DUN. 1576

MASTER

ORNL-4601 UC-80 — Reactor Technology

EVALUATING FUEL BEHAVIOR DURING

IRRADIATION BY FISSION-GAS RELEASE

R. M. Carroll O. Sismon



OAK RIDGE NATIONAL LABORATORY

UNION CARBIDE CORPORATION for the U.S. ATOMIC ENERGY COMMISSION

DISTRIBUTION OF THIS DOCUMENT IS UNITED

BLANK PAGE

· . . .

, **(** ,) ,

Frinted to the College States of Associate, Association from Clearinghouse for Federal Scientific and Technical Information, National Jurray of Standards, U.S. Department of Connerce, Springfield, Virgitie 22151 - Fries: Printes Cepy 53.04; Hierefliche \$0.65

۰,

• 1 1

F

This report was propared as an ecoluly of Sove muont sponsored work. Koither the United States, an the Commission, and any person anting on take if at the Commission:

LEGAL MITCE -

- station, expressed in haplied, with respect to the eccuracy, s any warranty or summer ness, or verificers of the information correlated in this report, or that the use of any information, personal without, or general discharged a this report may not infringe privately armed rights; at
- on any Habilitian with request to 🗠 and all or for demages resulting from 🖂 🛹 of . tas ory information, opportulue, stind, or process discioned in this report.

used in the uters, "person acting an behalf of the Commission" theredos any analogue or tractor of the Commission, or suplayed of ruch contentor, to vice excent that such employme or contractor of the Counstantion, or anglesis of such contractor property, diasoninates, or provides occess to, any information parsuant to his employment or contract with the Commission, er his employment with such contractor.

BLANK PAGE

ORNL-4601

Contract No. W-7405-eng-26

REACTOR CHEMISTRY DIVISION

EVALUATING FUEL BEHAVIOR DURING IRRADIATION BY FISSION-GAS RELEASE

R M. Carroll and O. Sisman

SEPTEMBER 1970

Province and the province of t

•

LEGAL NOTICE This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

OAK RIDGE NATIONAL LABORATORY Oak Ridge, Tennessee operated by UNION CARBIDE CORPORATION for the U.S. ATOMIC ENERGY COMMISSION

m

CONTENTS

Abstract	•	•	•	•	•	•	•	•	•	•	•	•	•	•	1
Introductio	n	•	•	•	•	•	•	•	•	•	•	•	•	•	1
Gas Release	by	Surf	ace	Fisa	sion	5	•	•	•	•	3	•		•	2
Gas Release	e fro	om th	e Fu	æl 1	Body	•	•	•	•	•	•	•	•	•	3
Experiments	il Me	ethod	•	•	•	•	•	•	•	•	•	•	•	•	4
Interpreta	ion	of I	soto	opi c	Ra t	i os	•	•	•	•	•	•	•	•	4
Interpretat	tion	of C	hane	ge in	n St	eady-	Ste	te Ge	s Re	eleas	æ	•	•	•	9
Gas Rele	ea se	Rate	Dec	rea	sing	•	•	•	•	•	•	•	•	•	9
Ges Rele	ea se	Rate	Inc	erea	sing	•	•	•	•	•	•	•	•	•	11
Interpretat	tion	of F	'issi	on-(Ges	Relea	se	When	the	Spec	imen	is	Cool	led	17
Summe ry	•	-	•	•	•	•		•	•	•	•	•	•	•	2 3
References	•	•	•	•	•	•	•	•		•		•	•	•	2 8

Page

1

-

- Marin

ł

I.

EVALUATING FUEL BEHAVIOR DURING IRRADIATION BY FISSION-GAS RELEASE

R M Cerroll and O. Sismen

ABSTRACT

If a nuclear fuel is being irraciated where evolved fission gas can be measured, considerable information about the physical condition of the fuel can be obtained from observations of how the fission-gas release rate responds to different irradiation conditions of the fuel. The unchanging release of fission gas as burnup progresses can assure the experimenter that the fuel has not changed physical characteristics. Alternatively, a variable behavior of gas release rate can imply a cracking fuel, irradiation sintering of porosity, chemical reactions, grain growth, breakaway swelling, or surface changes.

