Processing and Disposition of Remote-Handled Transuranic Liquid Waste Generated at Oak Ridge National Laboratory – 16031*

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ABSTRACT

Oak Ridge National Laboratory (ORNL) develops radiochemical processes for implementation in the US Department of Energy (DOE) production facilities and produces radioisotopes for medical and industrial applications. These activities result in the production of remote-handled liquid waste that is transuranic (TRU) and requires treatment for disposal at the Waste Isolation Pilot Plant (WIPP). Capabilities are needed to characterize, treat, package, and dispose of remote-handled liquid waste generated from research activities at ORNL. A project to implement these capabilities has been recently initiated, and activities are under way to support remote-handled liquid waste treatment at ORNL. Experimental data as well as model predictions are being used to support the conceptual design of the treatment facility.

INTRODUCTION

Since the 1980s, ORNL has been collecting, concentrating, and storing liquid low-level waste in underground storage tanks for future treatment. Since that time, the liquid low-level waste system has accumulated large quantities of relatively low-activity waste from environmental management activities and small volumes of newly generated mission waste that contain high levels of radioactivity. During storage, the concentrated liquid low-level waste has segregated into a sludge containing the TRU constituents and a supernatant layer primarily containing cesium and some strontium. In 2003, the US DOE Office of Environmental Management constructed the Transuranic Waste Processing Center facility to treat this large volume of accumulated legacy liquid low-level waste as well as the legacy inventory of solid TRU waste. Most of the supernate has been removed from the underground storage tanks, solidified, and disposed of at the Nevada National Security Site (NNSS). The Transuranic Waste Processing Center site is now being upgraded to process the legacy sludge for disposal at NNSS. After treatment of the legacy sludge, the existing collection and treatment systems are planned for shutdown and deactivation and decommissioning (D&D). The remote-handled liquid waste generated in the future from ORNL nuclear research and radioisotope production is expected to be lower in volume but will have a higher radioisotopic

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concentration. Studies are under way to evaluate treatment options for ORNL's future mission-generated remote-handled liquid waste for disposal at the WIPP. Impacts of waste composition and radioisotopic decay on the ability of the waste forms to meet the WIPP waste acceptance criteria (WAC) and/or transportation criteria are being examined.

DESCRIPTION OF THE WASTE STREAM

ORNL is the production, storage, and distribution center for the heavy-element research program of the US DOE. One of ORNL's main missions is the production of transuranic isotopes. The facilities that support the production of transuranic isotopes will be the primary sources of remote-handled liquid waste generation in the future at ORNL. The stream is expected to contain ~580,000 nCi/g of a wide range of radionuclides, including ~12,000 nCi/g of TRU, and will require treatment and disposal at WIPP. The estimated average concentration of select isotopes and chemical species in future ORNL remote-handled liquid waste is given in Table I.

TABLE I. Expected composition of future ORNL Remote-handled Liquid Waste

Example Isotopes	Concentration (Bq/L)	Chemical Component	Concentration (M)
Sr-90	3 E+08	Na_2CO_3	0.13
Cs-137	1 E+09	NaOH	0.19
Ru-103	2 E+09	NaCl	0.10
Ru-106	4 E+09	$NaNO_3$	0.49
Ce-144	4 E+09	$NaAlO_2$	0.03
Eu-154	3 E+07		
Pu-238	4 E+08		
Cm-244	1 E+07		
Zr-95	3 E+09		
Total	**2 E+10**		**0.94**

EVALUATION OF WASTE TREATMENT FLOW SHEET OPTIONS

Three flow sheets were evaluated for treatment of ORNL future remote-handled liquid waste [1], as shown in Fig. 1:

a. evaporation and solidification in a grout matrix with disposal of the solidified waste form at WIPP,
b. evaporation to a solid salt cake waste form for disposal at WIPP, and
c. separation of the TRU and high-gamma isotopes before solidification of the bulk liquid waste stream in a grout matrix for disposal at NNSS. The concentrated gamma liquid waste stream would also be treated for disposal at NNSS, and the concentrated TRU stream would be solidified for disposal at WIPP.

