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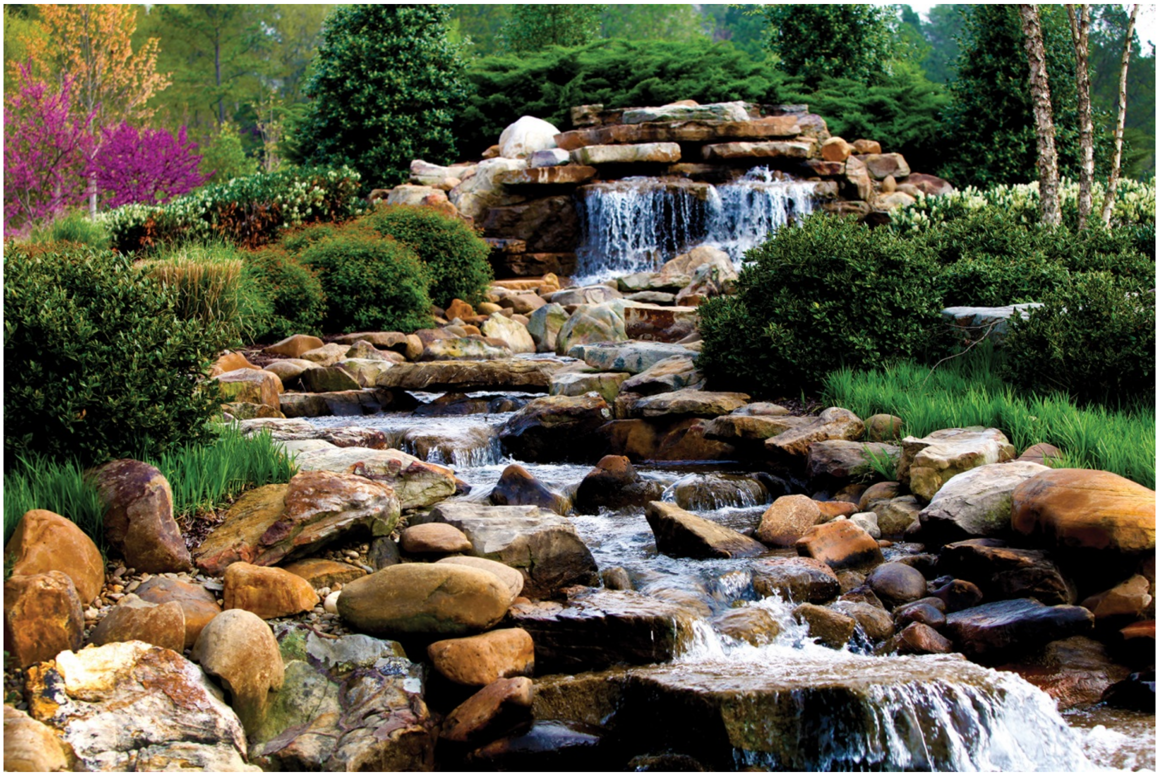
Advanced Low-Temperature Chlorination of Zirconium



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Nuclear Energy and Fuel Cycle Division

ADVANCED LOW-TEMPERATURE CHLORINATION OF ZIRCONIUM

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ABBREVIATIONS

HLW	high-level radioactive waste
ICP-MS	inductively coupled plasma mass spectrometry
LWR	light water reactor

SUMMARY

Recovery of Zr from nuclear fuel is of high interest because the Zr cladding material comprises up to 50% of the high-level radioactive waste (HLW) that requires disposition. An ideal Zr-recovery process would not only recover the bulk Zr from the fuel or cladding but would also provide decontamination from the components that give rise to the HLW designation. A simplified decontamination pathway providing recovered Zr in compliance with low-level radioactive waste acceptance criteria could significantly reduce the burden of HLW requiring geologic disposition.

A newly developed *advanced low-temperature chlorination* process presents an opportunity to recover a $ZrCl_4$ product that is effectively decontaminated from the elements of concern. This advanced low-temperature chlorination process uses a mixture of the sulfur-containing chlorination compounds sulfur monochloride (S_2Cl_2) and thionyl chloride ($SOCl_2$).

