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A Gamma Radiolysis Study of $\text{UO}_2\text{F}_2 \cdot 0.4\text{H}_2\text{O}$ Using Spent Nuclear Fuel Elements from the High Flux Isotope Reactor

January 2002

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Nuclear Science and **Technology** Division

**A GAMMA RADIOLYSIS STUDY OF $\text{UO}_2\text{F}_2 \cdot 0.4\text{H}_2\text{O}$ USING
SPENT NUCLEAR FUEL ELEMENTS FROM THE
HIGH FLUX ISOTOPE REACTOR**

**A. S. Icenhour
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ABSTRACT

The development of a **standard** for the **safe, long-term storage** of **^{233}U -containing** materials resulted in the identification of several needed **experimental** studies. These studies were largely related to the potential for the generation of **unacceptable** pressures or the **formation** of deleterious **products** during storage of uraniumoxides. The **primary concern** was that **these conditions could occur as a result of the radiolysis** of residual **impurities—specifically fluorides** and water-by the high radiation fields associated with **$^{233}\text{U}/^{232}\text{U}$ -containing** materials.

This **report documents** the results from a gamma radiolysis experiment in which $\text{UO}_2\text{F}_2 \cdot 0.4\text{H}_2\text{O}$ was loaded in helium. This **experiment** was performed using spent **nuclear** fuel elements from the **High Flux Isotope Reactor** as the gamma source and was a follow-on to experiments **conducted** previously.

It was found that upon gamma irradiation, the $\text{UO}_2\text{F}_2 \cdot 0.4\text{H}_2\text{O}$ released O_2 with an initial $G(\text{O}_2) = 0.01$ molecule $\text{O}_2/100$ eV and that some of the **uranium** was **reduced** from U(VI) to U(IV). The high total dose achieved in the SNF elements was **sufficient** to reach a damage limit for the $\text{UO}_2\text{F}_2 \cdot 0.4\text{H}_2\text{O}$. This **damage limit, measured in terms of the amount of the U(IV) produced, was found to be about 9 wt %**.

1. INTRODUCTION

The development of a standard for the safe, long-term storage of ^{233}U -containing materials' resulted in the identification of several **needed experimental** studies. These studies were largely related to the potential for the generation of unacceptable pressures or the **formation** of deleterious products during storage of uranium oxides. The primary concern **was** that these conditions **could occur** as a result of the radiolysis of residual **impurities**—**specifically** fluorides and water—by the high radiation fields associated with $^{233}\text{U}/^{232}\text{U}$ -containing materials.

The gamma radiolysis of **fluoride** impurities in uranium oxides has been studied previously, and the data **from** that study confirmed that such radiolysis will not result in the **overpressurization** of storage containers or the **formation** of deleterious products.² The radiolysis experiments in the previous study used both a ^{60}Co source and High Flux Isotope Reactor (HFIR) spent nuclear fuel (SNF) elements. The experiments with HFIR SNF elements involved a small nickel sample vessel that was inserted into the SNF element (below ~6 m of water) and was connected by a tube to a Bourdon pressure gage. The pressure gage was typically read during each shift by HFIR operations personnel. To **obtain** more accurate data and additional data points, an irradiation rig with a pressure transducer was used for the **current** measurements. This rig permitted irradiation of up to four samples simultaneously in an SNF element and provided the means for continuous collection of data with a **computerized** data acquisition system.

This report documents an experiment in which uranyl fluoride (UO_2F_2) was subjected to gamma radiolysis using the new irradiation rig. The work described here served as a follow-up irradiation experiment to the work reported in ref. 2.

2. EXPERIMENTAL METHODS

The HFIR irradiation facility, sample container, data acquisition system, irradiated material, and sampling and analysis techniques are described in this section.

2.1 SNF IRRADIATION FACILITY AT HFIR

Samples can be irradiated in the SNF pool at HFIR by inserting them inside SNF elements (Fig. 2.1), which are cylindrical with a hollow center. In its storage position in the SNF pool, a cadmium sleeve inside the hollow region of the element absorbs neutrons. Hence, the hollow region of the fuel element primarily provides a gamma field for irradiation. The neutron flux in this region is about $100 \text{ neutrons}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$. The contribution of neutrons to radiation damage is negligible when compared with the very large gamma field. Variable gamma-radiation fields are available based on the decay times of the elements. The facility provides a nominal 7.62-cm (3-in.) opening for placing the samples inside the SNF elements. Reported exposure rates range from about 10^8 R/h down to 10^7 R/h or lower, depending on the time elapsed since discharge of the SNF from the reactor. The gamma-ray energy spectrum for a HFIR SNF element 1 d after discharge from the reactor is shown in Table 2.1.³

ORNL Photo 3067-2000



Fig. 2.1. SNF elements in the HFIR SNF pool.

Table 2.1. Gamma-ray energy spectrum for HFIR SNF element 1 d after discharge from reactor^a

Energy group	Upper bound (MeV)	Average energy in group (MeV)	Percentage of total energy in group
1	0.02	0.01	0.44
2	0.03	0.025	0.44
3	0.045	0.0375	0.89
4	0.07	0.0575	0.56
5	0.1	0.085	1.04
6	0.15	0.125	2.66
7	0.3	0.225	5.66
8	0.45	0.375	4.48
9	0.7	0.575	26.94
10	1	0.85	26.82
11	1.5	1.25	6.89
12	2	1.75	21.06
13	2.5	2.25	0.88
14	3	2.75	1.24
15	4	3.5	0.01

Average energy = 0.93 MeV

^aD. F. Williams, G. D. Del Cul, and L. M. Toth, A *Descriptive Model of the Molten Salt Reactor Experiment After Shutdown: Review of FY 1995 Progress*, ORNL/TM-13 142, Lockheed Martin Energy Research Corp., Oak Ridge National Laboratory, Oak Ridge, Tennessee, January 1996.

