The Effect of Kr-85 Decay to Rb-85 on Waste Forms — 17105*

R. T. Jubin,[1] S. H. Bruffey,[2] B. B. Spencer,[3] and D. M. Strachan[4]

[1]*Oak Ridge National Laboratory, Oak Ridge, TN 37831, jubinrt@ornl.gov*
[2]*Oak Ridge National Laboratory, Oak Ridge, TN 37831, bruffeysh@ornl.gov*
[3]*Oak Ridge National Laboratory, Oak Ridge, TN 37831, spencerbb@ornl.gov*
[4]*Strata-G, LLC, Knoxville, TN 37932, dmsllc@yklc.net*

ABSTRACT

Krypton-85 (Kr-85) is a volatile radionuclide released during the reprocessing of used nuclear fuel (UNF). Prior to discharge of process off-gas streams to the environment, removal of Kr-85 is required to meet emission regulations. Once removed from the off-gas streams, Kr (containing stable Kr in addition to Kr-85) must be placed in storage until the Kr-85 has decayed to levels such that its potential release would meet regulatory guidelines. The current method of storage is high-pressure metal canisters. Historically, concerns have been expressed that the decay product of Kr-85, Rubidium-85 (Rb-85), could cause corrosion and weakening in the canisters that would ultimately lead to premature failure and release of Kr-85. Alternate methods and materials, such as matrix isolation in a vapor deposited metal and immobilization in zeolites, have been investigated as storage approaches for Kr. However, even these storage methods may become compromised due to the chemical effects of Rb-85 arising from the decay of Kr-85. A review was conducted of the available literature to examine topics relevant to the effects of Rb on the long-term storage of Kr-85, examine the source terms in the context of this concern, and suggest a path forward to resolve outstanding knowledge gaps.

In the course of this analysis, several knowledge gaps were identified. Examples include:
- The use of solid sorbents to potentially permit more Kr-85-bearing gas to be loaded into canisters, either at the pressures used in the current method or at significantly lower pressures.
- A cost-to-benefit analysis is needed to ascertain the practicality of manufacturing waste forms for Kr-85 storage with solid sorbents.
- The effects of radiation and chemical transmutation in solid sorbents or matrices for this application are virtually unknown.
- Fundamental data are needed on corrosion rates and mechanisms as functions of Rb concentration, storage temperature, etc., for various storage

approaches (e.g., as compressed gas or encapsulated in a getter material) and storage container materials.

The present analysis focused on the available information regarding storage of Kr-85 in high-pressure containers. It was judged unlikely that such containers would be compromised by corrosion from the Rb decay product, especially if the Kr recovery operations result in a gas composition with very low oxygen content. However, this conclusion should be revisited based on preliminary data gathered from analysis of legacy Kr waste forms. These legacy waste forms are a result of experiments conducted at Idaho National Laboratory over 30 y ago and have been stored for more than three Kr-85 half-lives. Evaluation of these forms indicates significant corrosion, the cause of which has not been conclusively determined.

INTRODUCTION

One of the volatile radionuclides that requires removal from process off-gas streams, during the reprocessing of used nuclear fuel (UNF) to meet regulations, is krypton (Kr). Of the Kr isotopes that are present in UNF, Krypton-85 (Kr-85) ($t_{1/2}$ = 10.7 y) is the only remaining radioactive isotope after the fuel has been out of the reactor for one year; all other isotopes (Kr-82, Kr-83, Kr-84, and Kr-86) are stable [1]. If UNF is reprocessed within about 50 y, Kr capture is needed to meet regulatory guidelines [2]. Once removed from the off-gas streams, Kr must be placed in storage for a period not less than that required for the Kr-85 to decay to levels such that its release would meet regulatory guidelines. The currently proposed method of storage is high-pressure metal canisters. However, concerns have been expressed that the decay product of Kr-85, Rubidium-85 (Rb-85), could cause corrosion and weakening in the canisters, ultimately leading to premature failure and release of Kr-85 [3]. Rubidium metal, with a melting point of about 39 °C, is very corrosive, and the decay heating of the canister is likely to raise the temperature above the melting point. Vapor deposition in a metal matrix, immobilization in zeolites, and immobilization in metal-organic framework (MOF) materials have all been investigated as alternative storage media for Kr [4-13]. Even these storage methods may still be susceptible to the chemical effects caused by decay of Kr-85 to Rb-85.

In the late 1970s, an research and development (R&D) effort to study Kr-85 encapsulation and leakage was performed at Idaho National Laboratory (INL) by Christensen, et al. [6-7, 14-15] Off-gas resulting from fuel dissolution underwent treatment, with the fission products sent to the Rare Gas Recovery Facility at the Idaho Chemical Processing Plant (ICPP) where the Kr-85 was removed with cryogenic distillation and collected in gas cylinders. A cylinder containing the Kr-85 was transferred to the Multi-Curie Cell where the encapsulation studies were completed.

The Kr-85 R&D encapsulation effort incorporated Kr-85 in numerous materials, including sodalite, "thirsty" glass, and zeolite 5A, with zeolite 5A reportedly showing

the best results. Some of these materials have recently been recovered for examination. Because the testing included numerous materials, there is a question as to the exact nature of the legacy samples. It is assumed that because the zeolite 5A material showed the most promise, these samples represent the zeolite material. Further support of this assumption was a verbal communication with one of the original researchers (retired), who stated that the samples included "loose" zeolite 5A encapsulating Kr-85 and zeolites hot isostatic pressed (HIPed) in a glass matrix contained in squashed metal tubes [16]. Fig. 1 shows one of the metal tubes.

