Recent Advances in Used Nuclear Fuel Reprocessing Off-Gas Treatment

Robert T. Jubin

Oak Ridge National Laboratory, 1 Bethel Valley Road, P.O. Box 2008, MS 6243,
Oak Ridge, Tennessee 37831 USA
Tel. (865) 594-4934, Fax. (865) 574-6872, Email: jubinrt@ornl.gov

ABSTRACT

The Off-Gas Sigma Team, formed in fiscal year 2010, brought together multidisciplinary teams from across the Department of Energy complex and academia to work collaboratively to solve the technical challenges of and to develop the scientific basis for the capture and immobilization of volatile radionuclides contained in the airborne effluents from used nuclear fuel reprocessing. The volatile radionuclides of concern are ^{3}H, ^{14}C, ^{85}Kr, and ^{129}I. This work has been performed with the ultimate objective of ensuring regulatory compliance for gaseous discharges by potential used nuclear fuel reprocessing facilities. Participating laboratories include Idaho National Laboratory, Oak Ridge National Laboratory, Pacific Northwest National Laboratory, and a number of university laboratories affiliated with the Department of Energy Nuclear Energy University Partnership Program.

The long-term scope of the Off-Gas Sigma Team includes the development, demonstration, and technical maturation of integrated off-gas treatment and immobilization systems for prototypical dissolver off-gas, vessel off-gas, tritium pretreatment off-gas, cell off-gas, and waste treatment off-gas arising from aqueous and electrochemical fuel treatment processes. The five major thrust areas include (i) iodine capture, (ii) iodine immobilization, (iii) tritium separations, (iv) krypton separations and storage, and (v) integrated off-gas treatment system development.

INTRODUCTION

The Off-Gas Sigma Team was formed in fiscal year 2010 (FY 2010) to bring together multidisciplinary teams from across the DOE complex and academia that would work collaboratively to solve the technical challenges of and develop the scientific basis for the capture and immobilization of volatile radionuclides contained in the airborne effluents from used fuel treatment processes. This work has been performed with an ultimate goal of clarifying the practical impacts of the regulations and development of technology that can facilitate regulatory compliance for gaseous discharges by potential used fuel treatment facilities. The team is focused on development of effective methods to capture and immobilize ^{129}I, ^{14}C, ^{85}Kr, and ^{3}H released from fuel treatment processes, thereby enabling a range of advanced fuel cycle options in the US. Participating laboratories have included Argonne National Laboratory (ANL), Idaho National Laboratory (INL), Oak Ridge National Laboratory (ORNL), Pacific Northwest National Laboratory (PNNL), and Sandia National Laboratories (SNL).

OVERVIEW OF OFF-GAS SIGMA ACTIVITIES

In very broad terms, the objectives of the Off-Gas Sigma Team include

1. developing materials and processes for capture of I, ^{3}H, Kr, and C that can be implemented across likely used fuel treatment processes (including both aqueous and electrochemical fuel treatment options);

2. developing a fundamental understanding of the generation, capture, and immobilization of radionuclides from used fuel treatment process off-gas to gain confidence in their performance and allow for new solutions to be developed as fuel treatment processes are better defined;

3. developing waste forms and processes to manage off-gas waste streams in an effective manner; and

4. designing and demonstrating the effectiveness of integrated off-gas treatment systems for likely fuel treatment processes.

To support the four primary objectives, research and development (R&D) is being conducted by the team in five primary areas:

1. Iodine capture—The objective of the iodine capture task is to develop materials and processes that ensure compliance with regulatory releases of radioiodine from a used fuel treatment facility. The off-gas streams within an aqueous reprocessing facility that have been identified as sources of volatile radioiodine include the dissolver off-gas, tritium pretreatment off-gas, vessel ventilation off-gas, melter off-gas, and cell off-gas. The materials development and testing should be performed on materials and systems that are cost-effective to deploy at an industrial scale, and testing should aim for both a fundamental understanding of the materials and processes and generation of data needed for future engineering studies.

2. Iodine immobilization—The objective for the iodine immobilization task is to develop suitable waste forms and processes capable of immobilizing radioiodine with characteristics that limit the release of radioiodine to levels consistent with a potential repository waste acceptance criteria. The process should be adaptable to specific capture methods and materials and be cost-effective.

3. Tritium separations from head-end off-gas generated by a tritium pretreatment process—The objective of the tritium separations task is to develop a viable process to separate tritium from the tritium pretreatment off-gas stream and from radioiodine.

4. Krypton separations and storage—The objective of the krypton separations and storage task is to develop a cost-effective method to separate and manage radiokrypton from off-gases in the head-end processes of a used fuel treatment plant so that the radiokrypton releases are below regulatory limits. The krypton capture method should separate krypton from the off-gas carrier stream and from xenon. Both aqueous and electrochemical plant off-gases are of interest, but the initial focus is on aqueous process off-gases. The effects of long-term storage of ^{85}Kr loaded waste forms must also be understood.

