Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

We put science to work.™

Savannah River
National Laboratory™

OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

A U.S. DEPARTMENT OF ENERGY NATIONAL LABORATORY • SAVANNAH RIVER SITE • AIKEN, SC

Ruthenium Volatilization During Dissolution of Spent Nuclear Fuels in H-Canyon Dissolvers

T. S. Rudisill

February 2019

SRNL-TR-2018-00073, Revision 0

SRNL.DOE.GOV

DISCLAIMER

Printed in the United States of America

Prepared for
U.S. Department of Energy

SRNL-TR-2018-00073
Revision 0

Keywords: *Ruthenium Volatilization, SNF Dissolution, H-Canyon*

Retention: *Permanent*

Ruthenium Volatilization During Dissolution of Spent Nuclear Fuels in H Canyon Dissolvers

T. S. Rudisill

February 2019

Prepared for the U.S. Department of Energy under contract number DE-AC09-08SR22470.

Savannah River National Laboratory™
OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

REVIEWS AND APPROVALS

AUTHORS:

T. S. Rudisill, Separations and Actinide Science Programs Date

TECHNICAL REVIEW:

J. M. Duffey, Separations and Actinide Science Programs Date

W. H. Clifton Jr., H-Canyon Engineering Date

APPROVAL:

T. B. Brown, Group Leader Date
Separations and Actinide Science Programs

J. E. Therrell, Manager Date
H-Canyon/Outside Facilities Engineering

S. D. Fink, Director Date
Chemical Processing Technologies

EXECUTIVE SUMMARY

During nuclear material processing, many radioactive elements form volatile species under certain process conditions which may allow release to the environment unless they are removed from offgas streams. Ruthenium, a product from the fission of U, will form a volatile compound (RuO_4) under highly oxidizing conditions which may be released during spent nuclear fuel (SNF) dissolution. The principal Ru isotopes of concern are ^{103}Ru and ^{106}Ru with half-lives of 39.27 and 372.6 days, respectively. Ruthenium-103 will decay to less than 1% of its initial activity within one year; therefore, its presence is not a concern for fuel cooled more than one year after discharge. However, the half-life of ^{106}Ru is sufficiently long that it will persist for many years at levels of concern. The isotope does not decay to less than 1% of its initial activity until approximately 6.8 years. Therefore, the dissolution of SNF cooled for less than approximately 7 years is defined as processing short-cooled fuel as it relates to Ru volatilization.

Ruthenium is a troublesome fission product during the reprocessing of SNF due to its potential to form volatile RuO_4 during fuel dissolution and extract into the tributyl phosphate (TBP) solvent used in the PUREX process. Ruthenium occurs in HNO_3 solutions of irradiated fissile material largely as ruthenium(III) nitrosyl (RuNO) complexes primarily with nitrato (NO_3^-) and nitro (NO_2^-) ligands. The behavior of Ru during dissolution of SNF and in downstream purification and waste management operations is highly dependent upon the speciation. The primary reason for changes in Ru speciation is associated with small changes in the concentration of HNO_2. The balance between the RuNO nitro and RuNO nitrato complexes is critically dependent upon the HNO_2 concentration in warm or heated solutions. Lower concentrations of HNO_2 in solution containing Ru favor the formation of the RuNO nitrato complex. The nitrato complex is more easily converted to volatile RuO_4 and extracted into TBP during solvent extraction operations. Higher concentrations of HNO_2 result in higher concentrations of the RuNO nitro complex which has higher stability in HNO_3 solutions (i.e., less likely to form volatile RuO_4) and is extracted to a lesser extent into a TBP solvent.

Significant releases of ^{103}Ru and ^{106}Ru have occurred from both F- and H-Canyon, mostly attributed to nonroutine operations and unrecognized changes in process chemistry which led to the volatilization of RuO_4 and its subsequent release to the environment. A common thread in two of the releases was the dissolution of Al metal prior to complete dissolution of the fissionable material. If Al is present in the dissolver, its dissolution will generate NO_2 and NO gases which serve as reducing agents. The NO_2 and NO gases are in equilibrium with HNO_2 which favors the formation of RuNO nitro complexes in solution. When short-cooled fuels were reprocessed in H-Canyon, a permanganate strike was performed in head-end treatment to remove Zr and Nb fission products. Generally, the permanganate strike resulted in small releases of ^{103}Ru and ^{106}Ru. To address this issue, an absorber packed with stainless steel mesh was designed and installed to remove RuO_4 from the sweep of air pulled through the strike tank by the vessel vent system. The Ru absorber remained in H-Canyon until the 1990's when the there was no longer a need to process short-cooled reactor fuels.

In previous work at the Savannah River Laboratory, an evaluation of the variables which affected the volatilization of RuO_4 during SNF dissolution was performed to develop a range of operating conditions which eliminated Ru volatilization. The formation of RuO_4 was controlled by the concentration of HNO_3 used in the dissolver and the temperature. The effect of the acid concentration and temperature on the fraction of RuO_4 volatilized during 20 h of heating was used to define the acceptable operating region. To prevent Ru volatilization, the temperature of the solution must be maintained significantly below the boiling point especially at higher HNO_3 concentrations used to dissolve SNF (e.g., 4-8 M). The use of reducing agents was also evaluated to prevent the volatilization of RuO_4 during dissolutions. The slow addition of nitrite (as $NaNO_2$) or NO_2 gas was recommended to increase the equilibrium concentration of HNO_2 which favors the formation of the more difficult to oxidize RuNO nitro complexes. The experimentally determined addition rates of NO_2^- and NO_2 prevented the volatilization of Ru from a dissolving solution containing 8 M HNO_3 at boiling.

The flowsheets developed for Material Test Reactor (MTR) and High Flux Isotope Reactor (HFIR) fuel dissolution were assessed for their potential to volatilize Ru during processing. The flowsheets recommended the use of 5 to 7.5 M HNO_3 containing 0.002 M Hg at the boiling point of the solution which is well within the operating region where Ru is volatilized. However, the release of Ru is mitigated to a large extent if the dissolution of Al is incomplete. The generation of gases which serve as reducing agents maintains significant concentrations of HNO_2 in solution, which favors the formation of more stable RuNO nitro complexes. As HNO_3 is consumed during the fuel dissolution, the driving force to oxidize Ru is reduced and at approximately 2 M HNO_3, the dissolving conditions are in the operating region where Ru volatilization does not occur. A bounding amount of ^{106}Ru which could be released from H-Canyon during the dissolution of short-cooled HFIR fuel was estimated for cores with cooling times from approximately 4 to 10 years. The estimates were based on the calculated ^{106}Ru activity in the fuel, the flowsheet conditions, previous SRNL work in which the fraction of RuO_4 volatilized during 20 h of heating was measured (in the absence of Al dissolution), and a decontamination factor of 30 across the H-Canyon ventilation system. The cumulative ^{106}Ru releases for the dissolution of five HFIR cores ranged from approximately 3300 mCi for 4.12 years cooling to 50 mCi for fuel cooled for 10.37 years.

If short-cooled MTR or HFIR fuels are processed in H-Canyon, modifications to the dissolution flowsheets would be required to minimize the volatilization of Ru. Dissolution of the SNF's using lower temperatures and concentrations of HNO_3 could increase the concentration of H_2 in the offgas; although, less aggressive dissolution conditions would be expected to generate less total offgas. Measuring the H_2 and total offgas generation rates in laboratory-scale dissolutions would be required to demonstrate that the H_2 concentration in the offgas from an H-Canyon dissolver does not exceed 60% of the lower flammability limit. Using less aggressive conditions will have a deleterious effect on the rate of dissolution and limit the number of bundles or cores which can be dissolved due to the consumption of HNO_3. Dissolving SNF at a sufficiently low temperature which prevents Ru volatilization but also provides a reasonable dissolution rate may be difficult to initially control due to the surge in temperature which occurs below the boiling point during dissolver heating and often results in solution boiling. Another flowsheet strategy which will minimize the volatilization of Ru is to ensure that a portion of the fuel remains undissolved prior to charging the next core. The continued dissolution of Al will maintain the concentration of HNO_2 at levels which maximize the presence of the more stable RuNO nitro complexes.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

DF	decontamination factor
Fe_2O_3	hematite
HFIR	High Flux Isotope Reactor
H_2O	aquo ligand
LFL	lower flammability limit
MTR	material test reactor
$(NH_4)_2Ce(NO_3)_6$	ceric ammonium nitrate
NO	nitric oxide
NO_2	nitrogen dioxide
NO_2^-	nitro ligand or nitrite ion
NO_3^-	nitrato ligand or nitrate ion
OH^-	hydroxo ligand
ORNL	Oak Ridge National Laboratory
PUREX	plutonium uranium reduction extraction
RuNO	ruthenium(III) nitrosyl
SNF	spent nuclear fuel
SO_2^{2-}	sulphito ligand or sulfite ion
SRS	Savannah River Site
TBP	tributyl phosphate