The general sequence of the work for examining the legacy samples was originally planned in three stages and was discussed at WM 2016 [17]. Stage 1 included non-destructive analyses (NDA) of the samples (radiation levels, contamination levels, and gamma signature) and neutron or x-ray imaging. X-ray tomography was performed and allowed the capsules and the interior to be viewed along the three axes. X-ray tomography was used in place of the originally planned neutron tomography due to greater availability of the instrument. A hold point was established at the completion of the NDA characterization to review the results and develop the detailed plans for the next phase of analysis that would involve opening one of the capsules and sub-sampling. These legacy samples are now being examined to determine the potential effects of the decay daughter Rb-85 on the encapsulation media and storage capsules and to compare the observed behavior to that anticipated based on the literature.

Fig. 1: Photo of a metal tube, presumably containing potentially loose zeolites that had not undergone HIP [18].

THE DECAY OF KR-85

Krypton-85 has a half-life (t$_{1/2}$) of 10.76 y and decays to stable Rb-85 by emission of a beta particle (ß), and an antineutrino (ū), both of equal energy (0.687 MeV).

Marks et al. [19] provide the formulas for calculating the contributions to the recoil energy from the ß⁻ and ū15. The effect of this recoil is discussed below.

With such a relatively short $t_{1/2}$, after 50 years of storage only 3.6% of the original stored amount of Kr-85 remains. Rubidium, a Group I alkali metal with a melting point of about 39 °C, is known to be corrosive as a liquid, especially in the presence of an oxidant. Historically, the potential corrosion of Kr-85 storage containers by the decay product Rb-85 has been of concern [20]. In addition, the energetic emissions resulting from ß⁻ decay and any potential electrical or chemical changes (i.e., production of Rb⁺, a charged particle, in place of neutral Kr) also have the potential to damage the waste form and, potentially, storage containers.

Storage of Kr in low-pressure (assumed 0.1 MPa) cylinders is attractive from the standpoint of reducing the risk of large gas releases in the event of corrosion-assisted rupture of a cylinder, but it can result in substantially higher storage volumes. The use of higher pressures (≥5 MPa) reduces the stored waste volume with a concomitant increase in the probability of leakage or cylinder rupture caused by Rb corrosion (and also a larger release per unit storage volume).

CHEMICAL PROPERTIES OF RUBIDIUM

Rubidium is a Group I metal with a melting point of about 39 °C and a boiling point of 688 °C. It is most common as a solid at room temperature, with a density of 1.53×10^3 kg/m³. Liquid metal Rb has a density of 1.46×10^3 kg/m³ at the melting point. Rubidium has two naturally occurring isotopes: stable Rb-85 (with a natural abundance of 72.2%) and radioactive Rb-87, (with a natural abundance of 27.8% and a $t_{1/2}$ of 4.92×10^{10} y). Like other alkali metals, Rb is strongly reactive with water and can ignite in air. For this reason, it is recommended that it be stored and handled in an inert atmosphere. Pure Rb metal may not cause substantial corrosion to materials it contacts when handled in this way, but in the presence of oxygen it will form highly corrosive rubidium oxides. Thus, when evaluating any potential storage mechanism the presence of oxygen should be considered.

KRYPTON SEPARATION

There are multiple processes that can be used to separate and purify Kr, and these methods can result in different oxygen concentrations in the product stream. The most common industrial separation is cryogenic distillation. When used to separate Kr-85, this process inherently results in very low oxygen contamination because O_2 should be removed prior to distillation to avoid the formation of ozone when Kr-85 is present in the liquid air [21]. Fluorocarbon adsorption has the capability to capture and purify multiple noble gases based on their solubility in CCl_2F_2 (commonly known as R-12). The product stream from fluorocarbon adsorption has higher levels of contaminants than that from cryogenic distillation, and the oxygen content is estimated to be 1.4% [22]. The third process that can be used to separate Kr is adsorption on solid sorbents, such as zeolites or MOFs. While Liu et

al. [23, 24] show that all air components pass through MOFs used to separate Kr and Xe from air, data regarding the oxygen content of both zeolites and MOFs when used to remove Kr were not found in publically available literature. It should also be noted that zeolites in particular have a high affinity for water.

STORAGE OPTIONS FOR Kr-85

Options for Kr-85 storage include storage as a compressed gas in gas cylinders, vapor co-deposition within a metal on the inside of a canister, or immobilization in a solid sorbent. If a solid sorbent is selected, the Kr-containing sorbent, once loaded, can then be disposed of through various methods such as direct disposal, hot press compaction, or other processing methods. In this section, the potential impacts of Kr-85 decay to Rb-85 on cylinder storage, vapor deposition, and immobilization on two common sorbents are discussed.

