

Feasibility Study of Direct Disposal of Electrorefining (ER) Salt Waste in a Salt Repository: Laboratory Tests

Fuel Cycle Research & Development

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SUMMARY

In FY10, the concept of direct disposal of salt from electrorefining (ER) of spent fuel into a salt repository was proposed. The theory is that the electrorefining salt waste would be thermodynamically stable in a salt repository, therefore potentially eliminating the need for waste form development for ER salt disposition by leveraging a key natural system attribute of a salt repository. FY12 work has been focused on laboratory testing and data collection on the leaching capability of surrogate ER salts and related waste forms in anticipated salt repository environments. Specifically, experimental investigation of factors affecting disposal of electrorefining salt in a generic salt repository was performed at Idaho National Laboratory (INL). Surrogate electrorefining salt samples, glass and ceramic waste forms were fabricated and assessed for leachability. At Sandia National Laboratories (SNL), the evaluation of the effect of radionuclide loading on salt encapsulation was conducted to confirm the threshold behavior of ER salt leaching as predicted by the percolation theory. In addition, a thermal gravity analysis was performed on water vapor sorption or condensation on surrogate ER salts. The key experimental results are summarized as follows:

- Given a typical repository time scale of 10 thousand to 1 million years, three materials tested - surrogate ER salt, tellurite glass, and sodalite – all dissolve very quickly in deionized water or WIPP brines (practically close to instantaneous release), although the latter two may have relatively slow dissolution rates on a laboratory time scale.
- The new experimental data on the leachability of salt composite materials prepared using WIPP salt further confirm the critical behavior of ER salt predicted with the percolation theory. The percolation thresholds are estimated to be about 10% and 20% initial iodine (I) loading, respectively, for evaporated and encapsulated salt samples.
- The different percolation thresholds imply that the threshold value is dependent on the scale and amount of heterogeneity present in the system at hand as well as the sample preparation procedures.
- For a WIPP salt sample, the amount of water adsorbed approached a plateau within ~ 6 days for all relative humidities except for 95% RH, which is consistent with the equilibrium RH (~75%) of a NaCl-saturated solution. When the environment has a RH higher than 75%, the moisture would continue to condense on salt surface to form a NaCl-saturated brine. Otherwise, the moisture will adsorb onto the salt surface, and the amount of water adsorbed is limited by the density of sorption sites.
- There is no adsorption plateau appearing for all relative humidities, because ER salt contains a highly hygroscopic Li-Cl salt. In comparison with WIPP salts, the ER salt has a much faster water adsorption rate.

It is planned that a comprehensive evaluation of the direct disposal concept will be performed in FY13 using the data collected in FY12. Additional data will also be collected on water uptake and desorption under elevated temperature conditions.

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Feasibility Study of Direct Disposal of Electrowinning (ER) Salt Waste in a Salt Repository: Laboratory Tests

1.0 Introduction

In fiscal year 2010 (FY-10), evaluation of the concept of direct disposal of salt from electrowinning (ER) of spent fuel was initiated (Wang, 2010). Electrowinning is currently being performed on spent fuel from the Experimental Breeder Reactor-II (EBR-II) using two systems. One is called the Mark-IV electrowinner (ER), which is used to process driver fuel containing high-enriched uranium. The other is the Mark-V ER, which is used to process blanket fuel containing depleted U and a relatively high concentration of Pu-239. There is approximately one metric ton (MT) of salt waste contained in these electrowinners. That amount will increase with further processing of spent fuel over the next 5 to 15 years. The salt is primarily LiCl-KCl in a eutectic ratio but also contains NaCl, UCl₃, and numerous fission product and transuranic chlorides. Active metal species from the spent fuel are converted to chlorides and partition into the salt phase during each electrowinning run. The current baseline for ER salt processing is to immobilize these salts into a glass ceramic waste form. The immobilization process consists of absorption of the salt in dehydrated zeolite-A followed by blending the salt-loaded zeolite in glass frit and finally heating to 915-950°C to form a ceramic waste ingot. The glass-ceramic waste form is comprised of sodalite regions interspersed in a glass matrix (Simpson and Law, 2010). Several steps are involved that require large equipment and significant space in a shielded, inert-atmosphere hot cell, which drives up the cost of the processing, since hot cell space is extremely expensive. The resulting ceramic waste form would then be considered high level waste (HLW) and eventually be shipped to a permanent geologic repository. There are concerns related to waste loading in the ceramic waste form, since lithium, potassium, and chlorine comprise a large mass fraction of the salt. Only a limited concentration of chloride can be loaded into the zeolite without impacting waste form durability. The other problem is that this process increases the mass of the waste by about a factor of 10 (Wang et al., 2011). For these reasons, alternative disposal approaches have been considered. As originally discussed in the FY-10 progress report (Wang, 2010), one interesting concept is to dispose of the salt directly into a repository in salt formation. The theory is that the electrowinning salt would be thermodynamically stable in a salt repository, potentially eliminating the need for waste form development for ER salt disposition by leveraging a key natural system attribute of a salt repository.

Furthermore, the direct disposal concept proposed may have an implication to the development of an advanced fuel cycle (Wang et al., 2011). In a closed or modified open fuel cycle, used fuels will be reprocessed and useful components such as uranium or transuranics will be recovered for reuse (e.g., Bodansky, 2006). Two general fuel reprocessing technologies are currently under consideration: the aqueous process and the ER process (e.g., Simpson and Law, 2010). For the case of aqueous reprocessing, used fuel is dissolved in nitric acid; the uranium (and also plutonium if needed) is then extracted through a complex set of aqueous chemical processes; and the fission products and minor actinides remain as wastes for permanent disposal. In the ER process, metallic used fuel (or reduced oxide fuel) is loaded into anode baskets of the electrowinner and lowered into a vessel containing molten chloride salt. When electric current is passed, the uranium, transuranics, and active metal fission products become oxidized and dissolved in the molten salt. Pure uranium is simultaneously recovered at the primary cathode where it deposits as a metal. A mixture of uranium and transuranics can be periodically recovered from the molten salt using a secondary cathode (e.g., Simpson and Law, 2010; Inoue and Koch, 2008).

The ER process has certain advantages over the aqueous process (Chang and Till, 1991; Chang, 2007). First of all, compared to the aqueous process, the ER is relatively compact and does not involve complex aqueous chemistry. Furthermore, since plutonium is always co-deposited with other actinides, the ER does not produce a pure plutonium product, which is a proliferation-resistant feature. Also, the ER process is highly compatible with metallic fuels needed for fast nuclear reactors, because the fuel components extracted from the ER will already be in a metal alloy form. Due to its compactness, the ER facility can be collocated with the fast reactor, thus eliminating the need for off-site shipments. In addition, for electrorefining all reagents such as electrolyte salts can be recycled, thus minimizing process waste streams (Chang, 2007). Thus, the ER seems to have a significant financial benefit over the aqueous process (Chang and Till, 1991; Chang, 2007).

Molten LiCl-KCl salt used for ER has high solubility for metal chlorides as well as high electrical conductivity and stability over a broad range of electrochemical potentials. As the ER runs, fission products and NaCl accumulate in the molten salt. The usable life of the salt may be dictated by one of the following concentration limits: (1) NaCl concentration in the case of sodium-bonded fuel due to an increase in melting point, (2) fission-product chloride concentration in the case of other fuel due to an increase in decay heat, and (3) plutonium concentration in the case of Pu-rich spent fuel (Simpson and Law, 2010). The accumulation of fission products and other contaminants in the ER salt eventually makes it necessary to remove salt for disposal. Based on the proposed direct disposal concept, such salt can potentially be disposed in a salt repository with no or minimal treatment. Therefore, there may be a synergistic effect among the development of fast reactor, the ER treatment of used fuel, and the development of a salt repository.

This report documents the FY12 work performed at Idaho National Laboratory (INL) and Sandia National Laboratories (SNL) toward a feasibility demonstration of the concept of direct disposal of ER salt in a salt repository. Using the experimental setup and procedures developed in the previous fiscal year, the FY12 work has been focused on laboratory testing and data collection on the leaching capability of surrogate ER salts and related waste forms in anticipated salt repository environments. Specifically, at INL, experimental investigation of factors affecting disposal of electrorefining salt in a generic salt repository was performed. Surrogate electrorefining salt samples and glass and ceramic waste forms were fabricated and assessed for leachability. Glass and ceramic waste forms were used as reference waste forms for a comparative analysis. At SNL, the evaluation of the effect of radionuclide loading on salt encapsulation was conducted to confirm the critical behavior of ER salt leaching as predicted by the percolation theory. In addition, a thermal gravity analysis was performed to evaluate water vapor sorption or condensation on surrogate ER salts. It is planned that a comprehensive evaluation of the direct disposal concept will be performed in FY13 based on the data collected in FY12.

2.0 Direct Disposal Concept

Durability and waste loading are two major attributes for waste form performance. Over the last few decades, significant effort has been devoted to develop durable waste forms (e.g., Peters and Ewing, 2007). However, it should be realized that the durability of a waste form is a relative concept and should be evaluated in the context of relevant disposal environments. In this sense, ER salt itself may be an ideal waste form for disposal in a salt repository. Using computer code EQ3/6 and the associated Pitzer parameter database (Wolery and Jarek, 2003), the saturation indexes of a brine (SB-3) from the Waste Isolation Pilot Plant (WIPP) with respect to various salt minerals have been calculated. As expected, the calculation shows that brine SB-3 is at or close to equilibrium with both major and minor minerals in the formation, including halite (NaCl), sylvite (KCl), glaserite [$\text{NaK}_3(\text{SO}_4)_2$], anhydrite (CaSO_4), and calcite (CaCO_3). Typical ER salt waste contains more than 50% of NaCl and KCl in the mass fractions. If it is

assumed that the rest of salt components are embedded as inclusions in the matrix of NaCl and KCl, the ER salt waste will be highly durable and even thermodynamically stable in a salt repository environment.

Direct disposal of ER salt waste in a salt repository also has other advantages. First of all, it does not require immobilization of salt waste into waste form matrix such as currently proposed glass-bonded sodalite (Simpson and Law, 2010). Therefore, the proposed concept will minimize the waste volume for final disposal. Based on composition of used fuel from Experimental Breeder Reactor-II (EBR-II) (Simpson and Law, 2010), it has been estimated that direct disposal can reduce waste volume by at least 10 fold as compared with the baseline approach. Increasing the waste content in the glass-bonded sodalite is possible, but it reduces the waste form durability (Simpson and Law, 2010). In direct disposal, however, high chloride concentrations in the salt would not pose any problem. Thus, direct disposal can potentially eliminate the need for all of the complex process steps currently proposed for waste separation and treatment. In addition, because of its plastic creep deformation behavior, salt as a waste form possesses a self-healing capability with regard to any mechanical or radiation damages.

On a microscopic scale, the effectiveness of ER salt as a waste form is expected to depend on the total volume fraction of NaCl and KCl in the salt. It is known that LiCl, another major component of the ER salt, is highly hygroscopic and may be unstable in brines from salt formations (Lide, 2003). The volume fraction of NaCl and KCl must be high enough to encapsulate radionuclides and mitigate the LiCl effect. The minimum volume of NaCl and KCl needed for effective encapsulation can be estimated from the percolation theory as shown schematically in Figure 1. For illustration purpose of the theory, consider LiCl and radionuclides as the waste component. At a low volume fraction of waste component, the embedded waste inclusions are separated from each other and completely isolated by the encapsulating matrix (NaCl and KCl). In this case, the leaching rate of the ER salt will be dictated by the dissolution kinetics of the matrix. Since both NaCl and KCl are stable in a salt repository, the dissolution rate is expected to be close to zero. As the waste component volume increases, the embedded inclusions become close to each other; and eventually at the threshold of waste loading, the inclusions are contiguous and form a connected network. Beyond this threshold, the leaching rate of the ER salt will be controlled mainly by the embedded inclusions. Therefore, the maximum volume of waste component is the value right below the percolation threshold. The actual threshold of a composite material depends on the geometry (e.g., shape and size) of the embedded phase. For spherical inclusions, the percolation threshold is calculated to be 29% volume fraction (Garboczi, 1995), implying that the minimum volume fraction of NaCl and KCl needed for effective encapsulation is approximately ~70%. Given a typical ER salt composition, recycle of certain amount of LiCl from ER salts may be necessary for direct disposal. The appropriate volume fraction can also be achieved by adding additional NaCl or KCl to ER salt waste. Determining the actual percolation threshold for ER salt is crucial for developing a source term model for the performance assessment of ER in a salt repository.

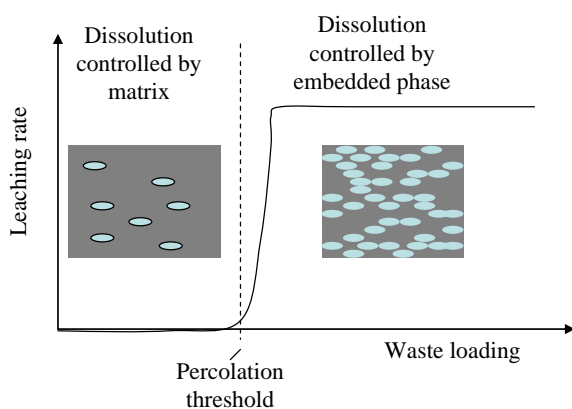


Figure 1. Schematic representation of leaching rate of ER salt waste form as a function of waste loading postulated from the percolation theory.

