Near-Complete Retention of Technetium During Vitrification of Hanford Low Activity Waste With Recycle-17315

Ian L. Pegg[1], Keith S. Matlack[1], Howard Abramowitz[1], and Innocent Joseph[2]
[1]*Vitreous State Laboratory, The Catholic University of America, 620 Michigan Av., NE, Washington, DC 20064*
[2] *Atkins Energy Federal EPC, Inc., Columbia, MD 21046*

ABSTRACT

Technetium is one of the more volatile radionuclides for which retention in low activity waste (LAW) glass under the high temperature conditions during vitrification can vary depending on feed composition, feed chemistry, and melter operating parameters. High retention of technetium in the glass is desirable in order to minimize the fraction of technetium that is directed to secondary waste streams, which include liquid off-gas treatment system effluents. Consequently, test data are crucial to assess and underpin the assumptions underlying such mass balance projections, the compositions of secondary waste streams, and, in particular, the effects of recycle on increasing the incorporation of technetium in the glass products and minimizing the fraction in non-glass waste forms. Previous testing performed on a scaled melter system that incorporates prototypical off-gas treatment components and real-time recycle of the liquid effluents back to the melter feed demonstrated that recycle resulted in significant increases of the incorporation of technetium into the LAW glass product and behavior that was in good agreement with process models for the system. However, the maximum technetium retention achieved in those tests was less than predicted. Tests conducted in the present work demonstrated, for the first time, near-complete retention of technetium in the LAW glass product with recycle. Holdup of technetium in the vacuum evaporator, which concentrates the liquid effluents prior to their recycle to the melter feed, was identified as the primary reason for the previously observed shortfall. The fate of other volatile constituents was also assessed. In particular, in addition to technetium, rhenium, which is frequently used as a non-radioactive surrogate for technetium, was included in these tests. This allows for a direct comparison of the behavior of rhenium and technetium during vitrification and in the down-stream processes.

INTRODUCTION

Technetium is a fission product that is present in used nuclear fuel and wastes generated from nuclear fuel reprocessing. At the Hanford site in Washington State, production of plutonium for nuclear weapons generated approximately 24,000 Ci of ^{99}Tc in about 56 million gallons of high-level waste (HLW) that is currently stored in aging underground tanks. In the Hanford Tank Waste Treatment and Immobilization Plant (WTP), this waste will be separated into LAW and HLW fractions and separately converted to glass by vitrification. The HLW glass is designed for disposal in a national HLW repository while the LAW glass will be disposed on site in the Integrated Disposal Facility (IDF). The long half-life of ^{99}Tc (211,000 yrs), coupled with the high environmental mobility of the very soluble pertechnetate anion, makes ^{99}Tc one of

the most significant risk contributors in performance assessments of the LAW disposal facility.

Technetium is one of the more volatile radionuclides for which retention in LAW glass under the high temperature conditions during vitrification can vary depending on feed composition, feed chemistry, and melter operating parameters. High retention of technetium in the glass is desirable in order to minimize the fraction of technetium that is directed to secondary waste streams, which include liquid off-gas treatment system effluents. These secondary waste streams will be treated and disposed in non-glass waste forms, which will also be disposed in the IDF. Since these waste forms are generally much less leach resistant than the LAW glass, the fraction of technetium in these waste forms can have significant impacts on the overall IDF performance assessment. Consequently, test data are crucial to assess and underpin the assumptions underlying such mass balance projections, the compositions of secondary waste streams, and, in particular, the effects of recycle on increasing the incorporation of technetium in the glass products and minimizing the fraction in non-glass waste forms.

The WTP design includes recycle, wherein the liquid waste streams from the primary off-gas system components are recycled back to the pretreatment facility and, ultimately, to subsequent melter feed batches. In this way, most of the radionuclides and other contaminants of concern that exit the melter in the off-gas stream are captured in the off-gas system and recycled back to the melter feed. Such a recycle loop can greatly increase the overall retention of volatile constituents in the glass product.

