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A Review of Selected Aspects of the Effect of Water Vapor on Fission Gas Release from Uranium Oxycarbide

B. F. Myers

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ORNL/TM-12641

Metals and Ceramics Division

A REVIEW OF SELECTED ASPECTS OF THE EFFECT OF WATER VAPOR ON FISSION GAS RELEASE FROM URANIUM OXYCARBIDE

B. F. Myers

Date Published: April 1994

Prepared for the
U.S. Department of Energy
Office of Advanced Reactor Programs
EE 51 01 00 0

Prepared by the
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37831-6285
managed by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
U.S. Department of Energy
under contract DE-AC05-84OR21400

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A REVIEW OF SELECTED ASPECTS OF THE EFFECT OF WATER VAPOR ON FISSION GAS RELEASE FROM URANIUM OXYCARBIDE*

B. F. Myers

ABSTRACT

A selective review is presented of previous measurements and the analysis of experiments on the effect of water vapor on fission gas release from uranium oxycarbide. Evidence for the time-dependent composition of the uranium oxycarbide fuel; the diffusional release of fission gas; and the initial, rapid and limited release of stored fission gas is discussed. In regard to the initial, rapid release of fission gas, clear restrictions on mechanistic hypotheses can be deduced from the experimental data. However, more fundamental experiments may be required to establish the mechanism of the rapid release.

1. INTRODUCTION

The effect of water vapor on the release of fission gas from coated fuel particles with exposed kernels under irradiation conditions has been measured and an analysis reported.[1] This seminal study has shown that the general sequential response of exposed fuel kernels† to the addition of water vapor consists of three stages: (1) a rapid release of fission gas with a concomitant increase in the steady-state release, (2) a period of constant steady-state release, and, upon cessation of water vapor injection, (3) a decline in the release to prehydrolysis values except where configuration changes occur in the particles with exposed kernels. The experiments were conducted in the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory and were designated as HRB-17 and HRB-18.

The interest shown in the published measurements and analysis, as well as related experimental and analytical efforts in other laboratories, has indicated that an elaboration and greater emphasis on certain aspects of the study would enhance understanding and promote a better coupling of all the results.

The aspects of the measurements and analysis on which focus will be placed here involve fuel composition, release mechanisms operative in stages 1 and 2 of the general sequential response, and the decline in release during stage 3.

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*Research sponsored by the U.S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

†The fuel is in the form of a sphere and is referred to as the fuel kernel of the coated fuel particle. The configuration of the fuel in the coated fuel particles dispersed in carbonaceous fuel compacts and the use of the so-called "designed-to-fail" particles to ensure a known source of exposed kernels for the irradiation experiment have been described previously.[1]

2. FUEL COMPOSITION

The fuel, uranium oxycarbide, designated as UCO, is a mixture of UO_2 and UC_2 (actually UC_{2-x} where x is small).[2] The initial overall composition of the fuel kernel was $UC_{0.4}O_{1.6}$; thus, the kernel was 80% UO_2 and 20% UC_2 before irradiation began. Of importance here is the changing composition of those particles with exposed kernels, i.e., those particles which contribute to fission gas release.

During the normal course of any irradiation, the carbide phase (i.e., UC_2) is consumed in several ways, namely: (1) by the fissioning of UC_2, (2) by reaction of the UC_2 with oxygen released in the fissioning of UO_2, and (3) by reaction of UC_2 with metal oxides (mainly BaO, SrO, and ZrO_2) formed by reaction of fission products with the oxygen released in the fissioning of UO_2. The fraction of UC_2 remaining at any time, in the absence of water vapor or other oxidants, can be shown, with an error of about 10%, to be:

$$f_{UC2} \approx f^{o}_{UC2} - (0.53 + 0.47 f^{o}_{UC2}) \cdot B_u \,, \tag{1}$$

where

f_{UC2} = mol fraction of UC_2 remaining,
f^{o}_{UC2} = initial mol fraction of UC_2,
B_u = burnup [(fima) = n_f/n_o; n_f is the number of heavy metal atoms fissioned, n_o is the number initially present].

