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Author(s):

Robert J. Donohoe, INC-14
David E. Morris, INC-14
John A. Musgrave, INC-9
C. Drew Tait, INC-14
Steven D. Conradson, MEE-11

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URANIUM SPECIATION IN FERNALD SOILS

Progress Report:

October 1, 1992 - May 31, 1993

Robert J. Donohoe, David E. Morris, John A. Musgrave, and C. Drew Tait

Isotope and Nuclear Chemistry Division

and

Steven D. Conradson

Mechanical and Electronic Engineering Division

Los Alamos National Laboratory

Los Alamos, New Mexico 87545

ABSTRACT

This interim progress report describes new experimental data collected from October 1, 1992 through May 31, 1993 as part of the Characterization Task of the Uranium in Soils Integrated Demonstration of the Office of Technology Development, Office of Environmental Restoration and Waste Management of the United States Department of Energy. X-ray absorption and optical luminescence spectroscopies have been used to analyze the uranium remaining in contaminated soils from the Fernald site after these samples were treated by various decontamination technologies under development within this Integrated Demonstration. The treatment technologies included soil washes with carbonate, citrate, Tiron, and Tiron / dithionite mixtures. The effectiveness of these procedures is discussed in a separate report from the Decontamination Task group. Our characterization results indicate that following the application of all of these treatment strategies the uranium remaining in the soil is still best characterized as being primarily in the hexavalent oxidation state. However, for the A-series soils from the Incinerator Area, the speciation of this remaining uranium is different than seen previously based on the x-ray absorption data. The luminescence data for these treated soils also demonstrate that there is a decrease in size and quantity of the particulate hexavalent uranium that gives rise to the structured green emission. Thus, all treatment technologies do seem to generate a more dispersed, finer-grained form of uranium. For several treated samples a characteristic luminescence signal was found indicative of a schoepite phase of uranium. New results are also reported for untreated Fernald soil samples and reference uranium mineral phases.

INTRODUCTION

A key factor in the successful and timely remediation of contaminated sites at United States Department of Energy (US DOE) facilities is the availability of speciation information on the contaminants. The actinides (uranium, neptunium, and plutonium) in particular can exist in a myriad of different chemical forms, and their response to remediation technologies, particularly those based on chemical intervention, will vary as the speciation varies. If there is an *a priori* characterization of the speciation (i.e., specification of the oxidation state, chemical form and structure, and physical state [surface precipitate, secondary mineral, or adsorbate]), several potential advantages accrue. The remediation technology can be devised with the specific form of the actinide in mind. This will lead to more efficient and less drastic remediation measures, and enhance the chance of leaving a viable soil substrate behind. In addition, a valid assessment can be made of the long-term stability of the actinide contamination in its present form. This information will be important to guide the remediation effort and to determine whether the contaminant site is stabilized with respect to transport and subsequent groundwater contamination.

We are presently involved in the determination of uranium speciation in contaminated soils at the United States Dept. of Energy's former uranium processing facility located at Fernald, Ohio as part of the Uranium in Soils Integrated Demonstration sponsored by the Office of Technology Development of the US DOE. This facility was formerly devoted to the processing of large quantities of uranium. Many different processes were employed at the site, and there are potentially many different source terms for uranium in the soils. While migration of uranium in general has not been extensive, the shallow subsurface requires significant remediation to bring the site into compliance with EPA standards. We have employed molecular-level probes including x-ray absorption spectroscopy and optical luminescence and Raman spectroscopies to characterize the uranium speciation in samples from several different sites at the facility. In this interim progress report, we present recent results from Phase 1 activities as described in the FY 93-94 Characterization plan (Morris, 1993). The focus of our Phase 1 efforts has been on characterizing the speciation in samples treated by the Los Alamos National Laboratory (LANL) Decontamination Task (J. R. Brainard, PI) and the Oak Ridge National Laboratory (ORNL) Decontamination Task (C. Francis, PI). As such, the speciation information we are attempting to obtain is principally that of the uranium that is left behind in the soils following the soil-washing treatment(s). Specific issues / concerns that we hope to address include determining whether the soil-washing selectively removes certain uranium species, whether the treatment alters all or most uranium phases, and how the treatment affects the dominant physical forms of the uranium. (For example, do the particulates decrease in size and / or become redistributed?) In addition, new results on untreated and reference materials are presented that help in understanding the speciation behavior and identifying uranium species in both treated and untreated samples.

