NRC Staff Comments on SRNL-STI-2015-00446

1. NRC supports DOE's efforts to better understand key radionuclide solubility at reasonable geochemical endpoints while attempting to produce representative conditions to the extent practicable. If DOE determines that solubility is risk-significant at an Eh/pH endpoint (e.g., at a reasonably bounding Eh endpoint for redox-sensitive radionuclides), DOE may then conduct additional experiments to identify the conditions where solubility is non-risk-significant (e.g., the Eh below which key radionuclide solubility decreases to non-risk-significant values while understanding any pH, complexing agent, etc., dependencies). For example, results of preliminary experiments indicated a risk-significant Pu solubility could occur. If real waste experiments show similar results, DOE could conduct additional experiments to show that key radionuclide solubility is non-risk-significant at conditions expected in the field (e.g., at Eh below the endpoint).

2. If there is a subset of realistic field conditions where solubility is not risk-significant, DOE should provide information to support if and when the favorable conditions will occur in the field, or show how other barriers can compensate for risk-significant solubility. Support can be provided through additional waste release experiments that attempt to mimic actual conditions in the field.

3. DOE has assumed that key radionuclides in the tank residual waste may be co-precipitated with iron. Further, the predicted solubilities for co-precipitated phases are significantly lower than the predicted solubilities for pure phases. These waste release experiments will provide effective solubilities, which may be representative of multiple phases. However, characterization data could help differentiate between potential multiple phases and therefore, reduce uncertainty in the long-term projections of radionuclide release from the residual waste. Does DOE intend to conduct any characterization to provide additional information on potential various phases present in the residual waste?

4. Based on the risk significance of the solubility of the Tank 18 waste, NRC staff agrees with the need for conducting the Tank 18 waste release experiments. However, the solubility of the residual waste in other tanks is also risk significant and could be significantly different from Tank 18 residual waste. For example, Tank 18 did not undergo oxalic acid cleaning and the use of oxalic acid could increase the solubility of key radionuclides. It is not clear to NRC staff that the results from the Tank 18 experiments will represent or bound the potential solubilities of key radionuclides from other tank farm tanks. If residual waste samples with oxalates are not tested, the results of this testing will be inconclusive for tanks that underwent oxalic acid cleaning. Additional discussion on what experiments are planned for other tank waste or how the results from the Tank 18 experiments will be applicable to other tanks would be helpful.

5. As discussed previously by NRC staff in comments on SRNL-RP-2013-00203, NRC staff is concerned that the synthetic infiltrate water is not in contact with a grout formulation containing admixtures. Contacting the synthetic infiltrate with a grout formulation that is as similar to as-emplaced materials as is practicable is expected to reduce experimental artifacts and uncertainties. Does DOE plan on conducting any experiments with grout containing admixtures? If DOE is not planning on conducting experiments with admixtures present, is there any information to indicate that the admixtures in the tank grout will not increase the solubility of residual tank waste?

6. In several experiments, $CaCO_3$ was added to represent an endpoint of cement degradation. It is not clear to NRC staff how the results from the waste release experiments will be influenced by the potential lattice substitution of trace elements during the formation of $CaCO_3$ compared to sorption of trace elements onto the surface of pre-existing $CaCO_3$. Additional discussion on any potential experimental artifacts due to the addition of $CaCO_3$ would be helpful.

7. The authors state that in these experiments sand was not added as a component of the monolith since both fly ash and slag contain significant quantities of silicon. In Section 2.4, the authors note that the target concentration of silicon was well below the target concentration for the Tank 18 surrogate solids. Additional discussion on how this difference may or may not influence the waste release experiments would be helpful.

8. DOE should address inconsistencies between model and experimental results (see Tables 1 and 2 below) and understand if the inconsistencies are due to the inability to mimic actual field conditions or due to uncertainty in the modeling results, or both.

Table 1. Modeled Geochemical Conditions and Controlling Phases/Species

Chemical Condition	Eh (V)	pH	Eh	pH
Reduced Region II	-0.47	11.1	FeS_2 (pyrite)	JenH/TobD
Oxidized Region II	+0.56	11.1	Oxygen	JenH/TobD
Oxidized Region III	+0.68	9.2	Oxygen	OH-hydrotalcite

Table 2. Recommended Geochemical Conditions and Controlling Phases/Species

Chemical Condition	Eh (V)	pH	Eh	pH
Reduced Region II	-0.100*	10.5	FeS (ferrous sulfide)	$Ca(OH)_2/CaCO_3$
Oxidized Region II	+0.515	10.8	Oxygen	$Ca(OH)_2/CaCO_3$
Oxidized Region III	+0.523	9.4	Oxygen	$CaCO_3$
*-0.100 mV is an approximation based on three different test conditions for Reduced Region II, which include zero headspace and continuous inert gas purge methods. Both of these methods may be used for testing with actual Tank 18 residual waste.				