Surface Complexation Modeling of U(VI) Adsorption on Granite at Ambient/Elevated Temperature: Experimental and XPS study

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The Beishan granitic formation is under investigation as the host rock for a high-level radioactive waste repository in China. It is important to understand the retention processes, including influential parameters such as the metal ion concentration, pH, ionic strength (*I*) and temperature. The present study deals with U(VI) adsorption on Beishan granite using batch-type experiments in a CO_2-free atmosphere. U(VI) adsorption on granite is shown to be insensitive to ionic strength. Temperature has a positive effect on U(VI) adsorption indicating that it is endothermic. Combining the X-ray photoelectron spectroscopy (XPS) analysis and adsorption data at 25 °C, a Generalized Composite (GC) model with only three surface complexes, $\equiv SOUO_2^+$, $\equiv SO(UO_2)_2(OH)_2^+$ and $\equiv SO(UO_2)_3(OH)_5$, was constructed. The experimental data at 40 °C and 60 °C were fitted by the proposed model to obtain the equilibrium constants (*K*) of surface reactions at these two temperatures. The enthalpy changes (ΔH) of the surface reactions were evaluated from the *K* obtained at three temperatures via the van't Hoff equation. Finally, blind modeling predictions were performed to test the robustness of the proposed model and ΔH. Satisfactory agreement with the literature data confirmed this GC model with ΔH proving a useful tool to predict U(VI) adsorption on granite samples, especially on Beishan granite at ambient/high temperature