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Plutonium Solubility Peer Review Report

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Prepared for DOE Savannah River Operations Office (DOE-SR)

Kirk Cantrell, David L. Clark, David R. Janecky, John Psaras, and Wolfgang Runde

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PURPOSE: The purpose of the Plutonium Solubility Peer Review was to obtain independent, expert technical advice related to the evaluated radiological risk posed by residual plutonium in Tank 18 in the F Tank Farm at Savannah River Site. External independent reviewers included Kirk Cantrell (PNNL), David L. Clark (LANL), David R. Janecky (LANL), Wolfgang Runde (LANL), and John Psaras (LANL contractor).

SCOPE: Two days of briefings and discussions were held at the Savannah River Site (SRS) on November 9 and 10, 2011. The review focused on the evaluation processes for risk posed by residual plutonium, particularly ^{239}Pu , remaining in Tank 18 planned for closure at SRS. This panel evaluated the solubility and modeling assumptions (waste release and transport) used in the Performance Assessment for F-Tank Farm at the Savannah River Site (PA) and Tank 18/Tank 19 Special Analysis for the Performance Assessment for the F-Tank Farm at the SRS; the technical basis for these assumptions and the body of knowledge available (Pu chemistry, solubility, sorption, leachability, etc.) to expand the technical basis for Pu modeling assumptions; and an evaluation of the contractor's plans for performing Tank 18 residual leachability testing.

Specific questions to be addressed by the peer-review included 1) Are the assumptions and technical basis for modeling plutonium release from tank residual waste reasonable and technically justified? 2) Are there any recommended changes to the plutonium modeling assumptions? 3) Is there additional evidence associated with plutonium chemistry or solubility to support, contradict or modify the assumptions in the risk analysis? and 4) Are there any recommendations related to the contractor's short term experiments planned for actual residual Tank 18 waste in terms of scope, methodologies, facilities, researchers, etc?

Executive Summary

During the Plutonium Solubility Peer Review, the review panel was presented with information on the plutonium solubility assumptions and calculations that were used to perform an assessment of Tank 18. Reviewers identified opportunities for improvement in nearly all areas of interest including assumptions for plutonium solubility, choice of database, adsorption data, geochemical parameters, conceptual modeling assumptions, quality assurance, and lack of internal peer-review. The current geochemical modeling based on Geochemist's Workbench is based on assumptions that are either not validated by experimental studies or by evaluation of peer-reviewed data available in the open literature. This deficiency can be readily rectified by using an internally consistent, internationally accepted thermodynamic database, modifying assumptions for the conceptual model and providing some validation to support critical assumptions will produce a much more defensible case that is based on current scientific knowledge.

The peer reviewers offer recommendations in five main areas:

- 1. Assumptions and technical basis for modeling plutonium:** Plutonium in residual tank waste is assumed to be co-precipitated with iron minerals in Region II, while solubility in Region III is assumed to be controlled by PuOH_4 . These assumptions are not consistent with current scientific understanding or supported by experimental data. It is recommended that the contractor employ a more technically valid model using amorphous $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ (am) as the solubility controlling phase.
- 2. Conceptual modeling:** The plutonium phase solubility calculations use an internally inconsistent and out-of-date thermodynamic database. It is recommended that a properly vetted, internationally-accepted thermodynamic database (such as the NEA-TDB) be used to calculate plutonium solubility¹. Modeling plutonium adsorption in the vadose zone currently employs K_d values measured under typical ambient (mildly acidic) conditions. The decomposing grout would likely result in a plume below the tank with a much higher pH (>8) than the typical acidic conditions of SRS soils. It is recommended that plutonium adsorption on decomposing grout material and sediments in the vadose zone are calculated using readily available literature data (K_d values) that are applicable to the relevant conditions. Important geochemical parameters need to be properly evaluated and considered, in specific the oxygen levels in infiltration water contacting

¹ As an alternative to directly building a data file for use with Geochemists Workbench from the NEA data, there are files available that can be validated against the NEA-TDB files and published experimental data. For example, JAEA developed geochemical data files in GWB format with the latest update in 2010 which is located at <http://migrationdb.jaea.go.jp/english.html>

the waste can be expected to be much lower while carbon dioxide partial pressures may be much higher. Slow carbonation of the grout as a result of diffusion of carbon dioxide into the grout with water seems like a low probability scenario. Fracturing of the grout with preferential flow through the cracks would appear to be a much more likely scenario and should therefore be evaluated.