5. Integrated off-gas treatment system development—The objectives of the integrated off-gas treatment system task are to develop and demonstrate integrated off-gas treatment systems for tritium pretreatment off-gas, dissolver off-gas, vessel off-gas, and melter off-gas. The task

includes the evaluation of materials and processes for capture and immobilization of each gaseous component and modeling of the entire gas system including heat and mass balances, equipment scaling and layout, and study of any interface issues. Development of fundamental understanding of adsorption, mass flow, and reaction modeling is also a sub-tier objective of this task.

HIGHLIGHTED R&D EFFORTS

Over the past four years the Sigma Team has conducted over 20 distinct R&D activities within the US Department of Energy national laboratory complex to advance the state-of-the-art in off-gas treatment. This has been supported by six closely linked Nuclear Energy University Program projects involving 10 universities. These activities focus on one or more of the Sigma Team's overarching objectives. Although there are numerous accomplishments that merit discussion, the balance of this paper will highlight four of the ongoing or recently completed R&D activities.

Engineering Evaluation of an Integrated Off-Gas Treatment System for Used Nuclear Fuel Reprocessing Facilities *[R. T. Jubin, S. H. Bruffey, J. A. Jordan, B. B. Spencer (ORNL), N. R. Soelberg, A. K. Welty, M. Greenhalgh (INL), D. M. Strachan (Strata-G LLC), and P. K. Thallapally (PNNL)]*

The capture and subsequent immobilization of the four regulated volatile radionuclides (^{3}H, ^{14}C, ^{85}Kr, and ^{129}I) and relevant semivolatile species from the off-gas streams of a used nuclear fuel (UNF) reprocessing facility has been a topic of significant research interest for the US Department of Energy and other international organizations. To that end, significant research and development (R&D) has been conducted over the past decade. Using the results from this R&D as well as the body of information available from earlier research an engineering evaluation of an integrated off-gas system was conducted in FY2016. This evaluation resulted in a report entitled Engineering Evaluation of an Integrated Off-Gas Treatment System for Used Nuclear Fuel Reprocessing Facilities [1], referred to as the "Engineering Evaluation."

This study was an initial engineering evaluation and design of the off-gas abatement systems required to ensure that air emissions from a reprocessing facility can meet or exceed emission requirements set forth by the US Environmental Protection Agency and US Nuclear Regulatory Commission. It built on a series of case studies that addressed various reprocessing options for UNF. This study focused on the capture and retention of the volatile radionuclides (^{3}H, ^{14}C, ^{85}Kr, and ^{129}I); selected semi-volatile radionuclides, specifically ^{106}Ru; and chemical species, such as NO and NO$_2$, generated during the dissolution of the UNF. The study examined the design of the combined head-end off-gas streams and the vessel off-gas stream.

The Engineering Evaluation covered only engineering options for the off-gas streams arising from head-end, fuel dissolution, and process vessels for aqueous reprocessing. It was not intended to be an exhaustive, detailed design effort but, rather, a first attempt to evaluate (1) the relative sizes, (2) the material balances, (3) energy requirements, (4) key instrumentation requirements, (5) support system requirements, (6) extended operating cycles and associated requirements, and (7) the extent to which key design data are or are not available for the off-gas systems. This engineering evaluation addressed the designs of the head-end off-gas, which

typically includes the shear off-gas, dissolver off-gas, and vessel off-gas systems. Subsequent reports will add similar assessments for the cell off-gas and melter off-gas systems.

A summary of the engineering evaluation results are (in no particular order):

1. Common processes used in industry and existing nuclear facilities are perceived as relatively easy to implement in a UNF reprocessing facility. These unit operations involve condensers, heat exchangers, scrubbers, valves, and particulate filters. These systems are well understood because of their widespread use in the nuclear industry; however, uncertainties remain, including the actual performance of the NO_x scrubber and the CO_2 scrubber under operating conditions.

2. There is a surprising lack of clarity in the understanding of the speciation of volatile ruthenium under various facility conditions. The amount of ruthenium volatilized during fuel processing, either directly from the dissolved fuel or during the treatment to remove tritium separately, does not appear to be well understood. Once ruthenium is volatilized, its chemical form under the various conditions that exist in the facility and the behavior of the species in the off-gas stream are sufficiently ambiguous and could result in large pathway and decontamination factor uncertainties. Silica gel has been proposed as the best ruthenium sorbent. However, the capacity and efficiency of this material under facility conditions do not appear to be well understood or at least not well articulated in the literature. This is important because of the very high ruthenium removal efficiency (3×10^7) that may be required to meet regulated releases even for UNF that has been cooled for 5 years.

3. There is a lack of pertinent engineering data, such as the length of the mass transfer zone, sorption kinetics, and desorption kinetics, about solid sorbents for the removal of xenon and krypton. This uncertainty means that the systems designed by this project require multiple large columns to achieve the desired decontamination of the off-gas stream and purity of the krypton gas for storage. These data are needed to ensure the continued viability of these solid sorbent materials for xenon and krypton as opposed to the cryogenic process for the removal of these gases).