Kohring⁴ measured the exposure rate inside HFIR SNF elements as a function of (1) axial location within the element and (2) time elapsed since discharge from the reactor. These measurements were made for elements that had been operated at 100 MW for 21.5 d (equivalent to a burnup of 2150 MWd). Figure 2.2 shows the peak exposure rate as a function of time since shutdown.⁴ This exposure rate can be corrected for the axial location of the sample by use of Fig. 2.3, which is adapted from ref. 4. In 1987, HFIR's operating power was reduced to 85 MW. This reduced power level necessitated an adjustment in the reported exposure rates, which was calculated by Kohring.⁵ Kohring used the ORIGEN computer code to determine the correction factors that needed to be applied to the measured exposure rates to account for the decreased operating power. These correction factors can also be calculated by use of the Borst-Wheeler formula,⁶ which has been shown to be in good agreement with the correction factors reported by Kohring.⁷ The correction factor is calculated by

$$CF(t) = 0.85 \frac{[t^{-0.2} - (t + T)^{-0.2}]}{[t^{-0.2} - (t + 21.5)^{-0.2}]}, \quad (2.1)$$

where

$CF(t)$ = correction factor at time t after shutdown, dimensionless;

t = time since shutdown, d;

T = time of operation at 85 MW [= burnup (MWd)/85 MW], d

The factor 0.85 is simply the ratio of the new operating power level (85 MW) to the original operating power level (100 MW).

To evaluate the exposure rate of a given sample, the exposure-rate data provided by Kohring are adjusted on the basis of burnup of the element and the axial location of the sample. To evaluate radiolytic yields (i.e., the number of molecules of a species produced per amount of energy deposited in a material), the energy deposited in the irradiated material (i.e., the dose) must be known. Hence, the exposure rate, which is a measure of the amount of ionization produced in air by gamma or X rays, must be converted to a dose rate in the irradiated material. The method established in ASTM Standard E666-91 (see Appendix A) was used to perform this conversion.⁸

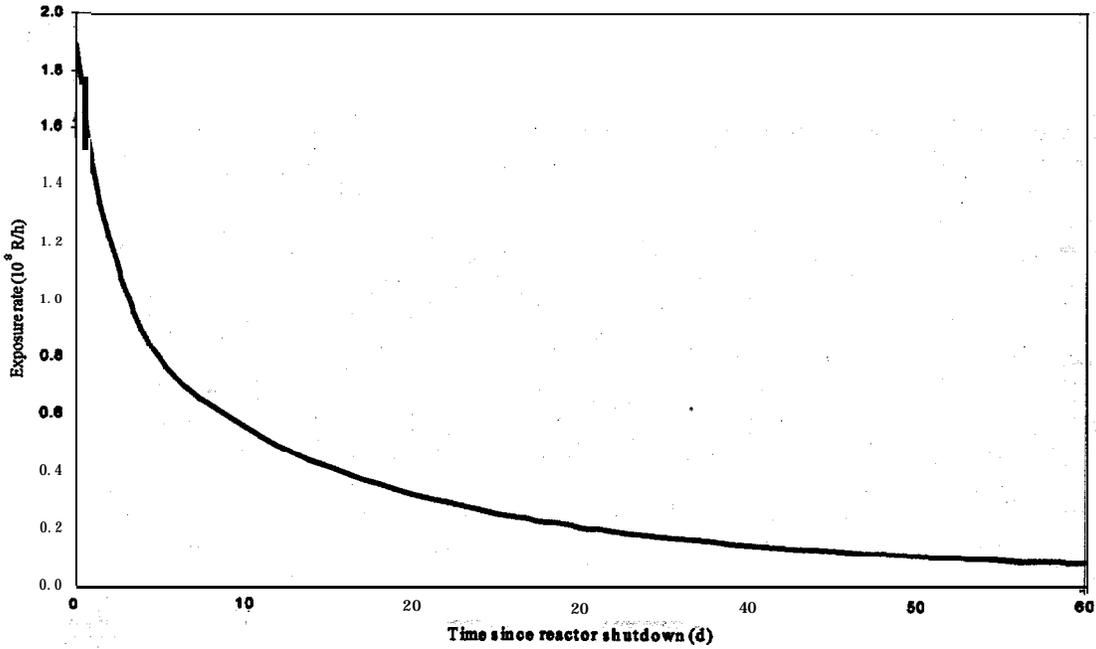


Fig. 2.2. Peak exposure rate in a HFIR SNF element as a function of time since reactor shutdown. (Adapted from ref. 4).