- 3. *Experimental verification:*** The assumptions need to be validated and verified where feasible. It is recommended that the residual solid waste be characterized using advanced spectroscopic techniques. Simplistic leach and solubility tests will provide further support of plutonium and grout chemistries assumptions used for modeling.
- 4. *Quality Assurance/Quality Control:*** The review team saw no evidence of a credible quality assurance program for geochemical modeling during their on-site review. A QA program is of high importance for convincing NRC, peer reviewers, and other entities that SRS has a robust evaluation process for potential release scenarios so that Tank 18 and other tanks at SRS can be safely closed.
- 5. *Communications:*** The reviewers found a lack of coordination and project integration across lines and contractors. This led to an apparent lack of engagement of plutonium chemists in the selection of thermodynamic data or peer review of the products prior to release of the reports. Management at all levels need to be vigilant and questioning the assumptions and conclusions of reports that carry multiple management level signatures prior to being issued, otherwise loss of credibility can have long lasting and painful consequences.

As our recommendations to perform revised geochemistry calculations are all computational studies, it is not envisioned that they should take a long time to complete. Samples of residual tank waste solids can be obtained for leaching and spectroscopic study and archived prior to commencing with tank grouting activities, such that the experimental verification of the modeling can take place while grouting activities are underway. This offers a reasonable path forward to a scientifically defensible closure model while allowing tank closure activities to continue in parallel.

Peer review observations and recommendations

1. Assumptions and technical basis for modeling plutonium

Plutonium solubility and speciation: The fate and transport of plutonium is governed by the solubility of its compounds in groundwater and surface waters, the tendency of plutonium compounds to be adsorbed onto mineral phases in soil particles, and by

processes that cause the colloidal forms of plutonium to be filtered by the soil or rock matrices, or to aggregate, adsorb and settle during transport.

Extensive field observations and research have been conducted internationally on the environmental behavior of actinide elements in diverse sets of environments over the past 30-40 years.^{2,3} This research has provided a good base for understanding the major types of species and their transport mechanisms in soils and natural waters. In natural waters plutonium solubility is limited by the formation of amorphous $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ (am) or polycrystalline PuO_2 , with aqueous $\text{Pu}(\text{OH})_4$ (aq) as the predominant solution species above the solid for a wide range of concentrations. The Nuclear Energy Agency (NEA) has performed an extensive peer-review of this literature and made recommendations for accepted values of solubility products (K_{sp}) for solubility-controlling solids, and formation constants for important aqueous species of plutonium.^{4,5} Literature values for the solubility of plutonium show a wide range of concentrations. It is now well known that when $\text{Pu}(\text{IV})$ concentrations exceed the solubility of amorphous $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ (am), $\text{Pu}(\text{IV})$ colloids are the predominant species in solution. The chemical form of freshly precipitated or aged $\text{Pu}(\text{IV})$ phases is not clear, and they have been referred to as either amorphous hydroxides $\text{Pu}(\text{OH})_4$ (am) or amorphous hydrous oxides $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ (am). Many earlier estimates of $\text{Pu}(\text{IV})$ solubility have been rejected as having been contaminated with colloids or the presence of other Pu oxidation states and/or complexes in solution.⁶ The currently accepted NEA solubility estimate puts an upper limit on the amount of dissolved (i.e. ionic/molecular) Pu that can be present, even if higher oxidation states such as $\text{Pu}(\text{V})$ or $\text{Pu}(\text{VI})$ are the more stable solution forms. Moreover, in many natural waters, extremely low Pu concentrations may be traced to sorption processes.² Sorption of hydrolyzed $\text{Pu}(\text{IV})$ in natural water on mineral surfaces and surfaces coated with organic material is accountable for the very low observed concentrations of dissolved Pu even in the absence of $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ (am) or PuO_2 (cr). The strong tendency of $\text{Pu}(\text{OH})_4$ (aq) to sorb on surfaces is a dominant and often controlling feature in plutonium geochemistry. Both solubility and sorption must be considered.