Previous testing performed on a scaled melter system that incorporates prototypical off-gas treatment components and real-time recycle of the liquid effluents back to the melter feed demonstrated that recycle resulted in significant increases of the incorporation of technetium into the LAW glass product and behavior that was in good agreement with process models for the system. Such models show that the long-term steady state should approach 100% retention in the glass product, with any shortfall reflecting inefficiencies in the capture in the off-gas treatment system and any holdup in the process (piping, vessels, etc.). However, the maximum technetium retention actually demonstrated in the previous tests was about 84%. For the tests described in this paper, modifications were made in order to more closely approach steady state and to reduce holdup in the process. The new test results demonstrate, for the first time, near-complete retention of technetium in the LAW glass product, as predicted. Holdup of technetium in the vacuum evaporator, which concentrates the liquid effluents prior to their recycle to the melter feed, was identified as the primary reason for the previously observed shortfall. The fate of other volatile constituents was also tracked. In particular, in addition to technetium, rhenium, which is frequently used as a non-radioactive surrogate for technetium, was included in these tests. This allows for a direct comparison of the behavior of rhenium and technetium during vitrification and in the down-stream processes. Data were also collected on other volatile species, including sulfur, chlorine, and fluorine, to assess their potential accumulation in the recycle stream and any consequent effects on salt formation in the melter.

These tests build upon previous tests using the same scaled melter and off-gas system [1-3] by incorporating the WTP-prototypical wet electrostatic precipitator (WESP) deluge and transition line wash [2], enhancing the efficiency of the evaporator and recycle systems to more quickly approach steady state, and using iron (II) oxalate as an additive to increase the single-pass Tc retention in order to increase the rate of approach to steady state Tc concentration in the glass, as identified in extensive previous studies on characterizing and optimizing the single-pass retention of Tc [3-5].

It is important to note that incorporation of technetium into the glass melt is limited by volatility and not solubility. Homogeneous LAW glasses with over 1500 ppm Tc have been made and characterized [6-8] and the solubility has been estimated at around 2500 ppm [9], as compared to the average concentration in the WTP LAW glass which is expected to be about 3 ppm. Studies using X-ray absorption [6, 7, 9] and Raman spectroscopy [8, 10] have shown that technetium is present in these glasses as both Tc(VII) and Tc(IV), with the former dominating under the redox conditions expected for LAW vitrification. Conversely, while Re(VII) is observed, Re(IV) is not [7].