EXPERIMENTAL

Samples

Most of the speciation characterization work that we have carried out during this period has been on three new sets of samples, in addition to the SP series of unwashed samples already received from Dr. S.Y. Lee of ORNL. The first set of samples was supplied by Dr. J.R. Brainard of LANL (Table I). These samples were from the A-14 lot from the Incinerator Area, and were treated according to the procedures under

development at Los Alamos. Specifically, washings were in Tiron only (100 mM solution), Dithionite only (100 mM solution), Tiron+Dithionite (100 mM in each), and Carbonate/Bicarbonate ($0.1 \text{ M CO}_3^{2-} + 0.1 \text{ M HCO}_3^-$). The second set was provided to us by Dr. S.Y. Lee and consisted of two soils (A-14 and B-16) washed with sodium carbonate (15 minute attrition time at room temperature) or sodium citrate (15 minute attrition time at room temperature) (Table II). The final set of new samples consists of specimens from the library of high purity uranium-bearing minerals maintained by the University of New Mexico (Professor Rodney Ewing, Department of Earth and Planetary Sciences, Albuquerque, NM). These samples are valuable for use in expanding our data base of luminescence spectra for comparison with Fernald data. The identity and purity of these minerals have been confirmed by XRD analysis. Table III lists the samples in this group.

Specific analytical data on the treated soil samples that we investigated were not available at the time this report was prepared. It will be included in the year-end summary report from the Characterization Task Team. However, from a qualitative perspective for the Los Alamos soil-washing samples, Tiron + dithionite is more effective than carbonate / bicarbonate, which is more effective than Tiron alone, and these three methods are significantly more effective than dithionite only. Treatment with deionized water removed no measurable amount of uranium from the A-14 soil.

Instrumentation

The spectroscopic instrumentation used to collect data in this report has been described in detail elsewhere (Tidwell et al., 1993). The scanning electron microscopy data were collected on an International Scientific Instruments Model ISI-SX-40 unit interfaced to a Tracor Northern Pulse Processing and Detector Electronics package. The x-ray diffraction data were obtained on a Philips Model XRG 3100 x-ray system using copper K_α radiation.

RESULTS AND DISCUSSION

Treated Soil Samples: X-ray Absorption Data

X-ray absorption data have been collected to date on eight samples from the Los Alamos group and five samples from the Oak Ridge group after treatment according to their respective uranium soil-washing strategies. Note that even though some of these samples are suspected to have uranium contamination removal of ~ 90% (e.g., the Tiron + dithionite treatment), adequate signal-to-noise was still achievable by x-ray absorption. We have found excellent reproducibility in collected data from the same sample at different times as well as from different samples treated by the same treatment strategy (e.g., Los Alamos samples Mar-32-9-A and Mar-32-9-J, Table I).

In general, the uranium that remains in all samples following treatment is still best characterized as predominantly hexavalent based on the position of the absorption edge and the prominence in the radial structure function of a peak at ~ 1.8 Å that is uniquely associated with the axial "yl" oxygen-uranium bonds. However, a difference in the hexavalent uranium speciation in most of these treated samples (relative to the speciation in untreated samples) is clearly evidenced by a relative increase in the

amplitude of the white line at ~ 17.18 eV and the loss of the characteristic shoulder in the near-edge region (~ 17.19 keV, Fig. 1). This shoulder has been observed in the spectrum of all untreated Fernald soils and most of the hexavalent uranium model complexes we have examined.

The treated samples which exhibit this new XAS spectrum (i.e., one with increased white-line amplitude and without the near-edge shoulder feature) are all from the A-14 lot and include those treated with Tiron only (e.g., Mar-32-9-A), those treated with Tiron plus dithionite (e.g., Mar-32-9-E), those treated with carbonate (e.g., AX-15-T20), and those treated with citrate (e.g., AY-15-T20). Samples treated with dithionite only (e.g., Mar-32-9-C) and the control samples treated only with deionized water (e.g., Mar-32-9-P) have XAS spectra similar to the untreated samples.

The A-14 sample that underwent the pretreatment processing only (dry sieving followed by ball milling) gave an XAS spectrum similar, but not identical, to those for completely untreated soil samples. While the shoulder in the XANES region is present in the spectrum of the pretreated soil, the total spectrum appears less sharply defined, and the first EXAFS oscillation is slightly shifted. Thus, the pretreatment appears not to be entirely innocuous with respect to the bulk uranium speciation.

