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Reactive transport modeling of radium transport and co-precipitation in fractured crystalline rocks

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Radium-226 (Ra) as a decay product of the uranium-238 present in deep geological disposal facilities for spent nuclear fuels can be one of the main dose contributors in the later stages of repository evolution. Its retention is primarily expected through co-precipitation with Ba sulfate, with Ra and Ba coming from the waste reacting with sulphate-rich pore waters in the repository host rocks to precipitate Ba-Ra-SO_4 solid solutions. While the thermodynamic properties of these solid solutions are well established by experiments and modeling, a realistic understanding of Ra migration and retention requires consideration of coupled hydrological and chemical processes in the repository host rocks. Fractured crystalline rocks are considered as potential host formations for deep geological repositories, e.g. in Finland, Sweden and Canada. However, a detailed understanding of Ra migration and co-precipitation in these rocks remains challenging due to their structural heterogeneity and complex groundwater chemistry.

Here, 3D reactive transport modeling with PFLOTRAN on high-performance supercomputers was conducted to simulate Ra migration in fractured crystalline rocks over long timescales. The models account for variations in fracture network characteristics and fluid composition to evaluate how their interplay affects the transport and co-precipitation of Ra. The results show that connected fracture networks act as permeable flow paths channeling Ra migration in the crystalline rocks. The spatial distribution of Ra co-precipitation is primarily controlled by the hydraulic properties of fractures, the number of connected fractures, location and number of dead-end fractures, and initial sulfate concentrations. Higher fracture density, permeability and connectivity, and fewer dead-end fractures enhance Ra transport due to more available high-permeability pathways for efficient advective transport of Ra. In contrast, lower fracture density, permeability, and connectivity, and more dead-end fractures tend to facilitate co-precipitation of Ra and retard Ra migration. Over a total of 10,000-year simulations within a 10-m domain, Ra precipitation fronts initially propagate through the primary flow paths of connected fractures, then extend into secondary flow paths and dead-end fractures, and eventually diffuse into the rock matrix. These observations suggest that Ra co-precipitation in the matrix and dead-end fractures is the primary mechanism for Ra retention in fractured crystalline rocks.