

Uptake of Pb^{II} by CaCO₃: the influence of polymorphism on the process

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Lead is a dangerous contaminant; its ability to induce disorders in living organisms makes lead pollution a serious issue for human health. Among the remediation techniques aiming at Pb^{II} uptake, carbonate minerals represent a promising option especially due to their abundance and low price.

Calcium carbonate is the most abundant carbonate mineral and it mainly forms two polymorphic phases: calcite and aragonite (CAL and ARG). CAL and ARG have very similar solubility products at normal temperature-pressure conditions. This allows deriving mechanistic information on the crystallographic control exerted by the substrate on the Pb-uptake process via a comparative study.

The interaction of Pb²⁺ ions with CaCO₃ was investigated with batch recrystallization experiments and using in situ AFM (Atomic Force Microscope) observation of surface reactivity in flow-through experimental setup. The first set of experiments allowed to measure different Pb-uptake kinetics for CAL and ARG. After the interaction period, the solid phase was filtered and the solution was analyzed by ICP-OES to determine the amount of Pb²⁺ removed and Ca²⁺ released. The solid phase was characterized with x-ray diffraction (XRD) and electron microscopy (SEM). The AFM experiments allowed observing the effect of Pb²⁺ ions on the dissolution of calcite. The experimental constraints of AFM fluid cell lead to the formation of a secondary Pb-bearing phase (hydrocerussite, HCER). The growth of HCER was observed in situ during the replacement process. Finally, geochemical modelling with PHREEQC was used to explain the experimental results and the thermodynamic properties of the Pb-Ca-CO₂-H₂O system.