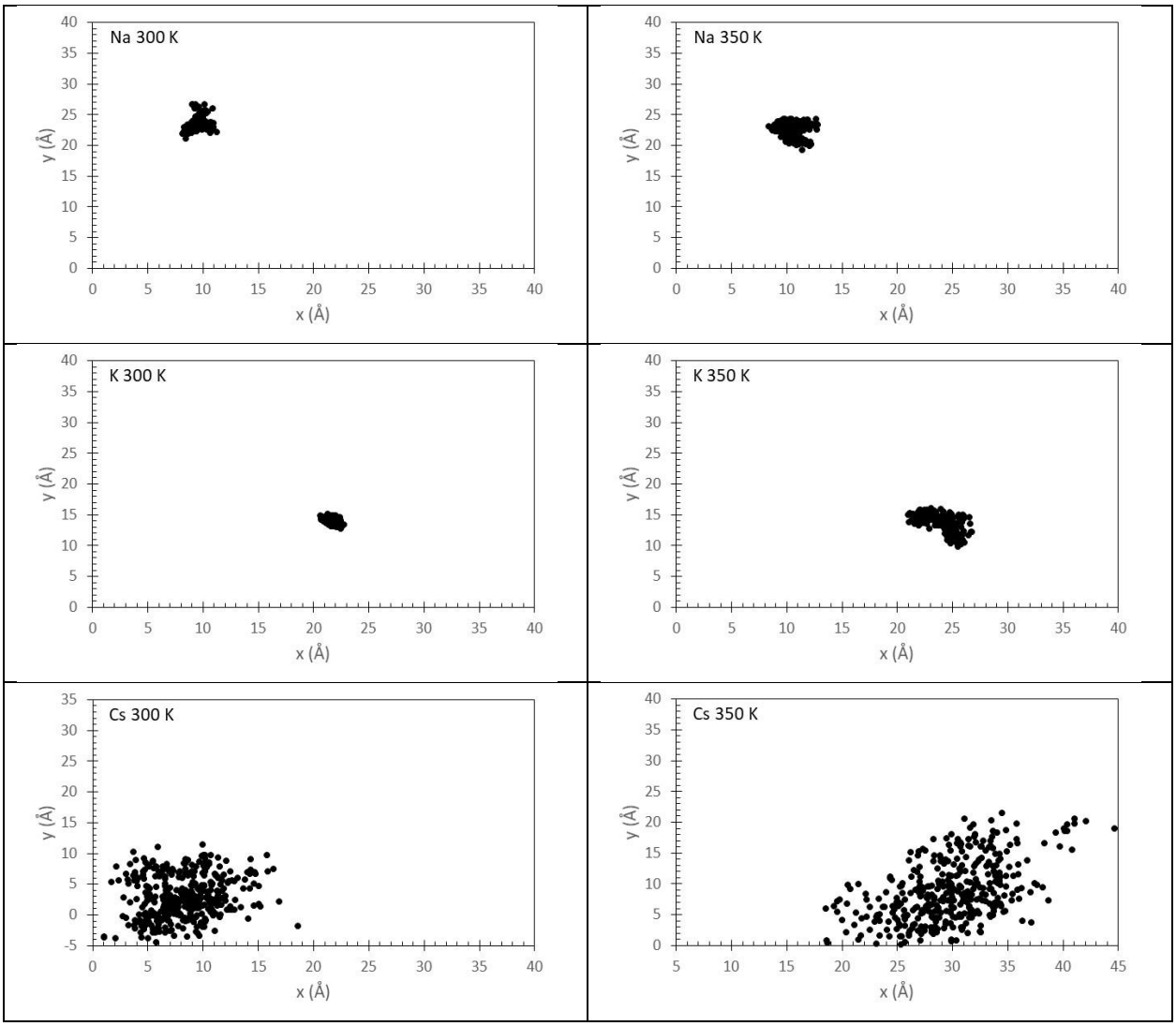


Figure S11. MD trajectories in the *xy* plane comparing the effect of temperature on AOT micelle mobility on the mica surface. Trajectories are shown from 32 AOT systems with monovalent counterions at 300 K and 350 K. Each point corresponds to the location of an AOT carbon atom every 0.05 ns during a 20-ns simulation.

