

Electronic Supplementary Material

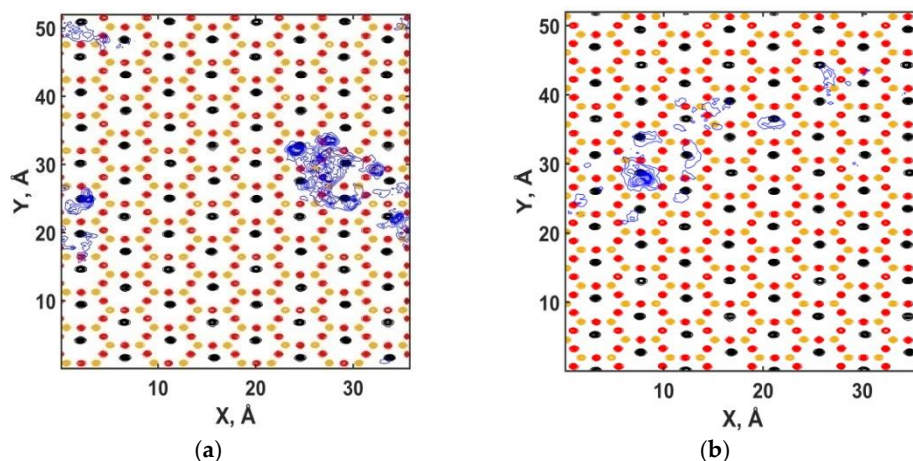


Figure S1. Time-averaged atomic density distributions of uranium at the basal surface of smectite. Red – bridging oxygen atoms, O_b ; yellow – tetrahedral Si; black – hydrogen atoms of structural hydroxyls, H_o ; blue – U. (a) ClayFF-orig; (b) ClayFF-MOH.

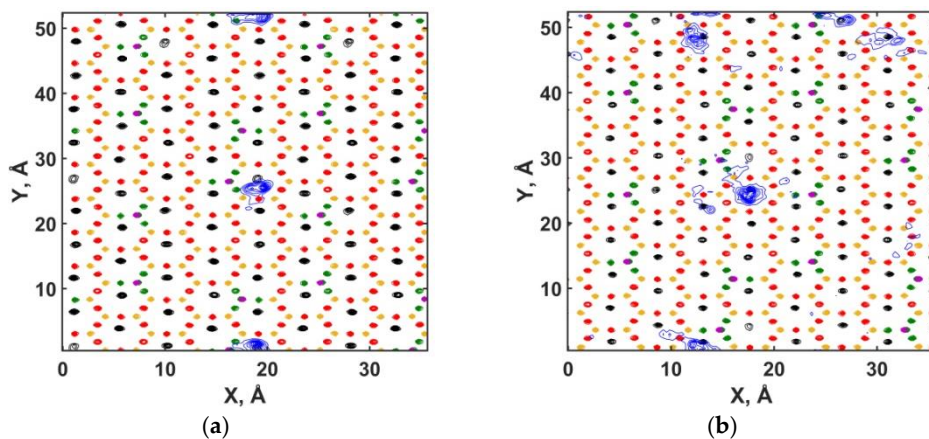


Figure S2. Time-averaged atomic density distributions of uranium on the basal surface of illite. Red – bridging oxygen atoms, O_b ; yellow – tetrahedral Si; black – hydrogen atoms of structural hydroxyls, H_o ; purple – Al of the tetrahedral substitutions; green – oxygen atoms next to the tetrahedral Al/Si substitutions, O_{bts} ; blue – U. (a) ClayFF-orig; (b) ClayFF-MOH.

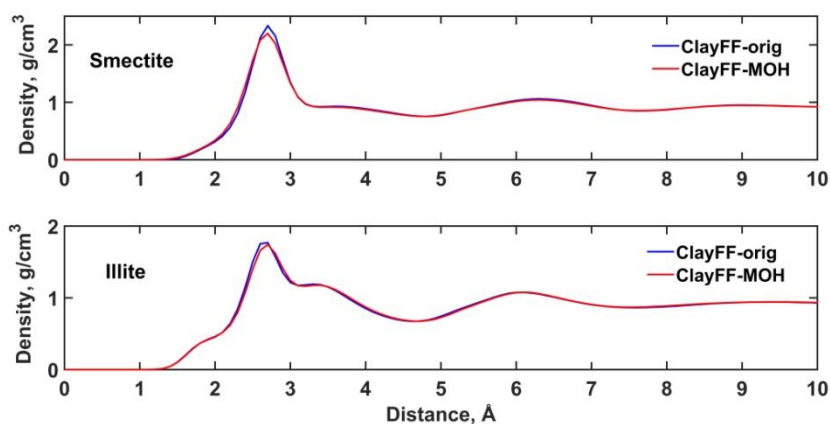


Figure S3. Atomic density profiles of O_w atoms of H_2O molecules for both versions of ClayFF force field as functions of their distance from the basal clay surface.