Development of the advanced low-temperature chlorination process has been focused on fundamental work that is necessary for understanding reaction mechanisms and reaction rates for pure metals and characterizing solubilities of the chlorinated products in $S_2Cl_2/SOCl_2$ mixtures. Building on that fundamental work, the next step is to understand how the advanced low-temperature chlorination process could partition particular contaminants in an irradiated fuel assembly. Toward that end, two proof-of-principle experiments were conducted. Scoping Experiment 1 chlorinated a mixture of Zircaloy-4 cladding segments and other elemental metals. Scoping Experiment 2 chlorinated a mixture of UO_3 and selected fission products representing simulated irradiated fuel. Together, these experiments were intended to demonstrate how the primary constituents of an irradiated spent fuel rod would behave in the advanced low-temperature chlorination process and to provide a preliminary assessment of $ZrCl_4$ product purity when using a more realistic and complex simulant.

Completing these two experiments yielded valuable findings related to the advanced low-temperature chlorination process. First, Scoping Experiment 2 confirmed the nonreactivity of uranium oxides within the process, indicating that advanced low-temperature chlorination could be used to separate Zr from solid feeds containing U. Second, Scoping Experiment 1 showed that a mixture of $SOCl_2/S_2Cl_2$ is a very effective and *selective* chlorination agent for quantitatively transforming the Zr in Zircaloy claddings into $ZrCl_4$, even in the presence of large amounts (>50,000 ppm) of radiological contaminants (stable isotope surrogates representative of those arising from irradiation). These experiments demonstrate the robustness of the advanced low-temperature chlorination process and indicate that a pure product may be easily obtained when using mixtures more representative of irradiated cladding and fuel.

These results indicate that the advanced low-temperature chlorination process holds promise for chemical decladding, hull processing, and $ZrCl_4$ purification processes and that it merits continued research and development. Suggestions for further research are provided.

1. INTRODUCTION

Recycling used nuclear fuel to recover U or other valuable materials must begin by exposing the fuel for chemical processing. Industrially, exposure is performed via mechanical bundle shearing, a process in which intact irradiated fuel assemblies are compressed and then segmented. In the case of light-water reactor (LWR) fuel, this process produces small open-ended cladding segments with at least one uranium oxide fuel pellet inside. The specific cladding material can vary, but it is generally a Zr-based alloy.

The shearing process takes place in the head end of the plant prior to separations. This process presents several disadvantages to operations, including being subject to periodic outages because of the mechanical nature of the process. When outages occur, remote maintenance is required because of the high radioactivity levels within the shear-containing hot cell. Shear process reliability can be an important factor in the total availability of the plant for operations because it is responsible for producing the initial feedstock. Beyond these reliability issues, the shearing process generates radioactive dust, embeds fuel particles into the cladding, and can result in crimped segment ends (reducing the surface area of the pellet for chemical reaction). This process also has a large shielded footprint compared to most other plant operations.

These disadvantages have prompted steady research and development on this head-end process through the decades, with most research and development centered on (1) improving the mechanical shear process by using concepts such as single-pin shearing or alternative blade designs and (2) developing alternative fuel exposure processes, such as chemical decladding. *Chemical decladding* refers to chemical processing of the irradiated fuel assembly to remove Zr and the other cladding alloying metals. A well-known form of chemical decladding is chlorination, in which solid Zr-based cladding is reacted with gaseous Cl_2 or HCl to form $ZrCl_4$. At typical dry chlorination temperatures, $ZrCl_4$ is volatile and can be transported in the gas phase away from the uranium oxide fuel pellets, which are not reactive under dry chlorination conditions. The $ZrCl_4$ is then recovered as a condensed solid, and the uranium is fed into the dissolution and separations portion of the processing facility.

Recovery of Zr from nuclear fuel is of high interest because the Zr cladding material comprises up to 50% of the high-level radioactive waste (HLW) requiring disposition.[1] For this reason, other potential Zr recovery pathways have been investigated, including processing the empty cladding segments (hulls) that are discharged following a fuel shearing and nitric acid dissolution process. Chlorination can also be used to recover Zr from alternative fuel types, such as from U–Zr alloy fuels. The Zr itself is not highly radioactive, but it is contaminated with embedded transuranics, embedded fission products, and activated alloying metals. An ideal Zr recovery process would not only recover the bulk Zr from the fuel or cladding but would also provide decontamination from the components that give rise to the HLW designation. An effective decontamination (or simplified decontamination pathway) providing recovered Zr in compliance with low-level radioactive waste acceptance criteria could significantly reduce the burden of HLW requiring geologic disposition. As an alternative to Zr decontamination and disposition, the potential exists for reusing the nuclear-grade (i.e., Hf-free) Zr. This material may be reused as recovered or can even be enriched in ^{90}Zr, which has more desirable neutron capture properties than Zr of natural isotopic abundances.

