

WPO 35197

# ACTINIDE STABILITY/SOLUBILITY IN SIMULATED WIPP BRINES PROJECT

## Stability of Pu(VI), Np(VI) and U(VI) in Simulated WIPP Brine

3/13/96  
Interim Report

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## Executive Summary

The initial results and observations from two ongoing Tasks being performed as part of the Argonne National Laboratory project entitled "Actinide Stability/Solubility in Simulated WIPP Brine" are briefly summarized. This summary is being submitted to fulfill a contractual milestone identified by the WIPP Actinide Source Term Program, that is being managed by Sandia National Laboratories. The data provided are preliminary and are subject to re-interpretation as more data are collected.

In the An (VI) stability/solubility experiments, the initial results obtained indicate that, in the absence of a reducing agent, the An(VI) oxidation state can exist as a stable species in simulated WIPP brine at actinide concentrations of ~ 0.1 mM. Many of the systems investigated are, however, not at steady-state. The concentration stability of the actinide depends on the redox stability of the actinide, the presence/absence of strong complexes (e.g., hydrolysis or carbonate), pH and brine composition. Systems that appear to be stable, and probably not at saturation, include Pu(VI) in ERDA-6 brine with/without carbonate present, Np(VI) at low pH and high pH in the presence of carbonate, and U(VI) at pH 5 in G-Seep and pH 10 with carbonate in ERDA-6. The stability in actinide concentration was supported by absorption spectrometry data that showed no change in the oxidation state.

Some of the systems investigated appear to already be approaching steady-state conditions. Uranium concentrations, at pH 7 in G-Seep and pH 8 in ERDA-6, are rapidly reduced to an apparent steady-state value that is 2-3 orders of magnitude lower than initial concentrations. Neptunium concentrations, at pH 10 in the absence of carbonate, are also reduced by an order of magnitude to an apparent steady-state value.

Redox instability, as evidenced by the formation of An(V) species, was evident in the Np(VI) and Pu(VI) experiments in G-Seep (i.e., at low pH). For neptunium, reduction to Np(V) was rapid so contribution to long-term solubility from the An(VI) species can be discounted. For Pu(VI), reduction was slow and appeared to be linked to autoradiolytic effects. In this context, arguments to discount Pu(VI) as an important species, in the absence of a reducing agent, are based on accounting for radiolysis effects. For uranium species, there was no spectral evidence for redox although analysis of the phases formed is likely to show predominantly An (IV) solid phases.

In Task II, the effect of organic chelators on the redox stability of the An(VI) species is being investigated. The presence of organic chelating agents, specifically oxalate, citrate and EDTA, destabilized the An(VI) oxidation state in all Pu and Np systems investigated. This led to the reduction of An(VI) organic complexes to form organic complexes of the An(V) and An(IV) oxidation state. The stable neptunium oxidation state, over the 30 day duration of the experiments, was Np(V). These Np (V)-organic complexes appeared to be at or near steady-state. For plutonium, after 10 days, the final oxidation state distribution has not reached steady state. Reduction is however proceeding to form Pu (IV)-organic complexes and it is predicted that Pu (IV) will be shown to be the predominant oxidation state when organics are present.

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## 1.0 BACKGROUND

The project entitled "Actinide Stability/Solubility in Simulated WIPP Brine" is being performed at Argonne National Laboratory (ANL) as part of the Dissolved Actinide Concentration Subprogram of the Actinide Source Term (AST) Program. The AST program is an effort to develop a comprehensive mathematical model based on the Pitzer methodology to describe the solubility of actinides in concentrated brine solutions. Brines, present at the WIPP site, have the potential to mix with emplaced waste and provide a pathway for actinide transport out of the WIPP disposal horizon. The Pitzer model is parameterized using binary and ternary electrolyte solution data only. Data on solubility for complex electrolyte solutions are not needed for developing the model, but are necessary to test the predictive ability of the model for these solutions.

The WIPP Empirical Actinide Stability/Solubility Project was implemented to provide experimental data on actinide solubility and oxidation state distribution to test and challenge the AST model. The experimental plan consists of determining the stability of actinides in two synthetic WIPP brines, G-Seep and ERDA-6, and measuring the solubility of selected actinides in these brines. The effects of carbonate and three organic ligands, oxalate, citrate, and EDTA, on actinide solubility are also evaluated. The solubility measurements are being conducted at ANL independent of other laboratory programs that are determining Pitzer parameters for the AST model. In addition to providing a challenge for the AST model, the empirical solubility measurements serve as a contingency plan for the AST program. In the event certain parts of the model cannot be completed by the deadline for submission of the WIPP compliance application to EPA, the empirical data will be used to supplement the AST model and provide a complete data set for actinide solubility.

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## 2.0 TASK I: STABILITY/SOLUBILITY OF THE AN(VI) OXIDATION STATE IN SIMULATED WIPP BRINE

The objectives of this Task are to establish the stability of the An (VI) oxidation state in simulated WIPP brines. Two brines, ERDA-6 at pH 8 and 10 and G-Seep at pH 5 and 7, are being investigated. Carbonate is present in the higher-pH ERDA-6 brines. The experiments are being conducted anoxically under a  $950 \pm 50$  torr hydrogen atmosphere at  $25 \pm 5$  °C. Initial actinide concentrations are ~ 0.1 mM. The stability of the An (VI) oxidation state is being established by absorption spectrometry. Total actinide concentrations are being measured by a combination of ICP-MS and alpha scintillation counting.

### 2.1 Summary of Experimental Results

The concentration data for the actinide stability/solubility experiments are tabulated in Appendix A (both data Tables and Figures). The spectra that show the oxidation-state distribution are in Appendix B.

For each actinide, six experiments are underway. These are 1) pH 10 with carbonate in ERDA-6, 2) pH 8 with carbonate in ERDA-6, 3) pH 10 in ERDA-6 without carbonate, 4/5) pH 7 in G-Seep, and 6) pH 5 in G-Seep. Two experiments are being done in G-Seep at pH 7 because carbonate will be added to one of them after a steady-state concentration has been observed. After two months, actinide concentrations in only a few of the experiments are clearly approaching steady-state.

#### Uranium Stability/Solubility Experiments

- ERDA-6:** Uranium, in the presence of carbonate at pH 10, is stable as a carbonate complex at ~ 0.1 mM concentrations (experiment U-E10). This is supported by the concentration data, which show no effect of filtration on total uranium measured, and the absorption spectroscopy which shown the dissolved species to be a carbonate complex. At pH 8, however, even in the presence of carbonate, concentrations are quickly reduced to almost three orders of magnitude lower ( $\sim 10^{-7}$  M). The concentration of uranium, in these experiments, appear to be approaching steady-state. When carbonate is not present (experiment U-E10-NC) the results are less clear-cut. Initially, solution concentrations are low, but a steady increase has been observed in both the filtered and unfiltered fraction.
- G-Seep:** A two order-of-magnitude decrease in total concentrations was observed in the two G-Seep experiments being conducted at pH 7 (experiments U-G7 and U-G7-NC). These concentrations appear to be approaching steady-

state values at  $\sim 2 \times 10^{-6}$  M. Identical results were obtained in both of these experiments. These concentrations are too low to see using conventional spectroscopy so no speciation data are available for these experiments. At pH 5 (experiment U-G5), the uranium is stable as an aquo/chloride species and is probably not at saturation. The time-dependent absorption spectra support this conclusion since there has been no change noted in the observed spectrum.

### Neptunium Stability/Solubility Experiments

- ERDA-6: In the presence of carbonate (experiments NP-E10 and NP-E8), total concentrations of neptunium are stable. The spectra obtained at pH 8 and pH 10 are very nearly identical but show spectral evidence for several Np (VI) and possibly Np (IV/V) species. The predominant species appear to be a carbonate species and a yet un-identified inorganic (possibly mixed-ligand) complex. As alluded to, there is some evidence of oxidation-state instability (<10%). In the absence of carbonate at pH 10 (experiment NP-E10-NC), the concentrations are decreasing in time to approximately an order of magnitude lower in concentration. There is also spectral evidence for reduction to Np(V) species.
- G-Seep: At lower pH (experiments NP-G5 and NP-G7/NC), Np (VI) is being reduced to Np(V). This reduction is proceeding more rapidly at pH 5 than at pH 7. At pH 5, the total concentration of neptunium is remaining stable. At pH 7, an  $\sim 20$ -30% decrease in concentration is associated with the redox instability being observed.

### Plutonium Stability/Solubility Experiments

- ERDA-6: At pH 8 and 10, in the absence and presence of carbonate, the total concentration of plutonium is stable at  $\sim 0.1$  mM. When carbonate is present (experiments PU-E10 and PU-E8), the absorption spectra are almost identical and correspond to a carbonate species. At pH 10, in the absence of carbonate (experiment PU-E10-NC), an undefined Pu(VI) - inorganic complex is present. There is no evidence for the reduction of Pu (VI) under the conditions investigated.
- G-Seep: The replicate experiments (PU-G7 and PU-G7-NC) being done at pH 7, show that total plutonium concentrations are essentially stable over two months with perhaps a slow, < 5%, decrease in concentration evident. This reduction is presumably due to autoradiolysis. There is spectral evidence for Pu(V) formation. At pH 5 (experiment PU-G5), total

concentrations have decreased by ~ 20%. The speciation, based on the absorption spectra is similar to that observed at pH 7 except that a lower concentration of Pu(V) is present.

## 2.2 General Observations and Conclusions

The An (VI) oxidation state can exist as a stable species in simulated WIPP brine at actinide concentrations of ~ 0.1 mM. The observed stability depends on the redox stability of the actinide, is increased by the presence of carbonate, and depends on the pH. Both carbonate species and "aquo-like" species (e.g., hydrolytic, sulfate, chloride) are observed spectroscopically. Some of the systems investigated appear to be approaching steady-state conditions. Most, however, are either not at saturation or are still undergoing change (e.g., precipitation and/or redox).

