WIPP TRU REPOSITORY: UPDATE OF THE SAFETY CASE

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WIPP Project and Actinide Solubility

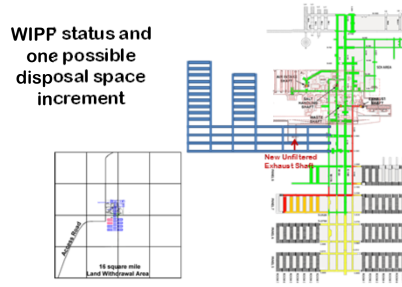
The Waste Isolation Pilot Plant (WIPP) transuranic repository remains a cornerstone of the U.S. Department of Energy's (DOE) nuclear waste management effort. The project continues to deal with operational issues that led to a shutdown of operations in February 2014, although some parts of the repository are back to being operational and some waste is currently being disposed. These operational issues, however, did not impact the long-term safety case for the permanent disposal of transuranic (TRU) waste which remains intact. The WIPP's third recertification (submitted in March 2014) has been declared complete by the Environmental Protection Agency (EPA) and should be approved by June 2017. The full recovery plan of the repository, however, is to install a second shaft to make up for lost volume due to the slight residual contamination and this is not expected to be completed for approximately five years (~ 2022) and this is when full waste disposal activities will resume. There continue to be ongoing discussions about a possible expanded mission for the WIPP and additional nuclear repository concepts in a Salt geology within the United States remain under consideration. Research to strengthen the scientific basis of the safety case for actinide containment in high ionic-strength brine systems continues and is ongoing.

An understanding of the actinide and brine chemistry in the WIPP is needed to address the low-probability scenario that brine inundation and release could occur due to human intrusions and is driven by regulatory requirements for the repository license case. The overall ranking of actinides, from the perspective of potential contribution to release from the WIPP, is: Pu ~ Am > U >> Th and Np and remains unchallenged from past recertification. Research emphasis in the past couple of years has focused on issues raised during the regulatory review process of the 2014 recertification application. The most important of these are: 1) oxidation state distribution of plutonium and overall redox conditions expected; 2) role and importance of site-specific solubility studies in assessing the overall source term modeling in PA; and 3) effects of organic complexation on the An(III) and An(IV) oxidation states; and 4) further progress in understanding microbial effects. All of these areas will be updated and progress made in addressing these issues will be presented. These data [2-7] continue to extend our understanding of high ionic-strength actinide chemistry and strengthen and improve the scientific basis for the safety case for a nuclear repository in salt. A brief summary of ongoing activities in these areas follows.

WIPP Status and Future Directions for Salt-based Nuclear Repositories

Status of the WIPP Project

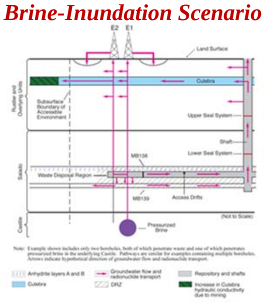
- After 15+ years of safe operation, a radiological release incident closed the WIPP in February 2014. Until this time, 24 waste storage sites in the US were cleaned up, ~ 12,000 CH and RH shipments were received, over 91,000m^3 of waste emplaced, and 6 panels are full.
- January 4, 2017 radioactive waste emplacement operations in the WIPP were safely resumed.
- July 19, 2017 the EPA recertified the WIPP for the third time.
- Although the WIPP is now open again, operations remain limited and plans continue to construct a second shaft that will allow full restoration of operations and provide additional waste disposal areas needed.



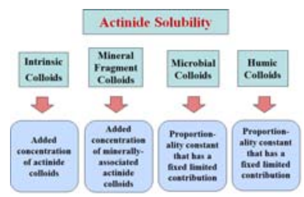
Impacts to the Core WIPP Safety Case

There has been effectively no change in the WIPP safety case due to the February 2014 release incident. The main pathways to release remain the low-probability brine inundation scenarios where human intrusion permits brine to enter the repository and react with the waste and barrier material to solubilize the TRU in the waste. This leads to high magnesium brines (GWB is the simulation of this) at an expected pC_{H+} of 9.5 ± 0.2. Actinide solubility is calculated at this pC_{H+} using the Pitzer approach with EQ3/6. The current calculational results and oxidations state assumptions are shown on the right. Actinide solubility has increased somewhat over time due to better accounting for the effects of organic complexation (primarily EDTA) in WIPP waste.