1.0 Ru Process Chemistry and Volatilization

1.1 Radioactive Decay of Ru Isotopes

During nuclear material processing, a number of radioactive elements form volatile species under certain process conditions which may allow release to the environment unless they are removed from offgas streams.[1] Ruthenium, a product from the fission of U, will form a volatile compound (RuO_4) under highly oxidizing conditions which may be released from solution during the dissolution of spent nuclear fuel (SNF).[2] The principle Ru isotopes present in SNF following reactor discharge are ^{103}Ru and ^{106}Ru. Ruthenium-103 has a half-life of 39.27 days[3] and will decay to less than 1% of its initial mass within one year; therefore, its presence is not a concern for fuel cooled more than one year after discharge. However, ^{106}Ru has a half-life of 372.6 days[3] and will persist for many years at levels of concern. Figure 1-1 shows the decay of ^{103}Ru and ^{106}Ru in terms of the mass remaining following the discharge of fuel as a function of time. The half-life of ^{106}Ru is sufficiently long that the isotope does not decay to less than 1% of its initial mass until approximately 6.8 year. Therefore, the dissolution of SNF cooled for less than approximately 7 years is defined as processing short-cooled fuel as it relates to Ru volatilization.

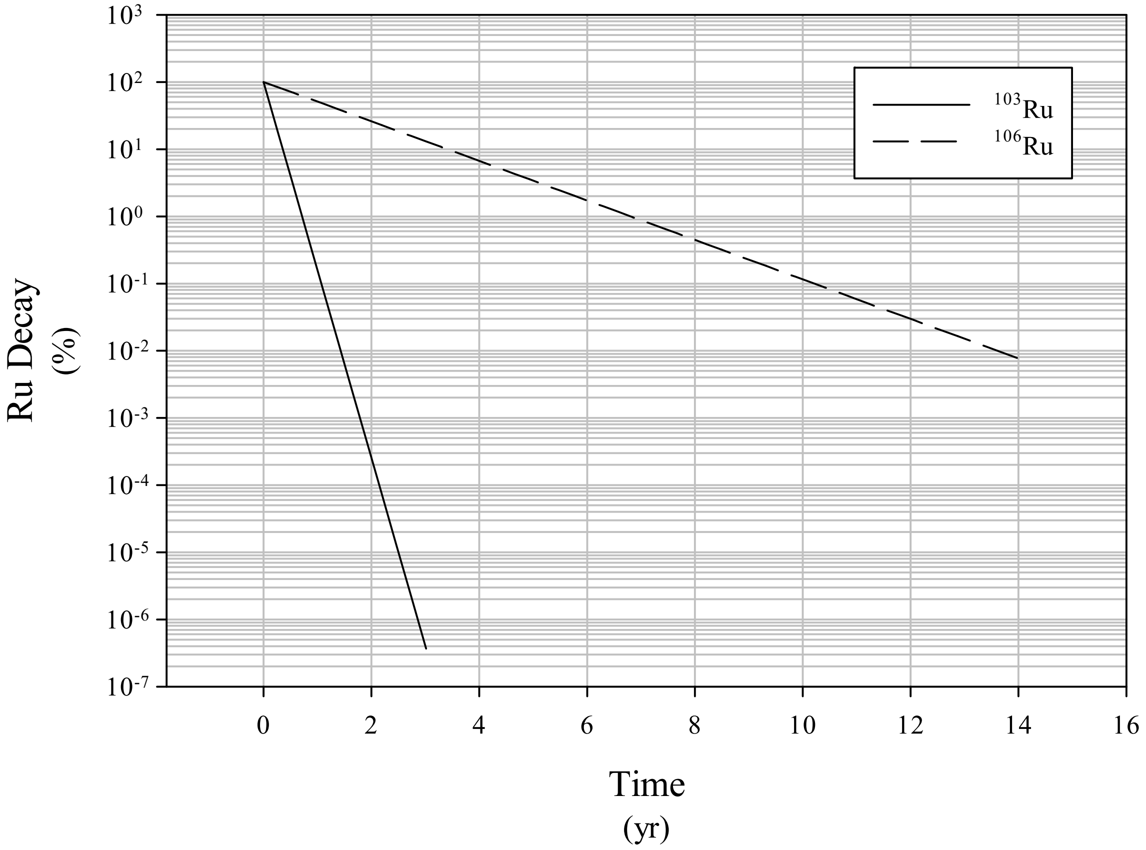

Figure 1-1. Radioactive Decay of ^{103}Ru and ^{106}Ru

1.2 Ru Chemistry in HNO_3 Solution

Ruthenium is a troublesome fission product during the reprocessing of SNF due to its potential to form volatile RuO_4 during fuel dissolution, evaporation processes, and waste vitrification, and extract into the tributyl phosphate (TBP) solvent used in the PUREX process.[4] Fission product Ru occurs in HNO_3 solutions of irradiated fissile material largely as ruthenium(III) nitrosyl (RuNO) complexes with nitrato (NO_3^-), nitro (NO_2^-), hydroxo (OH^-) and aquo (H_2O) ligands.[5] A large number of individual RuNO complexes are possible in HNO_3 solution depending on the solution composition and temperature.[6] Table 1-1 summarizes the key Ru species which are subject to oxidation–reduction reactions, extraction into TBP, and precipitate under conditions applied in effluent treatment (e.g., stirring with an alumino-ferric hydroxide floc at pH 10).[7]

Table 1-1. Properties of Ru Species Occurring in SNF Process Solutions

Species	**Oxidation to RuO_4 in HNO_3 solution**	**Behavior in HNO_3–HNO_2 mixtures at room temperature**	**Extractability by organic solvents**	**Ease of precipitation (effluent treatment)**
(1) RuNO nitrato complexes	Easy, but slow	Moderately stable, but conversion to (4) possible	Increases with degree of nitrato complexing	Good
(2) RuNO nitro complexes	Difficult	Stable	Moderate	Poor
(3) RuNO uncomplexed	Easy, but slow	Converted to (1) and (2) under appropriate conditions	Very low	Easy, but slow
(4) RuORu nitrate	Very easy	Stable in absence of HNO_2	Very low	Very easy

Ruthenium nitrosyl nitrato complexes are formed during fuel dissolution and are likely to persist throughout head end processing, solvent extraction, and evaporation of waste solutions. The RuNO nitrato complex is relatively weak and may be slowly changed by nitrous acid to RuNO nitro complexes, by sulfite (SO_2^{2-}) to RuNO sulphito complexes, by oxidation to Ru(IV), and reduction to Ru(II) or Ru(III). There are a series of these complexes ranging from the pentanitrato complex ($H_2[RuNO(NO_3)_5]$) which is strongly extracted into TBP to the mononitrato complex which is weakly extracted. The percentages of the various complexes at equilibrium depend upon the nitrate rather than the hydrogen ion concentration. High nitrate concentrations favor the higher nitrato complexes (Table 1-2). The first order reactions between the complexes, when equilibrium is disturbed by a change in nitrate concentration, have half-time values of a few minutes at 25 °C.[7]

Table 1-2. Percentages of Ru as Various RuNO Nitrato Complexes at Equilibrium in HNO_3–Nitrate Solutions at 20 ˚C and~10^{-3} M Ru

HNO_3 (M)	Added Nitrate	Total Nitrate (M)	Penta- and tetra-nitrato (%)	Tri-Nitrato (%)	Di-Nitrato (%)	Mono-Nitrato (%)
0.1	−	0.1	−	−	4	96
1.0	−	1.0	2.5	13	19	65
1.0	$UO_2(NO_3)_2$	2.7	8	−	−	−
3.0	−	3.0	8	24	33	34
1.0	$UO_2(NO_3)_2$	3.5	13	−	−	−
3.0	$UO_2(NO_3)_2$	4.7	22	−	−	−
3.0	$UO_2(NO_3)_2$	5.5	27	−	−	−
3.0	$NaNO_3$	5.5	20	−	−	−
5.5	−	5.5	28	26	26	20
7.5	−	7.5	44	24	22	10
11.3	−	11.3	65	18	12	5