The changing composition of the fuel kernel from the beginning of irradiation to the beginning of the second water-vapor injection test, test number 2,* is shown in Table 1 for experiment HRB-17.

During the second water-vapor injection test, however, the remaining carbide phase is completely hydrolyzed and is thereby not present during subsequent tests. The evidence for the absence of UC_2 in the subsequent test, test number 3, is based on (1) the absence of spikes in the ionization chamber record for test 3, which were observed in test 2 when carbide was present; (2) kinetic calculations involving the reaction rate of UC_2 with water vapor, which are consistent with the time interval between water addition and the initial spike in test 2 and consistent with the complete hydrolysis of the carbide portion of the fuel; and (3) the absence in the region of constant steady-state release, stage 2, of a detectable decrease in release in test 3 that was present (and resolvable) in test 2 when UC_2 was present.

Experimental evidence indicates that the fractional contribution of the carbide phase to fission gas release in the presence of water vapor depends on the mechanisms of release and the chemical changes induced by hydrolysis. The fractional contribution to release of fission gas atoms contained in bubbles is less than the nonzero fractions of remaining UC_2 as listed in Table 1. During and after hydrolysis, the contribution to the release of interstitial atoms from the newly formed, low-density UO_2 is larger than before.

In effect, experiments HRB-17 and HRB-18 involved mainly the response of UO_2 to the presence of water vapor. Fortunately, there was a sufficient quantity of UC_2 present in tests 1 and 2 to permit

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*The liberty is taken of referring to the water-vapor injection tests by the number associated in Table 1 with each test; thus, 1, 2, 3, or 4.

Table 1. Data on the sequence of water-vapor injection tests in HRB-17

Test order	EFPD[a]	P_{H2O}[b] (Pa)	T (°C)	Remaining UC_2[c] fraction	B_u[d] (fima)[e,f]
				0.20	
1	33	20.6	778	0.13	0.11
2	58	199.0	779	0.10	0.16
3	109	186.0	755	0.00	0.22
4	151	64.8	756	0.00	0.25

[a]EFPD = effective full power days (at the beginning of the test).
[b]P_{H2O} = the partial pressure of water vapor (Pa).
[c]UC_2 = The initial mol fraction of UC_2 was 0.20. As a result of fissioning, reaction with oxygen from UO_2 fissions, and via the formation of fission product carbides, the mol fraction of UC_2 would decrease in the absence of water vapor.[3] In the second water-vapor injection test, the remaining UC_2 was hydrolyzed as indicated by the ionization chamber records for the second and subsequent tests. The entries in column 5 were calculated using Eq. (1).
[d]B_u = burnup (fima = n_f/n_o).
[e]n_f = number of metal atoms fissioned.
[f]n_o = number of initial metal atoms.

Note also that the values given in column 5 represent the mol fraction of UC_2 remaining at the beginning of each water-vapor injection test.

observations to be made on the response of UC_2 to water vapor, including the important observation of a sequential interaction of the fuel kernels with water vapor.

3. RESPONSE OF EXPOSED KERNELS TO WATER VAPOR

3.1 THE INITIAL RESPONSE: STAGES 1 AND 2

As a basis for discussion, the R/B* profiles for ^{85m}Kr, ^{88}Kr, and ^{87}Kr during the water-vapor injection test (no. 3 of Table 1) at 186 Pa and 755°C in the period 109.07 to 109.45 d (EFPD)* are presented in Fig. 1. In order to understand the data, as presented, one must know that data prior to the start of water-vapor injection and the data in the plateau region, i.e., the data beyond 109.45 d in Fig. 1, are represented by the average value of R/B in the time intervals 107.20 to 109.07 d and 110.2 to 114.05 d, respectively. On Fig. 1, these are shown as the thick line segments drawn prior to 109.07 d and following 109.45 d. The average standard deviation for these data is 8%. See also Figs. 3 and 4 of ref. 1.