INTRODUCTION

In the past decade a model for the release of fission gas from reactor fuel material has evolved, which has now been fairly universally accepted. This model, which we call the "defect-trap" model, postulates the movement of fission gas through a combination of diffusion and trapping processes. There appears now to be no argument about the general principles of this model, although different investigators may have slight differences in the details of the process.^{1,2,3} Other models are discussed in reviews of the literature.^{4,5}

This report will deal with the physical changes in the fuel which can be inferred from changes in the release rate and composition of the fission gas. Examples of how these changes are evaluated are illustrated with data from our past experiments, some of which have already been published. This report is, however, not a review but rather a summary of the use of an evaluation technique, which we have found to be most useful.

In our experiments we have monitored the fission-gas release rate and composition, the temperature, and the neutron flux continuously during the irradiation of various fuel materials. Furthermore, we have the capability of changing the fission rate within the specimen and the

BLANK PAGE

specimen temperature independently. The discussion which follows assumes this capability, but much can be inferred about the condition of the fuel even if only the gas release and temperature are monitored. The defecttrap model was developed for UO^2 but will apply to any fuel with similar properties.

GAS RELEASE BY SURFACE FISSIONS

Fission-gas releate from fuel materials can be divided into two portions, release from the body of the fuel and release caused by fissions on the surface of the fuel. If fission occurs within about 10 microns of a surface it is possible for one of the fission fragments to recoil free of the fuel. Almost always, the fission fragment leaving the surface will emerge with considerable energy remaining. Thus, if the surface is part of a marrow crack or small pore, the fission fragment will likely reenter the fuel.

The amount of free surface (i.e., surface connected to the exterior of the specimen) of a ceramic specimen is usually much greater than the geometric surface area. However, because of the reembedding process, the release of fission gas by direct fission recoil is governed by the geometric surface area. Moreover, if the fuel is held in a close-fitting solid holder, a large proportion of the recoiling fission fragments will embed in the holder. The net result is that only a very small proportion of the fission fragments recoiling from the fuel surface will escape from the immediate vicinity of the fuel.⁶

When a fission fragment passes through a surface some of the surface molecules will be "knocked out." If the surface contains fission gas, then some of the fission gas will be liberated by the knockout process. The number of molecules knocked out of the surface depends on the energy of the fission fragment and the condition of the fuel surface. The fission-gas atoms liberated by the knockout process will have little energy and will not be embedded in nearby surfaces. This means that the knockout release will be related to the geometric surface area.⁶

During fissioning, some isotopes of fission gas are produced which have half-lives in the order of hours. For these isotopes, an equilibrium will soon be reached where the escape rate plus the radioactive

2

decay rate will equal the production rate. The production rate of fission gas is directly proportional to the fission rate and the equilibrium amount of short-lived isotopes of fission gas within the fuel is also directly proportional to the fission rate (if the escape rate is much smaller than the production rate). The knockout release rate for a given surface depends on both the rate of surface fissions and on the fission-gas concentration at the surface. Since both of these processes are directly proportional to the fission density, it follows that the knockout release is proportional to the square of the fission rate. In contrast, the direct recoil release is directly proportional to the fission rate.⁶

An important exception occurs when the surface of the fuel specimen becomes saturated with fission products. At that time an increase in fission rate will increase the surface fission rate but cannot increase the amount of fission products in the saturated surface layer. Under these conditions, the knockout release will increase directly with an increase in fission rate.⁷ However, the knockout release can still be distinguished from the direct-recoil release by comparing the relative amounts of the different isotopes, as described below.

GAS PELEASE FROM THE FUEL BODY

The isolated xenon and krypton atoms, created within the UO₂ structure by fission, are too large to fit into interstitial positions without lattice straining. These atoms alfuse rapidly but tend to cluster, to form bubbles, or to become trapped at imperfections within the fuel structure. In our model,^{6,8} the traps are divided into three categories: (1) intrinsic traps, which are voids, grain boundaries, or other large defects in the fuel caused by the manufacturing process: (2) point defects, which are formed in the wake of a fission fragment; and (3) clusters of point defects. The second and third types of defects are formed by irradiation, whereas the first is an inherent property of the material.