In the case of typical LWR fuel, common cladding formulations have varied over time and across specific reactor type. Three selected alloy formulations are listed in Table 1, adapted from Collins, et al. (2017).

Table 1. Composition of common zirconium cladding alloys.

Element	Concentration in alloy (µg/g Zr)		
	Zircaloy-2	**Zircaloy-4**	**High-Nb variations**
Sn	14,200	12,900	<35
Nb	<50	<50	10,100
Fe	1,720	2,140	400
Cr	1,020	1,110	<50
Ni	690	<35	<35

Of these alloying elements, Sn, Fe, and Nb (present as ^{94}Nb) have been the most challenging to separate from Zr using traditional chlorination or hydrochlorination processes.[2] These elements tend to form volatile chloride species that can transport with the $ZrCl_4$ product at process temperatures. Industrially, some of these metal chlorides are reduced by an Ar/H_2 mixture at 500°C to nonvolatile species.

Some other elements have also been found as contaminants in $ZrCl_4$. These include Sb (present as ^{125}Sb generated via activation of Sn) and Cs (present as ^{134}Cs and ^{137}Cs resulting from U fission).

The new *advanced low-temperature chlorination* process presents an opportunity to more simply recover a $ZrCl_4$ product that is effectively decontaminated from the elements of concern.[3] This advanced low-temperature chlorination process uses a mixture of the sulfur-containing chlorination compounds sulfur monochloride (S_2Cl_2) and thionyl chloride ($SOCl_2$). These compounds can chlorinate zirconium to $ZrCl_4$ at their reflux temperature (approximately 95°C for the mixture used). The $ZrCl_4$ is soluble in this mixture whereas other important contaminants (iron and cesium) are not, enabling separation of a purified $ZrCl_4$ product via filtration.

Development of the advanced low-temperature chlorination process has been focused on the chlorination of single components (either unirradiated cladding segments or pure metals) or on unirradiated UO_2- or U-containing rodlets that do not contain the contaminants present in irradiated clad fuel.[4, 5] This fundamental work has been necessary for understanding reaction mechanisms and reaction rates for pure metals and characterizing solubilities of the chlorinated products in S_2Cl_2/$SOCl_2$ mixtures.

Building on that fundamental work, the next step is to develop a more applied understanding of how the advanced low-temperature chlorination process could partition particular contaminants in an irradiated fuel assembly. Toward that end, two proof-of-principle experiments were conducted. The first experiment chlorinated a mixture of Zircaloy-4 cladding segments and other elemental metals. The second experiment chlorinated a mixture of UO_3 and selected fission products representing simulated irradiated fuel. Together, these experiments demonstrate how the primary constituents of an irradiated spent fuel rod would behave in the advanced low-temperature chlorination process and provide a preliminary assessment of $ZrCl_4$ product purity when using a more realistic and complex simulant.

2. MATERIALS AND METHODS

Experiments were performed in a dry N_2 atmosphere using Schlenk reaction tubes.

The reaction system, illustrated in Figure 1, includes an N_2 supply, the Schlenk reaction tube, a condenser, and a NaOH gas scrubber on the condenser effluent.

