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THE BEHAVIOR OF FISSION PRODUCT GASES IN HTGR FUEL MATERIAL

by

B. F. MYERS, N. L. BALDWIN, W. E. BELL,
and R. D. BURNETTE

Prepared under
Contract EY-76-C-03-0167
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GENERAL ATOMIC COMPANY

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ABSTRACT

This document is a status report on experimental data relating to the behavior of fission product gases in HTGR fuel material. It contains (1) a review of published experimental data on the behavior of gaseous fission products (noble gases, halogens, and related elements) in HTGR fuel materials and (2) new data on fission gas behavior. From the experimental data, the effects of fission product element, fuel configuration, nuclide half-life, temperature, pressure, neutron flux, neutron fluence, and fuel hydrolysis on fission gas release are deduced. Also on the basis of the experimental data, a model is formulated for the release of fission gases from fuel rods. The data presented in this report are being confirmed and extended through continuing work at General Atomic and other laboratories.

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1. SUMMARY

This is a status report containing a review and analysis of available experimental data on the behavior of gaseous fission products (noble gases, halogens, and related elements) in High-Temperature Gas-Cooled Reactor (HTGR) fuel materials. The data consist of published and new data. From the data presented herein, the effects of the following variables on fission gas release are deduced: fission product element, fuel configuration, nuclide half-life, temperature, pressure, neutron flux, neutron fluence, and fuel hydrolysis.

The data indicate:

1. Fission gas release from HTGR fuel elements is governed by the amount of uranium and thorium contamination in as-manufactured fuel rods (defined as exposed uranium and thorium and including the contribution of particles with missing, permeable, or broken coatings) and by the number of particles which experience coating failure during irradiation.
2. Fission gas release from fuel configurations of interest for short-lived nuclides (half-life $\lesssim 5.3$ d) occurs primarily by a mechanism involving recoil from the fissile material followed by gas phase and bulk diffusion in the surrounding material at temperatures up to 1300°C at least.
3. The gas phase contribution to the release of short-lived nuclides is dominant at temperatures below 800°C and the bulk diffusion is

dominant at temperatures above 900°C; the activation energies for the latter process in fuel rods are 139 kJ/mol (33 kcal/mol) for krypton isotopes and 112 kJ/mol (27 kcal/mol) for xenon isotopes.

4. The steady-state fractional release for the short-lived nuclides depends on a power of the nuclide half-life; for reactor conditions, the exponent of the half-life is about 0.2 at 300°C and approaches 0.5 at 1100°C.
5. The steady-state fractional release, R/B, value for Kr-85m at 1100°C for the configuration of failed fuel particles with dense kernels and constrained coatings in fuel rods at low burnup is 5×10^{-3} .
6. For short-lived isotopes, R/B is independent of burnup over the range 0% to 63% FIMA for failed particles with dense UC_2 kernels; for failed particles with dense ThO_2 kernels, the data are insufficient to determine the burnup dependence.
7. The Kr-85m R/B value at 1100°C for uranium and thorium contamination in fuel rod matrix material is 0.30.
8. For long-lived and stable isotopes, release from the kernel of the fuel particles is by a diffusion-related mechanism. The release can be described at temperatures above 900°C by an average effective reduced diffusion coefficient which is a strong function of burnup and which is derived on the basis of the equivalent sphere model.
9. R/B is not appreciably affected by neutron flux (fission rate density) and neutron fluence, except for a possible increase due to coating failure.
10. The recommended R/B for Kr-85m at 1100°C for failed particles containing hydrolyzed carbide fuel is 0.10.

11. Iodine and tellurium isotopes should be treated as if they were xenon isotopes; bromine and selenium isotopes should be treated as if they were krypton isotopes.

The data presented in this report are being confirmed and extended through continuing work at General Atomic and other laboratories.

2. INTRODUCTION

The purpose of this report is to review experimental data on the behavior of gaseous fission products (noble gases, halogens, and related elements) in HTGR fuel material.

In Section 3, measurement techniques are described; and in Section 4, discussions and relevant data are presented on the effects of the fission product element, fuel configuration, nuclide half-life, temperature, neutron flux, neutron fluence, and fuel hydrolysis on fission gas release.

HTGR fuel consists of coated fuel particles bonded into fuel rods. The fuel rods are contained in graphite fuel elements. The present reference particles consist of two types: (1) a BISO-coated fertile particle and (2) a TRISO-coated fissile particle. The fertile particle consists of a thorium oxide fuel kernel, an inner buffer coating of low-density pyrolytic carbon, and an outer coating of high-density isotropic pyrolytic carbon. The fissile particle consists of a uranium carbide fuel kernel and successive coatings of a pyrolytic carbon buffer, an isotropic pyrolytic carbon, silicon carbide, and a second isotropic pyrolytic carbon.