The surface of the specimen may be considered to be a grain boundary, and gas is trapped there, producing a high concentration rather than the depleted surface region expected from recoil and diffusion processes. Gas bubbles collect at grain boundaries and, with sufficient burnup, can

generate enough pressure to cause cracking or swelling. The release of gas from the body of the fuel, then, is either by a coupled diffusiontrapping process or by escape from a ruptured gas pecket.

EXPERIMENTAL METHOD

Some means must be provided for obtaining samples of fission gas for analysis. We use a continuous sweep of purified helium to transport the evolved fission gas outside the reactor. Samples of gas are obtained at intervals and the activity of the gas stream is measured continuously. In our case, the fuel specimen is heated by its own fission heat, which is regulated by moving the specimen capsule to different neutron flux levels.⁹ The temperature of the specimen is controlled, independent of the fission power, by air-cooling the outside of the specimen capsule (see Fig. 1).

INTERPRETATION OF ISOTOPIC RATIOS

Assuming that different isotopes of the same element behave in the same manner, considerable information can be obtained by comparing the relative amounts of the different isotopes of fission gas. The isotopes of xenon and krypton differ in their precursors, their fission yield, and their rate of radioactive decay. All of these factors can be used in evaluating the physical condition of the fuel.

If the escape rate of a short-lived radioactive isotope is small compared to the production rate, equilibrium will be reached when the death rate, $\frac{dN}{dt}$, is equal to the birth rate.

$$\frac{dN}{dt} = N(\lambda + \sigma \phi) = \gamma \Sigma_{f} \phi \qquad (1)$$

where:

N = number of atoms of the isotope.

 $\gamma = \text{decay constant of the isotope},$

 $\dot{\phi}$ = neutron flux,

 Σ_r = the fission cross-section of the fuel,

y = the fission yield of the isotope, and

 σ = the absorption cross-section of the isotope.

ORNL DWG 69-6068



Fig. 1. Schematic of Fuel Irradiation System.

I.

Since λ , ϕ , Σ_{f} , γ , and σ are known, then N, the amount of isotope trapped in the fuel, can be calculated.

$$N = \frac{\text{birth rate}}{\gamma + \sigma} = \frac{\gamma \Sigma_{f} \phi}{\gamma + \sigma \phi}$$
(2)

Except for ¹³⁵Xe, for which σ is high, $\lambda + \sigma \phi \approx \lambda$.

The ratio of two different isotopes trapped in the fuel is

$$\frac{N_1}{N_2} = \frac{\gamma_1 \Sigma_f \phi}{\gamma_2 \Sigma_f \phi} \cdot \frac{\lambda_2}{\lambda_1} = \frac{\gamma_1}{\gamma_2} \cdot \frac{\lambda_2}{\lambda_1}$$
(3)

when the isotopes are at radioactive equilibrium.

The amount of isotopes released by direct recoil from a given fuel surface will be directly proportional to the fission rate. Isotopes released by recoil will have ratios in direct proportion to their fission yields.

$$\frac{N_1}{N_2} = \frac{\gamma_1 \Sigma_f \Phi}{\gamma_2 \Sigma_f \Phi} = \frac{\gamma_1}{\gamma_2}$$
(4)

Isotopes released by the knockout process will have a ratio somewhat between that for direct recoil and that for trapped gas.

Isotopes which are released by diffusion follow the general relation

$$f = C \sqrt{\frac{D}{\lambda}}$$
 (5)

where

f = fractional release (rate of release/rate of formation),

D = diffusion coefficient for the isotope, and

C = a constant which depends upon the diffusion model used.