Each of the flow sheets shown in Fig. 1 includes an evaporation step before the solidification step to reduce the volume of the waste form requiring disposal. The preliminary designs for these flow sheets include potential interim storage of the liquid waste before solidification. The designs also assume that the waste stream would not be concentrated to the point that solids are formed at room temperature. This would allow the waste stream to be stored and processed before the solidification step without the presence of solids, which would reduce plugging and fouling problems in the process equipment and would likely make waste certification easier.

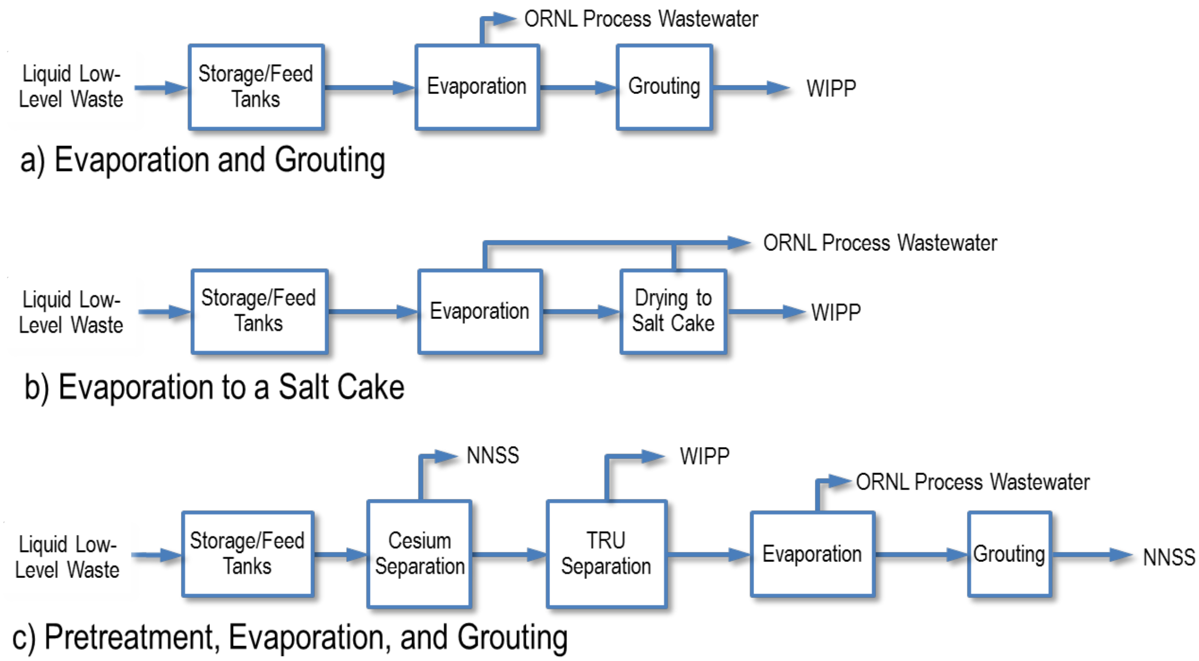

Fig. 1. Proposed Treatment Options for ORNL Remote-handled Liquid Waste.

Experimental tests with simulated waste indicate that a volume reduction factor of ~5 could be achieved for each of the evaporation steps without precipitation. Assuming that grouting the remote-handled liquid waste concentrate results in a volume increase of 1.6 and that the salt cake waste form can be further concentrated after evaporation to a specific gravity of 1.5, the final bulk waste forms for the flow sheets shown in Fig. 1 will be

a. 30% of the original remote-handled liquid waste volume for a grouted waste form for disposal at WIPP,
b. 4% of the original remote-handled liquid waste volume for the salt cake waste form for disposal at WIPP, and
c. 30% of the original remote-handled liquid waste volume for the pretreated grouted waste form for shipment to NNSS if sufficient TRU separations can be achieved to make the bulk waste non-TRU.