The primary barriers to release of fission gas into the primary circuit of the HTGR are the coatings on fuel particles. Both pyrolytic carbon and silicon carbide coatings are very effective in retaining fission gases under normal operating conditions (Refs. 2-1 through 2-7*). Thus, release of fission gases from fuel particles depends on the failure of coatings. In addition to this source of release, fission gas may escape

*References appear at the end of each section.

as a result of contributions to fissioning from uranium and thorium contamination (defined as exposed uranium and thorium including the contribution of particles with missing, permeable, or broken coatings) in as-manufactured fuel rods.

The release of fission gases (except for long-lived and stable isotopes) is described by the fractional release at steady state as given by the ratio, R/B , of the release rate, R , to the birth rate, B , for each fission gas isotope. The total fractional release for each isotope is represented as

$$(R/B) = (R/B)_{am} + f(R/B)_f, \quad (2-1)$$

where $(R/B)_{am}$ is the ratio of the release rate of fission gas from contamination to the birth rate in all the fuel of the as-manufactured fuel rod, f is the fraction of fissions that occur in particles with coatings that fail during irradiation, and $(R/B)_f$ is the fractional release from fuel particles with constrained failed coatings in fuel rods. Furthermore,

$$(R/B)_{am} = C(R/B)_c, \quad (2-2)$$

where C is the fraction by weight of uranium or thorium contamination in the fuel rod and $(R/B)_c$ is the fractional release of fission gases born in the contamination.

For long-lived and stable isotopes, a relationship similar to Eq. 2-1 applies. Thus,

$$H = H_{am} + fH_f, \quad (2-3)$$

where H is the time-dependent fractional release. In addition to Eq. 2-3, a contribution to the release of long-lived and stable isotopes occurs

when each particle fails; the long-lived and stable isotopes accumulated in the free volume of the particle coatings are rapidly released (see Section 4.2.2).

Equation 2-1 is often written with the $(R/B)_{am}$ term multiplied by $(1-f)$. The factor $(1-f)$ is necessary if the contamination is associated with the coatings of particles which fail during irradiation. If the contamination is primarily in the matrix material, which is the most likely case, then Eq. 2-1 is the preferred form. In the application of Eq. 2-1, the terms would represent the average gaseous release contribution, $\overline{(R/B)}$, for as-manufactured fuel rods and failed fuel particles in the core or a core segment of the HTGR.

At the beginning of fuel life, fission gas release from HTGR fuel elements is governed by the amount of uranium contamination (including all exposed uranium) in the fuel rods. Late in life, the release is governed by the amount of uranium and thorium contamination in the as-manufactured fuel rods and/or the number of particles which experience coating failure during irradiation. Accordingly, in order to calculate the fission gas release for fuel during irradiation it is necessary to know the following:

1. Fission gas release, $(R/B)_{am}$, for as-manufactured fuel rods. The value of $(R/B)_{am}$ is derived from quality control tests during fuel manufacture. Present specifications for as-manufactured fuel rods require an R/B value of $\leq 3 \times 10^{-5}$ for Kr-85m at 1100°C. This criterion was satisfied for Fort St. Vrain fuel rods (Ref. 2-8).
2. Fission gas release from failed fuel particles, $(R/B)_f$. A value for $(R/B)_f$ is derived from studies described in Section 4.1.
3. The fraction of failed particles in the fuel rods as a function of irradiation. This is derived from information provided in Ref. 2-9.

4. The dependence of $(R/B)_{am}$ and $(R/B)_f$ on half-life. This is derived from data provided in Sections 4.2 and 4.3.
5. The fractional release of long-lived and stable gaseous nuclides. This is needed to establish (1) the fractional release upon particle failure and (2) the subsequent fractional release from particles with failed coatings. The release of stable and long-lived nuclides is discussed in Section 4.2.
6. The effect of temperature on $(R/B)_{am}$ and $(R/B)_f$. This is derived from data given in Section 4.3.
7. The effect of the neutron flux on R/B . This effect must be considered in deriving R/B values for high flux level (reactor, loop, and capsule) systems from laboratory (low flux level) test data. This effect is considered in Section 4.5.
8. The effect of neutron fluence (and burnup) on $(R/B)_{am}$ and $(R/B)_f$. Data on this effect are presented in Sections 4.2 and 4.6.
9. The release behavior of iodine, tellurium, selenium, and bromine nuclides. (See Section 4.7.)
10. The effect of carbide kernel hydrolysis on $(R/B)_f$. This effect is discussed in Section 4.8.