3.0 Leaching Tests of Surrogate ER Salts and Other Related Waste Forms

To test this theory, preliminary tests were run in FY10 to evaluate the stability of LiCl-KCl based salts in water and representative brine solutions. Samples of pure eutectic LiCl-KCl salt were subjected to solubility and dissolution rate tests. Brine solutions were prepared that attempted to mimic WIPP brines (G-Seep and ERDA-6). The compositions and properties of these brines are given in Table 1. For the FY10 tests performed, the pH and Eh properties of the brine solutions were not maintained.

Table 1. WIPP brine compositions (Wang et al., 2011)

	G-Seep	ERDA-6
B ³⁺ (mM)	20	63
Br ⁻ (mM)	10	11
Ca ²⁺ (mM)	20	12
Cl ⁻ (M)	5.35	4.8
K ⁺ (mM)	770	97
Mg ²⁺ (M)	1.44	0.019
Na ⁺ (M)	1.83	4.87
SO ₄ ²⁻ (mM)	40	170
TIC (mM)	10	16
pH	6.5	6.17
Eh (mV)		-152

For the solubility experiments, LiCl-KCl salt was incrementally added to known volumes of each of the three liquids (water, simulated G-Seep, and simulated ERDA-6) at room temperature. Solubility limits were established at the point where some residual salt remained undissolved. For the dissolution rate tests, eutectic LiCl-KCl beads with a uniform 1.3 mm diameter were added in small amounts to the liquids. The liquids were continuously stirred, and the time required to dissolve all of the beads was recorded. It was found that the LiCl-KCl solubility in the brine solutions was a factor of 5 to 7 lower than that in water. Dissolution rates of the salt were also lower in the brine solutions than that for deionized water by about a factor of 10.

In FY11, more representative salt compositions (not just LiCl-KCl) were used in experiments. Non-radioactive surrogate salts were used to minimize the cost involved with the testing. These surrogate salts were prepared by melting ingredient salt components at 500°C and cooled at different rates followed by examination using scanning electron microscopy (SEM) and x-ray fluorescence (XRF). The SEM images showed what appeared to be two solid phases, but there were no significant differences in elemental composition of the phases based on SEM/EDX point analysis. Thus the salt continues to be considered to be a homogenous material. The affect of removing LiCl from the salt was also experimentally studied, since it was hypothesized that the more hygroscopic LiCl might be the cause for rapid dissolution of the LiCl-KCl based salts in brine solutions. But there was found to be no correlation between LiCl content and kinetic or equilibrium properties of the salt in contact with the brine.

In FY-12, this investigation was extended to include a waste form that could be easily formed by melting the ER salt with glass pre-cursors and rapidly cooling to form a glassy waste form. During the initial

development of the ceramic waste process for ER salt, it was found that the salt was incompatible with borosilicate glasses. However, Pacific Northwest National Laboratory has recently found that tellurite glasses (78 % tellurium dioxide (TeO_2) and 22 % lead oxide (PbO)) are compatible with the salt and appear to form stable waste forms containing up to 10 wt% salt. This could represent an optimal trade-off between waste processing complexity and waste form performance. Thus, samples of surrogate electrorefining salt, tellurite glass with surrogate salt, and glass bonded sodalite with surrogate salt were prepared, evaluated for the stability, and subjected to leaching experiments. In addition to the valuation of the stability of the two types of waste forms stabilize the salt compared to direct disposal, the data collected in this year of the project can also be used to support FY13 performance assessment calculations for ER salt disposed of in a generic salt repository.

Experimental Methods

Surrogate Salt Preparation

Simulated electrorefining (ER) salt was prepared by mixing the individual salts shown in Table 2 in a magnesia crucible and heating them to 500°C for 14 hrs to allow for the LiCl-KCl to melt and dissolve the other salt compounds. All salt handling and heating was performed in an argon atmosphere glove box containing less than 10 ppm water and oxygen gas.

Table 2. Composition of surrogate electrorefining salt

	<i>LiCl-KCl</i>	<i>NaCl</i>	<i>CsCl</i>	<i>RbCl</i>	<i>SrCl₂</i>	<i>LaCl₃</i>	<i>CeCl₃</i>	<i>NdCl₃</i>	<i>YCl₃</i>	<i>NaI</i>
Mass (g)	13.846	23.077	4.615	1.154	2.308	2.308	3.462	6.923	1.154	1.154
Mass Fraction	0.231	0.385	0.077	0.019	0.038	0.038	0.058	0.115	0.019	0.019

This salt composition has a relatively high concentration of contaminants, which would be representative of salt waste that had been used to its operating limit in an electrorefiner and perhaps further concentrated using a controlled freezing process to recycle a portion of the LiCl-KCl to the ER. After the mass of salt cooled to room temperature, it was removed from the crucible and broken into smaller pieces. Images of the resulting surrogate ER salt are shown in Figure 2. After cooling, the salt was broken into smaller pieces and subjected to milling in a blade-type coffee grinder located inside the argon atmosphere glove box.



Figure 2. Photographs of surrogate electrorefining salt prior to grinding and sieving.

Sodalite Ceramic Waste Form Preparation

Zeolite-4A granules from UOP (45-250 μm in diameter) were heated to 300°C for 6 hours in the glove box for dehydration of water from the zeolite micropores. The dried zeolite-4A was then mixed with the surrogate salt particulate at a 9:1 mass ratio of zeolite (30 g) to salt. The salt/zeolite mixture in an MgO crucible, was heated to 500°C for 18 hrs to form salt-loaded zeolite-4A. The salt-loaded zeolite-4A was mixed with glass frit at a mass ratio of 3:1 and heated to 915°C in a graphite crucible for 8 hrs to form a glass bonded sodalite ceramic waste form with a total mass of 20 grams. The salt content in the ceramic waste form was 7.5 wt%.

Tellurite Glass Waste Form Preparation

The glass utilized for this portion of the research is a lead/tellurite-based powder. It is composed of 78 wt% tellurium dioxide (TeO_2) and 22 wt% lead oxide (PbO). A mixture containing 90 wt% of that glass precursor material and 10 wt% of the surrogate ER salt was blended together and heated to 700°C in a platinum crucible (Figure 3) for 40 min. After heating for 40 min, the molten glass was poured onto a stainless steel sheet for rapid quenching. All steps in this process were carried out in the argon atmosphere glove box.



Figure 3. Photographs of platinum crucible used for tellurite glass melting.

Simulated Brine Solution Preparation

In addition to deionized water, two simulated brine solutions were prepared for performing the leach experiments of the salt, sodalite, and glass waste forms. Table 3 gives the compositions of these brine solutions.

Table 3. Compositions of Simulated WIPP Brines (Wang et al., 2011)

	Simulated G-Seep	Simulated ERDA-6
NaCl (M)	1.83	4.714
MgCl ₂ (M)	1.42	
CaSO ₄ (mM)	20	12
KCl (mM)	760	86
MgSO ₄ (mM)	20	19
KBr (mM)	10	11
Na ₂ SO ₄ (mM)		139

Leach Test Procedure

After grinding and sieving each of the waste form (salt, glass, and sodalite) to less than 250 μm diameter particle size, 1.0 g of the waste form was placed in 50 mL of leachate. In total, there were six waste-form/leachant combinations. Samples of nominally 0.5 mL were acquired from each of the solutions every 24 hours over seven days of leaching. Diluted samples for ICP-MS analysis were prepared by combining 43 μL of the leachate and 12.0 mL of deionized water.

Results and Discussion*Surrogate Salt Dissolution*

Leach tests were not really possible with the unstabilized surrogate salt, because it very rapidly dissolves in each of the leachant solutions. This was re-confirmed with tests of surrogate salt with particle sizes less than 250 μm diameter in contact with water, ERDA-6, and G-Seep. The particles were visually observed while they dissolved into the solution. Complete dissolution was observed at the times given in Table 4.

Table 4. Results of directly adding surrogate salt (<250 μm) to leachate solutions

Leachate solution	Deionized water	Simulated ERDA-6	Simulated G-Seep
Dissolution time (sec)	60	330	360

For the case where deionized water was used as the leachant solution. The concentrations measured using ICP-MS were compared to the calculated concentrations based on mass of salt and volume of water. That comparison is shown to be excellent in Figure 4.

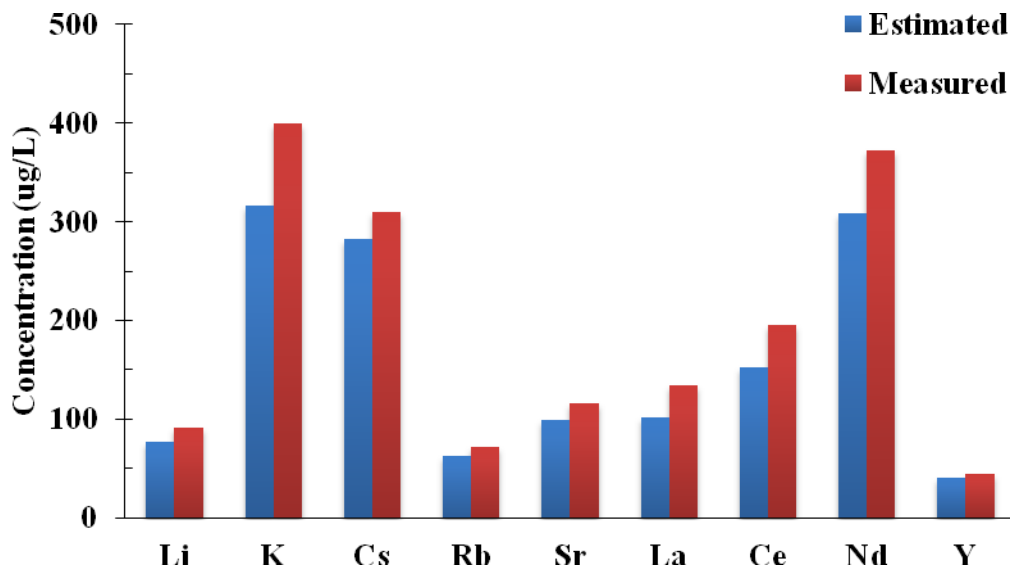


Figure 4. Concentration of leachate after complete dissolution of surrogate salt sample in deionized water- comparing the nominal calculated concentrations to those measured with ICP-MS

Leaching of Tellurite Glass Waste Form Sample

The photographs of the tellurite glass sample containing 10 wt% surrogate salt are shown in Figure 5. After crushing, milling, and sieving the glass waste form sample to particles less than 250 μm in size, leach tests were performed in deionized water, simulated ERDA-6 brine, and simulated G-Seep brine. Results are shown in the Figures 6 through 8. In these plots, the concentration of each species in the leachate solution is plotted versus time in days. Conditions for the leach tests are given in the experimental method section of this report. The potassium concentrations are plotted only for deionized water leaching, because K is a major component of both brine solutions. Its concentration dwarfs that of the other species and does not add value to waste form durability analysis.

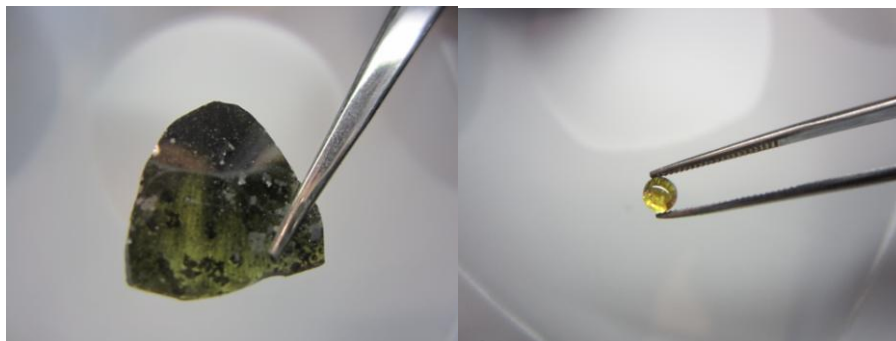


Figure 5. Photographs of tellurite glass sample containing 10 wt% surrogate ER salt

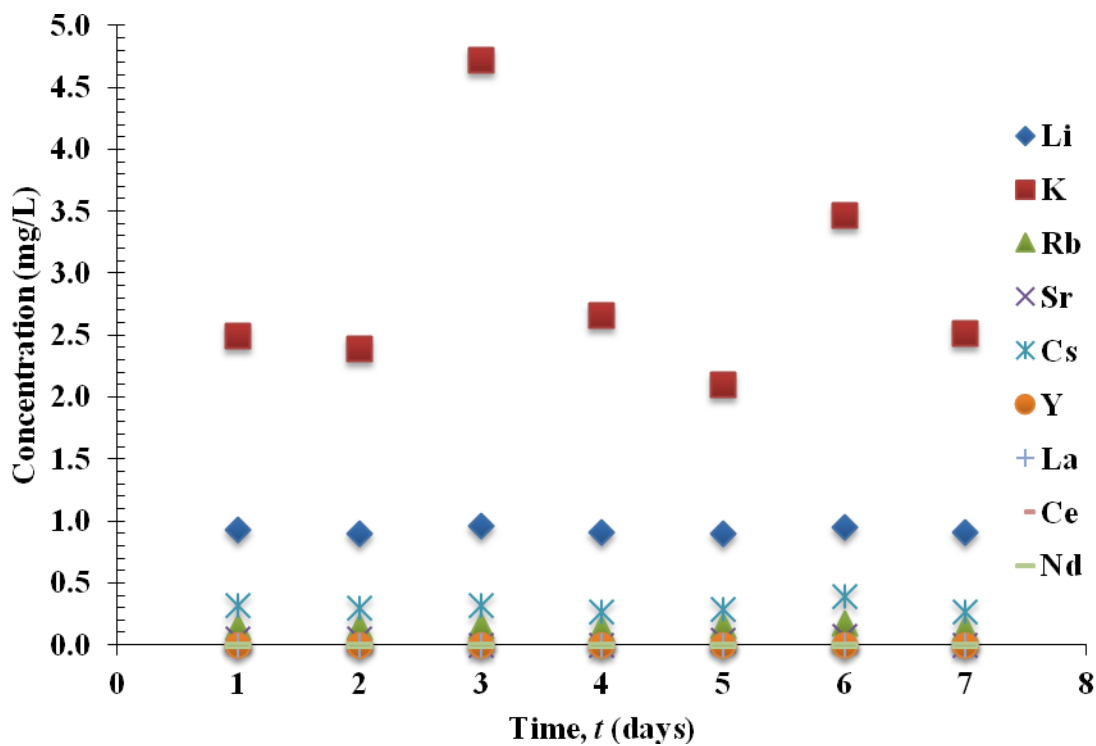


Figure 6. Leaching of tellurite waste form containing 10 wt% surrogate ER salt in deionized water

