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Microbial reduction of a U(V) complex with an organic ligand

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The molecular mechanism of hexavalent uranium (U(VI)) reduction to tetravalent (U(IV)) species remains partly unresolved for metal-reducing bacteria such as *Shewanella oneidensis* MR-1. All evidence points to the first step being the one-electron reduction of U(VI) to U(V)[1,2]. We confirm that finding by capturing the U(V) intermediate using the unusual properties of the aminocarboxylate ligand, dpaea[3], which forms a stable aqueous complex with U(V) at pH 7. Dpaea provides the opportunity to directly investigate the fate of the U(V) species. Previous work has attributed U(IV) formation to the disproportionation of 2 U(V) molecules to U(VI) and U(IV) when carbonate was the ligand[1,2]. However, the elucidation of this step requires more direct experimental evidence for other ligands. Here, we investigate the fate of U(V)-dpaea in the presence of *S. oneidensis* MR-1 by combining ion exchange chromatography, UV-vis spectrometry, and U M4-edge High Resolution X-ray Absorption Near Edge Structure spectroscopy. On the one hand, we probed the ability of wild-type and mutant strains of *S. oneidensis* MR-1 to interact with U(V)-dpaea. These mutant strains lacked part or all of their *c*-type cytochromes, proteins key to electron tranfer. On the other hand, we reacted U(V)-dpaea with purified outer-membrane *c*-type cytochrome MtrC, a terminal reductase. The combination of these two systems allowed us (i) to rule out disproportionation as a major mechanism for U(V)-dpaea transformation to U(IV), and (ii) to deduce that U(V)-dpaea is biologically reduced to U(IV) via an active electron transfer mediated by *c*-type cytochromes. Altogether, these results suggest that U(V) could be biologically reduced when bound to a stabilizing organic ligand.