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Interfacial Speciation of Ln(III) on Albite: Insights from Batch Sorption and Luminescence Spectroscopy Experiments

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The internationally accepted strategy for high-level nuclear waste disposal is containment in deep geological repositories [1]. Reliable long-term safety assessments require robust models describing radionuclide (RN) migration and retention, supported by accurate thermodynamic data and molecular-level understanding of RN–mineral interactions [2,3].

Crystalline rocks, alongside clay and rock salt, are considered as possible host forms for deep geological repositories. Quartz, mica, and feldspar make up the majority of granitic rocks. Among these, albite ($NaAlSi_3O_8$) is a major plagioclase feldspar and an abundant framework aluminosilicate in the Earth's crust, providing reactive surface area in repository environments. Despite its geological relevance, the sorption behavior of RNs on albite surfaces remains insufficiently understood, limiting reliable thermodynamic descriptions of radionuclide retention in crystalline host rocks.[4]

This study investigates retention mechanisms of Eu(III), Nd(III), and Lu(III) on albite as analogues for minor actinides. Batch sorption experiments were conducted at pH 3-9 and ionic strengths of 0.15 M NaCl under atmospheric CO_2(g) condition. Sorption is low at acidic pH and increases continuously with pH, reaching maximum uptake at alkaline conditions for all Ln(III). At constant pH, sorption increases from light to heavy lanthanides, consistent with increasing ionic charge density along the series.

Eu(III) sorption shows negligible dependence on ionic strength (0.01–0.1 M NaCl), indicating dominant inner-sphere surface complexation.

Eu(III) speciation was further investigated by time-resolved laser-induced fluorescence spectroscopy (TRLFS). Changes in the emission spectra compared to the aqueous reference clearly indicate surface complex formation. Fluorescence lifetime analysis reveals that at higher pH values a single surface species dominates, whereas at lower pH two distinct species are observed.

The combined sorption and spectroscopic results provides mechanistic insight into the direct Ln(III) coordination at albite surface sites. The experimentally derived parameters offer a quantitative basis for refining surface complexation models and enhance the thermodynamic description of trivalent radionuclide migration in granitic repository systems.

References:

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