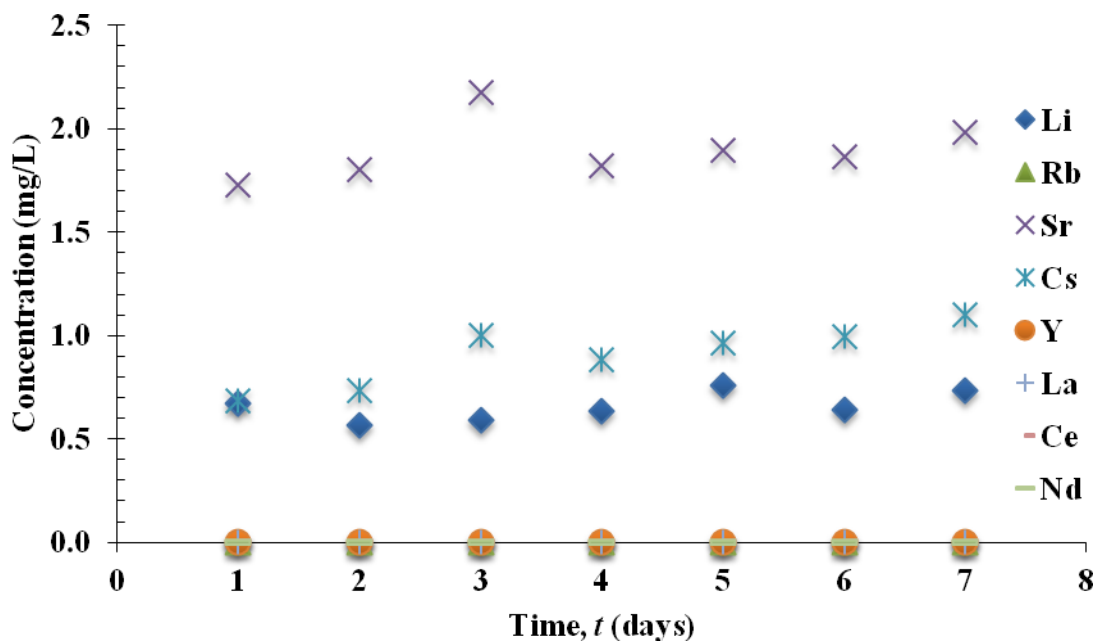


Figure 7. Leaching of tellurite waste form containing 10 wt% surrogate ER salt in ERDA-6

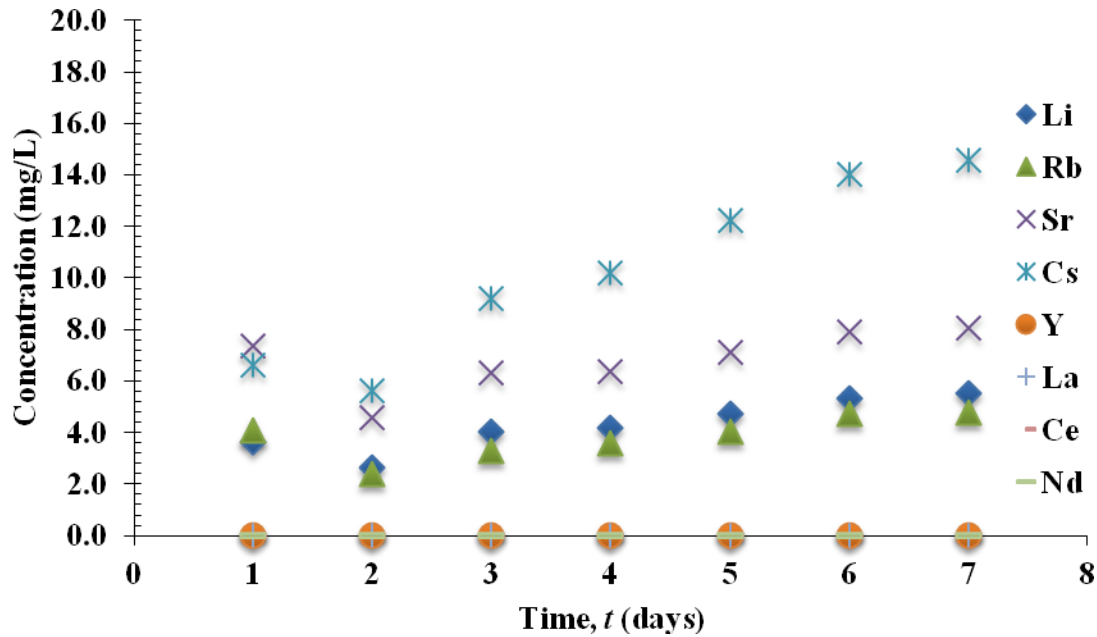


Figure 8. Leaching of tellurite waste form containing 10 wt% surrogate ER salt in G-Seep

Notice that there always appears to be a rapid increase in concentration in the first day of leaching. This is likely due to salt on the surface of the waste form particles that is readily accessible for rapid dissolution. The increase in concentration after the first day appears to be relatively slow for all elements except for Cs.

Leaching of Glass Bonded Sodalite Ceramic Waste Form Sample

The ceramic waste form sample containing 10 wt% surrogate ER salt is shown in Figure 9. After crushing, milling, and sieving the ceramic waste form sample to particles less than 250 μm in size, leach tests were performed in deionized water, simulated ERDA-6 brine, and simulated G-Seep brine. Results are shown in Figures 10 through 12. In these plots, the concentration of each species in the leachate solution is plotted versus time in days. Conditions for the leach tests are given in the experimental method section of this report. As for the tellurite glass waste form leach test results, the potassium concentrations are plotted only for the tests with deionized water.



Figure 9. Photograph of glass bonded sodalite ceramic waste form sample containing 10 wt% surrogate ER salt

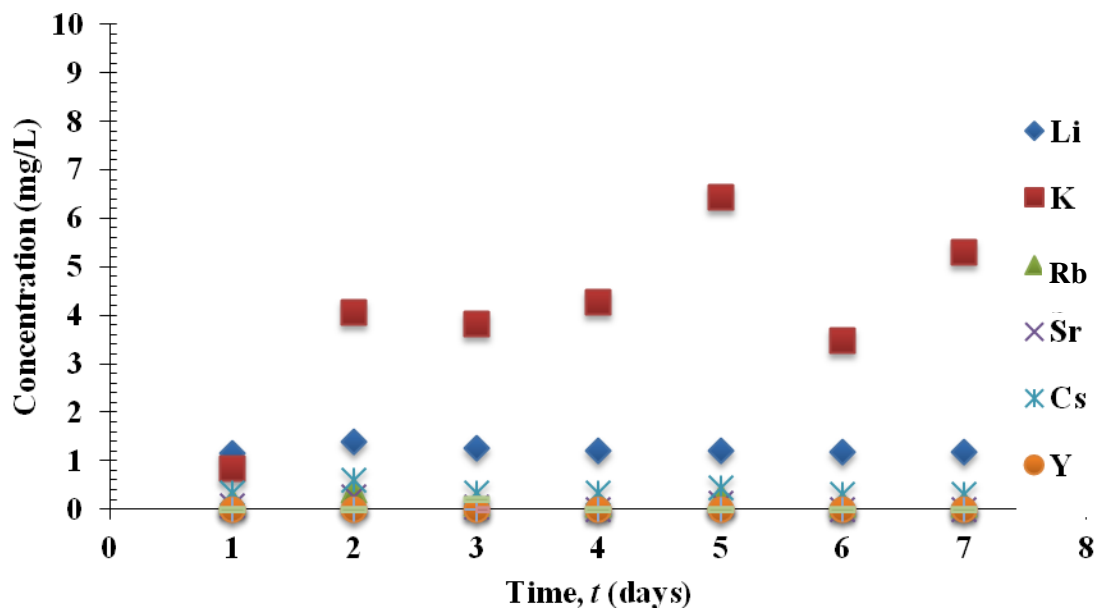


Figure 10. Leaching of sodalite ceramic waste form containing 7.5 wt% surrogate ER salt in deionized water

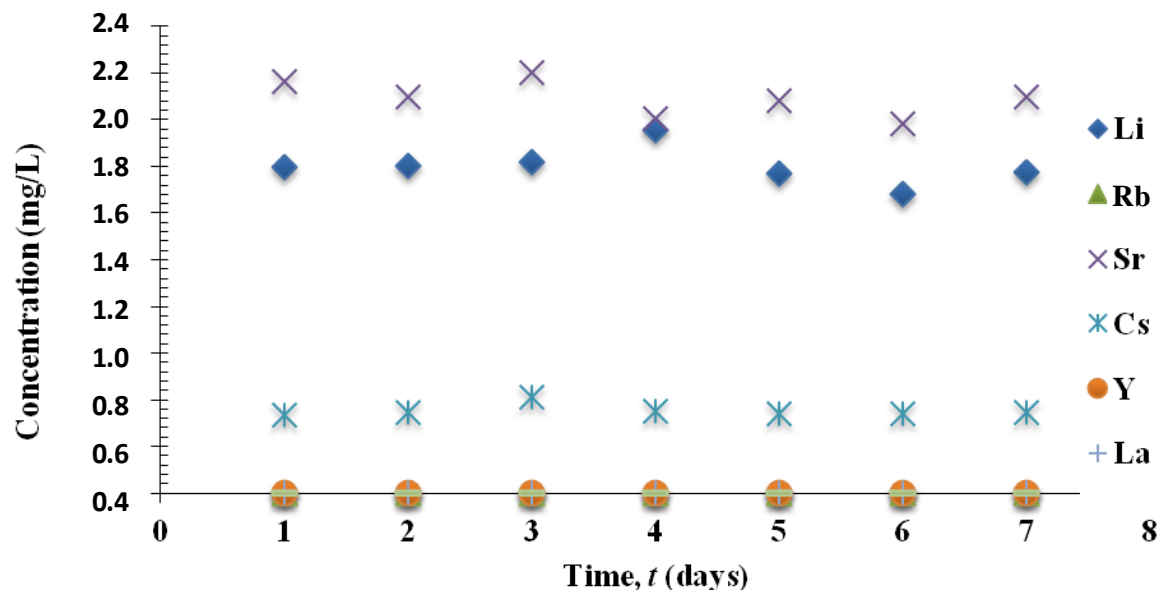


Figure 11. Leaching of sodalite ceramic waste form containing 7.5 wt% surrogate ER salt in ERDA-6 brine

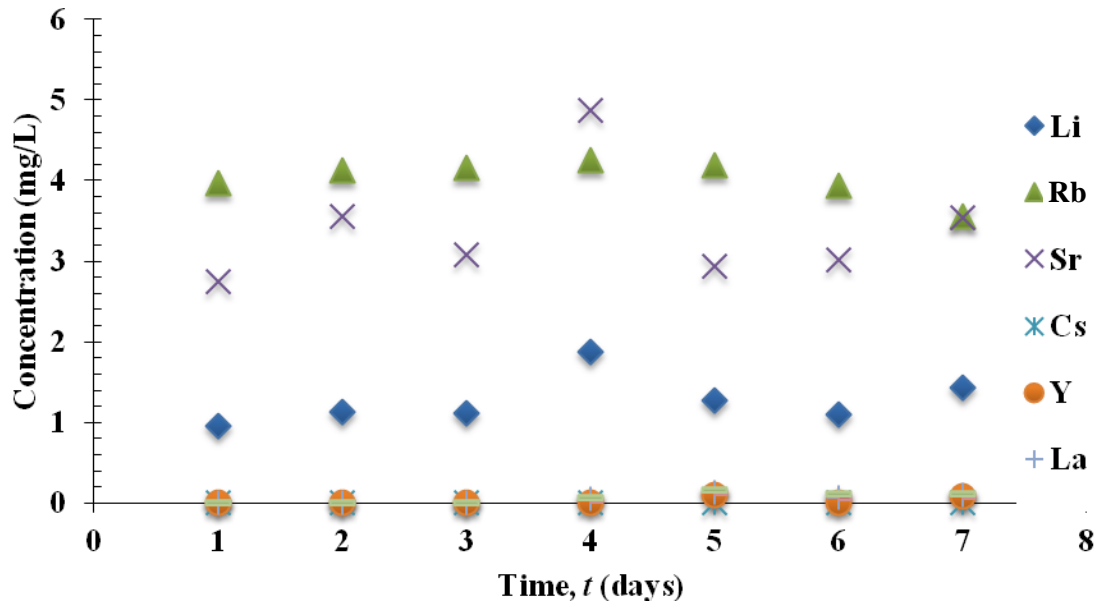


Figure 12. Leaching of sodalite ceramic waste form containing 7.5 wt% surrogate ER salt in G-Seep brine

As was the case for the tellurite glass waste form, there always appears to be a rapid increase in concentration in the first day of leaching. This is also likely due to salt on the surface of the waste form particles that is readily accessible for rapid dissolution. The concentrations appear to be stable after the first day. All release into the liquid appears to occur in the first day for each of the different types of leachant investigated.

