Effects of microbial activity and electron shuttles on the reduction of U(VI) under sulfidogenic conditions

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Introduction

Recent studies suggest that electron shuttles such as low molecular mass quinones and humic substances may play a role in many redox reactions involved in contaminant transformations and the biogeochemical cycling of redox active elements. This study investigates the effects of 9, 10-anthraquinone-2, 6-disulfonate (AQDS), a synthetic electron shuttle often used as a surrogate for quinone moieties in humic substances, on transformations of Fe, S, and U under reducing conditions.

Experimental Methodology

Experiments were conducted in defined mineral medium containing 30 mM Fe (III), 5 mM sulfate, and 10 mM acetate, with and without 100 μM AQDS and inoculated with sediment from the Rifle, CO, USA, Integrated Field Research Challenge (IFRC) Site. After the system reached steady state with respect to Fe (III) and sulfate reduction, aliquots of suspension were collected from each system and one set was pasteurized at 70 °C for 1 hr. The suspensions were then spiked with 500 µM U (VI).

Discussion of Results

After 48 h, 100% of the added U was removed from solution in the non-pasteurized AQDS system. However, only 58%, 25%, and 11% of added U was removed in the no AQDS non-pasteurized, AQDS pasteurized, and no AQDS pasteurized systems, respectively. U XANES analysis of the hydrated solids indicated that, with the exception of the pasteurized system without AQDS, the majority (85-95%) of the U associated with the solids was reduced to U (IV). The results of the EXAFS analysis of U (IV) in the systems with and without AQDS (not pasteurized) are consistent with the formation of nanoparticulate uraninite. The results of this study suggest that microbial reduction was the dominant process contributing to the reduction of U (VI) over the timescale of this experiment and that the presence of AQDS enhanced both biotic and abtiotic/microbially mediated U (VI) reduction.

Age and origin of the Nuvvuagittuq Greenstone Belt

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The Nuvvuagittuq belt is dominated by mafic and ultramafic rocks metamorphosed to at least upper amphibolite facies. Primary U-rich minerals that might provide reliable dates for rock formation have yet to be found in the dominant lithology called the Ujaraaluk unit. Metamorphic zircons, rutiles and monazites are, however, present in the unit locally and give variably discordant results with $^{207}Pb/^{206}Pb$ ages ranging from 2.8 Ga to 2.5 Ga. The younger ages overlap 2686±4 Ma zircon ages for intruding pegmatites and Sm-Nd ages for garnet formation in the Ujaraaluk rocks suggesting this era as the time of peak metamorphism and metasomatism in the Nuvvuagittuq belt, coeval with regional metamorphism of the Superior craton. ^{147}Sm-^{143}Nd data for Ujaraaluk whole rocks provide a statistically poor isochron of 3814±300 Ma, but when separated by compositional groups, this 'isochron' is seen to consist of a series of ~2.7 Ga slopes emanating from a baseline distribution older than 4 Ga. Metamorphism at 2.7 Ga will have less effect on the ^{146}Sm-^{142}Nd chronometer because of ^{146}Sm extinction prior to ~4 Ga. Expansion of the ^{142}Nd dataset for the Ujaraaluk rocks and associated ultramafic cumulates continues to show a good correlation between Sm/Nd and $^{142}Nd/^{144}Nd$ that corresponds to an age of 4.359^{+45}_{-67} Ga. The dataset now includes samples with superchondritic Sm/Nd ratios that extend the correlation to values of $^{142}Nd/^{144}Nd$ slightly higher than the terrestrial standard with a total range in $\varepsilon^{142}Nd$ of more than 24 ppm. The upper Sm/Nd end of this correlation is defined by rocks that are interpreted as cumulates to compositionally related extrusive rocks indicating that this crystal fractionation had to occur while ^{146}Sm decay was active, i.e. well before 4 Ga. Intruding gabbros give ^{143}Nd and ^{142}Nd isochron ages overlapping within error at 4.16 Ga also supporting an Hadean age for the belt. Eoarchean tonalites surrounding the Nuvvuagittuq belt show a deficit in ^{142}Nd compared to the terrestrial standard, but plot to the low Sm/Nd side of the Ujaraaluk isochron suggesting that they are remelts of this type of mafic basement. The Nuvvuagittuq belt thus preserves over 1.6 billion years of early Earth history including an expanse of mafic crust formed only ~200Ma after Earth formation.