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A comparative study on Li^+ adsorption by clay minerals with different structures

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Due to the rapid development of lithium-ion batteries in electric vehicles and energy storage systems, global lithium (Li) consumption is growing exponentially. Clay-type Li deposits are widely distributed, abundant, and inexpensive to mine, making them a significant potential source of Li resources. As the primary hosts for Li in clay-type Li deposits, the enrichment of Li are strongly controlled by the adsorption of Li^+ on clay minerals. However, the effect of clay mineral crystallographic features on Li^+ adsorption remains poorly understood. To address this gap, this study investigated the adsorption of Li^+ by four representative clay minerals: montmorillonite (Mnt), illite (Ilt), chlorite (Chl), and kaolinite (Kln). Through adsorption kinetics and thermodynamics experiments, the effects of pH, temperature, and ionic strength on Li^+ uptake were systematically examined. Additionally, desorption tests, ^7Li solid-state NMR, and FTIR spectroscopy were employed to identify adsorption mechanisms within different mineral structures. The results showed that the adsorption of Li^+ by the four clay minerals reached equilibrium within 48 hours. Mnt exhibited the highest Li^+ adsorption amounts, while Kln showed the lowest. Compared to the other three clay minerals, the adsorption of Li^+ by Mnt is less pH dependent. The desorption results showed that more than 70% of pre-adsorbed Li^+ could be desorbed from Mnt, while only a small portion of Li^+ pre-adsorbed in the other three minerals could be desorbed, suggesting that Mnt exhibited the weakest affinity to Li^+. Mnt adsorbs Li^+ primarily through outer-sphere complexation and cation exchange within its expandable interlayers, attributed to the diffuse negative charge generated by octahedral substitution. Li^+ may have entered the pseudo-hexagonal sites of Ilt due to the intense charge generated by tetrahedral substitution. Given the fixed interlayers of Chl and limited isomorphous substitution of Kln, inner-sphere complexation between Li^+ and surface hydroxyl groups is likely the main adsorption mechanism for these two minerals. These findings demonstrate that crystallographic features of clay minerals significantly influence Li^+ adsorption behavior, providing experimental insights into Li enrichment processes and offering implications for exploration and processing of clay-type Li resources.