Actinide Containment Safety Case in a TRU/HLW Repository in Salt

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ABSTRACT

The safety case for the permanent isolation of actinides in a salt nuclear repository is discussed. The need for actinide and brine chemistry research is driven by the regulatory process that requires an accounting of low-probability "worst case" scenarios to show that the release criteria are met under these unexpected conditions. An overview of the WIPP approach to establish the actinide source term, which is defined by the oxidation state distribution, solubility and tendency to form colloidal species, is presented for the key actinides. This approach is then discussed in the context of extending the salt repository concept to defense high level waste (HLW) and possibly spent fuel (SF) to identify key needs and gaps. It is an overall conclusion of this review that the WIPP serves as a good template for the HLW/SF case and the safety case can be readily extended to this repository concept with proper repository design and supporting experimental results.

INTRODUCTION

The overall concept to use bedded or domed salt for the permanent disposal of nuclear waste was recommended by the National Academy of Sciences quite some time ago (in 1950's). In salt, the expected outcome is that the excavated salt will rapidly re-seal (< 200 years) once the repository is closed. With little/no interconnected groundwater flow, the sealed repository should remain unsaturated with essentially zero radionuclide release for millions of years. The ability to rely heavily on the site geology for permanent isolation is the key advantage of the salt repository concept and, along with low cost and extensive mining experience, continues to make this an ideal medium for the permanent disposal of nuclear waste.

The salt repository concept was not fully implemented until the Waste Isolation Pilot Plant (WIPP) began operations as a transuranic (TRU) repository in 1999 (see Figure 1). The safety case for the permanent disposal of TRU waste in the bedded salt Salado Formation in southeastern New Mexico was first licensed by the EPA in 1998 [1]. It has since been recertified twice by the EPA in 2005 and 2010 [2, 3]. A third recertification was submitted in March 2014 and is currently under review. Salt repository options for nuclear waste are also under consideration in the German repository program. In all of these certification or safety case development activities, an actinide source term was needed to account for the low probability

scenarios where direct brine release may occur. The scientific basis that defines the actinide source term, for this reason, has been a key focus of research in the United States [4-14] and in Germany [15-19] over the past several years.   This was also the subject of joint US-German workshops [20-22].   An overview of the current understanding of the key factors that affect the actinide source term is provided herein.

It can also readily be argued that the WIPP TRU safety case is a template for the development of a safety approach to support the permanent geologic disposal of high-level nuclear defense waste (HLW) and spent fuel (SF) in a salt repository. In comparison to the WIPP TRU case, the need for a well-defined actinide source term should increase due to the much higher nuclear content in the HLW/SF waste forms that will also lead to a thermally heated repository with significantly higher levels of ionizing radiation.   The data gaps and key needs to address this expanded role for the salt repository concept are also briefly discussed herein.

Figure 1.   WIPP Salt Repository Concept and Implementation in the Bedded-Salt Salado Formation in Southeastern New Mexico.   This concept was first certified in 1998 and has been recertified two additional times in 2005 and 2010.
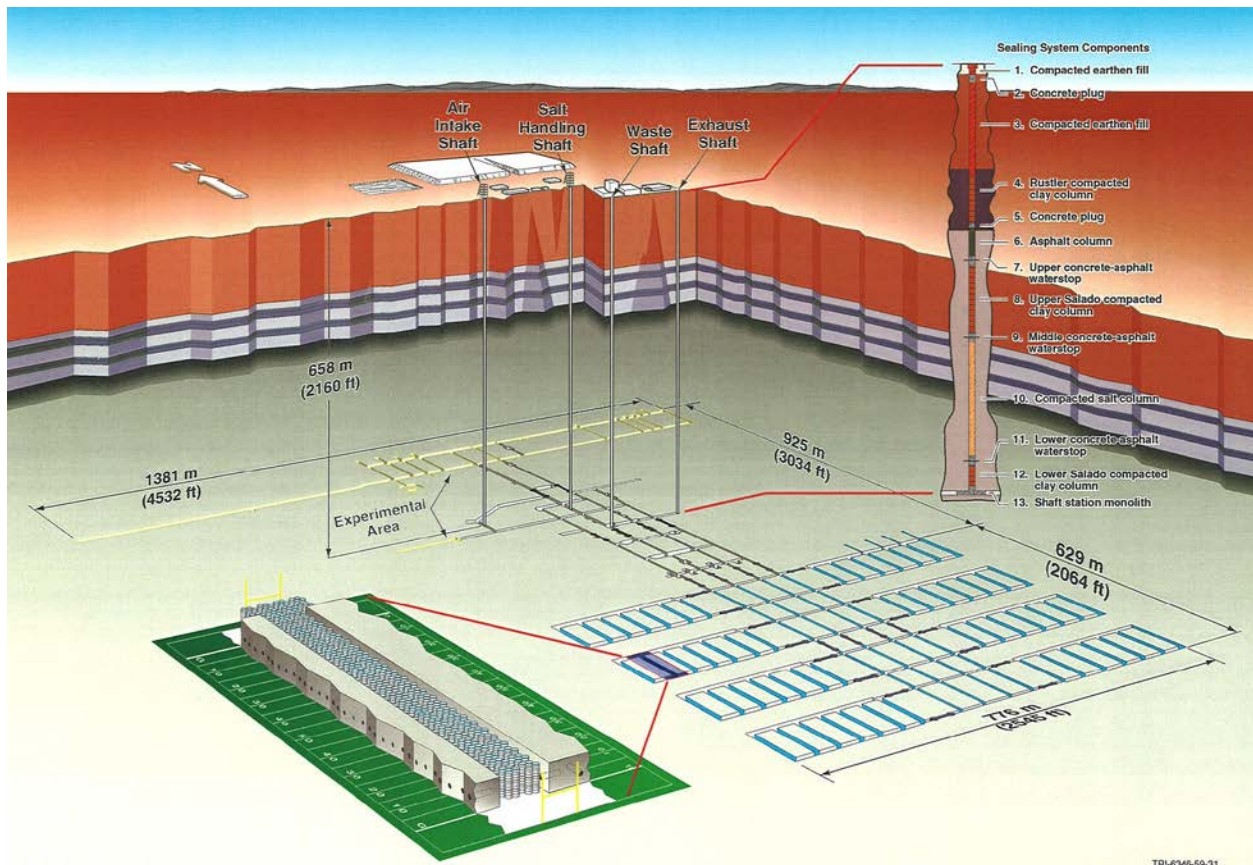
## ROLE AND IMPORTANCE OF ACTINIDE/BRINE CHEMISTRY IN THE OVERALL SAFETY CASE

