Stability of U(IV) in fresh and aged bioreduced sediments

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Bioreduction of hexavalent uranium [U(VI)] has been intensively investigated and proposed as a remediation strategy following uranium contamination in the subsurface. Initially, bioreduced uranium was believed to be solely uraninite [1], but later, other less crystalline species were documented [2]. It has also been shown that non-crystalline tetravalent U [U(IV)] species (NCU4) are the dominant products independently from the geochemical conditions and reductive pathway [3]. Because NCU4 appears to be more sensitive to reoxidation and remobilization to the aqueous phase [4], the use of bioreduction for remediation is now debated.

This study investigates the stability of freshly bioreduced U(IV) under geochemical conditions that are relevant to the case of Rifle site in Colorado (USA). Furthermore, we document the potential for ageing processes to transform NCU4 to more crystalline phases and how this may affect the rate of NCU4 oxidation.

For these purposes, uranium was reduced under both iron-reducing and sulfate-reducing conditions in continuous flow-through columns packed with Rifle aquifer background sediments. Artificial groundwater amended with uranium and multiple electron donors was the influent. The bioreduction phase was maintained for ~ 350 days until a significant amount of U was immobilized in the sediments. Prior to the re-oxidation phase, bioreduced sediments were characterized via X-ray fluorescence, X-ray absorption spectroscopy (XAS), and electron microscopy. The sediments harbouring sufficient amount of U(IV) were incubated in batch reactors under anoxic conditions and left to age for a minimum of 4 months to a maximum of 20 months. Fresh and aged samples were characterized with XAS to probe for evidence of mineralogical changes. In order to assess the stability of U(IV) species, bioreduced sediments were exposed to oxygen in flow-through reactors under various geochemical conditions. Preliminary results indicated that, in fresh sediments, the non-crystalline fraction of bioreduced U is most sensitive to oxidation.

[1] Lovley .D.R. and Philips E.J. (1992), AEM vol. 58 no. 3 850-856.
[2] R. Bernier-Latmani et al. (2010), ES&T 44 (24) 9456-9462.
[3] Stylo et al. (2013), ES&T 47 (21) 12351-12358.
[4] Cerrato et al. (2013), ES&T 47(17) 9756-9763.