It is interesting that the different treatment technologies that proved effective (i.e., all but the dithionite-only treatment), particularly the one involving a reducing agent (Tiron + dithionite), all seem to generate samples with comparable x-ray absorption spectral data. It is tempting to ascribe this spectral result to a single remnant uranium species that is unresponsive to all of the treatment technologies, but such a conclusion is premature. This is particularly true since there are slight but significant variations seen in the XAS spectra for the treated samples giving comparable signals, particularly in the extended fine-structure (EXAFS) region, suggesting that this new signal may be seen for several different species. However, it is clear that we are not seeing oxidation-state redistributions that might have been expected for a reprecipitated chemical product from the treatment (e.g., the precipitation of a U(IV) product from the dithionite treatment).

Only one treated sample from the B-16 soil lot [Plant 1 / Storage Pad Area] has been examined thus far. This sample from Oak Ridge [BX-15-T20] underwent carbonate treatment, and the spectrum of the small size fraction (< 20 microns) had the same general appearance as those from untreated samples. It is encouraging that we are still able to obtain sufficient signal-to-noise in the spectrum for this sample, because preliminary reports from both the LANL and the ORNL Decontamination Task teams indicate that very little uranium remains in the B-16 samples following virtually any treatment.

Treated Soil Samples: Optical Spectroscopic Data

The most notable result from the optical spectroscopic investigations of the treated soil samples is the substantial decrease in the quantity and size of the particulate uranyl species that give rise to the structured green luminescence spectra reported previously. We have employed the complete range of experimental perturbations for these studies including either a tightly focused laser excitation source or large-area laser and continuous-wave excitation sources and using either microscopic imaging or manual rastering of the sample in the excitation beam. In all cases we find no significant quantities of structured luminescent uranyl species. We have consistently found a broad-band emission similar to that seen in the SP-9 series of samples for all of the treated samples. Note that this broad emission is typically centered at $\sim 18,000$ cm^{-1} , well red-shifted from the typical energy of iron and / or organic impurity emission in the soil

samples. However, we have no definitive evidence to assign this broad-band emission to uranyl species.

Small green luminescent "spots" have been found for three of the samples using tightly focused 364 nm laser excitation. The luminescent spectra obtained from these spots are shown in Fig. 2. Note that these spectra are essentially identical suggesting that the same uranyl species is giving rise to the spectrum from each sample. We were unsuccessful in our efforts to obtain Raman vibrational data from these green "spots." In addition, we found no indications of the orange emissive particles that we have recently found to be prevalent in the untreated soil samples from several locations at the site (see below).

Clearly the diminished occurrence of discernible uranyl emission in the treated samples is consistent with the preliminary analytical results which show that as much as ~90% of the uranium can be removed from the soils. However, other factors must be operating as well, because the signal to noise of our results in unwashed soils was >20:1 suggesting that we should easily see the remaining 5-10% of the uranium in the soils. (Note that the x-ray absorption data indicate that hexavalent uranium still dominates. Therefore, complete oxidation-state conversion to non-emissive uranium(IV) can be excluded.) Furthermore, even those samples for which the treatment was not particularly effective (e.g., dithionite-only and deionized water-only treatments) yielded samples with diminished uranyl luminescence. It is possible that as a result of all treatments the uranium may be more evenly spread along the surface of the soil matrix rather than existing as micron-sized particulates. Such an effect may be caused by dissolution followed by re-precipitation and/or attrition to smaller particle sizes followed by redistribution. This latter effect might be promoted by the ball-milling process used in the pretreatment protocol by the LANL team. The more homogeneous distributions of uranium resulting from either of these mechanisms would lower the uranyl luminescence relative to the broad emission background from organics or iron species remaining in the soil making it more difficult to locate and isolate the emissive uranyl species for spectroscopic characterization. Alternatively, upon dissolution / redistribution, the uranium may become sorbed onto the surface of iron oxide particles, especially goethite, or organic-coated particles. In these cases, the uranyl luminescence may be effectively quenched.

Note that the uranyl luminescence spectrum observed for several of the samples is very similar in energy and band shape to a naturally-occurring, well-crystallized schoepite sample (Fig. 2). These spectra are very distinct from those for freshly precipitated uranyl hydroxides, suggesting that the uranyl species giving rise to the spectra in these treated samples are not simply reprecipitation products resulting from hydrolysis of dissolved uranyl species during the course of the treatment processes.