² Clark, D. L.; Hecker, S. S.; Jarvinen, G. D.; Neu, M. P.; "Chapter 7, Plutonium" in *The Chemistry of the Actinides and Transactinides*, 3rd Ed., Morss, L. R.; Edelstein, N. M.; Fuger, J., Eds. **2006**, Springer, New York, 813-1264.

³ Runde, W.; Neu, M. P.; "Chapter 32, Actinides in the Geosphere" in *The Chemistry of the Actinides and Transactinides*, 4th Ed., Morss, L. R.; Edelstein, N. M.; Fuger, J., Eds. **2010**, Springer, New York, 3475-3594.

⁴ Lemire, R. J.; Fuger, J.; Nitsche, H.; Potter, P.; Rand, M. H.; Rydberg, J.; Spahiu, K.; Sullivan, J. C.; Ullman, W. J.; Vitorge, P.; Wanner, H., *Chemical Thermodynamics of Neptunium and Plutonium*, Vol. 4. North-Holland, Amsterdam (**2001**).

⁵ Guillaumont, R.; Fanghänel, T.; Fuger, J.; Grenthe, I.; Neck, V.; Palmer, D. A.; Rand, M. H., *Update of Chemical Thermodynamics of Uranium, Neptunium, Plutonium, Americium and Technetium*, Vol. 5., North-Holland, Amsterdam (**2003**).

⁶ Knopp, R.; Neck, V.; Kim, J. I. *Radiochim. Acta* **1999**, 86, 101.

With this background, reviewers were surprised to learn that PA calculations were using old solubility values for $\text{Pu}(\text{OH})_4$, values that have been rejected by the NEA peer reviewers in 2001² and 2003.³ This can be traced to the fact that the geochemical modeling software was simply “used as received” with no effort made to verify the database, or insure that the internationally accepted data were being employed. We noted that the solubility of $\text{Pu}(\text{OH})_4$ used in calculations was as high as 10^{-5} M in the oxidizing region III of the model. This situation leads to a solubility estimate for plutonium that is orders of magnitude higher than expected, if the correct range of solubility were employed. In addition, $\text{PuO}_2(\text{OH})_2$ was calculated to have a ca. 2-4 orders of magnitude lower solubility than $\text{Pu}(\text{OH})_4$. This result would indicate that the Pu(VI) solid phase should be the solubility-controlling solid phase in the oxidizing region II and III. Unfortunately, this lack of attention to thermodynamic data quality raises questions and casts doubts as to the validity of other data used in calculations – mineral phases, redox potentials, sorption constants, etc. contained within the database used for PA.

Reviewers learned that the PA calculations took adsorption into account in the vadose zone beneath the tanks and onto the grout. The K_d values used in the vadose zone were from normal (mildly acidic) conditions, and these are not considered to be appropriate for the situation where grout will likely result in much higher pH conditions below the tank ($\text{pH} > 8$).