Comparisons between Waste Forms

Figures 23 through 15 show the comparison between the two waste forms (tellurite glass and glass bonded sodalite ceramic). Data from each type of waste form are overlaid for comparative purposes. The values reported here are the mass of each element measured in the leachate solution divided by the initial mass of that element in the waste form sample. The waste form leach rates are relatively comparable, but the tellurite glass does appear to be generally more leach resistant based on the observation that the rate of release of salt constituents is lower for the glass than the ceramic. No appreciable increase in net leaching appears to be occurring from Day 1 through Day 7. This would imply that the major release mechanism during the test duration may be dissolution of salt on the surface of the sample particles (i.e., not actually encapsulated in the waste form solid matrix). Because tellurite is actually melted during its fabrication, it may be slightly more effective at occluding all of the salt into its matrix. When sodalite is formed, it is believed that some of the salt components are ejected into the glass binder phase. Since that glass binder was never heated to its melting point or higher, the salt may not be actually dissolved in the glass binder phase in the ceramic.

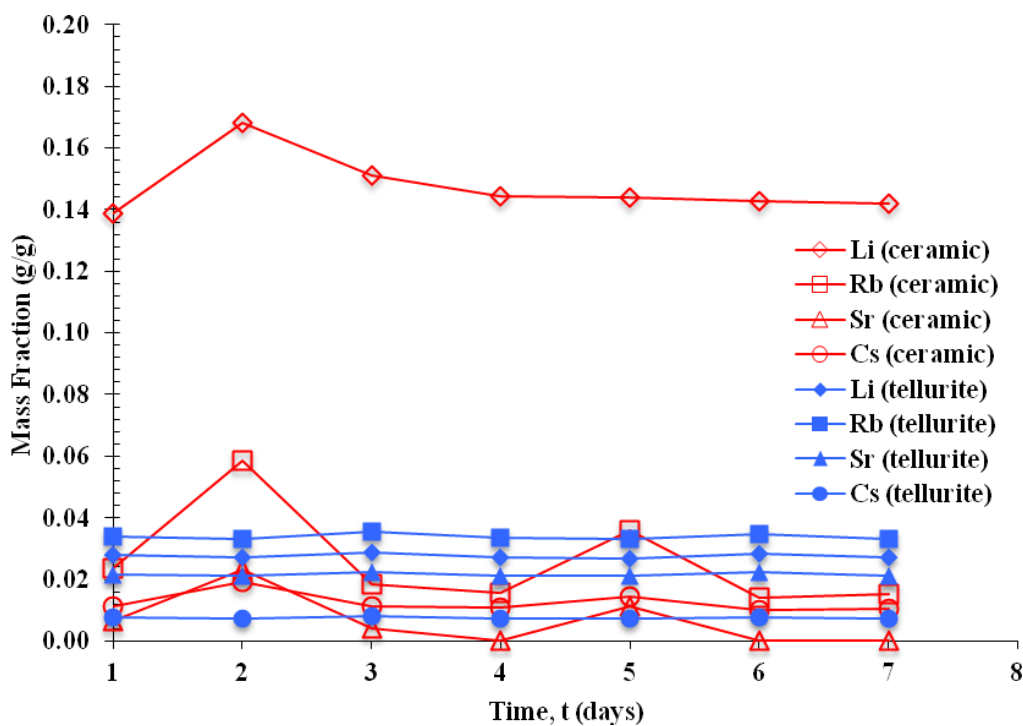


Figure 13. Leaching comparison between tellurite glass and glass bonded sodalite ceramic in deionized water

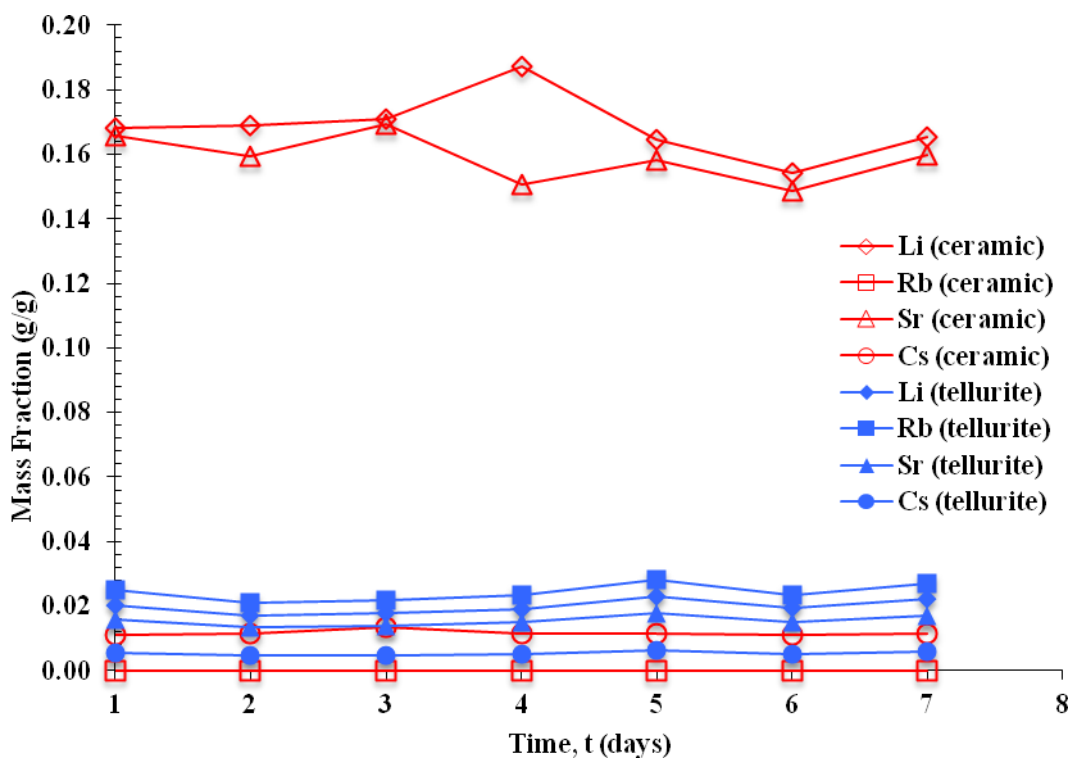


Figure 14. Leaching comparison between tellurite glass and glass bonded sodalite ceramic in ERDA-6

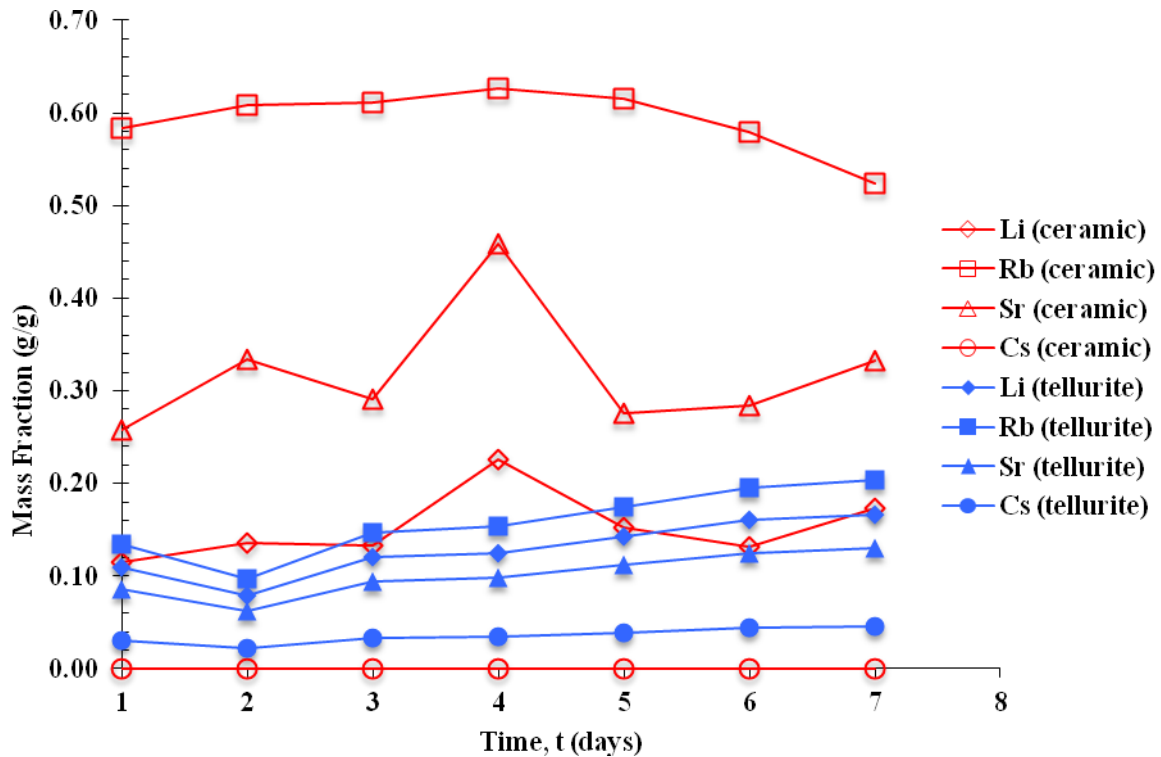


Figure 15. Leaching comparison between tellurite glass and glass bonded sodalite ceramic in G-Seep

In summary, experimental investigation of factors affecting disposal of electrorefiner salt in a generic salt repository continued in FY-12. In addition to surrogate electrorefiner salt samples, glass and ceramic waste forms were fabricated and assessed for leachability. The same three leachant solutions were used as in previous years of this project—deionized water, simulated ERDA-6 brine, and simulated G-Seep brine. In this year, the surrogate salt composition was made representative of waste salt after extensive electrorefiner operations and selective extraction of LiCl-KCl from the salt mixture. As in previous years, it was found that unstabilized salt dissolved quickly in a matter of minutes into each of the leachant solutions. In contrast, both the tellurite glass and sodalite waste forms have relatively low dissolution rates. Both waste forms performed comparably with the tellurite glass having a slight advantage. While tellurite is simple to produce, it does have the drawback of tellurium oxide being a very expensive material. The data from this study will be used in future performance assessment calculations to determine the long term benefit of the waste forms compared to direct disposed salt.

Based on these tests, two conclusions can be drawn. First of all, given a typical repository time scale of 10 thousand to 1 million years, the three waste form materials tested - surrogate ER salt, tellurite glass, and sodalite - all have very fast dissolution rates in WIPP brines, practically close to instantaneous release, relative to the repository time scale. Second, it is important to notice that materials used for these tests are fine particles less than 250 μm in size and the grinding process in sample preparation may have destroyed the original texture of the samples. Different from the other two waste forms, ER salt is a composite material with radionuclide-containing minor mineral phases encapsulated by Na-K-Cl salt matrix. In this case, the texture of the sample and the stability of matrix material are expected to play important role in determining the leaching behavior of ER salt (refer to Figure 1 and associated discussion). This effect is addressed in the next section.

4.0 Leaching tests on the effect of encapsulation and percolation

In many of the advanced nuclear reactor design concepts, it is anticipated that the electrorefining of used nuclear fuel will generate waste salt precipitates containing several possible fission products, most notably iodide. One possible route to disposition of this waste is disposal in a repository in salt formation. As discussed in Section 1.0, typical conceptual models for disposal of these salts require their placement in a robust waste form such as ceramics or glass. The addition of this material to the waste itself introduces a bulking factor of approximately one order of magnitude (Wang et al., 2011); by mass it requires about nine parts of waste form material to accommodate one part of ER salt waste. The creation of these waste forms also increases the total amount of handling and processing technologies required. The underlying assumption in waste form development is that relevant radionuclides could be incorporated into the lattice structures of one or multiple mineral phases that are supposed to be durable (i.e., with a low dissolution rate) to limit radionuclide release. As discussed in Section 2.0, if the ultimate disposition pathway is to place the waste in a salt repository, the validity of the disposal approach based on a durable waste form can be called into question. The Salado Formation, which is the salt bed surrounding WIPP, is stable on geologic time scales, and it can be surmised that a salt-based waste form with a composition similar to that of the salt host rock will also be thermodynamically stable on geologic timescales.

Despite this assumed stability, the waste loading into a salt based waste form can have significant impacts on the leaching rate of the waste. As shown in Figure 1, the hypothesis of the experiments described here is that there exists a threshold waste-loading value where the release rate of the waste switches from being controlled by the dissolution rate of the matrix encapsulating the waste to the dissolution of rate of the embedded waste itself. In order to evaluate the risk associated with such a disposal scheme, the dissolution rate of the source term must be known. Also, to mitigate the risk associated with the disposal option, it would be ideal to engineer the waste forms below the threshold value.