The safety case for the use of a bedded salt or salt dome for the permanent geologic isolation of nuclear waste is centered on the self-sealing properties of salt that lead to geologic isolation (see Figure 2).   This, under ideal conditions, will lead to a "dry" site that can geologically isolate the nuclear waste for millions of years.   The salt formations being considered or used for geologic isolation are hundreds of millions of years old and have remained unsaturated with no interconnected groundwater for much of this time.   In many respects this disposal concept is somewhat independent of the nature of the wasteform since it relies on geologic isolation and not container material and wasteform properties under the expected repository conditions.   There are also significant cost advantages to a salt repository approach since the technology for the mining of salt is well established and we have a ~14-year operational history and experience at the WIPP site.

Figure 2.   Conceptualization of the effect of salt creep on emplaced TRU waste containers in the WIPP.   The resultant self-sealing and compaction greatly reduces the reliance of the safety case on container material and wasteform properties.

Emplaced TRU Waste   Compacted Waste at 1000 Years
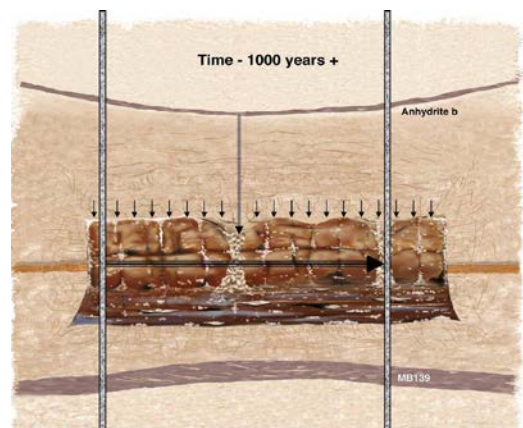
It is critically important that a repository concept, and its associated safety case, has a sound scientific basis to assure the public that the repository will perform as predicted.   For a salt-based nuclear repository, although primary safety reliance is on the self-sealing of its geology, there are low probability scenarios where brine intrusion leading to the solubilization of actinides/radionuclides in high ionic-strength brines and their subsequent release to the accessible environment is possible.   An understanding of actinide and brine chemistry, in this context, is mainly needed to account for these low-probability brine intrusion scenarios that may solubilize and mobilize the actinides present.   The driver for this understanding is therefore the regulatory process that requires low-probability scenarios to be addressed in the safety case to show that the repository will meet the target release limits even under these unexpected "worse case" scenarios.

ACTINIDE SOURCE TERM IN THE WIPP

The mobile actinide/radionuclide concentration that defines the source term, in the low-probability scenario of brine intrusion, is defined by the multitude of subsurface processes that impact actinide/radionuclide speciation that are described in this section.

