ORNL/LTR-2022/419

Oak Ridge National Laboratory: Quantify Contaminant Partitioning in Advanced Chlorination Process

Joanna McFarlane
Emory Collins
Jacob Hirschhorn
Craig Barnes
Breanna Vestal

June 2022

ORNL IS MANAGED BY UT-BATTELLE LLC FOR THE US DEPARTMENT OF ENERGY

DOCUMENT AVAILABILITY

Reports produced after January 1, 1996, are generally available free via OSTI.GOV.

Website www.osti.gov

Reports produced before January 1, 1996, may be purchased by members of the public from the following source:

National Technical Information Service
5285 Port Royal Road
Springfield, VA 22161
Telephone 703-605-6000 (1-800-553-6847)
TDD 703-487-4639
Fax 703-605-6900
E-mail info@ntis.gov
Website http://classic.ntis.gov/

Reports are available to US Department of Energy (DOE) employees, DOE contractors, Energy Technology Data Exchange representatives, and International Nuclear Information System representatives from the following source:

Office of Scientific and Technical Information
PO Box 62
Oak Ridge, TN 37831
Telephone 865-576-8401
Fax 865-576-5728
E-mail reports@osti.gov
Website https://www.osti.gov/

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

ORNL/LTR-2022/419

Nuclear Energy and Fuel Cycle Division

QUANTIFY CONTAMINANT PARTITIONING IN ADVANCED CHLORINATION PROCESS

Joanna McFarlane
Emory Collins
Jacob Hirschhorn
Craig Barnes
Breanna Vestal

June 2022

Prepared by
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, TN 37831
managed by
UT-BATTELLE LLC
for the
US DEPARTMENT OF ENERGY
under contract DE-AC05-00OR22725

ABSTRACT

Chlorination of the zirconium in spent fuel cladding has been identified as a significant means of Zr recovery and purification, enabling a 25% (wt.) or greater reduction in waste requiring geologic disposition. Several chlorinating agents have been identified over the last few years, including gases and liquids. In particular, the interactions of sulfur-chloride solvents with zirconium may allow the separation of zirconium from uranium with the selective formation of $ZrCl_4$. This report reviews what is known about reactions between chlorinating agents, uranium metal and oxide, Zircaloy and its alloying elements, and fission plus activation products. Plans for testing the feasibility of applying advanced chlorination methods developed for zirconium recycling to the front end of the recycling of spent nuclear fuel are also included.

1. INTRODUCTION

The proposed advanced chlorination work scope is to test a decladding process using sulfur-chloride reagents. This report briefly reviews work performed under the auspices of a Nuclear Energy University Program (NEUP) awarded to the University of Tennessee in which the seminal work on zirconium and zircalloy dissolution was carried out. The success of the NEUP project indicates that the chemical process is being considered for chemical decladding of metal-matrixed fuel as planned for material recovery for HALEU production. This fuel is different from the oxide fuels often considered for material recovery processing, such as voloxidation. Therefore, a brief synopsis of a metal-matrixed fuel is included in this introduction. The report continues with an outline of a possible flow sheet for Zircaloy removal and processing, including the parameters to be analyzed in the first round of tests at ORNL.

1.1 METAL-MATRIXED FUEL

Metal fuel has been considered for advanced reactor technology because it is relatively easy to fabricate, has a high density, and has a high thermal conductivity (Betzler et al. 2017). It can be used in research reactors such as the Experimental Breeder Reactors (EBRs) at Idaho National Laboratory (INL, operated by Argonne National Laboratory) (Stevenson 1987, Judd 2014). The fissile metal is often enriched to greater levels than for that used for light-water reactor (LWR) fuels such as the 48–65 wt.% ^{235}U used at EBR-II (Stevenson 1987). The fuel can be alloyed with Zr and noble metals that could include Mo, Pd, Ru, Rh, and niobium. The alloys impart superior structural strength, leading to a more stable geometry and a higher melting point. In the advanced chlorination process presented herein, a simfuel alloy of uranium with either 10 wt% zirconium or 10 wt.% molybdenum will be used. The properties of metal-matrixed fuel have been reviewed by Crawford et al. (2007).