The experiments that we have designed are aimed to test this threshold value hypothesis, and will lead to a quantification of that threshold value. WIPP salt samples were procured from the WIPP site. This WIPP salt was mixed with varying amounts of sodium iodide (NaI). WIPP salt was used to mimic the major components of ER salt, and NaI was used to represent radionuclide-containing minor mineral phases. Two methods were used to prepare WIPP salt-NaI composite samples. One method was simply a physical mixing of the two different salts; the other method involved the dissolution of both the WIPP and sodium iodide salts in deionized water and co-precipitation via evaporation. These mixtures were compressed into pellets, and the pellets were subjected to leach tests with WIPP brine solution.

Methods

Preparation of WIPP Brines

Instead of using a synthetic brine, a saturated brine solution was prepared by dissolving WIPP salts in deionized water. An excess amount of WIPP salt was added to a known volume of deionized water. The solution and solid were allowed to mix from several hours to overnight. The remaining salt was removed by decantation and vacuum filtration. The solid material was allowed to air dry and massed. Working in several batches, a total of 2.85L of brine were prepared with an overall solids concentration of 360.36g/L. The three separate batches were mixed prior to the dissolution experiments. However, for the evaporative pellet formation, as described below, a single batch with a solids concentration of 358.18g/L was used. There is no detectable amount of iodide in the WIPP brine.

Pellet Formation

Two methods were used to prepare salt pellets for the leaching tests: encapsulation and evaporation. For the encapsulation pellets, the crushed WIPP salt was physically mixed with different weight percents of NaI. Once the two salts were well mixed, a hydraulic press and cylinder form was used to make the pellet. Three grams of mixed salt was added to the form; several drops of water were added to the salt before pressing. The small amount of water added allowed for a good and coherent compaction. The pellets were subjected to a load of 20,000psi for two minutes. The pellet was removed, and placed in a plastic container in which the dissolution experiment took place. Pellet diameter was fixed at 1.3cm by the form used in the press. However, the height of pellet was variable depending on the amount of NaI used; the general trend is decreasing height with increasing NaI content. The masses of salt components and the calculated weight percent of I in each pellet are shown in Table 5.

For the evaporative pellet formation, the target was to create about five grams of evaporate salts. A known amount of NaI was dissolved in 14mL of the brine described above. For the case with larger NaI weight percents (>20% wt%), a small amount of water (~1 - 2 mL) was added to NaI weight percents (>20%) to enhance dissolution. The solution was allowed to evaporate for several days at room temperature, but due to slow evaporation the samples were subsequently moved to a 45 °C oven overnight. No visual segregation of the evaporate salts was observed. The masses and the weight percent of I are summarized in Table 6. From the approximately 5g of evaporate obtained, 3g were used to make the pellet, as described above. A similar, but slightly subdued, trend was also observed between pellet height and the amount NaI added in the pellet.

Table 5. WIPP salt and NaI masses for encapsulation pellets

Nominal I Loading	Replicate A		Replicate B		Replicate C	
	Grams WIPP salt	Grams NaI	Grams WIPP salt	Grams NaI	Grams WIPP salt	Grams NaI
1%	2.9715	0.0301	2.9682	0.0307	2.9695	0.0306
20%	2.4001	0.6008	2.4014	0.6001	2.4002	0.5999
30%	2.1008	0.901	2.1005	0.9001	2.1001	0.9012
40%	1.8011	1.2013	1.8004	1.1998	1.8003	1.2003
50%	1.5004	1.5003	1.5019	1.4985	1.5025	1.4986
75%	0.751	2.2498	0.7497	2.2496	0.7498	2.2501

Table 6. Sodium iodide masses for evaporation pellets

Nominal I Loading	Target grams NaI	Exact mass NaI #1	Exact mass NaI #2	Exact mass NaI #3
1%	0.059	0.0588	0.0597	0.0596
5%	0.295	0.2959	0.296	0.2958
10%	0.591	0.5917	0.5915	0.591
20%	1.181	1.1815	1.1816	1.1825
30%	1.772	1.7734	1.7723	1.7729
40%	2.362	2.3628	2.3621	2.3615

Experimental Procedures

Once made, the pellets were immersed in 50 mL of the brine solution. For the most of test duration the samples are left quiescent at room temperature in the dark. The only time that the samples are actively mixed is during sampling. Prior to sampling, a transfer pipette is used to thoroughly mix the solution. For each sampling event, 0.5 mL of solution are removed, and placed in a 15ml centrifuge tube for storage. The sample is diluted with 5.0 mL of deionized water giving a final dilution factor of 1:11. The samples are tightly capped and stored in a refrigerator for analysis. Samples were taken periodically for up to 18 days.

Results

The release rates for iodide from the encapsulated and evaporated pellets are shown in Figures 16 and 17, respectively. The percent of total I released is calculated from the total I released into solution normalized by the total I originally in the pellet. In both cases the total amount released is often much less than the theoretical maximum. For clarity, the percent of iodide released for the 20% I encapsulation and evaporation pellets are plotted as a function of time in Figure 18. In general for similar initial mass loading the encapsulation pellets release smaller amounts of iodide.

Dissolved iodide concentrations are shown as a function of initial mass loading in Figure 19 for all three replicates of both evaporative and encapsulated pellets. The evaporative pellets have much narrower error bars and much more predictable behavior than the encapsulated pellets. This is consistent with an increased amount of iodide physical heterogeneity expected in the encapsulated pellet formation process. However, when only a single replicate is plotted (Figure 20), the trend is much smoother. For a given amount of mass loading, the iodide concentration is usually higher in the evaporated pellets than in the encapsulated pellets. Figure 20 clearly demonstrates the critical behavior of ER salt leaching as predicted from the percolation theory (Figure 1). The percolation thresholds are estimated to be about 10% and 20% initial I loading, respectively, for the evaporated and encapsulated salt samples. These different values may imply that the percolation threshold is dependent on the scale and amount of heterogeneity present in the system at hand. Smaller waste particles more evenly distributed throughout the matrix leads to a smaller threshold value than when the waste is more locally concentrated in larger particles.

Apparently, the amount of I released is a function of the pellet preparation procedure. It can be assumed that the NaI is more evenly distributed in the evaporative pellets. The NaI near the surface can dissolve, opening up a pathway to the interior of the pellet, because of the low dissolution-percolation threshold. As the brine penetrates the pellet, more NaI can dissolve leading to higher total releases. In the encapsulated pellets, the amount of iodide released is theoretically limited by the amount that is initially exposed to the brine. As the NaI near the surface of the encapsulated pellet dissolves, the exposed surface (in low iodide loading densities) is more likely to be WIPP salt which may dissolve extremely slowly near the equilibrium state as it is in contact with its saturated brine. This could effectively limit the dissolution and release of the NaI leading to the lower release at equilibrium. The next logical step would be to take this information to develop a pellet formation method where the NaI is not present at the surface of the pellet, and instead is completely surrounded by the WIPP salt. This idea is simply reinforced in Figure 20 showing the difference between the pellet formation methods. The evaporative method is less efficient at controlling NaI release than is the encapsulation method.

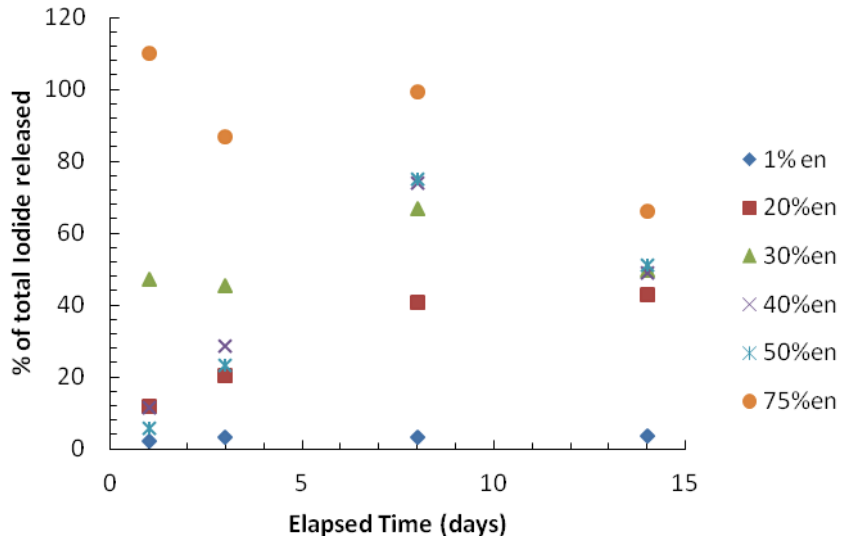


Figure 16. Percent of the total iodide released as a function of time for encapsulated pellets

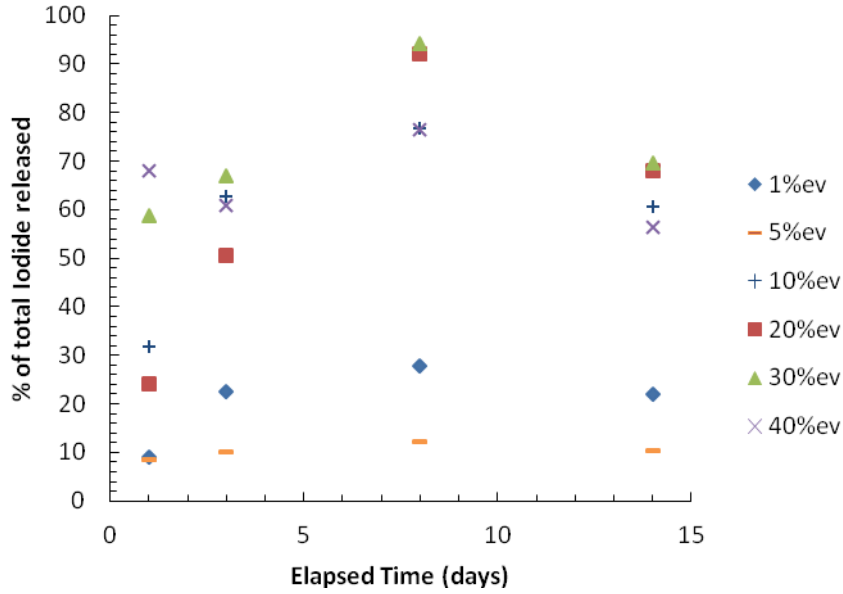


Figure 17. Percent of the total iodide released as a function of time for evaporated pellets

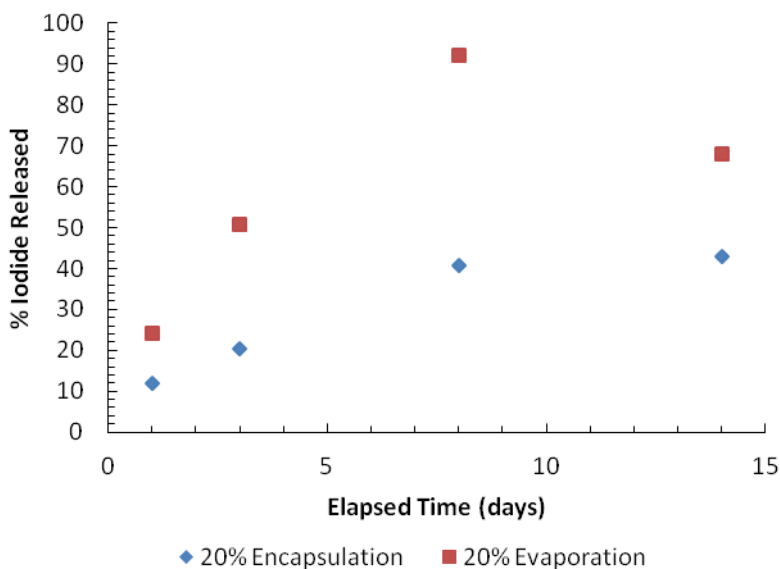


Figure 18. Comparison of the percent of I released between encapsulated and evaporated pellet formation

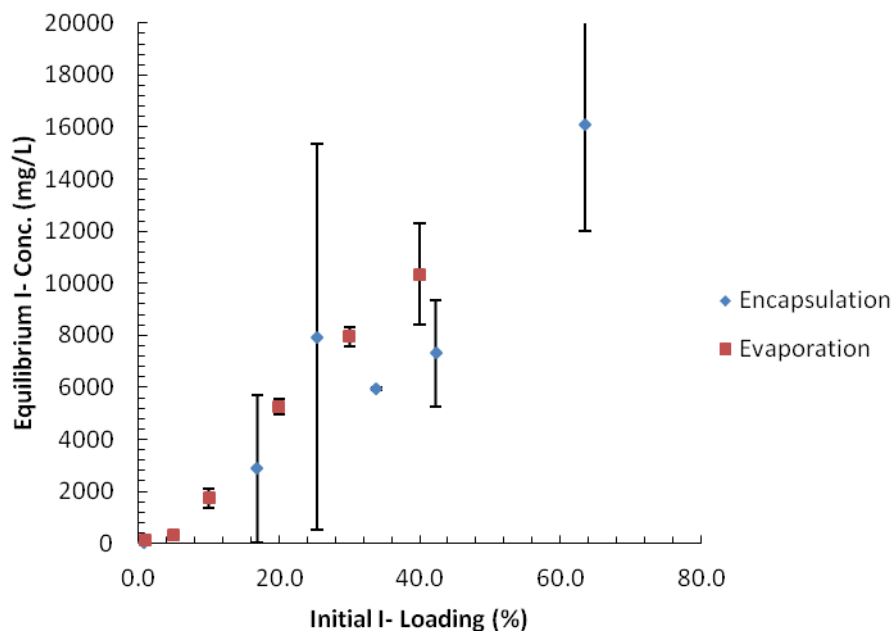


Figure 19. Equilibrium iodide concentration vs. initial iodide mass with error bars. Error bars represent plus/minus one standard deviation of the average of three pellets.