Treated Soil Samples: Ancillary Data

X-ray diffraction powder patterns (XRD) of the untreated soils identified quartz, feldspars (Ca, Na, and K), dolomite, and calcite as the principle minerals present, with quartz contributing the dominant pattern. Neither clay minerals nor apatites were important features, and an unassigned peak at $2\theta = 37^\circ$ was found. Many of the XRD features of the treated soils remained the same: quartz is still the dominant constituent in all of the XRDs, while feldspars also contribute [especially albite (Na), and anorthite (Ca)] in CaCO_3 washed samples, while orthoclase (K) became relatively less abundant. Furthermore, calcite and especially dolomite became more prominent in the treated

samples, especially those treated with carbonate and citrate. The XRDs of the small-size fraction from the carbonate treatment (BX-15-T20 <20; AX-15-T20 <20) changed the most of all samples, with the former showing dolomite and calcite becoming more predominant in the pattern. The unknown peak at 37° remained in all samples after treatment. The formation of any amorphous phases during the washing process is discounted as no broad scattering band is found in any of the XRDs. Several samples (AX-15-T20 <20; AX-15-T20 75-20; BX-15-T20 <20; BY2-15-T20; 32-9-F/D) developed a new peak at 3.5° . Because of the large d-spacing implied by this peak ($23\text{\AA}$), this species is likely to be a clay mineral, possibly a weathered product from original feldspars. It is very encouraging that these XRD data for the treated samples indicate that the treatment processes don't significantly alter the native soil characteristics.

Untreated Soils and Reference Uranium Minerals

Efforts continue to precisely identify the various forms of uranium in the Fernald soils. To this end, we continue to examine untreated samples in search of new uranium species and more precise spectroscopic data (e.g., Raman vibrational spectra) for previously identified species. We have also continued studies of reference uranium minerals and compounds. Most notably, we have established a collaboration with Prof. Rodney Ewing of the University of New Mexico that has enabled us to access the extensive collection of uranium minerals in their mineral library. Substantial new luminescence and Raman spectroscopic data have been collected on oxy-hydroxide, carbonate, phosphate, and silicate minerals for comparison to the Fernald soil data.

Several new emissive particulates have been identified in portions of the original SP series of samples. In general, the emission bands from these phases are broad, poorly structured, and shifted to lower energy with respect to the highly structured green phases noted previously. The color of these emissions is best described as orange. Examples of the orange emission from these particulates are shown in Fig. 3. For the SP2 series of samples, these orange phases are frequently found in close association with the green particulates. The variability in the spectra for these orange phases suggest that several uranyl species are responsible. In addition, some of the orange emissive phases appear to be quite sensitive to laser photolysis. Prolonged exposure to moderate to high intensity 364 nm excitation results in significant changes in the emission spectral profile. This result suggests that these uranyl species are either associated with organic species (i.e., as uranyl-organic ligand complexes) or they are undergoing some annealing process. We have also collected additional luminescence and Raman data on the typical green particulates in the SP2 series. The new Raman data reveal two distinct uranyl vibrational bands at $\sim 830\text{ cm}^{-1}$ indicating the existence of two populations of uranyl species in these particulates.

The luminescence data from the reference uranium minerals have enabled us to draw several important conclusions. First, it is unlikely that uranyl carbonates are the species responsible for the structured green emission seen in the soil samples. This is because the simplest uranyl carbonate, rutherfordine, yields only broad, structureless emission, and simple ternary minerals such as liebigite (calcium uranyl carbonate), while possessing structured emission, have significantly shifted emission energies. For similar reasons, we can eliminate becquerelite (a common uranyl hydroxide phase) as a possible source for the structured green emission. We continue to find that both silicate (sklodowskite, soddyite) and phosphate (meta-autunite, meta-ankoleite, phosphuranylite, saleeite, meta-torbernite) phases are candidate matches for the structured green emission. While these phases all have comparable emission spectra, their Raman spectra are much

more distinct, and we hope to be able to use the Raman data to make unequivocal assignments of the species in the Fernald soils.