Overall Approach

The overall approach used to establish the actinide source term (see Figure 3) in the WIPP is the following:

- Establish the key and relevant brine chemistry by sampling/analysis of brine that seeps into the repository and in the nearby brine pockets.   The interactions of this brine with waste and engineered barrier components is modeled and confirmed by experiment.   This established the range of brine composition, pH, and overall redox conditions.
- Down-select by two screening processes to prioritize the oxidation state and actinides that contribute most significantly to TRU release.   First, establish the isotope-specific inventory of the TRU content.   Second, establish the likely oxidation states and speciation that could persist throughout the 200-10,000 y repository performance period.   This establishes the prioritization for TRU species that need to be investigated experimentally and subsequently modeled.
- Determine by a combination of modeling and site-relevant experimental results the oxidation state-specific solubility and colloidal contribution of the key oxidation states for the expected range of brine composition that includes the potential impacts of complexing co-contaminants in the waste (specifically these are oxyanions, carbonate, and anthropogenic or biogenic organic chelators).
- Model, using the Pitzer approach, actinide solubilities and associated colloidal contributions using relatively few, but defensibly conservative, sets of reactions. This addressed the uncertainties in the data and provided an explainable and defensible description of the actinide source term used in performance assessment (PA) release calculations.
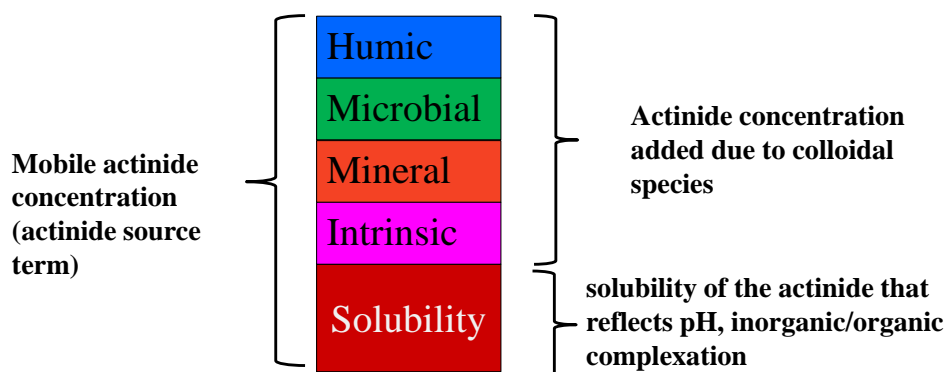
Figure 3.   Mobile actinide source term concentration in the WIPP safety case.

Brine Composition and Chemistry

Brine introduced into the WIPP will react with emplaced TRU waste (actinides, organics and salts), waste-affiliated components (e.g. iron and lead), and the engineered barrier material (MgO) to establish the brine chemistry that will define the actinide solubilities and colloidal species that contribute to the actinide source term. The composition of brine in and around the WIPP site prior to waste emplacement was established by sampling the nearby groundwater, analyzing the brine seeping into the repository, and determining the composition of the brine in the intergranular inclusions in the Salado, Castile and Dewey Lake Formations. The stability of these brines were investigated extensively [8, and the several reports referenced within]. A summary of the brine compositions used in the WIPP project over time and those predicted based on modeling studies is given in TABLE I.

Salado brine will enter the repository after closure, and can be supplemented by Castile brine, from brine pockets underneath the repository horizon, in some human intrusion scenarios. It is also possible that groundwater from the Rustler and Dewey Lake Formation could flow down the borehole into the repository, mix with the waste, and then be forced back up a borehole. The majority of WIPP-specific solubility studies were performed using GWB or ERDA-6 brines, since these brines bracket the expected compositional range of the brine.   More recently [8], repository studies have focused on pH-specific brine compositions to account for the large changes that occur as a function of pH (See Figure 4) with some of the brine components to provide a more defensible approach that addresses a broader pH range.

Figure 4.   Concentration of tetraborate and Mg^{2+} in WIPP-relevant brine as a function of pC_{H+} [8]. Experimentally-derived pH-specific brine formulations are used to account for current limitations in modeling the magnesium chemistry at pH> 10.
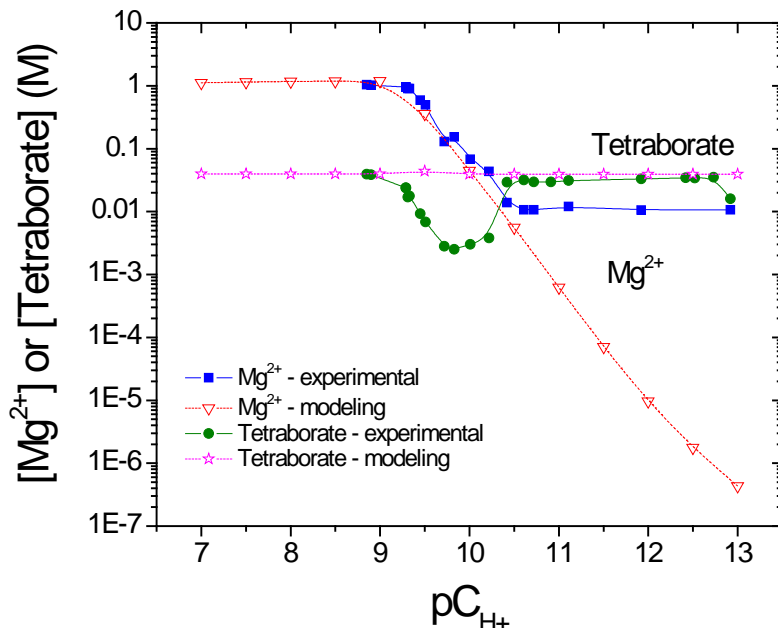
TABLE I.   Compositions of historical brines (Brine A, G-Seep), and GWB and ERDA-6 brines prior to and after equilibration with MgO [8].