Preliminary experimental data [2] shows that ~95% of the TRU components, 97% of the rare earths, and 99.9% of the cesium and strontium can potentially be removed by pretreatment. At these removal levels, the bulk pretreated remote-handled liquid waste would still be transuranic and would not be eligible for disposal at NNSS as proposed in the Fig. 1 (c) flow sheet. A TRU removal efficiency of 99.2% would have to be achieved to make the as-generated remote-handled liquid waste a non-TRU waste. For the Pu-238 isotope, which has a low TRU waste concentration threshold, the concentration would need to be reduced from ~600 ppb to <6 ppb for the stream to be non-TRU; the experimental pretreatment system would have only reduced the Pu-238 concentration to ~34 ppb.

The grout flow sheet shown in Fig. 1(a) is the most commonly used treatment method for liquid low-level waste streams; it would require the least amount of additional development effort. The salt cake flow sheet (Fig. 1[b]) has the advantage of potentially reducing the amount of waste destined for WIPP by a factor of ~7, but it would require more development work than the grout flow sheet. Initial evaluation of the pretreatment flow sheet (Fig. 1 [c]) has indicated that pretreatment will not result in making the bulk of the waste non-TRU, and disposal of the concentrated waste forms from pretreatment will be problematic. Alternative approaches to this flowsheet include segregation at the source of generation rather than pretreatment of the bulk waste stream. Either approach to this flow sheet would require considerably more development work before it could be considered for implementation. While all three flow sheets require additional development work, this paper focuses on further refining the conceptual design of the treatment facility for the grout flow sheet shown in Fig. 1(a) for disposal of remote-handled TRU waste at WIPP.

OPTIMIZATION OF GROUTING FLOW SHEET

The isotopic composition of the future remote-handled liquid waste stream impacts the ability to meet open road transportation limits and the WIPP WAC for the standard configuration of three 55-gal drums transported in a 72-B carrier. The parameters evaluated include the hypothetical accident condition limit for the 72-B, the decay heat limit for the 72-B, the surface dose of the 72-B, the surface dose of the unshielded 55-gal drums, the minimum transuranic content of the waste form, and the maximum curie limit of the waste form. Typically, the criterion that limits the concentration of ORNL remote-handled liquid waste in the final waste form is the surface dose for the unshielded 55-gal drum.

The isotopic concentrations of the future remote-handled liquid waste given in Table I are for the waste as it is generated at the ORNL research facilities. This newly generated waste contains short-lived isotopes, such as Zr-95 and Eu-154, that contribute significantly to the surface dose. Their concentrations will decrease fairly rapidly over time. The curve in Fig. 2 shows the impact of storage time for radionuclide decay on the volume of the final waste form required to meet the WIPP

WAC. It represents the maximum volume reduction that will result in a waste form that meets the WIPP WAC for a waste stream having the composition of the newly generated ORNL remote-handled liquid waste as a function of radioactive decay time. The dramatic reduction in the final waste form volume occurs during the first 2 years of storage; little additional volume reduction is achieved with increased storage time after ~4 years. A waste form falling above the curve in Fig. 2 would exceed the WIPP WAC and would require additional storage for decay. Any waste form falling below the curve would meet the transportation and disposal limits, so additional storage time would not be beneficial.

It is estimated that it will take a least a year to generate the remote-handled liquid waste and process it into a waste form. It is also likely to be staged in interim storage for 0.5–1 year awaiting pickup for transport to WIPP. The minimum volume of waste that could be shipped to WIPP after 1 year of decay would be ~15% of the original waste volume; a 2-year decay period would result in a minimum of ~10% of the original waste volume being shipped.