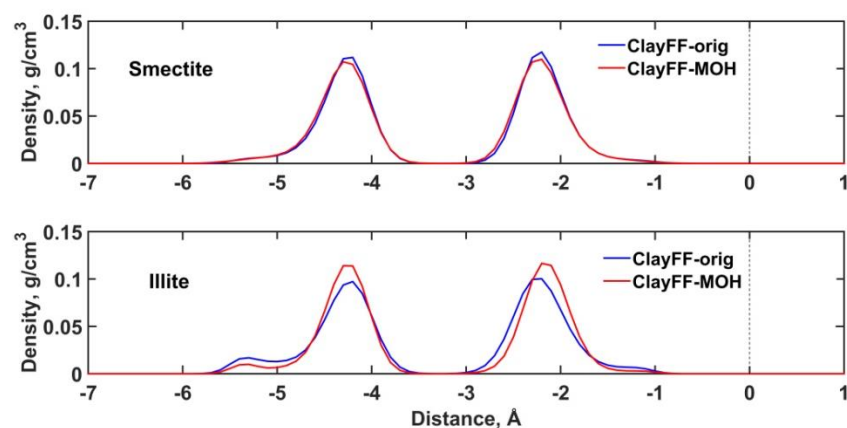


Figure S4. Atomic density profiles of Ho atoms as functions of their distance from the basal clay surface modeled with two versions of the ClayFF force field.

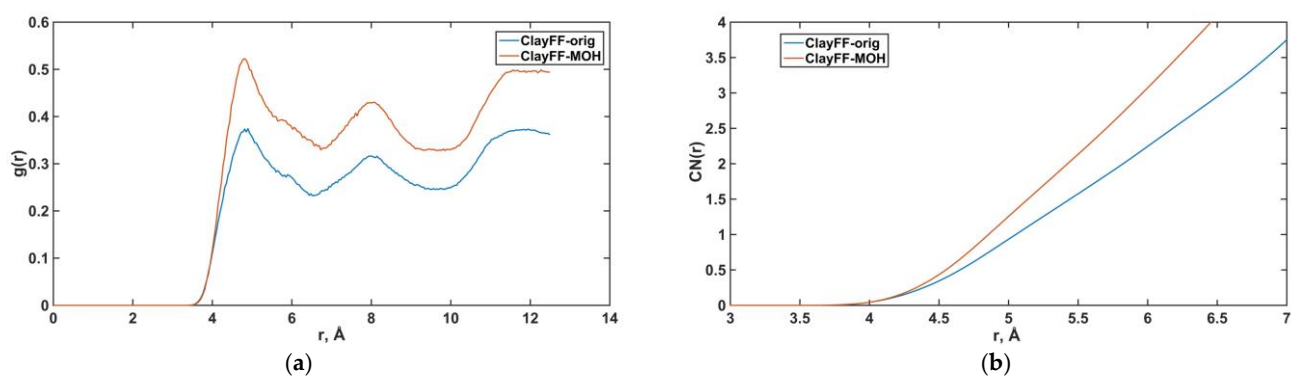


Figure S5. (a) Radial distribution functions for U-O_b pairs; (b) Running coordination numbers for U-O_b pairs (smectite model).

