

URANYL CARBONATE SPECIATION BY TIME RESOLVED LASER INDUCED FLUORESCENCE SPECTROSCOPY

O.Preedy, Loughborough University

O.Preedy@lboro.ac.uk

C.Buck¹, D.R.Worall¹, D.Read¹ and N.D.M.Evans¹

[1] Loughborough university

Time Resolved Laser Fluorescence Spectroscopy (TRLFS) is a very useful technique for differentiating the chemical species of a fluorescent metal ion through analysis of characteristic excitation and emission spectra and decay (relaxation) lifetimes. The principal advantage over other spectroscopic techniques is the ability to determine in-situ metal speciation at environmentally relevant concentrations in solution and on ultra-thin surfaces [1].

Due to the spectral and temporal data being orthogonal to each other it is possible to differentiate the metal species by two discrete variables allowing speciation to be determined, which is difficult by any other laboratory based method [2].

The technique is becoming a widely used method for fundamental actinide & lanthanide studies. TRLFS is unique in being able to determine in-situ metal speciation at picomolar concentrations, which is essential when dealing with incipient corrosion of speciality metals or the alteration of ceramic and other materials used in the nuclear fuel cycle[3], [4].

In this study the aqueous speciation of uranyl carbonate species is investigated at both near neutral and hyper alkaline pH. The addition of carbonate ligands has been known to quench the uranyl fluorescence, however it has been observed that at molar ratios less than 0.5 an additive effect has been observed.

[1] T. Reitz, A. Rossberg, A. Barkleit, R. Steudtner, S. Selenska-Pobell, and M. L. Merroun, "Spectroscopic study on uranyl carboxylate complexes formed at the surface layer of *Sulfolobus acidocaldarius*," Dalton Trans., vol. 44, no. 6, pp. 2684–92, Feb. 2015.

[2] C. Moulin and P. Decambox, "Uranium Speciation in Solution by Time-Resolved Laser-Induced Fluorescence," no. 28, pp. 348–353, 1995.

[3] G. Geipel, T. Reich, V. Brendler, G. Bernhard, and H. Nitsche, "Laser and X-ray spectroscopic studies of uranium-calcite interface phenomena," vol. 248, pp. 408–411, 1997.

[4] a Kowal-Fouchard, R. Drot, E. Simoni, and J. J. Ehrhardt, "Use of spectroscopic techniques for uranium(VI)/montmorillonite interaction modeling.," Environ. Sci. Technol., vol. 38, no. 5, pp. 1399–407, Mar. 2004.