Cylinder Storage

Assuming that Kr-85 is stored in high-pressure cylinders (canisters) to minimize waste volume, the resulting Rb-85 is likely to exist within the metallic cylinder as a uniformly distributed alloy on the metal walls. Corrosion by liquid metallic Rb has been studied for a variety of metals [4, 25 – 41]. Pinchback and Knecht [41] evaluated stainless steel alloys and other alloys for use as canisters to hold pressurized Kr, specifically looking at the potential that Rb corrosion could compromise the canister integrity. They subjected coupons of various metal alloys (Inconel, 316 stainless steel, 304 stainless steel, and others) to liquid Rb at elevated temperature (400-672 °K) for over 200 days. They found that several alloys (notably including 316 stainless steel) were not corroded. One exception was 304 stainless steel that was corroded by the liquid Rb. While these test results are promising, it is unclear how realistic they are as they were conducted in the absence of oxygen. Borgstedt et al. [26] conducted corrosion testing on several different metal alloys with non-purified Rb (containing Rb oxides and hydroxides). These tests were conducted at 150 °C and 200 °C for up to 1250 d. Borgstedt and coworkers concluded that corrosion was limited and did not impact the integrity of the metals. Based on the results of these studies, it is likely that the presence of Rb in high-pressure Kr-85 storage cylinders will not negatively impact the long-term integrity of the canisters provided oxygen is excluded.

Ion Implantation

Co-deposition of Kr with metal vapor (also known as ion implantation) has been considered by multiple researchers [4, 5, 42]. This type of process would result in a metal matrix containing about 5 mass% Kr [42] and, since the concentration of Kr-85 is about 4 mass% of all Kr collected from the off-gas, the resulting metal matrix would contain about 0.2 mass% Rb after all the Kr-85 decayed. At these concentrations, a separate liquid phase would not be expected. The Rb^+ would become part of the metal matrix as an interstitial particle either as an ion or metal

after gaining an electron from the metal through conduction, probably the conduction band. From a study of transmutation effects on crystalline materials potentially used for nuclear waste, it was determined that, even in these non-conducting materials, electrons were available from neighboring atoms or from the unoccupied conduction band and, at relatively high concentrations of the decaying atom, some defects were likely because of the changes in atomic volume [43 - 46]. At the low concentrations expected for the decay of Kr-85 to Rb-85, the electrons might be extracted from the conduction band, but little effect is expected on the overall crystal structure of the vapor deposited metal.

Immobilization in Solid Sorbents

One option to decrease the cost of Kr storage is to pack the canisters with the sorbent used to remove Kr from the off-gas. It is also possible to put more Kr in a canister at a given pressure if a sorbent is present. For example, Mueller et al. [13] showed that more Xe could be placed in cylinders packed with an MOF. Work at the Pacific Northwest National Laboratory has shown that MOF materials have an affinity for Kr, as they do for Xe, especially in the absense of Xe [24]. The effects on these sorbent materials will be discussed below. It is assumed that if they degrade significantly the Rb would come into contact with, and possibly compromise, the canister. Similarly, Kr-85 could be isolated in a zeolite. Few or no data have been reported on the behavior of Rb ingrowth within a consolidated zeolite structure.

Effects Kr-85 Decay on the Structure and Chemistry of Solid Sorbents

When Kr-85 decays to Rb-85, there are two primary processes that may affect the solid sorbent structure and its long-term viability as a Kr waste form. The first process is that of the decay itself – the potential damage to the solid that can result from either β^- emission or from the recoil of the Rb-85. The second is that of the charge distribution that must result when Kr-85 decays to $Rb-85^+$ with the release of a β^- particle.

In the case of radioactive decay the damage to ceramic materials, such as a zeolite, are unknown. The full radiation damage effects of the ß$^-$ on the solids have not been fully investigated [19]. However, Marks et al. [19] have worked out the energies associated with the recoiling decay daughter in $SrTiO_3$ as ^{90}Sr decays (β^- decay) through ^{90}Y to ^{90}Zr. The bond strengths (threshold displacement energies) in strontium titanate are sufficiently strong (typically 50 to 150 eV) to avoid radiation damage from the recoiling Y-90 and Zr-90 atoms. For the decay of Kr-85, the average recoil energy for the Rb-85 daughter is 1.2 aJ[1] (7.3 eV) from the ß$^-$ and 0.5 aJ (3 eV) from the antineutrino. It is expected that the bond strengths for zeolite materials would be similar to those in the titanate. It can be concluded from this information regarding decay energetics that the decay of Kr-85 (which is

---

[1] aJ = atto Joule (1×10^{-18} J)

energetically of the same order of magnitude as Sr-90 decay) is unlikely to result in radiation damage in zeolites.

Secondly, the effects of the redistribution in charge must be considered. For $SrTiO_3$, the transmutation from Sr to Zr had some surprising structural stabilities that could not have been anticipated without the thorough examination of the possible phase transitions and the energetics associated with each. One comment at this point is that Marks et al. [19] did not consider the kinetics of the phase transformations; it is unlikely that this information exists. The other transmutation that Marks et al. [19] discussed was SrH_2 to ZrH_2 where much more information is available, both end members exist, and there is a clear phase transformation pathway (radioparagenesis) from SrH_2 (PbCl$_2$-type) to ZrH_2 (face-centered tetragonal). Thus, for the chemical transformation from Kr to Rb in both zeolites and MOFs, it is unclear what the overall effect is and how to calculate or experimentally determine it. Because Kr and the daughter Rb are isolated in cages within the structure, rather than being incorporated into the structure, the effects in these materials are probably much different than if the Kr were integral to the overall structure of the material. There is likely to be a structural effect of Kr transmuting to Rb based just on the size change [Kr (0.112 nm) and Rb (0.216 nm atomic; 0.161 nm ionic)] and how the charge change is compensated. A more thorough examination of the effect should be considered if this approach is utilized for the immobilization of Kr.

