



# The biogeochemistry and bioremediation of uranium and other priority radionuclides



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## ABSTRACT

Microbial metabolism has the potential to alter the solubility of a broad range of priority radionuclides, including uranium, other actinides and fission products. Of notable interest has been the biostimulation of anaerobic microbial communities to remove redox-sensitive radionuclides such as uranium U(VI) from contaminated groundwaters at nuclear sites. Particularly promising are bioreduction processes, whereby bacteria enzymatically reduce aqueous U(VI) to insoluble U(IV) coupled to oxidation of an organic electron donor; and uranium phosphate biomineralisation, in which bacterial phosphatase activity cleaves organophosphates, liberating inorganic phosphate that precipitates with aqueous U(VI) as uranyl phosphate minerals. Here we review the mechanisms of uranium bioreduction and phosphate biomineralisation and their suitability to facilitate long-term precipitation of uranium from groundwater, with particular focus on in situ trials at the US Department of Energy field sites. Redox interactions of other priority radionuclides (technetium, neptunium, plutonium, americium, iodine, strontium and caesium) are also reviewed.

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## Contents

1. Introduction . . . . .	165
2. Microbe–radionuclide interactions . . . . .	165
2.1. Bioreduction . . . . .	166
2.2. Biomineralisation . . . . .	167
2.3. Bioaccumulation . . . . .	167
2.4. Biosorption . . . . .	167
3. Uranium bioreduction . . . . .	167
3.1. Early work & mechanisms . . . . .	167
3.1.1. Cytochromes . . . . .	168
3.1.2. Nanowires . . . . .	168
3.1.3. Extracellular electron carriers . . . . .	168
3.2. Mineralogical endpoints of bioreduction . . . . .	168
3.3. Field studies . . . . .	169
3.3.1. US DOE Rifle site, Colorado . . . . .	170
3.3.2. US DOE Oak Ridge site, Tennessee . . . . .	172
3.3.3. US DOE Hanford site, Washington . . . . .	173
3.4. Stability of bioreduced U(IV) and reoxidation . . . . .	173
3.4.1. Reoxidation by exposure to oxygen . . . . .	173
3.4.2. Reoxidation by exposure to nitrate . . . . .	174
4. Uranium phosphate biomineralisation . . . . .	174
4.1. Early work & mechanisms . . . . .	174
4.2. Mineralogical endpoints . . . . .	175

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4.3.	Demonstration at the US DOE Oak Ridge site, Tennessee	175
4.4.	Limitations	176
5.	Other priority radionuclides	176
5.1.	Technetium	176
5.1.1.	Early work & mechanisms	176
5.1.2.	Reduction and reoxidation studies	176
5.2.	Neptunium	177
5.3.	Plutonium	177
5.4.	Americium	177
5.5.	Iodine	178
5.6.	Strontium and caesium	178
6.	Conclusions and future directions	178
	Acknowledgements	178
	References	179

## 1. Introduction

Decades of nuclear activities have left a legacy of environmental contamination. Elevated concentrations of uranium and other radionuclides are present in mining and milling areas, at sites where uranium ore was processed, and where uranium was enriched. This contamination potentially represents an uncontrolled source of radiation, and therefore regulatory bodies may require it to be remediated to acceptable levels.