Brine-Inundation Scenario



Actinide "Mobile" Concentration



Actinide Solubility in Brine-Inundation Scenarios

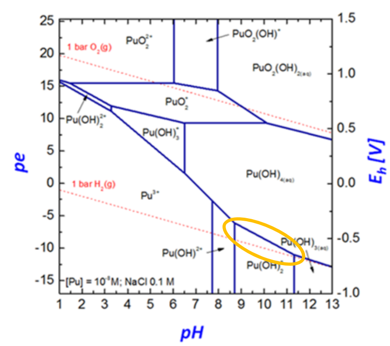
Calculated Oxidation-Specific Solubility of Actinides in Equilibrated WIPP Brine (GWB – high Mg brine, ERDA-6 – NaCl brine) [Brush and Domski, 2013]				
Actinide Oxidation State, and Brine	**Actinide/ Analog Used**	**CRA-2004 PABC (M)**	**CRA-2009 PABC (M)**	**CRA-2014 PA (M)**
III, GWB	Am/Nd	3.87×10^{-7}	1.66×10^{-6}	2.59×10^{-6}
III, ERDA-6	Am/Nd	2.88×10^{-7}	1.51×10^{-6}	1.48×10^{-6}
IV, GWB	Th	5.64×10^{-8}	5.63×10^{-8}	6.05×10^{-8}
IV, ERDA-6	Th	6.79×10^{-8}	6.98×10^{-8}	7.02×10^{-8}
V, GWB	Np	3.55×10^{-7}	3.90×10^{-7}	2.77×10^{-7}
V, ERDA-6	Np	8.24×10^{-7}	8.75×10^{-7}	8.76×10^{-7}

Plutonium Oxidation State Distribution

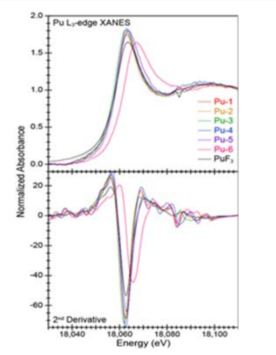
The distribution of actinide oxidation states within the range of expected conditions in the WIPP was set by expert opinion within potential E_h range. The current distribution of key multivalent actinides and the speciation data used to model each oxidation state is shown to the right. This is now being challenged by long-term WIPP-specific data, as well as reported studies in the literature, that show that the low-valent iron chemistry in the WIPP is effective in establishing more strongly reducing conditions. Some ~ 10 year data recently obtained are shown to the right. These data, obtained for Pu-242 in simplified systems, show that Pu(III) can be produced and sustained in WIPP brine but this appears to be a very narrow Eh range and under more realistic conditions (e.g., Pu-239 and higher-activity isotopes, presence of organics, and Fe(III) and nitrate phases) the Eh is increased in favor of the Pu(IV) oxidation state. These data confirm our current model assumptions by showing that Pu(III) is observed under extremely reducing conditions and Pu(IV) prevails and remains the predominant oxidation state under more realistic conditions. This issue continues to be the focus of continued investigation within the WIPP project with variable isotope studies and more realistic long-term conditions.

Currently Assumed Actinide Oxidation-State Distribution

Oxidation State Distribution of Key Actinides in WIPP Performance Assessment					
Actinide	**Oxidation State**				**Speciation Data used in Model Predictions**
	III	**IV**	**V**	**VI**	
Uranium		50%		50%	Thorium for U(IV), 1 mM fixed value for U(VI)
Plutonium	50%	50%			Am/Nd for Pu(III) and thorium for Pu(IV)
Americium	100%				Americium/neodymium



E_h, pH diagram for Pu (provided by KIT/INE – ref [8]) that illustrates the very narrow E_h regime where Pu(III) would be expected at the WIPP repository conditions – see yellow circle. Under more realistic conditions Pu(IV) is expected to predominate.