Organic molecules, including MOFs, are more likely to be ionized from the recoiling Rb-85 atom. The bond strength for a C-H bond is roughly 0.7 aJ (4.4 eV) with the C-C bond being slightly less. Ionization potentials are about twice that of the bond energies [47]. Also, the change in chemical state from a neutral atom to a charged ion may enhance the overall susceptibility to Kr-85 transmutation.

Certain MOFs currently under examination for Kr-85 capture [48] have been demonstrated to hold five to six atoms of Kr per cage. As only 5% of the total Kr captured will be Kr-85, it is likely that the solids may be able to absorb the associated radiation damage and possible charge redistribution without significant effect upon Kr retention and long-term storage. This is a hypothesis that requires further investigation because MOFs are likely to be more damaged than zeolites by the chemical and radiative events.

Penzhorn and coworkers [10] determined that Ar, Kr, and Xe can be isolated in zeolite A, mordenites, and chabazites even when they are hydrothermally vitrified between 340 °C and 650 °C. These gases remained fixed even in the presence of liquid Rb. They measured activation energies for the diffusion of Kr and found the diffusion coefficients to be very small. Depending on the formulation, zeolites can hold two to four Kr atoms per cage unit; Daniel and coworkers [49] found two Xe atoms per formula unit for Ag-loaded zeolite.

The results summarized here seem to indicate that as the Rb concentration increases in the zeolite, the materials become more stable thermodynamically and

the lattices become more rigid, thereby isolating the remaining Kr in the zeolite cage. The latter might be of less importance than other radiation effects since roughly 5% of the total Kr is Kr-85. Hence, it is unlikely that any zeolite cage will contain more than one Kr-85 atom, and blocking the cage opening with the Rb daughter results in a minor benefit.

As indicated above, the bond strengths in zeolites are significantly stronger than the recoil energies from the ß⁻ and antineutrino emission from a Kr-85 decay. This coupled with the low concentration of Kr-85 suggests that the recoil effects are likely to be minor to negligible. Aluminosilicate materials are generally resistant to ß⁻ irradiation. Thus, the effects of pure ß⁻ irradiation from the decay should not cause significant structural damage. Since the damage to the zeolite structure is likely to be minimal, the release of Kr over time would not be expected to increase over the leakage rate from a zeolite containing no radioactive Kr.

As indicated above, there are effects on the sorbent material resulting from the transmutation of one atom into another [43 – 46]. The charge can be compensated though transfer of electrons from the Fermi band or from a multivalent atom with available oxidation states, e.g., Fe(II) and Fe(III). In a zeolite, the matrix is generally constructed from Al and Si: These elements do not have readily available alternate oxidation states. The amount of Kr-85 present in a zeolitic waste form, however, is small, ~ 0.3 mass%. Most solids can tolerate this level of defects without deleterious effects.

PRELIMINARY EXAMINATION OF SELECTED LEGACY Kr-85 STORAGE CAPSULES

The legacy Kr-85 storage capsule samples described in the Introduction section of this paper provide a unique opportunity to examine the potential effects of the decay daughter Rb-85 on the encapsulation media and storage capsules. Two capsules, Capsules 2 and 5, are the first to be examined since these were previously compromised. Each capsule was approximately 3 cm in diameter and 4 cm tall (not including the stem). Capsule 2 was sectioned vertically, i.e., cutting through the stem and the vertical sides of the right circular cylinder with a diamond rotary saw. Water was used to cool the blade during cutting. The sectioned capsule was examined visually with the aid of an optical microscope within the hot cell. Composite images from the optical microscope showing the interior of a halved Capsule 2 are shown in Fig. 2. This figure shows areas of probable corrosion damage on the inside of Capsule 2. Fig. 3 is a magnification of the lower portion of Section A of the capsule that shows pitting and corrosion at multiple points on the inner walls of Capsule 2.

One half of this capsule was then cut into smaller sections that allowed it to be mounted for scanning electron microscopy analyses. The mounted sample from Capsule 2 is shown in Fig. 4. This section is from the lower right portion of the section shown in Fig. 2. The mount was polished to a 1-µm finish. Results from the

optical analysis of the capsule are striking. The extensive corrosion that had been observed in the macro images taken through the stereo-microscope appear to extend through the entire wall thickness. At 50× magnification (Fig. 5), the extent of the corrosion is obvious.

DISCUSSION AND PATH FORWARD

Reprocessing of UNF that has been out of the reactor for less than about 50 y requires the removal of Kr-85 from the process off-gas streams. The decay of Kr-85 to Rb-85 presents challenges to the materials that will potentially be used to remove and store the Kr recovered from the off-gas. To address some of these problems, a literature survey was completed, and the results of have been summarized here.