4. Although the xenon and krypton capacities on the solid sorbent materials studied to date are quite high, they also have high capacities for air components, such as nitrogen, oxygen, and argon. These components are desorbed when the xenon and krypton are desorbed. In the case of the krypton desorption, these air components have a significant impact on the quality of the krypton-rich gas to the extent that it is not sufficiently pure (<50%) to be sent directly to storage. Hence, a krypton purification unit operation is installed to increase the krypton purity to acceptable levels and reduce the total gas volume for storage.

5. Iodine recovery based on silver-loaded mordenite requires multiple columns of material. This is because of the rate the sorbent is consumed in removing iodine and bromine released to the off-gas during UNF dissolution and the chlorine contained in the acid used to dissolve the fuel. The dimensions of the hot cell also limited the height of the columns and required the use of multiple columns. In addition, airflow dynamics through the column and engineering norms imposed dimensional limitations that resulted in the need for multiple columns.

6. The alternative iodine sorbent, silver-functionalized silica aerogel (Ag^0-aerogel), is less dense than the mordenite material, but it contains four to five times more silver (the active ingredient for removal of iodine). The combination means that the number of columns could be reduced or the time between column change out could be lengthened by roughly a factor of two.

7. Important to the sizing and use of the iodine sorbent is the amount of physisorbed iodine (i.e., the iodine that is not chemically bound to the sorbent). The absolute quantities of physisorbed iodine are uncertain for all the materials proposed as iodine sorbents under actual operating conditions. Little physisorption was observed on mordenite and negligible on Ag^0-aerogel in laboratory tests with a thin bed.

8. There is also lack of pertinent engineering data such as the length of the mass transfer zone, sorption kinetics, and sorption capacity for the capture of iodine at the much lower concentration found in the vessel off-gas systems.

The Engineering Evaluation drew upon the available literature to conduct the equipment sizing and sorbent usage. A number of assumptions were required to complete this analysis, and major gaps in the available data were identified that, if resolved, could increase the fidelity of the engineering design. In FY 2017, an assessment was conducted that examined the identified data gaps in the Engineering Evaluation and the assumptions that were required to complete that evaluation. This assessment provided a detailed look at the specific data needs for each major system [2]. Across the off-gas control systems evaluated, several common data gaps became apparent.

- Capacity and adsorption rate data needs are similar for most of the unit operations considered. These two parameters dictate the dimensions and operating conditions of the sorbent beds and wet scrubber systems. The capacity dictates the mass and volume of Ru, ^{3}H, I, and Kr/Xe sorbent beds for a given adsorption duration; the adsorption rate dictates the required depth of the solid sorbent beds and height of the wet scrubbers. Co-adsorption of other species is important to the extent that it interferes with the adsorption of the target species, exceeds the sorbent capacity; affects process operation; impacts handling, recycle, or disposal of the spent sorbent/scrub solutions; and influences the waste form and disposal of the captured species. This is of secondary importance compared to demonstrating and optimizing capacity and adsorption rates for the target species; but co-adsorption and its effects are areas where generally even less is currently known for both sorbent beds and wet scrubber systems.

- Sorbent particle and bulk densities impact the volume of the adsorption system for a given mass of sorbent, but these physical properties data are readily measured.

- For systems that include regenerable sorbents, data on desorption and multicycle operations is also critical for sizing the subsequent recovery systems, designing additional purification steps, determining the useful or practical lifetime of the sorbent, or determining how many cycles it might last, and converting it to a waste form. In general, obtaining this type of data requires larger, more complex test beds and must be able to test the effects of co-adsorbed species. Regenerable sorbents are being tested for the separation of krypton and xenon and

the removal of tritium. The scrub solution used in the CO_2 scrubber is recycled back to the scrubber after precipitating out the captured CO_2 and may also be considered regenerable.

Direct Consolidation of Iodine-loaded Ag^0-Aerogel to Fully Densified Waste Form *[J. Matyáš, E. Ilton, N. Canfield, G. Kroll, and S. E. Sannoh (PNNL)]*

Silver-functionalized silica aerogel has been shown to have a high decontamination factor in excess of 10,000 and a high sorption capacity (~480 mg/g) for iodine due to its high silver content. Current activities are focused on the development of an engineered form of this sorbent and on the high-temperature consolidation processes to convert the iodine-loaded sorbent to a silica-based waste form.

Small-scale (<1.5 g of material) Ag^0-aerogel consolidation experiments with hot uniaxial pressing, hot isostatic pressing (HIP), and spark plasma sintering (SPS) demonstrated that AgI-SiO_2 is as an attractive choice for long-term immobilization of radioiodine [3-5]. Of these, HIP and SPS are the two most promising methods [6].