There are, in Fig. 1, three clearly distinct regions; two of these, indicated by the thick line segments, represent periods of steady fission gas release. The third, intermediate region, represents a growth and decline of fission gas release. The decline to a steady release suggests that, to some extent, the gas released in this region comes from a limited source or availability of fission gas. These three regions are also distinguished by the parameter n in the equation:

$$R_i = g(b,F,T) \cdot B_i/\lambda^n , \qquad (2)$$

where

- R_i = release rate for isotope i (atom/s),
- g = a function of burnup, b, fission rate density, F(fission/$m^3 \cdot s$), and temperature, T(K),
- B_i = birth rate for isotope i (atom/s),
- λ = the isotopic decay constant (1/s),
- n = a parameter.

3.2 A MEASURE OF RELATIVE RELEASES

During the short time interval of the third, intermediate region, to a good approximation, the quantities b, F, T, and B of Eq. (2) may be regarded as constant. For a given isotope then, $R = K/\lambda^n$. Thus, n can be viewed as a measure of the relative release of the isotopes of a given element, in this case, Kr or Xe. A low value of n corresponds to comparable release rates or quantities over a specific time interval for the isotopes of a given element; a large value of n corresponds to a preponderance of long-lived isotopes and a relative scarcity of short-lived isotopes released in the specific time interval.

The three regions of Fig. 1 are not only distinguished by the R/B values but also by the values of n (see Fig. 5 of ref. 1). A consequence of this is indicated by values of the ratio R_i/R_j where i and j

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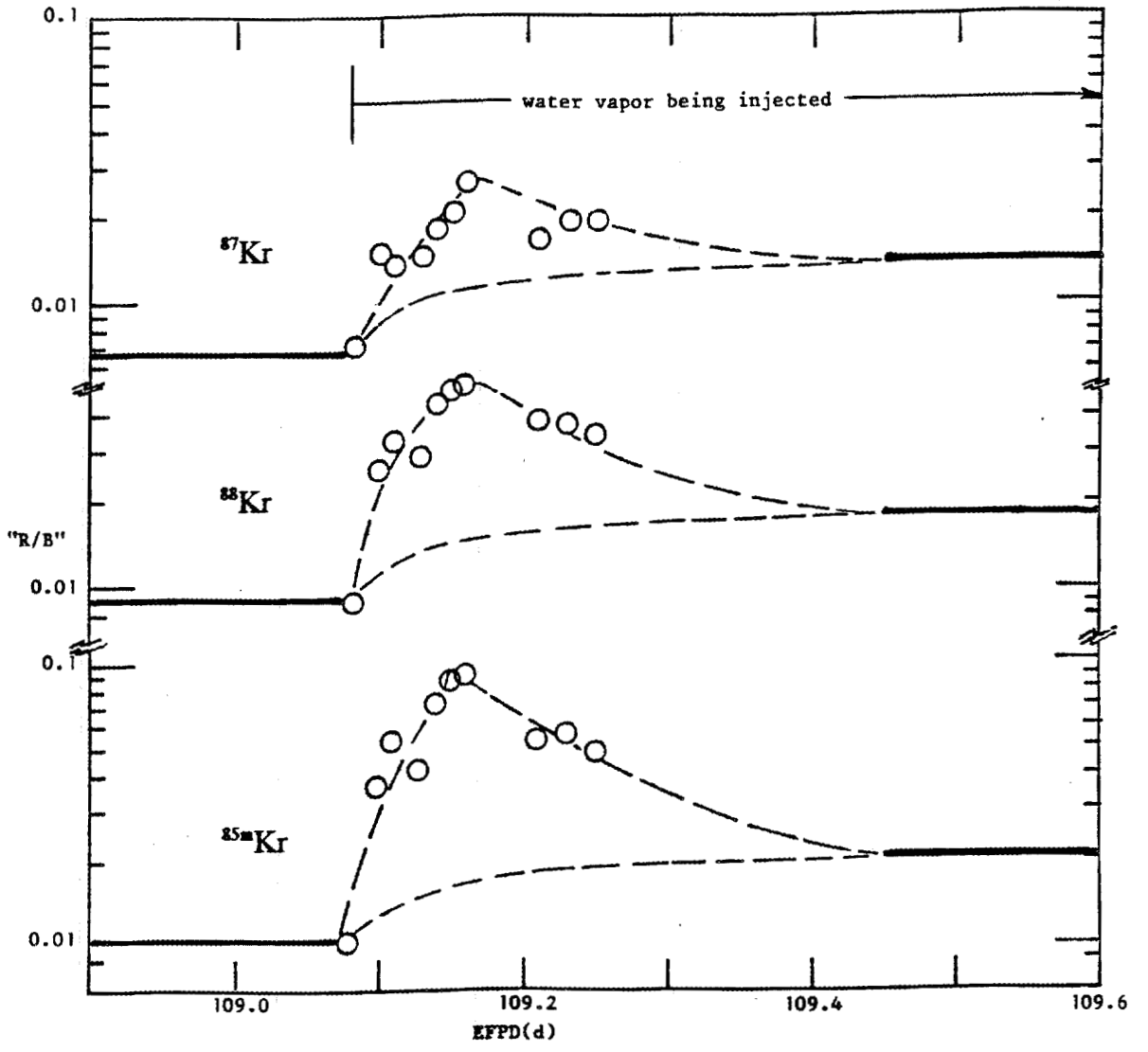
*R/B represents the ratio of the release rate to the birth rate; EFPD represents effective full power days.