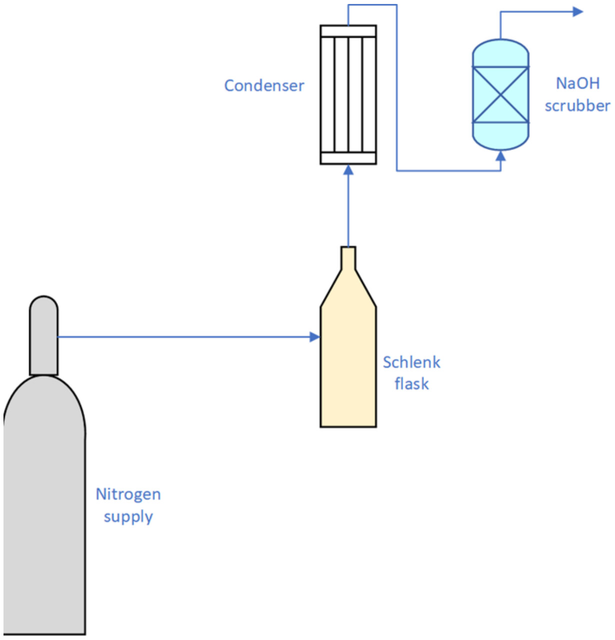

Figure 1. Bench-scale equipment configuration for the advanced low-temperature chlorination process.

The general chlorination protocol is depicted in Figure 2. The first step is the addition of a 42/58 mixture of $SOCl_2/S_2Cl_2$ to a Schlenk flask that contains the solids to be chlorinated. If desired, the reaction can be started at 0°C by using chilled solvent, resulting in a decreased reaction rate. When desired, the solvent is brought to reflux temperatures (approximately 95°C). The reaction progress can be monitored via gas evolution because the Zr chlorination reaction evolves SO_2.

