**Development and Quantification of UV-Visible and Laser Spectroscopic Techniques for
Materials Accountability and Process Control**

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Final Report (Sep 2008-Sep 2010)

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Principal Investigator: Ken Czerwinski, University of Nevada, Las Vegas
Phil Weck, Frederic Poineau, University of Nevada, Las Vegas

Graduate Students: Nick Smith, Jamie Warburton, Troy Robinson, Guy Dutech
Collaborators: Samuel Bryan, PNN, Robert Jubin, ORNL, Monica Regalbuto, ANL

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1. Summary

This project was continued as a NEUP funded effort that began in September 2009. Research in the final no-cost extension year of this project was performed coincidentally. The results demonstrated the utility of optical spectroscopy for determining the concentration of U and Cm under process conditions. The methods were also demonstrated to be applicable to assessing nitrate concentration in the uranium extraction aqueous phase. Efforts were extended to apply on-line methods for the analysis of dry reprocessing conditions.

The project supported student research. Nick Smith was awarded a Ph.D. in September 2010 from research supported by this project. A student lead publication resulted from this effort “Warburton, Jamie, Smith, Nick and Czerwinski, Ken(2010) 'Method for Online Process Monitoring for Use in Solvent Extraction and Actinide Separations', Separation Science and Technology, 45: 12, 1763 — 1768”. A number of student presentations were given at DOE working group meetings, American Chemical Society Conferences, Plutonium Futures 2009, and presentations at ANL.

2. Project Objectives

Ultraviolet-Visible Spectroscopy (UV-Visible) and Time Resolved Laser Fluorescence Spectroscopy (TRLFS) optical techniques can permit on-line, real-time analysis of the actinide elements in a solvent extraction process. UV-Visible and TRLFS techniques have been used for measuring the speciation and concentration of the actinides under laboratory conditions. These methods are easily adaptable to multiple sampling geometries, such as dip probes, fiber-optic sample cells, and flow-through cell geometries. To fully exploit these techniques for GNEP applications, the fundamental speciation of the target actinides and the resulting influence on

spectroscopic properties must be determined. Through this effort detection limits, process conditions, and speciation of key actinide components can be establish and utilized in a range of areas of interest to GNEP, especially in areas related to materials accountability and process control.

In this project UV-Visible and TRLFS will be evaluated for use in proposed GNEP solvent extraction based separations (Figure 1). Research in the UNLV radiochemistry laboratory has previously examined the efficacy of UV-Visible spectroscopy to examine uranium speciation under UREX conditions. The success of the uranyl nitrate UV-Visible spectroscopy study supports the utility and expansion of spectroscopic methods for the evaluation of actinide concentrations and solution conditions for other aspects of the UREX+ solvent extraction scheme. Studies in this project will examine the influence of Pu absorbance on the previously examined uranyl nitrate spectroscopy and the application of Cm laser fluorecence to TRUEX and TALSPEAK analysis. The Pu studies will examine conditions from previous uranyl experiments, UREX extraction conditions. Peak ratios will be determined as a function of nitrate concentration to assess Pu speciation and solution conditions. The studies will also include any impact Pu has on the uranyl system as well as the impact of uranyl on Pu spectroscopy. The use of TRLFS to examine Cm concentration and speciation under extraction conditions applicable to TRUEX and TALSPEAK will be investigated. The basis for this technique has been investigated in the UNLV laboratory, providing fundamental evidence for the utility of the application. Through this research effort, tools and spectroscopic techniques will be developed to evaluate and determine solution extraction conditions and concentrations of Pu and Cm in extraction processes, addressing areas on process control and materials accountability.

Figure 1. Schematic of potential UREX based separation

The UNLV program is actively engaged with Dr. Sam Bryan of PNNL on spectroscopic studies of extraction systems. The UNLV program has previously interacted with Dr. Robert Jubin through projects on UV-Visible spectroscopy of uranium and Tc separations related to CETE. It is expected these collaborations will continue with this project, with UNLV providing

support for both student interactions at the DOE facilities and DOE researchers at UNLV. It is expected that the developed spectroscopic techniques will be examined in detail at UNLV. Through the collaborators the application of the developed techniques to DOE facilities, such as CETE, will be accomplished. This project will also train the next generation of radiochemistry researchers necessary for the long-term success of GNEP and other DOE projects. The research team will include graduate students in the UNLV radiochemistry Ph.D. program. This project will expose the students to DOE researchers and facilities, providing a basis for introducing future researchers to DOE needs and scientific opportunities.

