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## Article

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## A combined DFT and molecular dynamics study of U(VI)/calcite interaction in aqueous solution

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**Abstract** Here we present a combined DFT and molecular dynamics study of uranyl (U) interaction mechanisms with the calcite (104) surface in aqueous solution. The roles of three anion ligands ( $\text{CO}_3^{2-}$ ,  $\text{HCO}_3^-$ ,  $\text{OH}^-$ ) and solvation effect in U(VI) interaction with calcite have been evaluated. According to our calculations, water adsorbed on the calcite (104) surface prefers to exist in molecular state rather than dissociative state. Energy analysis indicate that the positively charged uranyl species prefers to form surface complexes on the surface, while neutral uranyl species may bind with the surface via both surface complexing and ion exchange reactions of  $\text{U(VI)} \rightarrow \text{Ca(II)}$ . In contrast, the negatively charged uranyl species prefer to interact with the surface via ion exchange reactions of  $\text{U(VI)} \rightarrow \text{Ca(II)}$ , and the one with  $\text{UO}_2(\text{CO}_3)_2(\text{H}_2\text{O})^{2-}$  as the reactant becomes the most favorable one in energy. We also

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