The RuNO nitro complexes are more stable than the RuNO nitrato complexes and may involve 30-70% of the total Ru at 2-4 M HNO_3 containing 10^{-2} to 10^{-3} M HNO_2. Factors such as the HNO_2 concentration, temperature, and the gases in the free space above the solution fix the chemical equilibrium (equation 1).

$$\text{RuNO nitrato complexes} \underset{HNO_3}{\overset{HNO_2}{\rightleftharpoons}} \text{RuNO nitro complexes} \quad (1)$$

Once the RuNO nitro complexes are formed in solution, the complexes are difficult to destroy and tend to persist through all stages of fuel reprocessing and may even survive prolonged boiling in concentrated HNO_3.[7]

A portion of the Ru present in process solution will exist as uncomplexed RuNO primarily when the total nitrate concentration is low. The uncomplexed RuNO will serve as a reservoir of material for the formation of the nitrato and nitro complexes if increases in the concentrations of HNO_3 and HNO_2 occur. However, if the acidity of the solutions falls, polymers derived from the uncomplexed RuNO cation may form. The formation of compounds with more than one Ru atom (species 4 in Table 1-1) are rare in processes based on HNO_3 since at elevated temperatures, these compounds are rapidly converted by HNO_2 (or oxides of nitrogen) to RuNO species.[7]

1.3 Volatilization of RuO_4 during SNF Dissolution

Ruthenium tetroxide has been established as the primary volatile species released from boiling HNO_3 solutions during nuclear fuel dissolutions.[8,9] Pure RuO_4 crystals melt at 24.4 ˚C.[10] The literature provides values for the boiling point which range from 40 to 133.4 ˚C. A value of 129.6 ˚C reported by Koda was judged the most reliable.[11] The rate of oxidation of Ru to the volatile RuO_4 is dependent on the chemical speciation. The oxidation of the RuNO nitrato complex proceeds at a much faster rate than the RuNO nitro complex consistent with the stability of the nitro complex in HNO_3 solution (Figure 1-2).[7]

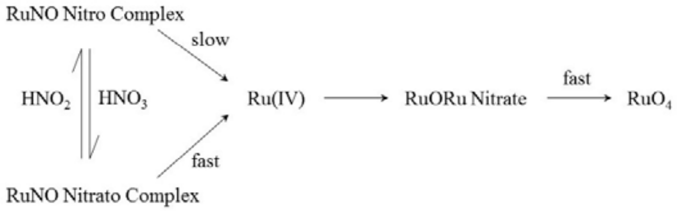

Figure 1-2. Oxidation of Ru Species in HNO_3 Solution to RuO_4

The rapid oxidation of the nitrato complex in HNO_3 was illustrated during a study performed by Brown et al.[7] in which an excess of ceric ammonium nitrate ($(NH_4)_2Ce(NO_3)_6$) was added as an oxidant to solutions containing the RuNO nitrato, RuNO nitro, and RuORu nitrate complexes (at 20 ˚C) and the percentages of Ru converted to RuO_4 were measured. The results from the study (Table 1-3) showed complete conversion of the RuORu nitrate complex to RuO_4 and that a much higher proportion of the RuNO nitrato complex was oxidized to RuO_4 when compared to the amount of the RuNO nitro complex which was oxidized.

Table 1-3. Oxidation of Ru Species in HNO_3 to RuO_4 with $(NH_4)_2Ce(NO_3)_6$

Ru Species	Ru Conc.	Percent Ru Converted to RuO_4					
		Oxidation in 0.1 M HNO_3			Oxidation in 2.4 M HNO_3		
		1 min	10 min	1 h	1 min	10 min	1 h
RuORu nitrate	10^{-3} M	100	100	100	100	100	100
RuNO nitrato complexes	10^{-3} M	51	74	96	50	70	93
	10^{-8} M	61	82	100	10	31	62
RuNO nitro complexes	10^{-3} M	3.6	13	32	0	0	0
	10^{-8} M	3.9	14	30	0	0	0

The behavior of Ru during dissolution of nuclear fuels and in downstream purification and waste management operations is highly dependent upon the Ru speciation in the process solutions. The primary reason for changes in Ru speciation is associated with small changes in the concentration of HNO_2. The balance between the RuNO nitro and RuNO nitrato complexes is critically dependent upon the HNO_2 concentration in warm or heated solutions. Lower concentrations of HNO_2 in solution containing Ru favor the formation of the RuNO nitrato complex. The nitrato complex is more easily converted to volatile RuO_4 and extracted into TBP during solvent extraction operations. Higher concentrations of HNO_2 result in higher concentrations of the RuNO nitro complex which has higher stability in HNO_3 solutions (i.e., less likely to form volatile RuO_4) and is extracted to a lesser extent into a TBP solvent.[7]

2.0 Ru Volatilization at the SRS

2.1 Historical Releases of Ru from SRS Canyons

Significant releases of ^{103}Ru and ^{106}Ru mostly attributed to nonroutine operations have occurred from both F-Canyon[12,13] and H-Canyon[14-18] during past operations. The circumstances leading to a number of these releases are discussed below to illustrate how process chemistry changes resulted in the Ru volatilization and subsequent release to the environment. A major release totaling approximately 30 Ci of ^{106}Ru occurred in 1978 from an F-Canyon dissolver. During the processing of Mark 41 PuO_2-Al cermet targets under a Test Authorization, the Al bundle/cladding and the fuel core were dissolved in boiling 7 M HNO_3 containing 0.01 M Hg and 0.01 M KF. Following the dissolution, analyses indicated that about 2% of the Pu predicted to be present remained undissolved. The dissolver was decanted and a 6-7 M HNO_3 solution containing 0.06 M KF complexed with Al in a 6:1 (fluoride to Al) ratio was added to dissolve residual PuO_2. The dissolver was initially heated at 75 ˚C for a total of 20 h, but PuO_2 was believed to still be present.

The dissolver was heated to boiling (105 ˚C) for 4 h to determine if the PuO_2 would dissolve. At this temperature, ^{106}Ru began volatilizing from the dissolver and was released from the stack due to the highly oxidizing conditions in the solution.[12,13] Since the Al present in the bundle, cladding, and fuel core had already dissolved and the PuO_2 dissolves slowly, there was no source of HNO_2 (including NO and NO_2 gases) which favors the formation of the RuNO nitro complex. Much of the Ru was likely converted to the RuNO nitrato complex which is easily oxidize to Ru(IV) and subsequently to the volatile RuO_4 species.

A release of ^{106}Ru during the dissolution of enriched U scrap in an H-Canyon dissolver occurred in 1976 due to similar process circumstances. The dissolution was also being performed using a Test Authorization. The unirradiated U metal in Al cans was charged to the High Flux Isotope Reactor (HFIR) insert in the 6.4D dissolver one can at a time. Prior to the U metal dissolution, the dissolver was used to dissolve HFIR fuel. At the end of the HFIR fuel campaign, an acid flush was left in the dissolver; however, when the acid charge for the U scrap was made, it was assumed that the last dissolver flush was process water. Consequently, the first can of U scrap was inadvertently dissolved in a higher HNO_3 concentration (6 M) than planned (3.5 M). About 60% of the U dissolved following 24 h of boiling, but an additional 28 h of dissolver operation only dissolved 10% more of the material. During this time ^{106}Ru was observed in the offgas going to the 221-H stack. Initially, the releases were small, but eventually reached 85 mCi/day.[16] The slow dissolution rate and the ^{106}Ru release were due to the highly oxidizing conditions in the dissolver and depletion of the HNO_2 (including NO and NO_2 gases) in the solution following dissolution of the Al can. Ruthenium remaining in the dissolver from the HFIR fuel dissolution was converted to the RuNO nitrato complex which was easily oxidized to volatile RuO_4. The remaining U metal dissolved slowly due to the absence of small amounts of HNO_2 in solution which catalyze the dissolution.[19]

In 1982, a release of ^{103}Ru and ^{106}Ru occurred during the decontamination of a failed H-Canyon head end strike tank. An estimated 45 mCi of activity were released over an eight-day period. At the time of the release, the failed canyon tank was being decontaminated by steam sparging a solution of 12 wt % (2.1 M) HNO_3 containing 0.75 wt % (0.05 M) $KMnO_4$.[14] Since the tank was used for permanganate strikes during head end processing,[20] a majority of the Ru in the tank was likely present as Ru(IV) (e.g., $RuO_2{\cdot}xH_2O$) which is formed by the decomposition of RuO_4; although, other species (including peroxy-bonded polymeric $(RuO_4)_n$ and RuO_3) have also been suggested.[21] Ruthenium tetraoxide is a very strong oxidizing agent. It is reduced to RuO_2 by traces of organic materials, stainless steel surfaces, and even metal surfaces coated with RuO_2.[13] Although, the dissolution rate of $RuO_2{\cdot}xH_2O$ in HNO_3 is slow (31 µg/L/min in 3 M HNO_3 at 100 ˚C),[22] the hot, acidic $KMnO_4$ solution would easily oxidize the RuNO nitrato complex formed upon dissolution in HNO_3 to volatile RuO_4.