average value of R/B in the time intervals 107.20 to 109.07 d and 110.20 to 114.08 d (the latter interval encompasses all the R/B data between 109.45 and 114.08 d)

Fig. 1. R/B profiles in the early portion of the water-vapor injection test at a partial pressure of 186 Pa and at 755°C in experiment HRB-17.

represent different isotopes. The value of the ratio R_{85mKr}/R_{87Kr} in the intermediate region is about three times larger than in the regions of constant R/B; for R_{133Xe}/R_{138Xe}, the corresponding factor is 12. These experimental results are an important indicator of the mechanisms of release and set the challenge for interpretation.

4. MECHANISMS OF FISSION GAS RELEASE

4.1 DIFFUSIONAL RELEASE

Diffusional release is associated with relatively smaller values of n. The value required by the standard diffusion model, i.e., the Booth model, is 0.5. Values of n in the range 0.2 to 0.4 are common in the experience of the U.S. high-temperature gas-cooled reactor (HTGR) community when measuring fission gas release in the absence of oxidants and from configurations involving coated fuel particles with exposed kernels, often dispersed within a fuel compact. The basis of the values of n smaller than 0.5 has not been definitely established. However, among possible contributory causes are release of fission products recoiled into the buffer coating surrounding the fuel kernel, the relatively high burnups (~ 25% FIMA) of irradiated fuel kernels in conformance with prismatic core, gas-cooled reactor designs, and, perhaps, the effect of high fission rate densities.[4] For example, the diffusional release of recoiled fission products from the buffer is more rapid than from the fuel kernel;* the more rapid release results in a lower value of n.

In the two regions of steady R/B values of Fig. 1, the value of n is about 0.35 (see Fig. 5 of ref. 1). Thus, in these regions, diffusional release is dominant. Furthermore, the value of n is the same in the regions of constant steady-state release before and during water-vapor addition. This suggests that the same mechanism or mechanisms are dominant in these two regions and that the presence of water vapor does not alter the diffusional mechanism or mechanisms dominant in the two regions.

For a diffusional mechanism, one expects, in the water-vapor injection tests conducted at constant temperature, that R/B will rise as more exposed kernels are reached by the water vapor† and will rise at a continually decreasing rate until the rate of increase approaches zero when all exposed kernels are interacting with the water vapor. There is no expectation that a peak in the R/B profile will occur. The lower dashed curves in Fig. 1 are drawn in accord with the attribution that (1) the initial and final R/B values for the diffusive release are given by the thick line segments, (2) the curve connecting the initial and final values is monotonically increasing, and (3) the shape of the curve is consistent with ionization chamber traces and analysis of the R/B profile for ^{135}Xe.‡

Thus far, the analysis, referenced and graphically presented data, and the discussion lead to the conclusion that the thick line segments and the connecting (lower) dashed curves of Fig. 1 represent

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*During the course of irradiation, the buffer does densify and the release rate of recoiled fission gas atoms declines to some degree.

†That water vapor reaches each kernel sequentially over a substantial period of time is consistent with ionization chamber chart records and with the profile peaks such as shown in Fig. 1. See ref. 1.

