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Sorption of Nickel on Calcite based on Surface Chemistry Studies

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Calcite is a common mineral in the environment, comprising approximately 4% of the earth's crust. The surface reactions of calcite play an important role in many geological and environmental systems, including the production of oil and gas, the geological storage of nuclear waste and CO₂ etc. For example, the sorption of Ni-63 on calcite affects significantly its mobility and removal from aqueous phases. Ni-63 is one of the most important radionuclides that will determine the radiation risk of spent nuclear fuel for the first several hundred years of the final disposal. A clear understanding of the interactions between Ni-63 and calcite surfaces is vital for the safety assessment of a spent nuclear fuel repository.

However, despite numerous studies that have been performed, the fundamental parameters of calcite surface charge in aqueous solutions, especially at conditions relevant to natural systems, remain poorly understood. The calcite surface composition and charge are linked to its reactivity and sorption properties, and thus, they are the primary data source to understand the sorption properties of Ni-63 on calcite surfaces. In this work, the surface properties of Bukov calcite are studied by a combination of experimental and basic-stern surface complexation modelling methods. The surface properties and cross-sections of crushed calcite samples are characterized experimentally by cation exchange capacity (CEC), B.E.T. (Kr) for specific surface area (SSA), scanning electron microscope (SEM) and zeta potential measurements (ZP). The structural and surface characterization measurements will answer several uncertainties associated with understanding calcite surface structures and dissolutions, including the dissolution kinetics, surface charges, the contribution of grain boundaries, potential determining ions etc. The next step in the work is to develop a basic Stern surface complexation model together with our colleagues in Gesellschaft für Anlagen- und Reaktorsicherheit (GRS), Germany, to provide data for the surface species and the Stern layer thickness.

With the understanding of the surface properties, three sets of sorption experiments are performed. The titration experiments are carried out in a batch-wise manner to measure the calcite surface site capacities and intrinsic surface protonation and deprotonation constants. A backtitration approach is performed to compensate for the effect of dissolution. Sorption edge experiments are carried out by measuring the sorption of 10⁻⁹ M Ni-63 on calcite from pH 7 to 11. Sorption isotherm experiments are carried out at pH around 8 with Ni concentrations from 10⁻⁹ M and 10⁻³ M.

A multi-site surface complexation model of Ni-63 sorption on Bukov calcite is used to provide explanation and prediction of Ni-63 sorption behaviour under various conditions. The model is based on the above experimental data (titration data, sorption edge data and sorption isotherm data) and molecular DFT modelling which will provide basic model parameters like surface site types and surface site densities.

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