Plutonium solubility and sorption recommendations: As described in the Performance Assessment discussion below, it is recommended that SRS obtain the OECD NEA-TDB database, and perform a new set of PA calculations using this database. The solubility controlling solid should be the amorphous $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ (am). It is further recommended that SRS PI’s performing thermochemical modeling calculations establish a close working relationship with the plutonium chemists at SRS to insure that plutonium chemistry assumptions and data are reasonable. It is recommended that K_d values used to model adsorption in the vadose zone be re-evaluated. The impact of grout would likely result in a plume below the tank with a much higher pH (>8) than the typical acidic conditions of SRS soils. A range of K_d values under more relevant conditions should be used. The range of K_d values for a variety of conditions has been documented in the open literature.⁵

2. Conceptual Modeling

Performance assessment models: Performance assessment studies for radioactive waste need to consider all reactive interactions between waste and its surroundings. Reactive interaction of the various wastes and the grout materials with the complex

geological environment have to be predicted over thousands of years. Chemical thermodynamics with reaction modeling represents the most developed tool for such long term predictions and is the accepted standard practice worldwide. Modern numerical codes have no principal problems in solving the respective equations for calculation of the reaction and phase equilibria even in the presence of hundreds of chemical species and transport along a concentration gradient. *The reliability and quality of the results depends first of all on the quality of the thermodynamic database underlying the calculations.*⁷ The peer review team was surprised to learn that a commercial numerical code (Geochemists Workbench) was acquired and calculations performed using thermodynamic data that was supplied with the code. There was no effort made to verify and validate the thermodynamic data used or to acquire a thermodynamic database appropriate for plutonium. Peer reviewers discovered this error by recognizing that solubility values for plutonium that were used in the calculations (see discussion under plutonium chemistry) differed by several orders of magnitude from the peer-reviewed and universally accepted values. The Conceptual Model and PA reports cite the NEA accepted thermodynamic data for U, Np and Pu, yet made no effort to insure that these data were actually being employed in PA calculations. This situation is unacceptable, and raises significant questions about the validity of *all* calculations used to support tank closure. This oversight highlights the question of whether SRS has a quality assurance program in place to insure defensibility of its PA for Tank 18 closure.

Modeling recommendations: There are a number of current database projects available for use in PA studies. Of these, the Nuclear Energy Agency Thermochemical Database (NEA-TDB) is the most widely accepted by the international community. It is recommended that SRS obtain this database, and recalculate all its PA calculations using this database. The NEA-TDB was initiated in 1986 as an international effort to make available a comprehensive, internally consistent, internationally recognized, and quality-assured chemical thermodynamic database for selected elements to meet the modeling requirements for safety assessments of radioactive wastes.⁶ For the reviewed elements, international review teams produced critical evaluated thermodynamic standard data following strict NEA-TDB guidelines for the review procedure and data selection, the extrapolation to zero ionic strength, the assignment of uncertainties, temperature corrections to thermodynamic data and enthalpy calculations, standards and conventions for TDB publications, and the independent peer review of TDB reports. To date, there have been 11 published volumes of peer-reviewed and internationally accepted thermodynamic data for actinides (including plutonium) and many organic ligands.^{2-3,7-14} The NEA-TDB is

⁷ Voigt, W.; Brendler, V.; Marsh, K.; Rarey, R.; Wanner, H.; Gaune-Escard, M.; Cloke, P.; Vercoeur, Th.; Bastrakov, E.; Hagemann, S., *Pure Appl. Chem.*, Vol. 79, No. 5, pp. 883–894, (2007).

available online from www.oecd-nea.org and databases¹ have been generated for direct use in the Geochemist's Workbench modeling software being applied by SRS for this work.^{4,5 8-16}

It is further recommended that other geochemical modeling parameters need to be properly evaluated and considered. For example, oxygen and carbon dioxide in infiltration water contacting the waste will not be in equilibrium with air. Oxygen can be expected to be much lower and carbon dioxide partial pressures much higher as a result of contact with organic rich soil horizons undergoing microbial decomposition.

Slow carbonation of the grout as a result of diffusion of carbon dioxide into the grout with water seems like a low probability scenario. Fracturing of the grout with preferential flow through the cracks would appear to be a much more likely scenario and should therefore be evaluated.