The presence of carbonate in ERDA-6, with one exception, led to the formation of a stable actinide carbonate complex that was not at saturation for all three actinides investigated. The one exception was uranium at pH 8 where a significant decrease in total concentration was noted. In the absence of carbonate, at high pH, the behavior of the actinide system was much less predictable. In the uranium system, an initial decrease in concentration was followed by a slow increase in concentration. This phenomena is as yet unexplained. For plutonium, a stable Pu (VI) species was observed. In the case of neptunium, an order-of-magnitude decrease in concentration to a lower steady-state value is being observed.

In the lower pH systems investigated in G-Seep brine, greater overall instability was noted. Total concentrations were stable for uranium at pH 5, plutonium at pH 5 and 7, and neptunium at pH 5. Although total concentrations were stable, there was evidence for reduction in both neptunium and plutonium at pH 5 and 7. The extension of these experiments in time, especially in the case of plutonium, should lead to a lowering of the total concentration. Uranium was stable at pH 5 and unstable at pH 7. The reduction in concentration is presumably due to solubility limitations although redox effects cannot be precluded.

### 3.0 TASK II: REDOX STABILITY OF THE VI OXIDATION STATE IN THE PRESENCE OF ORGANIC CHELATING AGENTS

The objectives of this Task are to establish the effect of organic chelating agents, present in TRU waste, on the redox stability of the An(VI) oxidation state and identify the "stable" oxidation state in the presence of the organic species. Scoping experiments for Np and Pu are in progress and not yet at steady-state. The uranium experiments have not been initiated.

The scoping experiments were performed at near neutral pH (pH of 6-7) at room temperature ( $23 \pm 2$  °C), in high-purity water (HPW), and in the dark. Four experiments were conducted for  $\sim 10^{-4}$  M solutions of each actinide in the following systems: 1) high-purity water (i.e., no organic species), 2) 0.005 M Citrate, 3) 0.005 M oxalate, and 4) 0.0005 M EDTA. The stability of the oxidation state was established by time-dependent absorption spectrometry.

#### 3.1 Summary of Experimental Results

The absorption spectra obtained at various times are given in Appendix C. The observations made, based on these spectral data, are:

##### Neptunium-Organic System

- HPW: Initially, the neptunium was present as predominantly (>95%) Np(VI) with some Np(V) evident. In time, a slow reduction from Np(VI) to Np(V) is taking place. After 30 days, approximately 30% of the Np(VI) initially present was reduced to Np(V). This reduction is presumably due to autoradiolysis.
- Oxalate: In the presence of oxalate, Np(VI) is rapidly reduced to Np(V). Approximately 70% of the Np(VI) is reduced in < 2 h with greater than 95% reduced in < 1 day. After 29 days, the only complex present is the Np(V)-oxalate complex with no evidence for Np(IV) species.
- Citrate: The reduction of Np(VI) proceeds more rapidly in the presence of citrate than in the presence of oxalate. Essentially all the Np(VI) is converted to Np(V)-citrate complexes in < 1.5 h. Over time, two Np(V)-citrate complexes are spectrally evident. The relative concentration of these two species is changing slowly (time frame of days) with the longer-wavelength species growing in as a function of time. After 29 days, although the reduction is complete, the final species distribution is not at steady-state. There is no spectral evidence for Np(IV) species.

EDTA: EDTA, although a factor of ten lower in concentration relative to oxalate and citrate, rapidly reduced Np(VI) to form Np(V)-EDTA complexes. No Np(VI) was evident in solution after 2.3 h. As was the case with citrate, there is evidence for two Np(V) complexes. The shorter wavelength species, evident as a shoulder in the earlier spectra obtained, is converted to a single longer-wavelength species. After 29 days, there is only evidence for one Np(V)-EDTA complex with no evidence for Np(IV) species.

### Plutonium-Organic System

HPW: In the absence of organic complexants, over a period of 10.3 days, the Pu(VI) is stable at concentrations of 0.1 mM.

Oxalate: The presence of oxalate resulted in a slow reduction of Pu(VI)-oxalate to Pu(V)-oxalate. After 10.1 days the system is still not at steady-state and approximately 60% of the Pu(VI) had been reduced. This increase is due to reaction with oxalate. There was no evidence of Pu(IV) species after 10 days.

Citrate: The presence of citrate led to two changes in the Pu(VI)-citrate complex initially present. First, there was a relatively rapid (~ 2-3 day) change in the speciation of the Pu(VI)-citrate complex which was indicated by a blue-shift in the absorption spectrum. Second, there was a slower reduction of Pu(VI)-citrate to a Pu(IV)-citrate complex with only, at most, trace amounts of the Pu(V) complex evident. After 10 days, the system is not at steady state and approximately 70% of the Pu(VI)-citrate complex has been reduced to the Pu(IV) complex.

EDTA: The presence of EDTA, even though a factor of ten lower in concentration than citrate and oxalate, resulted in a rapid reduction to Pu(V)-EDTA complex. There was no evidence of the Pu(VI) species after 0.13 h. The Pu(V)-EDTA complex is not stable and is undergoing a slow reduction to Pu(IV)-EDTA species. The Pu-EDTA system, after 10 days, is not at steady-state with ~35% of the Pu present as a Pu(IV)-EDTA complex and ~65% as the Pu(V)-EDTA complex.



### 3.2 General Observations and Conclusions

The presence of organic chelating agents led to the destabilization of the An (VI) oxidation state for both neptunium and plutonium. Significant differences in rates of reduction were noted for the three organics investigated: oxalate, citrate and EDTA. The actinide-organic systems investigated were not at steady-state so it was not possible to establish the long-term/final oxidation state of the stable actinide complex. Evidence-to-date, however, is consistent with Np(V) and Pu(IV) as the predominant oxidation states in the presence of organics. These data need to be confirmed for the simulated WIPP brines being used for the stability/solubility experiments underway which is the next step in Task II.

There were some similarities in the behavior of the three organic ligands investigated towards neptunium and plutonium. Oxalate, in both cases, led to significantly slower rates of reduction of An(VI) to what appeared to be a single, spectrally distinct, An(V) species. For neptunium, reduction to Np (V) in the presence of both citrate and EDTA was rapid. The resultant An(V) speciation was however more complex as two spectrally distinct species were noted. For plutonium, Pu(VI) was reduced more rapidly by EDTA than citrate. The stability of the An(V) species generated, however, was much greater for EDTA than citrate leading to a slower reduction of the EDTA complex to form a Pu(IV) species. Overall reduction of An(VI) to An(IV) actually proceeded more rapidly for the citrate complex.

**APPENDIX A  
CONCENTRATION DATA FOR THE  
TASK I PU, NP AND U EXPERIMENTS**

The actinide concentration data are tabulated in this appendix. The uranium data is tabulated in Table A.1 and displayed in Figure A.1. These data are based on ICP-MS analyses for total uranium before and after filtration with a 0.2  $\mu$  filter. The neptunium data is tabulated in Table A.2 and displayed in Figure A.2. These data are based on alpha scintillation counting and are being re-analyzed by low-level gamma counting to increase their accuracy. The plutonium data is tabulated in Table A.3 and displayed in Figure A.3. These data were also obtained using alpha scintillation counting of filtered and unfiltered samples.

Table A.1 Concentration Data ( $\pm 15\%$  Uncertainty) for the Uranium Task I Experiments

Experiment Designation	Experiment Description	Time (Days)	*Uranium Concentration in M	
			Unfiltered	0.2 $\mu$ Filtered
U-G7-NC	G-Seep, No Carbonate, pH 7	0	$9.52 \times 10^{-5}$	ND
		14	$3.43 \times 10^{-6}$	$3.00 \times 10^{-6}$
		29	$2.79 \times 10^{-6}$	$2.57 \times 10^{-6}$
		51	$1.91 \times 10^{-6}$	$1.76 \times 10^{-6}$
U-G7	G-Seep, No Carbonate, pH 7	0	$9.52 \times 10^{-5}$	ND
		14	$3.21 \times 10^{-6}$	$3.21 \times 10^{-6}$
		29	$2.79 \times 10^{-6}$	$2.57 \times 10^{-6}$
		51	$2.36 \times 10^{-6}$	$2.36 \times 10^{-6}$
U-G5	G-Seep, No Carbonate pH 5	0	$9.52 \times 10^{-5}$	ND
		14	$9.86 \times 10^{-5}$	$9.21 \times 10^{-5}$
		29	$0.43 \times 10^{-5}$	$9.21 \times 10^{-5}$
		51	$9.86 \times 10^{-5}$	$8.79 \times 10^{-5}$
U-E10-NC	ERDA-6, No Carbonate, pH 10	0	$1.01 \times 10^{-4}$	ND
		14	$2.04 \times 10^{-5}$	$7.29 \times 10^{-7}$
		29	$2.36 \times 10^{-5}$	$1.82 \times 10^{-6}$
		51	$8.79 \times 10^{-5}$	$1.76 \times 10^{-5}$
U-E10	ERDA-6 Carbonate, pH 10	0	$7.84 \times 10^{-5}$	ND
		14	$9.43 \times 10^{-5}$	$9.21 \times 10^{-5}$
		29	$9.21 \times 10^{-5}$	$8.79 \times 10^{-5}$
		51	$9.21 \times 10^{-5}$	$9.21 \times 10^{-5}$
U-E8	ERDA-6 Carbonate pH 8	0	$8.40 \times 10^{-5}$	ND
		14	$5.14 \times 10^{-7}$	$2.36 \times 10^{-7}$
		29	$1.16 \times 10^{-6}$	$1.07 \times 10^{-6}$
		51	$2.61 \times 10^{-7}$	$2.76 \times 10^{-7}$

ND - not determined  
# - the uranium data reported were obtained by Steve Wolf (CMT) using ICP-MS

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Table A.2 Concentration Data ( $\pm 50\%$ ) for the Neptunium Task I Experiments