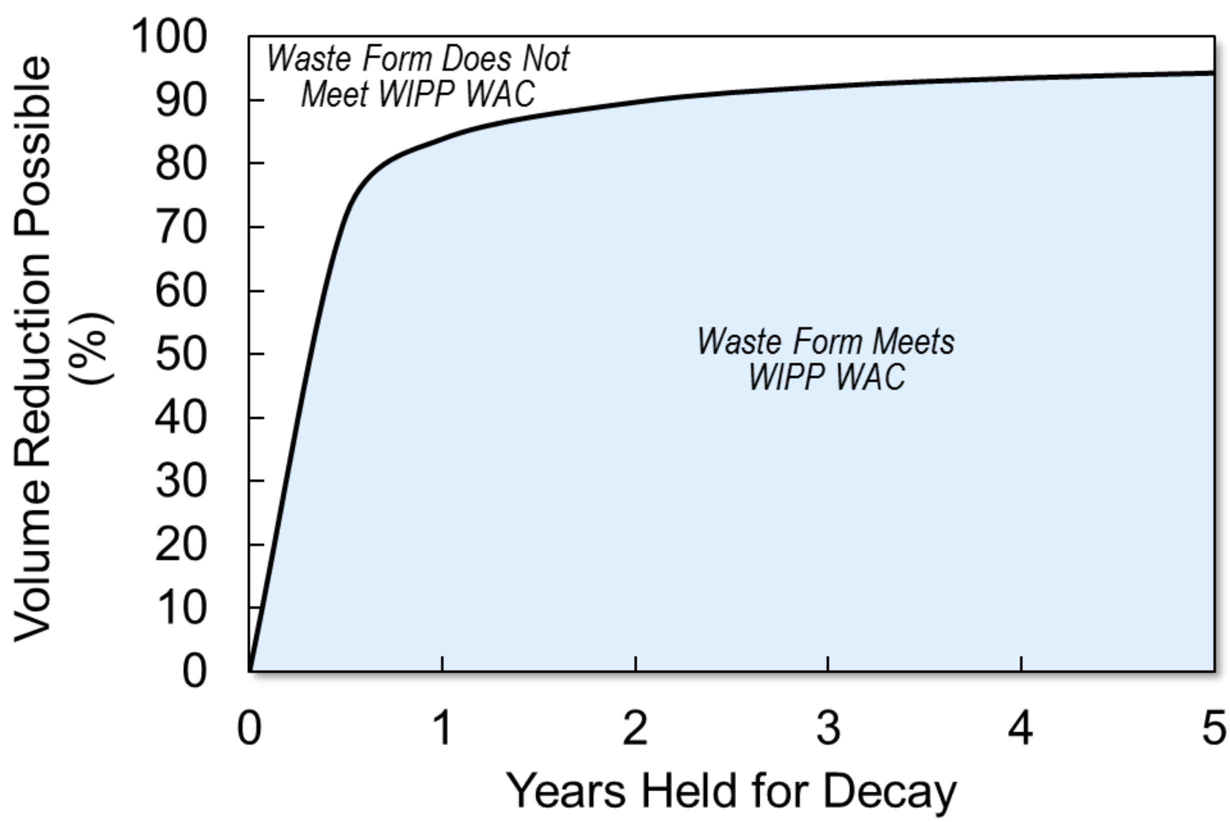

Fig. 2. Impact of Radioactive Decay on Waste Form Volume.

The grout flow sheet produces a waste form that is ~30% of the original waste volume. This is approximately double the minimum volume that could be shipped to WIPP if the remote-handled liquid waste decayed for a year and increases the volume approximately three times if the remote-handled liquid waste decayed for 2 years before shipment. Efforts are under way to evaluate potential ways to reduce the volume of grouted waste to be within the minimum range required for shipment to WIPP after a radioactive decay period of 1–2 years.

The preliminary grout flow sheet designs assume that the waste stream will be concentrated by a factor of 5 to the point that solids will not be formed prior to solidification to reduce plugging and fouling problems in the process equipment and to make waste certification easier as described in detail below. Two avenues are being considered to reduce the volume of waste requiring disposal by an additional factor of 2 to 3: (1) investigating the solubility limits of the remote-handled liquid waste and (2) investigating design of equipment that will allow successful handling of solids in the waste stream.

The aluminum content is a significant consideration regarding the potential for solids precipitation in liquid streams with a high salt content. The aluminum content of remote-handled liquid waste is expected to increase from its present value of 0.019 M to 0.034 M, and this may impact solids formation in the future remote-handled liquid waste.