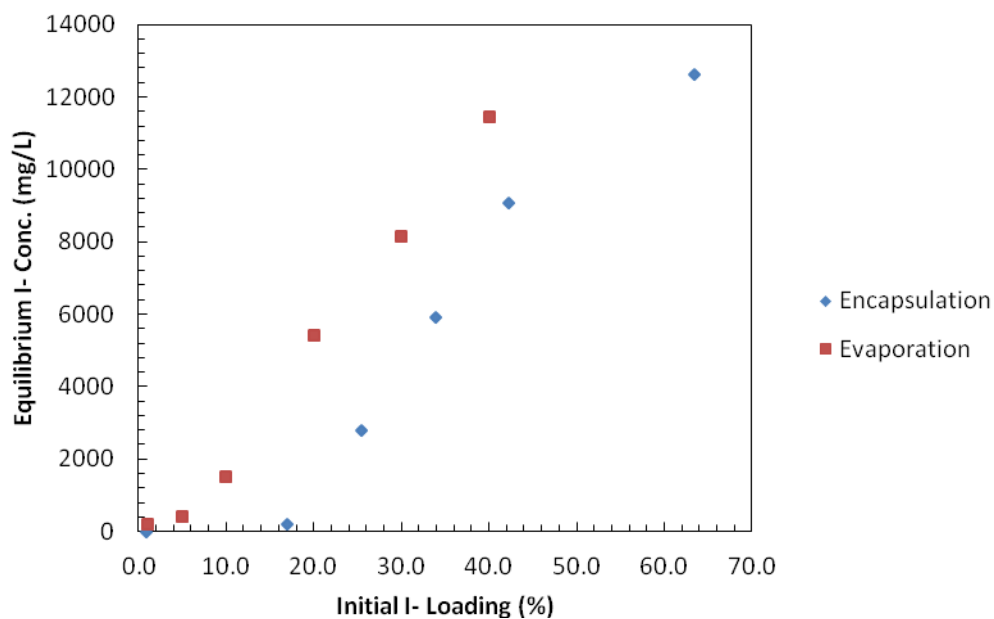


Figure 20. Iodide concentration vs. initial iodide loading; results from a singlet experiment.

The testing results point out the importance of textural characterization of actual ER salt. In order to actually predict the leaching behavior of ER salt in a salt repository, the following key information is required: (1) mineral phase distribution, (2) the geometry (e.g., shape and size) of embedded radionuclide inclusions, (3) the volumetric fraction of the embedded radionuclide inclusions in the salt, and (4) the related percolation threshold for salt dissolution. Such information may also help design an engineering approach to conditioning ER salt waste to obtain optimal performance for the long-term disposal of ER waste in a salt repository. For example, it is known that LiCl, another major component of the salt, is highly hygroscopic and may be unstable in brines from salt formations (Lide, 2006). The volume fraction of NaCl and KCl must be high enough to completely encapsulate LiCl salt phases and radionuclide inclusions, which can be achieved by either recycling LiCl or adding extra amounts of NaCl-KCl to ER salt waste. The key information to be obtained will allow us to estimate the minimal amount of LiCl to be removed from or the minimal amount of NaCl to be added to original ER salt waste.

5.0 Measurements of Moisture Adsorption on ER and WIPP Salts

To simulate adsorption of moisture by electrorefining waste salt during storage and transportation at different temperatures and various relative humidities (RH), experiments were conducted at temperatures of 9 °C, 25 °C, and 35 °C with RH ranging from 33% to 95%, simulating climate in winter, spring and fall, and summer, respectively. Such data will be useful for modeling water vapor transport in the near field of a salt repository. In FY12, the related effort was focused on the analytical technique development for the measurements. The measurements were performed without an explicit consideration of the effect of radioactive decay heat, that is, samples were not heated to high temperatures. This effect will be addressed in FY13.

Experimental Methods

Two salts were used in experiments: the analog ER salt and the WIPP salt. The WIPP salt was crushed and sieved as different size fractions including US Standard < 60 Sieve Number (size < 0.250 mm), US Standard 60 Sieve Number (0.250 mm < size < 0.425 mm), US Standard 40 Sieve Number (0.425 mm < size < 0.710 mm), US Standard 25 Sieve Number (0.710 mm < size < 1.18 mm), and US Standard 16 Sieve Number (1.18 mm < size < 2.30 mm). Because there were only small amounts of the analog salt, it was crushed and ground, but not sieved.

Before the water adsorption experiments, the salt samples were placed into an oven at 90 °C overnight to remove any moisture from the samples that might be present. Experiments were set-up in the following steps. First, the masses of empty glass vials were measured by using a balance accurate to subgram values (precise to four decimal places). Second, exact amounts of salts (about 5 grams) were weighed out, and placed in glass vials. Third, glass vials with salts were placed into a larger plastic container. Fourth, a separate glass beaker containing a saturated RH buffer solution was also placed in the plastic container. Four saturated RH buffers were used. They were a saturated K₂SO₄ solution with a RH of 95%, a saturated NaCl solution with a RH of 75%, a saturated NaBr solution with a RH of 58%, and, a saturated MgCl₂ solution with a RH of 33%, all at 25 °C. Finally, the plastic container was sealed. Experiments were set-up as replicates. After the desired equilibration time, the plastic container was opened, and the masses of the glass vials with salts were measured. Then, the amounts of moisture adsorbed were determined by subtracting the original sample mass.

Results

The experimental results are presented in Figures 21 through 24. In Figure 21, moisture absorption by the analog ER salts as a function of time at 25 °C and at various RH is shown. It is shown in Figure 21, adsorption has a strong dependence on RH, with the experiment at RH = 95% having the highest amounts of moisture absorbed. Notice that there is a gap between 10 and 25 hours; this gap occurred overnight after work hours. Figures 22A through 22E display experimental results for different size fractions of the WIPP salt at 9 °C. It seems that the effect of size on absorption is not significant. In comparison with the results for the analog ER salts, the absorption kinetics for the WIPP salt is much slower.

Similarly, Figures 23A through 23E, and Figures 24A through 24E display experimental results for different size fractions of the WIPP salt at 25 °C and 35 °C, respectively. In comparison with the results at 9 °C, it seems that absorption increases with temperature. Notice that for the size fractions larger than 0.425 mm, the reproducibility for experiments at RH = 95% was not good. This may be due to heterogeneous distributions of impurities in different samples, as the WIPP salt is known to have gypsum (anhydrite), polyhalite and clays as impurities.

It is interesting to notice that, for a WIPP salt sample, the amount of water adsorbed approached a plateau within ~ 6 days for all relative humidities except for 95% RH, which is consistent with the equilibrium RH (~75%) of a NaCl-saturated solution. When the environment has a RH higher than 75%, the moisture would continue to condense on the salt surface to form a NaCl-saturated brine. Otherwise, the moisture will adsorb onto the salt surface, and the amount of water adsorbed is limited by the density of sorption sites. For an ER salt, as shown in Figure 21, there is no adsorption plateau appearing for all relative humidities, because ER salt contains highly hygroscopic Li-Cl salt.

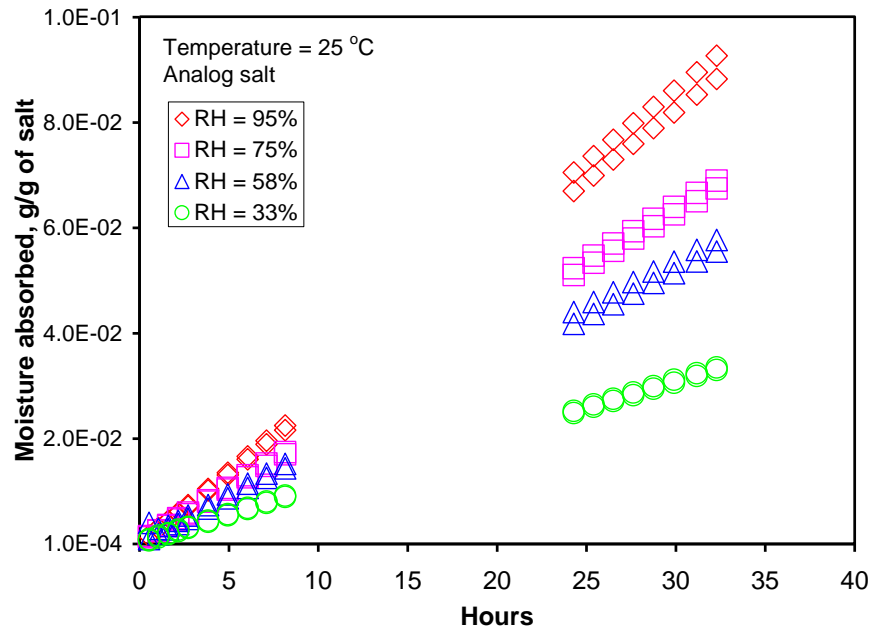


Figure 21. Sorption of moisture by analog ER salt as a function of time at different relative humidities at 25 °C

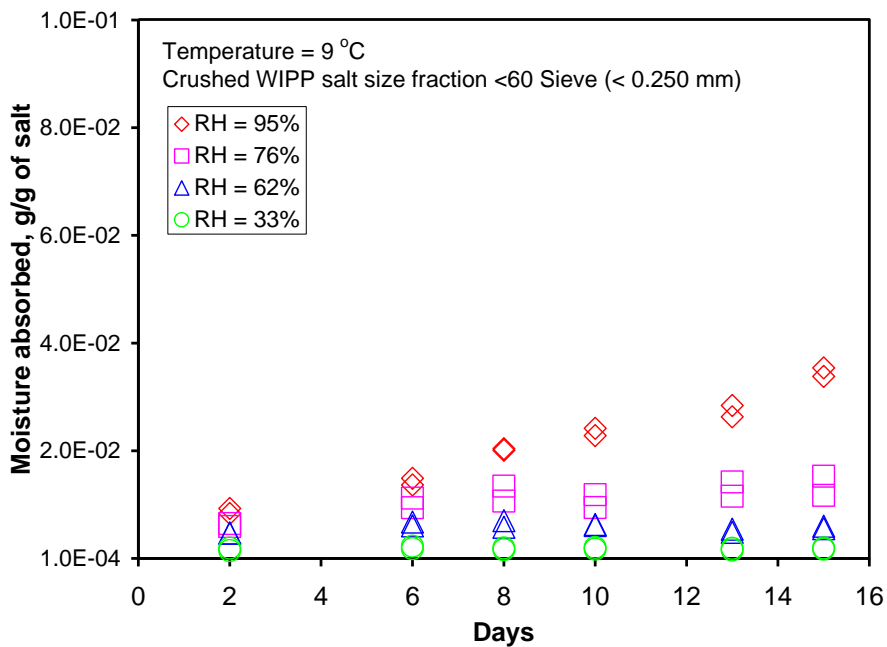


Figure 22A. Sorption of moisture by the WIPP salt with size less than 0.250 mm as a function of time at different relative humidities at 9 °C

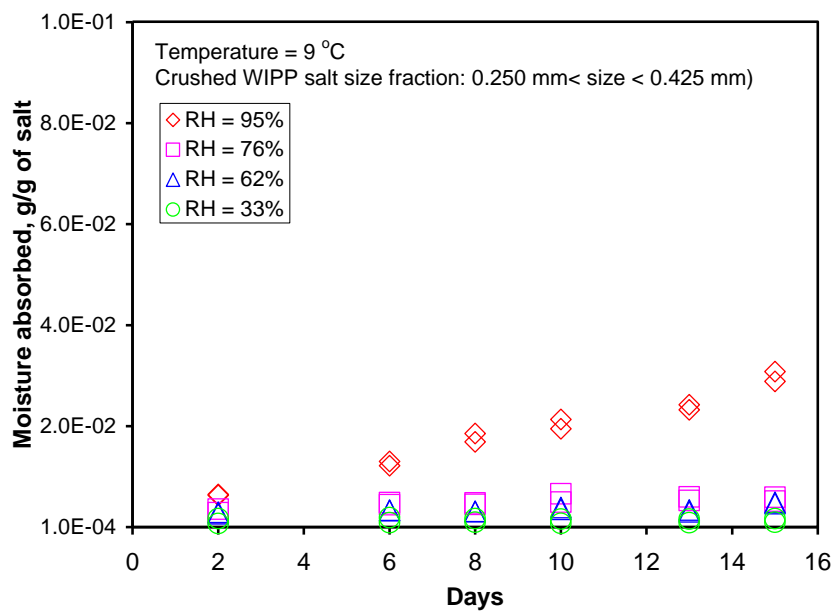


Figure 22B. A plot showing absorption of moisture by the WIPP salt with size between 0.250 mm and 0.425 mm as a function of time at different relative humidities at 9 °C