Small-scale successfully demonstrated that an iodine-loaded Ag^0-aerogel could be processed using HIP at 1200°C for 30 min under 207 MPa to produce a fully consolidated waste form. The apparent density was ~3 g/cm^3, and the sample had an iodine loading of ~36 mass%. Scanning electron microscopy examination revealed the presence of AgI inclusions predominantly distributed inside the silica grains. Samples processed with SPS resulted in a product with low or no porosity. Fully consolidated samples with apparent densities ranging from ~2.4 to 2.8 g/cm^3 were obtained after 120 min at 1100°C, and after 30 and 120 min at 1200°C. The AgI inclusions were predominantly distributed inside of the silica grains with some along the grain boundaries. Lower iodine loadings did not change the distribution of AgI inclusions in the SPS samples examined. They were still mostly concentrated inside of the silica grains (Matyáš et al. 2017).

Recent scaled-up consolidation of highly iodine-loaded Ag^0-aerogel with HIP and SPS has also been successfully demonstrated [6]. HIP processing of a ~115 g pellet at 1200°C with a 30-min hold and under 207 MPa pressure produced a puck ~4.8 cm in diameter and ~2.2 cm thick. The sample contained ~43.8 mass% iodine, which was retained in the form of uniformly dispersed nano- and micro-particles, and micrometer sized "veins" of AgI surrounding individual grains. The sample had a bulk density of 2.90 ± 0.01 g/cm^3; it contained a small amount of open porosity (~7%), but this porosity is expected to diminish with a better container design. SPS processing of ~115 g powder at 1200°C with a 30-min hold and under 32 MPa pressure produced a puck ~6 cm in diameter and ~7 mm thick. The sample contained 41 mass% iodine, had a bulk density of 2.94 ± 0.02 g/cm^3, and exhibited a small amount of open porosity (~2.8%). Figure 1 shows representative samples from these tests.

HIPing of iodine-loaded Ag^0-aerogel at 1000°C resulted in porous sample with void volume fraction of 22%. Increase of pressure from 207 to 379 MPa at 1000°C did not decrease the amount of open porosity but slightly increased the size and number of AgI inclusions at the interface of grains. The higher pressure caused plastic deformation of grains at this temperature and resulted in expulsion of AgI inclusions outside of silica grains, producing a network of thick veins of AgI throughout the sample. These results suggest that a combination of lower

temperature and high pressure during HIP processing is not recommended for production of a waste form with AgI inclusions entrapped inside the silica grains [7].

Figure 1: Pucks of iodine-loaded Ag0-aerogel produced with HIP (left) and SPS (right) (Matyáš et al. 2015)

Consolidation tests showed that a fully densified waste form can be produced and 100% retention of iodine achieved without using any additives. However, if needed, a small amount (~5 wt%) of additives, such as colloidal silica or silica aerogel, can be added to significantly limit or prevent accumulation of AgI inclusions outside of silica grains [4].

Demonstrated Separation of Krypton and Xenon by a Multi-Column AgZ-PAN/HZ-PAN Solid Sorbent System *[T. G. Garn, M. Greenhalgh, A. K. Welty, T. L. Watson (INL)]*

Recent studies conducted at INL demonstrated the separation of xenon from krypton in a mixed gas feed stream using silver- and hydrogen-mordenite-polyacrylonitrile (AgZ-PAN and HZ-PAN) solid sorbents. The results of initial testing of a multi-column system with AgZ-PAN and HZ-PAN indicated that an excellent separation of xenon from krypton could be attained with these sorbents [7]. These initial test results were then extended by demonstrating scaled-up sorbent beds, testing of varied feed gas compositions, and improving analysis of the individual capture columns' compositions.

A schematic of the multi-column experimental test setup is shown in Figure 2. This design allows controlled gas flow initiated from the feed gas cylinders to be routed through both columns, to be directed through either column, or to bypass both columns and flow directly to the gas chromatography–thermal conductivity detector (GC-TCD). All gas flow can be routed through the GC-TCD in any valve lineup configuration. Locations for sample bomb connections to analyze individual column compositions are also shown in Figure 2 near V05 and V20 located on the lines from SC#1 and SC#2 respectively.