Schoepite + phosphate phases or schoepite + silicate phases are still the closest spectral matches to those found in the Fernald soils. The presence of schoepite has been confirmed by XRD data from hand-concentrated sample of the green emitting particulates. However, the structured emission can not be attributed simply to phosphate or silicate phases. The energy dispersive SEM of the concentrated Fernald green emitters showed both silicon- and phosphorous-containing anions (any carbon or oxygen would not be detected because of their low emission energy), as well as calcium, potassium, and magnesium cations, so a possible combination of phosphates and silicates can not be eliminated. The crystal habit of the green emitters in the Fernald soil can be described as platy. This habit is consistent with an oxy-hydroxide (e.g., schoepite) and / or a phosphate phase (e.g., autunite), whereas silicates (e.g., sklodowskite) tend to have acicular crystal habits.

We have also attempted to identify the orange emitters found in the SP2 samples. Two reddish minerals from the library samples, kasolite (lead uranyl silicate) and curite (hydrated lead-oxide uranate), showed no emission from the bulk phase, but orange impurity emissions were noted. The impurity in curite is similar (but not identical) to that found in the orange emitter from Fernald SP2. On the basis of previous studies, it has been suggested that orange and red uranium emitters are uranate " UO_3 " structures such as ianthinite rather than uranyl ($[\text{UO}_2]^{2+}$) structures, but many historically " UO_3 " structures have been re-formulated as uranyl structures. Finally, alkyl-phosphates uranium compounds have been shown to emit orange to red upon optical excitation. The SP2 samples are the ones to exhibit the orange-red luminescence, and these were also taken near the Plant 2/3 area where TBP was used in the PUREX process. We are presently pursuing luminescence spectra from a series of known U-phosphate complexes to check this hypothesis.

The energy dispersive SEM for hand-concentrated orange emitters is much simpler than for the green emitters: silicon is greatly reduced in importance, phosphorous is not present at all, and the cations Mg and Ca are almost absent. In fact, the only cations in abundance are K and Al, with trace Fe also present. Clearly the orange emitters present in small quantities in untreated Fernald soil are not phosphates, nor are they likely to be silicates. This is in contrast to the green emitters, where uranyl species containing these anions are likely candidates for the structured emission. The lack of Ca and Mg in the orange emitting phases is especially strange because non-luminescent particles that get picked out with the orange emitters look like calcite/dolomite under a microscope and contain a carbonate peak (not calcite) in the Raman spectrum. In any case, the XRD spectrum from hand-concentrated Fernald orange emitters is consistent with the ianthinite species, which would also account for the lack of anions and cations associated with this uranium phase. This identification is questionable, however, as these peaks are also present in the hand concentrated green emitters and may therefore be from the soil matrix rather than a uranium species.

SUMMARY

X-ray absorption and optical luminescence spectroscopic data show that the uranium remaining in Fernald soils following all tested decontamination treatment technologies is still predominantly in the hexavalent oxidation state. For the A-14 series of samples, all treatment strategies that are effective in removing the uranium appear to

leave in the treated soil one or more species of uranium that give rise to a distinctive new x-ray absorption spectral pattern. It is unclear at this time if this signal is attributable to remnant uranium species that are intractable, but it is likely that it is not solely attributable to a precipitated product from the treatment because of the similarity of the x-ray absorption signal in samples treated by very different chemistries.

The optical luminescence data demonstrate that there is a decrease in size and probably quantity of the particulate hexavalent uranium that gives rise to the structured green emission. Thus, all treatment technologies do seem to lead to a more dispersed, finer-grained contamination. Some instances of particulate green emission still exist in these samples, and the spectral band structure suggests that this is attributable to a schoepite phase. A pervasive broad-band emission is also seen in all samples, but this is not confirmed to arise from uranium species.

Several new phases of uranyl contaminant in the untreated Fernald soils have also been identified. These are quite common in the SP2 soil, but they have also been found in soils from other sites. Recent luminescence and Raman spectroscopic data collected on reference uranium minerals have enabled us to solidify our assignments of the emissive uranyl species found in the Fernald soils. The best candidates continue to be silicates and phosphates. However, new Raman data may allow us to make quite specific assignments in the near future.