Ion or property[a]	Brine A	G-Seep	GWB before reaction with MgO, halite, and anhydrite	GWB after reaction with MgO (phase 5), halite, and anhydrite	ERDA-6 before reaction with MgO, halite, and anhydrite	ERDA-6 after reaction with MgO (phase 5), halite, and anhydrite
[b]$B(OH)_x^{3-x}$	20 mM	144 mM	158 mM	180 mM	63 mM	62.4 mM
Na^+	1.83 M	4.11 M	3.53 M	4.52 M	4.87 M	5.28 M
Mg^{2+}	1.44 M	0.630 M	1.02 M	0.463 M	19 mM	136 mM
K^+	770 mM	350 mM	0.467 M	0.532M	97 mM	96.1 mM
Ca^{2+}	20 mM	7.68 mM	14 mM	10 mM	12 mM	11.2 mM
SO_4^{2-}	40 mM	303 mM	177 mM	214 mM	170 mM	176 mM
Cl^-	5.35 M	5.10 M	5.86 M	5.37 M	4.8 M	5.23 M
Br^-	10 mM	17.1 mM	26.6 mM	28.3 mM	11 mM	10.9 mM
Total Inorganic C (as HCO_3^-)	10 mM	11.5 mM	Not reported	0.358 mM	16 mM	0.448 mM
pH	6.5	6.1	Not reported	8.74	6.17	8.98
Relative Density	Not reported	Not reported	1.2	1.233	1.22	1.22
Ionic Strength (m)	Not reported	Not reported	7.56	7.64	6.05	6.77

[a] Ions listed represent the total of all species with this ion.
[b] Boron species will be present in brine as boric acid, hydroxy polynuclear forms ($B_3O_3(OH)_4^-$), and/or borate forms (e.g., $B_4O_7^{2-}$)

Actinide Inventory and Oxidation State Distribution: Prioritization of Actinide Species as to their Importance to Potential Release

The collection of waste inventory data is done annually as part of the WIPP project. The projected inventory, based on the 2012 data [22] that is the basis of the pending 2014 WIPP recertification application, is given in TABLE II as a function of time after repository closure.

The assessment of this inventory data is a critical first screening step to establish the prioritization of the actinide species that define the actinide source term. These data,

in the case of the WIPP project, show that both Am and Pu initially dominate the TRU source term.   In the longer term, the relative importance of Pu increases.   This screening was also used to eliminate Np and Cm based on their low inventory (this is independent of speciation arguments) and established Th and U as minor contributors to release although their overall mass is relatively high.

TABLE II.   Radionuclide inventory in the WIPP at different times.   Pu/Am are the dominant TRU actinides with respect to the source term [23]

Element	2033 (0 years) Ci (Kg) and %Activity	2133 (100 years) Ci (Kg) and %Activity	3033 (1000 years) Ci (Kg) and %Activity	12033 (10,000 years) Ci (Kg) and %Activity
Th	7.04 (1.35×10^4) 0.00%	8.52 (1.35×10^4) 0.00%	22.5 (1.35×10^4) 0.00%	127 (1.35×10^4) 0.03%
U	528 (2.26×10^5) 0.02%	645 (2.26×10^5) 0.04%	746 (2.26×10^5) 0.09%	769 (2.28×10^5) 0.15%
Np	23.2 (32.5) 0.00%	44.8 (62.9) 0.00%	140 (197) 0.02%	170 (238) 0.03%
Pu	2.02×10^6 (1.20×10^4) 63.5%	1.03×10^6 (1.19×10^4) 60.9%	7.24×10^5 (1.16E4) 83.0%	5.00×10^5 (9.12×10^3) 99.8%
Am	7.05×10^5 (203) 22.2%	6.20×10^5 (179) 36.6%	1.47×10^5 (42.4) 16.9%	21.1 (0.0994) 0.00%
Cm	9.97×10^3 (0.122) 0.31%	216 (2.65×10^{-3}) 0.01%	2.32×10^{-13} (2.84×10^{-18}) 0.00%	0.00 (0.00) 0.00%
Cs	2.35×10^5 (2.67) 7.39%	2.33×10^4 (0.265) 1.38%	2.17×10^{-5} (2.46×10^{-10}) 0.00%	0.00 (0.00) 0.00%
Sr	2.09×10^5 (1.51) 6.57%	1.78×10^4 (0.129) 1.05%	4.21×10^{-6} (3.05×10^{-11}) 0.00%	0.00 (0.00) 0.00%
Total Ci for these isotopes	3.18×10^6	1.69×10^6	8.72×10^5	5.01×10^5