One storage option is high-pressure gas in canisters. Based on the literature data, it appears unlikely that Kr-85 storage in high-pressure containers will be compromised by corrosion from the decay product Rb, especially if the Kr recovery operations result in a gas composition with very low oxygen content. Similarly, the choice of immobilization through ion implantation is also unlikely to be significantly affected by Kr-85 decay. However, this conclusion is based on past studies that were not performed to the quality assurance rigor needed to qualify a waste form (method) or the processes for a UNF reprocessing facility. Contrary to our assessment based on the literature, recent studies of small legacy capsules that contained Kr-85 encapsulated in a collapsed zeolite appear to show significant corrosion of the capsule wall. These capsules are the subject of significant further analysis.

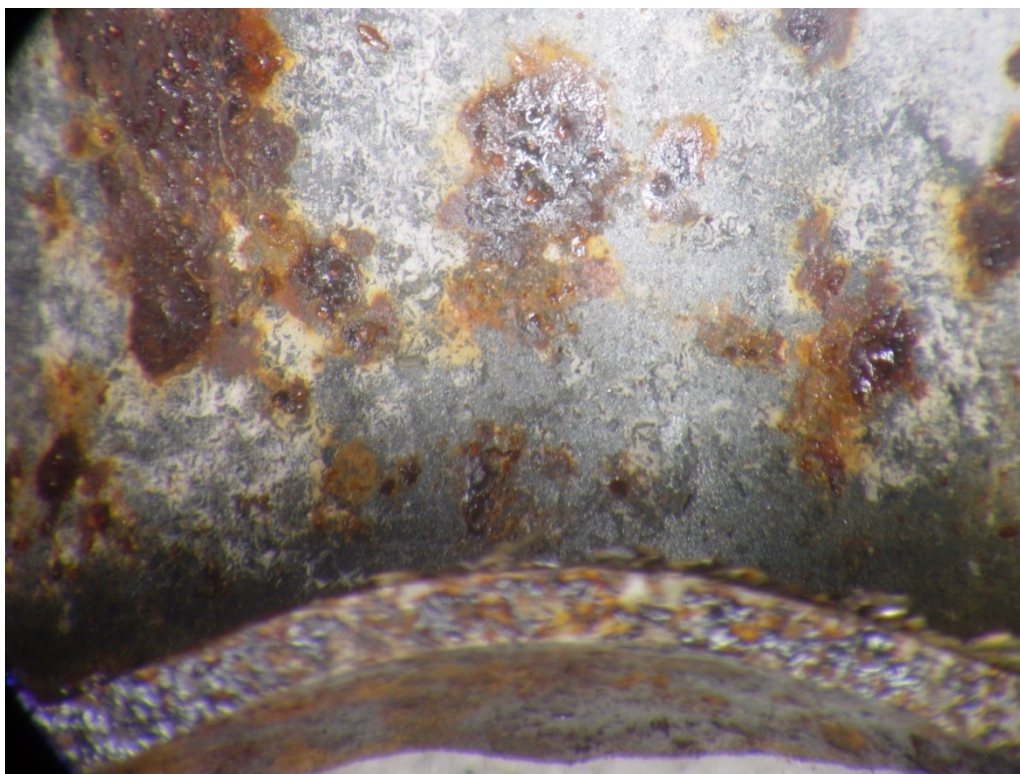Fig. 2: Composite optical microscope image of Section "A" of Capsule 2.

Fig. 3: Detail optical microscope image of Section "A" of Capsule 2 at low
magnification. (This is lower portion of full section shown in Fig. 2.

Fig. 4: Optical microscope image of portion of Section "A" of Capsule 2 wall
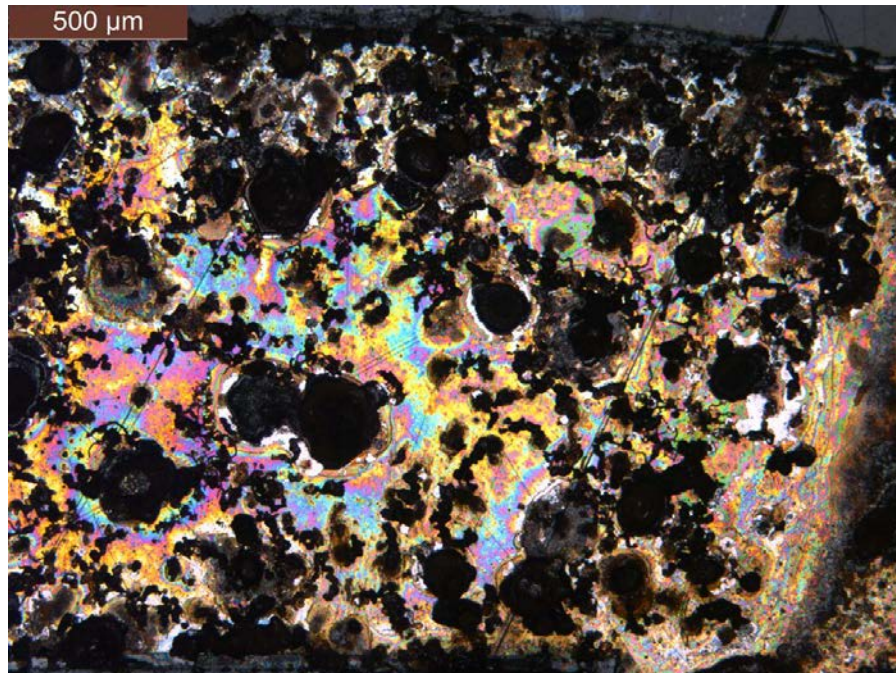(Image MM2B2-0MS-001).