Experiment Designation	Experiment Description	Time (Days)	Neptunium Concentration in M		
			Unfiltered	0.2 $\mu$ Filtered	10,000 MW $\sim$ 20 nm Filtered
NP-G7-NC	G-Seep, No Carbonate, pH 7	0	$9.3 \times 10^{-5}$	ND	ND
		6	$6.86 \times 10^{-5}$	$6.45 \times 10^{-5}$	ND
		20	$6.27 \times 10^{-5}$	$6.12 \times 10^{-5}$	ND
		61	$6.24 \times 10^{-5}$	$6.04 \times 10^{-5}$	$4.64 \times 10^{-5}$
NP-G7	G-Seep, No Carbonate, pH 7	0	$9.3 \times 10^{-5}$	ND	ND
		6	$1.22 \times 10^{-4}$	$6.65 \times 10^{-5}$	ND
		20	$6.36 \times 10^{-5}$	$6.27 \times 10^{-5}$	ND
		61	$7.85 \times 10^{-5}$	$6.05 \times 10^{-5}$	$5.76 \times 10^{-5}$
NP-G5	G-Seep, No Carbonate pH 5	0	$9.3 \times 10^{-5}$	ND	ND
		6	$9.14 \times 10^{-5}$	$8.54 \times 10^{-5}$	ND
		20	$8.93 \times 10^{-5}$	$8.1 \times 10^{-5}$	ND
		61	$8.92 \times 10^{-5}$	$8.89 \times 10^{-5}$	$8.6 \times 10^{-5}$
NP-E10-NC	ERDA-6, No Carbonate, pH 10	0	$9.3 \times 10^{-5}$	ND	ND
		6	$8.99 \times 10^{-5}$	$4.38 \times 10^{-5}$	ND
		20	$8.36 \times 10^{-6}$	$7.88 \times 10^{-6}$	ND
		61	$8.98 \times 10^{-6}$	$8.86 \times 10^{-6}$	$6.34 \times 10^{-6}$
NP-E10	ERDA-6 Carbonate, pH 10	0	$9.3 \times 10^{-5}$	ND	ND
		6	$9.06 \times 10^{-5}$	$9.17 \times 10^{-5}$	ND
		20	$9.25 \times 10^{-5}$	$9.1 \times 10^{-5}$	ND
		61	$8.58 \times 10^{-5}$	$8.51 \times 10^{-5}$	$9.51 \times 10^{-5}$
NP-E8	ERDA-6 Carbonate pH 8	0	$9.3 \times 10^{-5}$	ND	ND
		6	$6.92 \times 10^{-5}$	$6.93 \times 10^{-5}$	ND
		20	$9.18 \times 10^{-5}$	$9.21 \times 10^{-5}$	ND
		61	$9.36 \times 10^{-5}$	$9.18 \times 10^{-5}$	$9.86 \times 10^{-5}$

ND - not determined

Table A.3 Concentration Data ( $\pm 10\%$ ) for the Plutonium Task I Experiments

Experiment Designation	Experiment Description	Time (Days)	Plutonium Concentration in M		
			Unfiltered	0.2 $\mu$ Filtered	10,000 MW ~ 20 nm Filtered
PU-G7-NC	G-Seep, No Carbonate, pH 7	0	$9 \times 10^{-5}$	ND	ND
		8	$8.31 \times 10^{-5}$	$8 \times 10^{-5}$	ND
		22	$8.25 \times 10^{-5}$	$8.17 \times 10^{-5}$	ND
		58	$7.59 \times 10^{-5}$	$7.7 \times 10^{-5}$	$7.35 \times 10^{-5}$
PU-G7	G-Seep, No Carbonate, pH 7	0	$9 \times 10^{-5}$	ND	ND
		8	$8.88 \times 10^{-5}$	$8.78 \times 10^{-5}$	ND
		22	$9.15 \times 10^{-5}$	$8.95 \times 10^{-5}$	ND
		58	$8.47 \times 10^{-5}$	$8.25 \times 10^{-5}$	$8.53 \times 10^{-5}$
PU-G5	G-Seep, No Carbonate pH 5	0	$9 \times 10^{-5}$	ND	ND
		8	$8.62 \times 10^{-5}$	$8.59 \times 10^{-5}$	ND
		22	$1.12 \times 10^{-4}$	$1.12 \times 10^{-4}$	ND
		58	$5.06 \times 10^{-5}$	$5 \times 10^{-5}$	$3.04 \times 10^{-5}$
PU-E10-NC	ERDA-6, No Carbonate, pH 10	0	$9 \times 10^{-5}$	ND	ND
		8	$8.26 \times 10^{-5}$	$7.94 \times 10^{-5}$	ND
		22	$8.17 \times 10^{-5}$	$8.17 \times 10^{-5}$	ND
		58	$7.56 \times 10^{-5}$	$7.51 \times 10^{-5}$	$7.19 \times 10^{-5}$
PU-E10	ERDA-6 Carbonate, pH 10	0	$9 \times 10^{-5}$	ND	ND
		8	$8 \times 10^{-5}$	$7.95 \times 10^{-5}$	ND
		22	$7.81 \times 10^{-5}$	$7.63 \times 10^{-5}$	ND
		58	$7.26 \times 10^{-5}$	$7.26 \times 10^{-5}$	$7.38 \times 10^{-5}$
PU-E8	ERDA-6 Carbonate pH 8	0	$9 \times 10^{-5}$	ND	ND
		8	$8.25 \times 10^{-5}$	$8.25 \times 10^{-5}$	ND
		22	$8.14 \times 10^{-5}$	$8.14 \times 10^{-5}$	ND
		58	$8.03 \times 10^{-5}$	$7.71 \times 10^{-5}$	$7.65 \times 10^{-5}$

ND - not determined

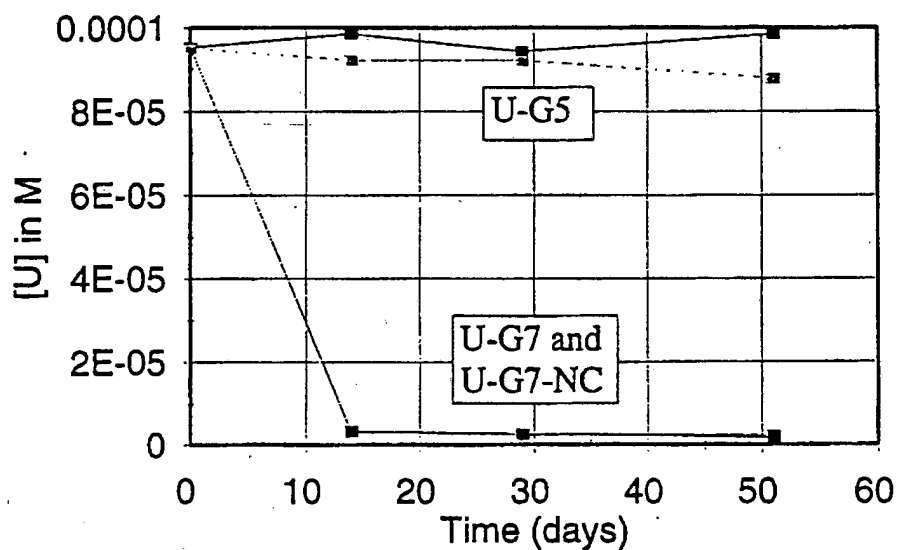
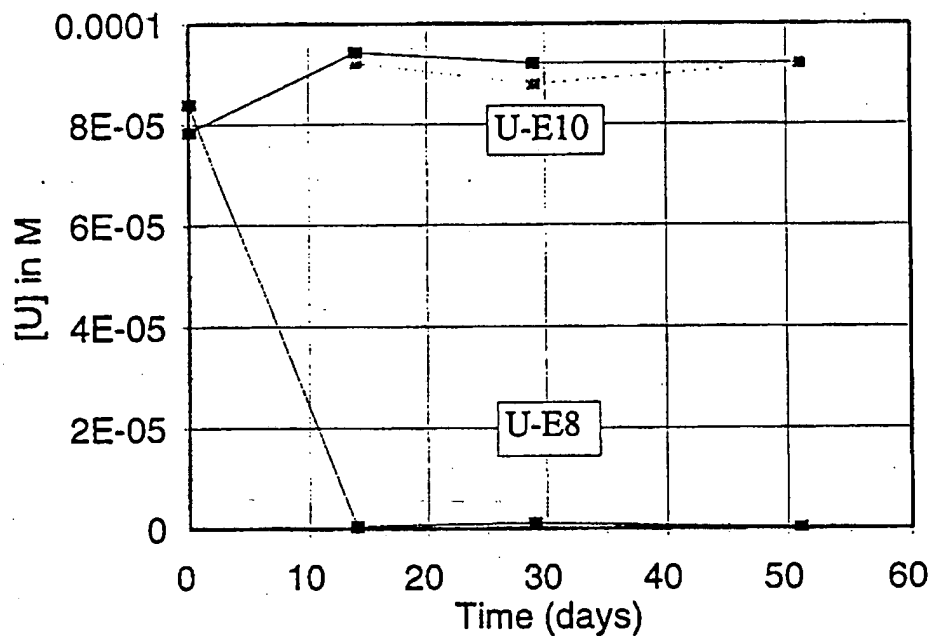


Figure A.1 Total and 0.2  $\mu$ -filtered concentration data as a function of time for uranium in ERDA-6 (a) and G-Seep (b) brine.