Two separation tests were performed with the AgZ-PAN (xenon column) temperature held at either 295 or 253 K while the HZ-PAN (krypton column) was held at 191 K. The tests were performed with a feed gas of 400 ppmv xenon and 40 ppmv krypton in air, with a superficial velocity of 23 cm/min. The adsorption phases of the tests were operated for ~300 minutes, and the effluent from the columns was monitored using a gas chromatography–thermal conductivity detector. Neither xenon nor krypton was detected in the column effluent. The gases captured on both columns were sampled with evacuated sample bombs and subsequently analyzed via gas chromatography–mass spectrometry for both xenon and krypton. The results of the gas chromatography–mass spectrometer (GC-MS) analysis are presented in Table 1.
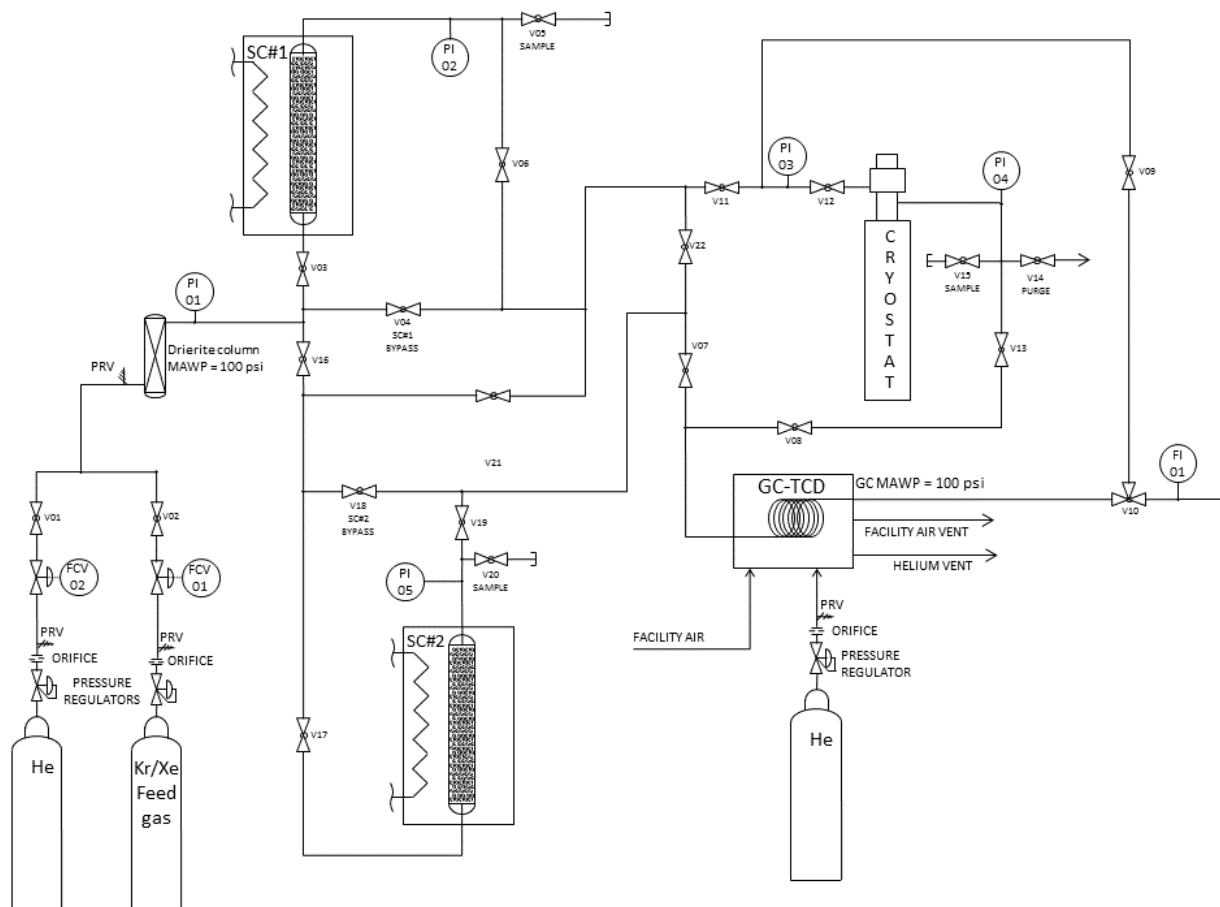
Figure 2: Multi-column experimental test bed schematic for separation of Kr/Xe [9]

Table 1. Xe/Kr concentrations from sample bombs [10]

Test #	Temperature (K)	Column Sample	Average Xe (ppmv)	Average Kr (ppmv)
1	295	Xe Column	41.3	81.2
	191	Kr Column	BDL	367
2	253	Xe Column	25.6	179
	191	Kr Column	BDL	586

Note: BDL (below detection level)

These results indicated that an excellent separation of xenon from krypton was achieved, as there was no detectable xenon found in the krypton column samples. The low xenon concentration for the xenon column samples along with the desorption data revealed that removal of xenon from the AgZ-PAN was ineffective. The sample bomb gas collection procedure provided inconsistent results, even though a combination of high temperature and purge gas flow was used during the sampling of the columns.

These initial tests of the multi-column system were followed by a series of tests to demonstrate the selective separation of xenon and krypton from each other and from the bulk air stream at gas flow velocities representative of an operating nuclear fuel reprocessing facility [1]. The feed gas for these tests contained 150 ppm krypton and 1,000 ppm xenon with the balance air. The feed rates were either 500 or 2,000 sccm to the multi-column test bed. This resulted in superficial velocities 10–40 times higher than previous work. The first column contained AgZ-PAN to capture xenon. The AgZ-PAN column temperatures tested were 295 K and 253 K. The xenon-free krypton effluent stream was routed to a second column packed with HZ-PAN operating at 191 K to capture krypton [9].