2.1. Project Tasks

The success of the uranyl nitrate UV-Visible spectroscopy study supports the utility and expansion of spectroscopic methods for the evaluation of actinide concentrations and solution conditions for other aspects of the UREX+ solvent extraction scheme. Studies in this project will examine the influence of Pu absorbance on the previously examined uranyl nitrate spectroscopy and the application of Cm laser fluorescence to TRUEX and TALSPEAK analysis. The collaborative partners will focus on different aspects of the project. As a graduate student from the UNLV program is currently working with Dr. Bryan at PNNL, experimental studies will include these efforts within the project. The results and spectroscopic conditions will be communicated to Dr. Jubin to evaluate its applicability for inclusion in CETE. Furthermore, spectroscopic data sharing between CETE and UNLV is expected to continue within this project. This work will be conducted in accordance with the requirements specified in the GNEP Quality Assurance Program Document.

2.1.1. Task 1: UV-Visible spectroscopy of Pu under UREX conditions

The UV-Visible absorbance spectroscopy of Pu will be examined under this task. The Pu studies will examine conditions from previous uranyl experiments, UREX extraction conditions. Peak ratios will be determined as a function of nitrate concentration to assess Pu speciation and solution conditions. The studies will also include any impact Pu has on the uranyl system as well as the impact of uranyl on Pu spectroscopy. The expected molar absorptivities for Pu(IV) are around 55 L mol⁻¹cm⁻¹ at 470 nm and 35 L mol⁻¹cm⁻¹ at 650 nm, requiring concentrations in the mM range. The UNLV radiochemistry program has the capability to perform such experiments with Pu. The techniques and methodologies employed in this task will be based on those demonstrated in the UNLV laboratory.

2.1.2. Task 2: Cm TRLFS studies under TRUEX and TALSPEAK conditions

The use of TRLFS to examine Cm concentration and speciation under extraction conditions applicable to TRUEX and TALSPEAK will be investigated in this task. The basis for this technique has already been demonstrated in the UNLV laboratory. The Cm spectroscopy will be examined in both the aqueous and organic phase. A range of nitric acid aqueous phases, based on the output from CCD-PEG, FPEX, and NPEX, will be assessed. The organic phase will be based on the TRUEX and TALSPEAK schemes. Curium excitation will use both 434 nm and 375 nm, with fluorescence monitored near 595 nm. As only a single fluorescence peak is expected, peak ratios will not be determined. Efforts in this task will focus on measuring the exact fluorescence wavelength, intensity, and lifetime against solution conditions. As with the previous studies on the U system with UV-Visible spectroscopy, it is expected that the Cm concentration and solution condition can be determined by the task data.

2.2. Schedule and Resources

The facilities and resources of the UNLV radiochemistry program will be available for this project. Currently experiments with macroscopic and microscopic quantities of radionuclides are routinely performed in UNLV radiochemistry. The spectroscopic methods and radionuclides to be used in this project are established at the UNLV radiochemistry program.

The project schedule is:

- Task 1: First data on the Pu nitrate system (by month 3)
- Task 1: UV-Visible studies on UREX based system with U and Pu (by month 10)
- Task 1: Evaluation of Pu and U peak ratio correlation (by month 12)
- Task 2: Initial Cm spectroscopy studies on TRUEX and TALSPEAK system (by month 6)
- Task 2: Use of Cm fluorescence wavelength and lifetime data in evaluating TRUEX and TALSPEAK conditions (by month 12)
- Delivery of results to Dr. Robert Jubin for initial CETE development of applications (by month 12)

3. Work Performed in 1st Quarter (October –December 2008)

Research efforts in the 1st quarter focused on UV-Visible spectroscopy, Task 1. Efforts in Task 2 examined fundamental chemistry of HDEHP in the organic phase as a basis for TALSPEAK studies.