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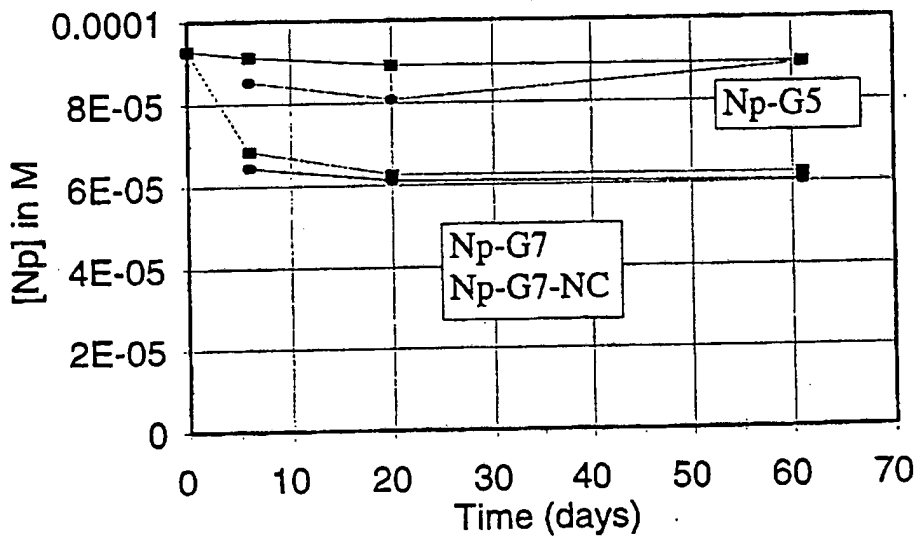
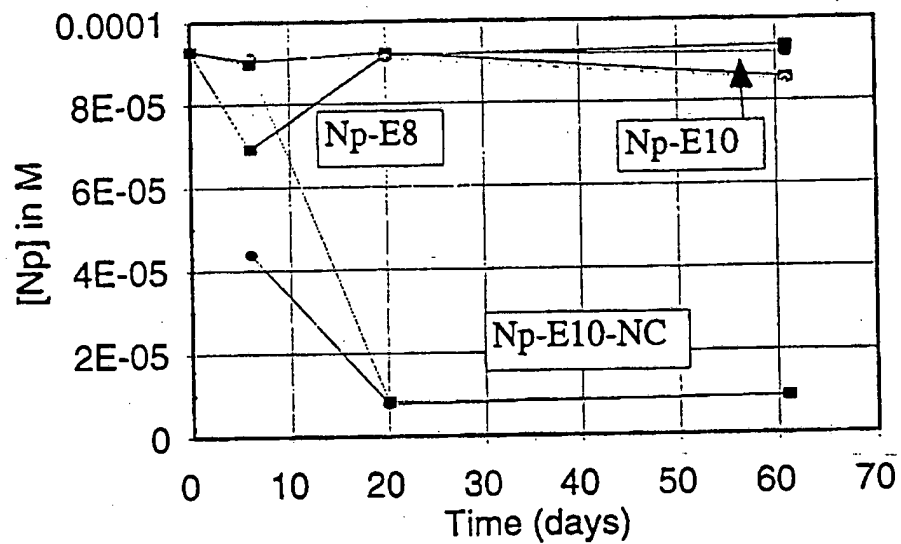


Figure A.2 Total and 0.2  $\mu$  -filtered concentration data as a function of time for neptunium in ERDA-6 (a) and G-Seep (b) brine.

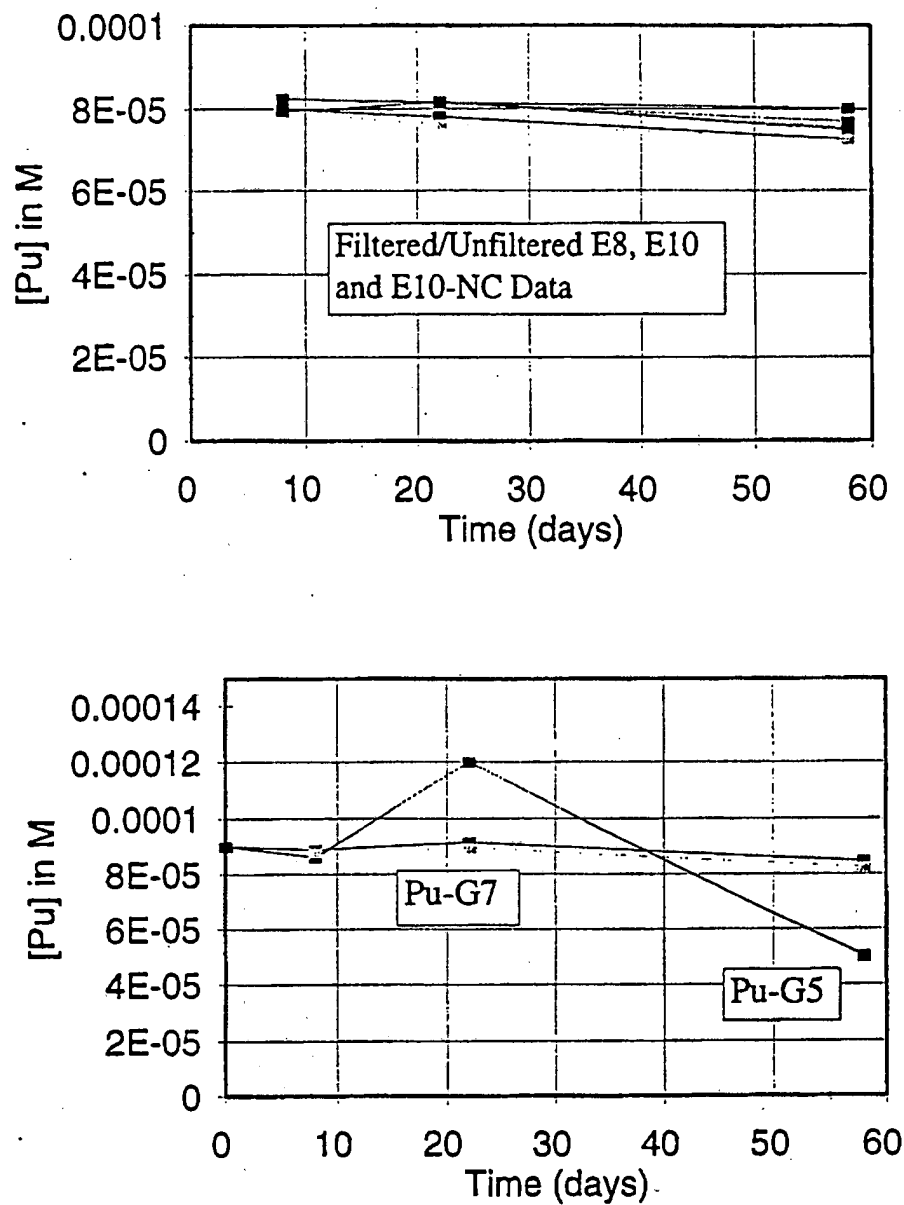


Figure A.3 Total and 0.2  $\mu$  -filtered concentration data as a function of time for plutonium in ERDA-6 (a) and G-Seep (b) brine.

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### APPENDIX B

## ABSORPTION SPECTRA OF URANIUM, NEPTUNIUM AND PLUTONIUM IN THE TASK I EXPERIMENTS

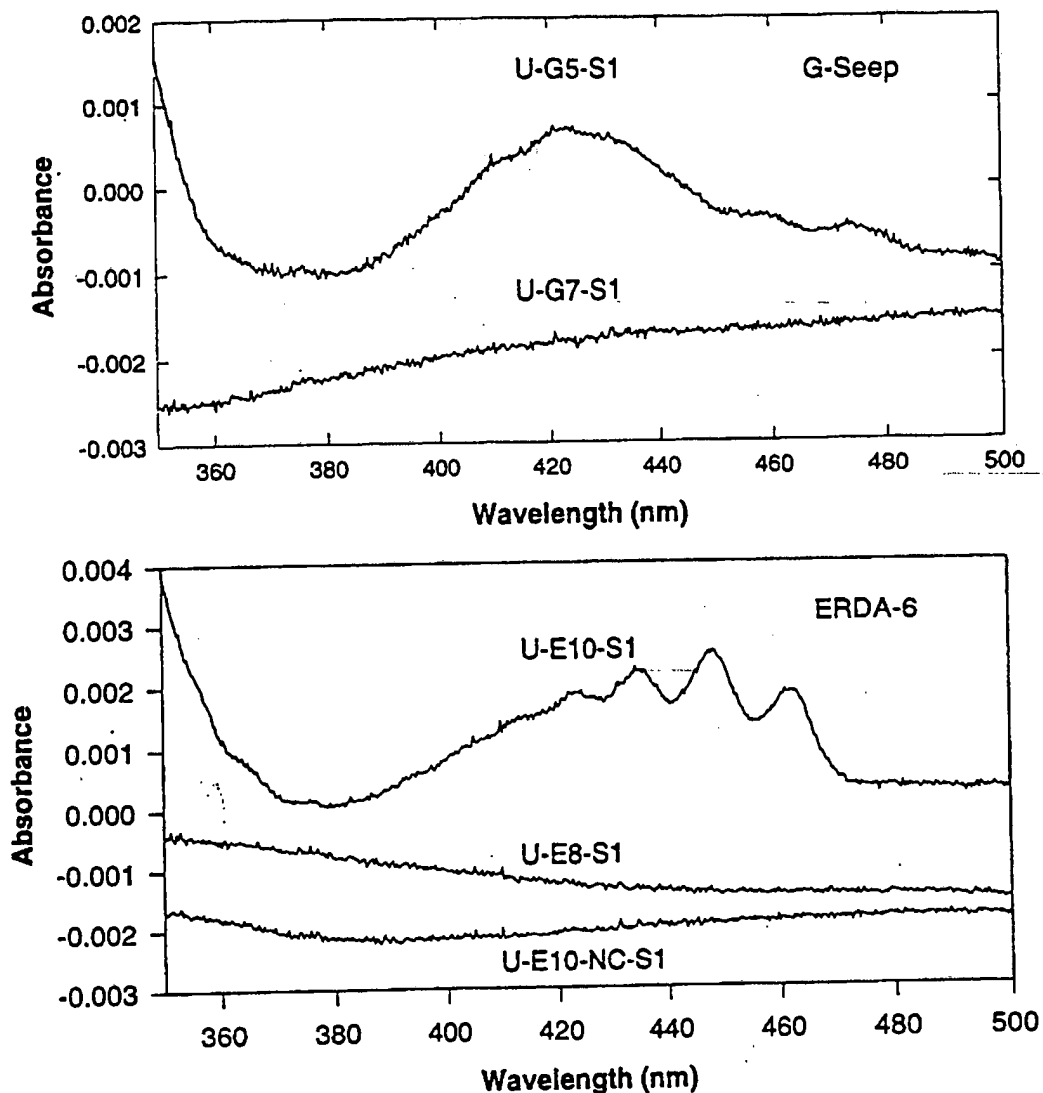


Figure B.1 Absorption spectra for uranium in ERDA-6 (a) and G-Seep (b) brine at 13 days.

