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Atomistic simulations of hydrogen adsorption and transport in hydrated Na- and Ca-montmorillonite: Effects of temperature, pressure, pore size and interlayer cations

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Hydrogen can be formed in the geological repository of radioactive waste as the most prevalent gas through anoxic corrosion of stainless steel waste containers ($3Fe+4H_2O=Fe_3O_4+4H_2(g)$) and water radiolysis reactions caused by alpha decay under radioactive waste storage conditions. The accumulation of hydrogen gas presents a potential risk of overpressure and fracture of the surrounding clay host rock within the repository. To prevent this, it is crucial that gases are either absorbed, diffused or react with the constituent host rocks.

At the same time, the problems of geological hydrogen storage are not significantly different from the aforementioned environment and conditions. Thus, a molecular-scale comprehension of H_2 adsorption and transport processes in clays is crucial not only to ensure safe geological disposal and storage of radioactive waste, but also to facilitate the development of the geological H_2 gas storage technologies.

We apply the techniques of classical molecular simulations to quantitatively investigate the adsorption and diffusional transport of H_2 gas in hydrated clayey materials and improve the fundamental comprehension of the physical and chemical processes governing the interactions between H_2, water, and clay minerals. Our study aims to improve the fundamental understanding of the molecular mechanisms underlying these processes. We performed Grand Canonical Monte Carlo (GCMC) simulations to investigate H_2/H_2O binary mixture adsorption in Na- and Ca-montmorillonite at temperatures of 25, 50, and 90°C and high pressure. Furthermore, molecular dynamics simulations of H_2 adsorption and transport in hydrated Na- and Ca-montmorillonite interlayers are conducted to investigate not only the structural properties of the nano-confined H_2/H_2O binary fluid but also its dynamic properties. The simulation results show that the adsorption of H_2 gas exhibits a proportional increase with increasing pressure, while a decrease is observed with increasing temperature in accordance with the solubility behavior of hydrogen gas in water. It has been noted that the mole fraction of adsorbed hydrogen in hydrated clay interlayers is higher in comparison to its solubility in bulk water.