In fission gas release calculations, the relationship between R/B and variables such as temperature and half-life is usually based (Refs. 2-10, 2-11) on a simple diffusional model, which is described by the following relationship (Refs. 2-12, 2-13):

$$(R/B) = 3(D'/\lambda)^{1/2} [\coth(\lambda/D')^{1/2} - (D'/\lambda)^{1/2}] \quad , \quad (2-4)$$

where $D' \equiv D/a^2$, D' being the reduced diffusion coefficient, D the diffusion coefficient of the model, a the diffusion distance, and λ the nuclide decay constant. Strictly (Ref. 2-12), Eq. 2-4 represents classical (Fickian) diffusion in a sphere of homogeneous material in which the diffusant is generated at a constant rate in position and time under isothermal conditions with zero concentration of diffusant in the sphere initially and at the surface at all times. The actual diffusional mechanism of fission gas release from fissile material (Ref. 2-14) and from coated fuel particles (Ref. 2-15) is more complex. However, on the basis of the fission gas release data presented in Section 4, Eq. 2-4 will be useful in formulating a description of fission gas release from fuel rods.

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3. MEASUREMENT TECHNIQUES

The purpose of this section is to describe various measurement techniques utilized at General Atomic Company (GA) to provide fission gas fractional release (R/B) data for HTGR fuel. Laboratory measurements provide data for use in evaluating fuel quality and in predicting fission gas release behavior in loop, capsule, and reactor systems. In-pile (reactor, loop, and capsule) measurements provide information on fission gas release behavior for use in evaluating fuel performance and validating calculational methods.

3.1. LABORATORY TECHNIQUES FOR FISSION GAS RELEASE DETERMINATION

Laboratory measurement techniques relating to fission gas release consist of (1) TRIGA irradiation tests, used to perform fission gas release (R/B) measurements; (2) hydrolysis tests, used to measure heavy metal contamination in fuel specimens; and (3) the laser-drilling technique, used to simulate constrained failed fuel particles in fuel rods.

3.1.1. TRIGA Irradiation Tests

Fission gas release measurements are routinely performed on HTGR fuel specimens over a broad range of physical test parameters, such as temperature, flux, and geometry, by using a King furnace facility mounted in a TRIGA research reactor (Refs. 3-1, 3-2). For example, the TRIGA test was utilized to measure the quality of as-manufactured Fort St. Vrain fuel rods from the standpoint of fission gas release (Ref. 3-3).

Fission gas release values are obtained by irradiating fuel samples in the King furnace, which is a resistance-heated, graphite-tube furnace designed to fit into a fuel element position in the TRIGA reactor core. The samples, consisting of loose particles, fuel rods, or fuel bodies, are

irradiated to produce approximately 10^{14} fissions, regardless of uranium loading, by adjusting the reactor power level. During irradiation, the fission gases released are swept with helium from the furnace into a liquid-nitrogen-cooled charcoal collection trap. At the end of the irradiation period, normally 30 min, the collection trap containing the released fission gases is gamma-counted using a lithium-drifted germanium detector. The gamma-counting and R/B calculations are done by use of a Sigma II computer-analyzer and appropriate computer codes. The test yields a direct measure of the noble gas release, both krypton and xenon isotopes, at irradiation temperatures to 1800°C. The R/B values are calculated by comparing the release of gaseous radionuclides from the fuel with the release from a powdered sample of uranium stearate, irradiated and gamma counted under the same conditions. Uranium stearate has been shown to be 100% releasing for gaseous nuclides with half-lives >30 s (Ref. 3-4). An absolute counting procedure using the latest literature values of branching ratios, decay schemes, half-lives, and fission yields has shown excellent agreement with the stearate comparison method for determining R/B values (Ref. 3-5).

Nuclides with half-lives greater than about 5 min do not attain their steady-state release values during the routine 30-min TRIGA irradiation. Steady-state R/B values can be determined using equations developed to correct the short-term irradiation data (Ref. 3-6). Recently, a more direct approach has been to subject samples to long-term (>100 h) irradiation in the TRIGA reactor to obtain steady-state conditions; from these experiments, a series of steady-state correction factors were derived (a factor of 1.9 for Kr-85m at 1100°C) for the noble gas isotopes of interest. The data reported herein for fresh fuel samples subjected to 30-min irradiations have been corrected to steady-state conditions using the experimentally derived factors. Analogous tests on highly irradiated fuel samples have shown virtually no change in activity with length of irradiation. Therefore, the R/B values reported for highly irradiated fuel were not corrected for steady state.