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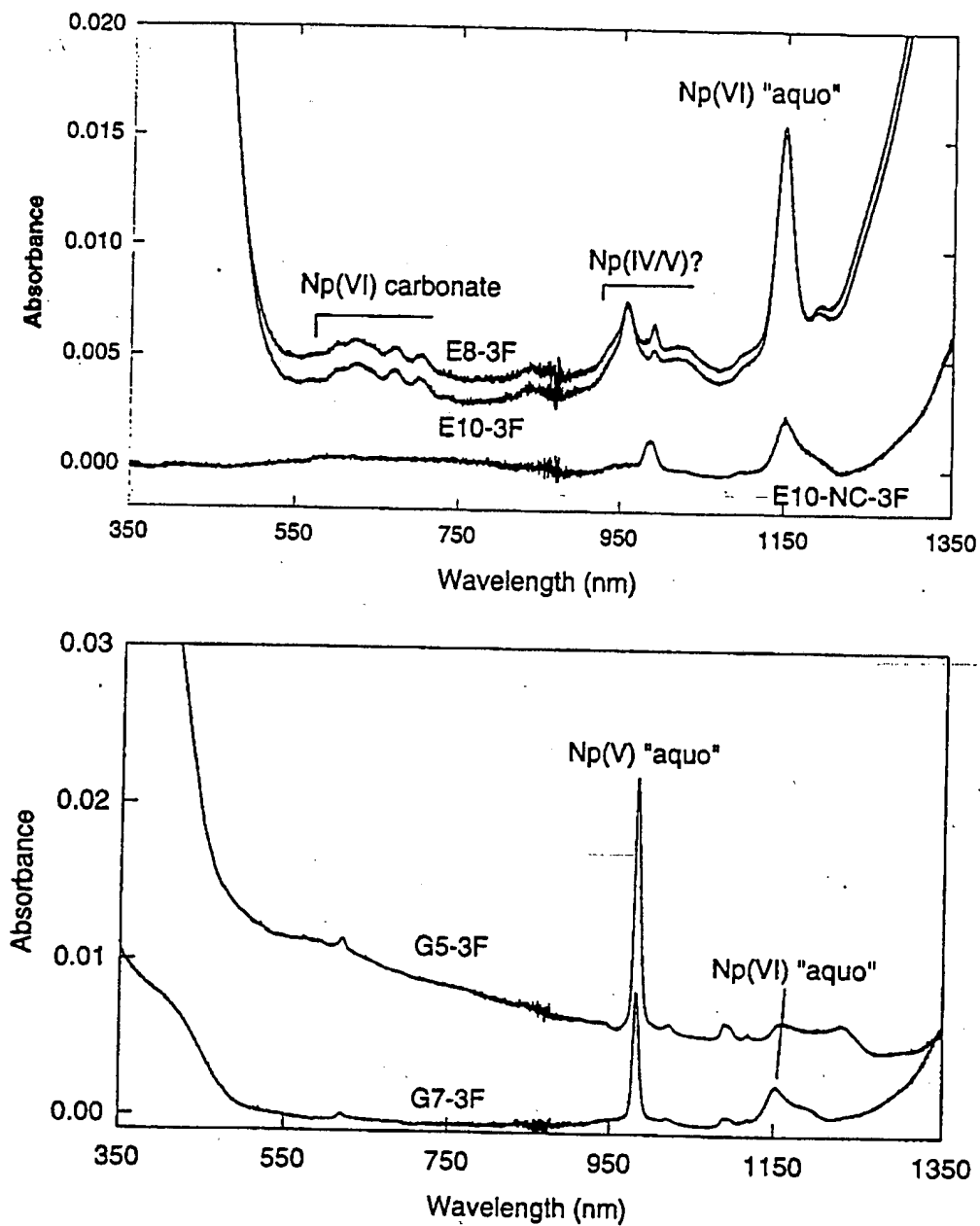


Figure B.2 Absorption spectra for neptunium in ERDA-6 (a) and G-Seep (b) brine at 60 days.

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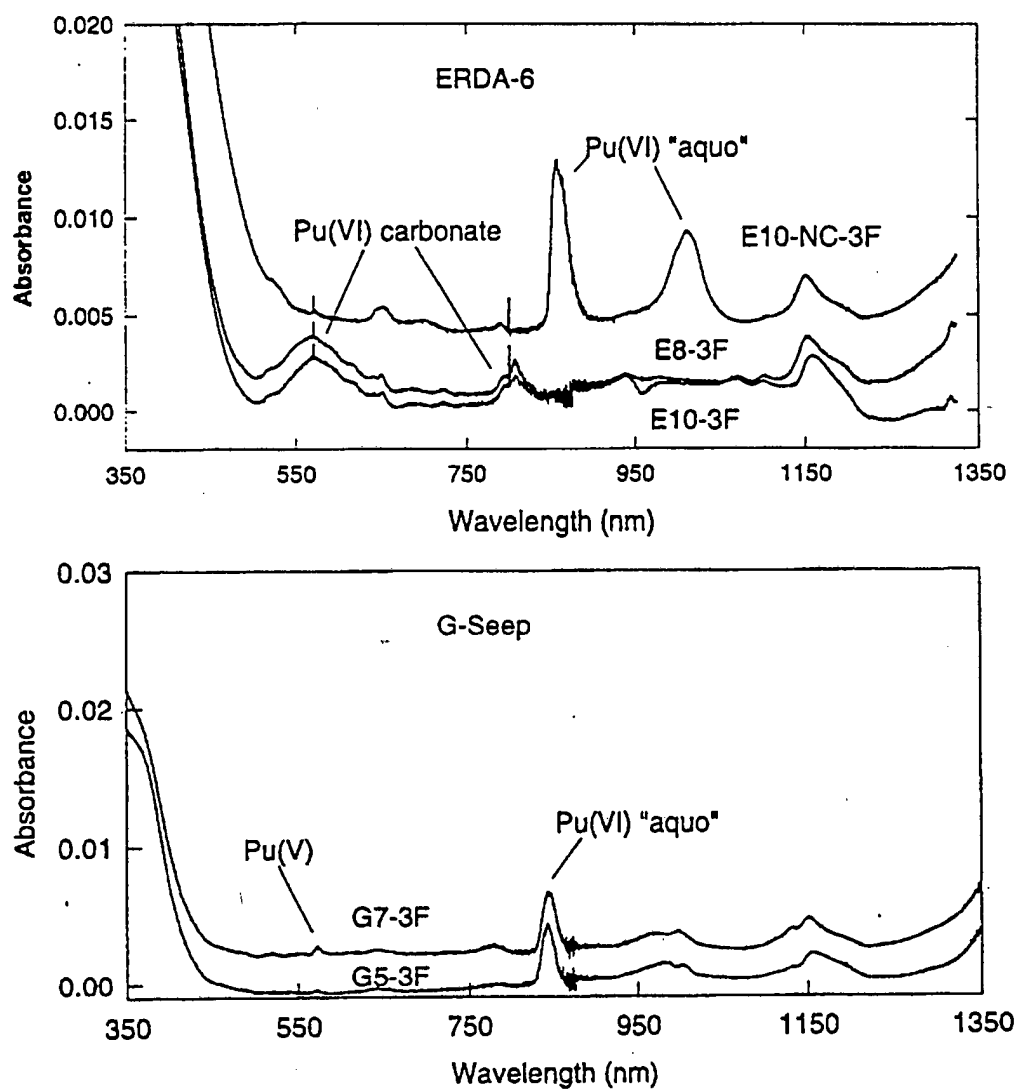


Figure B.3 Absorption spectra for plutonium in ERDA-6 (a) and G-Seep (b) brine at 58 days.