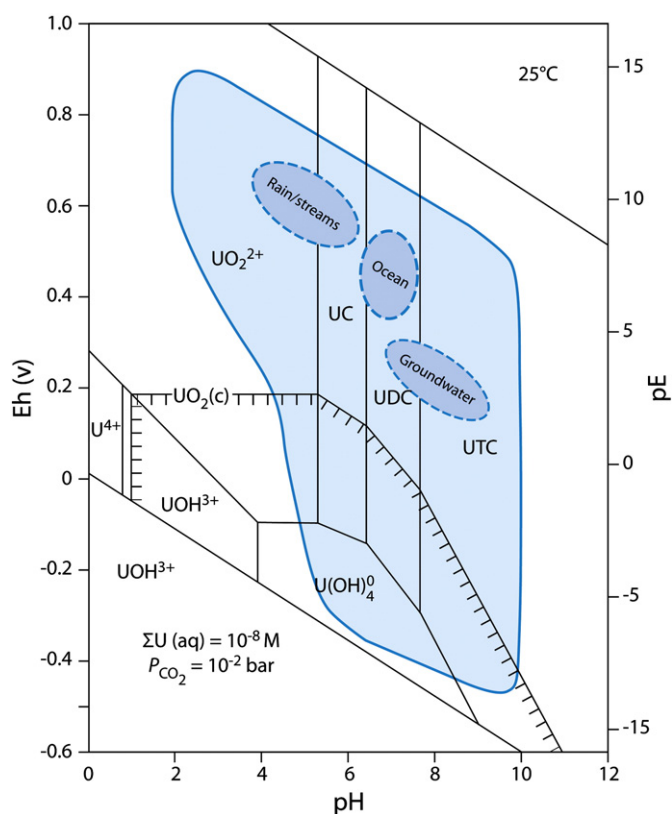
The mobility of uranium in the environment depends on its speciation and redox state (Fig. 1). It is present as mobile U(VI) in oxidising conditions, predominantly as the uranyl ion ( $\text{UO}_2^{2+}$ ) or hydroxyl complexes below  $\sim\text{pH}$  6.5, or as uranyl carbonate complexes at higher pH

(Choppin et al., 2002). In the absence of carbonate, the uranyl ion and its complexes sorb strongly onto the surface of iron oxides and organics (Hsi and Langmuir, 1985; Andersson et al., 2001; Koch-Steindl and Pröhl, 2001) and onto the edge sites of clay minerals (Pabalan et al., 1998; Davis et al., 2004). Sorption decreases in the presence of complexing ligands such as humic and fulvic acids, and in the presence of competing cations such as  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  (Siegal and Bryan, 2003). Under reducing conditions, relatively insoluble and immobile U(IV) predominates, typically as the mineral uraninite, but recently a different, non-uraninite U(IV) phase has been reported (Bernier-Latmani et al., 2010; Bargar et al., 2013). U(V) is generally considered to be transient although evidence is emerging to suggest that it might be stable for periods of weeks under certain conditions (Docrat et al., 1999; Behrends et al., 2012; Ilton et al., 2012). Depleted uranium metal, used in anti-tank penetrators and present in the environment as a legacy of military activities, is relatively immobile, but may be oxidised to uraninite and consequently U(VI) depending on the redox conditions (UNEP, 2003; Parrish et al., 2008; Handley-Sidhu et al., 2010).

Biogeochemical interactions play a key role in controlling the speciation and mobility of uranium and other redox sensitive radionuclides (such as Tc, Np and Pu), through direct metabolic processes such as microbial respiration, or indirectly by changing ambient redox/pH conditions, producing ligands or new biominerals, or altering mineral surfaces. In addition to controlling radionuclide mobility via “natural attenuation”, these biogeochemical processes can be stimulated to accelerate clean-up of contaminated environments through bioremediation. This review focuses on microbe–radionuclide interactions, how they may control radionuclide (especially uranium) mobility in natural environments, and how they can be applied to bioremediate legacy metal and radionuclide contamination.

## 2. Microbe–radionuclide interactions

Microbial interactions with uranium and other radionuclides have been documented extensively e.g. Gorby and Lovley (1992), Lloyd and Gadd (2011), Lloyd and Macaskie (2000), Lloyd and Renshaw (2005), Lovley et al. (1991) and Merroun and Selenska-Pobell (2008). Most study the interactions between uranium and bacteria; these are the focus of this review. There is emerging interest in the use of microbial “bioleaching” mechanisms to extract uranium from low grade ores e.g. Choi et al. (2005) and Qiu et al. (2011), although these are not covered further in this review. Preventing uncontrolled dispersion and transport of radionuclides in groundwater is the overarching remediation goal at many nuclear sites. Stimulating bacterial interactions to fix aqueous uranium into insoluble minerals in situ may provide a relatively inexpensive and non-intrusive solution to remediating radionuclide contamination. The mechanisms of the different microbe–uranium interactions are illustrated in Fig. 2 and discussed below, along with their suitability to facilitate long-term uranium removal.



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