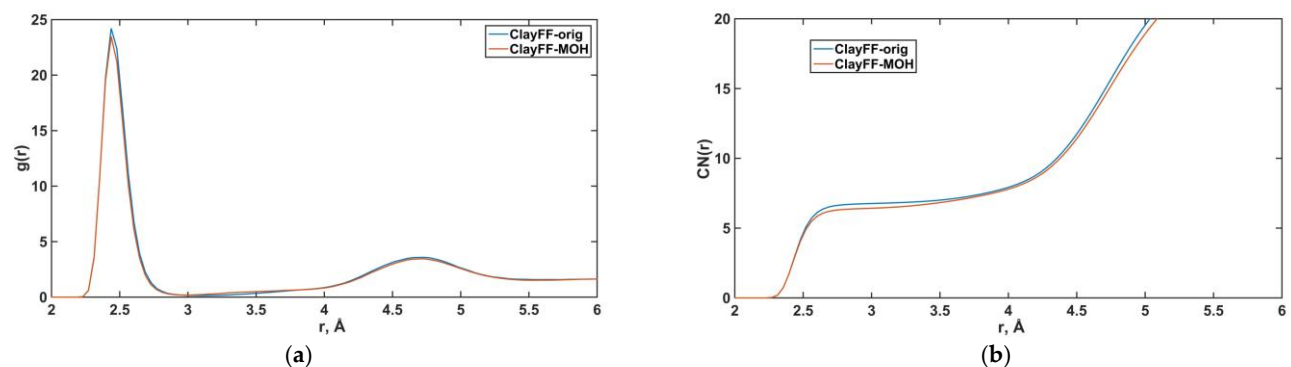
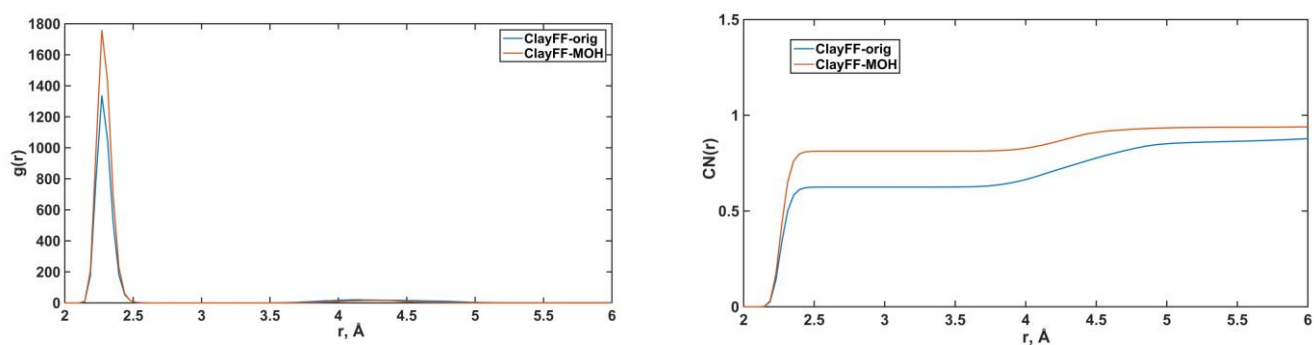


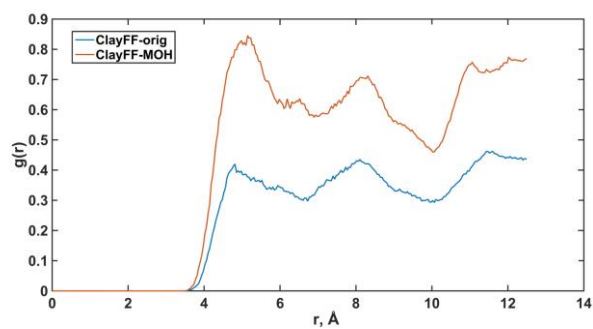
Figure S6. (a) Radial distribution functions for Ca-O_w pairs; (b) Running coordination numbers for Ca-O_w pairs (smectite model).



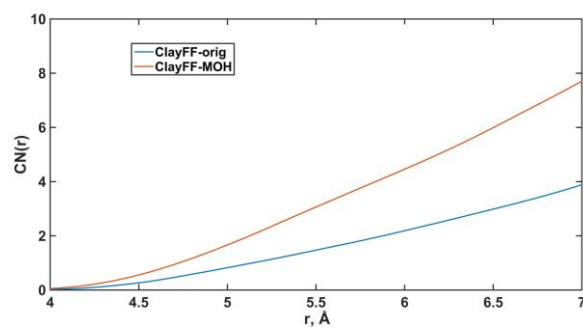
(a)

(b)

Figure S7. (a) Radial distribution functions for Ca-O_{hw} pairs; (b) Running coordination numbers for Ca-O_{hw} pairs (smectite model).



(a)



(b)

Figure S8. (a) Radial distribution functions for U-O_b pairs; (b) Running coordination numbers for U-O_b pairs (illite model).

