Reaction of U(VI) with iron sulfides

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In sulfidic environments, iron oxides undergo reductive dissolution by S(-II) leading to the formation of FeS. Iron oxides are important sorbents for U(VI) and their reductive dissolution may lead to the mobilization of adsorbed U(VI). However, mobilization of uranium might be counteracted by reduction of U(VI) by FeS [1]. In previous experiments, reduction of U(VI) to U(IV) was observed when S(-II) solution was added to iron oxide suspensions with adsorbed U(VI). Uranium reduction coincided with FeS formation and in this study we aimed at establishing the role of FeS as reductant for U(VI) in these systems.

For this purpose, U(VI) was added to FeS suspensions, which were either produced by combining Fe(II) and S(-II) containing solutions, or by complete reduction of the iron oxide lepidocrocite by S(-II). The solids from these experiments were collected and analyzed by X-ray absorption spectroscopy.

Sorption of U(VI) to the solids and the extent of U(VI) reduction varied considerably between the different FeS substrates and depended on experimental conditions. When U(VI) was added to FeS suspensions, which were formed by reducing lepidocrocite and aged for several weeks, very little U(VI) adsorption and no reduction of U(VI) was observed during two weeks incubation at pH 8. On the contrary, U(VI) was complete reduced within a time scale of hours when added to freshly precipitated FeS suspensions and reacted at pH 6.

The data indicate that pronounced U(VI) reduction occurs in experiments in which the FeS phases are only metastabile. That is, U(VI) reduction might predominately take place during FeS precipitation or FeS dissolution. This implies that not FeS, the endproduct of iron oxide reduction by S(-II), but an imtermediate, which forms during the reductive dissolution of iron oxides, may be the most effective reductant for U(VI).

[1] Wersin.*et al.* (1994) *GCA* **58(13)**, 2829-43.

The systematics of ^{36}Cl production rate calculations in limestone and dolomite

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One of the major radionuclides that are measured by Accelerator Mass Spectrometry (AMS) is chlorine-36 ($T_{1/2}$ = 301kyr). It is produced in very low concentrations (^{36}Cl/Cl~1E-12 - 1E-15) by the cosmic rays in atmosphere and upper meters of lithosphere, mainly in limestone and dolomite.

There are several production pathways of ^{36}Cl in limestone and dolomite: spallation of Ca by fast neutrons, capture of slow negative muons on ^{40}Ca, fast-muon induced reactions on Ca, capture of thermal and epithermal neutrons on ^{35}Cl, where these neutrons are also produced by several pathways. The complexity of ^{36}Cl production makes it difficult to calibrate each separate pathway and ultimately to use this radionuclide in the exposure dating. Here we summarize our investigation in the ^{36}Cl production systematics and draw an outline for the recommended calibration constants and calculation procedure.