# Mechanistic Study of $CO_2$ Adsorption and Desorption on $Li_4SiO_4$

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Lithium orthosilicate ($Li_4SiO_4$) is considered as one of the promising material for high temperature $CO_2$ capture. However, there is no clear understanding about the reaction mechanisms for adsorption and desorption processes, which include the formation of two solid phases (i.e. $Li_4SiO_4(s) + CO_2(g) \leftrightarrow Li_2SiO_3(s) + Li_2CO_3(s)$). In this study, the $CO_2$ adsorption and desorption mechanisms were investigated by density functional theory calculation. In the adsorption process, we explored the reaction pathways for the formation of thin bilayer structure consisting of $Li_2CO_3$ (above) and $Li_2SiO_3$ (below). After the thin bilayer formation, thermal diffusion of $Li^+$ and $O^{2-}$ enabled the further reaction to occur and made a double shell for complete adsorption. In the desorption process, we observed the successive formation of $Li_4SiO_4$ nuclei at the $Li_2CO_3$–$Li_2SiO_3$ interface, which was induced by the desorption of $CO_2$ from $Li_2CO_3$ layers. The detailed reaction energetics on each step were confirmed by Gibbs free energy calculation. These findings into underlying mechanisms for CO2 capture will provide a way to improve the catalytic performance for $Li_4SiO_4$–based sorbents at high temperature.