Metal fuels samples for research can be fabricated by casting the alloy into a quartz mold, which is then clad. For instance, U-10 wt% Zr alloy pins were cast into a mold with a diameter of 0.5 cm and were then clad with HT-9 tubing in preparation for study after irradiation (Harp et al. 2017). The cladding can include Zircaloy, stainless steel, or aluminum. Studies have been undertaken with aluminum serving as a diffusion barrier between the uranium and the cladding. The cladding can interact with fission products, namely lanthanides, that escape from the fuel. The stability of the fuel matrix will also be affected by the presence of oxidants such as corium, or reductants such as B_4C (Fukasawa and Tamura 2007).

Processing of spent fuel requires an understanding of where the fission products are located in the matrix. Most fission products have a lower reactivity with oxidizers than uranium and will remain in the fuel alloy (Fukasawa et al. 2005). However, with metal alloy fuels, swelling caused by the accumulation of gaseous and semi-volatile fission products during burnup is a concern. Volatile fission products such as fission gases and their daughters escape the metal fuel to the fuel-cladding gap. Fission product analysis of irradiated HEU has identified Sr, Y, Cs, La, Ce, Pr, Nd, Pm, and Sm isotopes in small, dissolved samples to be in good agreement with ORIGEN predictions, except for cesium (Roach et al. 2019). The low values for cesium were attributed to the escape of the parent xenon isotopes. Fuel rods can be engineered to limit fuel-cladding mechanical interaction that might arise from swelling. Fuel rods that use metallic fuels frequently have larger plenum volumes to accommodate the fission gases released from the fuel. Therefore, these semi-volatile radionuclides will be easily accessible during decladding.

These studies will analyze the effect of chlorinating agents on the fuel, the cladding, and fission products.

1.2 CHLORINATION OF ZIRCONIUM AND FISSION PRODUCTS

The goal of chemical decladding is to replace the dusty, hazardous mechanical processes typically used for cladding removal. Recycling of zirconium from spent fuel has been investigated in a study by Collins et al. (2013) in which three feedstock forms, empty hulls from nitric acid leaching, empty hulls from voloxidation of the fuel, and chemical decladding of Zr-clad spent nuclear fuel were compared. In the case of chemical decladding, conditions are kept sufficiently mild to remove the zirconium and leave the spent fuel intact. Gases that have been tested for this application include iodine (Collins et al. 2012), chlorine (Collins et al. 2012, 2017) and HCl. (Collins et al. 2012, Del Cul and Hunt 2019, McFarlane et al. 2020, 2021). Although aluminum dissolution in caustic-nitrate proceeds well at 100°C, aluminum chlorination was investigated in later tests with HCl(g). For aluminum, the gaseous chlorination reaction with HCl occurs at 200°C, well below any involvement of the fuel (McFarlane et al. 2020, 2021). Gasous zirconium chlorination requires a higher temperature of 350°C for HCl to react to form $ZrCl_4$. Uranium metal is less inert at this higher temperature and may form some UCl_4 in the hydrochlorinator.

1.3 ADVANCED CHLORINATION OF ZIRCONIUM

The Material Recovery and Waste Form Development campaign has a goal of reducing high-level waste from LWRs. Zircaloy comprises about 25% (wt) of the intermediate-level waste (ILW) from an LWR (Collins et al. 2012). Because it is a large portion of the waste stream, there has been research into the separation of Zircaloy from the fuel pellet and purification by removal of fission and activation products. Elements of concern in various Zircaloy alloys include Sn, Nb, Fe, Cr, and Ni (Collins et al. 2017). Fission product Sb and Cs also form volatile chlorides when the gaseous reagents discussed in Section 1.2 are used: HCl(g) and Cl_2(g).