SYSTEM AND TEST DESCRIPTION

Testing was performed on a continuously-fed DM10 joule heated ceramic melter (JHCM) system and off-gas treatment system [1-5]. The DM10 is a ceramic refractory-lined melter fitted with two Inconel 690 plate electrodes that are used for joule-heating of the glass pool and a bubbler for stirring the melt to increase the glass production rate. Thermocouples installed in a thermowell provide temperature measurements at various locations within the glass pool and in the plenum space. The nominal operating temperature is 1150°C. The glass product is removed from the melter periodically by means of a prototypical air-lift discharge system. The DM10 unit has a melt surface area of 0.021 m^2, a glass inventory of about 8 kg, and a glass production rate of about 50 kg per day. The off-gas system maintains the melter under slight negative pressure (typically about 2 inches W.C.) with respect to ambient. The melter was fitted with an off-gas treatment system that included a submerged bed scrubber (SBS), WESP, and high efficiency particulate air (HEPA) filtration, which is representative of the primary components used in the WTP. The liquid effluents from the SBS and WESP were concentrated in a vacuum evaporator in real time and the concentrate was recycled back to the melter feed. A schematic diagram of the system is shown in Fig. 1.
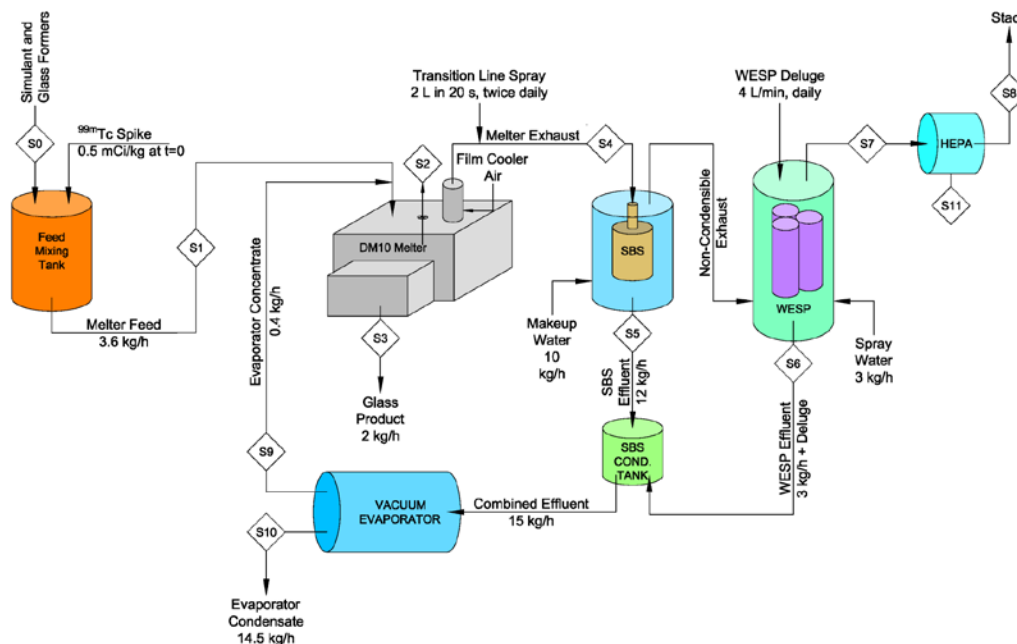
Figure 1. Schematic diagram showing DM10 system components and process flows to implement recycle loop. Also shown are the sampling point (S0 – S11) and the approximate process flow rates.

The SBS is the first unit operation in the DM10 off-gas system. The SBS serves multiple functions including cooling the off-gas via contact with water and condensing moisture in the gas; removing large particulates from the off-gas stream; scrubbing out soluble acid gases; and dissolving soluble particulates into the aqueous phase. The DM10 SBS consists of a vertically oriented, cylindrical, packed-bed column that is submerged in an outer tank that is partially filled with water. The water pool serves to quench the off-gas and collect particulates and soluble species. A gas plenum space above the packed bed serves as a disengagement volume to remove entrained liquid from the exiting gas stream. As condensed water accumulates, it overflows from the SBS into an external collection tank. In the present tests, the SBS sump was spiked with its projected steady state concentration of technetium at the start of each test in order to increase the rate of approach to steady state; the constancy of its concentration over the course of the test confirmed that spiking level was appropriate.

The WESP consists of an array of vertical collection tubes that run the length of the vessel, each with a center electrode. A high electric field between the electrodes ionizes particles in the gas stream, which are then accelerated and collected on the electrodes. The WESP typically operates at about 20,000 volts depending on the gas composition and process conditions. A water spray at the off-gas inlet ensures that the collection surfaces are coated in a wet film during operation that prevents the collected particulates from permanently adhering. The WESP can be flushed

4

(deluged) with fresh water from the top in order to wash any particulate material that has built up on the electrodes into the vessel sump, as is the case for the WTP.

Liquid effluents from the SBS and WESP are routed through a collection tank to a vacuum evaporator, which concentrates the effluents by evaporation under reduced pressure, as is the case at the WTP. The concentrate is then recycled into the melter feed. To facilitate more rapid achievement of steady concentrations in the system, the evaporator is designed to have a very small liquid inventory. The vapor flows through a pumping system, which maintains the vacuum on the system and condenses the vapor. The condensate accumulates in a reservoir where it can be sampled.

The tests employed three simulated LAW streams representing pretreated supernate from Hanford tanks AN-105, AN-104, and AZ-102, and associated glass formulations (LAWE4H, LAWE6H, and LAWE10H, respectively) that were developed and tested previously [11, 12]. These streams are essentially high-sodium salt solutions containing many components but predominantly nitrate, nitrite, hydroxide, aluminum, phosphate, potassium, sulfate, and chloride as well as various organics. The melter feed material was an aqueous slurry of the simulated waste mixed with glass forming chemicals (which provide sources of Si, B, Al, Fe Ca, Mg, Ti, Zn, Zr, Li), which is pumped continuously onto the surface of the molten glass pool in the melter.