3. Experimental Verification

Analysis of tank residues: Extensive presentations were made about the chemical conditions in Tanks at SRS in an attempt to understand what chemical forms of Pu may have precipitated in the tanks. While interesting, the discussion only provided possible clues as to the potential nature of plutonium speciation in the precipitates and residues left in the tanks after extensive cleaning. In addition, the tank inventory has a high degree of uncertainty due to the inexact method of estimating the volume of residues left in the tanks. Analyses were described as taking samples, grinding and dissolving the samples for analysis. The problem with this procedure is that the act of grinding will homogenize the material. More and more experimental data from fieldwork at contaminated sites points to

⁸ Grenthe, I.; Fuger, J.; Konings, R. J. M.; Lemire, R. J.; Muller, A. B.; Nguyen-Trung, C.; Wanner, *Chemical Thermodynamics of Uranium, Vol 1.* H., North-Holland, Amsterdam (1992).

⁹ Silva, R. J.; Bidoglio, G.; Rand, M. H.; Robouch, P. B.; Wanner, H.; Puigdomenech, I., *Chemical Thermodynamics of Americium, Vol 2.* North-Holland, Amsterdam (1995).

¹¹ Rand, J. A.; Rand, M. H.; Anderegg, G.; Wanner, H., *Chemical Thermodynamics of Technetium, Vol. 3.* North-Holland, Amsterdam (1999).

¹² Olin, A.; Noläng, B.; Öhman, L.; Osadchii, E.; Rosén, E., *Chemical Thermodynamics of Selenium, Vol. 7.*, Elsevier, Amsterdam (2005).

¹³ Brown, P. L.; Curti, E.; Grambow, B.; with the collaboration of Ekberg, C., *Chemical Thermodynamics of Zirconium, Vol. 8.*, Elsevier, Amsterdam (2005).

¹⁴ Hummel, W.; Anderegg, G.; Puigdomenech, I.; Rao, L.; Tochiyama, O., *Chemical Thermodynamics of Compounds and Complexes of U, Np, Pu, Am, Tc, Se, Ni and Zr with Selected Organic Ligands, Vol. 9.*, North-Holland, Amsterdam (2005).

¹⁵ Bruno, J.; Bosbach, D.; Kulik, D.; Navrotsky, A., *Chemical Thermodynamics of Solid Solutions of interest in Nuclear Waste Management, Vol. 10.*, OECD online bookstore (2007).

¹⁶ Rand, M.; Fuger, J.; Grenthe, I.; Neck, V.; Rai, D., *Chemical thermodynamics of Thorium, Vol. 11.*, OECD online bookstore (2009).

the heterogeneity of plutonium speciation. There is no real understanding of the nature of plutonium speciation in tank precipitates.

Recommendations for tank residue analysis: SRS should perform some spectroscopic characterization on plutonium precipitates/residues to generate a more scientifically defensible understanding of the nature of these precipitates. Due to its element specificity and ability to probe amorphous materials, x-ray absorption fine structure (XAFS) would seem to be ideally suited to this task. This technique is not limited to Pu, but could be used to identify other elements (such as Fe) that may be associated with Pu in residues.

Residual leachability testing plans: To provide a stronger scientific foundation to justify the use of geochemical modeling in the tank closure performance assessment, validation and verification of the model and assumptions is required. This involves both solid phase characterization of the residual wastes (as described previously) and leachability testing. Leachability tests should be conducted under conditions that are as close as reasonably possible to those expected for actual tank closure scenarios.

Recommendations for leachability testing. It is recommended that leaching tests using leachant solutions that represent both fresh grout and aged grout (including impacts of carbonation and loss of reducing capacity) be evaluated. In some cases it can be useful to conduct leaching studies with deionized water. This approach minimizes precipitation and geochemical modeling of leachates can sometimes be used to identify phases in equilibrium that could not be identified with solids characterization techniques, either because the solid phases are amorphous or occur at concentrations that are below the method detection limit. Leachate analyses should include all major ions, pH, alkalinity, Eh, and appropriate trace components (Pu, Fe, sulfide). Special precautions should be taken in the case of the Pu and Fe analysis to avoid colloids. For example, it is recommended that supernatants be filtered through Amicon Centricon filters (Amicon Corp.) with a 30,000 molecular-weight cutoff (approximately 0.004 μ m pore size). It is also recommended that solids characterization be conducted after leaching because new phases may have precipitated or some phases may have dissolved completely. With the leachate data as input, geochemical modeling of the leachates can be conducted to validate and verify the geochemical model and certain assumptions used in the model.