Therefore, the University of Tennessee, Knoxville (UTK) has been testing low-temperature, 95°C, solution-phase processes using $SOCl_2$ and S_2Cl_2 solvents for the chlorination of zirconium alloys. They have achieved success in targeted production of $ZrCl_4$ with little chlorination of other metals in the same solution (Vestal et al. 2021, Vestal et al. 2020a, Vestal et al. 2020b)

Preliminary studies with $SOCl_2$ and S_2Cl_2 were undertaken at ORNL and were reported by Bruffey and colleagues in 2021 (Bruffey et al. 2021). At ORNL, two sets of experiments were performed. One test exposed a mixture of contaminants to the chlorinating solvents along with the zirconium. The additional metals included Fe, Cr, Sn, and Nb metals, as well as CsCl. The reaction time under reflux was 30 min. Volatile $SnCl_4$ was captured in a NaOH(aq) trap on the reaction vessel. Sulfur was the byproduct of the chlorination reaction. The purity of the zirconium went to 94%, with the main contaminants being Fe and Cs, likely in chloride form. For the purposes of reducing high-level waste, the focus will be to remove those isotopes that increase the radiological hazard of the spent zirconium over the long term.

The other test involved exposure of UO_2 and selected fission product oxides—Eu_2O_3, SrO, and Cs_2O—to a mixture of S_2Cl_2 and $SOCl_2$ for 5 h. No reaction was observed with the metal oxides.

The focus of these studies at UTK and at ORNL were to produce purified zirconium and not to target the decladding process discussed in Section 1.2. In the follow-on work, the objective is to apply these or similar processes to U-Zr or U-Mo fuel, with the ultimate goal of separating and recovering the uranium.

2. ADVANCED CHLORINATION FOR REMOVAL OF METAL MATRIX FROM URANIUM

The Zircex process uses a dry chlorination process for separating zirconium or aluminum from spent fuel (Tripp et al. 2017). The work to be conducted in this current project will be to evaluate whether the decladding process that currently uses HCl(g) could be replaced with a sulfur-chloride liquid solvent under reflux conditions. A flow sheet was developed for the chlorination of zirconium (Figure 1) based on the process developed at UTK. The separation of uranium and the implementation of advanced chlorination as a dry decladding process is presented in Figure 2. Note that the recovery of purified $ZrCl_4$ is still possible as shown in the second flow sheet, but the unit operations have been removed to simplify the diagram. Other cladding materials may be considered, such as aluminum or stainless steel. Preliminary experiments conducted at UTK have shown that the kinetics are much more rapid with aluminum than zirconium. Therefore, the reagent S_2Cl_2 may be diluted in $SOCl_2$ to better control the reaction to remove aluminum cladding and aluminum-cladding liners / diffusion barriers. Process scale up requires control of the dissolution kinetics.

