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Mechanism of surface reactions and dissolution of fluorite surface in an aqueous electrolyte solution

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Fluorite, a mineral form of calcium fluoride (CaF₂), is semi-soluble in water, which makes it an interesting material for simultaneous studying both interfacial properties and kinetics of dissolution. Although the dissolution process of fluorite has been studied by different experimental techniques, the mechanisms and especially the pH-dependence of the dissolution are not entirely clarified. Motivated by this fact, the dissolution of the fluorite (111) crystallographic plane and fluorite colloidal particles was studied as a function of pH. The process was examined by measuring the concentration of released calcium and fluoride ions by ion-selective electrodes. Additionally, the electrokinetic and the inner surface potentials were measured by means of electrophoresis and fluorite single crystal electrode, respectively. These complementary methods indicated that the mechanism of fluorite dissolution includes a series of elementary steps: the reaction of surface groups with H⁺ ions, the formation of F⁻ vacancies, the dissociation of surface groups, the release of calcium and fluoride ions into the interfacial region as well as the diffusion of ions from the interfacial region to the bulk of solution. The proposed reaction mechanism indicates that the H⁺ ions perform an important role in the fluorite dissolution, a concept not possible to confirm by looking at the overall equation of fluorite dissolution. The order of the reaction with respect to H⁺ ions was found to be 0.37 which is in good accordance with the value derived from the reaction mechanism. The experimentally determined rate coefficient of fluorite dissolution was found to be $k_{dis} = 9 \times 10^{-6}$ mol^{2/3} dm m⁻² s⁻¹.