

Aqueous speciation and sorption properties of beryllium in carbonate-containing systems

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Beryllium bearing waste has been stored in the Aube Storage Center (CSA), the French surface facility dedicated for the disposal of low- and intermediate-level, short-lived radioactive waste. As for other chemo-toxic elements, the evaluation of Be release toward the biosphere is required for safety and environment impact assessment. This study aims a better characterization of Be migration in the sandy aquifers (Aptian sands) below the waste cells. Be(II) is characterized by a strong hydrolysis and the formation of sparingly soluble hydroxide phases [1]. In the presence of carbonate, Be(II) forms stable binary/ternary complexes Be(II)-OH-CO₃(aq), although the aqueous species prevailing in near-neutral to alkaline conditions remain ill-defined. Groundwater in Aptian sands is characterized by a near-neutral pH and carbonate concentrations ~ 1 mM. Be(II) uptake by Aptian sands has been quantified associated to the characterization of the aqueous speciation in carbonate-containing porewaters.

All experiments were conducted in Ar-gloveboxes at $T = (22 \pm 2)^\circ\text{C}$. Sorption experiments were performed with two samples of Aptian sands collected in the environment of CSA. Rietveld refinement of XRD data shows that the Aptian sands are rich in quartz (91–92%), with presence of microcline (3%), glauconite (4–2 %), kaolinite (<2%), muscovite (<2%) and trace amount of smectite clays. Synthetic groundwater was characterized by pH = 6.4, $C_{\text{tot}} = 1.2 \cdot 10^{-3}$ M and low ionic strength ($I = 1.8 \cdot 10^{-3}$ M). Sorption isotherms were constrained by the upper Be(II) concentrations defined by solubility and the detection limit of ICP-MS (~ $1 \cdot 10^{-10}$ M). Solubility samples were prepared using a well-defined $\alpha\text{-Be(OH)}_2(\text{cr})$ in aqueous solutions containing $1 \cdot 10^{-3} \text{ M} \leq C_{\text{tot}} \leq 0.4 \text{ M}$, with $C_{\text{tot}} = [\text{HCO}_3^-] + [\text{CO}_3^{2-}]$. Total Be(II) concentration was quantified by ICP-MS after ultracentrifugation or ultrafiltration.

Solubility experiments suggest that the equilibrium constant previously reported for $\text{Be(OH)}_2\text{CO}_3^{2-}$ might be largely overestimated, and that Be(II) hydrolysis species prevail in near-neutral to alkaline systems with $C_{\text{tot}} \leq 1 \cdot 10^{-3}$ M. Both Be(II) aqueous speciation and sorption data will be extensively discussed in this contribution.

[1] Cevirim-Papaioannou et al., 2020, Appl. Geochem., 98, 237-246