Preliminary calibration studies with Hellma flow cells (1 cm & 1 mm) have begun using the peristaltic pump and DI water. The peristaltic pump will be calibrated first for its settings as the units are unknown and must be experimentally determined. Next, flow cell calibrations will be conducted to determine maximum flow rates for each path length (without bubbling, excess internal pressure). Calibrations will be reproduced. After calibrations are complete, uranyl concentrations will be stepped through in the following increments: 0.01, 0.126, 0.25, 0.5, 0.63, 0.75, 1.0, 1.26 M [U]. Each uranyl concentration will be examined at nitric acid increments: 0.01, 0.03, 0.1, 0.5, 1, 2, 3, 4, 5, 6 M [HNO₃].

Primary method development for thermodynamic titrations of the uranyl-nitrate-acetate system was finalized. Initial experiments suggest that the method is viable and will produce defendable, repeatable results. The method utilizes a program developed by Peter Gans (Hyperquad, UK) for IUPAC called GLEE or GLass Electrode Evaluation to calibrate the electrode at variable ionic strengths.

The use of the Liquid Waveguide Capillary Cell (LWCC) was proposed for studying plutonium visible spectroscopy with 100x less plutonium than what would be required with a conventional cell. The goal was to compare the uranium Peak Ratios Algorithm results with a standard cuvette with the LWCC, then move on to plutonium. With an effective pathlength of ~100 cm, most species will have some absorbance. Nitric acid shows a marked absorbance below 450 nm, in the sensitive range of the uranyl ion. The uranyl spectra need to be corrected for this contribution. A series of spectra are included which illustrate the corrections needed. Finally, the peak ratio analysis was applied to the LWCC data. Inside of the calibrated range (1-5 M HNO₃), the LWCC data matches to within 4% of the 1-cm data. This shows that the LWCC can be used to probe the spectroscopy of uranyl and plutonium species at much lower levels.

Studies were performed on HDEHP chemistry as part of the efforts in Task 2 on TALSPEAK chemistry. Studies examined the details on the HDEHP organic phase, including volume change, water extraction and vapor pressure.

4. Work Performed in 2nd Quarter (January-March 2009)

In the 2nd quarter efforts focused on Task 1. The evaluation of uranyl nitrate complexation constants continued with the determination of the first nitrate complexation constant. This result agreed with the limited literature data and established the utility of the titration method to evaluate the desired data. The determined thermodynamic data will be used to correlate speciation with spectroscopic results. The damaged laser system for use in Task 2 was returned to UNLV. Upon setup the laser scorched several mirrors and optical components. The entire system was sent back to OPOtek for testing and realignment. Continuous spectroscopic analysis switched from dip probes to flow through cuvettes. Data provided on the uranyl ion at differing metal concentrations, cuvette pathlength, and nitrate concentrations in flow-through cuvettes were analyzed. The data set shows good agreement with the 1 cm pathlength data collected in a static system and used as the basis for the peak ratio analysis. Importantly, the method is insensitive with respect to metal ion concentration ranging from 0.01 to 1.25 M UO_2^{2+} . Interestingly, the peak ratio levels out at $\sim 7 \text{ M NO}_3^-$, suggesting that the speciation stabilizes near the end of the examined range. Peak Ratio method was modified prior to examining flow cell data. The method was modified to be dependent on the nitrate concentration only considering data from systems using nitric acid only. This removed any effects that alternate counter ions (i.e. Na^+) might add and more closely approximates the feed stream of the UREX process. Trivalent actinide-lanthanide separations focused on cloud-point methods. Studies and literature reviews were initiated to provide more detail on the 8-hydroxyquinoline:Ln(III) system. As the efforts in Task 2 will focus on TALSPEAK, details from these studies are excluded from this quarterly report.

