Molecular dynamics simulations of cesium adsorption on illite

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Radioactive cesium isotopes ($^{134}\text{Cs}$, $^{137}\text{Cs}$) released during the Fukushima nuclear disaster on March 11, 2011 remain the main source of external radiation dose, five years after the nuclear disaster, in a region covering 3 to 8 % of the land area of Japan. Cesium has a well-known affinity for clay minerals and is likely to be primarily adsorbed on 2:1 clay minerals (illite, vermiculite) in surface soils near the disaster site. Experimental data indicate that illite, in particular, carries a small density of surface sites with a very strong affinity for cesium. These sites are widely thought to be located on the “frayed edges” of nanoparticles, but they are difficult to characterize at molecular scales. Here we discuss the methodological challenges associated with atomistic simulations of illite edge surfaces and present both large-scale molecular dynamics (MD) and thermodynamic integration calculations of cesium adsorption by illite nanoparticles contacting liquid water. New insights into the identities of different illite surface sites and their respective affinities for cesium are provided by our results.

Figure 1. Molecular dynamics (MD) simulation snapshot showing our model illite nanoparticle in liquid water. Blue spheres are adsorbed or interlayer K⁺ ions.