A series of comparison tests were made on the same fuel specimens by GA and Oak Ridge National Laboratory (ORNL) (Ref. 3-7). The fuel sample exchange included both fuel particle and rod samples. The ORNL and GA R/B data show agreement within a factor of three, with the exception of measurements obtained for one sample which showed extremely low R/B values, approaching the limits of detection in the GA test. In general, there appears to be good agreement between the two sets of laboratory data, providing a measure of verification for the fission gas release measurements conducted at the two laboratories.

3.1.2. Hydrolysis Test for Determining Contamination

In the hydrolysis test, which is an out-of-pile test, the fraction of thorium contamination (i.e., exposed thorium) in fuel rods is measured (Ref. 3-8). The exposed thorium could be in the form of (1) contamination in the rod matrix material, (2) contamination associated with coatings, or (3) broken fertile particles. The hydrolysis test was developed for use as a quality control test to evaluate the level of thorium contamination in as-manufactured Fort St. Vrain fuel rods.

The main purpose of the hydrolysis test is to demonstrate that as-manufactured fuel rods conform to specified values for the level of thorium contamination. Meeting the specified values assures that the R/B values will not change appreciably with U-233 buildup in the reactor, provided that particle coating failure with time is not appreciable.

The test involves the chromatographic measurement of ethane evolved during hydrolysis of exposed thorium carbide in a fuel rod. Fuel rods submitted for testing are first annealed in vacuum at 1800°C for 1 h to convert exposed thorium oxide to the dicarbide, after which the rods are hydrolyzed and the gaseous decomposition products (including ethane) are collected for chromatographic analysis. A standard containing a known amount of thorium carbide is hydrolyzed using the identical procedure. To determine the fraction of exposed thorium, the amount of ethane evolved from the sample is compared with that from the standard.

Actually, the test measures the total of exposed thorium and exposed uranium, since exposed uranium carbide hydrolyzes as well as exposed thorium carbide. However, the uranium content of fuel rods is normally much lower than the thorium content; consequently, the level of exposed uranium is normally small compared with that of thorium. The level of uranium contamination can be derived from the TRIGA test (described above) utilizing a relationship between fission gas release (R/B) and fraction of exposed uranium (see Section 4.1).

The hydrolysis test has a shortcoming when applied to fuel rods containing exposed ThO_2 kernels. Long times are required to convert exposed ThO_2 kernels to ThC_2 because of their high density and relatively large size. Complete conversion requires large amounts of carbon, and carbon coatings on adjacent particles can be attacked resulting in a cascade reaction. To prevent the latter, the time at high temperature is set so that only partial conversion occurs and a correction factor is applied. This yields an incorrect (high) result if no exposed ThO_2 kernels are present (i.e., if only thorium particulate contamination is present). Because of this shortcoming and since reference-type fuel rods contain coated ThO_2 particles, alternative methods for measuring exposed thorium are being sought.

3.1.3. Laser-Drilling Technique

The laser-drilling technique was developed to provide simulated failed fuel particles for studying the effect of coating failure on fission gas release. Laser-failed particles are considered to be representative of constrained failed particles in fuel rods (see Section 4.1).

During the laser-drilling of irradiated particles, the particles are contained in a sealed compartment, which has a window for the beam to enter. The compartment is purged with helium, which flows into an ionization chamber. A burst of radioactive gas (mainly Kr-85) shows that the laser beam has penetrated the coating of the irradiated particles.

Metallographic examination of sister fuel particles after laser-failing can be used to determine the depth of penetration. The depth of penetration is found to be proportional to the laser power output.

3.2. IN-PILE MEASUREMENT OF FISSION GAS RELEASE

Fission gas release (R/B) measurements during operation of reactor, loop, and capsule systems provide data for evaluating fuel performance. For example, in the Peach Bottom HTGR, routine R/B measurements were made to evaluate fuel performance by measuring the activity of selected noble gas isotopes in the fuel element purge stream, which contained essentially 100% of all the activity released by the fuel (Refs. 3-9, 3-10).

The release rate, R , is derived from measurements of the specific activity $[Bq/m^3 \text{ (ntp)}]^*$ of an isotope in the helium coolant gas by means of gamma ray spectrometry. The following relationship is used to evaluate R for reactor, loop, and capsule systems which have a purge system, including the Peach Bottom HTGR:

$$R_i = \frac{A_i F}{\lambda_i} \text{ (atoms/s)} \quad , \quad (3-1)$$

where A_i = specific activity of the isotope i in the purge stream gas sample $[Bq/m^3 \text{ (ntp)}]$,

F = fuel element purge flow rate $[m^3 \text{ (ntp)}/s]$,

λ_i = decay constant for isotope i (s^{-1}).