The results from these tests indicate that increasing superficial velocity does not impact sorbent capacity or initial breakthrough for AgZ-PAN. For HZ-PAN, however, there is some indication that the higher feed rate (2,000 sccm) may result in an earlier initial breakthrough of krypton. The data obtained in these tests indicate that HZ-PAN capacity for krypton does not appear to be affected by the presence of xenon in air. It is unknown whether this will remain true at higher concentrations of krypton and xenon, such as those that would be seen if adsorption columns are staged for additional krypton concentration or polishing. A complete operating cycle has not yet been optimized but will be the focus of future work [9].

Analysis of Legacy Krypton Waste Form Samples *[R. T. Jubin and S. H. Bruffey (ORNL)]*

Legacy samples composed of ^{85}Kr encapsulated in solid zeolite 5A material and five small metal tubes containing a mixture of the zeolite combined with a glass matrix resulting from hot isostatic pressing have been preserved. The samples were a result of krypton R&D encapsulation efforts in the late 1970s performed at the Idaho Chemical Processing Plant. These samples were shipped to ORNL in mid-FY 2014 and repackaged into individual glass sample bottles for further analysis. Visual examination and x-ray imaging conducted in 2015 revealed that two of the five capsules had been breached. Capsule 2 was observed to have saw marks on the capsule, and a quantity of bead-like loose pellet material remained in the capsule. Capsule 5 had been opened at an undetermined time in the past. The end of this capsule appears to have been cut off, and there are additional saw marks on the side of the capsule. X-ray images confirmed the presence of residual material within this capsule. The material appears to be compacted but still retains some of the bead-like morphology.

Based on these initial images and the fact that there are at least two breached samples, it was proposed that FY 2016 efforts be directed at analysis of the breached specimens before opening the three intact capsules. Portions of material removed from Capsules 2 and 5 (Figure 3) were analyzed to determine the fraction of krypton/xenon remaining in the matrix and the amount of rubidium remaining in the matrix. The presence of rubidium, the sole decay product of ^{85}Kr, was of special interest because its corrosive nature could have significant long-term impacts on the integrity of container structures for any proposed krypton waste form. Owing to the high radiation levels of these samples, all analyses beyond the initial imaging were performed in ORNL hot-cell facilities.
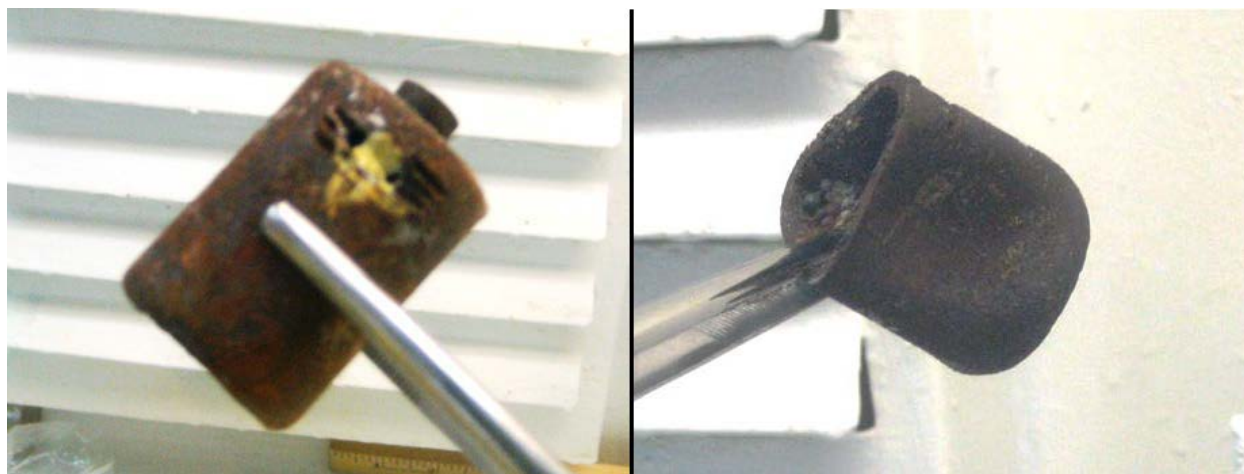
Figure 3: Krypton-85 legacy waste form capsule 2 (left) and capsule 5 (right)

The materials contained in Capsules 2 and 5 were examined by scanning electron microscopy–electron dispersive spectrometry (SEM-EDS) and x-ray diffraction. There appears to be a relatively uniform distribution of krypton and rubidium throughout the pellets examined (Figure 4). The EDS-derived chemical analysis indicates a chemical composition of the recovered materials consistent with zeolite 5A. The elemental mapping indicates that krypton and xenon are coincident with the Si, Al, Ca, and Na implying that the noble gases have remained within the collapsed zeolite structure (Figure 4) [11]. The material contained within Capsule 5 showed ~1 at.% lead. The origin of the lead is currently indeterminate. X-ray diffraction analysis shows a significant shift from the 5A structure, most likely due to the krypton encapsulation/sintering process that occurred when the samples were made.