Fig. 5: Optical microscope image at 50× magnification of portion of Section "A" of Capsule 2 wall (Image MM2B2-050X-002).

Study of the immobilization and storage of Kr-85 on solid sorbents has been severely limited, and almost no literature was available on this topic or on related systems. The effects of Kr-85 decay in solid sorbents is likely both an interesting and challenging scientific topic, as both radiation damage and chemical transmutation must be considered when evaluating long-term stability. Based on limited data, it was concluded that zeolites are likely to experience less radiation damage than MOFs from the decay events and that the presence of Rb may be thermodynamically beneficial for long-term retention of Kr within the zeolite matrix. These effects are somewhat ameliorated by the small amount of Kr-85 present. In most materials, this leads to small amounts of Rb in the solid as the Kr decays. It is recommended that the benefits of solid sorbent immobilization for Kr-85 storage be analyzed and that careful consideration be given to answering the myriad unknowns surrounding the effects of Kr-85 decay (both chemical and radiative) on the solids being considered. Again, irrespective of the decision to incorporate MOFs or zeolites in storage canisters, the resistance of these materials to radiation-induced property changes is important to their use as sorbents to remove Kr from the off-gas streams.

On-going is the study of legacy material that was used in the 1970s to immobilize some Kr-85 in the laboratory. While these studies are in their preliminary stages, results indicate that the steel walls of the canisters used to isolate the Kr-85-bearing zeolite 5A material are severely corroded. Based on the results from the

literature, this is an unexpected result. Further investigation should reveal the nature of the corrosion and if Rb-85 participated in the corrosion process.

Further experimental work to verify the viability of cylinder and ion implantation storage methods, and to better characterize the effects of Kr-85 decay on solid sorbents, will provide assurance that Kr-85 can be safely stored for extended periods of time as is required by United States regulation. The work already underway at ORNL to analyze legacy Kr waste forms will be highly beneficial to better understanding the effect of Kr-85 decay on storage methods that allow for contact between metals and Rb. Work has also been planned at INL that will investigate the resiliency of MOFs when exposed to radiation. These two current research efforts should be leveraged to answer outstanding questions regarding the impact of Kr-85 decay on storage methods, and future work should be planned based on the results of these studies.

REFERENCES

1.   R. JUBIN, N. SOELBERG, D. STRACHAN, and G. ILAS, *Assessments and Options for Removal and Immobilization of Volatile Radionuclides from the Processing of Used Nuclear Fuel*. FCR&D-SWF-2011-000305, Oak Ridge National Laboratory, Oak Ridge, TN, (2011).
2.   R. JUBIN, N. SOELBERG, D. STRACHAN, and G. ILAS, *Fuel Age Impacts on Gaseous Fission Product Capture During Separations*. FCRD-SWF-2012-000089, PNNL-22550, Oak Ridge National Laboratory, Oak Ridge, TN, (2012).
3.   R. T. JUBIN, D. M. STRACHAN, and Authors from ORNL, PNNL, INL, and SNL. *Assessments and Options for Removal and Immobilization of Volatile Radionuclides from the Processing of Used Nuclear Fuel*, FCR&D-MRWFD-2015-000429, ORNL-SPR-2015/115, (31 March 2015).
4.   D. S. WHITMELL, "Immobilization of Krypton in a Metallic Matrix." *Nuclear Energy* (British Nuclear Energy Society) 21(3):181-7 (1982).
5.   D. S. WHITMELL, "Choice of Materials for the Immobilization of Krypton-85 in a Metallic Matrix by Combined Ion Implantation and Sputtering." In *Proceedings of the Proceedings of the 18th DOE/NRC Nuclear Air Cleaning and Treatment Conference*, 667-82 pp. The Harvard Air Cleaning Laboratory, Cambridge, Mass (1985).
6.   A. B. CHRISTENSEN, J. A. DEL DEBBIO, D. A. KNECHT, and J. E. TANNER, "Immobilization and Leakage of Krypton Encapsulated in Zeolite or Glass," *Materials Research Society Symposia Proceedings*, v 6, p 525-532, (1982).
7.   A. B. CHRISTENSEN, J. A. DEL DEBBIO, D. A. KNECHT, J. E. TANNER, and S. C. COSSEL. "Immobilization of Krypton-85 in Zeolite 5A," *Proceedings of the DOE Nuclear Air Cleaning Conference*, v 1, CONF-820833), p. 333-356, (1983).
8.   D. A. KNECHT, *Evaluation of Methods for Immobilizing Krypton-85*. ICP-1125, Idaho National Engineering Laboratory, Idaho Falls, ID, (1977).

9.   M. MIYAKE, T. KONISHI, T. SUZUKI, S. OSADA, and H. KOJIMA, "Fixation of Krypton in Type-A Zeolites and Its Leakage Characteristics." *Zeolites* 4(3):291-4. 10.1016/0144-2449(84)90041-1, (1984).

10. R. D. PENZHORN, R. D., P. SCHUSTER, H. LEITZIG, and H. E. NOPPEL, "Noble Gas Immobilization in Zeolites." *Berichte der Bunsen-Gesellschaft-Physical Chemistry Chemical Physics* 86(11):1077-82. 10.1002/bbpc.198200024, (1982).