5. Work Performed in 3rd Quarter (April-June 2009)

In the 3rd quarter efforts focused on Task 1 with initial investigation of the TALSPEAK system in Task 2. Thermodynamic titrations of the uranyl-nitrate-acetate system were extended to 2.0 molal ionic strength. The methodology for production of the samples was based on previous work on the 1.0 molal ionic strength system. The results provided a value of $\log \beta_{(1,1)} = -0.399 \pm 0.022$ for uranyl nitrate. Experiments to determine complexation constants involving UV-Visible spectroscopy were initiated and will exploit the program Hyperquad. The laser returned and was re-installed and aligned by early May. The system performance has been mostly stable though there have been some issues. As of the end of this quarter, the system is still being tested and adjusted in house. Three samples were sent to the APS for EXAFS in June. Initial analysis of the data indicated a drastic shift in the equatorial U-O bond distance. It is currently theorized that this is due to the third nitrate bonding in a bidentate fashion. UV-Visible samples were prepared at 2, 4, 5, 7, 8 M HNO_3 with 0.01, 0.126, 0.262, 0.63 and 1 M UO_2^{2+} , in an effort to close the data gap from previous work. A student in the program performed an internship at INL and information on TRUEX studies performed at part of this internship is presented.

6. Work Performed in 4th Quarter (July-September 2009)

In this quarter all tasks were examined. A UNLV student researcher participated in TRUEX studies at INL. Contactors were used to examine the TRUEX flowsheet, thermal profiles, and separation of key elements. Evaluation results indicated the pilot plant equipment performed satisfactorily, however, additional flowsheet testing is warranted to better define operating

throughputs, rotor speeds, solution flow path between stages and potential off-gas design changes

The uranyl nitrate system was investigated by UV-Visible spectroscopy and EXAFS. The formation of nitrate species was evaluated as a function of nitric acid concentration. Peak ratios in the obtained spectra were correlated with previous results. The EXAFS data indicates bidentate nitrate complexation. Thermodynamic measurement on the nitrate uranyl system continued. This included the use of spectrophotometric titrations to evaluate the complexation constant.

The spectroscopic studies of the TALSPEAK systems included UV-Visible and Laser Fluorescence examination of both lanthanides and actinides. Initial studies were performed on Er by UV-Visible spectroscopy. From these results lactic acid was not expected to form Er species under TALSPEAK conditions. However complex formation is evident in the DTPA studies. The system was examined with Tb by laser fluorescence, showing the same behavior as Er with the ligands. Curium was examined by TRLFS with DTPA. The lifetime showed a dramatic difference for the Cm species. The free ion lifetime was measured at 80.14 ± 4.21 μsec , while the Cm DTPA complex is 220 μsec . This demonstrates that TRLFS can be used to evaluate Cm speciation on line in the TALSPEAK process. As the excitation wavelength is selective, this method can also be used to examine Tb speciation during TALSPEAK separation as well, provide the spectroscopic system is optimized for Tb.

7. Work Performed in 5th Quarter (October-December 2009)

Efforts in this quarter of the project focused on absorbance spectroscopy. The uranyl system was investigated by UV-Visible spectroscopy, X-ray absorption fine structure spectroscopy (EXAFS), and density functional theory. From the contributions of the EXAFS experiments and the DFT modeling it was hypothesized that the uranyl mononitrate was thermodynamically unfavored due to the low symmetry of the molecule. Instead, it was theorized that the dinitrate species would be prevalent, even at low (<0.5 M) nitrate levels. This would help to explain the extraction behavior seen in the TBP/Dodecane extraction. The results of the first set of experiments showed the need for 2-3 different conditions at a given molality to completely cover the intended range. This arrangement allowed the molal concentration not to vary while changing the nitrate concentration in each sample. Currently, the molal value for the dinitrate conditional constant at 1 molal ionic strength is 0.8873 ± 0.0024 . In an effort to elucidate the uranyl nitrate speciation, 14 samples were prepared for EXAFS analysis. Changes can be seen across the 4-10 M HNO_3 region. The samples showed an increase in the U-O equatorial bond length which increased until ~ 10 M HNO_3 . The change in bond length corresponds to an increase in the U-O-N multiscatter suggesting that the increase in bond length is due to the formation of the sterically hindered trinitrate complex which has no water molecules in its inner sphere. The development of contactors to evaluate separations by spectroscopy was initiated. Contactors were purchased and setup.