The second important screening step is that of oxidation state distribution for the actinides considered in release.   There is no universally accepted method for doing this and this was extensively addressed recently in the European community ReCosy project [23].   In the WIPP, oxidation states that bracket the most reduced and most oxidized expected scenarios were determined by expert opinion and this distribution, shown in TABLE III, has been the same since the initial license application.   From a potential release perspective, the oxidation states and actinides that are important are:

Am(III) ~ Pu (III/IV) > U(IV/VI) >> Th(IV) and Np(IV/V)

The only technical challenge to this "expert" opinion approach was the assumption that higher-valent Pu(V/VI) species do not persist in the WIPP. This was addressed extensively by the WIPP project (see Figure 5) where it was shown that the Fe(0,II) present in the WIPP lead to the rapid reduction of Pu(V/VI) to Pu(III/IV) [4, 12].

TABLE III. Actinide Oxidation-State Distribution Used in WIPP PA.

Actinide Element	Oxidation States, Abundance (%), and Analog Used (If Any)				Model Speciation Data Used
	Oxidation State[a,b]				
	III	IV	V	VI	
Thorium	—	100 %	—	—	Thorium
Uranium	—	50 %	—	50 %	1 mM assumed for VI, Th for IV
Neptunium	—	50%	50 %	—	Np for V Th for IV
Plutonium	50 %	50 %	—	—	Am for III Th for IV
Americium	100 %	—	—	—	Americium
Curium	100 %	—	—	—	Americium

[a] Oxidation state distributions (percentages) refer to the percent of PA vectors that have 100% of the specified oxidation state.

[b] In PA calculations the distribution of oxidation states is correlated for U, Np, and Pu such th the states for all three elements are simultaneously either in the lower oxidation state (U(IV), Np(IV), and Pu(III)) or in the higher oxidation state (U(VI), Np(V), and Pu(IV)).

Figure 5. The Concentration of Pu as a Function of Time in the Presence of Iron Powder, Iron Coupon, Ferric Oxide, and Magnetite (Mixed Iron Oxide) [9]. These data show that Pu(V/VI) is rapidly reduced by the iron (0, II) present in the WIPP.
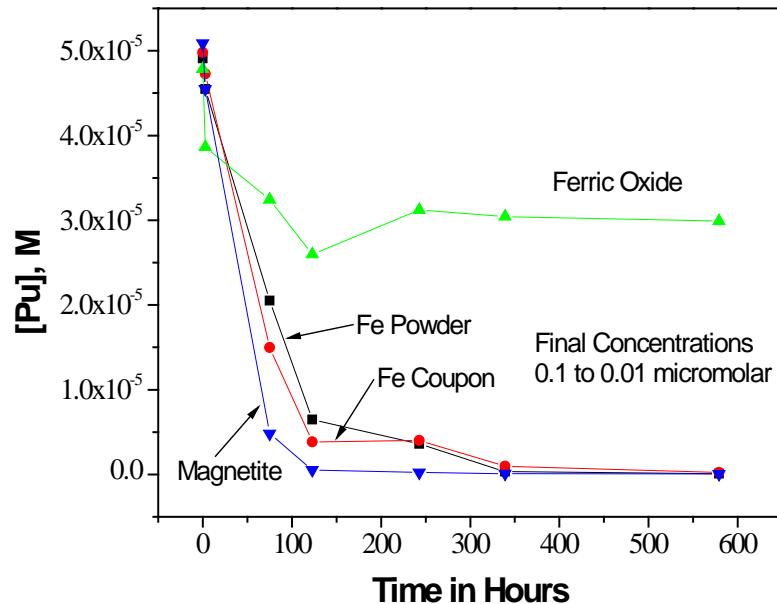
Actinide Solubility and Colloidal Species

Actinide solubility and colloidal species concentrations to support WIPP PA were established by a combination of Pitzer approach modeling [24, 25], experimental measurements [11, 13-18] and confirmed by WIPP-specific studies [4-7]. The overall agreement between these various approaches provides a sufficient basis to define the actinide source term that supports the repository safety case.   It was critical in this approach to be conservative to address potential uncertainties in the regulatory process especially since all aspects of the chemistry are not fully understood for the duration of the repository performance period.