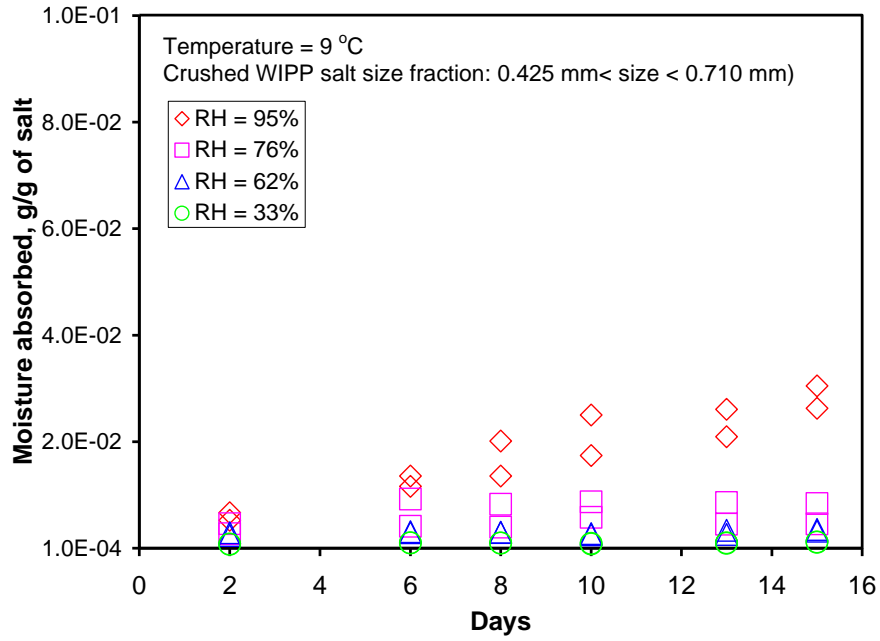


Figure 22C. Sorption of moisture by the WIPP salt with size between 0.425 mm and 0.710 mm as a function of time at different relative humidities at 9 °C

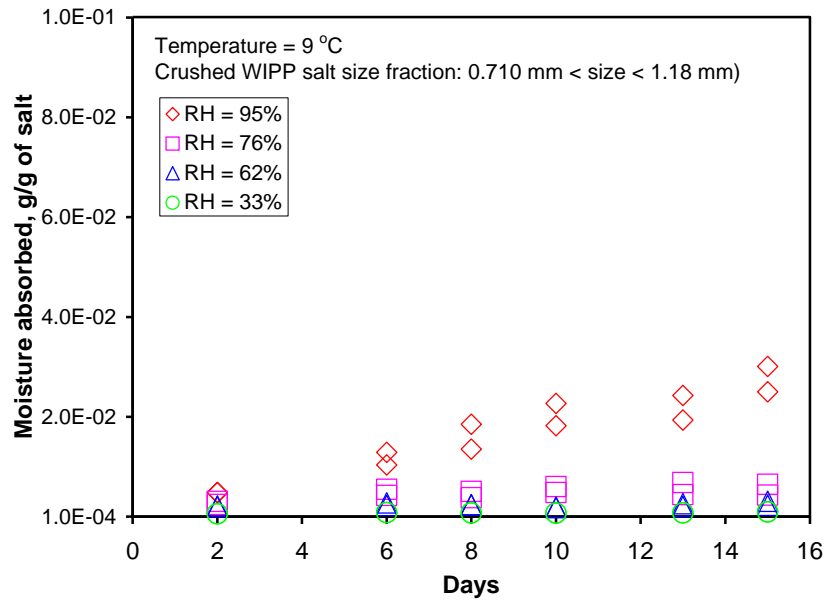


Figure 22D. Sorption of moisture by the WIPP salt with size between 0.710 mm and 1.18 mm as a function of time at different relative humidities at 9 °C

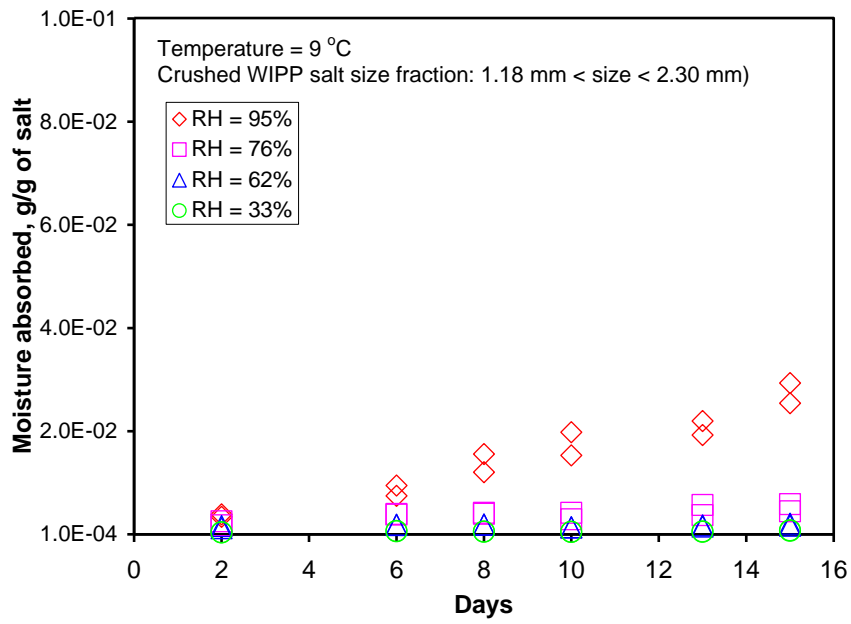


Figure 22E. Sorption of moisture by the WIPP salt with size between 1.18 mm and 2.30 mm as a function of time at different relative humidities at 9 °C

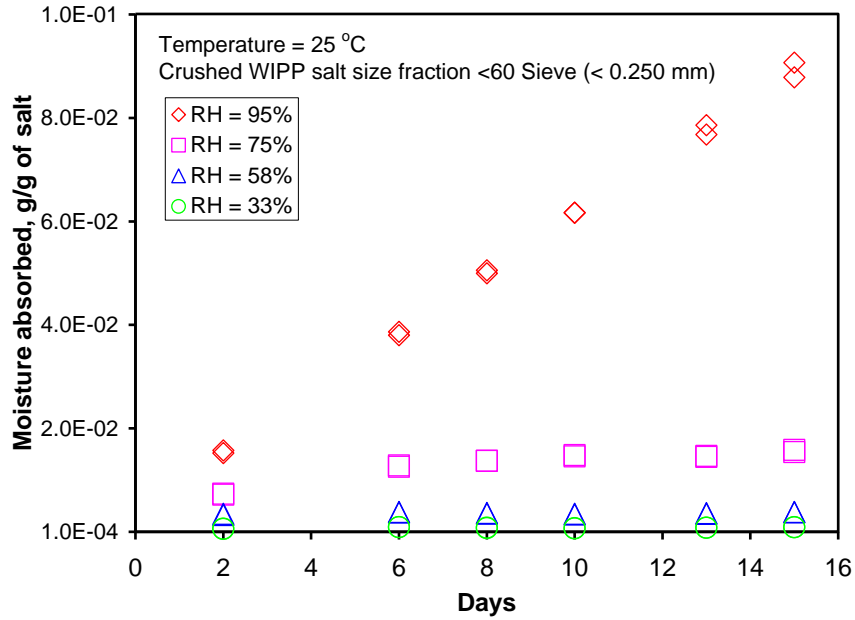


Figure 23A. Sorption of moisture by the WIPP salt with size less than 0.250 mm as a function of time at different relative humidities at 25 °C

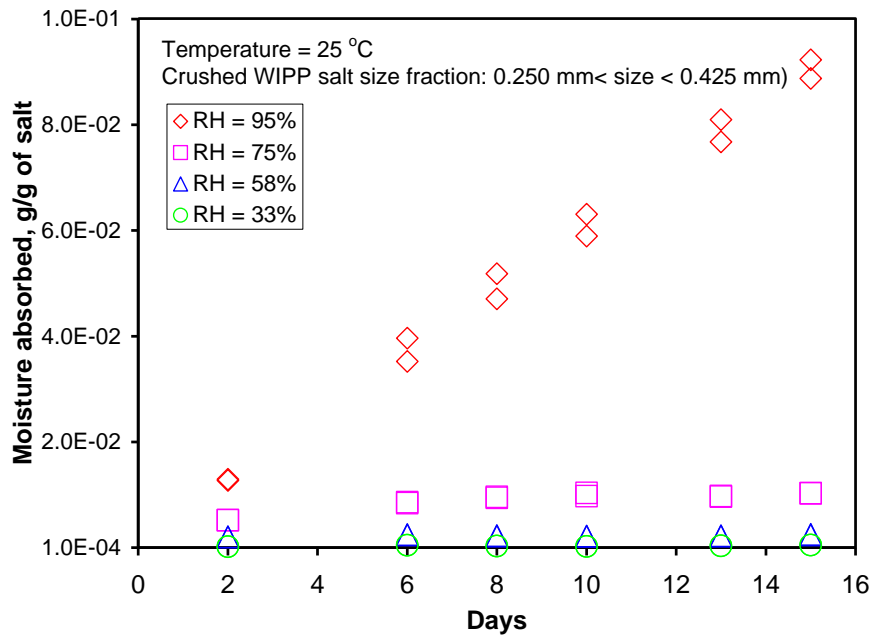


Figure 23B. Sorption of moisture by the WIPP salt with size between 0.250 mm and 0.425 mm as a function of time at different relative humidities at 25 °C

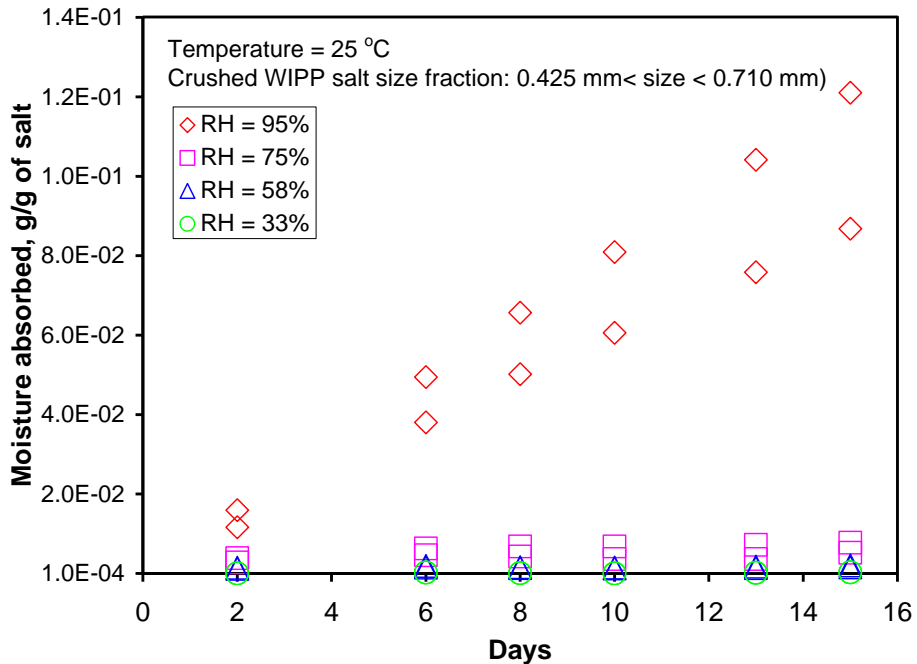


Figure 23C. Sorption of moisture by the WIPP salt with size between 0.425 mm and 0.710 mm as a function of time at different relative humidities at 25 °C

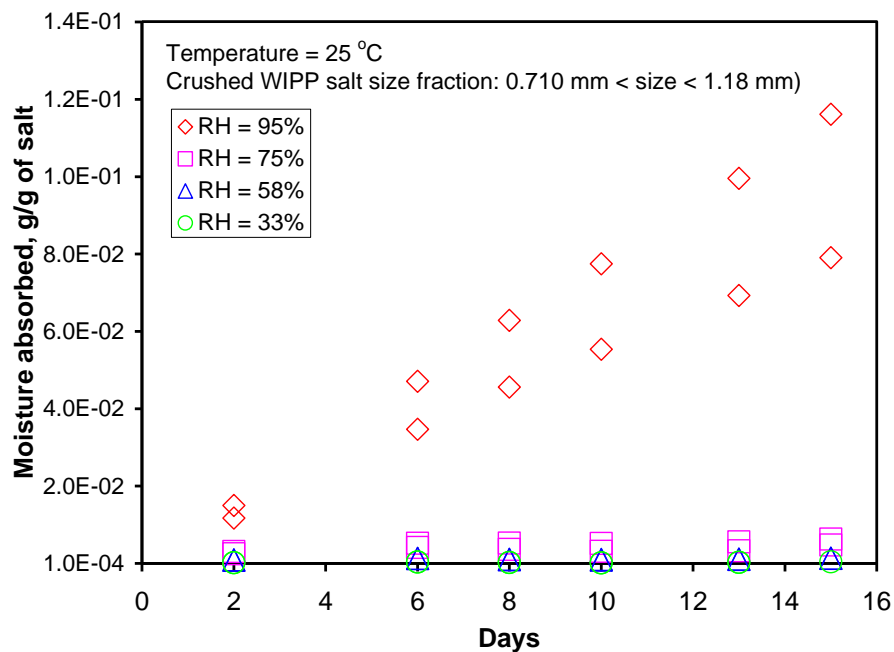


Figure 23D. Sorption of moisture by the WIPP salt with size between 0.710 mm and 1.18 mm as a function of time at different relative humidities at 25 °C