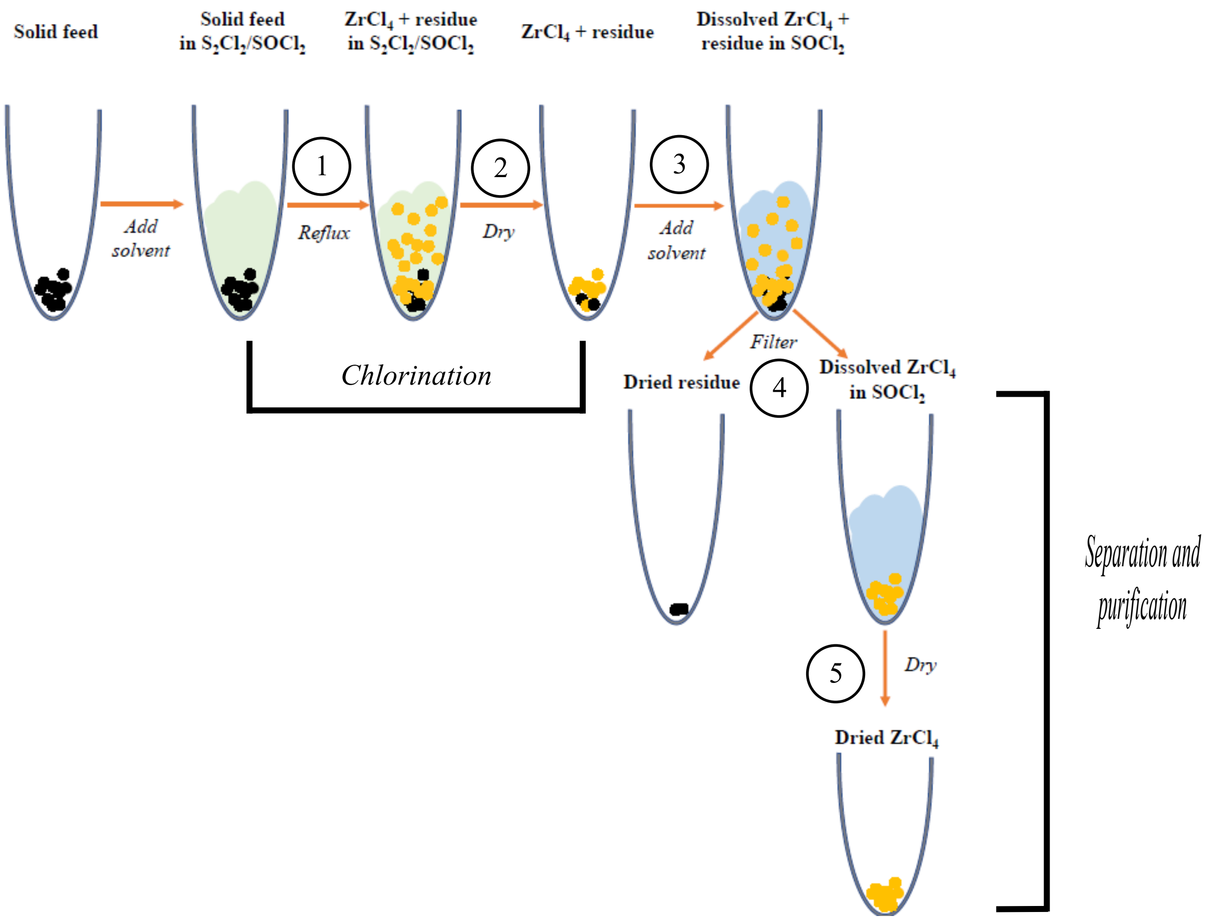

Figure 2. Conceptual representation of the advanced low-temperature chlorination process.

After a typical induction period of about 30 min at reflux temperatures, vigorous chlorination of Zr can be inferred from gas production, solid particulates floating in solution, and a slight rise in temperature. This period of vigorous reaction typically lasts 10–15 min, depending on the amount of Zr present. To ensure complete chlorination, the mixture is refluxed for an additional 3–4 hours after solution foaming and gas evolution have decreased.

The solvents are volatile and can be removed by vacuum. Other volatile species could be removed during this drying process. Removing the solvents (step 2) leaves solid $ZrCl_4$, along with any residual unreacted solids, within the reaction flask. This $ZrCl_4$ is dissolved into $SOCl_2$ (steps 3–5) for purification and recovery at approximately 60°C. Some of the common contaminants of concern are not soluble in $SOCl_2$ and will remain as solids or particulates (e.g., CsCl and $FeCl_3$). After the $ZrCl_4$ is dissolved into $SOCl_2$, the solution is filtered via cannulation to remove any residual unreacted solids or solid chlorinated material. This step effectively transfers dissolved $ZrCl_4$ and other soluble components into a clean new Schlenk flask, thereby separating them from the unreacted solids. The undissolved materials remaining in the original Schlenk reaction vessel are washed with additional warm $SOCl_2$, which is then also cannulated into the separate flask to ensure complete $ZrCl_4$ recovery. Both the unreacted solids and the $ZrCl_4$ in the new reaction flask are dried under vacuum. The $SOCl_2$ removed from these materials can be condensed as liquid waste or recycled.

All solid samples are analyzed using gravimetric analysis and inductively coupled plasma mass spectrometry (ICP-MS) to determine the elemental partitioning via the advanced low-temperature chlorination process.

The off-gas scrubber for this system is a mini-scrubber with a total volume of less than 25 mL NaOH.

2.1 MATERIALS

Dry N_2 was delivered to the chlorination apparatus from a gas cylinder plumbed into a nitrogen outlet in a hood. Atmospheric pressure was used in these experiments.

Reagent-grade samples of Fe, Cr, Sn, and Nb metals were used without pretreatment, with additional details provided in Table 2.

Reagent-grade CsCl was used as received.

S_2Cl_2 and $SOCl_2$ were procured from a commercial supplier (Alpha) and used without any purification or pretreatment.

Table 2. Composition of Scoping Experiment 1 irradiated cladding simulant.

Material	Physical form	Weight (g)	Concentration	
			(µg/g)	(mg/g)
Zircaloy-4 cladding	Unirradiated segments	9.2	5.6×10^5 (Zr only)	5.6×10^2 (Zr only)
Fe	Thin wire, 0.009 in. diameter	0.97	60,000	60.0
Sn	Powder, 0.016 in. mesh	0.995	61,600	61.6
Nb	Powder, 0.0029 in. mesh	1.0012	62,000	62.0
CsCl	Coarse powder	3.0123	186,000	186
Cr	Pellets	0.98	60,680	60.7

2.2 SCOPING EXPERIMENT 1: IRRADIATED CLADDING SIMULANT

The solid feed for this experiment was intended to represent irradiated cladding and included Zircaloy-4 cladding segments and a mixture of other key contaminants (Fe, Sn, Nb, Cs [as CsCl], and Cr). The composition of the solid feed is shown in Table 2. The contaminants were included at levels significantly higher than what would be expected in irradiated cladding, to both challenge the process and simplify the analysis methods.