The ratio of the amounts of two isotopes escaping the fuel by diffusion will be,

$$\frac{f_1}{f_2} = \frac{C\sqrt{\frac{D_1}{\lambda_1}}}{C\sqrt{\frac{D_2}{\lambda_2}}} = \sqrt{\frac{D_1\lambda_2}{D_2\lambda_1}} \quad . \tag{6}$$

If the diffusion coefficients are equal,

$$\frac{f_1}{f_2} = \sqrt{\frac{\lambda_2}{\lambda_1}} \tag{7}$$

The fractional release $f = \frac{R}{\gamma \Sigma_{f} \phi}$; R = release rate

and

$$\frac{\mathbf{f_1}}{\mathbf{f_2}} = \frac{\mathbf{R_1} \mathbf{\gamma}_2 \boldsymbol{\Sigma}_{\mathbf{f}} \boldsymbol{\phi}}{\mathbf{R_2} \mathbf{\gamma}_1 \boldsymbol{\Sigma}_{\mathbf{f}} \boldsymbol{\phi}}$$

The ratio for the equilibrium release rates of two isotopes with the same diffusion coefficients is then

$$\frac{R_1}{R_2} = \frac{\gamma_1}{\gamma_2} \sqrt{\frac{\lambda_2}{\lambda_1}} \qquad (8)$$

In most fuels which we have examined the xenon and krypton behave in the same manner, consistent with a trapping process. It is erroneous, however, to suppose that xenon and krypton will have the same behavior in all materials. For example, in pyrolytic-carbon-coated fuel particles $(UO_2 \text{ or } UC_2)$ we found that krypton had a proportionally much greater escape rate than xenon.¹⁰ (See Fig. 2) This is apparently caused by the easier diffusion of the smaller sized krypton atom through the carbon coating, since the effect of the smaller mass on the diffusion rate cannot account for the magnitude of the difference. We knew the coating was not broken, because, when the coating did break, the ratio of the xenon to krypton emerging from the fuel became the same as that for uncosted fuel particles.¹⁰ Thus the failures of the costings were detected first by bursts of fission gas released at the moment the particle ruptured, and coating failure was confirmed by the subsequent higher steadystate release rate and the different ratio of the senon to krypton. In addition, an analysis of the isotopes of the fission gas comprising the bursts showed the isotopes to be in the ratio given in Eq. (3), indicating stored gas.



į

2 P. N. Secondon Bul

Fig. 2. A Comparison of Fractional (R/B) Release from WO_2 Coeted with Buffered Isotropic Pyrolytic Carbon (BISO) to the Fractional Release from Bare WO_2 .

INTERPRETATION OF CHANGE IN STEADI-STATE GAS RELEASE

During irradiation, steady-state fission-gas release is attained when the specimen has been irradiated at constant conditions long enough to establish radioactive equilibrium for the isotopes being measured (Eq. 1). Equilibrium is assumed when the release rates of the isotopes are not changing significantly over a period of time equal to several half-lives. When the specimen has been at constant fission rate and temperature long enough to establish radioactive equilibrium but the fissiongas release is variable, then the physical characteristics of the specimen are changing. Listed below are some interpretations of the significance of variable gas release while the specimen is being irradiated under steady-state conditions.

Gas Release Rate Decreasing

Steady decrease in gas release can be caused by two factors; surface fissioning causing a change in the specimen surface, or by tiny cracks or pores in the fuel specimen being sintered closed by fission-spike welding.

The observation that the knockout release rate appears to decrease with time has been explained with three different theories.4,5 (1) Irradiation is smoothing the microscopic irregularities in the surface, thus decreasing the surface area. (2) An equilibrium between ejection and redeposition of the surface molecules is occurring. (3) Passage of the fission fragments through the surface causes recrystallization, which reduces the ejection rate of molecules from the surface. In any event, only the surface of the specimen is affected.⁶ Any escaping gas must, however, pass through this surface; thus, the hightemperature gas release rate is affected in the same manner as the knockout release rate. An extreme example of a decrease in ⁸⁸Kr by knockout release with irradiation time is illustrated in Fig. 3. Although each specimen behaves in a different manner, the decrease is usually not signii nt after about 30 days of irradiation and the subsequent gas relea .s about 1/3 to 1/2 the starting release; by contrast, the example





The decline in steady-state gas release because of surface changes is to be expected. Athough the extent of the decline cannot be predicted accurately, it is a smooth exponential function of irradiation time, or burnup, and apparently is little affected by the temperature of the specimen. This decline does not mean that the body of the specimen is changing.