8. Work Performed in 6th Quarter (Jan.-Mar. 2010)

The titration to determine uranyl nitrate stability constants have been completed for the low nitrate samples (up to ~ 1 M) at ionic strengths of 1, 2, 3, 4, and 6 molal. The results indicate that the dinitrate species is dominant over the mononitrate species by a wide margin and is generally the main species that will be found in solution. The residuals of the higher molal titrations indicated that a new species was starting to appear near the highest nitrate concentrations. If it is

assumed that this species is the trinitrate, then it should be visible at higher nitrate concentrations. The work with the uranyl system, coupled with the literature available, as well as the current data on the plutonium nitrate system has suggested a possible theory explaining the uneven nature of actinide nitrate speciation. It is hypothesized that in the absence of other driving factors the speciation of uranyl and plutonium with weak ligands can be significantly influenced by the symmetry of the formed species.

A primarily ^{239}Pu stock solution was prepared to evaluate the interaction with nitrate. By analytically adding the raw spectral data from a uranyl and comparable matrix plutonium solutions with a process-appropriate U/Pu ratio, the overlaps in the 350-500 nm region were examined. This makes spectral deconvolution difficult, primarily for Pu. The Pu(IV) absorbance near 650 nm can also be utilized, as the uranyl absorbance is non-existent at this wavelength.

In TALSPEAK studies, due to previous contacts ambiguity of whether water transport was due to pH change, or due to Na^+ transfer, follow-up experiments were performed to decouple the change of pH from the change in Na^+ concentration. There appear to be two regimes of water extraction behavior. The two behaviors occur when the final solution pH falls either within the lactic acid pH buffer region ($\text{pH} < 3.8$) or above where the buffering by lactic acid is not occurring ($\text{pH} < 4.4$). Contact experiments with Nd were performed to understand extreme process conditions that could be encountered in the TALSPEAK process. The solutions run the range from high-loading to more normal loading levels and seek to understand how speciation in the organic phase changes in these two regimes. In an attempt to quantify the extraction of Nd the visible absorbance spectrum of the aqueous phase was observed. The trend in the absorbance shows that as the ligand concentration increases the aqueous phase absorbance decreases due to increasing extraction to the organic phase.

Ongoing efforts to setup the three countercurrent contactors included collecting and examining various tubing connectors to enable contactor flow, then ordering additional tubing from Cole Parmer and investigating Swagelok fittings for Ocean Optics dip probe in-stream mount. Following the completion of physical setup, 2 phase flow will be demonstrated through all 3 stages and the Ocean Optics UV-Vis dip probe will be mounted at various points to acquire data from uranium aqueous phase experiments as a proof of concept. All of these efforts will culminated in a scaled UREX-type demonstration with up to $\sim \text{M}$ level UO_2^{2+} , actinides (U, Pu, Np), lanthanides (perhaps Nd due to excellent UV-Visible activity), fission products/Fe (for investigation of interferences), AHA (UREX vs PUREX) and the UV-Vis online monitoring.

9. Work Performed in 7th Quarter (April-June 2010)

The spectrophotometric titration results from the previous quarter were used to extrapolate the zero ionic strength stability constant. A program suite provided by the IUPAC, "Aqueous Solutions," was used to perform the Specific Ion Interaction Theory calculations. The data from 1-4 molal produced a value of $\log \beta_2^0 = 3.44 \pm 0.13$.