Historically, when short-cooled fuels were reprocessed in H-Canyon, a permanganate strike was performed in head end treatment to remove Zr and Nb fission products. Generally, the permanganate strike resulted in small releases of ^{103}Ru and ^{106}Ru.[18] To perform a permanganate strike, $Mn(NO_3)_2$ was added to the dissolved fuel as a 25 wt % solution to achieve a Mn(II) concentration of approximately 0.014 M. The solution in the head end strike tank was then heated to approximately 70 ˚C followed by the addition of a 3 wt % $KMnO_4$ solution until sufficient permanganate was added to precipitate 90% of the $Mn(NO_3)_2$ as MnO_2 (equation 2). The permanganate strike was generally combined with a gelatin strike to remove silica and polysilicic acids and improve fission product decontamination.[20]

$$3Mn(NO_3)_2 + 2KMnO_4 + 2H_2O \rightarrow 5MnO_2 + 2KNO_3 + 4HNO_3 \tag{2}$$

The presence of excess $Mn(NO_3)_2$ during the permanganate strike reduces the amount of RuNO nitrato and nitro complexes which are oxidized to RuO_4. Even though the $KMnO_4$ is added at a slow, controlled rate below the surface of the solution with maximum agitation, up to 1% of the Ru present in the strike tank escapes as volatile RuO_4.[20,23]

2.2 Mitigation of Ru Volatilization from SRS Processes

In the late1950's, personnel at the Savannah River Site (SRS) recognized that the permanganate strike used in head end processing to remove Zr and Nb fission products from dissolved Pu production targets and reactor fuels would result in the volatilization of Ru. The addition of the highly oxidizing $KMnO_4$ to generate freshly precipitated MnO_2 resulted in the volatilization of 80 to 100% of the Ru depending on how long the oxidizing conditions were maintained in the tank. To address this issue, an absorber packed with stainless steel mesh was designed to remove RuO_4 in the sweep of air pulled through the strike tank by the vessel vent system.[24]

During the development of the basic data for the design of a Ru absorber, both transition and noble metal surfaces were tested for their ability to reduce and absorb volatile Ru. These tests demonstrated that stainless steel packing was an acceptable material for a practical absorber.[24] The appropriateness of stainless steel was subsequently validated in commercial fuel reprocessing and waste management operations where the deposition of volatilized Ru in stainless steel pipework is a potential problem due to the radiological hazards of ^{103}Ru and ^{106}Ru.[21] The absorber deployed in the SRS canyons utilized a standard 6 ft (diameter) x 6 ft (height) tank packed with knitted stainless steel demister screen. Coils were installed in the vessel for cooling if a high temperature was generated by the radioactive decay of Ru; however, the vessel was heated with steam to 145 ˚C to minimize condensation.[25]

In the mid 1980's Shook[25] performed a review of the existing Ru absorber operations to determine if the existing design was optimum or if a new technology was available to reduce releases. A review of the literature on the trapping of Ru in offgas streams showed that other technologies had been tested. Systems using silica gel, hematite (Fe_2O_3), or alkaline earth metal carbonates had received much study. Replacement of the stainless steel mesh in the existing absorber with a system based on the use of Fe_2O_3 was recommended. In laboratory studies, removal efficiencies greater than 99.9% were obtained. Hematite will absorb Ru from offgas streams with high removal efficiencies in the 300 to 550 ˚C range; however, it is ineffective at temperatures as low as 200 ˚C.[26] Hematite was never deployed in the H-Canyon Ru absorber. The absorber was removed in the 1990's when there was no longer a need to process short-cooled reactor fuels.

3.0 Elimination of Ru Volatilization during SNF Dissolution

In a study performed at the Savannah River Laboratory, Ondrejcin[13] evaluated variables which affected the release of RuO_4 during SNF dissolution. In this work, he found that the formation of RuO_4 was controlled by the concentration of HNO_3 used in the dissolver and the temperature. Once formed, the amount of RuO_4 dispersed was dependent upon the efficiency of the condenser and the use of air sparges and purges in the dissolver. The effect of the HNO_3 concentration and temperature on the fraction of RuO_4 volatilized over 20 h (without the presence of Al metal) is shown in Figure 3-1. The data extend to the boiling points of the HNO_3 solutions and were extrapolated to temperatures where no RuO_4 was volatilized. The extrapolated values were subsequently used to define an operating region (as a function of the HNO_3 concentration and temperature) which would result in zero volatilization of Ru (Figure 3-2).

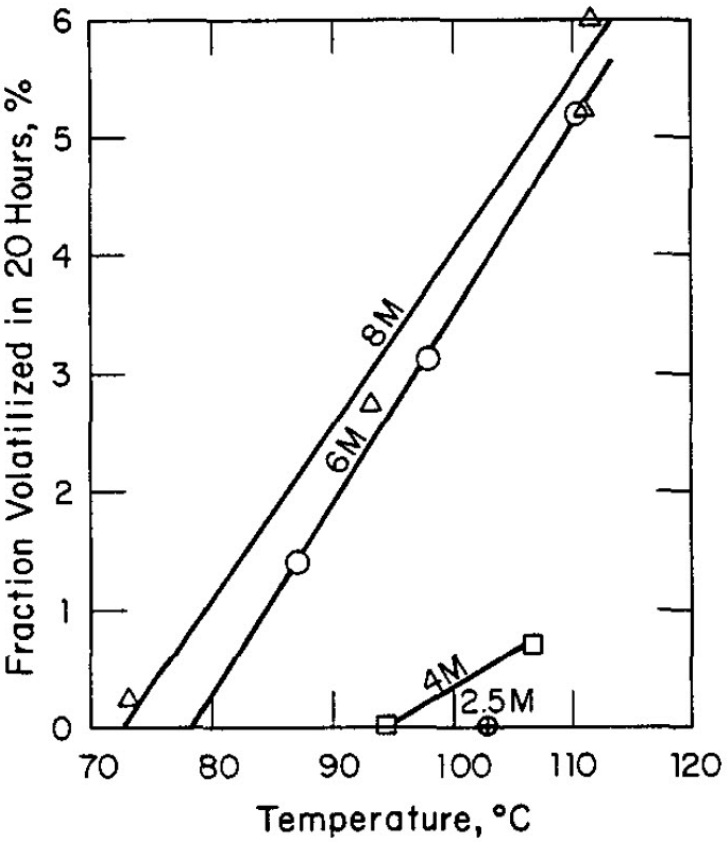

Figure 3-1. Volatilization of Ru from Heated HNO_3 Solutions

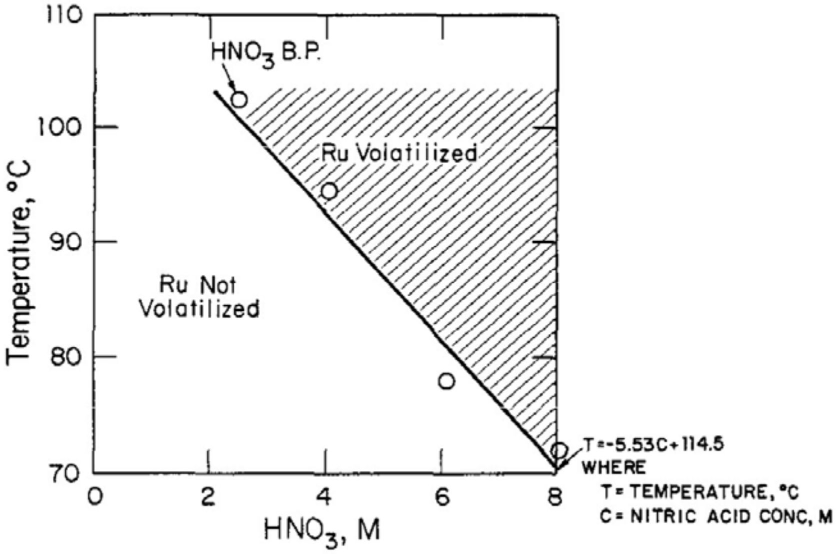

Figure 3-2. Ru Volatility from HNO_3 Solutions

The operating region was defined by a linear regression line. If no reducing agents are present in the dissolving solution, the HNO_3 concentration and temperature must be below the line to prevent the volatilization of Ru.