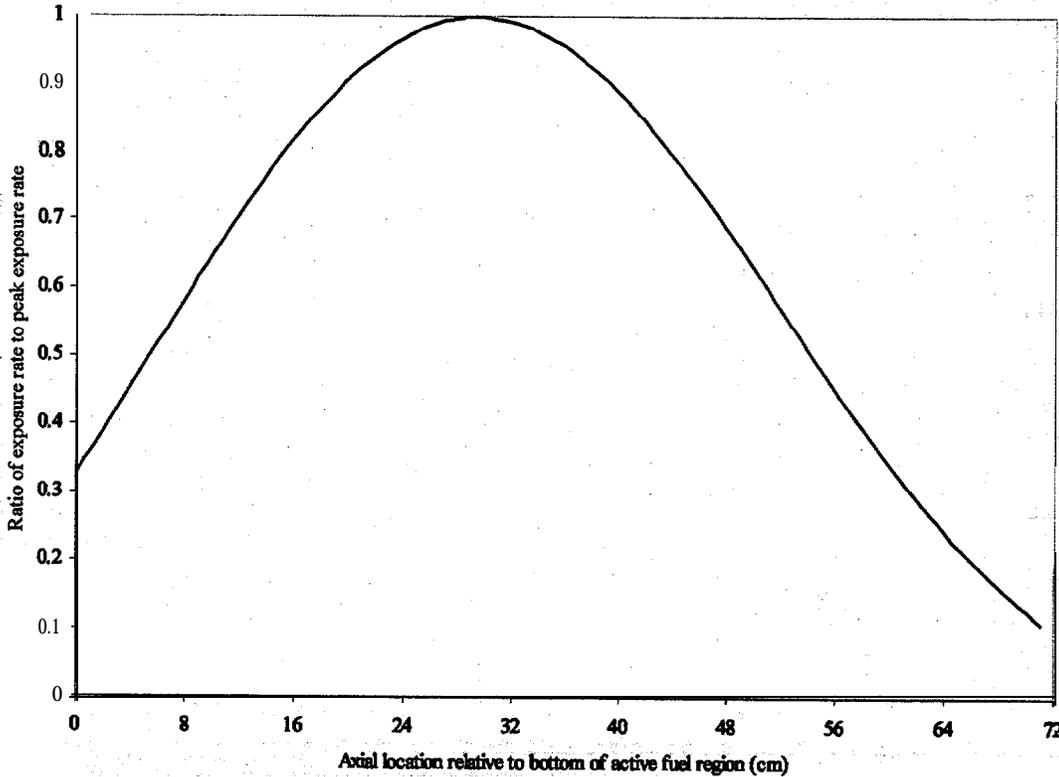


Fig. 2.3. Exposure rate relative to the peak exposure rate as a function of axial position in a HFIR SNF element. (Adapted from ref. 4).

2.2 SAMPLE CONTAINER

A multiple-radiation container was used for the simultaneous irradiation of up to **four samples** (Fig. 2.4). Small sample containers consisting of **1.27-cm (0.5-in.)-diam monel** tubing were placed inside an **outer container, which was fabricated from 8.9-cm (3.5-in.)-diam, 44-cm (17.19-in.)-long stainless steel pipe**. The outer container was closed at one end and had a **Conflat flange on the other end**. The flange had five penetrations, **four of which were used to connect the smaller inner containers to 0.318-cm (0.125-in.)-diam tubing (constructed of nickel or stainless steel, depending on the material to be irradiated)**. The **fifth penetration connected the void volume of the outer container to 0.318-cm (0.125-in.)-diam stainless steel tubing**. In each case, the **0.318-cm (0.125-in.)-diam tubing was about 6.1 m (20 ft) long and was connected to a pressure transducer and a valve**.

One of the sample containers (designated **HFIR-6-4**), which had a volume of **36.7 cm³**, was used in the experiment **described** in this report. (The other three sample containers were used for **the irradiation** of other materials and are reported on separately.) The sample was loaded in helium, and the outer container was **pressurized to 10 psig with helium before the experiment was transported to HFIR for insertion into an SNF element**.

2.3 DATA ACQUISITION SYSTEM

Sensotec[®] (Model FPA, 0-50 psia) pressure transducers were used to monitor the pressure within the sample containers. A **Kobold (KPK-30100, 30 in. Hg to 100 psig)** compound pressure transducer was used to monitor **the pressure in the outer container**. The **pressure transducers and valves for gas sampling (Fig. 2.5) were mounted to a bracket that was clamped to the edge of the HFIR SNF pool wall**. A computerized data acquisition system was used to record the pressure throughout **the experiments**.

2.4 MATERIAL USED IN GAMMA RADIOLYSIS STUDY

The **UO₂F₂•0.4H₂O** used in the experiment was obtained **from the East Tennessee Technology Park Material from this same batch was also used in the earlier experiments?**

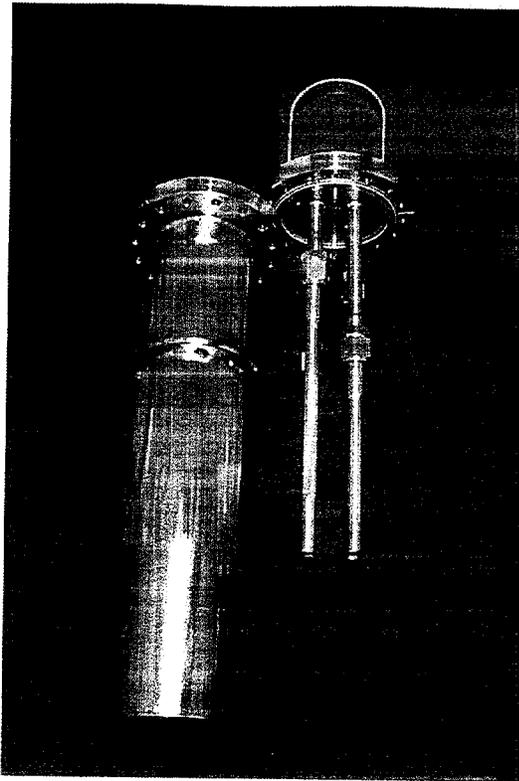


Fig. 2.4. Photograph of multiple-irradiation container used in HFIR SNF irradiations.