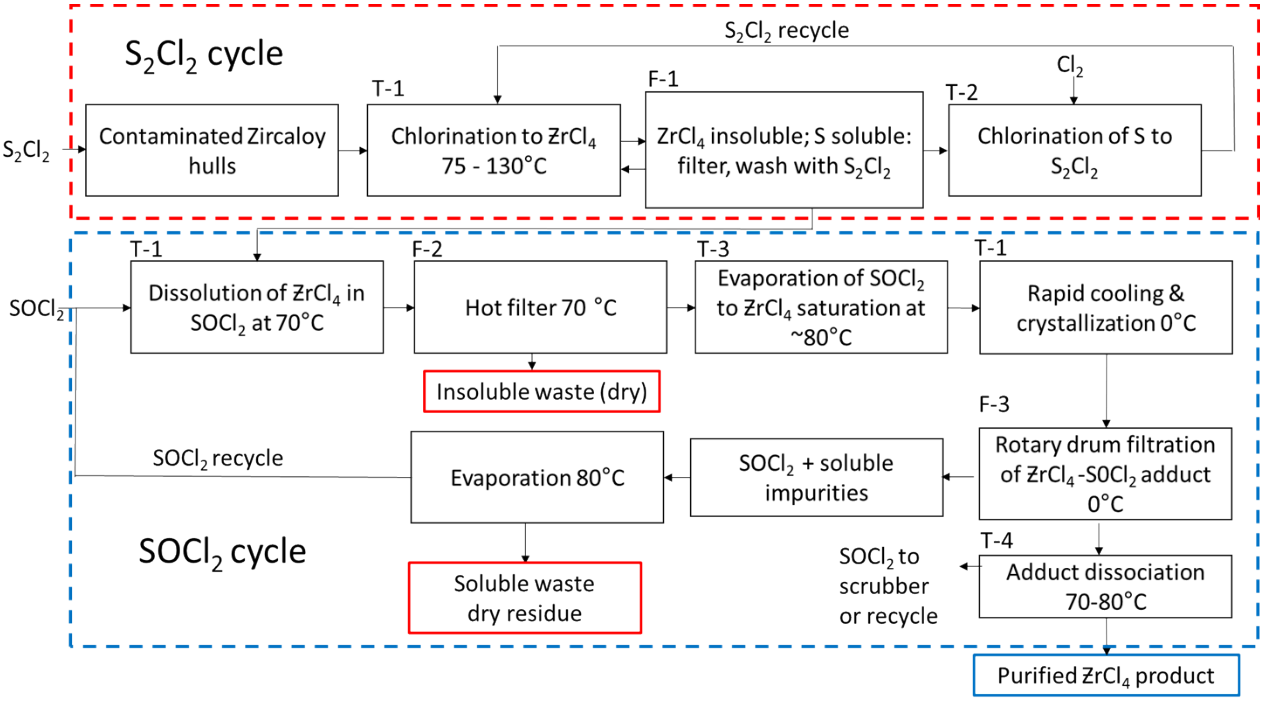

Figure 1. Flow sheet for the purification of zirconium using the advanced chlorination process.

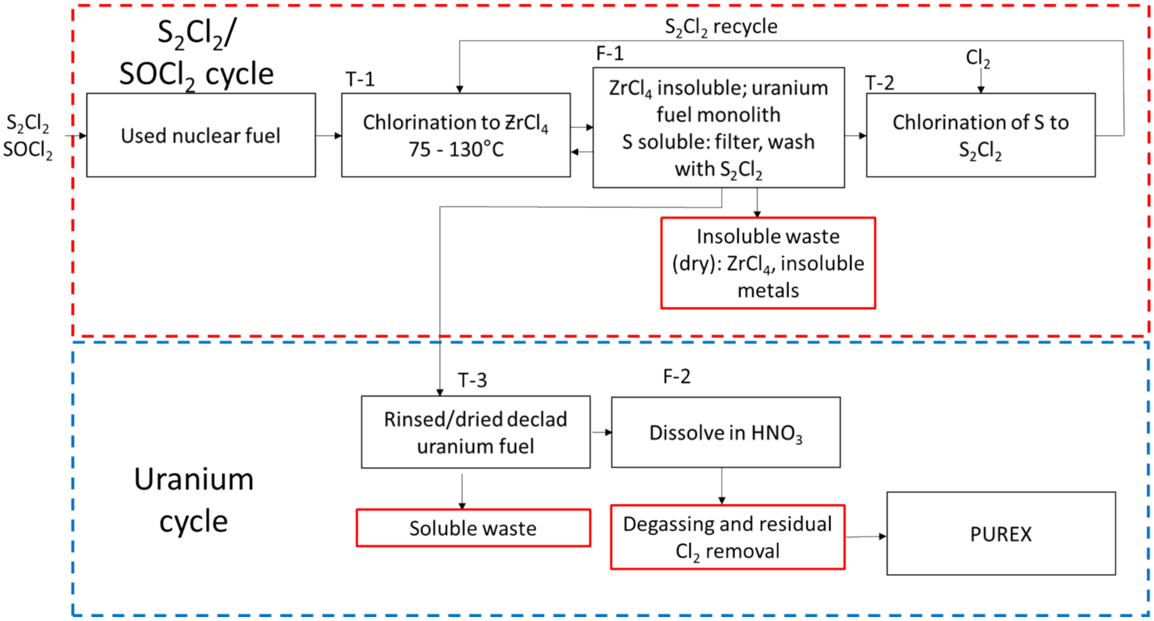

Figure 2. Advanced chlorination for decladding of used nuclear fuel. If Zr recovery is desired, then the $SOCl_2$ process from Figure 1 must be added.

The planned experiments will investigate the form of the uranium after the zirconium chlorination step. If the uranium does not react and remains as an intact monolith, then separation from the fine $ZrCl_4$ powder should be relatively easy. If the spent fuel is introduced in a porous basket, the larger uranium chunks can be retained in the basket while the $ZrCl_4$ powder is rinsed away. If the uranium as an alloy forms a crumbled mixture, then an additional separation step will be required, likely involving an additional step involving the dissolution of $ZrCl_4$ in $SOCl_2$ and filtration of the uranium particulates.