11. W. J. MAECK, and D.T . PENCE, "Application of the Metal Zeolites to Radioactive Air Cleaning Problems." In *Proceedings of the Eleventh AEC Air Cleaning Conference* (CONF-700816), 607-18 pp. The Harvard Air Cleaning Conference, Cambridge, Mass., (1970).

12. N. SOELBERG, M. ABBOTT, D. R. HAEFNER, and R. T. JUBIN. "Gaseous Fission Product Emissions Control During Spent Nuclear Fuel Recycling." Presented at 235th American Chemical Society, American Chemical Society, Washington, D.C., (2008).

13. U. MUELLER, M. SCHUBERT, F. TEICH, H. PUETTER, K. SCHIERLE-ARNDT, and J. PASTRE, "Metal-Organic Frameworks - Prospective Industrial Applications." *Journal of Materials Chemistry* 16(7):626-36, (2006).

14. A. B. CHRISTENSEN, J. A. DEL DEBBIO, D. A. KNECHT, and J. E. TANNER, "Loading and Leakage of Krypton Immobilized in Zeolites and Glass," *Scientific Basis for Nuclear Waste Management*, v 3, p 251-258, (1981).

15. R. W. BENEDICT, A.B. CHRISTENSEN, J.A. DEL DEBBIO, J.H. KELLER, and D.A. KNECHT, *Technical and Economic Feasibility of Zeolite Encapsulation for Krypton-85 Storage* (Exxon Nuclear Idaho Co., Inc., Idaho Falls.), Sponsor: US Department of Energy, 96 p, (Sept. 1979).

16. Personal Communications, Deiter Knecht, Oct. 2010.

17. R. T. JUBIN, and S.H. BRUFFEY, "Analysis of Legacy [85]Kr Waste Form Samples – 16319," WM2016 Conference, Phoenix, Arizona, (2016).

18. T. GARN, M. GREENHALGH, and J. LAW, *Strategy for Analysis of Legacy Kr-85 Samples*, FCRD-SEPA-2011-000027, INL/MIS-10-20449, (Dec. 23, 2010).

19. N. A. MARKS, D. J. CARTER, M. SASSI, A. L. ROHL, K. E. SICKAFUS, B. P. UBERUAGA, and C.R. STANEK, "Chemical Evolution Via Beta Decay: A Case Study in Strontium-90." *Journal of Physics - Condensed Matter* 25(6):065504/1-04/15. 10.1088/0953-8984/25/6/065504 (2013).

20. T. R. PINCHBACK, J. R. WINKEL, D. K. MATLOCK, and D.L. OLSON, "Corrosion of Structural Alloys in Liquid Rubidium." *Nuclear Technology* 54(2):201-07, (1981).

21. R. VON AMMON, W. BUMILLER, E. HAUSS, E. HUTTER, G. KNITTEL, C. MAS, and G. NEFFE, "Considerations on Cryogenic Krypton Separation with or without Preceding Oxygen Removal," Complementary Paper #3 in Hebel, W. and Cottone, G., editors, Leudet, A. et al., "Methods of Krypton-85 Management," Proceedings of a Meeting Organized by the Commission of the European Communities and Held in Brussels, 29 June 1982 (February 1983).

22. D. K. LITTLE, et al. "Noble Gas Removal and Concentration by Combining Fluorocarbon Absorption and Adsorption Techniques," *Proceedings of the 17th DOE Nuclear Air Cleaning Conference*, CONF-820833, pg. 694-717, (1983).

23. J. LIU, P. K. THALLAPALLY, and D. STRACHAN, "Metal-Organic Frameworks for Removal of Xe and Kr from Nuclear Fuel Reprocessing Plants." Langmuir 28(31):11584-89. 10.1021/la301870n, (2012).

24. J. LIU, C. A. FERNANDEZ, P. F. MARTIN, P. K. THALLAPALLY, and D. M. STRACHAN, "A Two-Column Method for the Separation of Kr and Xe from Process Off-Gases." Ind. Eng. Chem. Res. 53(32):12893-99. 10.1021/ie502156h, (2014).

25. Y. PONTILLON, G. DUCROS, and P. P. MALGOUYRES, "Behaviour of Fission Products under Severe PWR Accident Conditions VERCORS Experimental Programme-Part 1: General Description of the Programme." *Nuclear Engineering and Design* 240(7):1843-52. 10.1016/j.nucengdes.2009.06.028, (2010).

26. H. U. BORGSTEDT, H.E. NOPPEL, and R. D. PENZHORN, *Compatibility of Materials for Pressurized Cylinders with Unpurified Rubidium*. Report No. KfK-3717, Kernforschungszentrum Karlsruhe, Karlsruhe, Germany, (1984).

27. R. D. PENZHORN, *Alternative Processes for the Long-Term Storage of Krypton-85*. KFK-2482, Kernforschungszent. Karlsruhe, Karlsruhe, Germany, (1977).

28. O. ASAI, "Corrosion of Steel in Water at Elevated Temperature and Pressure. VI. Corrosion of Mild Steel in High-Temperature Solutions Containing Two Kinds of Alkali Hydroxides." *Denki Kagaku* 35(12):857-61. (1967).