The advanced low-temperature chlorination process was performed as described in the general chlorination protocol (see Figure 2), with the reaction started at 0°C and a solvent mixture of 31 mL S_2Cl_2 and 44 mL $SOCl_2$. A vigorous reaction period was observed about 30 min after bringing the solution to reflux. During this period, the flask was allowed to cool back to ambient room temperature to slow the reaction slightly and ensure adequate headspace in the Schlenk reaction flask. A second vigorous reaction period was observed approximately 10 min after the conclusion of the first reaction event while the flask was still at ambient temperature. The cause of this second reaction period is unknown, but, given the heterogenous nature of the solid materials in the flask, it could have resulted from an unreacted metal (e.g., Sn) becoming exposed and chlorinating quickly.

After $ZrCl_4$ production was complete, the solvent was removed and the residue was dried, as depicted in Figure 2. After drying, the residual solids mass comprised a mixture of fully chlorinated (e.g., $ZrCl_4$) and partially chlorinated materials. Unreacted Fe, Cr, and Nb were visually present. Fifty-five milliliters of $SOCl_2$ was used to dissolve the $ZrCl_4$ and any other materials that could be soluble in $SOCl_2$, such as $NbCl_5$. This solution was filtered via cannulation to separate the dissolved $ZrCl_4$ from the residual solids. During filtration, there was some concern that the filter paper experienced a small tear that would have allowed some residual solids through. For this reason, the filtered solution was re-dried, dissolved again in $SOCl_2$, and filtered a second time. After the second filtration, the solvent was removed, leaving $ZrCl_4$ solids in the flask. The weights of the dried $ZrCl_4$ and the dried solid residue were obtained. Understanding that potential impurities could be present in the $ZrCl_4$ in heterogenous fashion, the $ZrCl_4$ solids were sampled from three different locations within the flask, and this material was sent for ICP-MS analysis to determine the concentrations of Zr and other metal impurities.

The NaOH scrubber on the effluent was sampled twice from the 25 mL solution and sent for analysis of Sn. Because it is highly volatile, $SnCl_4$ is expected to transfer to the scrubber, where it can oxidize to SnO_2. Evidence of SnO_2 formation was present after reaction was complete based on the observation of colloidal material in the scrubber.

2.3 SCOPING EXPERIMENT 2: IRRADIATED FUEL SIMULANT

The solid feed for this experiment simulated irradiated fuel and contained a mixture of UO_3 and three potential fission products representing Group 1 elements, Group 2 elements, and the lanthanides. The composition of the solid feed is shown in Table 3.

Table 3. Composition of Scoping Experiment 2 irradiated fuel simulant.

Oxide basis	Elemental basis	Elemental concentration (wt%)
23.96 g UO_3	19.94 g U	83.2
0.23 g Eu_2O_3	0.20 g Eu	0.8
0.24 g SrO	0.20 g Sr	0.8
0.21 g Cs_2O	0.20 g Cs	0.8

The advanced low-temperature chlorination process was initiated as described in the general chlorination protocol (see Figure 2), with 7 mL of S_2Cl_2 and 13 mL of $SOCl_2$ added to the solid feed. The mixture was brought to reflux at approximately 95°C for 5 hours. No reaction event (as evidenced by gas evolution or other visual markers) was observed, nor was one expected given that most metal oxides are not expected to chlorinate at these temperatures. Following reflux, the solvent was removed, and the solid residue was weighed. No difference in the weights of the starting material and the residue was observed, indicating that no chlorinated compounds were produced within experimental error. Therefore, filtration and purification steps were not performed, and the experiment was considered complete.

3. ANALYSIS AND OBSERVATIONS

Both experiments were completed successfully, and they provided an opportunity to better understand the practical aspects of the advanced low-temperature chlorination process. To understand how the components of the solid feed for each experiment were partitioned via the process, both gravimetric and ICP-MS analyses were performed.

3.1 GRAVIMETRIC ANALYSIS

Given the practicalities of weighing tared vessels containing radioactive material under nitrogen, weight measurements are estimated as having error bars between 1% and 10%.