2.1 TRACKING CONTAMINANTS

A key aspect to a flow sheet's feasibility is to understand where the minor species end up and in which product or waste stream. These studies have been performed for many of the chlorination processes mentioned earlier including the research published by Collins et al. (2017) and documented in McFarlane and Walker (2021). The difference in this case will be the starting material, namely spent uranium fuel. The spent fuel will be simulated in a cast alloyed form.

2.1.1 Alloying Elements

The major contaminants to be tracked through the chlorination process are the alloying elements. Preliminary results from hydrochlorination with spent nuclear fuel showed that the $ZrCl_4$ produced from hydrochlorination contained Sn, Cr, and Fe, as well as fission products (Tripp et al. 2017), thus corroborating earlier ORNL studies (Collins et al. 2012). Collins and colleagues observed that tin as $SnCl_4$ was carried with the $ZrCl_4$ through the chlorination with Cl_2 at 200°C. About 30% of the $SnCl_4$ was condensed along with the $ZrCl_4$, necessitating a resublimation to remove the $SnCl_4$. Other alloying elements—Nb and Co—were reduced to a very low concentration during the process. Fe, Ni, and Cr from Zircaloy-2 were also greatly reduced.

2.1.2 Sulfur and Chlorine

The flow sheets in Figures 1 and 2 show that the UTK researchers have considered how to contain sulfur and chlorine. In Figure 1, the solvents used in the chlorination process are separated and cleaned for reuse. Sulfur generated during the initial process is soluble in the solvent S_2Cl_2 and can be re-chlorinated to S_2Cl_2. These concepts will be incorporated into the flow sheet for uranium recovery. The uranium solution must be chlorine free after undergoing the downstream nitric acid dissolution before the PUREX process. Therefore, recovery of sulfur and solvents and the removal and control of chlorine will be important aspects to study in the next round of experiments.

2.1.3 Uranium

The ultimate goal of this project is to test the advanced chlorination process as a route to front-end processing of metal-clad metal fuel. However, there may also be interest in removing cladding from oxide fuels. As mentioned in Section 1.3, the advanced chlorination process was tested in proximity to UO_2, which was introduced into the reactor as a Zircaloy-clad rodlet. The solvent tested for this process was $SOCl_2$. As in previous studies, the Zircaloy readily reacted to form $ZrCl_4$ and went into solution as an adduct $ZrCl_4 \cdot SOCl_2$. The UO_2 appeared to be untouched during the processing. In addition, recent studies involving uranium metal and low-temperature hydrochlorination also saw little reaction with the metal to produce UCl_4 (McFarlane et al. 2021). There did appear to be a small amount of uranium relocation, perhaps associated with iron chloride or aluminum chloride volatility. The extent to which uranium metal alloy is inert to chlorination will be an important test of the feasibility of zirconium/Zircaloy removal from spent fuel.

2.1.4 Fission Products

The chlorination of fission products associated with Zircaloy cladding has been studied at INL (Tripp et al. 2017), as well as ORNL (Collins et al. 2012, McFarlane and Walker 2021). The results from Tripp are more representative of Zircex, because actual samples of spent fuel were investigated in the hydrochlorination process, but they do not include an analysis of fission product transport except for tritium. The ORNL experiments used spent UO_2 (Collins et al. 2012) or fission product chlorides in contact with zirconium and aluminum metals (McFarlane and Walker, 2021).

In the work reported by Collins and coworkers (2017), they tracked the radioisotopes ^{241}Am, ^{244}Cm, ^{137}Cs, ^{94}Nb, ^{125}Sb, ^{60}Co, ^{154}Eu. In particular, the cesium was present at a high level in the spent fuel cladding. The chlorination process reduced the activity level by three orders of magnitude, with the off-gas being passed through a cesium trap, but some residual activity remained, primarily from ^{137}Cs.

Recent testing at ORNL (McFarlane and Walker 2021) looked specifically at the formation and transport of fission product chlorides under representative flows of HCl(g) at 200°C for aluminum chlorination and at 350°C for zirconium chlorination. The results indicate that enhanced volatility was observed for Sr, Sn, Mo, and Te in the presence of the cladding materials. Iodine appeared to transport with the tin to the off-gas scrubber.