‡As a result of an (n,γ) reaction, the effective lifetime of ^{135}Xe is sufficiently small, under HFIR conditions, to prevent storage in bubbles or voids. This is in contrast to the other monitored isotopes as illustrated by the interpretation presented in Fig. 7 of ref. 1.

the diffusive release of fission gas. This diffusive release is that normally encountered in the absence of water vapor both from fuel specimens and from coated fuel particles with exposed kernels. The operative mechanisms for the former case have been pointedly reported;[4] in the latter case, these mechanisms, plus the additional mechanism of fission product recoil into the buffer with subsequent diffusive release, apply. A consequence of the conclusion is the applicability of the value of n ≈ 0.35, not only to the region of the thick line segments but also to the lower dashed curves in the intermediate region.

The fact that the n value is the same in the regions of steady-state release, represented by the thick line segments, and is deduced to be the same in the intermediate region has an implication for the release process. The argument previously used[1] can be applied here. Consider a distance of order $\sqrt{D\tau}$ where D is a diffusion coefficient and τ is the lifetime of an isotope (defined as a time by which only a specified small fraction remains radioactively undecayed). The argument for a constant n is as follows. If an isotope is within a distance of order $\sqrt{D\tau}$ of a surface, it can escape from the fuel; otherwise, it will decay radioactively before escape. The quantity that escapes, therefore, is proportional to the volume of the surface layer. When water vapor interacts with the fuel, the density decreases and D can increase. Consequently, the quantity of an isotope escaping will increase by a factor independent of the particular isotope of an element. Thus, the relative quantities of the isotopes escaping and, therefore, the n value will remain unchanged. The increasing diffusive release in the intermediate region represents the contribution of the increasing number of particles with exposed kernels interacting with the water vapor. The argument used here implies that the n value remains constant in the intermediate region as well as in the regions of steady-state release.

4.2 STORED FISSION GAS RELEASE

In the intermediate regions of Fig. 1, in contrast to the regions of the thick line segments, the n values increase from 0.35 to larger values and then decline to 0.35. The maximum value is ≈ 1.1. If the diffusionally released gas has an n value of ≈ 0.35 throughout the time period considered in Fig. 1, as concluded above, then the occurrence of larger n values indicates a contribution to the intermediate region from a release or releases of fission gas in which diffusion of individual fission gas atoms is not the dominant or significant mechanism.

In the analysis of the experiments, the large area between the two dashed curves was attributed to the release of stored fission gas. The stored fission gas is regarded as contained in a closed pore or bubble. The relative concentrations of the isotopes within the bubble, of which n is a measure, are maintained by the balance between entering fission gas atoms and radiodecay of the contained atoms. (Re-solution of the contained gas atoms does not significantly affect the n value.) The n values of stored fission gas can be as large as 1.5. The n value of the mixture resulting from the release of fission gas by diffusion and from stored gas would yield intermediate n values. As shown in Fig. 5 of ref. 1, the n values in the intermediate region ranged, for Kr, from 0.35 to ≈ 1.1 at the peak, consistent with expectations.

If stored fission gas were being released directly, one would expect the release as a fraction of the inventory to be the same for all isotopes at a given temperature, partial vapor pressure of H_2O, irradiation history, and for a given fuel. In the present experiments, only the constancy of the fractional release at constant conditions and the variation with the partial pressure of H_2O could be adequately examined. The dependency on temperature and irradiation history can be examined during the analysis of the recently completed experiment HFR-B1. Establishing the dependency on fuel composition would require new experiments.

The experimental results shown in Table 2 verify that the release of fission gas as a fraction of the inventory is the same within experimental error for all isotopes of both krypton and xenon under fixed conditions and that the fraction varies with the partial pressure of H_2O. Therefore, the results are consistent with the direct release of stored fission gas.

A further relevant experimental result[1] is the constancy of the time interval between the injection of water vapor and the peak in the release curve; the average time interval and standard deviation for all isotopes monitored are 2.0 ± 0.2 h.* This constancy is also evident in Fig. 1 for the krypton isotopes in one of the water-vapor injection tests. If diffusion of individual fission gas atoms were the dominant mechanism, this time interval would be expected to vary with the half-life of the isotope. It is necessary to consider this matter in sightly more detail.