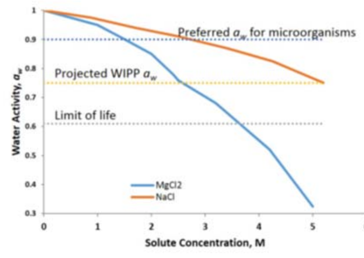


~ 10 year data on WIPP-specific Pu-Fe interaction studies that show that the edge position matches a PuF3 standard in all cases where there is available Fe(0/II). A higher oxidation state (Pu (V) and/or (VI) is observed when only Fe(III) is present. [data collected by Dan Olive, analyses performed by Sharon Bone and Stosh Kozimor - all Los Alamos]

Microbial Effects in the WIPP

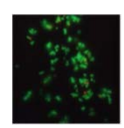
The microbial ecology and the interactions of WIPP-relevant microorganisms with actinides continues to be the subject of ongoing studies. This has now expanded to include emplaced organisms (e.g., found in the waste) to account for pre-inundation microbial activity (see *Arthrobacter* sp. Isolated from INL WIPP-bound waste). Once inundation occurs within the WIPP (via the low-probability intrusion scenarios) this effectively sterilizes the WIPP or renders most microorganisms inactive. These indigenous organisms can survive long-term at high salt concentrations but may not be active waste degraders and may be limited to early oxic or suboxic phase of repository history. At this time, no site-specific studies have detected or isolated anaerobic, halophilic organisms in subterranean salt. Emplaced organisms are better equipped to degrade waste but may not be active, or even viable, at high salt concentrations. Degradation is likely to occur within waste drums prior to inundation, but limited by lack of moisture and levels of radioactivity.

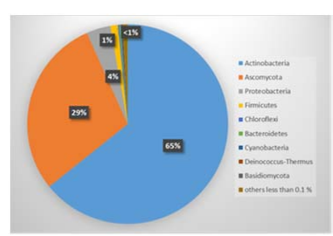
The bioassociation of actinides/analogs and long-term stability the these colloidal species is a key contributor to the WIPP mobile actinide concentration source term in PA. Halophilic bacteria continue to show relatively strong sorption at moderately high pH. Archaea, in contrast, undergo much lower levels of bioassociation. The long-term effects of this bioassociation remains unclear and is the subject of further studies.



Effects of Water Activity and Chaotropicity: Impacts of Natural Brine Constituents

***Arthrobacter* sp. Isolated from WIPP-bound TRU waste**





Emplaced Microorganisms

Acknowledgements

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[1] Van Soest, G. *Performance Assessment Inventory Report – 2012* (PAIR 2012). INV-PA-08, LANL-CO\Inventory Report, Los Alamos, NM: Los Alamos National Laboratory (2012).
[2] Swanson et. al., *Geomicrobiology Journal*, vol. 30:3, 189-198 (2013).
[3] Swanson et al., Los Alamos National Laboratory report LA-UR-13-26280. Los Alamos National Laboratory, Los Alamos NM, USA (2013).
[4] Ams et al. *Geochimica et Comoschimica Acta*, vol. 110: 45-57 (2013).
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[6] Reed et al., Los Alamos National Laboratory report LA-UR 13-20858, Los Alamos National Laboratory, Los Alamos NM, USA (2013).
[7] Swanson et al., "The Microbiology of Subsurface, Salt-Based Nuclear Waste Repositories: Using Microbial Ecology, bioenergetics, and Projected Conditions to Help Predict Microbial Effects on Repository Performance." Los Alamos National Laboratory report, LA-UR-16-28895. Los Alamos National Laboratory, Los Alamos NM, USA (2017).
[8] Altmaier et al., "Assessment of Np(IV) and U(IV) as Improved Analogs for Pu(IV) in High Ionic-Strength Brine Systems," KIT/INE report, 2017.