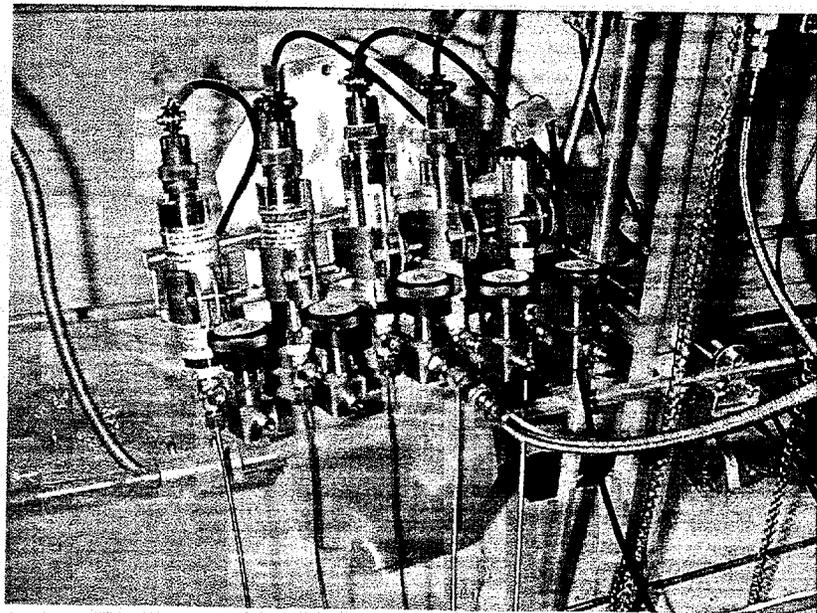


Fig. 2.5. Photograph of pressure transducer assembly that was connected to the multiple-irradiation container and used in HFIR SNF irradiations.

2.5 SAMPLING AND ANALYSES

At the completion of the irradiation, a gas sample was withdrawn and analyzed by mass spectrometry (MS) and Fourier Transform **Infrared (FTIR)** spectroscopy. Prior to withdrawal of gas samples, the irradiation rig was connected to a sampling rig (Fig. 2.6), which consisted of a sample cell for mass spectrometry connected in series to a **10-cm FTIR** gas cell. Zinc selenide windows were used in the FTIR gas cell. The uranium valence of the sample was analyzed by Davies-Gray titration both before and **after irradiation**.⁹⁻¹¹

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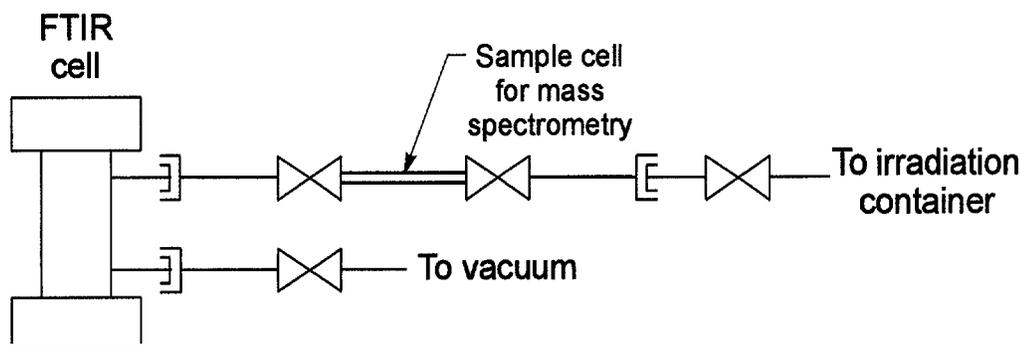


Fig. 2.6. Sampling rig used to withdraw gas samples from the irradiation container.

3. RESULTS

A 19.5-g sample of $\text{UO}_2\text{F}_2 \cdot 0.4\text{H}_2\text{O}$ was irradiated consecutively in two **HFIR** SNF elements until it had received a total dose of 3.1×10^{10} rad. Results from this experiment are presented in Sects. 3.1-3.3.

3.1 PRESSURE MEASUREMENTS

The pressure within the sample container was monitored throughout the irradiation. The data for this experiment are shown in Fig. 3.1, which is a plot of the pressure and gas yield (**mmol** gas/g sample)