Crack healing, or the irradiation sintering of interconnected porcsity, car occur during fissioning at much lower temperatures than required for sintering during manufacturing. This can be distinguished from the surface change process described above because the irradiation sintering is strongly temperature dependent; whereas, the surface change process is not. An example of ⁸⁸Kr release from (Pu,U)O₂, as interconnected porosity was sintered closed, is shown in Fig. 4. Crack healing usually does not produce a smooth exponential function of gas release vs time for very long. In Fig. 4, after 1.8 x 10¹⁹ fissions/cm³, new cracks caused an increased gas release rate and the cracks healed rapidly only to crack again. The points show measured values with the line segments showing the activity trend. It should be noted that this and subsequent figures involve both continuous and grab-sample measurements with the points indicating grab-sample data. Temperature perturbations will cause stress cracking which also heals rapidly. Crack healing progresses more rapidly at higher temperatures.

In Fig. 4, as in some subsequent figures, the data were normalized to some specific temperature. This means that some of the data were measured at some other temperature and then, since the temperature dependence of the gas release was known, the data values were adjusted for the selected temperature. The standard temperature was selected so that minimum adjustments were necessary.

Gas Release Rate Increasing

Cracking of the specimen will produce a small burst of fission gas and then a subsequently higher fission-gas release rate because of the increased surface area. Fission-gas pressure within grain boundaries and closed pores can produce cracking after the accumulation of enough burnup. As an example, the steady-state gas release from fused-crystal spheres of



.

Fig. 4. ⁸⁸Kr Release from (Pu,U)02.

UO₂ is shown in Fig. 5. The initial decrease in ⁸⁸Kr release rate is caused by changes in the specimen surface. The abrupt increase at 1.9%burnup is caused by the continued breaking of the spheres into even smaller particles ε s a result of the buildup of fission-gas pressure.¹¹

Increasing gas release can also be caused by a chemical reaction of the fuel with some portion of its environment.¹² Unless the reaction causes the fuel to crack, the gas release will increase smoothly as the reaction progresses. Cracking causes an irregular increase.

Equiaxed grain growth will cause increased fission-gas release during grain growth. This is illustrated in Fig. 6, where a UC₂ specimen was irradiated at 1700°C for 5 days.¹¹ Apparently gas trapped at grain boundaries can escape along the mobile grain boundaries during grain growth. This type escape can be confirmed by lowering the temperature of the fuel below the grain growth temperature. If the gas release at lower temperatures is the same as before, then grain growth has caused the increase in release rate. In the example shown in Fig. 6, existing cracks were healed, resulting in a lower release rate after grain growth had occurred. A measurement of knockout release will confirm whether or not the surface area of the specimen has changed.

If the gas release rate increases rapidly in an exponential fashion it is most likely caused by breakaway gas release; where the pressure of the trapped gas exceeds the fuel-matrix strength and the fuel begins to swell. The bubbles of trapped gas pushing their way to the surface will produce pips of gas release and the resulting porosity will increase the steady-state release rate.

The breakaway gas release can be distinguished from equiaxed grain growth because grain growth does not affect the subsequent gas release rate;¹¹ breakaway gas release will occur at lower and lower temperatures as burnup progresses because of the accumulation of fission gas. An example of this is shown in Fig. 7, where the temperature of spherical $(U, Pu)O_2$ specimens was maintained just around the breakaway gas release point as irradiation progressed. This experiment was performed after a cylindrical specimen of the same material had shown almost explosive release of fission gas when the specimen temperature was suddenly increased from $1100^{O_2} \pm 0.1450^{O_2-3/2}$











Fig. 7. Temperature vs Burnup for Breakaway Gas Release from Unrestrained Mixed Oxide Fuel (75% UO2, 25%, PuO2).

