A numerical simulation of dissolution and precipitation of uraninite during infiltration alteration of granite

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Equilibrium processes of dissolution and precipitation of unaninite during infiltration alteration of granite have been numerically simulated by MIX99 (Hoshino et al., 2000). Initial compositions of inlet fluids are assumed as the same as an average composition of river water with pH = 6 and log fO2 = -1.1. A model granite is composed of quarts (60 mole %), K-feldspar (25 %), plagioclase (albite (9.1 %) + anorthite (3.9 %)), biotite (annite (1.4 %) + phlogopyte (0.6 %)) and trace amounts of uraninite with or without pyrite. Plagioclase and biotite are assumed as ideal solid solutions of albite and anorthite and of annite and phlogopite, respectively.

The simulation results show that a dissolution front of plagioclase moves towards the downstream side quite rapidly, followed by slow dissolution fronts of biotite and then K-feldspar. Quartz precipitates at the dissolution fronts of plagioclase and K-feldspar. Alteration minerals appear in the analyses are muscovite, kaolinite and ferrihydrite. Smectite may be formed when ferrihydrite is inactive.

It should be noted that ferrihydrite does not equilibrate with pyrite under analyzed conditions. Practically impossible amounts of dissolved ferrous species are required for the equilibrium of ferrihydrite and pyrite due to very low oxygen fugacities at pyrite dissolution fronts. Ferrihydrite precipitates where dissolved ferrous species yielded by annite dissolution are oxidized by infiltrating fluids, resulting in the formation of the redox front of the most upstream side. Hereafter, the front will be called as the first redox front. Pyrite dissolves at a downstream side of the first front and forms a second redox front. When the dissolution front of pyrite moves faster than that of plagioclase due to its small initial content, pyrite dissolves quite rapidly because of high pH (above 11) of the fluids equilibrated with plagioclase.

Uraninite dissolves at the first redox front or at the downstream side adjacent to the front. It reprecipitates at an analytical slab being the next (downstream side) to the dissolution front and does also at the second redox front. Secondary enrichments of uraninite occur at the both precipitation areas.

It should be emphasized that a possibility of uranium enrichment without any preexistent reductant such as pyrite has been shown by the present numerical simulations for the equilibrium alteration processes of granite. The uraninite dissolution occurs at or nearby the first redox front immediately followed by its precipitation at the downstream side even when pyrite is absent initially. An amount of the enrichment increases with a development of the first redox front when the initial content of uraninite is large, while it is restrained at a certain amount when the initial one is small.

The simulation results have also shown that the dissolution and reprecipitation of uraninite are strongly controlled by the dissolution of annite and the following oxidation of dissolved ion species resulting in the precipitation of ferrihydrite. Smectite may act for ferrihydrite when the latter is inactive due to its slow reaction rate.