29. O. ASAI and N. KAWASHIMA, "Corrosion of Steel in Water at Elevated Temperature and Pressure. II. Corrosion of Mild Steel in Aqueous Solutions of Potassium Hydroxide, Rubidium Hydroxide, and Cesium Hydroxide." *Denki Kagaku* 34(3):201-5 (1966).

30. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Aluminum-Rubidium System," *Solubility Data Series 64 (Metals in Liquid Alkali Metals, Pt. 2)*:158 (1996).

31. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Bismuth-Rubidium System." *Solubility Data Series 64 (Metals in Liquid Alkali Metals, Pt. 2)*:327-30, (1996).

32. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Chromium-Rubidium System." *Solubility Data Series 63 (Metals in Liquid Alkali Metals, Pt. 1)*:195-96, (1996).

33. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Iron-Rubidium System." *Solubility Data Series 63 (Metals in Liquid Alkali Metals, Pt. 1)*:289-90, (1996).

34. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Lead-Rubidium System." *Solubility Data Series 64 (Metals in Liquid Alkali Metals, Pt. 2)*:290-92, (1996).

35. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Molybdenum-Rubidium System." *Solubility Data Series 63 (Metals in Liquid Alkali Metals, Pt. 1)*:218-19, (1996).

36. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Nickel-Rubidium System." *Solubility Data Series 64 (Metals in Liquid Alkali Metals, Pt. 2)*:45, (1996).

37. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Silver-Rubidium System." *Solubility Data Series 64 (Metals in Liquid Alkali Metals, Pt. 2)*:81, (1996).

38. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Tantalum-Rubidium System." *Solubility Data Series 63 (Metals in Liquid Alkali Metals, Pt. 1)*:175-76, (1996).

38. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Tin-Rubidium System." *Solubility Data Series 64 (Metals in Liquid Alkali Metals, Pt. 2)*:253-54, (1996).

40. H. U. BORGSTEDT and C. GUMINSKI, "Solubility Data. Zirconium-Rubidium System." *Solubility Data Series 63 (Metals in Liquid Alkali Metals, Pt. 1)*:112-13, (1996).

41. T. R. PINCHBACK, and D. A. KNECHT, *Development of Process and Storage Materials Suitable for Krypton-85 Waste Management.* CONF-7811213, EG&G Idaho, Inc., Idaho Falls, Idaho, (1978).

42. G. L. TINGEY, E. D. MCCLANAHAN, M. A. BAYNE, and W. J. GRAY, "Solid State Containment of Noble Gases in Sputter Deposited Metals and Low Density Glasses," IAEA Symposium on Management of Gasesous Wastes from Nuclear Facilties, Vienna, IAEA-SM-245/31, (1980).

43. W. JIANG, R. M. V. GINHOVEN, L. KOVARIK, J. E. JAFFE, and B. W. AREY, "Superlattice Structure and Precipitates in O^+ and Zr^+ Ion Coimplanted $SrTiO_3$: A Model Waste Form for ^{90}Sr." *Journal of Physical Chemistry C* 116:16709−15, (2012).

44. W. JIANG, L. KOVARIK, Z. ZHU, T. VARGA, M. H. ENGELHARD, M.E . BOWDEN, T. M. NENOFF, and T. J. GARINO, "Microstructure and Cs Behavior of Ba-Doped Aluminosilicate Pollucite Irradiated with F+ Ions." *J. Phys. Chem. C* 118(31):18160-69. 10.1021/jp5045223, (2014).

45. R. M. VAN GINHOVEN, J. E. JAFFE, W. JIANG, and D. M. STRACHAN, *Chemical and Charge Imbalance Induced by Radionuclide Decay: Effects on Waste Form Structure.* PNNL-20049, Pacific Northwest National Laboratory, Richland, Wash (2010).

46. R. M. VAN GINHOVEN, J. E. JAFFE, W. JIANG, and D. M. STRACHAN. *Chemical and Charge Imbalance Induced by Radionuclide Decay: Effects on Waste Form Structure.* PNNL-20312, Pacific Northwest National Laboratory, Richland, Wash, (2011).

47. R. T. MORRISON, and R.N. BOYD, *Organic Chemistry. 6th ed.*, Prentice-Hall, Inc., Englewood Cliffs, N.J., (1992).

48. S. K. GHOSE, Y. LI, A. YAKOVENKO, E. DOORYHEE, L. EHM, L. E. ECKER, A-C DIPPEL, G. J. HALDER, D. M. STRACHAN, and P. K. THALLAPALLY. 2015. "Understanding the Adsorption Mechanism of Xe and Kr in a Metal−Organic Framework from X-Ray Structural Analysis and First-Principles Calculations." *Journal of Physical Chemistry Letters* 6:1790-94, (2015).

49. C. DANIEL, A. ELBARAOUI, S. AGUADO, M-A SPRINGUEL-HUET, A. NOSSOV, J-P FONTAINE, S. TOPIN, T. TAFFARY, L. DELIERE, Y. SCHUURMAN, and D. FARRUSSENG, "Xenon Capture on Silver-Loaded Zeolites: Characterization of Very Strong Adsorption Sites." *Journal of Physical Chemistry C* 117(29):15122-29. 10.1021/jp403934r, (2013).

ACKNOWLEDGMENTS