Ondrejcin[13] also evaluated the use of reducing agents to prevent the volatilization of RuO_4 during SNF dissolution. When present, suitable reducing agents favor the formation of RuNO nitro complexes over RuNO nitrato complexes; the RuNO nitro complexes are oxidized to volatile RuO_4 at a much slower rate. Ondrejcin tested the use of nitric oxide (NO), nitrogen dioxide (NO_2), and nitrite ion (NO_2^-) as reducing agents for application in H-Canyon dissolvers. Each of the nitrogen oxides is compatible with the downstream solvent extraction process and would be expected as a product in most dissolutions where Al metal is present. At equilibrium, it would not matter whether NO, NO_2, or NO_2^- is added to the solution since the chemical equilibria in HNO_3 involves all three species (equations 3 and 4).

$$3NO_2 + H_2O \rightleftharpoons 2H^+ + 2NO_3^- + NO \tag{3}$$

$$2HNO_2 \rightleftharpoons H_2O + NO_2 + NO \tag{4}$$

In Ondrejcin's work, it appeared that NO_2^- was more efficient in reducing Ru volatilization than NO_2. The use of NO_2^- with an air purge was about equivalent to NO with an inert gas sparge. Nitric oxide was not recommended for use in an H-Canyon dissolver due to its reaction with O_2 (in air) to form NO_2. Recommended addition rates to eliminate Ru volatilization are provided in Table 3-1 for a dissolving solution containing 8 M HNO_3 at boiling. Less oxidizing conditions would require a smaller amount of the reducing agent.

Table 3-1. Addition Rates for Reducing Agents to Eliminate Ru Volatilization

Reducing Agent	Addition Rate (mol/h/mol Ru)
NO_2^-	1.4
NO_2	4.8

4.0 Assessments of MTR and HFIR Dissolution Flowsheets

4.1 Potential for ^{106}Ru Release from an H-Canyon Dissolver

The flowsheets developed for the dissolution of MTR and HFIR fuels recommended the use of 5 to 7.5 M HNO_3 containing 0.002 M Hg at the boiling point of the dissolving solution.[27,28] The boiling point of pure 7.5 M HNO_3 is approximately 110 ˚C.[29] Therefore, if short cooled fuel is dissolved in H-Canyon using the MTR or HFIR flowsheets, the combination of the initial HNO_3 concentration and temperature of the solution would be well within the region of Figure 3-2 where Ru is volatilized. However, if the dissolution of Al metal is incomplete, the volatilization of RuO_4 is minimized due to the generation of gases (e.g., NO_2 and NO) during Al dissolution which serve as reducing agents (equations 5 and 6).

$$Al + 6HNO_3 \rightarrow Al(NO_3)_3 + 3NO_2 + 3H_2O \tag{5}$$

$$Al + 4HNO_3 \rightarrow Al(NO_3)_3 + NO + 2H_2O \tag{6}$$

The NO_2 and NO gases produced during Al dissolution are in equilibrium with HNO_2 (equation 4) which favors the formation of RuNO nitro complexes in the solution. Ruthenium(III) nitrosyl nitro complexes are more stable than RuNO nitrato complexes and as a result are less likely to oxidize to volatile RuO_4.

As HNO_3 is consumed during fuel dissolution, the driving force to oxidize Ru is reduced. Based on the boiling point of pure HNO_3 solutions (Figure 4-1),[29] the concentration must be reduced to approximately 2.2 M before the dissolving conditions would be in the region of Figure 3-2 where Ru volatilization does not occur. However, since the actual boiling point of the solution is elevated due to the dissolved salt content, the HNO_3 concentration must be reduced to a value lower than 2.2 M to prevent Ru volatilization.

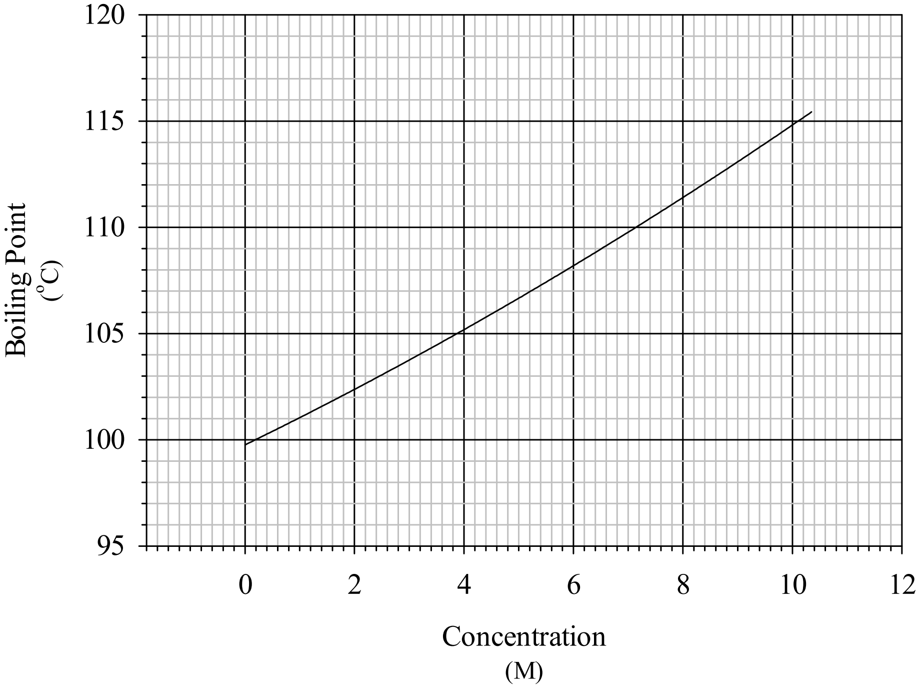

Figure 4-1. Boiling Point of Pure HNO_3

4.2 Estimated Release of ^{106}Ru from H-Canyon during HFIR Fuel Dissolution

A bounding estimate for the amount of ^{106}Ru which could be released from H-Canyon during the dissolution of short-cooled HFIR fuel was calculated for cores with cooling times from approximately 4 to 10 years. The activity of ^{106}Ru in a HFIR core was calculated (by Oak Ridge National Laboratory (ORNL) personnel) using SCALE Version 5.1[30] from a recently discharged core (Table 4-1). The thermal power and U isotopic concentrations were compared with data in an Appendix A for a HFIR core stored in the L-Area basin.[31] Comparison of the data showed good consistency (Table 4-2); therefore, the ^{106}Ru activity calculations should be representative of all HFIR cores which operated at the same thermal power. Each HFIR core is operated for one cycle of approximately 24 days at 85 MW, and then removed from the reactor.[31]

Table 4-1. Activity of ^{106}Ru per HFIR Core Following Discharge

Time Following Discharge (y)	^{106}Ru Activity (Ci)
4.12	813
5.12	411
6.12	208
7.37	89
8.37	45
9.37	23
10.37	11

Table 4-2. U Isotopic Concentrations of HFIR Fuel

Data Source	**SCALE 5.1**	**Appendix A**
HFIR Core	**I-480, O-481**	**I-358, O-358**
Thermal Power (MWd)	**2112.4**	**2108.7**
Isotope	**Composition**	**Composition**
	(wt %)	**(wt %)**
U-234	1.1	1.1
U-235	85.0	85.6
U-236	6.6	6.6
U-238	7.3	6.7
U-total	100	100

The amount of ^{106}Ru volatilized from an H-Canyon dissolver can be estimated from Figure 3-1 as a function of the solution temperature and the HNO_3 concentration. The temperature of the 6.4D dissolver solution following the completion of the dissolution of each charge for three batches of HFIR fuel are provided in Table 4-3. The solution temperature was nearly constant during the dissolution of each charge due to the offsetting effects of the decrease in the HNO_3 concentration and an increase in the dissolved salt content. For conservatism, the highest temperature recorded (110 ˚C) was used to calculate a bounding value for the ^{106}Ru volatilized following the dissolution of a charge of HFIR fuel.