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TABLE I. Washed Soil Samples from Los Alamos National Laboratory

Sample Designator	Processing ^a	Treatment
Mar-32-9-A	Y	Tiron only
Mar-32-9-B	Y	Tiron only
Mar-32-9-C	Y	Dithionite only
Mar-32-9-D	Y	Dithionite only
Mar-32-9-E	Y	Tiron + Dithionite
Mar-32-9-F	Y	Tiron + Dithionite
Mar-32-9-G	Y	D.I. Water
Mar-32-9-H	Y	D.I. Water
Mar-32-9-I	Y	Tiron only
Mar-32-9-J	Y	Tiron only
Mar-32-9-K	Y	Dithionite only
Mar-32-9-L	Y	Dithionite only
Mar-32-9-M	Y	Tiron + Dithionite
Mar-32-9-N	Y	Tiron + Dithionite
Mar-32-9-O	Y	D.I. Water
Mar-32-9-P	Y	D.I. Water
Mar-33-9-A	Y	Carbonate/Bicarbonate
Mar-33-9-B	Y	Carbonate/Bicarbonate
Mar-33-9-C	N	Tiron + Dithionite
Mar-33-9-D	N	Tiron + Dithionite

^a Processed = 2 mm sieving and ball milling, unprocessed = drying only

TABLE II. Washed Soil Samples from Oak Ridge National Laboratory

Sample Designator	Treatment	Size
AX-15-T20	Carbonate	20-75 microns
AX-15-T20 <20	Carbonate	< 20 microns
AY-15-T20	Citrate	20-75 microns
AY-15-T20 <20	Citrate	<20 microns
BX-15-T20	Carbonate	20-75 microns
BX-15-T20 <20	Carbonate	<20 microns
BY2-15-T20	Citrate	20-75 microns
BY2-15-T20 <20	Citrate	<20 microns

TABLE III. U-Mineral Library Samples from UNM Radioactive Minerals Research Collection

	Formula	Comments
<u>OXY-HYDROXIDES</u>		
Becquerelite	$\text{Ca}(\text{UO}_2)_6(\text{OH})_{14}$	
Billietite	$\text{Ba}(\text{UO}_2)_6(\text{OH})_{14}(\text{H}_2\text{O})$	Sits as a crust on rutherfordine (suspected). Billietite is golden-yellow, pseudo-hexagonal crystals.
Curite	$(\text{PbO})_2(\text{UO}_3)_5 \cdot 4\text{H}_2\text{O}$	Acicular reddish crystals. Black areas reflect loss of water. Yellow impurities, if any, are soddyite and can be distinguished by their blocky habit.
Schoepite	$(\text{UO}_2)_2(\text{OH})_2 \cdot \text{H}_2\text{O}$	Separated from Rutherfordine co-phase
<u>CARBONATES</u>		
Kametoite	unknown	A REE uranly carbonate (structure has not yet been solved)
Liebegite	$\text{Ca}_2\text{UO}_2(\text{CO}_3)_3 \cdot 10\text{H}_2\text{O}$	
Rutherfordine	UO_2CO_3	Separated from Schoepite co-phase
<u>PHOSPHATES</u>		
Meta-autunite	$\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 10-12\text{H}_2\text{O}$	
Meta-torbenite	$\text{Cu}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 8\text{H}_2\text{O}$	Green, platy phase.
Saleeite	$\text{Mg}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 8-10\text{H}_2\text{O}$	
<u>SILICATES</u>		
Kasolite	$\text{PbUO}_2\text{SiO}_3(\text{OH})_2$	Blocky, reddish-brown crystals.
Soddyite	$(\text{UO}_2)_5(\text{SiO}_2) \cdot 6\text{H}_2\text{O}$	Bright yellow crystals. Forms a coating on torbenite (green, platy phase).

Figure 1. Background-corrected x-ray absorption spectra for treated Fernald soils. Sample Mar-32-9-A was treated at Los Alamos with Tiron only. Sample BX-15-T20 was treated at Oak Ridge with carbonate and size fractionated. Both samples were run as dry powders.

Figure 2. Optical luminescence spectra obtained for treated Fernald soils and a reference uranyl mineral using focused 364 nm excitation from an argon ion laser. Sample Mar-33-9-A was treated at Los Alamos using carbonate / bicarbonate. Sample Mar-32-9-B was treated at Los Alamos using Tiron only. Both soils were examined as dry powders. The mineral schoepite $[\text{UO}_2(\text{OH})_2 \cdot n\text{H}_2\text{O}]$ is a well-crystallized material co-existing with a rutherfordine (uranyl carbonate) phase.

Figure 3. Orange luminescent phases ubiquitously found in Fernald soil SP2. Spectra were generated using focused 364 nm argon ion laser excitation. The two spectra were generated from two different particulate masses within the sample. The solid line spectrum also shows the highly structured green emissive phase (uranyl silicate or phosphate) in intimate contact with the orange phase within this excitation area (~ 100 micron diameter).

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