The walls of each capsule were also examined and showed extensive corrosion throughout (Figure 5). Elemental mapping of the capsule material appeared consistent with carbon steel, but the weld material appeared consistent with a stainless steel. The interior surface of the capsule appeared to have a layer of material containing Al, Si, and Ca similar to the 5A molecular sieve. Analysis for rubidium, a strong oxidizer and ^{85}Kr daughter, within the corrosion sites was inconclusive.
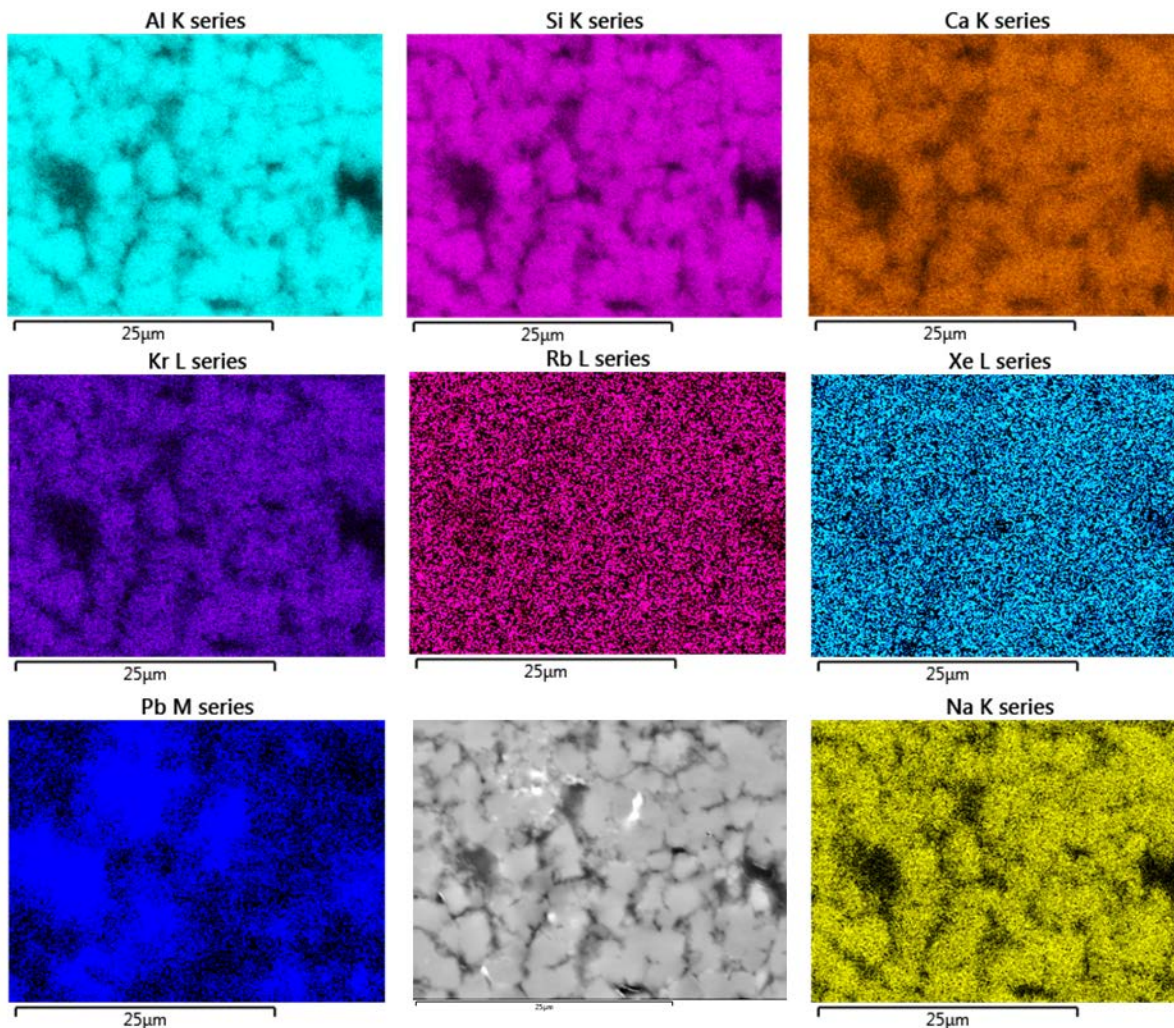
Figure 4: Elemental maps of an area near the center of a pellet recovered from Capsule 5 (sample K5)
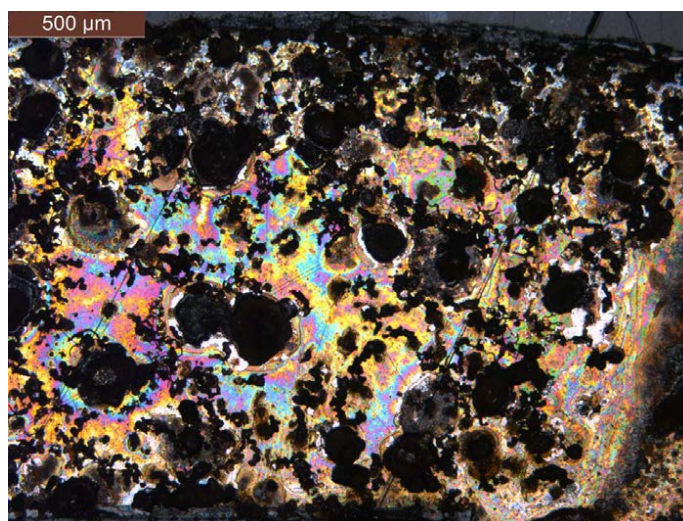