Table 4-3. 6.4D Dissolver Solution Temperature following HFIR Fuel Dissolution

Charge	**Batch 1**	**Batch 2**	**Batch 3**	**Charge Number**	
Number	**Temp**	**Temp**	**Temp**	**Average**	**Std Dev**
	(˚C)	**(˚C)**	**(˚C)**	**(˚C)**	**(˚C)**
1	108.6	107.7	108.1	108.1	0.5
2	108.4	108.4	107.9	108.2	0.3
3	108.2	107.8	107.6	107.9	0.3
4	109.2	107.7	107.6	108.2	0.9
5	110.0	108.1	107.9	108.7	1.2
Average	108.9	107.9	107.8	108.2	0.7
Std Dev	0.7	0.3	0.2		

The HNO_3 concentration following the dissolution of each HFIR core can be estimated based on the initial HNO_3 concentration and the amounts of Al and U_3O_8 dissolved using the stoichiometry given by equations 7 and 8.[20,28]

$$\mathrm{Al} + 3.75\mathrm{HNO_3} \xrightarrow{\mathrm{Hg^{++}}} \mathrm{Al(NO_3)_3} + 0.225\mathrm{NO} + 0.15\mathrm{N_2O} + 0.11\mathrm{N_2} + 1.9\mathrm{H_2O} \tag{7}$$

$$\mathrm{U_3O_8} + 8\mathrm{HNO_3} \rightarrow 3\mathrm{UO_2(NO_3)_2} + 2\mathrm{NO_2}\ \ 4\mathrm{H_2O} \tag{8}$$

If the initial HNO_3 concentration used for the dissolution of the first HFIR core is 7.5 M, the acid concentration after the dissolution of each of five successive cores in shown in Table 4-4. The Al concentrations are based on each HFIR core containing 129.1 kg of Al,[32] and a total Al mass of 6.3 kg for the inner and outer carriers.[33] A 14,000 L working volume of the H-Canyon 6.4D dissolver was used as the basis for evaluating the ^{106}Ru volatilization.[33]

Table 4-4. HNO_3 Concentration during HFIR Fuel Dissolution

HFIR Core/Charge	HNO_3
	(M)
1	6.1
2	4.8
3	3.4
4	2.1
5	0.7

The fractions of ^{106}Ru volatilized (Table 4-5) during the dissolution of five successive charges of HFIR fuel (i.e., five cores) were estimated using Figure 3-1 at a temperature of 110 ˚C using the HNO_3 concentrations calculated for the dissolving solution at the completion of each charge (Table 4-4). Implicit in the use of the data in Figure 3-1 are the assumptions that all Al in a charge has dissolved (as the fraction of ^{106}Ru volatilized was based on experiments in which Al metal was not present in the solution[13]) and the solution is heated for an additional 20 h.

Table 4-5. Fraction of ^{106}Ru Volatilized from the 6.4 D Dissolver

HFIR Core/Charge	Fraction Volatilized in 20 h
	(%)
1	5.1
2	2.9
3	0.5
4	0
5	0

The total amount of ^{106}Ru which is volatilized from the 6.4D dissolver will not be released from H-Canyon. Ruthenium tetraoxide generated in the dissolver will be reduced in the offgas system to RuO_2 which will deposit on stainless steel surfaces in offgas piping.[21] Kyser estimated the decontamination factor (DF) across the offgas system for the 30 Ci ^{106}Ru release which occurred in 1978 from an F-Canyon dissolver[12] was in the range of 30 to 50.[34] If a DF of 30 is assumed across the 6.4D Dissolver offgas system, the^{106}Ru activity released during the dissolution of 5 HFIR cores can be estimated using the calculated activity in Table 4-1 for a single core and the release fractions given in Table 4-5. The ^{106}Ru releases as a function of the fuel cooling time and the charge number are provided in Table 4-6 and are shown graphically in Figure 4-2.

Table 4-6. Bounding Cumulative Release of ^{106}Ru during HFIR Fuel Dissolution

HFIR Fuel Charge No.	Cumulative ^{106}Ru Release (mCi)						
	Time After Fuel Discharge (y)						
	4.12	5.12	6.12	7.37	8.37	9.37	10.37
1	1382	699	354	151	76	39	20
2	2913	1474	746	317	161	81	41
3	3305	1673	847	360	182	92	47
4	3305	1673	847	360	182	92	47
5	3305	1673	847	360	182	92	47

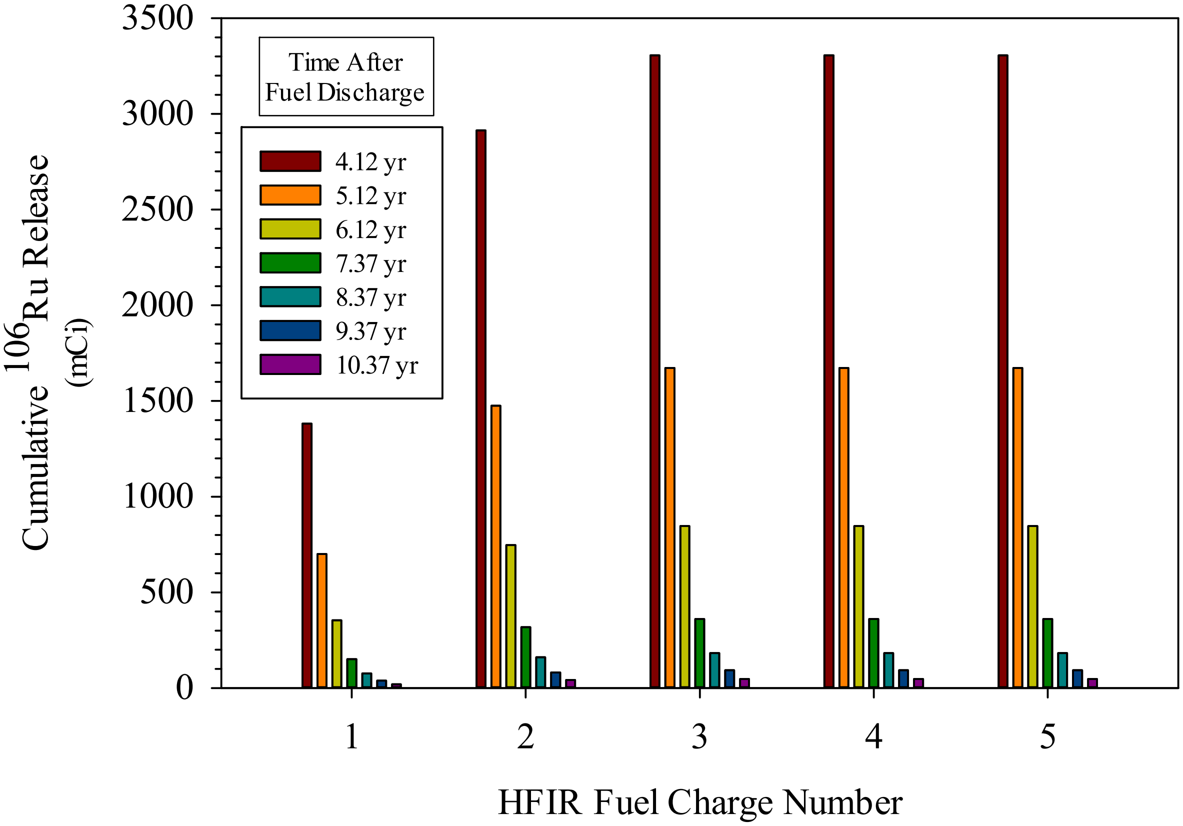

Figure 4-2. Bounding Cumulative Release of ^{106}Ru during HFIR Fuel Dissolution

5.0 Flowsheet Modifications for Dissolution of Short-cooled MTR and HFIR Fuels

The dissolution of short-cooled MTR or HFIR fuels is not anticipated in the next several years; however, if a decision is made to dissolve these fuels using existing flowsheets prior to the decay of the ^{106}Ru activity, volatilization of RuO_4 from the dissolver and subsequent release from H-Canyon would occur. To address this issue, revised flowsheets would need to be demonstrated using conditions (i.e., HNO_3 concentrations and temperatures) which preclude the volatilization of Ru from the dissolver (Figure 3-2). Dissolution of SNF using lower temperatures and concentrations of HNO_3 could increase the concentration of H_2 in the offgas; although, less aggressive dissolution conditions would be expected to generate less total offgas. During the dissolution of some metals (e.g., Be), lower acidities increase the generation rate of H_2 which is consistent with the dissolution mechanism shifting away from increased production of nitrogen oxides by nitrate (NO_3^-) oxidation at high acid concentrations to a typical metal/acid reaction at low acid concentrations.[35] Measuring the H_2 and total offgas generation rates during laboratory-scale dissolutions of Al coupons would be required to demonstrate that the H_2 concentration in the offgas from an H-Canyon dissolver does not exceed 60% of the lower flammability limit (LFL).[36]

Using lower temperatures and concentrations of HNO_3 to dissolve SNF would have a deleterious effect on the rate of dissolution and likely the number of MTR bundles or HFIR cores which can be dissolved in a batch. In the absence of dissolved Al in the solution, the dissolving rate of U-Al alloy is directly proportional to the concentration of dissociated HNO_3 as shown in Figure 5-1.[37]