The literature suggests that much fewer fission products are associated with the cladding than the fuel. Even so, chlorination of the cladding indicates that certain volatile and semi-volatile fission products will be released, even if the fuel alloy remains intact. A detailed assessment of radioactivity transport will require an understanding of the degree to which the fuel alloy will degrade with the chlorination process.

3. FEASIBILITY STUDY

Several questions must be addressed in the feasibility study in order to develop the draft flowsheet presented in Section 2. These questions can be divided into topics that relate to engineering and process flow and to the underlying science and disposition of fission products. Given the relatively short time available, the focus will be on engineering the process and testing the feasibility on a bench-scale level. If these results show promise, then a more in-depth program can be conducted into the science of how the spent fuel will behave in an advanced chlorination process.

The engineering questions to be addressed include tracking the mass balance for sulfur species and volatile chlorides, potentially including $SnCl_4$. For recovery of U, residual S and Cl must be removed after dissolution in HNO_3. Capture of stannic chloride requires an off-gas system. The studies conducted at UTK suggest that the chlorination process for Zr is more effective with S_2Cl_2, than with $SOCl_2$. However, a secondary dissolution in $SOCl_2$ is required for the separation. The use of S_2Cl_2 generated solid sulfur rather than SO_2, but SO_2 will be generated in downstream processing. For aluminum clad fuel, the reaction with S_2Cl_2 is too vigorous, so dilution with $SOCl_2$ may be required to slow the kinetics.

More fundamental questions are outlined below.

Oxidation of the uranium occurs in U-10 wt.% Zr fuels to form an oxide layer, even after brief exposure to air. Tripp et al. (2017) noted an induction period as the hydrochlorination reagent penetrated the ZrO_2 layer. Results from UTK also suggest that solution-phase chlorination is also a surface-mediated process for Zircaloy. If surface oxidation of U metal to UCl_4 occurs within advanced solvents, then the flow sheet must be modified to separate the surface layer UCl_4 from the $ZrCl_4$ Although surface layers may help limit interdiffusion and fuel-cladding chemical interaction, they may not be a reliable means of doing so because of cracks and gaps in these layers.

Experiments with complex alloys and fuel matrices will require careful consideration for understanding the chlorination process. During burnup, Zr migrates from the intermediate radii to the central zone and uranium moves in the other direction. Phase separation and formation of UZr_2 was observed. These tests at INL showed that migration of fission products through the uranium-alloy occurred, with different inclusions corresponding to lighter elements or heavier elements (Harp et al. 2017). The cross-sectioned pellet showed behavior with a strong radial dependence. On the other hand, fuel-cladding chemical interactions (FCCIs) have been observed even for lightly irradiated (1–2% FIMA) fuels, U-10 wt.% Zr, in HT-9 (Harp et al. 2017). Lanthanide elements migrated to austenitic grain boundaries in the cladding and in some cases were found to be associated with noble metal particles.

Porosity structure, porosity interconnection, and infiltration of liquid bone-sodium into the interconnected pore and cracks (particularly near the periphery of the fuel) may have significant impacts on the chlorination process and reactivity of the alloying element to the chlorinating agent. The reactivity of the fission products associated with the cladding and the retention of the fission products in the residue will be dependent on the chemistry and phase behavior of each element. It is not clear whether these phenomena can be replicated with simfuel, so it is likely that spent fuel experiments will be required. A decision will be made on continuation with spent fuel, following feasibility studies with unirradiated simulants.

3.1.1 Test Matrix for Feasibility Studies

This research will focus on the separation of the zirconium as $ZrCl_4$ from uranium metal. Key operations specified in the flow sheet in Figure 2 will be tested at the bench-scale to determine the pathways for zirconium, uranium, sulfur, chlorine, and major fission product groups. It may be difficult to generate a metal simfuel with representative fission products; however, preparation and testing of a U-10Zr or U-10Mo alloy is possible. Testing with legacy materials from the EBR-II program will provide a more realistic assessment, but this will only be implemented if the preliminary experiments show feasibility. The test matrix for advanced chlorination with unirradiated materials is given in Table 1. Feasibility testing will be conducted over a period of 6 months, likely from July through December in 2022. Irradiation experiments will be planned for later.