The short-lived isotope of technetium, ^{99m}Tc (half-life = 6 hrs) in the pertechnetate form, was used in place of ^{99}Tc (half-life = 210,000 yrs). ^{99m}Tc has easily detectable gamma emissions around 140 keV permitting analysis by gamma counting, which is fast and accurate. The short half-life means that the test systems and associated wastes are essentially decontaminated simply by allowing time for decay. However, the samples have to be collected and analyzed quickly, which, for a complex system that generates many samples, presents logistical challenges that have to be overcome. All measured activities were corrected to a common time.

The melter feeds were spiked with ^{99m}Tc at typically 1 mCi per kg of glass if all were retained; similarly, Re (in the perrhenate form) was added at a concentration of 500 ppm and iodine (as iodide) was added at a concentration of 1000 ppm. Previous tests with Tc and Re together, and with Tc alone have shown no interaction between these components with respect to retention [4].

A variety of sampling points are included in the DM10 system to support a mass balance for all constituents of interest. The sampling locations (S0 – S11) are shown in Fig. 1 and include melter feed, glass, off-gas, and all of the process fluids throughout each test. At the end of each test, all of the sumps were sampled, sump and line flushes and rinses were performed, and samples of all of these fluids were collected for analysis. In some cases two or three rinses were performed; the technetium activity typically dropped by about a factor of ten after each rinse. In addition, the film cooler and transition line were removed and the components were rinsed with water and the rinsates were sampled and analyzed. The results from analysis of this suite of samples were used to support a mass balance over each test.

RESULTS AND DISCUSSION

Over the course of these three tests it became evident that a significant fraction of the technetium was being held up in the plenum space of the evaporator where the liquid influent is introduced into the heated headspace. Evidently, solids were being dried and deposited in that region and therefore any material in those deposits was not available for recycle. This is the case for Test 1, which shows a relatively low technetium mass balance closure over the entire test as well as a relatively low technetium retention in the product glass at steady state, as shown in Table I. Once this was identified, at the end of Test 2, a modified procedure was employed that more effectively washed that portion of the evaporator plenum space at the end of the test. As can be seen in Table I, this increased the technetium mass balance closure significantly. With these findings in mind, Test 3 employed a further modified procedure which allowed for periodic washing of the evaporator plenum space during the test. Table I shows that this increased both the technetium mass balance closure and the concentration of technetium in the product glass significantly, with essentially complete retention of technetium at steady state. These results are consistent with the projections from process modeling when holdup in the system is insignificant [1] and demonstrate that recycle can indeed achieve essentially complete incorporation into the product glass.

TABLE I. Technetium Mass Balance and Steady State Concentrations in Product Glasses.

Test	Evaporator Plenum Wash?		Tc Mass Balance	Tc in Glass at Steady State
	During Test	At End		
Test 1, LAWE10H	No	No	84%	70%
Test 2, LAWE6H + ferrous oxalate	No	Yes	97%	75%
Test 3, LAWE4H + ferrous oxalate	Yes	Yes	106%	100%