3.1.1 Gravimetric analysis of Scoping Experiment 1

Based on previous studies, tin is expected to completely volatilize as $SnCl_4$ during chlorination and should transfer to either the NaOH off-gas scrubber or through the solvent as it is removed under vacuum. Therefore, tin was not included when comparing the masses of the undissolved residue or the soluble materials. The weight of the undissolved residue from Scoping Experiment 1 was equal to the sum of the masses of each starting material (i.e., Fe, Cr, Nb, and Cs [as CsCl]) within measurement error. Additionally, the mass of the recovered $ZrCl_4$ residue was within 10% of the mass predicted for the complete chlorination of the original Zircaloy-4 segments in the starting metal mixture, which implies that Zr chlorination was complete and that less than 10% of the minor components were chlorinated under the conditions used. Note that the chlorination of tin to $SnCl_4$ produces a stoichiometric equivalent of S that was also included in the calculation of the theoretical values. This result is consistent with previous experimentation that indicates the reaction pathways for each component as shown in Table 4. These reaction pathways are the reactions expected to occur in the initial chlorination step with the mixed solvent system.

Table 4. Proposed reaction pathways for components of irradiated cladding simulant.

$3\ Zr + 8\ SOCl_2 \rightarrow 3\ ZrCl_4 + 4\ SO_2 + 2\ S_2Cl_2$
$Sn + 2\ S_2Cl_2 \rightarrow SnCl_4 + 4\ S$
Nb → Nb (no reaction)
Cr → Cr (no reaction)
Fe → Fe (no reaction)
CsCl → CsCl (no reaction)

3.1.2 Gravimetric analysis of Scoping Experiment 2

After chlorination, the solid residue was weighed. No difference in the weights of the starting material and the residue was observed within measurement error, indicating no or very limited chlorination. This result was expected based on previous results and thermodynamic estimations.

3.2 ICP-MS ANALYSIS

Because no chlorination was observed in Scoping Experiment 2, only samples from Scoping Experiment 1 were analyzed by ICP-MS. The results of this analysis are shown in Table 5.

Table 5. ICP-MS results of dried and filtered digestion product.

Material	Starting wt% metal	ICP-MS results wt% metal	Decontamination factor
Zircaloy-4 cladding	56.9	93.85	—
Fe	6.00	3.55	1.69
Sn	6.16	<0.032	>193*
Nb	6.20	0.050	124
CsCl	14.7	2.47	5.96
Cr	6.19	<0.077	>80.4*
** Indicates complete removal of the material to below the instrument detection limit*			

The purity of the $ZrCl_4$ product was very high considering that Scoping Experiment 1 was the first demonstration of this advanced low-temperature chlorination process using a simulated feed and non-optimized equipment. The most significant contaminants were Cs (2.47 wt% ± 0.2 wt%) and Fe (3.55 ± 0.4 wt%). Because CsCl and $FeCl_3$ are insoluble in $SOCl_2$, these contaminants may have resulted from an imperfect solids filtration step, rather than from reactive impurities traveling with the $ZrCl_4$ product. Sn was completely removed. Nb and Cr were present in the product at 1 μg/mL levels. This analysis indicates quantitative chlorination and recovery of Zr as the desired tetrachloride product.

The Sn concentrations measured in NaOH were 40.7 and 37.9 μg/mL for duplicate samples. The scrubber was not designed to quantitatively remove Sn, but rather to simply ensure that Sn partition to the gas phase could be confirmed. In this respect, the off-gas trapping was successful. A full mass balance was not performed. Future experiments could include a larger scrubber to ensure complete Sn capture and could perform ICP-MS analysis to determine whether other metals are also transferred to the gas phase.

3.3 OFF-GAS TREATMENT

Off-gas treatment of the advanced low-temperature chlorination process would have to account for a radioactive source term that includes $SnCl_4$, some ^3H (as HCl), and Kr/Xe (if breaching spent fuel rods). Other components, such as iodine, could be present in limited amounts. The capture efficiencies required for these components will likely be relatively low because their release rates should be low if the fuel remains intact during the cladding breach.

The most significant component of the stream will be SO_2 evolved during chlorination of Zr with the mixed solvent system (Table 4). The high concentration of SO_2 is not similar to other common nuclear-related off-gases, but it is common to industrial emission points such as coal-fired power plants. Common SO_2-scrubbing technologies use Ca-containing aqueous scrubbers or slurries to form $CaSO_4$. These SO_2-scrubbing technologies are expected to transfer easily to this application.