4. Quality Assurance/Quality Control

Quality assurance: The conceptual model of waste release for a closed radioactive waste tank is a complex problem involving geochemical, physical structures and hydrologic flow path aspects. To demonstrate that it can meet long-term performance standards, SRS has been conducting scientific and technical studies at Tank 18 that serves as supporting documentation for the planned closure of the F-tank Farm tank wastes. One of SRS's most important tasks is to demonstrate the adequacy of its data, software, and models. NRC requires sites to develop a quality assurance program that ensures that the technical information submitted in support of a closure—such as scientific data, models, and details on design and construction—is well documented and defensible. In NRC parlance, data used to support conclusions about the safety and design of the closure must meet transparency and traceability standards. DOE O 414.1d together with 10CFR 830 Part A define the quality assurance program and its implementation by DOE and its contractors. The guidance applies to all aspects of DOE related work as called for in 10CFR830.3 *the performance of work, such as design, manufacturing, construction, fabrication, assembly, decontamination, environmental remediation, environmental restoration, waste management.....*” The site or contractor Quality Assurance Plan (QAP) applies at all levels of personnel and tools they employ in the execution of work.

During their on-site review the peer review team saw no evidence of a quality assurance program for geochemical modeling. As a part of the quality assurance evaluations, there needs to be evaluation of the integrated verification/validation that transparently connects results presented in dose plots over time to basic components of the system such as mass balance (e.g. for Pu and geochemically controlling components). A QA program is of high importance for convincing NRC, peer reviewers, and other entities that SRS has a robust evaluation of the future processes and potential releases so that Tank 18 can be safely closed.

Whereas the PA was peer reviewed by DOE prior to its transmittal to NRC one of the basic science documents *“Conceptual Model of Waste Release from the Contaminated Zone of Closed Radioactive Waste Tanks.WSRC-STI-2007-00544, Rev.1”* upon which the PA is based was not adequately peer reviewed.

Quality assurance recommendations: Basic science documents and all documents transmitted to Federal and state agencies require a level of peer review commensurate with the graded approach guidance given in DOE O 414. 1d.

Appropriate verification/validation of modeling results needs to be completed and documented. For example, conducting mass balance calculations for Pu and other key

geochemical parameters to confirm that model realizations are correct. These checks will confirm that the Pu dose plots and assumptions and simplifications of the PA integrated model are sound and properly constrained (e.g. controlling reactions for redox and pH do not violate mass balance for solid or aqueous components).

5. Communications

Communication: During the two day deliberations it became apparent to the review panel that during the preparation of the PA, a number of organizations contributed to the final document. Yet it also became obvious that communications across lines were targeted to specific points without consideration to the overall scheme of things and thus suffered from the effects of tunnel vision. As a case in point, no SRNL plutonium chemist(s) were consulted for input or peer review to WSRC-STI-2007-00544, Rev.1. The reviewers also learned that for the planned presentations of November 9, 2011 the SRNL plutonium experience presented was hurriedly put together the previous day.

This lack of coordination and project integration across lines and contractors often leads to situations similar to those found by the team and detailed in previous sections of this report. For example, the project lead for the PA assumed that the basic science used to develop the geochemical model was peer reviewed and passed muster. However, the geochemical model report came from another organization which did not carry out the necessary due diligence. This issue was further compounded by the fact that the plutonium model results were not peer reviewed by another group within the same organization.

Communications recommendation: Management at all levels need to be vigilant and questioning the assumptions and conclusions of reports that carry multiple management level signatures prior to being issued, especially to external organizations, otherwise loss of credibility can have long lasting and painful consequences.

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