The individual particles with exposed kernels interact with the water vapor sequentially;[1] this was attributed[1] essentially to a slow movement of water vapor into the fuel compacts. If the result were a rapid release of gas from each exposed kernel (say, $\Delta t \approx 1$ min), the profile would consist, generally, of a series of well-separated spikes ($\langle \Delta t \rangle \approx 18$ min). This is illustrated by the ionization chamber chart profile for test 2 during the rapid hydrolysis of the carbide phase in the fuel kernel (see Fig. 8 of ref. 1). By contrast, the R/B profiles, such as those in Fig. 1, show roughly a smooth rise to a peak and then a decline.† This indicates that the release from the oxide phase of a single exposed kernel occurs over a much larger time period than that of the carbide phase, at least as indicated by the ionization chamber chart record. Furthermore, a constancy in the peak location of the profile of fission gas release is incompatible with a rapid sequential release of the fission gas from each exposed kernel. The time period for stored gas release, as can be shown for a pattern of sequential release, is equal to the time interval between the injection of water vapor and the peak in the release curve; thus, as given above, the release of the fission gas from each exposed kernel occurs over a period of 2 h.

5. DISCUSSION OF MECHANISMS

5.1 GENERAL ASSESSMENT

In the preceding text, the focus has been on distinguishing two mechanisms of fission gas release referred to, for convenience, as "diffusional release" and "stored gas release." The intention in writing the present text was to present in more detail than previously[1] the evidence on which the distinction is based and to stress the basic nature of it. Without an acceptance of this distinction, the

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*The average time interval and standard deviation are derived from tests 3 and 4. For test 2, fission gas samples were not taken beyond the 2-h time interval so that one can state only that the time interval of test 2 could be 2 h but cannot be demonstrated to be so for lack of data. For test 1, the scatter of fission gas release data was too large to determine a definitive value of the time interval; an estimate, based on a least-squares fit of the scattered data, yields an average of 1.8 h.

†The contribution of the spikes of fission gas release in test 2 to the R/B profiles for this test are negligible on the grounds that (1) the frequency of release measurements (from which R/B values are derived) is significantly smaller than that of the occurrence of the spikes and (2) the contribution of the carbide-phase hydrolysis to fission gas release via the spikes is no more than 10% (assuming that the quantities of fission gas contained in the UC_2 and UO_2phases are proportional to the quantities of each phase).

Table 2. The fractional release, f_R, of stored fission gas for each isotope as a function of its inventory

Test order	1	2	3	4	
P_{H2O} (Pa)	20.6	199.	186.	64.8	
Fraction of UC_2 remaining	0.13	0.10	0.00	0.00	
Isotope	**f_R 10^{-4} [a]**	**f_R 10^{-2}**	**f_R 10^{-2}**	**f_R 10^{-3}**	**half-life**
^{85m}Kr	4.4	4.1	3.2	2.7	4.48 h
^{87}Kr	8.1	4.0	3.5	3.3	1.27 h
^{88}Kr	4.5	3.7	3.1	2.9	2.84 h
^{133}Xe	[b]	4.3	3.4	[c]	5.24 d
^{138}Xe	[b]	3.8	3.7	3.0	0.24 h
$\langle f_R \rangle$ [d]	5.7	4.0	3.4	3.0	
std [e]	2.1	0.24	0.24	0.25	
%std [f]	37.	6.	7.	8.	

[a] f_R = If $(f_R)/10^{-4} = 4.4$, then $f_R = 4.4 \cdot 10^{-4}$, for example.
[b] Necessary data lacking for the calculations of fractions.
[c] Inventory not established at time of stored gas release.
[d] $\langle f_R \rangle$ = the average f_R for all isotopes (Kr + Xe isotopes).
[e] std = standard deviation.
[f] %std = percent standard deviation.
Note: Data for test 2 are slightly different from data in ref. 1. These data should have appeared in ref. 1 for being based on the final calculational method. Data in ref. 1 were based on an earlier version of the calculational method.

interpretation remains open, but only to a degree, as the experimental observations provide strong bounds for any interpretation. Furthermore, as will be shown in future reports, the interpretation presented here is sustained over a wider range of temperatures and water-vapor pressures than encountered in experiments HRB-17 and HRB-18.