The following relationship is used to calculate the release rate, R_i , for reactor, loop, and capsule systems where the helium coolant circulates and is purified in a side stream:

$$R_i = \frac{A_i V_o (\lambda_i + r)}{\lambda_i} \text{ (atoms/s)} \quad , \quad (3-2)$$

*Disintegrations per second per cubic meter of helium at 0.1 MPa (1 atm) and 298K.

where V_o = volume of the primary coolant,

r = purification rate of cleanup system (fraction/s).

This relationship is applicable to large HTGR systems.

The value of B is calculated from a knowledge of the steady-state thermal power of the system. Thus,

$$B_i = P k_2 Y_i \text{ (atoms/s)} \quad , \quad (3-3)$$

where P = reactor power (MWt),

k_2 = conversion constant (3.2×10^{16} fissions/MW·s),

Y_i = chain fission yield of isotope i .

It follows that for purged systems

$$(R/B)_i = \frac{A_i F}{P k_2 Y_i \lambda_i} \quad , \quad (3-4)$$

and for circulating systems

$$(R/B)_i = \frac{A_i V_o (\lambda_i + r)}{P k_2 Y_i \lambda_i} \quad . \quad (3-5)$$

3.3. DETERMINATION OF R/B FOR IODINE NUCLIDES IN HTGR SYSTEMS

Iodine release measurements cannot be made in the same straightforward manner as the noble gas measurements because the iodine nuclides sorb in graphite and plate out on reactor components and sample lines. Two methods are available for iodine analysis: one method involves measurement of the decay products of condensed iodine, and the other method involves the use of a diffusion tube or other device to sample the helium coolant.

The decay method for iodine analysis involves determining the xenon daughter activity following a shutdown of the HTGR system. Once the fuel has cooled to ambient temperature and fission processes have ceased, the release of krypton and xenon emanating from the fuel will drop to a negligible value. The xenon isotopes present in the gas stream after cooldown are there as a result of decay of iodine isotopes condensed on surfaces outside the fuel. This has been shown, for example, by taking purge stream grab samples at the Peach Bottom HTGR at various times after cooldown and noting that the specific activity of a xenon isotope decreases with the half life of the iodine precursor (Ref. 3-9).

Only two isotopes of iodine (I-135 and I-133) can be determined by this method, since only these two yield detectable radioactive xenon daughters:

The disintegration rate of the iodine isotopes thus becomes the production rate (atoms/s) of the xenon daughters, which are immediately released into the coolant or purge stream. Relationships similar to those given in Section 3.2 are applicable.

The decay method tends to overestimate the fractional release of iodine nuclides into the helium stream because the method measures the decay products from the iodine sorbed in the fuel element graphite as well as that plated out in the circuit. (The sorption of iodine by graphite is discussed in Section 4.7.)

In the diffusion tube method (Ref. 3-11), a portion of the coolant stream passes through a small-diameter tube. From the total amount of each fission product deposited in the tube, partial pressures in the coolant

and R/B values can be calculated. Application of the diffusion tube method in the GAIL IV test is described in Ref. 3-12.

R/B values determined by the diffusion tube method tend to be too low because of iodine condensation in the primary circuit between the fuel element (or core) and the diffusion tube. However, the diffusion tube method is believed to be more accurate than the decay method for measuring the release of iodine nuclides into the helium coolant.

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4. FISSION GAS BEHAVIOR IN HTGR FUEL MATERIAL

4.1. EFFECT OF FUEL CONFIGURATION ON FISSION GAS RELEASE

In this section, which is an extension of an earlier presentation by Burnette et al. (Ref. 4-1), fission gas release characteristics are described for the following fuel configurations: (1) intact coated fuel particles, (2) fuel contamination, (3) particles with failed coatings, and (4) bare kernels. Included is a description of release mechanisms and a discussion of delay times of fission gas nuclides in graphite.

Information on the fission gas release characteristics of fuel contamination and particles with failed coatings is of particular importance because, as pointed out in Section 2, fission gas release from fuel rods is governed by the amount of uranium and thorium contamination in as-manufactured fuel rods and/or by the number of particles which experience coating failure during irradiation.

The reference fission gas nuclide has been taken to be 4.4-h Kr-85m; most of the data presented on the effect of fuel configuration are for this nuclide. The other fission gas nuclides with half-lives not much greater than 5 days are known to behave similarly. The relative release behavior of all the noble gas nuclides is discussed in Sections 4.2 and 4.3. The relative release behavior of xenon and iodine nuclides is discussed in Section 4.7.