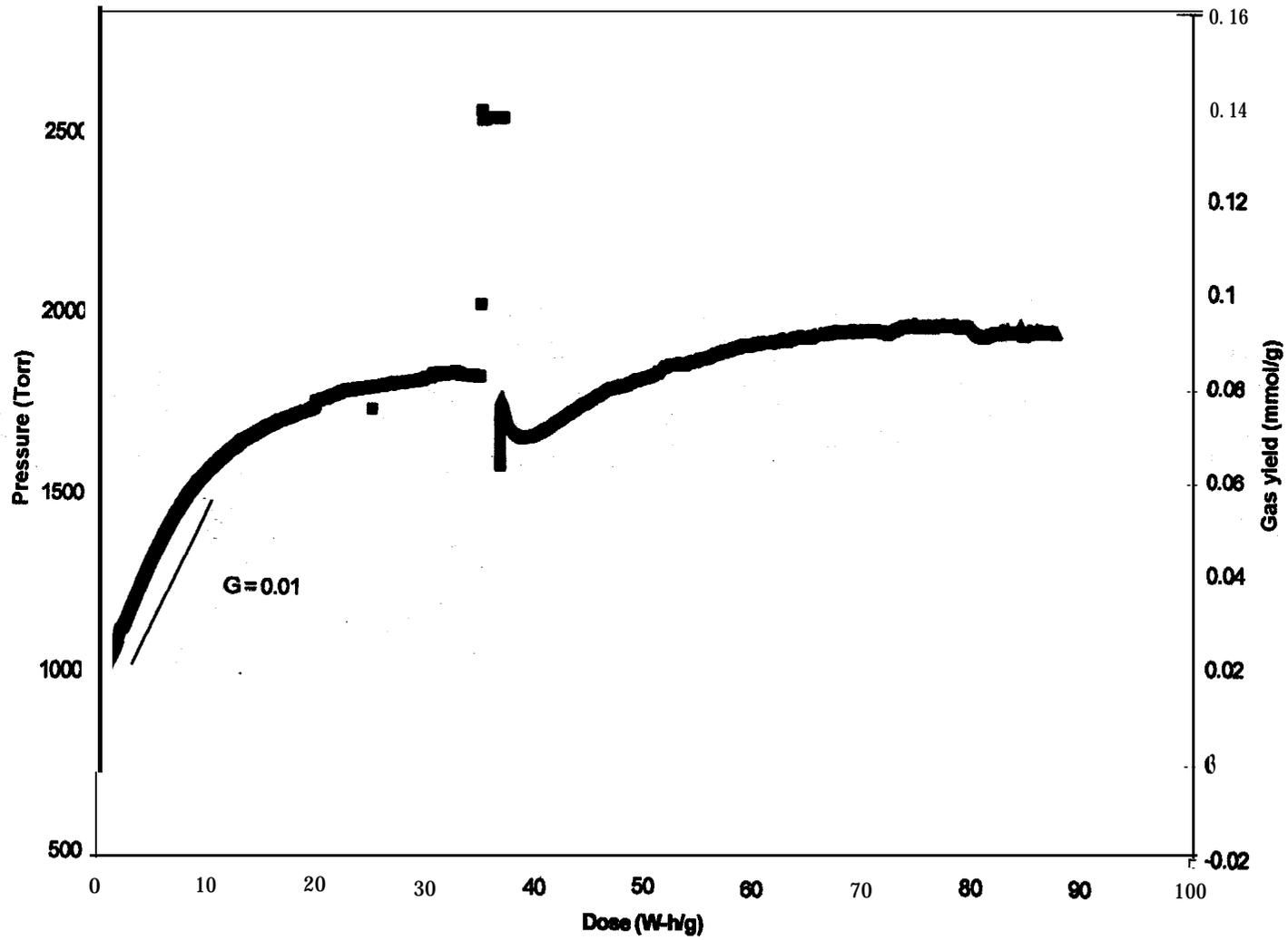


Fig. 3.1. Pressure and gas yield as a function of dose for sample HFIR-6-4 (HFIR SNF-irradiated $\text{UO}_2\text{F}_2 \cdot 0.4\text{H}_2\text{O}$, loaded in helium).

as a function of dose. The gas yield was calculated using the ideal gas law, and the temperature was estimated from the change in pressure of the outer vessel that surrounded the sample container.

The pressure exhibited a steady increase and eventually reached a plateau. After a dose of about 35 W-b/g had been attained, the sample was placed in a freshly discharged SNF element (-5 d since shutdown). The heat generated by this element caused the pressure in both the sample container and the outer vessel to increase rapidly. Because the pressure in the containers was greater than a predetermined action level, the sample was removed from the SNF element. After the element had cooled for an additional 12 days, the sample was reinserted. The pressure then increased until it again reached the plateau.

The initial G-value [i.e., the number of molecules of a species produced (or destroyed) per 100 eV of energy deposited] for this material can be computed from the linearly increasing portion of the curve shown in Fig. 3.1. The initial G-value was estimated to be about 0.01 molecule gas/100 eV.

3.2 GAS ANALYSIS

Results obtained from the analysis of a gas sample taken after irradiation of the $\text{UO}_2\text{F}_2 \cdot 0.4\text{H}_2\text{O}$ are shown in Table 3.1. The initial atmosphere for this sample was helium. Clearly, O_2 and CO , were produced by the gamma irradiation. Neither F_2 nor HF was present.

3.3 SOLIDS ANALYSIS

Because of previous experience with the gamma irradiation of uranyl fluoride² and the light green color produced in the irradiated sample, the valence of the uranium in the sample was determined.⁹⁻¹¹ The results obtained by analyzing three samples of the irradiated material (see Table 3.2) show that U(IV) was produced, reaching a level of about 9 wt % of the total uranium.