To investigate the potential for solids formation, experimental tests were performed with remote-handled liquid waste simulant having the chemical composition shown in Table I but with an initial aluminum concentration varying from 0.02 to 0.6 M [1]. Precipitation did not occur when the simulants were produced, and precipitation has not been detected in the simulants as they have sat over time. These simulants were further concentrated by evaporation. Precipitation occurred in the samples with initial aluminum concentrations of 0.02 M, 0.1 M, and 0.6 M at volume reductions of 4.6, 2.9, and 2.1, respectively.

The saturation points for these three ORNL simulants are shown graphically on Fig. 3, compared with predicted solubilities of aluminum species as a function of sodium hydroxide concentration [3,4]. The curve with the dotted line in Fig. 3 is based on literature values for gibbsite solubility in a Na_2O-Al_2O_3-H_2O system at 30°C; this curve predicts that precipitation could occur in the future as-generated remote-handled liquid waste, and additional precipitation of aluminum hydroxide would be expected if the waste stream is concentrated by evaporation. The solubility curve with the solid line is for a simulated waste stream that contained saturated sodium nitrate, sodium sulfate, and sodium carbonate in addition to sodium hydroxide and aluminum compounds for data taken over a temperature range of 20–80°C. This curve suggests that the presence of other chemical compounds in solution shifts the aluminum solubility curve to the left, allowing aluminum to remain in solution at lower hydroxide concentrations.

Although the data points for the ORNL simulants are in the extreme lower left part of the graph, which is not covered well by the two solubility curves found in the literature, the data points indicate that the other salts in the ORNL RLW could potentially allow more aluminum to stay in solution for the range of expected RLW sodium nitrate concentrations. Additional tests are recommended on both RLW simulants and actual waste samples to determine the solubility limits of the as-generated RLW and the waste stream after it has been concentrated by evaporation.

Aspects of equipment design that will successfully accommodate handling of solids in the waste stream are also under consideration. The preliminary grout flow sheet designs assume that the waste stream would not be concentrated to the point that solids will be formed prior to solidification to reduce plugging and fouling problems in the process equipment and to make waste certification easier. The preliminary design concept is that the waste forms will be certified for disposal based on chemical analysis of concentrated RLW in a holding tank upstream of the grouting system. It is assumed that multiple 55-gal drums of solidified waste would be produced from a single tank of concentrated waste. A particular concern for design of the RLW treatment system is how to reliably prove that the RLW in the holding tank is homogenized for chemical analysis and then uniformly transferred to the grouting process. Recent advances in Europe of