Fig. 2 shows the evolution of the technetium concentration in the product glass over the course of Test 3. Also shown in Fig. 2 are the results from a previous test with the same feed and glass composition (LAWE4H) but without ferrous oxalate and using a system configuration that took longer to reach steady state [1].
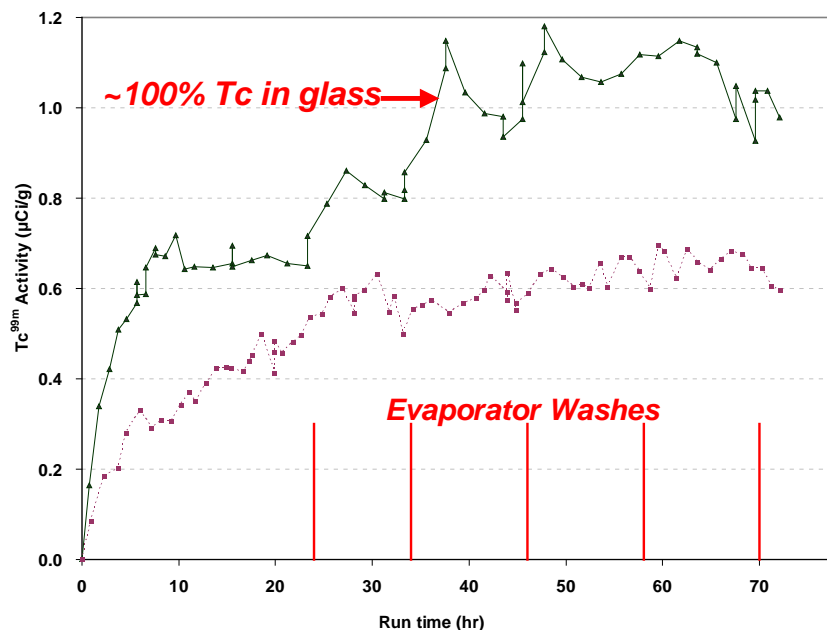
Figure 2. Technetium concentrations in product glasses from Test 3 with LAWE4H. Data from the present work (black triangles) lie above those from a previous test with LAWE4H (red squares) [1].

The data from the present test in Fig. 2 show a much faster rise as a result of the shorter system time constant and an initial steady state at about 63% retention before the implementation of the periodic washes of the evaporator plenum space at about 23 hours run time. Subsequently, a sharp rise is seen in the technetium concentration in the product glass after each wash of the evaporator plenum space, clearly demonstrating holdup of technetium-rich material in that region. On each wash cycle, technetium in those solids is mobilized and introduced into the evaporator concentrate, which is then recycled to the melter feed, thereby increasing the concentration in the product glass. Once holdup in this location is mitigated, essentially complete retention of technetium in the product glass is observed. Similar behavior was observed for other constituents, including rhenium.

Mass balance closure and retention of volatile constituents in the glass product for Test 3 (LAWE4H) was the highest of the three tests as a result of the periodic washes of the evaporator plenum space. The results for Test 3 are summarized in Table II. It should be noted that the results in Table II are cumulative of the entire test whereas those in Fig. 2 are instantaneous values. Thus, while the steady state technetium retention from Fig. 2 is about 100%, the cumulative amount in the entire product glass inventory was about 95% (Table II) as a result of the lower values early in the test; this value would be expected to rise and converge to the steady state value as the test duration is increased.

TABLE II. Mass Balance and Distribution of Key Constituents at End of Test 3 with LAWE4H.

%	Tc	Re	I	Cl	S
Glass	94.95	75.10	59.30	120.80	108.40
Transition Line/ Film Cooler	1.93	2.24	0.09	2.02	0.33
SBS Sump	(3.44)[a]	3.22	2.59	4.31	0.77
WESP Tube Walls	0.35	0.32	0.01	0.56	0.14
Containment Tank	1.08	0.99	0.43	1.16	0.08
Evaporator Concentrate	7.52	9.60	1.16	10.93	3.30
Evaporator Condensate	<0.001	0.05	18.80	4.39	0.51
Recycle Line	0.17	0.16	0.03	1.83	0.02
Test Samples	0.18	-	-	-	-
WESP Emissions	0.20	0.14	9.22	0.30	1.38
Makeup Water[b]	-	-	-	-22.48	-7.35
Total	106.4	91.8	91.6	123.8	107.6

[a] Not included in total since this was spiked in at the beginning of the test.
[b] Correction due to impurities in process water.