4. CONCLUSIONS

Advanced chlorination will be tested as an alternative chemical decladding process for spent metal-clad metal-alloy nuclear fuel. Advanced chlorination using solvents based on sulfur and chlorine has been shown to effectively remove zirconium from alloying elements. These studies were performed to assess the potential for recycling zirconium from Zircaloy to reduce waste volumes. Although this objective is still of interest, the focus of this study will extend to the separation of alloying elements and fission products with uranium as the targeted product. A test matrix has been developed to demonstrate key decontamination processes for alloying elements and fission products.

Table 1. Test matrix for testing the feasibility of advanced chlorination in removal of zirconium cladding from uranium metal

Project task	Purpose	Scheduling
1. Prepare depleted uranium alloy for testing, cast into a Zircaloy tube	This approach will best represent major components of metal-clad metal fuel. Pre-test characterization of constituent redistribution, phase structure and porosity will be performed.	Months 1 and 2
2. Design and construct an advanced chlorination apparatus	The apparatus will be made of glass to allow the process to be monitored. Other sensors will include temperature and pressure. The off-gas will be collected in a water trap and analyzed.	Months 1 and 2
3. Testing of apparatus with Zircaloy 4	Analysis will determine if the tin alloying element in Zircaloy-4 is volatile as a tin chloride; sulfur species will also be tracked.	Month 3
4. Chlorination process with uranium alloy material	The susceptibility of uranium as a metal and as an alloy will be determined. Post-test characterization of constituent redistribution, phase structure and porosity will be performed.	Month 4
5. Chlorination process with uranium alloy cast into a Zircaloy-4 tube	The kinetics of decladding uranium alloy fuel will be followed.	Month 5
6. Chlorination process with added representative fission products	Materials will be analyzed before and after the chlorination to perform a mass balance on important fission products.	Month 6
7. Revision of flow sheet and assessment of feasibility	If feasible, then (a) an engineering scale-up will be implemented, and (b) the science questions will be addressed in FY23/FY24.	Month 7

5. ACKNOWLEDGMENTS

This program was funded by the US Department of Energy (DOE) Office of Nuclear Energy Nuclear Technology Research and Development Program, which is managed at ORNL by L. Martin. This manuscript was authored by UT-Battelle LLC under contract DE-AC05-00OR22725 with DOE.

6. REFERENCES

Barnes, C., Orick, M., Styskalik, A., Del Cul, G. B. D., McLaughlin, D. F. 2017. “Purification of Zirconium Tetrachloride ($ZrCl_4$) from UNF Cladding - A Progress Report.” Annual Meeting of the American Nuclear Society, San Francisco, 11–15 June 2017, *Trans. Am. Nucl. Soc*., 116.

Braase, L., May, W. E., Todd, T. 2014. *Material Recovery and Waste Form Development*, FCRD-MRWFD-2015-00004, INL/EXT-14-33548.

Betzler, B.R., Chandler, D., Davison, E.., Ilas, G. 2017. “High-Fidelity Modeling and Simulation for a High Flux Isotope Reactor Low-Enriched Uranium Core Design, *Nucl. Sci. Eng.* 187(1), 81–99.

Bruffey, S. H., Hunt, R. D., Vestal, B. K., Barnes, C. E. 2021. *Advanced Low Temperature Chlorination of Zirconium*, ORNL/SPR-2021/2003.

Crawford, D. C., Porter, D. L., Hayes, S. L. 2007. “Fuels for Sodium-Cooled Fast Reactors: US Perspective,” *J. Nucl. Mater.* 371, 202–231.

Collins, E. D., Del Cul, G. D., Spencer, B. B., Brunson, R. R., Johnson, J. A. 2013. “Establishment of the Roadmap for Chlorination Process Development for Zirconium Recovery and Recycle,” *Proceedings of Global 2013 Conference, American Nuclear Society*, October 2013.

Collins, E. D., Spencer, B. B., DelCul, G.D., Brunson, R.R., Johnson, J.A. 2012. *Road Map for Zirconium Recovery/Recycle Process Development*, ORNL, FCRD-SWF-2013-000074.