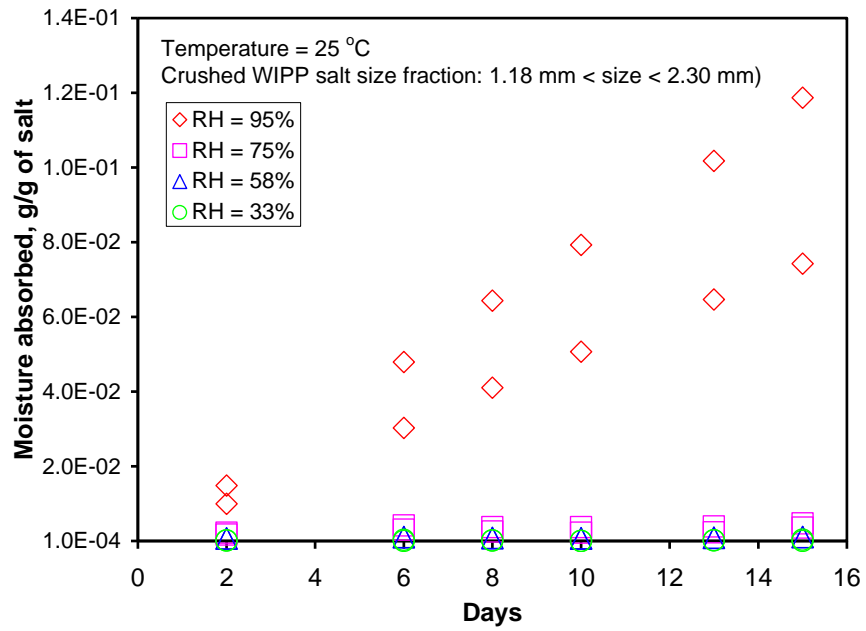


Figure 23E. Sorption of moisture by the WIPP salt with size between 1.18 mm and 2.30 mm as a function of time at different relative humidities at 25 °C

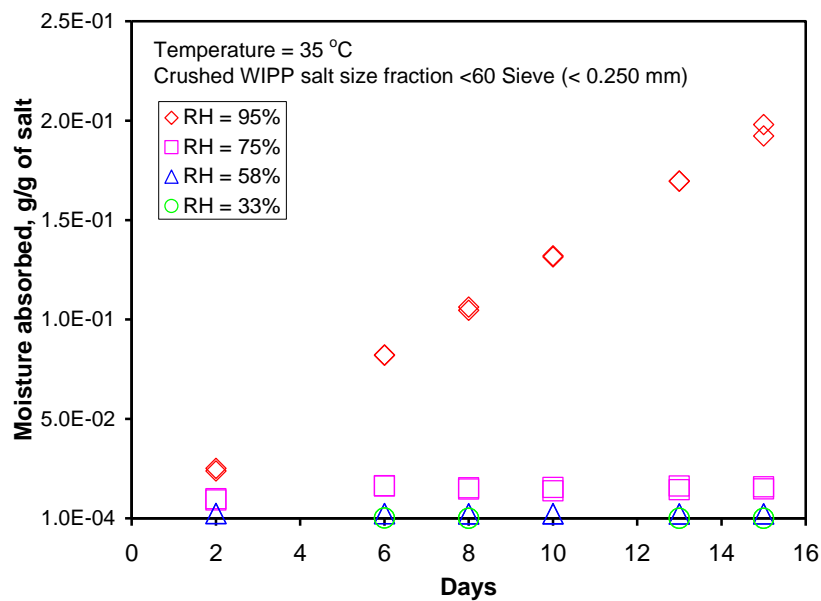


Figure 24A. Sorption of moisture by the WIPP salt with size less than 0.250 mm as a function of time at different relative humidities at 35 °C

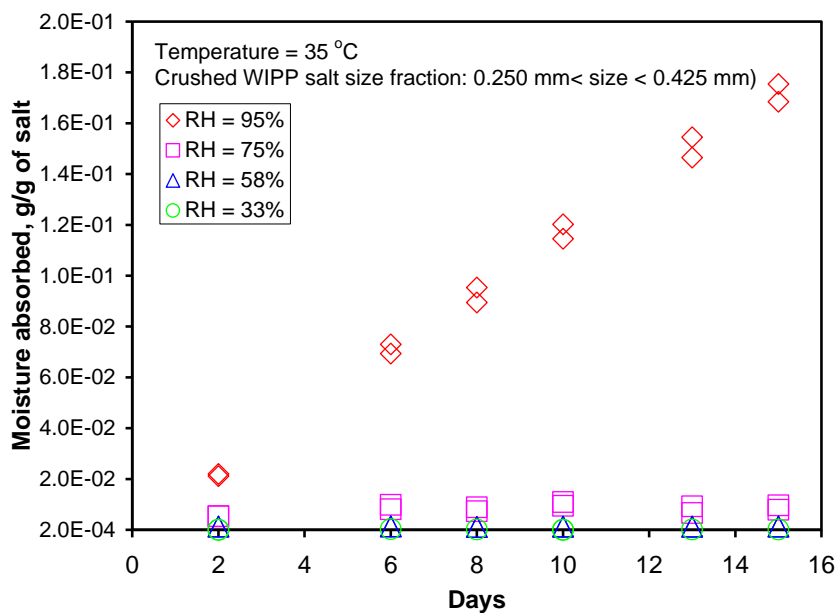


Figure 4B. Sorption of moisture by the WIPP salt with size between 0.250 mm and 0.425 mm as a function of time at different relative humidities at 35 °C

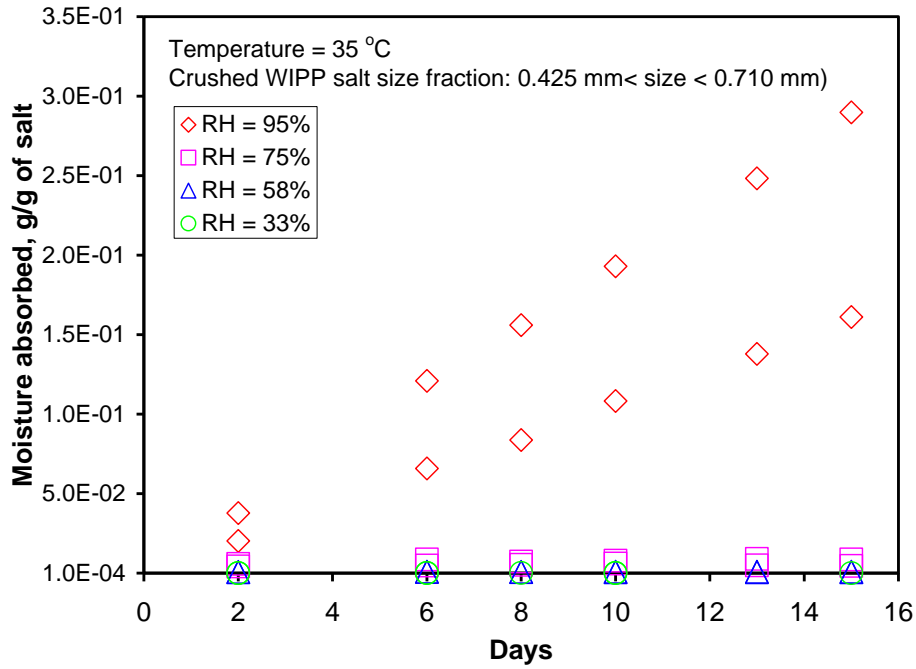


Figure 24C. Sorption of moisture by the WIPP salt with size between 0.425 mm and 0.710 mm as a function of time at different relative humidities at 35 °C

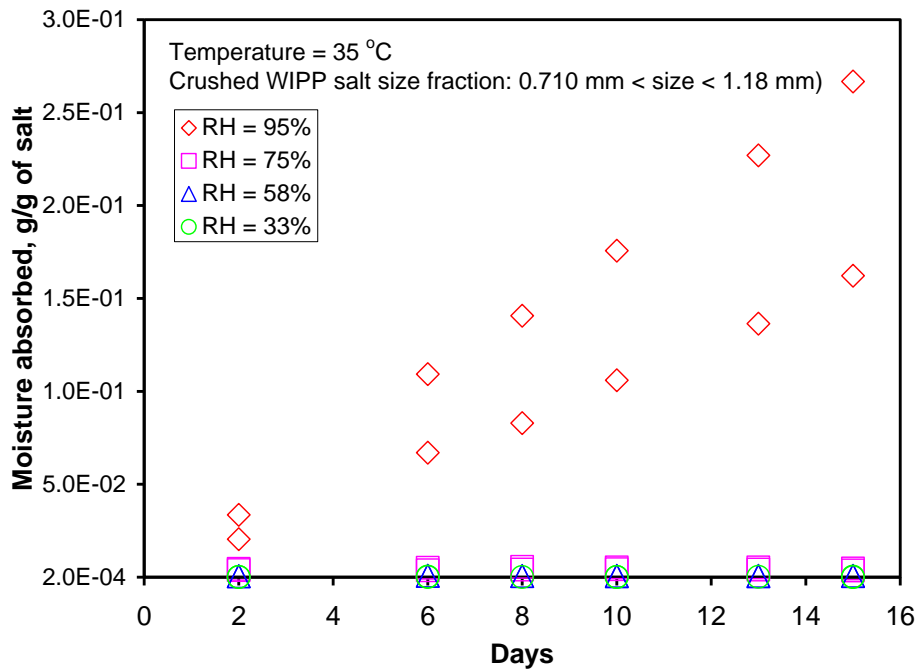


Figure 24D. Sorption of moisture by the WIPP salt with size between 0.710 mm and 1.18 mm as a function of time at different relative humidities at 35 °C

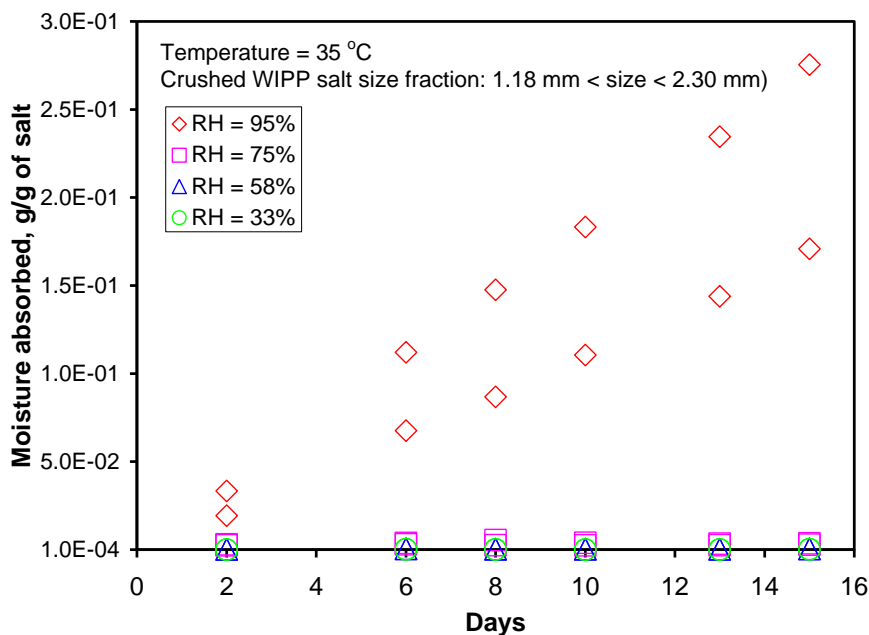


Figure 24E. Sorption of moisture by the WIPP salt with size between 1.18 mm and 2.30 mm as a function of time at different relative humidities at 35 °C

6.0 Summary and Future Work

FY12 work have been focused on laboratory testing and data collection on the leaching behavior of surrogate ER salts and related waste forms in salt repository environments. Specifically, at INL, experimental investigation of factors affecting disposal of electrorefining salt in a generic salt repository was performed. Surrogate electrorefining salt samples, glass and ceramic waste forms were fabricated and assessed for the leachability. At SNL, the evaluation of the effect of radionuclide loading on salt encapsulation was conducted to evaluate and confirm the waste loading threshold behavior of ER salt leaching as predicted by the percolation theory. In addition, a thermal gravity analysis was performed on water vapor sorption or condensation on surrogate ER salts and WIPP salts was performed. The key experimental results are summarized as follows:

- Given a typical repository time scale of 10 thousand to 1 million years, three waste form materials tested - surrogate ER salt, tellurite glass, and sodalite – all would dissolve fast in deionized water or WIPP brines (practically close to instantaneous release relative to a repository time scale), although the latter two may have relatively slow dissolution rates on a laboratory time scale.
- The new experimental data on the leachability of salt composite materials prepared using WIPP salt further confirm the waste-loading threshold behavior of ER salt leaching predicted with the percolation theory. The percolation thresholds are estimated to be about 10% and 20% initial I loading, respectively, for the evaporated and encapsulated salt samples.
- The different percolation thresholds imply that the percolation threshold is dependent on the size and amount of heterogeneity present in the system at hand as well as the sample preparation procedures.

- For a WIPP salt sample, the amount of water adsorbed and/or absorbed approached a plateau within ~ 6 days for all relative humidities except for 95% RH, which is consistent with the equilibrium RH (~75%) of a NaCl-saturated solution. When the environment has a RH higher than 75%, the moisture would continue to condense on the salt to form a NaCl-saturated brine. Otherwise, the moisture adsorbs onto salt surface, and the amount of water absorbed may be limited by the density of sorption sites.
- For ER salt there is no absorption plateau appearing for all relative humidities, because ER salt contains highly hygroscopic Li-Cl salt. In comparison with WIPP salts, the ER salt has a much faster water absorption rate.

It is planned that a comprehensive evaluation of the direct disposal concept will be performed in FY13 using the data collected in FY12. Additional data will also be collected on water uptake and desorption by salts under elevated temperature conditions.

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