Table 3.1. Results of mass spectrometric analysis of gas sample from $\text{UO}_2\text{F}_2 \cdot 0.4\text{H}_2\text{O}$ that was loaded in helium and irradiated in HFIR SNF elements

Component	vol %
N_2	0.42
He	45.58
H_2	co.01
CO_2	5.34
Ar	<0.001
O_2	48.65
HF/Ar^{2+}	<0.01
F_2	<0.01
CH_4	~0.001
c o	co.01
N O , ' ,	co.01

Table 3.2. Results of analysis of HFIR-6-4 samples for U(IV) before and after gamma irradiation

Sample	Weight percentage of total uranium as U(IV)	
	Before irradiation	After irradiation
1	0.114	9.521
2	0.114	8.768
3	0.114	8.557

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4. DISCUSSION AND CONCLUSIONS

This experiment was similar to previous work performed using uranyl fluoride,² and the results are consistent with those from the earlier work. The pressure in the sample container was seen to increase steadily during the irradiation, eventually reaching a plateau. The initial G-value for the radiolytic reaction was estimated to be about 0.01 molecule O₂/100 eV. The earlier experiments yielded G-values ranging from 0.007 to 0.03 molecule O₂/100 eV, which were shown to be dependent on the degree of hydration of the sample.²

The gas analysis revealed that both O₂ and CO₂ were produced. The O₂ was released (as 0 radicals) upon the irradiation of the UO₂F₂•0.4H₂O, and some of the O₂ reacted with carbon impurities to produce CO₂. The role of carbon impurities was clearly demonstrated in studies reported in ref. 2, where removal of carbon from samples prior to irradiation resulted in increased O₂ production and reduced CO₂ production. Unlike other fluoride salts (e.g., those described in refs. 3 and 12–14), F₂ was not released. Uranyl fluoride is characterized as a crystalline solid with mixed bonding, having both covalent and ionic characteristics. This experiment has shown that the oxygen in the covalently bonded uranyl (UO₂²⁺) group is more susceptible to radiation damage than is the ionically bonded fluorine. This same result has been found by the irradiation of other crystals with mixed bonding [e.g., Ba(NO₃)₂, NaNO₃, KNO₃, and KClO₃].^{15,16}

The valence of the uranium after irradiation was measured by Davies-Gray titration. It was found that some of the uranium was reduced from U(VI) to U(IV) by the gamma radiation, and this result provides further evidence that O₂ was released by the irradiation. Additionally, the amount of U(IV) produced provides a measure of the damage to the UO₂F₂ matrix. After irradiation, -9 wt % of the total uranium was found to be U(IV). Because a pressure plateau was reached, this value represents the damage limit to this material. This result compares well with the range of 7-9 wt % obtained in the earlier experiment with HFIR SNF elements.²

If all of the O₂ (or CO₂) produced is captured in the gas space above the sample, then the ratio of the moles of O₂ + CO₂ to moles of U(IV) should be 0.5. Based on the pressure increase and the valence analysis, this ratio is estimated to be about 0.3, indicating that some of the oxygen may remain in the lattice as an intermediate reaction product or be scavenged by an impurity or wall reaction. The ratios measured from the earlier experiments ranged from 0.08 to 0.4.

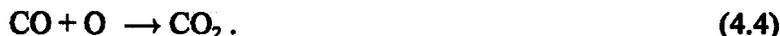
The observed pressure increase, gas composition, and valence change, when considered in total, give a clear picture of the radiolytic effects on the UO₂F₂•0.4H₂O. A mechanism for these effects was established

in ref. 2 and was further confirmed by this experiment. This mechanism is ~~indicated~~ in the following paragraphs.

Gamma radiation interacts with the **uranyl group** of the UO_2F_2 , releasing O radicals and **reducing** the uranium to **U(IV)**, as indicated in the following equation:



The O radicals react with one another to produce O_2 or react with carbon impurities in the sample to produce CO_2 :



The UO_2F_2 consists of stacked layers, with the linear UO_2^{2+} ions normal to each layer (with a **double-bonded oxygen** above and below each plane) and **fluorine atoms surrounding the uranium in its equatorial plane**. This relatively **open structure** allows for the **molecular oxygen** to be readily released **from the matrix and explains the lack of an induction period that was seen in other materials**.^{3,12-14} The released O_2 (or O_2 in the form of CO) causes a pressure increase in the irradiation container, **which** can be used to estimate the gas yield. The initial $G(\text{O}_2)$ -value for the gamma irradiation of $\text{UO}_2\text{F}_2 \cdot 0.4\text{H}_2\text{O}$ was 0.01 molecule $\text{O}_2/100 \text{ eV}$. The G -value depends on the amount of hydration of the UO_2F_2 and the **atmosphere** over the sample. **Back reactions** with oxidizing species **Produced from the radiolysis of water or moist air** may lower the reduction rate of the **uranium**.²

During irradiation, back reactions such as the following may occur:



At some point, a steady state is reached for a given dose rate in which the forward reaction [Eq. (4.1)] rate equals the back reaction [Eq. (4.5)] rate. A change in the dose rate would result in a change in the steady-state level. Note that during the irradiation, the sample was moved to a **fresher** (i.e., more recently discharged) fuel element. The higher dose rate resulted in a high rate of radiolytic production (Fig 3.1). Because the pressure **increase** in the container was above a **predetermined** action level (a standard procedure for **HFIR safety** purposes), the sample was removed **from** the SNF element and was then reinserted **after** the element had cooled for an additional 12 d. As shown in Fig. 3.1, **after reinsertion** into

the element, the pressure **initially** decreased prior to risii back to the plateau This decrease occurred as the excess radiolytic products (generated by the much higher dose rate) were **consumed through back** reactions at a lower dose rate. Once the excess products were **consumed**, the system returned to the steady state.