The total amount of technetium recovered was about 106%, which in view of the number of samples and analyses that make up this number corresponds to essential complete mass balance closure. About 95% of the technetium reported to the product glass inventory (averaged over the entire test) and the remainder was mostly contained in the evaporator (7.5%), with lesser amounts distributed among the transition line, film cooler, containment tank, and WESP internals. The total amount of rhenium recovered was about 92% with 75% reporting to the glass, 9.6% accumulated in the evaporator, and 1 to 3.2% associated with the film cooler/transition line, SBS, WESP internals, and containment tank. The total amount of iodine recovered was 92% with 59% reporting to the glass, about 19% in the evaporator condensate, and 9% in the WESP emissions. Chlorine and sulfur mass balance closure was over 100%, with over 100% of the target feed chlorine and sulfur reporting to the glass. Mass balance for these constituents was complicated by their presence in the process water. Upper transition line and film cooler deposits, which cannot readily be recycled back to the feed, constituted about 2% of the technetium, rhenium, and chlorine fed to the melter. Constituents emitted from the WESP, which are not recycled back to the melter feed, constituted 0.2% and 0.14% of the technetium and rhenium fed to the melter, respectively; 9.2% of the iodine fed to the melter was measured in the WESP off-gas emissions. Another 18.8% of the iodine fed to the melter was detected in the evaporator condensate with only 0.05% of the rhenium and no detectable technetium; the evaporator condensate is also not recycled back to the melter feed.

The effect of deluging the WESP was to reduce the amount of each constituent on the internals of the WESP at the end of the test by over an order of magnitude for technetium (4.6 to 0.35%) and rhenium (6 to 0.32%) as compared to previous tests without the deluge [1]. Another consequence of deluging the WESP is that during the deluge the WESP is not powered and therefore is not removing particulate from the exhaust stream. As a result, the amount of material emitted from the WESP is higher in the present tests which include deluges, as observed for technetium (0.2% vs. 0.045%), than in previous tests without the deluge [1]. The effect of the use of ferrous oxalate in the present tests is most discernable for the increase in iodine retention in the glass (59.3 vs 41.5%). Given the amount of iodine exiting the recycle loop to the evaporator condensate and WESP exhaust, the increase in iodine concentration in the glass product is attributable primarily to the enhanced single pass-retention resulting from the use of ferrous oxalate [4, 5].

The retentions of technetium and rhenium in the discharge glasses are compared in Fig. 3 for the three tests. For each test, the results for glass samples from early in the test fall towards the lower left (lower retention), while those from later in the test, corresponding to closer to steady state, fall towards the upper right (higher retention).
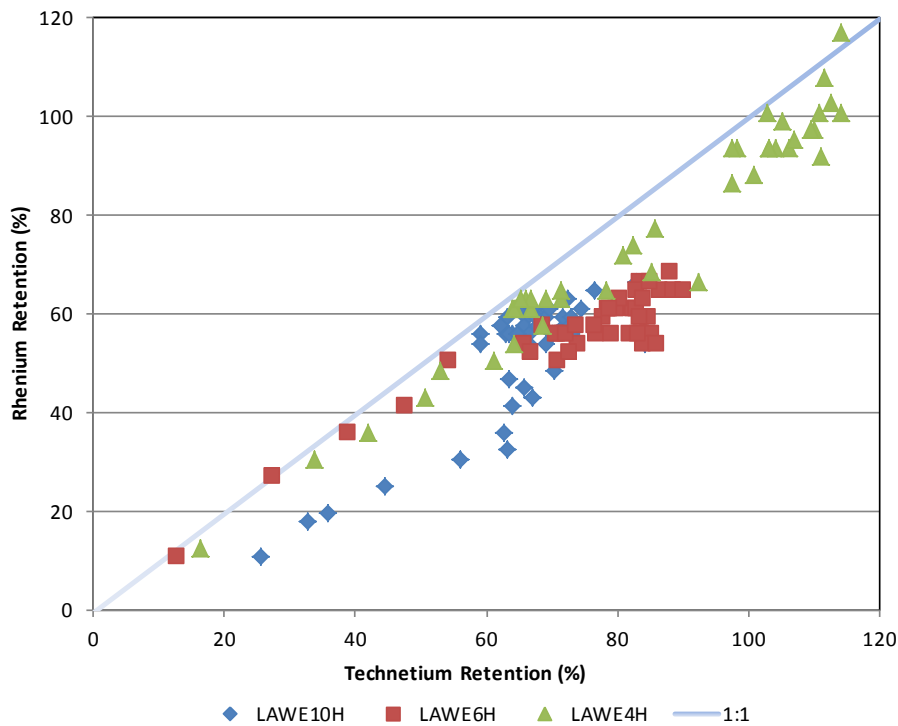
Figure 3. Relationship between technetium and rhenium retention in glasses for recycle tests.
Line illustrates 1:1 relationship.