4.1.1. Fission Gas Release from Intact Coated Fuel Particles

Many fission gas release experiments have been performed on intact coated fuel particles. The results show that silicon carbide and high-density pyrolytic carbon are very effective barriers for fission gas

nuclides at reactor temperatures and that release from intact coated particles under reactor conditions is primarily from fissioning of heavy metal contamination in the coatings or on the surface of coatings, not from the kernel by diffusion through the coatings. The fission gas release fraction (Kr-85m R/B at 1100°C) for as-manufactured coated fuel particles is typically around 10^{-5} or lower.

Reagan et al. (Ref. 4-2) studied fission gas release from coated fuel particles at temperatures between 1093° and 1370°C and burnups to about 20% heavy metal. Pyrolytic carbon coatings on thorium-uranium carbide kernels and on uranium carbide kernels were tested. Eight of the nine samples tested showed good ability to retain fission gases. Kr-85m R/B values at end-of-life for these samples were in the range 4×10^{-5} to 2×10^{-6} . One sample exhibited relatively high release (R/B value of 4×10^{-3}) because of coating failure. The level of uranium contamination in the particle coatings was sufficient to account for all the fission gas release observed from intact particles.

Samples of unbonded, Fort St. Vrain production-type particles were irradiated in capsule F-30. Fission gas release (R/B) data for the samples, given in Table 8 of Ref. 4-3, were obtained from measurements in the TRIGA King furnace facility. (See Section 3.) Beginning-of-life Kr-85m R/B values at 1100°C were around 10^{-5} for most of the particle samples, and most end-of-life values were in the range of 10^{-5} to 10^{-4} , indicating satisfactory performance.

Baumann and Reagan (Ref. 4-4) used mathematical models for interpretation of fission gas release from pyrolytic-carbon-coated particles. They concluded that (1) below 1600°C release of krypton, and probably iodine and xenon, was due to coating contamination, (2) above 1600°C release from the kernel followed by diffusion of krypton through the coating became significant, and (3) an overall increase in release rate with time occurred due to migration of U-235 through the coating.

The relationship between fission gas release through intact pyrolytic carbon coatings and coating density has been studied (Refs. 4-5, 4-6). Results indicate that (1) for coating densities greater than approximately 1.6 mg/mm^3 (bulk density), the fission gas release decreases very sharply with increasing density; (2) for coatings having densities greater than 1.90 mg/mm^3 , release through the coating becomes immeasurably low; and (3) BISO particles with outer isotropic coating densities of $\geq 1.80 \text{ mg/mm}^3$ meet the design criterion of 3×10^{-5} R/B for Kr-85m at 1100°C .

When coated fuel particles are mixed with matrix material, the fission gas release (R/B) increases if coating contamination is the predominant source of release. The release is higher for the mixture because recoil atoms, resulting from fissions of heavy metal contamination in or on the particle coating, may stop in the highly releasing, porous matrix material rather than in the more retentive, dense pyrolytic carbon coating material of an adjacent particle. (See Sections 4.1.4 and 4.1.5 for discussion of similar behavior for mixtures of bare kernels and matrix material.)

4.1.2. Fission Gas Release from Contamination

The source of fission gas release from as-manufactured fuel rods is considered to be heavy metal (uranium and thorium) contamination, i.e., exposed heavy metal not protected by intact coatings. Contamination, as defined in this report, includes (1) heavy metal distributed in the fuel rod matrix, (2) heavy metal in particle coatings within recoil range of the coating surface, and (3) coated particles with missing, broken, or permeable coatings in as-manufactured fuel rods. Contamination in the fuel rod matrix is probably the predominant source of fission gas release.

Burnette et al. (Ref. 4-1) simulated fuel contamination by adding UO_2 powder to loose carbon matrix (Fort St. Vrain fuel rod matrix material) or by diffusing uranium into carbon matrix material at high temperatures. Measured Kr-85m R/B values (at 1100°C) were 0.50 for the former configuration (UO_2 powder) and 0.30 for the latter (diffused uranium in carbon matrix).

The relationship between Kr-85m release and uranium contamination in fuel rod matrix material (Fort St. Vrain type) has been studied (Refs. 4-7, 4-8). Composites of loose fuel particles mixed with matrix material were annealed at 1800° and 2000°C. A value of the fractional release for Kr-85m at 1200°C was then measured for each composite utilizing the TRIGA facility (see Section 3). The irradiation time in each case was 1 h. Subsequently, the particles and matrix material in each composite were separated and counted. From the count data, fractions of uranium and thorium in the matrix material were determined.

Values of the fractional release are plotted in Fig. 4-1 (which was taken from Ref. 4-8). The data show an approximately linear relationship; however, they are not consistent with an expected slope of 45°. The data are believed to be relatively high at the lower concentrations owing to some contribution from uranium on or near the surface of the fuel particles.