While the steady state pressure attained in these and other experiments provides ample evidence for the existence of a back reaction, the **simplified** form of Eq. (4.5) **does** not satisfactorily **explain** the discrepancy in the ratio of **oxygen-containing** product (O_2 or CO_2) to moles of U(IV) formed We are led to **conclude** that there is an **oxygen-containing intermediate** that remains in the **lattice**—**either** trapped or non-volatile. Further work will seek to elucidate details regarding such intermediate **radiolytic** products.

At the steady state, the maximum damage limit to the UO_2F_2 matrix is realized; this limit is **~9 wt %** of the total U as U(IV) for the high dose rates available with HFIR SNF elements (**~ 10^8 rad/h**). Hence, the radiation damage reaches a **saturation point**, similar to other **crystalline** solids. Additionally, the damage limit to the ionic LiF- BeF_2 crystals has been shown to be about **2%** at the same dose rate as that used for the $UO_2F_2 \cdot 0.4H_2O$ sample.¹⁴ The higher limit for the UO_2F_2 is further evidence that the **covalently** bonded uranyl group is more susceptible to radiation damage than is the **ionically** bonded fluorine.

The **significance** of these findings is found in view of their application to the long-term storage of uranium oxides that may contain residual **fluoride compounds**—**recalling** that the goal of this work was to **evaluate** the radiolytic effects on such materials and thereby establish criteria for their safe storage. For uranium oxides placed in storage, UO_2F_2 is **present** in **very small quantities** (typically less than 1 wt %). For this experiment, the UO_2F_2 sample was exposed to high gamma doses that exceeded the total expected dose in 50 ye& by at least a **factor** of 100. Based on the results of this work, it is clear that the gamma radiolysis of this impurity in the uranium oxide **would be** inconsequential. Additionally, $^{233}U/^{232}U$ -containing materials have both alpha and gamma radiation fields that can cause radiolysis. The work performed to date has evaluated the **effects** of gamma radiation. Other work currently underway at ORNL will **evaluate** the effects of alpha radiation. The results **from** both the gamma and alpha radiolysis experiments will complete the understanding on the **effects** of radiation on **fluoride** impurities in ^{233}U oxides.

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Appendix A

ESTIMATION OF ABSORBED DOSE FROM EXPOSURE

Appendix A. ESTIMATION OF ABSORBED DOSE FROM EXPOSURE

Exposure rates are reported for the HFIR SNF elements that were used in the irradiation experiments. *Exposure* is a measure of the amount of charge produced in air per unit mass of air. However, in the case of radiolytic experiments, the quantity of interest is the absorbed dose, which is the energy absorbed by a material per unit mass. To compute the absorbed dose, the method presented in ASTM E666-91 (*Standard Practice for Calculating Absorbed Dose from Gamma and X Radiation, American Society for Testing and Materials, Philadelphia, 1991*) is used. The following formula is used to convert exposure rate to dose rate:

$$\dot{D}_y = 8.69 \times 10^{-3} \frac{\left(\frac{\mu_{en}}{\rho}\right)_y}{\left(\frac{\mu_{en}}{\rho}\right)_{air}} \dot{X} \exp\left[-\left(\frac{\mu_{en}}{\rho}\right)_y x\right], \quad (\text{A.1})$$

where

\dot{D}_y = dose rate in material y at depth x , Gy/h;

$\frac{\mu_{en}}{\rho}$ = mass energy absorption coefficient, m^2/kg ;

\dot{X} = exposure rate, R/h.

The value 8.69×10^{-3} converts roentgens to grays (Gy) in air.

For small samples, the sample thickness is neglected; the equation then reduces to

$$\dot{D}_y = 8.69 \times 10^{-3} \frac{\left(\frac{\mu_{en}}{\rho}\right)_y}{\left(\frac{\mu_{en}}{\rho}\right)_{air}} \dot{X}. \quad (\text{A.2})$$

For samples that consist of mixtures of elements, the mass energy absorption coefficient is calculated by Hubbell (Complete citation provided in note to Table A. 1):

$$\frac{\mu_{en}}{\rho} = \sum_i w_i \left(\frac{\mu_{en}}{\rho} \right)_i, \quad (\text{A.3})$$

where

- w_i = the proportion by weight of the *i*th element, dimensionless;
- $\left(\frac{\mu_{en}}{\rho} \right)_i$ = mass energy absorption coefficient for the *i*th element, m^2/kg .

Selected values of $\frac{\mu_{en}}{\rho}$ for several elements and energies are presented in Table A.I.

Table A.I. Selected mass energy absorption coefficients^a

Energy (MeV)	μ_{en} / ρ ($10^{-3} \text{ m}^2/\text{kg}$)				
	U	O	F	H ₂ O	air
0.93	4.978	2.824	2.675	3.137	2.820
1	4.473	2.791	2.643	3.100	2.787
1.25	3.748	2.669	2.528	2.966	2.666
2	2.612	2.346	2.223	2.604	2.342

^a J. H. Hubbell, "Photon Mass Attenuation and Energy-Absorption Coefficients from 1 keV to 20 MeV," *Int. J. Appl. Radiat. Isot.* 33, 1269-90 (1982).

To account for the slight attenuation of the photon flux by the irradiation containers, the computed dose rate was multiplied by the attenuation factor $e^{-\mu x}$, where μ is the attenuation coefficient (cm^{-1}) and x is the wall thickness of the container. For the HFIR SNF irradiations, the attenuation factor was based on the average energy of 0.93 MeV. Selected attenuation coefficients are presented in Table A.2.