As an example of this approach, thorium is used as the oxidation-state invariant analog for the An(IV) oxidation state.   In WIPP PA, thorium solubility data and colloidal properties are used to model all An(IV) actinides with the predominant actinide of concern being Pu(IV).   Thorium, in fact, is not a good analog for Pu(IV) since it is much less strongly hydrolyzed, has relatively high sorptive interactions and a higher tendency to form colloidal species (see Figure 6).   These well-known chemical disconnects lead to a significant overestimation of the An(IV) contribution to the actinide source term for Pu(IV).   This overestimation, however, works very well in the regulatory process as a defensible analog for Pu(IV) due to its multiple layers of conservatism with respect to potential release.

In WIPP PA, the actinide source term is, in the end, determined by model calculations/predictions that are confirmed by experimental results and supported by empirically determined parameters [24, 25].   The results calculated for the key actinide species are summarized in TABLE IV.   This is combined with an uncertainty analysis that includes the range of "relevant" literature results to provide a distribution that is used in PA release realizations to demonstrate the extent that the target release criteria are met.

The overall implementation of the WIPP PA model and predictions, as currently configured, has provided a defensible argument that dissolved brine release, should it occur during brine intrusion scenarios, will still meet the regulatory goals. Work continues within the project to quantify the conservatism of the current approach to strengthen the overall safety case and fill in the knowledge gaps that are currently addressed by conservative assumptions.   These may or may not lead to model implementation change but positively impact the scientific integrity and lower the uncertainty in the predicted performance of the repository.   There are also project-specific and international efforts to strengthen and make more robust the Pitzer-approach modeling that supports all these high ionic-strength brine systems. All of these strengthens the overall safety case, but also lays a strong foundation for the possibility of expanded uses of the salt repository concept for nuclear waste management and the final disposition of nuclear waste that is beyond the current waste acceptance criteria for the WIPP.

Figure 6.   Thorium solubility in simulated WIPP Brine as a function of time and pH (top) approached from above/below saturation.   The size distribution of the thorium species is also shown for selected systems (bottom).   Thorium is used as the An(IV) analog in WIPP PA but the long-term disequilibrium and unclear contribution of colloidal species make this a very difficult system to interpret although it is clear that it is conservatively overestimating the An(IV) contribution to the actinide source term.
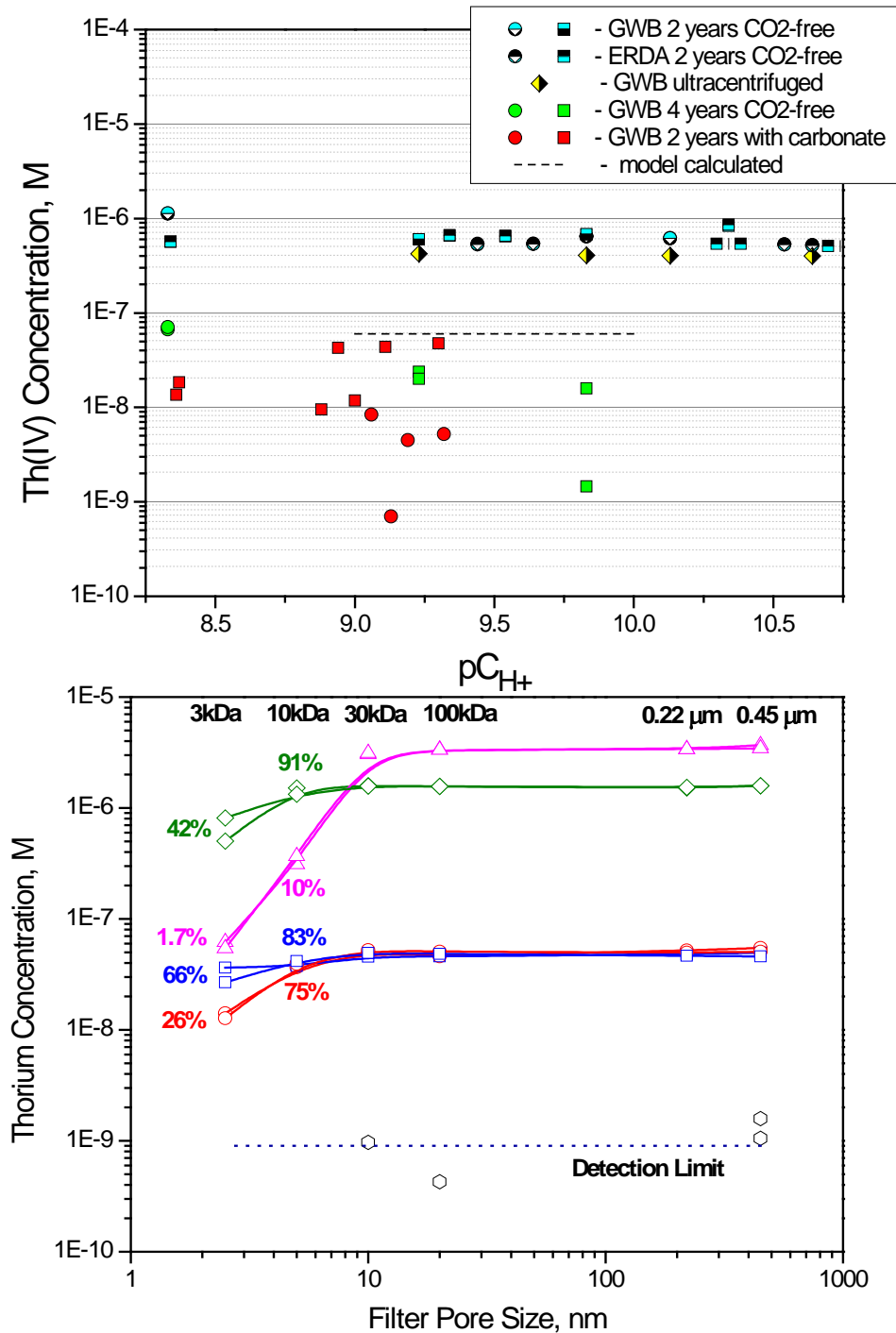
TABLE IV. Concentrations (M) of Dissolved, Colloidal, and Total Mobile Actinides Obtained Using Median Parameter Values for the CRA-2014 PA