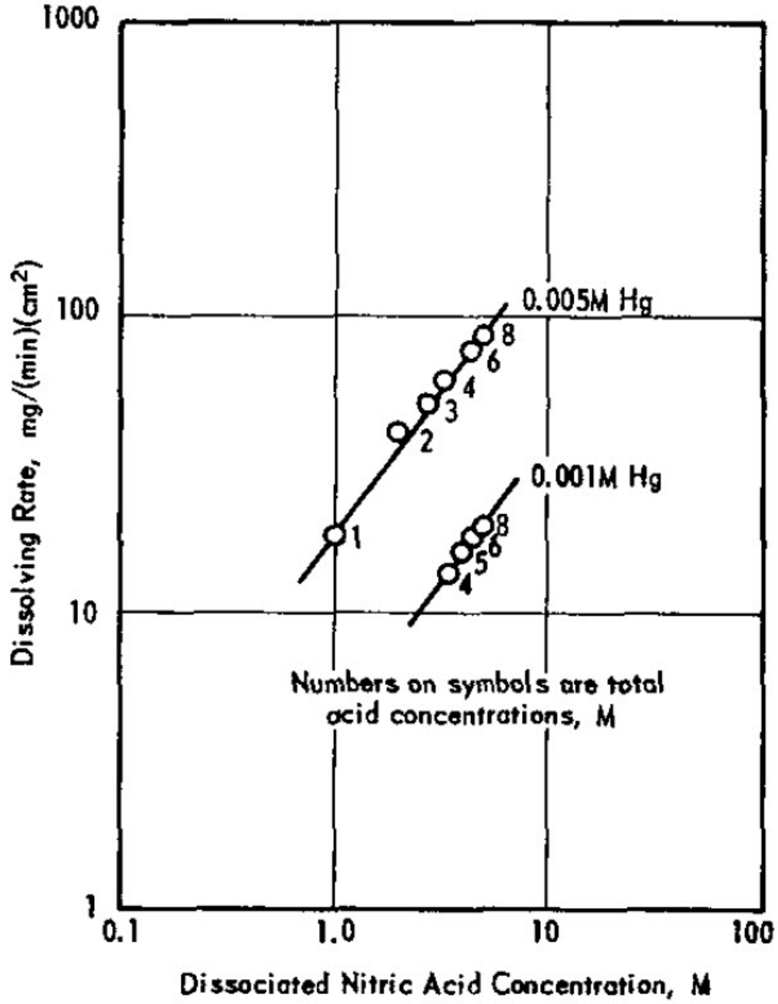

Figure 5-1. Effect of HNO_3 Concentration on the Dissolution Rate of Unirradiated 16U-Al Alloy

The temperature of the solution generally only has a weak influence on the dissolution rate of Al.[38] However, it may be difficult to operate at temperatures which are slightly below the region of temperatures (and nitic acid concentrations) which result in the volatilization of Ru (Figure 3-2). At low temperatures (<50 ˚C), the dissolution rate of Al is very slow. The dissolution rate increases moderately until a critical temperature is reached. At this point the dissolution rate increases very rapidly and may heat the solution to the boiling point. During the development of a dissolution flowsheet for HFIR control plates at the ORNL, the temperature at which rapid dissolution started varied from 60 to 85 ˚C.[39] To evaluate if the heat generated during a HFIR or MTR fuel dissolution could heat the solution in an H-Canyon dissolver to a temperature approaching the boiling point, it would likely be necessary to develop a heat transfer model for the vessel. However, if the heat of reaction results in the solution reaching a temperature (in the absence of reducing gases) above the dividing line in Figure 3-2, Ru volatilization from the dissolver will occur.

A reduction in the starting HNO_3 concentration for a HFIR or MTR fuel dissolution to prevent Ru volatilization will have impact on the amount of Al which can be dissolved per batch of fuel. The dissolution of one mole of Al requires nominally 3.75 moles of HNO_3 (equation 7).[20] If the starting acid concentration for a HFIR fuel dissolution is reduced from 7.5 to 5 M (in combination with a lower solution temperature) to prevent Ru volatilization, then only three HFIR cores can be dissolved before the lower limit on HNO_3 concentration (0.5 to 1 M) is reached. This scenario is based on a 14,000 L working volume in the 6.4 D dissolver. Depending on the extent of evaporation losses, it may be possible to add concentrated HNO_3 to the dissolver and dissolve additional fuel.

Another flowsheet strategy for the dissolution of HFIR or MTR fuel to minimize the volatilization of Ru is to ensure that a portion of the fuel remains undissolved prior to charging the next core. If Al is present in the dissolver, its dissolution will generate NO_2 and NO gases (equations 5 and 6) which serve as reducing agents. The NO_2 and NO gases are in equilibrium with HNO_2 (equation 4) which favors the formation of RuNO nitro complexes in solution.[7] This flowsheet strategy would take advantage of the stability of the RuNO nitro complexes which are more resistant to Ru oxidation than the RuNO nitrato complexes. If a HFIR core dissolves before termination of dissolver heating, Al is no longer present to generate NO_2 and NO gases. As these gases are removed from the dissolver, the equilibrium concentration of HNO_2 decreases. The high HNO_3/NO_3^- concentrations in the dissolver would now favor the formation of the RuNO nitrato complex which is easy to oxidize to the volatile RuO_4 species (Table 1-1).

Implementation of this strategy is made more difficult by the need to stop the dissolver and manually probe the HFIR insert wells to determine if fragments of the fuel remain. However, the dissolution times for the HFIR cores should be very similar given the same dissolving conditions which should allow prediction of the time required to achieve the desired extent of dissolution. The maximum fragment height of HFIR fuel that can be present in the insert wells must be controlled to ensure that the predicted H_2 concentration in the offgas does not exceed 60% of the calculated LFL.[36] Maximum fragment heights were calculated by Daniel and Rudisill as a function of the Al concentration for a HFIR core. The calculations showed that it was theoretically possible to charge at least two HFIR cores to the dissolver after reaching an Al concentration of 0.47 M. At Al concentrations greater than or equal to this value, it would not be necessary to control the height of the fuel fragments prior to charging another HFIR core.[33]

One potential issue to consider prior to implementing a dissolution strategy which depends on incomplete dissolution of a HFIR core to minimize the volatilization of Ru is the potential for volatilization during the dissolution of the final charge. Complete dissolution of all fuel fragments would be required before the dissolved fuel is transferred from the dissolver. Ending the dissolver heating cycle as soon as possible after complete dissolution would minimize the oxidation of Ru; however, the HNO_3 concentration and temperature would probably be in the operating region where Ru volatilization does not occur. Regardless, predicting the endpoint of each dissolution cycle based on previous history or available instrumentation would be necessary.

6.0 References

1. R. T. Jubin, D. M. Strachan, G. Ilas, B. B. Spencer, and N. R. Soelberg, *Radioactive Semivolatiles in Nuclear Fuel Reprocessing*, INL/EXT-14-33122, Idaho National Laboratory, Idaho Falls, ID (September 2014).
2. R. M. Wallace, DPST-64-483, E. I. du Pont de Nemours & Co., Aiken, SC (October 1964).
3. E. M. Baum, H. D. Know, and T. R. Miller, *Chart of the Nuclides*, Knolls Atomic Power Laboratory, KAPL Inc., Niskayuna, NY, 16th edition – Revised to 2002.
4. G. G. J. Boswell and S. Soentono, *Ruthenium Nitrosyl Complexes in Nitric Acid Solutions*, J Inorg Nucl Chem, 43, 1625-1632 (1981).
5. P. G. M. Brown, *Nitro Complexes of Nitrosylruthenium*, J. Inorg Nucl Chem, 13, 73-83 (1960).
6. L. Maya, *Ruthenium(IV) in Nitric Acid Media*, J. Inorg Nucl Chem, 41, 67-71 (1979).
7. P. G. M. Brown, J. M. Fletcher. C. J. Hardy, J. Kennedy, D. Scargill, A. G. Wain, and J. L. Woodhead, *The Significance of Certain Complexes of Ruthenium, Niobium, Zirconium, and Uranium in Plant Processes*, in Progress in Nuclear Energy Series III, Process Chemistry, Vol 3, F. R. Bruce, J. M. Fletcher, and H.H. Hyman, Eds, Pergamon Press, New York, NY (1961).

