An expanding view on plutonium oxide nanoparticles

E. Gerber[1,2,3], A. Yu. Romanchuk[3], I. Pidchenko[1,2], L. Amidani[1,2], A. Rossberg[1,2], C. Hennig[1,2], G. Vaughan[4], A. Trigub[5], T. Egorova[3], S. Bauters[1,2], T. Plakhova[3], M O.J.Y. Hunault[6], S. Weiss[2], S.M. Butorin[7], A.C. Scheinost[1,2], S.N. Kalmykov[3,5] and K.O. Kvashnina[1,2]

[1]Rossendorf Beamline at ESRF – The European Synchrotron, CS40220, 38043 Grenoble Cedex 9, France, [2]Helmholtz Zentrum Dresden-Rossendorf (HZDR), Institute of Resource Ecology, PO Box 510119, 01314, Dresden, [3]Lomonosov Moscow State University, Department of Chemistry, 119991 Moscow, Russia, [4]ESRF - The European Synchrotron, CS40220, 38043 Grenoble Cedex 9, France, [5]National Research Centre "Kurchatov Institute", 123182 Moscow, Russia, [6]Synchrotron SOLEIL, L'Orme des Merisiers, Saint Aubin BP 48, 91192 Gif-sur-Yvette, France, [7]Molecular and Condensed Matter Physics, Department of Physics and Astronomy, Uppsala University, P.O. Box 516, Uppsala, Sweden, **evgeny.gerber@esrf.fr**

The nanoscience field often produces results more mystifying than any other discipline. It has been argued that changes of the plutonium dioxide (PuO_2) particle size from bulk to nano can have a drastic effect on PuO_2 properties [1-2]. Here we report a full characterization of PuO_2 nanoparticles (NPs) at the atomic level and probe their local and electronic structure. The particles were synthesized from precursors with different oxidation states (Pu III, IV and V) under various environmentally and waste storage relevant conditions (pH 8 and pH 12). Synthesized PuO_2 nanoparticles were studied by complementary methods: X-ray diffraction (XRD), atomic pair distribution function analysis (PDF), high energy resolution fluorescence detection (HERFD) X-ray absorption spectroscopy at the Pu L_3 and M_4-edges [3] and extended X-ray absorption fine structure (EXAFS) spectroscopy. This approach - the application of multifold synchrotron methods - is crucial to confirm results, obtained with individual methods. By combinations of these techniques it was found that small (2 nm) nanoparticles are formed from the Pu(III), Pu(IV), Pu(V) aqueous solutions at pH 8 and 12, with the crystal structure close to PuO_2, without any other Pu-O contributions or oxidation states of Pu except Pu(IV) [4].

References

[1] - A.P. Novikov, S.N. Kalmykov, S. Utsunomiya, R.C. Ewing, F. Horreard, A. Merkulov, S.B. Clark, V.V. Tkachev, B.F. Myasoedov, Science **314**, 638 (2006).

[2] - A. Yu. Romanchuk, T.V. Plakhova, A.V. Egorov, T. B. Egorova, P.V. Dorovatovskii, Ya.V. Zubavichus, A.A. Shiryaev, S.N. Kalmykov, Dalton Trans. **47**, 11239 (2018).

[3] - K.O. Kvashnina, A. Romanchuk, I. Pidchenko, L. Amidani, E. Gerber, A. Trigub, A. Rossberg, S. Weiss, K. Popa, O. Walter, R. Caciuffo, A. Scheinost, S. Butorin, S. Kalmykov, Angewandte Chimie, doi.org/10.1002/anie.201911637 (2019).

[4] - E. Gerber A. Romanchuk, I. Pidchenko, L. Amidani, A. Rossberg, C. Hennig, G. Vaughan, A. Trigub, T. Egorova, S. Bauters, T. Plakhova, M. Hunault, S. Weiss, S. Butorin, A. Scheinost, S. Kalmykov, K. Kvashnina, submitted, (2019).