Table A.2. Selected attenuation coefficients (μ) for materials used in irradiation containers

Energy (MeV)	μ (cm^{-1})	
	Ni ^a	Fe ^b
0.93	0.241	0.5004
1	0.238	0.4807
1.25	0.234	0.4362
2	0.220	0.3421

^a E. Storm and H. I. Israel, *Nuclear Data Tables A7*, 565 (1970).

^b J. H. Hubbell, "Photon Mass Attenuation and Energy-Absorption Coefficients from 1 keV to 20 MeV," *Int. J. Appl. Radiat. Isot.* 33, 1269–90 (1982).

For the HFIR SNF element irradiations, exposure-rate data, as a function of time, were provided. These data were fit to a curve (e.g., Fig. A. 1), which was integrated to determine the total exposure during an irradiation. This exposure was then converted to dose by using Eqs. (A.2) and (A.3). The attenuation of the gamma field by the walls of the sample container was accounted for by multiplying the computed dose by the attenuation factor, $e^{-\mu x}$.

The HFIR SNF element emits a spectrum of gamma-ray energies. The average gamma energy (1 d after SNF discharge from the reactor) is 0.93 MeV (see Table 2.1 in the body of this report), and attenuation factors (and, hence, dose) are calculated on the basis of this energy. Additional calculations have been performed using different energies for the gamma rays: 1 and 2 MeV. "In each case, the computed G-value for the gas yield was the same; hence, the G-value computation was relatively insensitive to the gamma energy for the HFIR SNF irradiations.

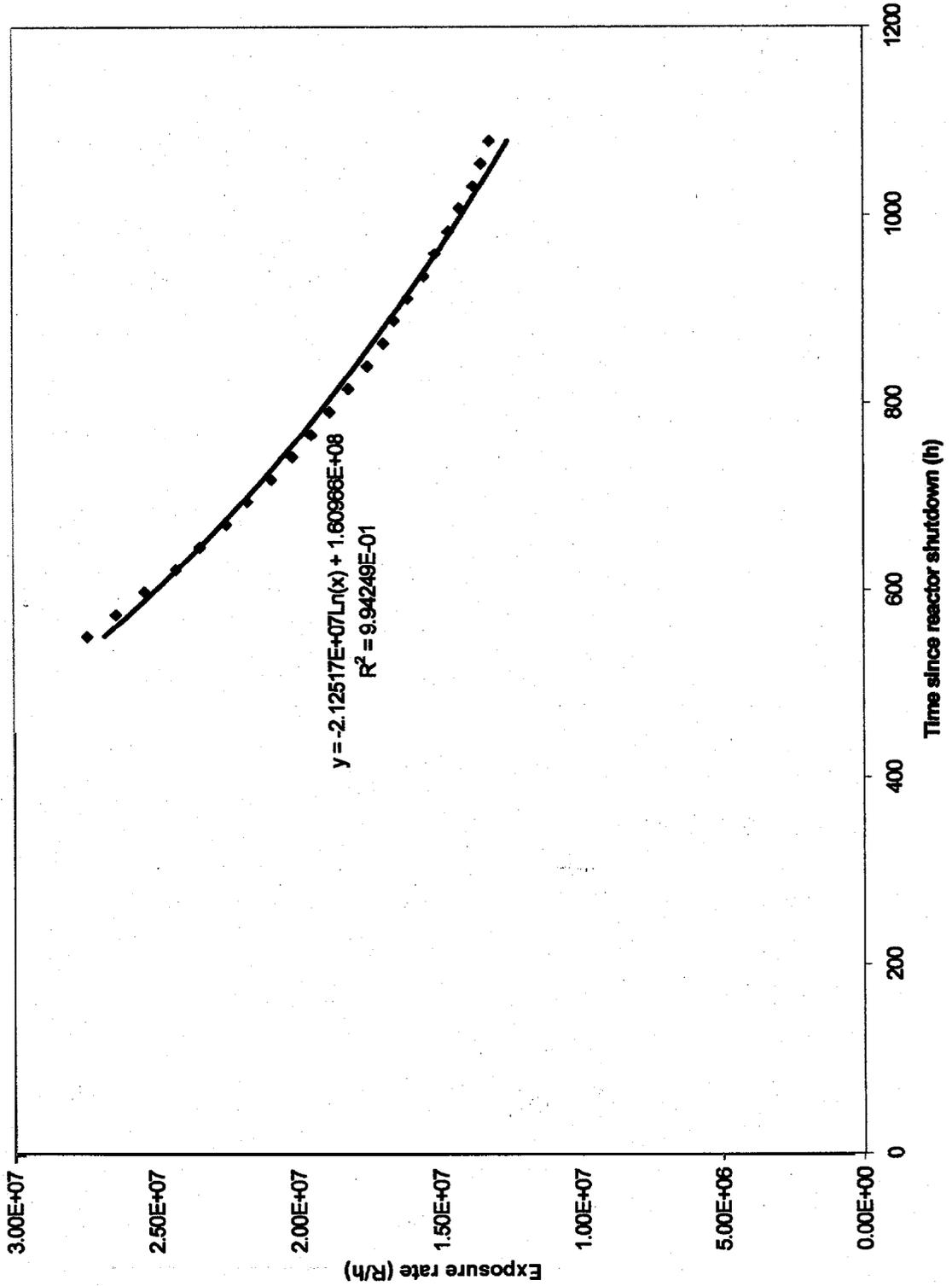


Fig. A.1. Exposure-rate curve fit for HFIR SNF element from cycle 380.

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