Actinide Oxidation State and Brine	Solubility (M)	Colloidal contribution (M)	Total mobile Concentration (M)
Pu(III), Salado brine	3.46×10^{-7}	7.21×10^{-7}	1.07×10^{-6}
Pu(III), Castile brine	1.98×10^{-7}	6.65×10^{-7}	8.62×10^{-7}
Am(III), Salado brine	3.46×10^{-7}	9.57×10^{-8}	4.42×10^{-7}
Am(III), Castile brine	1.98×10^{-7}	3.01×10^{-7}	4.98×10^{-7}
Th(IV), Salado brine	6.46×10^{-7}	4.12×10^{-6}	4.76×10^{-6}
Th(IV), Castile brine	7.50×10^{-7}	4.77×10^{-6}	5.52×10^{-6}
U(IV), Salado brine	6.46×10^{-7}	4.13×10^{-6}	4.77×10^{-6}
U(IV), Castile brine	7.50×10^{-7}	4.78×10^{-6}	5.53×10^{-6}
Pu(IV), Salado brine	6.46×10^{-7}	4.12×10^{-6}	4.76×10^{-6}
Pu(IV), Castile brine	7.50×10^{-7}	4.77×10^{-6}	5.52×10^{-6}
U(VI), Salado brine	1.00×10^{-3}	1.11×10^{-5}	1.01×10^{-3}
U(VI), Castile brine	1.00×10^{-3}	1.11×10^{-5}	1.01×10^{-3}

POTENTIAL IMPACTS OF HLW/SF ON THE ACTINIDE SOURCE TERM: EXTENSION OF SAFETY CASE BEYOND THE WIPP TRU CONCEPT

The WIPP safety case has been extensively reviewed and defended to EPA which is the regulator for the WIPP repository.   This addressed the brine and actinide chemistry that comprise the actinide source term since this is important in the overall TRU repository safety case to account for low-probability brine intrusion scenarios. There remain, however, many conservatisms within the current modeling approach to account for known gaps in our understanding and/or ability to predict long-term performance with respect to actinide release.

At the time of this writing, the WIPP repository is shut down and in the process of recovery from two significant operational problems that occurred in February 2014. These issues, although quite serious from an operational point of view, have had little/no effect on the overall safety case and do not weaken the strategy for using bedded salt for the permanent isolation of TRU waste.   This has however raised legitimate questions about the WIPP safety case and we are in the processing of re-affirming that the current safety case remains adequate.   These operational issues, perhaps more importantly, also raise questions about the wisdom of considering the salt geology for the permanent disposal of higher activity HLW and SF

waste.   Although there are clearly some technical issues that need to be addressed, it still remains that many of these same arguments used in the WIPP safety case extend to the HLW/SF case the WIPP remains a template for these extended repository concepts.