The "diffusional release" subsumes different specific mechanisms for the movement and eventual release of individual fission gas atoms from the fuel kernel and buffer layer of the HTGR coated fuel particles with exposed kernels. In so far as this mechanism has been applied in the interpretation of the HRB experiments, any corresponding questions are regarded as relatively minor in the context of this discussion.

Given the distinction between the "diffusional release" and the "stored gas release," the major questions involve the mechanism of the release of stored fission gas in the presence of water vapor and the limited time interval over which the stored fission gas is released from each particle with an exposed kernel. It is unlikely, on the basis of the experimental observations to date, that a mechanism involving the effect of water vapor on the release of stored fission gas can be satisfactorily established. Nevertheless, the consistency of the observations with the expectations based on the release of stored fission gas should sustain continued, strong interest in pursuing a mechanism.

5.2 HISTORICAL PERSPECTIVE

As stated above, the stored fission gas is contained in bubbles. The study of fission gas bubbles in fuel has been extensive and of long standing.[5] The general processes involved include nucleation, growth, re-solution, migration, coalescence, interaction with crystal defects, and release of the contained fission gas. The intensive interest has been motivated by the fuel swelling induced by the formation of fission gas bubbles and the release of fission gas via bubbles under the influence of strong gradients, particularly temperature gradients. A lack of such interest has existed in the HTGR community; the coated fuel particle is designed to accommodate the swelling of the fuel kernel, and the release of stored fission gas has been observed only at temperatures beyond the range of interest. However, in the presence of oxidants, the release of fission gas from bubbles becomes important for the HTGR under otherwise normal operating conditions.

5.3 THE DIRECT RELEASE OF STORED FISSION GAS

By the direct release of stored fission gas is meant the release of the isotopes of a given element having the same n value as exists within the bubble in which the stored gas resided before release. This definition is an idealization which cannot be confirmed easily by measurement. In practice then, the values of n deduced from the measurement of the quantities of released fission gas are required only to have high values, consistent with the values expected for stored fission gas, after taking into account the effects of mixing with the "diffusional release" gas which has lower values of n.*

Establishing a mechanism of the release of stored fission gas begins with the consideration of the motion of bubbles in the fuel kernel and with the recognition, as Table 2 demonstrates, that the release of the monitored fission gas isotopes with high n values, as a fraction of their inventories, is independent of the half-life. Therefore, if the released fission gas comes from bubbles, the release tends to be direct.

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*This suggests that a further analysis to obtain n values for the stored fission gas release could be undertaken, as it has not been to date. For the present, reliance is placed on the argumentation employed in the preceding text (see Sects. 4.1 and 4.2).

Consider the bubble motion as the sum of movement within the fuel kernel and escape from it. During movement within the fuel kernel, the value of n is apparently preserved in some manner. If the n value varied significantly, assuming the same inventories as in the case of constant n, the half-life independence of the fractional release of the isotopes would not be observed. A variation in n would result from changes in the rate at which the isotopes enter the bubble. The sensitivity of the independence on half-life to variations in n and the time profile of n during bubble motion need to be addressed in more detail. During the release phase, the bubbles reach a surface where conditions promote, in effect, escape of the stored fission gas into the externally connected pore system in a time such that the monitored isotope with the smallest half-life decays negligibly.*

The movement of the bubbles can be random, or in the presence of a force, directed. Random movement of bubbles in regard to the release of fission gas is clearly not important under the conditions of the HRB experiments; there is no evidence for significant direct release of fission gas from bubbles in the absence of water vapor. Apparently, the temperature gradients across the fuel kernel are too small to provide a significant, directed force.