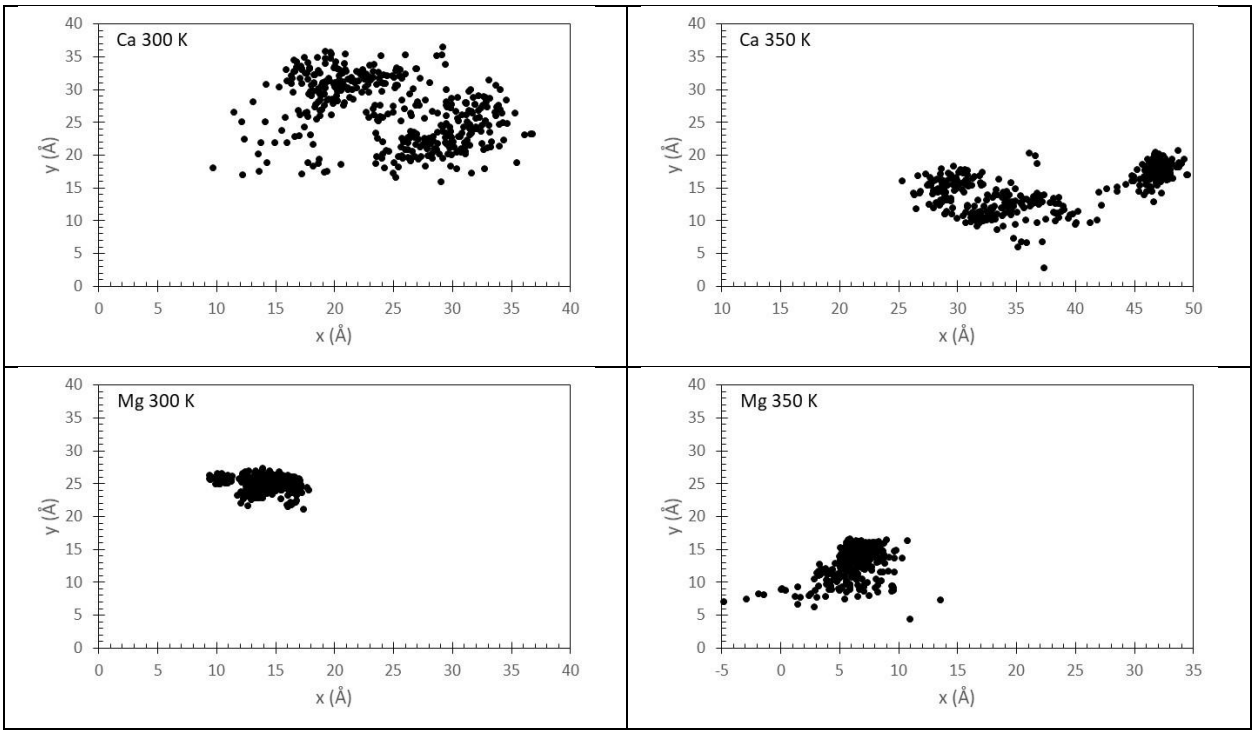


Figure S12. MD trajectories in the *xy* plane comparing the effect of temperature on AOT micelle mobility on the mica surface. Trajectories are shown from 32 AOT systems with divalent counterions at 300 K and 350 K. Each point corresponds to the location of an AOT carbon atom every 0.05 ns during a 20-ns simulation.

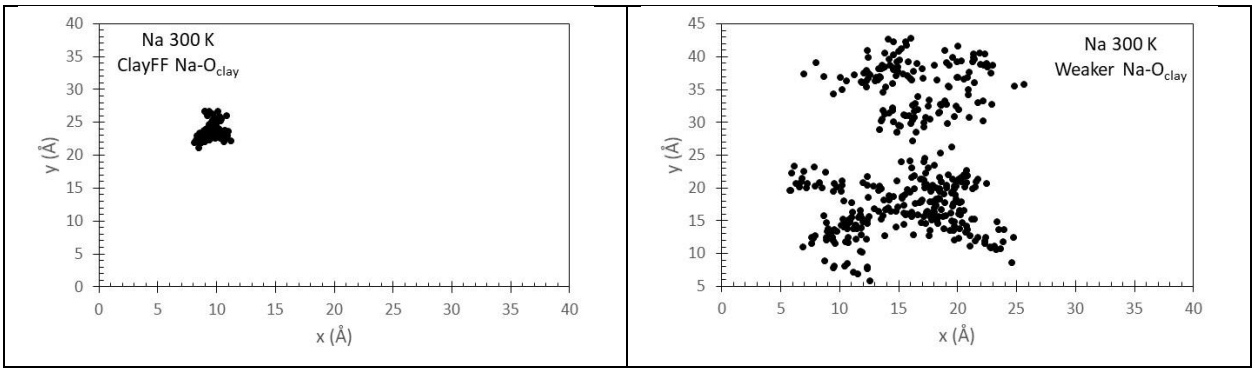


Figure S13. MD trajectories in the *xy* plane comparing the effect of cation-mica interaction potential on AOT micelle mobility on the mica surface. Trajectories are shown from 32 AOT systems with Na^+ counterions at 300 K. Each point corresponds to the location of an AOT carbon atom every 0.05 ns during a 20-ns simulation. Results are shown for Na^+ at 300 K with original (ClayFF) Na-O_{clay} Lennard-Jones parameters and Na^+ at 300 K with modified (weaker) Na-O_{clay} Lennard-Jones parameters.[1]

References

1. Ho, T. A.; Criscenti, L. J.; Greathouse, J. A., Revealing Transition States during the Hydration of Clay Minerals. *J. Phys. Chem. Lett.* **2019**, *10*, 3704-3709.