In all but one glass sample in Fig. 3, the technetium retention was equal to or higher than the rhenium retention. On average, rhenium retention was about ten to twenty absolute percent lower than the technetium retention. Notable exceptions, when the ratio of technetium to rhenium was closer to 1:1, were the early portions of tests with LAWE6H and LAWE4H as well as the latter portion of the test processing LAWE4H during periods with evaporator rinsing. In the previous tests with recycle the opposite trend was observed, with rhenium retention equal to or higher than the technetium retention for all compositions other than LAWE10H [1], in agreement with the results from the present test with LAWE10H. The difference in behavior for LAWE6H and LAWE4H between the present and previous tests [1] is due to the use of ferrous oxalate in those tests for the present work, since ferrous oxalate enhances technetium retention more so than rhenium retention [5]. The same relationship between technetium and rhenium retentions, with technetium being higher, was observed in previous single-pass tests with ferrous oxalate for the majority of the compositions tested [5], in agreement with results for the LAWE4H and LAWE6H compositions with ferrous oxalate in the present work.

The results in Fig. 3, when considered together with those in Table II, which show remarkably similar distributions of technetium and rhenium across the glass and the off-gas and evaporator system effluents, suggest that rhenium performs reasonably well as a surrogate for technetium. However, for retention in the glass in particular, the detailed relationship can be complicated and depends on both the waste and additive composition, particularly when redox effects are significant [4, 5, 7].

CONCLUSIONS

Key findings from this work include:

- Steady state concentrations in the glass product and off-gas sump solutions of technetium, rhenium, and most constituents of concern were achieved in each test. The technetium concentration reached steady state within the first day of processing with the modified test system. Once periodic rinsing of the evaporator head space was instituted during the last test, considerable increases in the steady state concentrations of these components in the product glass were seen. Without such rinsing, technetium and other species accumulate in solids deposits making them unavailable for recycle, which decreases their steady state concentrations in the product glass. The combination of the increased rate of approach to steady state and the mitigation of these accumulations led to near 100% retention of technetium in the product glass. These observations explain the lower technetium concentrations measured in the product glasses in previous tests conducted with recycle [1].

- Complete retention of technetium and rhenium in the glass product was demonstrated once the periodic rinsing of the evaporator head space was implemented with significant increases over the respective single-pass values.

- The holdup of material in the system, particularly in the WESP internals and the transition line, was mitigated in the present tests with periodic transition line rinses and the daily deluge of the WESP. Further mobilization of material in the evaporator headspace was achieved during the last test with the implementation of periodic evaporator rinses.

- Overall, technetium and rhenium showed remarkably similar distribution across the various system sumps, glass, and exhaust samples.

- Iodine retention in the glass for the present tests with recycle showed enhancements over the corresponding single-pass tests. A larger increase in iodine retention was observed with the use of ferrous oxalate as an additive. The impact of recycle on increasing iodine retention was lower than that for technetium and rhenium as a result of the much higher iodine emissions from the WESP and loss of iodine to the condensate during evaporation.

- Sulfur and chlorine showed near complete retention in the glass product and excess mass balance closure due in part to excesses in the melter feed and their presence as contaminants in the process water used for makeup water and sprays in the off-gas system.

In the WTP system, technetium can exit the recycle loop via two routes: the off-gas stream exiting the WESP and the liquid condensate stream from the vacuum evaporator. Constituents in the off-gas stream from the WESP are further removed in the down-stream packed bed scrubber (PBS). The PBS effluent and evaporator condensate secondary waste from the WTP are directed ultimately to non-glass waste forms. Consequently, the fraction of technetium and other constituents of concern exiting the recycle loop via these routes is a significant factor in the Performance Assessment (PA) for the IDF. There have been few previous measurements of this fraction [1]. Data from the present tests permit such estimates to be made including:

- The fraction of feed technetium exiting the recycle loop through the evaporator overheads and WESP exhaust ranged from <0.0001 to 0.0004% and 0.2 to 0.29%, respectively.