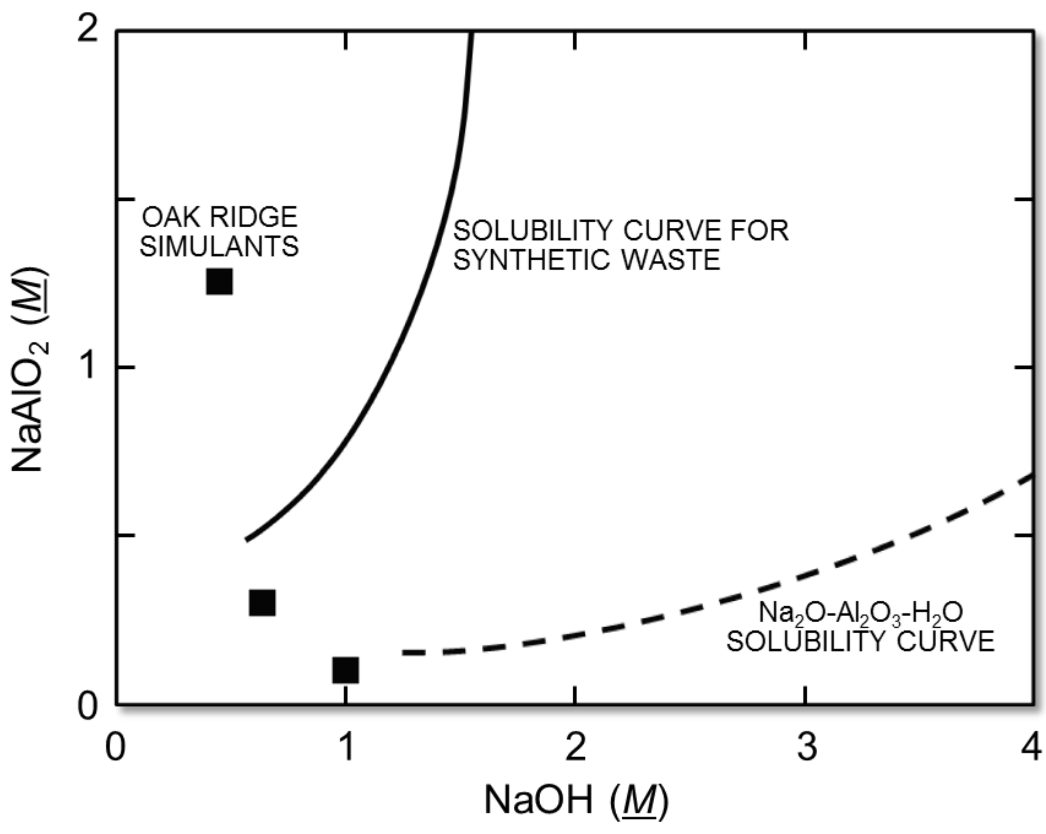

Fig. 3. Solubility Curves for Gibbsite.

process modeling and scale-up methods [5,6] are improving the designs of grouting systems for more reliable performance of tank mixing equipment and grouting systems to produce homogenous waste forms from waste streams containing solids. Recent literature publications indicate that careful selection and design of equipment could potentially result in a RLW treatment system that can operate in a remote environment and successfully produce a grouted waste form from a waste slurry that can be certified for disposal at WIPP. More investigation is recommended.

CONCLUSIONS

Studies are under way to evaluate treatment options for ORNL's RLW for disposal at WIPP. A conceptual design for a processing system consisting of evaporation and grouting has been selected for more detailed development. Impacts of chemical and radiological waste composition and decay time on the waste form and its ability to meet the WIPP WAC and/or transportation criteria are being examined. Evaluations to date indicate that a volume reduction factor on the order of 5 should be achievable when processing newly generated ORNL RLW without the production of solids. Studies show that storage of less than 1 year for radioactive decay of short-lived radionuclides is required for this amount of waste. Additional storage for radioactive decay of 0.5 years could reduce this volume by half, and storage for an additional year could reduce it by a factor of 3. Efforts are presently under way to evaluate the formation of solids during evaporation of RLW and identify equipment designs that could potentially allow these lower waste form volumes to be achieved.

REFERENCES

1. S. ROBINSON et al., "Processing and Disposition of Remote-Handled Transuranic Liquid Waste Generated at Oak Ridge National Laboratory," WM2015 Conference, Phoenix, Arizona (2015).

2. R. R. BRUNSON et al., *Operation and Testing of the Right Rack 7 Waste Treatment Module at the Radiochemical Engineering Development Center*, ORNL/TM-2005/64, Oak Ridge National Laboratory, Oak Ridge, Tenn. (2005).

3. D. A. REYNOLDS, *Practical Modeling of Aluminum Species in High-pH Waste*, WHC-EC-0872, Westinghouse Hanford Company, Hanford, Washington (1995).

4. J. G. REYNOLDS and D. A. REYMONDS, *A Modern Interpretation of the Barney Diagram for Aluminum Solubility in Tank Waste*, WRSP-44083-FP R0, Washington River Protection Solution, Hanford, Washington (2009).

5. C. MACQUERON et al., "Waste Homogenization Tank: Experimental Validation of a Numerical Study," Global 2015 Conference, Paris, France (2015).

6. E. TRONCHE et al., "Up-Scaling Methodology to Provide Knowledge for a Process Book: Application to a Cementation Process," Global 2015 Conference, Paris, France (2015).