INTERPRETATION OF FISSION-GAS RELEASE WHEN THE SHECIMEN IS COOLED

During irradiation some of the iodine precursors of xenon will escape from the fuel and deposit on cooler surfaces outside the fuel. If the fuel is withdrawn from the neutron flux, it will cool rapidly and the release of fission gas will stop. However, the deposited iodine will continue to decay into xenon. An example of the behavior of UO_2 in good condition during such a cooling period is shown in Fig. 8.

There are three ways to confirm that the source of the xenon is the decay of iddine outside the fuel specimen. (1) The krypton release ceases while the xenon release continues (if the xenon were emerging from the fuel, so would the krypton). (2) The emission rate of xenon decreases with the decay rate of the iddine precursor. (3) The isomer, 135m Xe, which has only a 15-minute half-life, is strongly present in the sweep gas. (If the xenon were diffusing from the fuel, most of the 135m Xe would have decayed before being released.)

Rapid cooling of ceramic fuel creates thermal stresses. If the fuel is in good condition, the fission-gas release will respond as illustrated in Fig. 8. If the fuel contains microcracks in the surface or grain boundaries strained by trapped fission-gas bubbles, then cracks will open during the cooling period and release trapped fission gas. Such a cooling burst is shown in Fig. 9, where a fine-grain specimen of UO_2 was quickly cooled from 1700°C to about 250°C.¹¹ In this cooling burst it is obvious that iodine was also released since the ¹³⁵ Xe is emitted in greater amounts after the burst than before.

A spontaneous burst is shown in Fig. 9. Thermal stresses were not responsible for the spontaneous burst because the specimen had been cool for more than two hours. The ratio of 133 Xe to 135 Xe in the spontaneous burst showed that a store of trapped gas (Eq. 3) was released. Again, some iodine was released in this spontaneous burst. It seems most likely that the spontaneous burst was caused by trapped iodine which built up pressure as it decayed into xenon until a microcrack was opened in the fuel specimen, releasing the accumulated xenon.

Although microcracks opened and closed during the cooling stresses as the UO₂ specimen of Fig. 9 was withdrawn from the flux, the specimen



Fig. 8. Xenon Born of Iodine Decay After a Reactor Scram During Irradiation of Fused-Crystal UO2.

.

.



Fig. 9. ¹³³Xc and ¹³⁵Xe Release when Fine-Grain Specimen (C1-20) was Withdrawn from Neutron Flux after 5 Days Irradiation at $1700^{\circ}C$ ($\phi = 6.0 \times 10^{13}$).

did not have interconnected porosity or a network of large cracks. This is shown by the clean relation of the emitted xenon to the half-life of the iodine precursor.

The response to cooling stresses is shown in Fig. 10 for a UN specimen which has interconnected porosity. Here the cooling stresses caused a large cooling burst, and several bursts in the last phase of the cooling. The interconnected porosity allowed the fission gas to dribble out over a period of hours as shown by the constant ratio of the 133 Xe to 135 Xe. The major source of the fission gas is a result of the leaking of gas from the porous specimen. Gas from iodine decay is a minor portion of the total released, as shown by the absence of dependence on precursor decay.

The character of the release from the UN specimen of Fig. 10 changed during irradiation, beginning with the type release associated with clean UO_2 , and then early in the irradiation changing to a larger steady-state release accompanied by pips of gas. Knockout release indicated an increasing surface area. From cooling curves, such as Fig. 10, we were able to conclude that a chemical reaction was occurring, causing penetrating cracks in the specimen surface. Postirradiation examination confirmed that there had been a reaction between the UN specimen and an Al_2O_3 holder.¹² Our observations showed that the reaction started at temperatures under $600^{\circ}C$.

In contrast, gas release from some $(U, Pu)O_2$ specimens indicated interconnected porosity at the very beginning of irradiation. As shown in Fig. 4, the amount of porosity was reduced by fission sintering, showing that it consisted of small passages, since large cracks are not sintered by fission spikes. Thermal stresses during cooling, as shown in Fig. 11, opened surface cracks or grain boundaries, and secondary bursts occurred after the specimen was cooled. Gas dribbling from the specimen showed some porosity remaining, with some dependence on iodime decay also apparent. By comparing Fig. 11 with Figs. 10, 9, and 8, one can see that the amount of open porosity of the $(U, Pu)O_2$ specimen was less than that of the UN specimen (after the chemical reaction) but more than that for UO_2 specimens in good condition.