5.4 OUTLINES OF A WORKING MODEL FOR STORED FISSION GAS RELEASE

In the experiments, a constant vapor pressure of water was maintained. As a consequence, there was a steady distribution between the water molecules in the vapor phase and those adsorbed, or perhaps, dissociatively adsorbed, on the surfaces of the fuel kernel. The surface-adsorbed water molecules can reasonably be expected to have undergone further configurational changes, interactions, and movement into the bulk fuel. The presumption is adopted, as a working model, that the inward movement of the water-derived species, and their interactions with the fuel as penetration increases, results in gradients which induce bubble movement toward the surfaces. The nature of the gradients remains to be determined. Possibilities include density gradients and other stress gradients established, perhaps, as a result of the concentration gradients of the inward moving species and of their effect on the local fuel structure. After a certain relaxation time, the entire system, i.e., the bulk, surface, and vapor phases, will be in a steady-state relation, and the phases will each be homogeneous on a macroscale, both in some approximation. In approaching the latter conditions, the gradients are expected to decline and become insignificant; correspondingly, the movement and escape of the bubbles will decline and become insignificant. The adopted working model is consistent with the release of stored fission gas via bubble movement and with the limited time over which stored fission gas is released. If the working model is basically correct, then its elaboration will account for the limited time period over which the release of stored fission gas from each kernel is deduced to occur.

The working model must principally account for the dependence of the fractional release of the stored fission gas on the partial pressure of water vapor. This implies that the inverse fraction of the bubbles containing fission gas is not affected by the interactions of the water-derived species, at the least, to the extent of inducing fission gas release or the processes that lead to release.

In an alternate working model,[6] less emphasis is placed on the motion of bubbles, and more is placed on the chemical effects of the water-derived species on the local structure of the fuel kernel. The postulate is made that the porosity of the fuel increases and thus permits the escape of fission gas from those bubbles for which the balance between the bubble gas pressure and the stress and surface-tension of the surrounding solid is sufficiently perturbed by the chemical effects.

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*Note that the values of the release rate, R, measured at the gas sampling station, have been corrected for the transit time from the capsule to the sampling station and therefore represent the fission gas release rates in the capsule at the fuel compact locations.

6. DECLINE OF R/B IN STAGE 3

Stage 3 begins when the water-vapor injection is terminated. Following this, the R/B declines to the prehydrolysis value provided the configurations of the particles with exposed kernels have not changed during either stages 1 or 2. This return to the prehydrolysis R/B value has been attributed to the sintering and densification of the fuel kernel.[1]

In a preliminary analysis, a characteristic time for the recovery has been determined. The decrease in the R/B as a function of time is governed by the function $\exp\{-\delta(t - t_T)\}$ where δ is a constant, t the time, and t_T the time at which the water-vapor injection is terminated.[1] The values of δ, determined from the experimental data, are given in Table 3.

The time for the function $\exp\{-\delta(t - t_T)\}$ to decline to 0.05, at which time the approach to the prehydrolysis R/B value is 95% complete, is given by:

$$\Delta t_{95} = 3/\delta \ . \tag{3}$$

In Eq. 3, $\Delta t_{95} = t - t_T$. These times, Δt_{95}, are given in Table 3. If the assumption is made that Δt_{95} is inversely proportional to the fission rate density, i.e.,

$$\Delta t_{95} = K/F \ , \tag{4}$$

where

K = constant (fissions$\cdot$ d)/(m$^3\cdot$ s) = $2.27\cdot10^{21}$ fissions$\cdot$ d/m$^3\cdot$ s,
F = fission rate density (fissions/m$^3\cdot$ s),

the calculated values of Δt_{95} according to Eq. 4, as shown in Table 3, are essentially the same as the values determined with Eq. 3.

Table 3. The R/B recovery times and the relation to fission rate density

Experiment	δ(1/d)	F(10^{20}f/m$^3\cdot$ s)[a]	T(°C)	Δt_{95}[d] Eq. (3)	Δt_{95}[d] Eq. (4)
HRB-17	0.425[b]	3.2[c]	770	7.1	7.1
HFR-B1	0.109[d]	0.84[e]	925[f]	27.5	27.0

[a]f = fissions.
[b]See ref. 1.
[c]F at 90 EFPD (108 elapsed days).
[d]B. F. Myers, unpublished data.
[e]F at 100 EFPD (cycle 88.10).
[f]Estimated fuel temperature, subject to change upon further thermal analysis.

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