Experiments were performed to add more data points at the higher loading conditions (0.025, 0.075, 0.125 M HDEHP) for TALSPEAK studies. The aqueous phase was modeled according to the Debye-Huckel activity and the complexation of lactate with Nd so that the ionic strength of the solution was a total of 1.2 M. The distribution of Nd was quantified using visible spectroscopy. The organic phase from each phase was run on a Nicolet 6700 series FTIR. The spectra are an average of 64 scans taken using absorbance mode. These results showed some similar trends to the visible spectroscopy. There were some unique bands that formed during

high loading that disappeared when the loading decreases. For the ^{14}C labeled Lactate distribution experiments, small samples of 0.4 mL of each phase were placed in vials, and then spiked with 30 μL of a ^{14}C labeled lactate tracer. The solutions were contacted for 16 hours, separated and counted by liquid scintillation counting. Again, under high loading conditions it appears that the lactate is extracted very strongly. The same organic phase used for the visible spectroscopy and the FTIR was titrated using a Karl Fischer titrator. This data shows that the water in the organic phase appears to be a function of Nd, Lactate, and Na in the organic phase. Organic phase from the batch contacts was also analyzed by ^{31}P NMR. It was discovered almost immediately that the samples with Nd present caused paramagnetic shielding that prevented a peak from being resolved. The same sample was cooled and run with a longer collection time and the resulting spectra were obtained. These results have been correlated with HDEHP concentration showing a linear trend.

Volatility based separations were also examined. Discharged fuel can be recycled in a dry process and utilized as fuel for other reactors. The general recycling process is based on AIROX and OREOX technologies previously studied for recycle of used LWR fuel. Volatile and semi-volatile fission products are removed in various fractions by flowing gas over the oxidized used fuel under a variety of temperatures. Conservative estimates state that 100% of tritium, K, Xe, I and ^{137}Cs , 90% of Cs and Ru, and 75% of Te and Cd are removed through this processing. Lower volatility fission products (Ba, Sr, Ce, La, Pd, Zr) stay in the fuel along with the uranium, plutonium and actinides [1,2]. This advanced fuel cycle would reduce the mass of used nuclear fuel needing to be disposed of as HLW, extract more energy from the original uranium ore and potentially deplete weapons-material inventories, while maintaining proliferation resistance [3]. Safeguarding dry fuel cycles is inherently simpler than wet fuel cycles mainly because when material is moved, it is moved as a discrete mass - no liquid is flowing through lines or passed through valves. Even waste and scrap are handled as discrete, tracked and weighed items [4]. The use of laser ablation techniques for the on-line analysis of dry reprocessing will be explored to expand the applicability of the safeguards research performed in this project.

10. Work Performed in Final Quarter (July 2010 to September 2010)

Finalization of the analysis and evaluation of uranium spectroscopy was performed. This work has shown the potential and use of visible spectroscopy, specifically UV-Visible and Time Resolved Laser induced Fluorescence, for the monitoring of nuclear fuel reprocessing streams. Both uranium and plutonium concentrations can be measured directly by UV-Visible spectroscopy. The total plutonium(IV) concentration can also be determined in the absence of process chemistry information by using one of the isosbestic points. Finally, in select process streams, TRLFS monitoring can be used to detect millimolar concentrations of uranium and lower concentrations of curium.

Figure 2: Elemental comparison of EM² and LWR used fuel

Evaluation of dry reprocessing techniques continued. An evaluation of dry reprocessing techniques is presented. Efforts to develop on line methods to examine the chemistry during dry processing will be based on known

literature and experiments. Proposed methods include laser ablation mass spectroscopy and Raman/IR methods. The majority of previous research on dry processing technologies has been for application to the LWR fuel cycle, therefore in order to show the relevance of these works a fuel cycle, an elemental comparison a model EM² and LWR UNF is shown in Figure 2 (assuming 140 GWd/t and 10 year decay for EM², 60 GWd/t and 10 year decay for LWR).

Figure 3: UNF discharge mass for EM² and LWR, both for 1200 GWd and 30 year buildup

Literature is available on the radionuclides in a LWR core and the resulting gaseous, liquid and solid chemical species present both during normal operation and at discharge [5,6]. A mass comparison between EM² and LWR discharges is found in Figure 3.

In general, LWR discharge fuel is assumed to have 60 GWd/t burnup and have the composition of 95% U, 1% Pu and minor actinides, and 4% fission products, with a mass distribution reported in the previous quarterly report [7,8]. Variations in reactor type, burnup, core life and decay time mean that isotopics of LWR UNF will likely be unique to each case, however the rough composition estimates should remain the same even within these variables.

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