Information Only

### APPENDIX C

#### TIME-DEPENDENT ABSORPTION SPECTRA FOR THE TASK II ORGANIC INTERACTION EXPERIMENTS WITH NEPTUNIUM AND PLUTONIUM

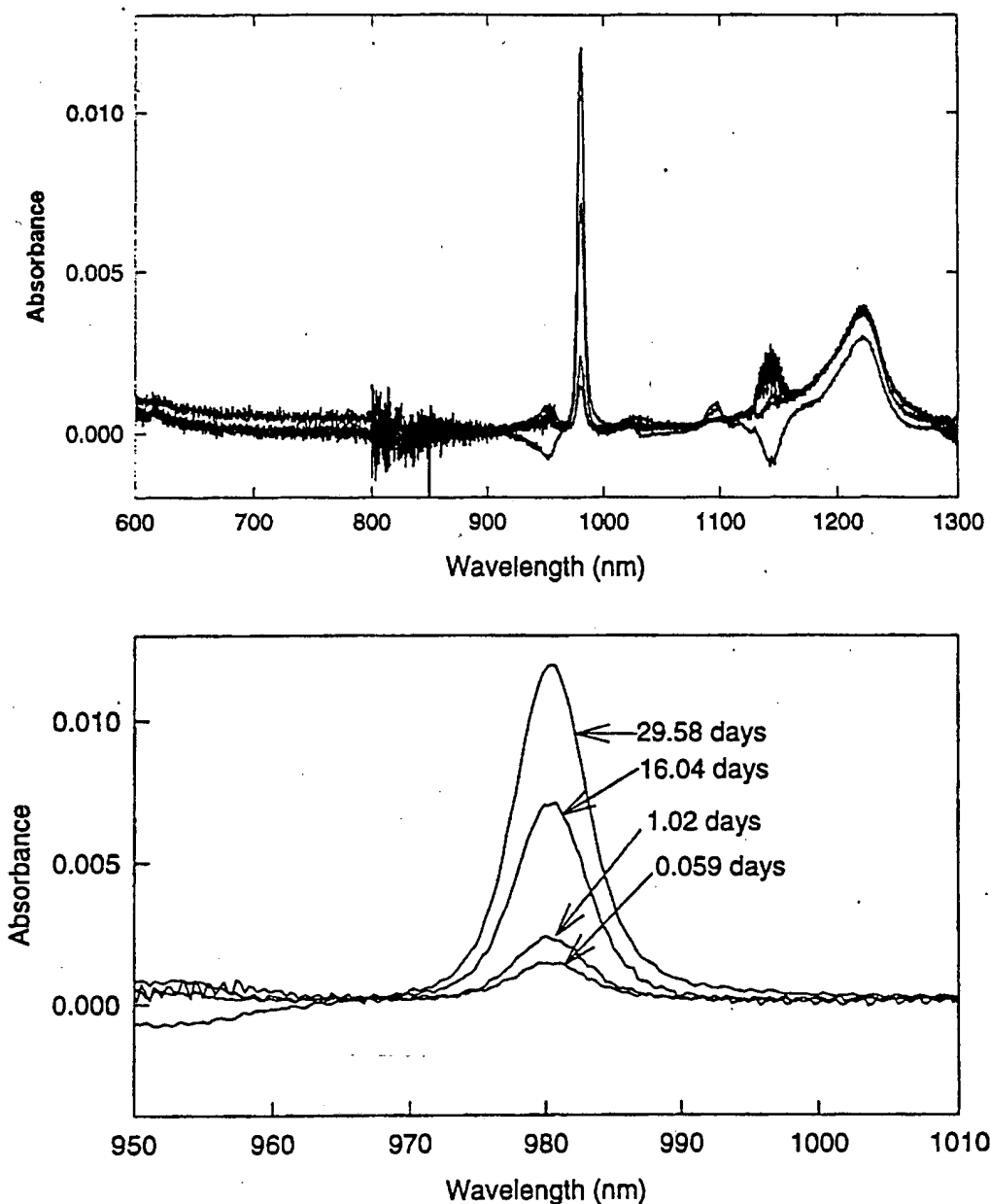


Figure C.1 Time-dependent absorption spectra for ~ 0.1 mM neptunium (VI) in high-purity water.

Information Only

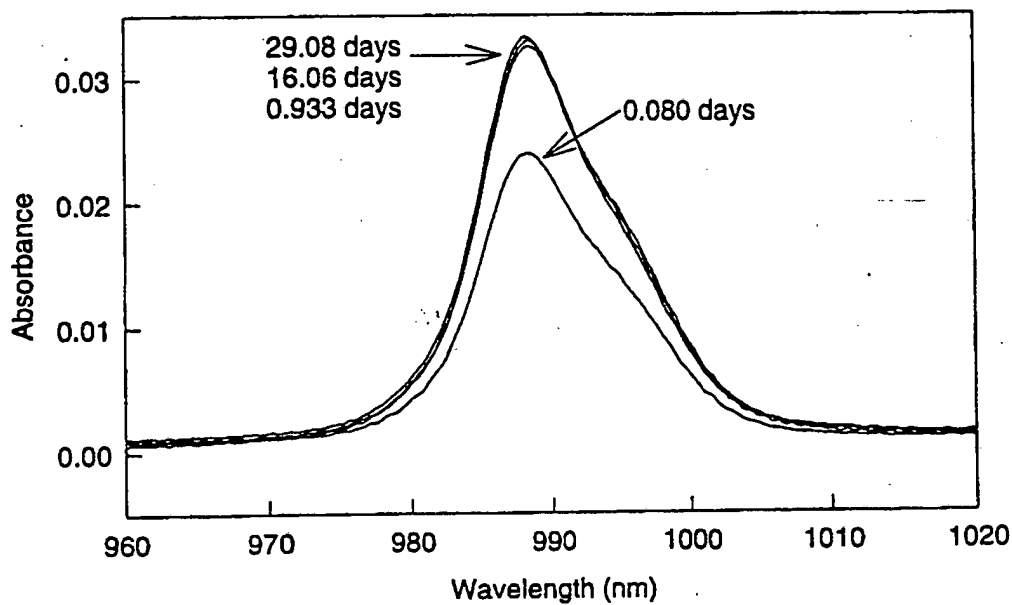
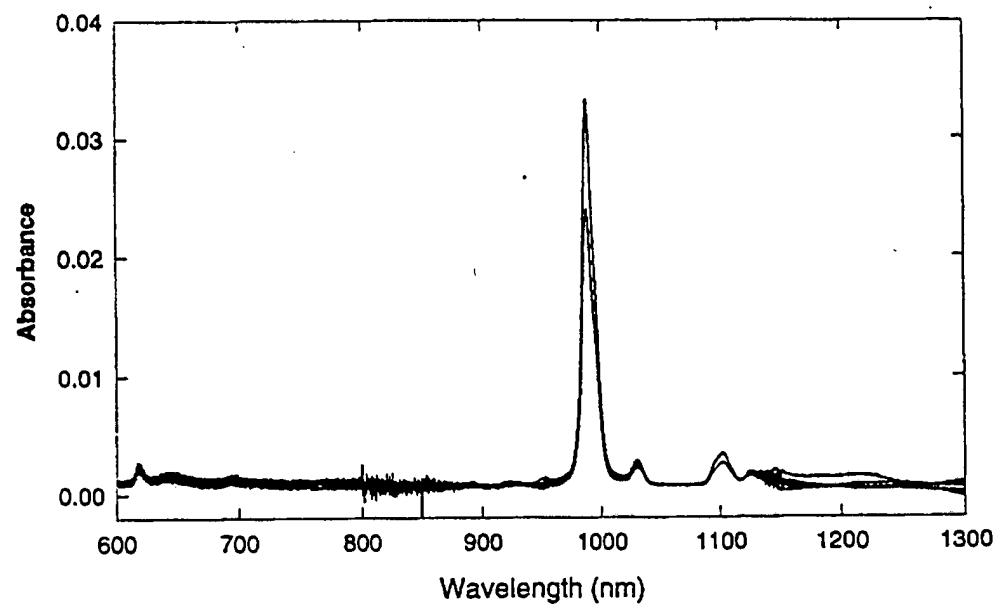


Figure C.2 Time-dependent absorption spectra for 0.1 mM neptunium (VI) in the presence of 0.005 M oxalic acid.

Information Only

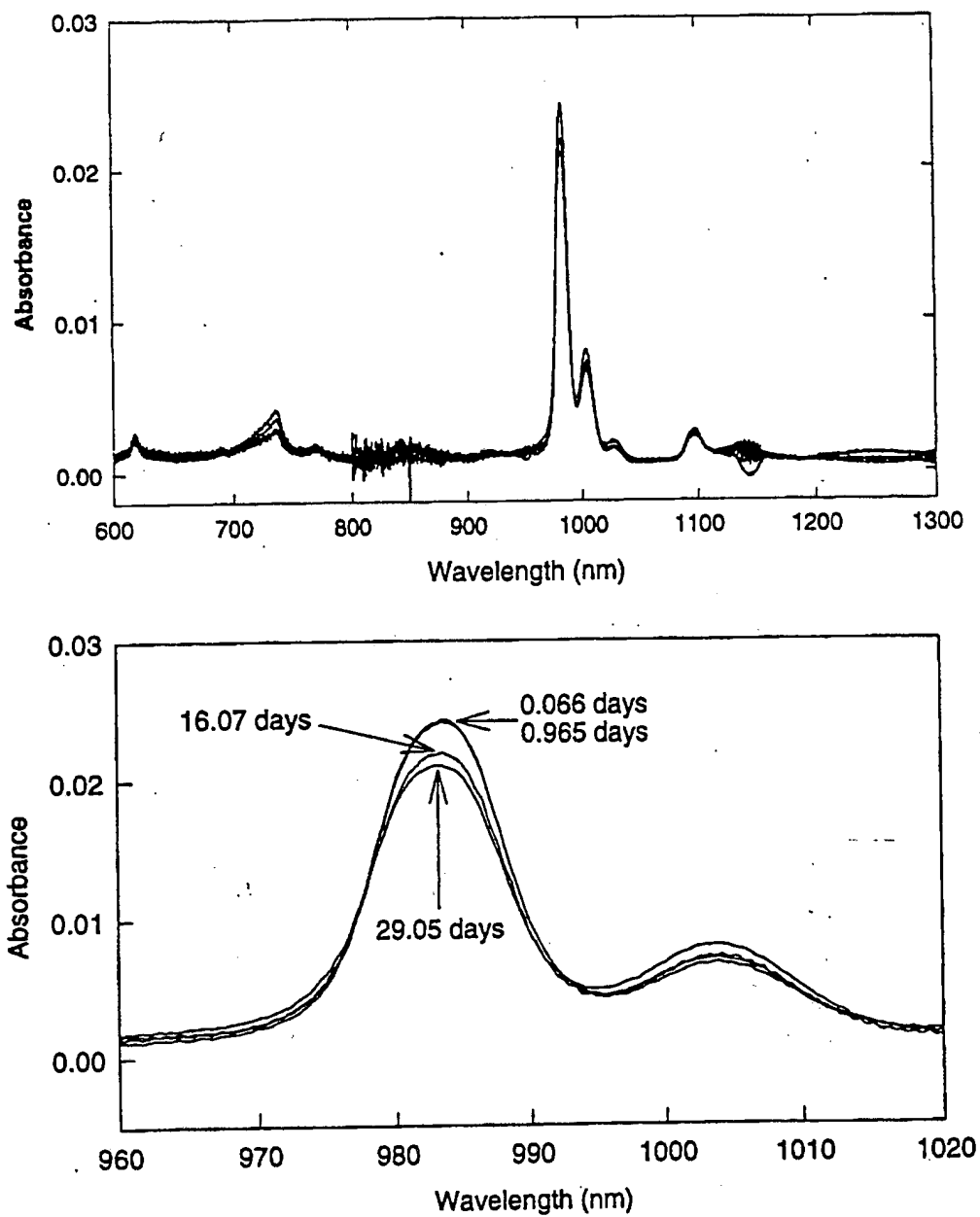


Figure C.3 Time-dependent absorption spectra for 0.1 mM neptunium (VI) in the presence of 0.005 M citric acid.