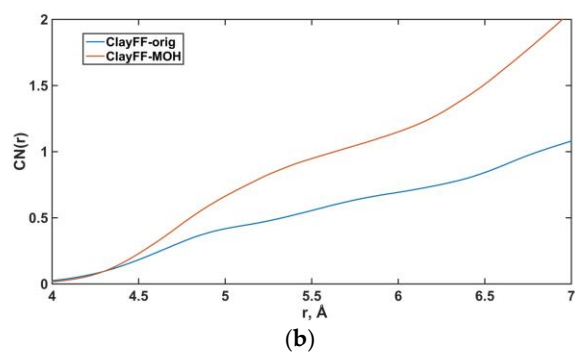
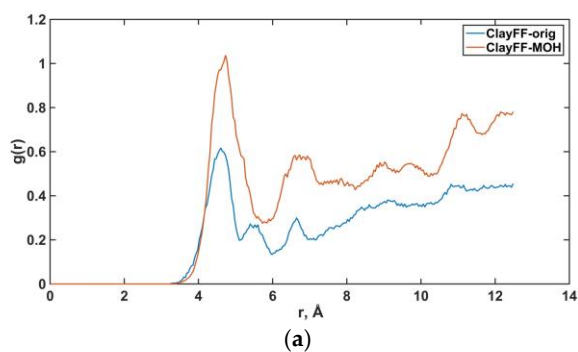


Figure S9. (a) Radial distribution functions for U-O_{bts} pairs; (b) Running coordination numbers for U-O_{bts} pairs (illite model). O_{bts} – Oxygen atom near tetrahedral substitution.

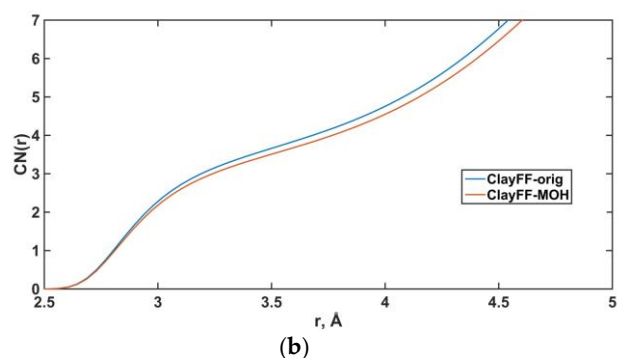
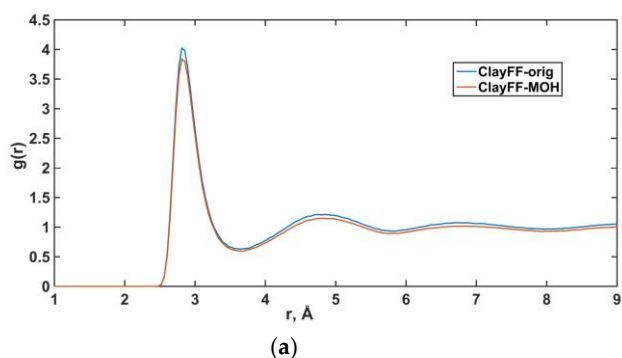


Figure S10. (a) Radial distribution functions for K-O_w pairs; (b) Running coordination numbers for K-O_w pairs (illite model).

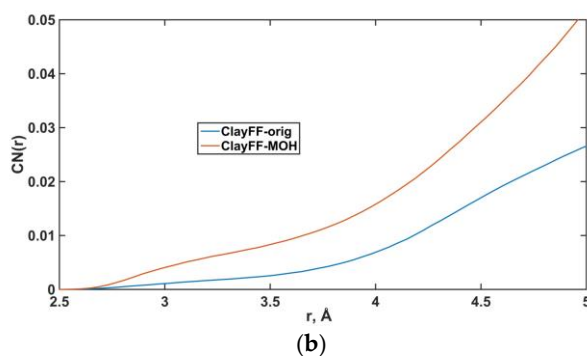
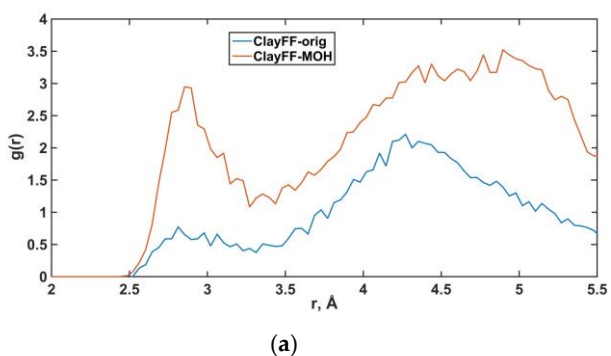


Figure S11. (a) Radial distribution functions for K-O_{hw} pairs; (b) Running coordination numbers for K-O_{hw} pairs (illite model).