8. K. J. Schneider, Ed., *Waste Solidification Program Volume I. Process Technology – Pot Spray, and Phosphate Glass Solidification Processes*, BNWL-1073, Battelle Memorial Institute Pacific Northwest Laboratories, Richland, WA (August 1969).
9. P. W. Cains, K. C. Yewer, and S. Waring, *Volatilization of Ruthenium, Caesium and Technetium from Nitrate Systems in Nuclear Fuel Processing and Waste Solidification*, Radiochim Acta, 56, p 99-104 (1992).
10. J. A. Dean, Ed., *Lange's Handbook of Chemistry*, 13th ed., McGraw-Hill Book Company, New York, NY, p 4-102 (1985).
11. Y. Koda, *Boiling Points and Ideal Solutions of Ruthenium and Osmium Tetraoxides*, J Chem Soc Chem Comm, Issue 17, p 1347-1348 (1986).
12. D. F. Chostner, *Release of Ruthenium from 6.1D Dissolver Offgas Stack*, SI-78-11-134, E. I. du Pont de Nemours & Co., Aiken, SC (November 14, 1978).
13. R. S. Ondrejcin, *Volatile Ruthenium Chemistry, An Answer to RTA 925-S*, DPST-80-254, E. I. du Pont de Nemours & Co., Aiken, SC (January 31, 1980).
14. T. F. Severynse, *Atmospheric Release of ^{106}Ru from 221-H Canyon*, SI-82-3-20, E. I. du Pont de Nemours & Co., Aiken, SC (March 23, 1982).
15. T. F. Severynse, *High Activity in 291-H Stack Releases*. E. I. du Pont de Nemours & Co., Aiken, SC (February 19, 1981).
16. W C. Perkins and R. M. Wallace, *Plant Problems during Dissolution of Uranium Metal Scrap*, DPST-77-204, E. I. Du Pont de Nemours & Co., Aiken, SC (January 10, 1977).
17 E. R. Norton, *Release of ^{103}Ru-^{106}Ru to the Atmosphere*, SEP(E)CAN-173-2, E. I. du Pont de Nemours & Co., Aiken, SC (March 29, 1972).
18. G. Hayes and D. S. Monk, *Stack Releases of Ruthenium to Environment*, E. I. du Pont de Nemours, Aiken, SC (September 4, 1968).
19. J. R. Lacher, J. D. Salzman, and J. D. Park, *Dissolving Uranium in Nitric Acid*, Ind Eng Chem, 53, p 282 (1963).
20. M. L. Hyder, W. C. Perkins, M. C. Thompson, G. A. Burney, E. R. Russell, H. P. Holcomb, and L. F. Landon, *Processing of Irradiated Enriched Uranium Fuels at the Savannah River Plant*, DP-1500, E. I. du Pont de Nemours & Co., Aiken, SC (April 1979).
21. P. W. Cains and S. J. Barnes, *Deposition of volatilized ruthenium on stainless steels*, J Nucl Mater, 186, p 83-86 (1991).
22. F. Mousset, C. Eysseric, and F. Bediour, *Studies of dissolutions of ruthenium metal, oxide and mixed compounds*, ATALANTE 2004, Nîmes, France (June 21-25, 2004).
23. E. R. Norton, *Release of ^{103}Ru and ^{106}Ru to Atmosphere*, SEP(E)CAN-173-2, E. I. du Pont de Nemours & Co., Aiken, SC (March 29, 1972).
24. L. H. Meyer, *Absorber for Volatile Ruthenium*, DP-223, E. I. du Pont de Nemours & Co., Aiken, SC (1957).
25. H. E. Shook, RTA 1184-S: *Improved Ruthenium Absorber Final Report*, E. I. du Pont de Nemours & Co., Aiken, SC (December 20, 1984).
26 B. J. Newby and V. H. Barnes, *Volatile Ruthenium Removal from Calciner Off-gas using Solid Sorbents*, ICP-1078, Idaho National Engineering Laboratory, Idaho Falls, ID (July 1975).
27. W. E. Daniel, T. S. Rudisill, and P. E. O'Rourke, *Dissolution of Material Test Reactor Fuel in an H-Canyon Dissolver*, SRNL-STI-2016-00725, Rev 1, Savannah River National Laboratory (May 2018).
28. W. E. Daniel, T. S. Rudisill, P. E. O'Rourke, and N. S. Karay, *Dissolution Flowsheet for High Flux Isotope Reactor Fuel*, SRNL-STI-2016-00485, Rev 1, Savannah River National Laboratory, Aiken, SC (December 2017).
29. *200 Areas Process Guidebook*, DPSOP 250, Rev 1, E. I. du Pont de Nemours & Co., Aiken, SC (January 1990).
30. *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluations*, ORNL/TM-2005/39, Version 5.1, Vols. I–III, November 2006. Available from Radiation Safety Information Computational Center at Oak Ridge National Laboratory as CCC-732.

31. Appendix A, Spent Nuclear Fuel Acceptance Criteria, High Flux Isotope Reactor, DOESRAAD-10-095.1, United States Department of Energy, Washington, DC (September 10, 2010).
32. G. H. Sykes and R. W. Zeyfang, *High Flux Reactor (HFIR) Fuel Processing*, Test Authorization No. 2-685, E. I. du Pont de Nemours & Co., Aiken, SC (May 9, 1969).
33. W. E. Daniel and T. S. Rudisill, *Analysis of Maximum Allowable Fragment Heights during Dissolution of High Flux Isotope Reactor Fuel in an H-Canyon Dissolver*, SRNL-STI-2017-00372, Savannah River National Laboratory, Aiken, SC (July 2017).
34. E. A. Kyser, *Ruthenium Volatilization in FB-Line Processes*, WSRC-TR-92-00387. Westinghouse Savannah River Company, Aiken, SC (July 1992).
35. T. S. Rudisill and M. L. Crowder, *Dissolution of FB-Line Residues Containing Beryllium Metal*. WSRC-TR-2005-00042, Rev 1, Westinghouse Savannah River Company, Aiken, SC (October 2005).
36. NFPA® 69, *Standard on Explosion Prevention Systems*, 2008 Edition, NFPA, Quincy, MA.
37. C. S. Schlea, *The Dissolution of Uranium-Aluminum Alloy*, DP-629, E. I. du Pont de Nemours & Co., Aiken, SC (September 1961).
38. R. W. Rice and D. V. Sarode, *Mercury-Catalyzed Dissolution of Aluminum in Nitric Acid*, Ind Eng Chem Res, 40, 1872-1878 (2001).
39. J. C. Posey, *Dissolution of HFIR Control Plates*, ORNL/TM-9025, Oak Ridge National Laboratory, Oak Ridge, TN (March 1984).

Distribution

timothy.brown@srnl.doe.gov
alex.cozzi@srnl.doe.gov
david.crowley@srnl.doe.gov
a.fellinger@srnl.doe.gov
samuel.fink@srnl.doe.gov
nancy.halverson@srnl.doe.gov
erich.hansen@srnl.doe.gov
john.mayer@srnl.doe.gov
daniel.mccabe@srnl.doe.gov
gregg.morgan@srnl.doe.gov
frank.pennebaker@srnl.doe.gov
amy.ramsey@srnl.doe.gov
william.ramsey@srnl.doe.gov
luke.reid@srnl.doe.gov
michael.stone@srnl.doe.gov
boyd.wiedenman@srnl.doe.gov
bill.wilmarth@srnl.doe.gov

nicholas.bridges@srnl.doe.gov
kim.crapse@srnl.doe.gov
gene.daniel@srnl.doe.gov
jonathan.duffey@srnl.doe.gov
harris.eldridge@srnl.doe.gov
daniel.jones@srnl.doe.gov
nicholas.karay@srnl.doe.gov
eddie.kyser@srnl.doe.gov
kevin.mccann@srnl.doe.gov
robert.pierce@srnl.doe.gov
lindsay.roy@srnl.doe.gov
tracy.rudisill@srnl.doe.gov
john.scogin@srnl.doe.gov
thomas.shehee@srnl.doe.gov
tara.smith@srnl.doe.gov
christopher.south@srnl.doe.gov

richard.burns@srs.gov
steven.brown@srs.gov
kenneth.burrows@srs.gov
bill.clifton@srs.gov
brett.clinton@srs.gov
perry.levin@srs.gov
michael.lewczyk@srs.gov
john.lint@srs.gov
omar.rodriquez@srs.gov
gdavid.sanders@srs.gov
jeffrey.schaade@srs.gov
james.therrell@srs.gov
timothy.tice@srs.gov
kevin.usher@srs.gov
hayley.williams@srs.gov
stephen.yano@srs.gov

Records Administration (EDWS)