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Aspects of environmental actinide chemistry



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- 1985 Diploma Sc., University of Saarbrücken, Germany
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- 1993-2003 Staff scientist at the Institute for Nuclear Disposal (INE) at the Karlsruhe Research Centre (FZK)
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Abstract

Since their application in various technologies, notably in nuclear energy, actinide elements have gained significant interest by environmental scientists (e.g. [1, 2]). Besides uranium and thorium, also transuranium elements, notably plutonium, can be found in the natural environment. While the naturogenic plutonium isotopes ^{244}Pu and ^{239}Pu are only abundant at ultra-trace levels in the earth crust, antropogenic releases between the 1950s and the 1970s via nuclear weapon tests and from military processing installations into the environment amount to more than 20 tons [3]. In addition, quite a number of countries using nuclear energy have decided to dispose spent nuclear fuel in a geological repository without separating uranium and plutonium. Understanding environmental actinide behaviour is certainly of high relevance not only to appraise actinide propagation in the biosphere but also to assess their behaviour in a deep geological repository over

long time scales.

It is well known, that plutonium exists in its tetravalent oxidation state in many environmental compartments and thus is considered of low mobility. This is consistent with observations made for plutonium already abundant in the environment, where e.g. plutonium deposited as weapon test fallout can be found located in narrow regions of sediment layers [3, 4]. However, formation of colloidal species, complexation with organic ligands, redox transitions have been reported to enhance the mobility of the early actinide series elements under specific conditions (e.g. [5, 6]). Respective reactions are often characterized by strong kinetic hindrances.

Frequently, lanthanides, uranium or thorium are taken as surrogates for americium, neptunium and plutonium. Such analogies, though being adequate in some cases, may be misleading in other cases. Ionic radii vary and thus speciation in solution can strongly differ. The capability of forming e.g. covalent bondings significantly varies between 4f and 5f elements and within the 5f element series (see e.g. [7, 8]). Consequently, thermodynamic data and speciation schemes of chemical homologues are not always applicable, but have to be determined element specific.

Our knowledge on actinide environmental chemistry has certainly improved tremendously over the years. There is, however, still space for research on a group of radioelements with exciting properties.

[1] A.Yu. Romanchuk et al., 2016, Russ. Chem. Rev., 85, 995

[2] A. B. Kersting, 2013, Inorg. Chem. 52, 3533

[3] H. Geckeis, M. Zavarin, B. Salbu, O. Lind, L. Skipperud, 2018, Environmental chemistry of plutonium in P. A. Stevens, D.L. Clark, Plutonium Handbook, *in print*

[4] Saito-Kokubu, et al. 2008, Journal of Environmental Radioactivity, 99, 211

[5] A.B. Kersting et al., 1999, Nature, 397, 56

[6] A.P. Novikov et al., 2006, Science, 314, 638

[7] M.L. Neidig et al., 2013, Coord. Chem. Rev., 257, 394

[8] T. Vitova et al., 2017, Nature Comm., 8, 16053

Selected Publications

1. Baumann, A; Yalcintas, E; Gaona, X.; Polly, R; Dardenne, K; Prussmann, T; Rothe, J; Altmaier, M; Geckeis, H. *DALTON TRANSACTIONS*. **2018**, 47, 4377.
2. Bahl, S; Peuge, S; Pidchenko, I; Pruessmann, T; Rothe, J; Dardenne, K; Delrieu, J; Fellhauer, D; Jegou, C; Geckeis, H; Vitova, T. *INORGANIC CHEMISTRY*. **2018**, 56, 13982.
3. Polly, R; Heberling, F; Schirmelpfennig, B; Geckeis, H. *JOURNAL OF PHYSICAL CHEMISTRY C*. 2017, 121, 20217.
4. Vitova, T; Pidchenko, I; Fellhauer, D; Bagus, PS; Joly, Y; Pruessmann, T; Bahl, S; Gonzalez-Robles, E; Rothe, J; Altmaier, M; Denecke, MA; Geckeis, H. *NATURE COMMUNICATIONS*. **2017**, 8, 16053.
5. Geckeis H., Lützenkirchen J., Polly R., Rabung T., Stumpf T. Mineral-water interface reactions of actinides, *CHEMICAL REVIEWS*, **2013**, 113, 1016.

Honors and Awards

1. Since 2014 member of the Nuclear Waste Management Commission (ESK) which advises the Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Safety (BMUB) in matters of nuclear waste management.

2. Since 2013, Chairman of the German association for repository research (DAEF)
3. Chairman of the International Steering Committee of the MIGRATION conferences (International Conference on the Chemistry and Migration Behaviour of Actinides and Fission products in the Geosphere)
4. Until 2014, Chairman of the Nuclear chemistry division of the German Chemical Society (GDCH).
5. Advisory Board Member of Radiochimica Acta