An R/B value for contamination in matrix can be derived from the curve drawn through the data. First, the data are multiplied by a factor to obtain steady-state R/B values; then the data are divided by a factor to adjust them to the reference temperature of 1100°C. Both factors are around 1.5 and just about offset each other; therefore, the curve as drawn can be taken to represent the relation between Kr-85m R/B at 1100°C and the fraction of uranium contamination. The curve indicates that the reference Kr-85m R/B value of 3×10^{-5} for as-manufactured fuel (see Section 2) corresponds to a value of about 1.1×10^{-4} for the fraction of uranium contamination. This leads to a Kr-85m R/B value of 0.27 at 1100°C for uranium contamination in fuel rod matrix material. This value is consistent with the value of 0.30 measured by Burnette et al. (see above).

In a similar study performed earlier, calcined petroleum coke was used as the matrix material (Ref. 4-7). However, the reported R/B data are questionable. The R/B values found appear to be numerically equivalent to the fractional contamination values, suggesting, incorrectly, 100% release of Kr-85m from the matrix material.

Fig. 4-1. Kr-85m fractional release values as a function of uranium contamination in fuel rod matrix material

On the basis of the above information, a Kr-85m R/B value of 0.30 at 1100°C is recommended for uranium contamination in fuel rod matrix material. One would expect the value to be the same for thorium contamination in fuel rod matrix material.

Studies have been performed to relate fission gas release and contamination associated with the outer coating of intact coated particles. Coating contamination can be in the form of (1) heavy metal in the outer pyrolytic carbon coatings, within recoil range of the surface, and (2) heavy metal on the coating surface [defined as the fraction of heavy metal that can be leached (Ref. 4-9)].

Results indicate that contamination on the coating surface is the predominant source of fission gas release from intact coated fuel particles (Ref. 4-2). The level of contamination on the coating surface is generally higher than the level within the coating (Ref. 4-9). For an isolated coated particle, the fraction of recoils that can escape from the interior of the outer pyrolytic carbon coating is only about 0.10, based on a recoil range of 10 μm in pyrolytic carbon and a coating thickness of 50 μm and assuming uniform distribution of contamination in the coating. Recoils occurring on the coating surface have a fractional release probability of about 0.5.

ORNL investigators studied the relationship between fission gas release and coating surface contamination (Ref. 4-10). They determined the fuel contamination at or near the surface of pyrolytic carbon coatings by alpha counts of each batch of particles before irradiation and found a correlation between contamination level and fission gas release, shown in Fig. 2-12 in Ref. 4-10. The investigators concluded that fission gas release from intact pyrolytic carbon particles is from contamination in the coating (at least up to 1400°C) and does not come from the fuel kernel.

In studies at GA (Refs. 4-8, 4-9, 4-11, 4-12), levels of uranium and thorium surface contamination and the levels of these elements in the pyrolytic carbon coating outside of the intact SiC coatings were determined

on samples of TRISO-coated particles. Calculated release values, based on uranium contamination levels, were compared with observed values obtained in TRIGA tests (Refs. 4-9, 4-12). By assuming that loose fissile particles released 20% of the gases and that loose fertile particles released 10% of the gases produced by contamination, good agreement was obtained between the calculated and observed Kr-88 release values. These assumptions are reasonable for a bed of loose particles in view of the known reduction in release due to recoils stopping in the dense coating material of adjacent particles (see Ref. 4-9 and Section 4.1.1).

4.1.3. Fission Gas Release from Particles with Failed Coatings

As described in Section 2, the fission gas release characteristics of failed particles must be known in order to predict the fission gas release behavior of fuel rods during irradiation. Considerable fission gas release data are available for failed particles; and for the purpose of discussion, the data are divided into three categories: (1) failed particles in reactor, loop, and capsule tests, (2) loose failed particles, and (3) laser-failed particles.

4.1.3.1. Fission Gas Release from Failed Particles in Reactor, Loop, and Capsule Tests. Fission gas release data for failed particles derived from reactor, loop, and capsule experiments are given in Table 4-1. The data represent measurements on fuel particles having various types of coatings and kernels, including oxides and carbides. All the particles had dense kernels. The average $(R/B)_f$ value for the data is 0.0044 for Kr-85m at 1100°C.