Information Only

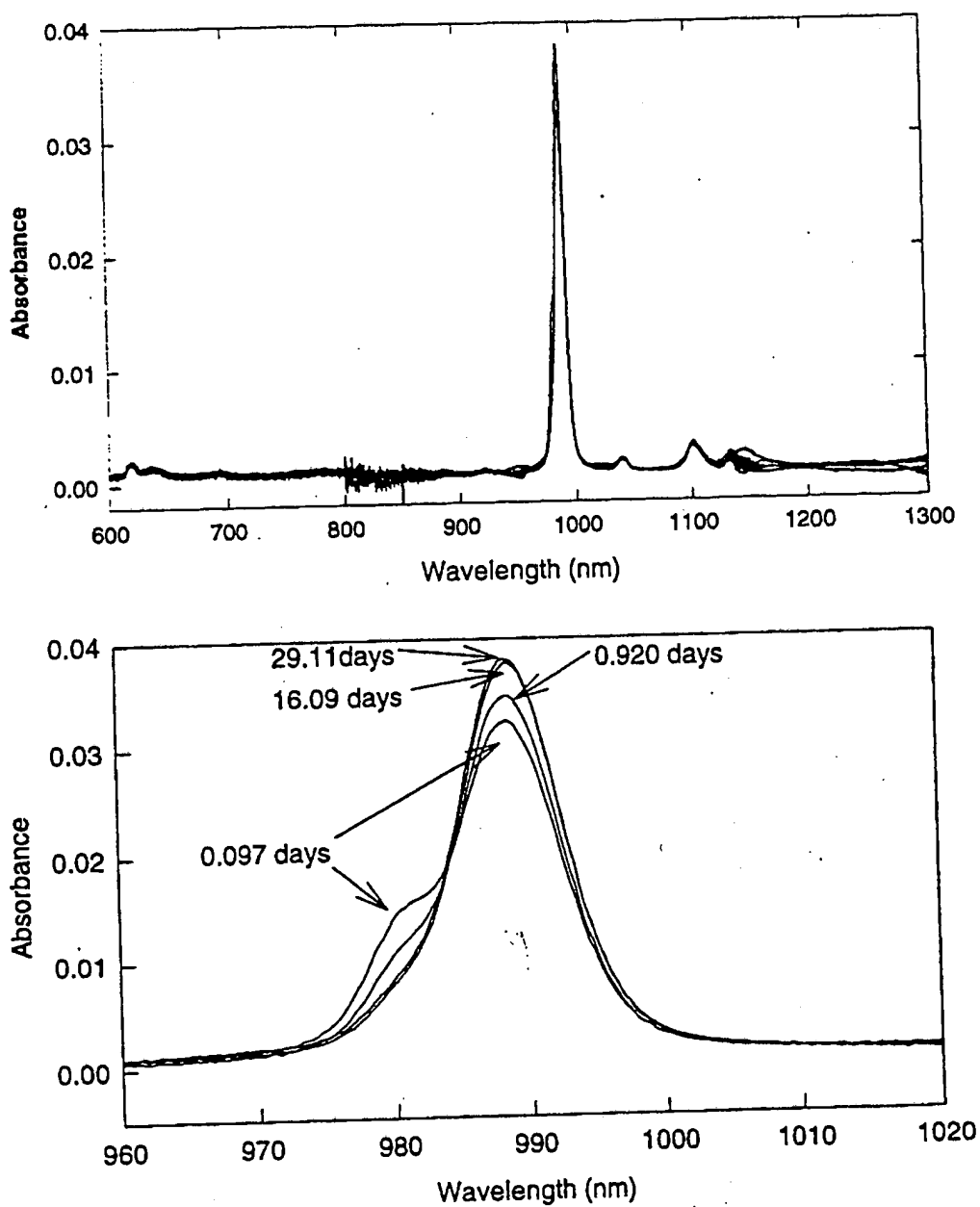


Figure C.4 Time-dependent absorption spectra for 0.1 mM neptunium (VI) in the presence of 0.0005 M EDTA.

Information Only

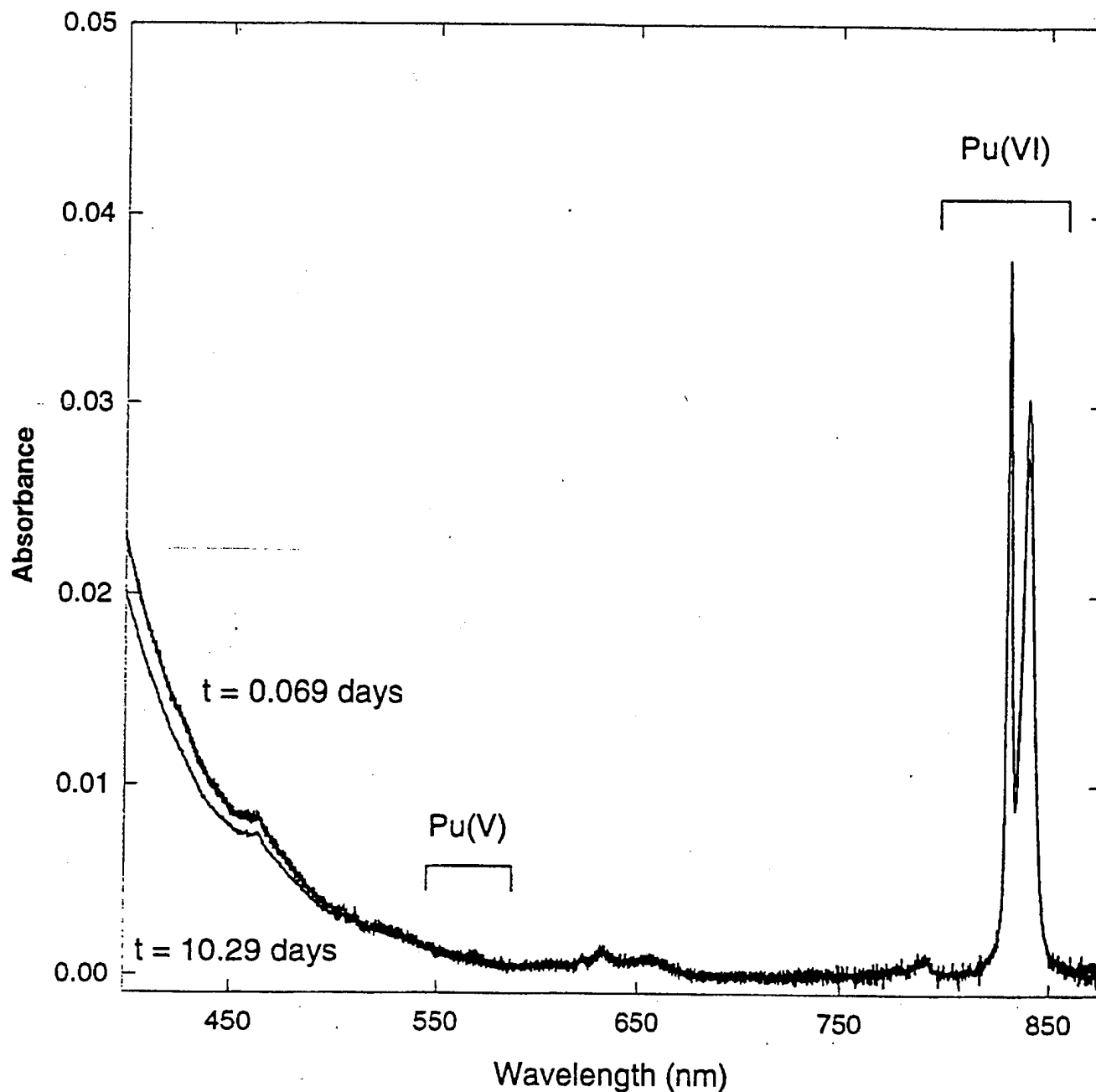


Figure C.5 Time-dependent absorption spectra for  $\sim 0.1$  mM plutonium (VI) in high-purity water. No change in oxidation state is being noted.

Information Only



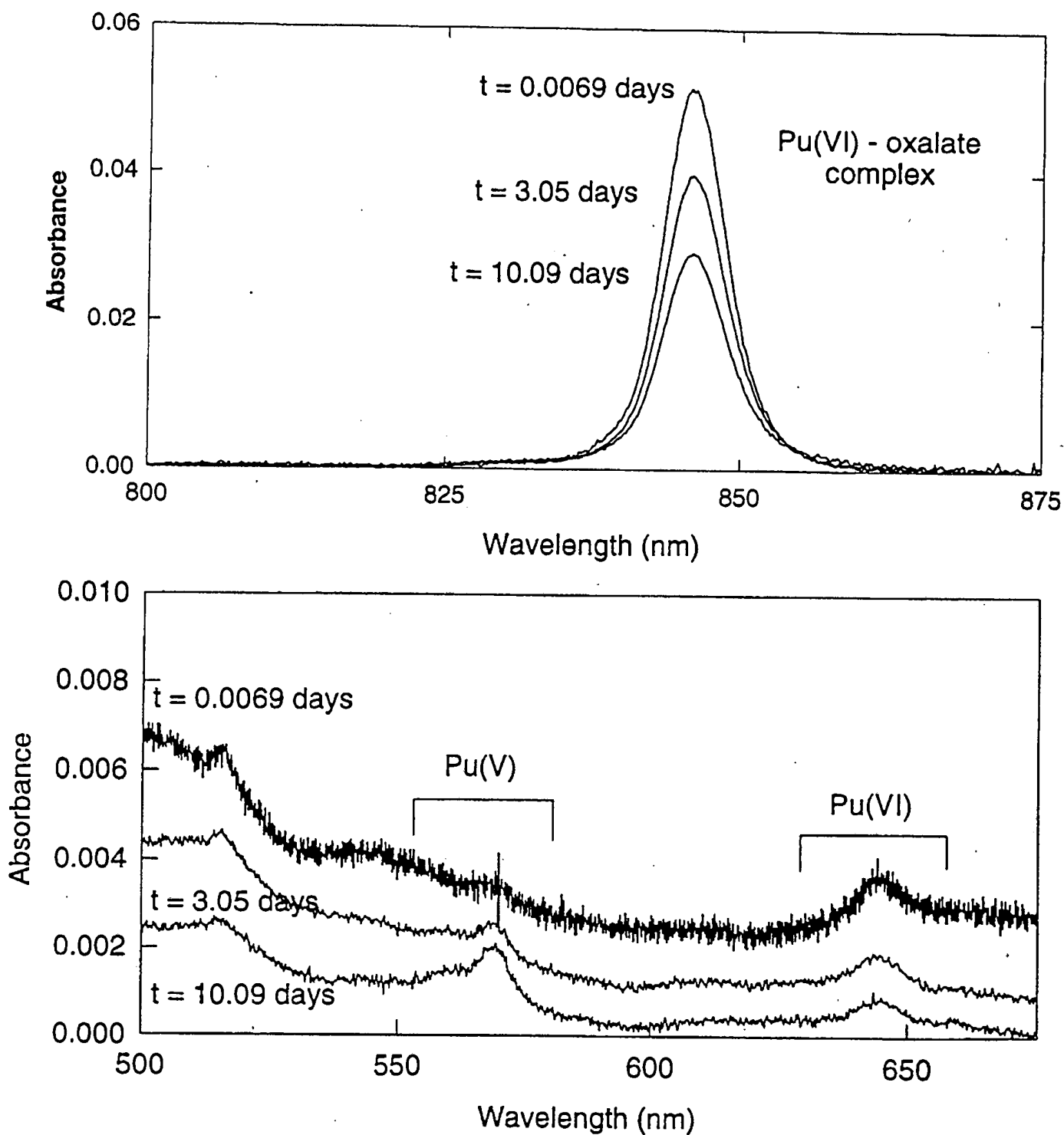


Figure C.6 Time-dependent absorption spectra for 0.1 mM plutonium (VI) in the presence of 0.005 M oxalic acid.