ORNL-DWG 68-11903

į

ź,

į









ALL CONTRACTOR

SUMMARY

Because of the difference in properties of the various Xe and Kr isotopes and their precursors, the physical condition of the fuel can be fairly accurately deduced by studying the magnitude and isotopic ratios of the fission gas released. This technique can be especially useful in showing changes in the physical condition of the fuel.

For best results a sweep gas system is used to carry the fission gas from the fuel to a monitoring station, and the fuel temperature and fissioning rate are continuously monitored and controlled. However, it is, often possible to infer the condition of the fuel from fission-gas release data and a guess at the temperature and power density. This can be a very useful tool for in-pile experiments and for reactor monitoring. A summary of the interpretation of the gas-release rate, temperature, and burnup data is given in Table 1.

One important use of fission-gas monitoring is to ensure that the physical condition of a fuel specimen has not changed. If you wish to measure a physical property, such as thermal diffusivity, during irradiation, then the observation that the fission-gas release rate has not changed during the time interval of the tests will confirm that the specimen remained intact during the tests.

It should be noted that it is seldom possible to fully evaluate the condition of the fuel by a single observation. The irradiation conditions and the time-dependent behavior must be considered as well as the ratio of the isotopes of the fission gas and the emission rate as a function of temperature. For example, the gas release characteristics of grain growth and breakaway swelling are very similar, requiring gasrelease measurements at lower temperatures to determine which has happened.

3

Observation	Irradiation Condition	Burnup Level	Interpretation	Confirming Tests		
Slow decrease in gas release rate	Steady state Any temperature	Initial to small	Surface fissions causing change in fuel surface.	Change in gas release rate inde- pendent of temperature. No cooling bursts and only xenon release after reactor shutdown. Isotopic composition constant.		
Slow decrease in gas release rate	Steady state Moderate to high temperature	Initial to high	Interconnected porosity is sin- tering closed by fission spikes and heat.	Change rate depends on tempera- ture. Frequent spontaneous bursts. Targe heating and cooling bursts. Rate of release has some depen- dence on immediate irradiation history. Isotope composition constant except during bursts.		
Rate of gas re- leuse increases in a step manner.	Steady state or variable Any temperature	Any	Specimen cracked	Low-temperature irradiation shows greater release rate. Isotopic composition constant except at moment of cracking.		
Rate of gas re- lease increases quickly, accom- panied by small bursts of gas.	Steady state High temperature	Any	Grain growth	When returned to lower tempera- ture, the gas release rate is no higher than it was before. During the event the isotopic com- position was shifted toward the longer-lived isotopes.		

Table 1. Changes in Fission-Gas Release Caused by Changes in Physical Properties in the Fuel

Table 1 (Cont'd)

Observation	Irradiation Condition	Burnup Level	Interpretation	Confirming Tests Low-temperature irradiation shows greater surface area. Small bursts composed of long- lived fission gases. Rate of breaking increased by thermal shock.		
Rate of gas re- lease increases quickly, accom- panied by small bursts of gas.	Steady state Any temperature	High	Fission-gas pres- sure breaking the specimen.			
Same as above	Steady state Any temperature	Low	Chemical reaction	Low-temperature irradiation shows greater surface area Higher temperature accelerates effect, which is irreversible. Only a small shift in isotopic composition toward longer-lived isotopes.		
Rate of gas re- lease increases in rapid exponen- tial manner.	Steady state Relatively high temperature	High	Fission-gas pres- sure exceeding the strength of the matrix, causing break- away gas release.	Slightly lower temperature will stop the increase in gas release rate, but the steady-state level will be increased compared to established levels. During the time of increased re- lease rate the gas shows a large shift in composition toward long-lived isotopes.		