Many core aspects of the WIPP safety case are not changed by considering HLW/SF. The repository as a whole will be elevated in temperature which should leave it dry (meaning unsaturated) in the expected long-term scenario.   Self-sealing should still occur, probably at a faster rate than predicted in the WIPP case.   Even though the repository is dry, intrusion scenarios will over time need to be considered and this will likely lead to pressurized and heated brine and associated reactions that will affect actinide concentrations.   The much higher activity, much higher radiation levels, and more stringent regulatory process likely all point to the need for a greater and more detailed understanding of the brine and actinide chemistry over a broader range of brine compositions and repository temperature.   For all these reasons, it will remain necessary to define the actinide source term and this will require some additional data beyond what is currently in the WIPP model.

In this context, and from the narrow point of view of the actinide source term, the following is an assessment of the most significant remaining issues that relate to the HLW/SF case.

Actinide model and speciation:

The WIPP geology and geochemical data used in PA pertain to temperature conditions at ~ 25°C since little/no heating occurs in the WIPP.   For HLW/SF it is clear that elevated temperatures (~ 70°C for HLW, perhaps 250°C for SF) will be present and the Pitzer-approach modeling will need to be extended to these higher temperatures to address this effect for the brine and actinide aqueous chemistry was well as the stability and formation of key solubility limiting phases.   There are little/no data on the effect of temperature on Pitzer formulations in high ionic strength systems. There is much discussion in the international community on practical ways to do this and elevated temperature is likely to have a slightly beneficial effect on the overall actinide solubilities measured.   Selected experiments coupled with strategic extrapolation from existing room temperature data may be sufficient to extend the Pitzer model to the higher temperatures expected.

In the WIPP, the focus, as just summarized, is on the TRU components and in particular Am and Pu.   For HLW/SF this however will need to be extended to other actinides, which will not be screened out due to inventory limitations, and moderate-activity fission products.   From an actinide point of view, it would be expected that there will be greater emphasis on uranium as its chemistry will impact the availability of other radionuclides (especially for SF), improved analogs for An(IV) are needed, and it will be likely that a wider range of oxidation states will need to be considered.   In the end, therefore, a broader set of radionuclides will also need to be

addressed to establish what will now be the radionuclide source term as input into the overall PA.

Colloidal species impact the actinide source term in the WIPP model, but this has a relatively small overall impact on PA realizations.   This is because the key impact of colloidal species in a salt repository is its effect on direct brine release scenarios rather than radionuclide migration because there is little/no interconnected groundwater once self-sealing occurs.   This needs to be evaluated as a function of temperature but is also expected to be less significant as higher temperatures will likely kinetically accelerate the formation of stable less soluble, and therefore less colloidal, radionuclide species.

The lack of a good understanding of the microbiology associated with salt formations was a critical limitation in the safety case that forced very conservative assumptions and engineering decisions that were relatively costly for the WIPP project.   Microbial effects, for the important reasons of the higher temperature and much greater nutrient limitations, should have a much smaller effect on the actinide source term, but data that demonstrates this needs to be obtained to avoid unrealistic assumptions that may be required in the regulatory process.

Redox Chemistry in the Repository

The redox conditions in a repository are key in defining the multivalent radionuclide source term.   Specifically, there is great benefit to the repository safety case if it can be shown and argued that reducing conditions will prevail.   The salt geology helps this process by sealing the repository and thereby limiting oxygen availability but this by itself is not sufficient.   In the WIPP example, significant credit is take for the excess zero-valent iron that is inadvertently there as the container material for the emplaced TRU waste.   Without this iron, it would be much more difficult to prove that reducing conditions prevail.   Generally, a salt geology does not contain significant quantities of active reductants as can be found in other geologies such as clay or granite where ferrous minerals actively impose reducing conditions.   For this reason, the addition of redox-active constituents should be a central part of repository design so that reducing conditions are readily established after repository closure and self-sealing. These reducing conditions should prevail in the low-probability case of brine intrusion and lead to a significantly lower radionuclide source term with respect to key multivalent radionuclides.

The redox issue for HLW/SF is also more complex that in the WIPP case due to the much higher radiation levels expected and the higher temperatures that may be present.   In the case of higher temperature, the redox half reactions for actinides and iron (or any redox active mineral/component) will likely be quite close and their relative position may change with temperature.   In this context the impacts of temperature on the redox reactions and relative reaction pathways needs to be established and used as part of the selection process in repository design.   The impacts of radiolysis are appropriately ignored in the WIPP case due to the low overall

activity and the WIPP project has shown that the residual auto-radiolysis effects of Pu and Am are overwhelmed by the iron chemistry present. This, however, may not extend to the HLW/SF case where the overall radiation levels may be much higher. Radiolysis will primarily impact the oxidation state distribution of the multivalent radionuclides rather than their complexation/speciation, so it will be important to account for this impact as the source term is defined. Investigations that confirm that the redox mechanisms and controls that lead to highly reducing conditions in the WIPP, primarily reactions with Fe, extend to the higher temperatures and radiation levels expected in the HLW case need to be performed.

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