Figure 5: Optical microscope image at 50× magnification of Section "A" of Capsule 2

CONCLUSIONS

The Off-Gas Sigma Team has made considerable progress over the past 8 years in advancing off-gas capture technology needed for UNF reprocessing. The numerous accomplishments include development of new sorbents for iodine; investigation of iodine capture from very dilute streams; development of solid sorbents to capture and separate xenon and krypton at near room temperature; demonstration of krypton recovery using multiple columns at moderate temperatures; and performance of overarching systems analyses. Several of these have been highlighted as examples of the work performed by this team of dedicated professionals.

Work continues to address the remaining data gaps that were identified as part of the Engineering Evaluation. These ongoing activities include iodine capture for the various iodine-containing process streams, continued development of iodine sorbents, tritium capture systems for advanced head-end processes, the scale-up of the krypton/xenon capture and purification systems, and systems to capture semivolatile radionuclides.

REFERENCES:

1. Jubin, R. T. J. A. Jordan, B. B. Spencer, N. R. Soelberg, A. K. Welty, M. Greenhalgh, D. M. Strachan, and P. K. Thallapally, Engineering Evaluation of an Integrated Off-Gas Treatment System for Used Nuclear Fuel Reprocessing Facilities, FCRD-MRWFD-2016-000313, ORNL/TM/2016-447, INL/LTD-16-39698, Oak Ridge National Laboratory, Oak Ridge, TN, September 2016. (Note: This report is Official Use Only–Export Controlled Information.)

2. Jubin, R. T., S. H. Bruffey, J. A. Jordan, B. B. Spencer, N. R. Soelberg, A. K. Welty, and M. Greenhalgh, Data Requirements and Test System Needs for Development of an Integrated Off-Gas Treatment System, FCRD-MRWFD-2017-000422, ORNL/TM/2017-314, Oak Ridge National Laboratory, Oak Ridge, TN, June 2017.

3. Matyáš J. and R. K. Engler, Assessment of Methods to Consolidate Iodine-loaded Silver-functionalized Silica Aerogel, FCRD-SWF-2013-000589, Pacific Northwest National Laboratory, Richland, WA, 2013.

4. Matyáš J. and A. V. Walters, Consolidation of Iodine-loaded Ag^0-functionalized Silica Aerogel with HUP, HIP, and SPS, FCRD-MRWFD-2015-000480, Pacific Northwest National Laboratory, Richland, WA, 2015.

5. Matyáš, J., N. Canfield, S. Sulaiman, and M. Zumhoff, "Silica-based Waste Form for Immobilization of Iodine from Reprocessing Plant Off-gas Streams," *Journal of Nuclear Materials*, 476: 255–261, 2016.

6. Matyáš, J., S. E. Sannoh, G. R. Kroll, and N. Canfield, Scaled-up Consolidation Processing of Iodine-loaded Ag^0-functionalized Silica Aerogel, FCRD-MRWFD-2016-000042, Pacific Northwest National Laboratory, Richland, WA, 2016.

7. Matyáš, J., S. E. Sannoh, and N. Canfield, Impact of Iodine Loading, Additives, Temperature, and Pressure on the Microstructure of HIP- and SPS-processed Ag^0-Aerogel, NTRD-MRWFD-2017-000218, Pacific Northwest National Laboratory, Richland, WA, 2017.

8. Welty, A. K., T. G. Garn, and M. Greenhalgh, Superficial Velocity Effects on HZ-PAN and

AgZ-PAN for Kr/Xe Capture, FCRD-MRWD-2016-000024, April 01, 2016.

9. Welty, A. K., K. L. Lyon, M. Greenhalgh, T. G. Garn, A. Ladshaw, and A. Wiechhert, Multi-Column Kr/Xe Separation Testing at Representative Superficial Velocities with DGOSPREY Model Validation, NTRD-MRWFD-2017-000033, July 24, 2017.

10. Greenhalgh, M., T. G. Garn, A. K. Welty, and T. Watson, Multi-Column Xe/Kr Separation with AgZ-PAN and HZ-PAN, FCRD-MRWFD-2016-000046, August 19, 2016.

11. Jubin, R. T. and S. H. Bruffey, Analysis of Selected [85]Kr Legacy Samples, FCRD-MRWFD-2016-000047, ORNL/TM-2016-350, Oak Ridge National Laboratory, Oak Ridge, TN, September 2016.

ACKNOWLEDGMENTS