- The fraction of feed rhenium exiting the recycle loop through the evaporator overheads and WESP exhaust ranged from 0.0004 to 0.052% and 0.14 to 0.34%, respectively.

- The fraction of feed iodine exiting the recycle loop through the evaporator overheads and WESP exhaust ranged from 2 to 31% and up to 9.22%, respectively.

REFERENCES

[1]   K.S. MATLACK, H. ABRAMOWITZ, M. BRANDYS, I.S. MULLER, R.A. CALLOW, N. D'ANGELO, R. CECIL, I. JOSEPH and I.L. PEGG, *Technetium Retention in WTP LAW Glass with Recycle Flow-Sheet: DM10 Melter Testing.* VSL-12R2640-1, Rev. 0, Vitreous State Laboratory, The Catholic University of America, Washington, DC, 9/24/12.

[2]   K.S. MATLACK, H. ABRAMOWITZ, I.S. MULLER, K. GILBO, M. PENAFIEL, and I.L. PEGG, *Wet Electrostatic Precipitator Performance and Technetium-Rhenium Behavior in LAW Recycle Flow-Sheet.* VSL-13R2800-1, Rev. 0, The Catholic University of America, Vitreous State Laboratory, Washington, DC, 3/1/13.

[3]   I.L. PEGG, J. Radioanal. Nucl. Chem., 305, 287 (2015).

[4]   K.S. MATLACK, I.S. MULLER, I. JOSEPH, AND I.L. PEGG, *Improving Technetium Retention in Hanford LAW Glass – Phase 1*, VSL-10R1920-1, Rev. 0, Vitreous State Laboratory, The Catholic University of America, Washington, DC, 3/19/10.

[5]   K.S. MATLACK, I.S. MULLER, R. CALLOW, N. D'ANGELO, T. BARDAKCI, I. JOSEPH, and I.L. PEGG, *Improving Technetium Retention in Hanford LAW Glass – Phase 2.* VSL-11R2260-1, Rev. 0, Vitreous State Laboratory, The Catholic University of America, Washington, DC, 7/20/11.

[6]   D.A. MCKEOWN, A.C. BUECHELE, W.W. LUKENS, D.K. SHUH, and I.L. PEGG (2007) Environ. Sci. Technol. 41:431.

[7]   W.W. LUKENS, D.A. MCKEOWN, A.C. BUECHELE, I.S. MULLER, D.K. SHUH, and I.L. PEGG (2007) Chem. Mater. 19:559.

[8]   D.A. MCKEOWN, A.C. BUECHELE, W.W. LUKENS, I.S. MULLER, D.K. SHUH, and I.L. PEGG (2007) Radiochimica Acta 95:275.

[9]   C.Z. SODERQUIST, M.J. SCHWEIGER, D.-S. KIM, W.W. LUKENS, and J.S. MCCLOY (2014) J. Nucl. Materials, 449:173.

[10]   P.L. GASSMAN, J.S. MCCLOY, C.Z. SODERQUIST, and M.J. SCHWEIGER (2014) J. Raman Spectrosc. 45:139.

[11]   K.S. MATLACK, W. GONG, I.S. MULLER, and I.L. PEGG, *DuraMelter 100 Tests to Support LAW Glass Formulation Correlation Development*, VSL-06R6480-1, Rev. 0, Vitreous State Laboratory, The Catholic University of America, Washington, DC, 3/21/06.

[12]   K.S. MATLACK, I.S. MULLER, W. GONG, and I.L. PEGG, *Small Scale Melter Testing of LAW Salt Phase Separation*, VSL-07R7480-1, Rev. 0, Vitreous State Laboratory, The Catholic University of America, Washington, DC, 08/20/07.

ACKNOWLEDGEMENTS