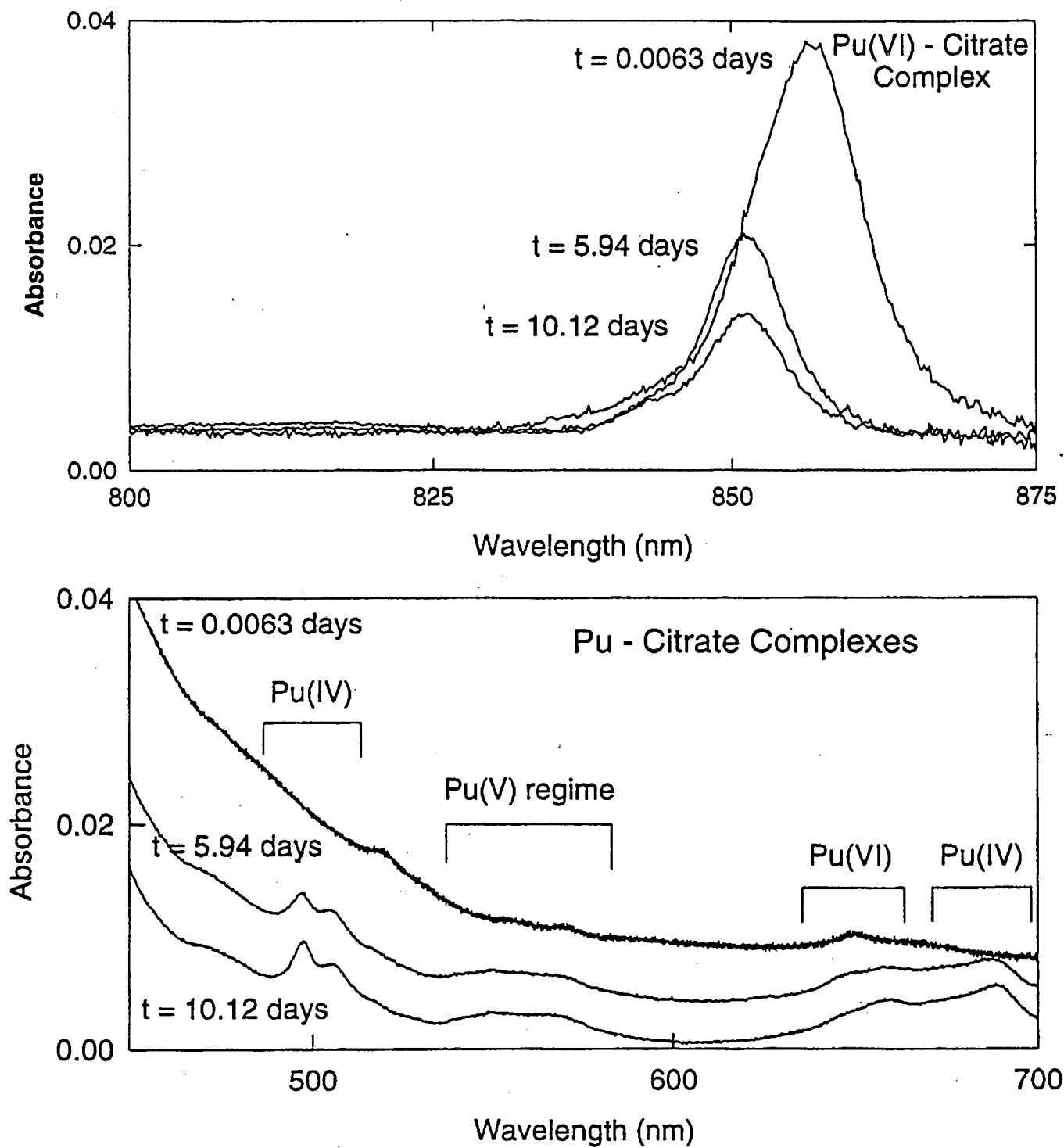


Figure C.7 Time-dependent absorption spectra for 0.1 mM plutonium (VI) in the presence of 0.005 M citric acid.

Sandia National Laboratories

Albuquerque, New Mexico 87185-1341

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date: 3/15/96

to: Richard V. Bynum

*Robert C Moore*

from: Robert C. Moore, Dept. 3748, MS-1341, 848-0146

subject: Actinide Stability/Solubility Program Milestone AC081

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The attached report entitled "Actinide Stability/Solubility in Simulated WIPP Brines Project: Interim Report" is documentation of milestone AC081 WBS number 1.1.1.1.4 (Radioactive Disposal Room Chemistry).

The report indicates some of the systems have not reached steady-state conditions. These experiments will continue until steady-state is reached.

*Exceptional Service in the National Interest*

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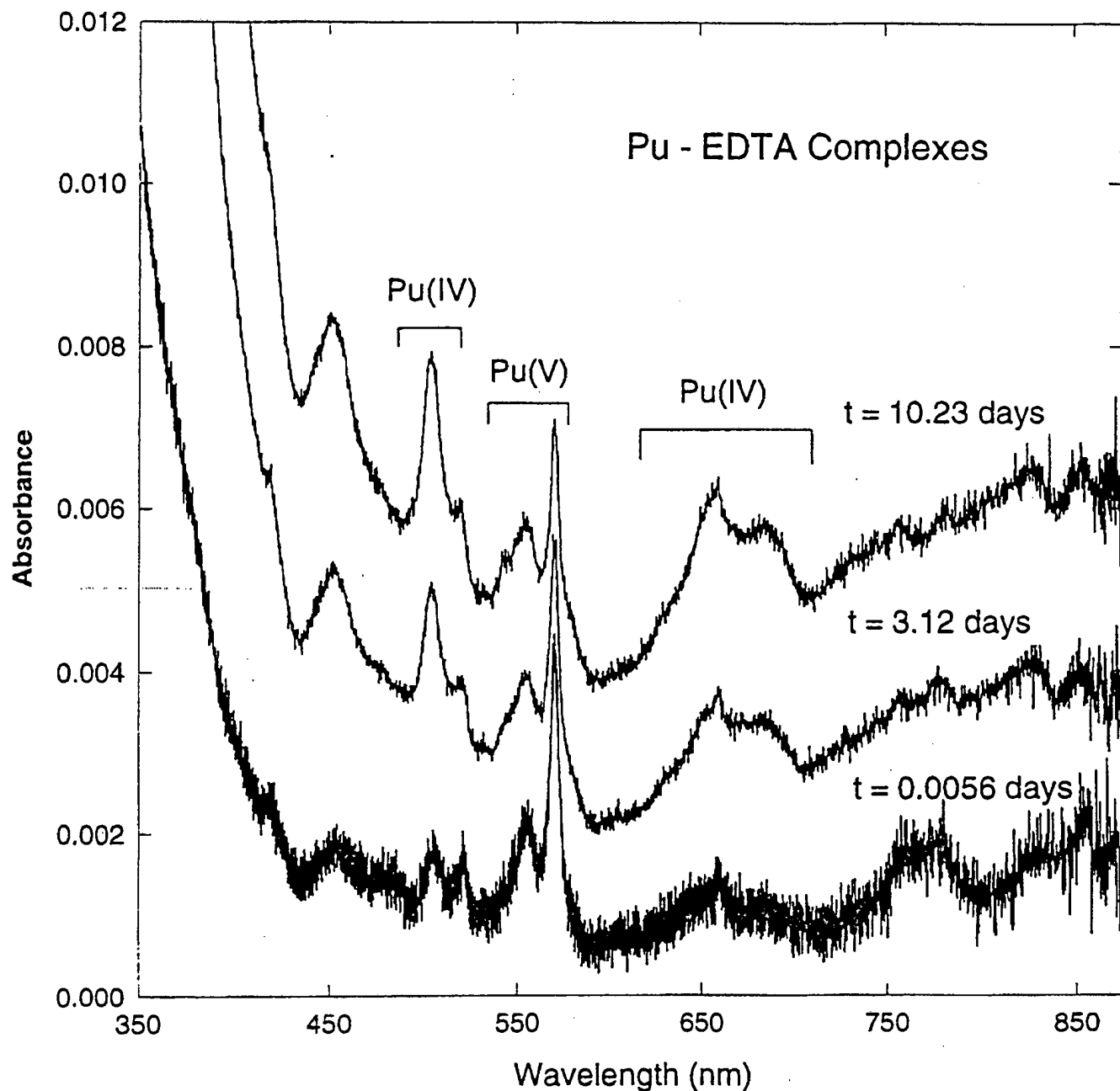


Figure C.8 Time-dependent absorption spectra for 0.1 mM plutonium (VI) in the presence of 0.0005 M EDTA.

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