Future research and development should ensure that the radioactive source term is quantified and that needed removal efficiencies are estimated. The potential benefits of reagent recycling should be evaluated, because both elemental S and SO_2 byproducts can be easily recycled for reuse as S_2Cl_2 and $SOCl_2$, respectively. A prototypical scrubbing system using industrially demonstrated SO_2-scrubbing technology should be investigated for use against a simulant stream. Some partitioning of the contaminant volatile radionuclides could occur. If acceptable removal efficiencies are not demonstrated, future research and development may be necessary.

4. CONCLUSIONS AND TECHNOLOGY DEVELOPMENT PATH

Scoping Experiments 1 and 2 resulted in several valuable findings related to the advanced low-temperature chlorination process. Scoping Experiment 2 confirmed results obtained by the University of Tennessee regarding the nonreactivity of uranium oxides within the typical parameters of the advanced low-temperature chlorination process. This result indicates that advanced low-temperature chlorination could be used for separating Zr from solid feeds containing U, whether these are alloy-type fuels, common LWR fuels, or other materials. No chlorination of the included fission product simulants introduced as oxides (Cs, Sr, and Eu) was observed within experimental error.

Second, Scoping Experiment 1 showed that a mixture of $SOCl_2/S_2Cl_2$ is a very effective and *selective* chlorination agent for quantitatively transforming the Zr in Zircaloy claddings into $ZrCl_4$, even in the presence of large amounts (>50,000 ppm or >5 wt%) of radiological contaminants arising from irradiation. The purity of the recovered $ZrCl_4$ was 93.85%. Note that the levels of contaminants used in this experiment were artificially high (roughly 100× what might be expected from irradiated cladding). This result further confirms the robustness of the advanced low-temperature chlorination process and indicates that an even more pure product may be easily obtained when using less challenging mixtures. Solids filtration via cannulation is challenging in the current test system, and improvements to this step may also improve product purity.

It follows then, that mixtures of $SOCl_2/S_2Cl_2$ are not effective as chlorination agents for metals such as Fe, Cr, and Nb in the time that is required to completely chlorinate Zircaloys (i.e., Zr). By contrast, recovery of Sn in the off-gas scrubber confirms that tin metal (coarse powder) is effectively chlorinated by $SOCl_2/S_2Cl_2$ mixtures at time scales comparable to those required for Zircaloy chlorination and that it easily transfers to the process off-gas. All of these results are in complete accord with the results that have been described in previous milestone reports and conference proceedings.[4, 5].

These results indicate that the advanced low-temperature chlorination process holds promise for chemical decladding, hulls processing, and $ZrCl_4$ purification processes and that it merits continued research and development. The advanced low-temperature chlorination process provides a simple pathway to a clean Zr product that requires limited additional purification. As the process moves from basic to more applied science, several important aspects of the process should be investigated. First, a detailed flowsheet should be developed that better defines how this chemical process could be implemented at an engineering scale. Next, the integration of this process into complete spent fuel processing methods should be demonstrated. Scale-up and integration of any nuclear-related process must address multiple technical issues, such as the robustness of the chemicals to radiolysis, reaction kinetics, the construction materials that could be used in a hot cell for this process, the potential impacts of corrosion, radiological containment, and the treatment of solid, liquid, and gaseous effluents.

In addition to flowsheet development and analysis, three types of experiments would be especially beneficial to continued technological development of this process. First, scoping-level testing should be expanded to more realistic simulant compositions and more comprehensive and precise analysis. The testing described here, as a scoping-level effort, is limited. More rigorous experiments with realistic low-level contaminants, trace-level contaminant analysis, solvent analysis, and complete quantification of off-gas constituents would provide full quantification of product and waste streams such that realistic benefit assessments of the advanced low-temperature chlorination process can be performed. Second, a Zr recovery experiment should be completed using irradiated cladding segments. This experiment could be performed with cladding amounts that can be handled in a glovebox. Previous work on irradiated Zr processing experienced unexpected results when full hot-cell demonstrations were performed. It is recommended that hot testing be performed for this technology early in the development cycle to screen more quickly for potential technical issues. Finally, a SO_2-scrubbing demonstration should be performed using a realistic simulant off-gas stream containing tritium (as ^{3}HCl), Sn, and other off-gas contaminants at levels expected from the advanced low-temperature chlorination process. Early demonstration of manageable off-gas streams arising from the process will reduce risk from waste management.

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