Collins, E. D., Hylton, T. D., DelCul, G. D., Spencer, B. B., Hunt, R. D., Johnson, J. A. 2017. “Completion of a Chlorination Test Using 250 Grams of High-Burnup Used Fuel Cladding from a North Anna Pressurized Water Reactor,” ORNL, ORNL/LTR-2017/327.

Collins, E. D., Del Cul, G. D., Spencer, B. B., Brunson, R. R., Johnson, J. A., Terekhov, D. S., Emmanuel, N. V. 2012. “Process Development Studies for Zirconium Recovery/Recycle from Used Nuclear Fuel,” *Procedia Chemistry* 7, 72–76.

Del Cul, G. D, Hunt, R. D. 2019. “Chlorination Scoping Tests on Aluminum 6061 Coupons,” ORNL/SPR-2019/1252.

Fukasawa, M., Tamura, S. 2007. “Thermodynamic Analysis for Molten Corium Stratification Test MASCA with Ionic Liquid U-Zr-Fe-O-B-C-FPs Database.” *J. Nucl. Sci. Technol.* 44(9), 1210–1219.

Fukasawa, M., Tamura, S., Hasebe, M. 2005. *J. Nucl. Sci. Technol.* 42(8), 706-716. DOI: 10.1080/18811248.2004.9726440

Harp, J. M., Porter, D. L., Miller, B. D., Trowbridge, T. L., Carmack, W. L. 2017, Scanning Electron Microscopy Examination of a Fast Flux Test Facility Irradiated U-10Zr Fuel Cross Section Clad with HT-9,” *J. Nucl. Mater.* 494, 227–239.

Judd, A. M. 2014. *An Introduction to the Engineering of Fast Nuclear Reactors*, Cambridge University Press, New York, NY.

McFarlane J., Walker, T.W. 2021. *Fission Product Hydrochlorination*, ORNL/TM-2021/2312.

McFarlane, J., Greaney, A. T., Hunt, R. D., Del Cul, G. D., Braatz, A. D., 2020. *Complete Al Chlorination Lab-Scale Testing with Non-Fuel Materials*, ORNL/SPR-2020/1551.

McFarlane, J., Braatz, A. D., Birdwell, J. F., Jr., Bruffey, S. H., Del Cul, G. D., Greaney, A. T., Hunt, R. D., Ngelale, R. O., Taylor, P. A., Walker, T. B. 2021. "Hydrochlorination at 200°C Relevant to the Removal of Cladding from Spent Nuclear Fuel," *Trans Am. Nucl. Soc*. June 2021, 124(1), 100–102.

Roach, B. D., Fenske, E. K., Glasgow, D. C., Partridge, J. D., Keever, T. J., Giaquinto, J. M. "Rapid Concentration and Isotopic Measurements of Ultra-Trace ^{235}U Fission Products with Comparison to an ORIGEN Isotope Depletion Model." *Talanta* 205, 120079.

Stevenson, C.E. 1987. *The EBR-II Fuel Cycle Story*, American Nuclear Society, La Grange Park, IL.

Tripp, J., Demmer, R., Tillotson, R., Olson, L., Warner, M. 2017. *CFA/RAL ZIRCEX Test Report*, INL/LTD-16-40322.

Vestal, B., Barnes, C., DelCul, G.D. 2021. "Recycling Spent Nuclear Fuel Rods Using Sulfur Chloride Reagents," American Nuclear Society Annual Meeting, Virtual.

Vestal, B. K., Barnes, C. E., DelCul, G. D. 2020a. *Solubilities of Metal Chlorides in* $SOCl_2$ *and* S_2Cl_2 *to Determine Recrystallization Conditions for Purifying Metal Chlorides*, NEUP (DE-NE00087) Milestone Report: 87M2NU-18-TN-UTK-020701-023, 2020.

Vestal, B. K., Barnes, C. E., DelCul, G. D. 2020b. *Reactions of the Components of Spent Fuel Rods with Sulfur–Chloride Reagents*, NEUP (DE-NE00087) Milestone Report: M2NU-18-TN-UTK-020701-023, 2020.