Alunite and basaluminite dissolution: Comparison and insights from batch experiments and atomistic computer simulations

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Alunite (KAl₃(SO₄)₂(OH)₆) and basaluminite (Al₄(SO₄)(OH)₁₀·4-5H₂O) are two of the main minerals controlling the mobility of aluminium in natural systems and especially in mining environments and soils under acidic conditions. Despite their importance, the dissolution rates and mechanisms of alunite and basaluminite under typical conditions in such environments are still unknown. In this work, the dissolution of both aluminium phases under acidic conditions (pH between 3 and 4.8) and at temperatures similar to the ones commonly found in surface environments (between 6 and 40°C) was studied by means of batch stirred dissolution experiments. In all the cases, the solutions were prepared by adding H₂SO₄ to the desired pH value.

The obtained results show that both alunite and basaluminite tend to dissolve incongruently under the target conditions. Released aluminium concentrations, compared with K and SO₄ for alunite and with SO₄ for basalumite, are clearly lower than the ones corresponding to the mineral stoichiometries. This can be partly attributed to the precipitation of Al-secondary phases (e.g., diaspore; AlO(OH), gibbsite; Al(OH)₃).

Dissolution rates within the studied range are between 10⁻¹⁰ and 10⁻¹¹ mol·m⁻²·s⁻¹ for alunite and between 10⁻⁸ and 10⁻⁹ mol·m⁻²·s⁻¹ for basaluminite. This is consistent with the much lower crystallinity of basaluminite. The dissolution rates for both minerals seem to increase with both the increase of pH and temperature within the studied range. However, the dependence of the alunite dissolution rate from both parameters seems to be much weaker than in the case of basaluminite.

Atomistic computer simulations for alunite show that Al is rarely exposed at the most stable solvated surfaces, which suggests that the detachment of aluminium is probably rate-limiting. The observed increase of dissolution rates for both minerals with pH decrease can be attributed to the parallel increasing formation of AlSO₄⁺ dissolved complexes, which would help aluminium to be detached from the mineral structure.