Table 1. (Cont'd)

the second of the second second

د بو ويه در طوعه د

and a second a construction and a second states

_

Ob servati on	I rradia tion Condition	Burnup Level	Interpretation	Confirming Tests Later irradiation shows higher release rates from the same conditions. Low-temperature irradiation shows increase in surface area.		
Gas release rate constant but large burst when speci- men is cooled rapidly.	Rapid cooling from moderate or high temperature	Any	Specimen is crack- ing because of com- bination of thermal and fission-gas stresses.			
Momentary burst of gas when specimen is heated rapidly.	Start of irradiation	Any	Normal response of fuel	Will happen always in about the same manner.		
After irradiation stops, the xenon release continues at a decreasing rate.	Specimen cool Reactor down	Any	lodine released during irradiation and plated out on- to the walls of the sweep system is decaying into xenon.	Xenon dies off with the half-life of the iodine precursor. Xenon-135m is present. Krypton isotopes are not present.		
After irradiation stops the xenon and krypton re- lease continues at a decreasing rate.	Specimen cool Reactor down	Any	Xenon and krypton diffusing through interconnected porosity in the specimen.	Ratio of fission gas isotopes continually changing as short- lived isotopes decay away. Xenon isotopes do not decrease as the half life of the iodine precursor		

Table 1. (Cont'd)

.

......

4 **a** 1

Otservation	Irr a di a tion Condition	Burnup Level	Interpretation	Confirming Tests		
Pip or burst of fission gas after the irradiation has stopped and the specimen has cooled.	Reactor down and specimen cool for some time. Specimen has previously been irradi- ated at high temperature.	Medium to high	Iodine collected into a closed pocket at the surface of the specimen during the high-tempera- ture irradiation. Decay of iodine into xenon caused pressure to rupture pocket.	Pip contains mostly xenon iso- topes in the ratio of trapped gas. Later irradiation shows no surface area change.		

N

REFERENCES

- J. R. MacEwan and P. A. Morel, "Migration of Xenon Through a UO₂ Matrix Containing Trapping Sites," <u>Nucl. Appl.</u> 2(2): 158-170 (1966).
- T. S. Elleman, L. D. Mears, R. P. Christman, "Rare Gas Diffusion in Cesium Iodide - The Use of Fission Recoil Doping Techniques,"
 <u>J. of Amer. Caram. Soc.</u>, 51(2): 560-564 (1968).
- 3. R. S. Nelson, "The Influence of Irradiation on the Nucleation of Gas Bubbles in Reactor Fuels," J. Nucl. Mater., 25: 227-232 (1968).
- 4. R. M. Carroll, "Fission-Gas Behavior in Fuels," <u>Nucl. Safety</u>, 10(3): 210-216 (May-June 1969).
- 5. R. M. Carroll, ibid, 8(4): 345-353 (Summer 1967).
- 6. R. M. Carroll and O. Sisman, "In-Pile Fission-Gas Release from Single-Crystal UO₂," Nucl. Sci. Eng., 21(2): 147-158 (1965).
- 7. R. M. Carroll and O. Sisman, "Fission-Gas Release During Fissioning in UO₂," Nucl. Appl. 2(2): 142-150 (1966).
- 8. R. M. Carroll, R. B. Perez, O. Sisman, 'Release of Fission Gas During Fissioning of UO₂," <u>J. Amer. Cersm. Soc</u>., 48(2): 55-59 (1965).
- 9. R. M. Carroll and P. E. Reagan, "Techniques for In-Pile Measurements of Fission-Gas Release," <u>Nucl. Sci. Eng.</u>, 21(2): 141-146 (1965)
- P. E. Reagan, J. G. Morgan, and O. Sisman, "Fission-Gas Release from Pyrolytic-Carbon Coated Fuel Particles During Irradiation at 2000 to 2500°F," Nucl. Sci. Eng., 23(2): 215-223 (1965).
- 11. R. M. Carroll, J. G. Morgan, R. B. Perez, and O. Sisman, "Fission Density, Burnup, and Temperature Effects on Fission-Ges Release from UO₂," Nucl. Sci. Eng., 38(2): 143-155 (1969).
- R. M. Carroll <u>et al.</u>, Fuels and Materiels Development Program Quart. Progr. Rept. March 31, 1959, USAEC Report ORNL-4420, pp 72-76.