Harmon and Scott (Ref. 4-5) reported fission gas release data for failed fertile particles. These data, given in Table 4-2, were derived from TRIGA tests on fuel rods irradiated in capsules P13P and P13N. The fertile particles in these fuel rods had relatively high coating failure fractions as determined by metallography. The fissile particles in the rods did

TABLE 4-1
(R/B)_f VALUES DERIVED FROM LOOP, REACTOR, AND CAPSULE TESTS

Experiment	Kernel Type	Coating Type	Failed Fraction	Burnup (% FIMA)	(R/B) ^(a)	Ref.
Pluto 3A loop	UC ₂	PyC ^(b)	0.2	8.7	0.005	4-1,-13,-14
GA capsule 309 series	(Th,U)C ₂	Thin PyC	~1.0	~3-8	0.005	4-1,-13,-15
GA 6F2-1 capsule	(Th,U)C ₂	Multilayer, PyC-SiC	~0.25	7.2	0.004	4-1,-13,-16
GA 6F2-2 capsule	(Th,U)C ₂	Single layer, PyC	1.0	7.6	0.003	4-1,-13,-16
GA 6F2-3 capsule	(Th,U)C ₂	Single layer, PyC	1.0	5.7	0.004	4-1,-13,-16
GAIL III A loop	(Th,U)C ₂	Single layer, PyC	~1.0	1.0	0.002	4-1,-13,-17
GAIL III B loop	(Th,U)C ₂	Single layer, PyC	~1.0	4.8	0.005	4-1,-13,-18
GAIL IV loop	(Th,U)C ₂	Multilayer, PyC	0.008	2.5-4.8	0.006	4-1,-13,-19
Peach Bottom Core 1	(Th,U)C ₂	Single layer, PyC	0.8	~3.5	0.008	4-1,-20
Fuel rod, P13N capsule	ThO ₂ ^(c)	BISO ^(c)	0.2	~3	0.002	4-1,-21
				Average	0.0044	

(a) Kr-85m R/B value for failed particles at 1100°C.

(b) PyC stands for pyrolytic carbon.

(c) Failed particles were predominantly fertile BISO-coated particles.

TABLE 4-2
FISSION GAS RELEASE (R/B) DATA FOR FAILED FERTILE PARTICLES IN FUEL
RODS IRRADIATED IN CAPSULE P13P AND P13N^(a)

Particle Type	Capsule and Position	Fast Fluence ($\times 10^{25}$ n/m ²) (E>29 fJ)	Burnup ^(b) (% FIMA)	Sample Size ^(c)	Coating Failure ^(d) (%)	R/B ^(e) Kr-85m	(R/B) _f
ThO ₂ BISO	P13P-5A	3.9	0.8	3330	0.7	6.6×10^{-5}	0.009
ThO ₂ BISO	P13P-4D	5.3	0.9	3470	0.7	1.6×10^{-5}	0.002
ThO ₂ BISO	P13P-4B	5.9	1.0	4290	22.8	2.2×10^{-4}	0.001
ThO ₂ BISO	P13P-2D	8.1	1.9	2720	97.0	2.0×10^{-3}	0.002
ThO ₂ BISO	P13P-2B	7.9	1.9	3330	96.2	1.6×10^{-3}	0.002
ThO ₂ BISO	P13P-2A	7.7	1.8	3550	83.3	1.3×10^{-3}	0.002
ThC ₂ BISO	P13P-3D	7.5	1.6	7550	6.5	3.2×10^{-4}	0.005
ThC ₂ BISO	P13P-3C	7.6	1.6	4120	1.9	1.5×10^{-4}	0.008
ThC ₂ BISO	P13P-1B	5.9	1.2	--	0.4	3.0×10^{-5}	0.008
ThC ₂ TRISO	P13P-1A	5.7	1.2	--	3.6	5.1×10^{-4}	0.014
ThC ₂ TRISO	P13N-3D	4.7	1.8	--	15.7	1.3×10^{-3}	0.008
						Average	0.0055

(a) Data from Table 16 of Ref. 4-5.

(b) Heavy metal burnups calculated with FISSIN computer program.

(c) Number of fuel particles in fission gas release sample.

(d) Failure fractions determined by visual and metallographic examination.

(e) Postirradiation TRIGA R/B for Kr-85m at 1100°C. The data were not corrected for steady state. The fuel rod R/B data were adjusted to reflect only the fissions occurring in the fertile particles. These R/B values differ from those in Table 9-1 of Ref. 4-22 because of revised end-of-life uranium loadings.

not fail to a measurable degree; thus, the fission gas release observed was attributed to the fertile particles. The average $(R/B)_f$ value given in Table 4-2 for the fertile particles is 0.0055. This value is not significantly different from the average value for the data in Table 4-1.

4.1.3.2. Fission Gas Release from Loose Failed Fuel Particles. Fission gas release measurements have been performed at GA on beds of loose fuel particles with failed coatings. Cracks were introduced into particle coatings by gently squeezing particles between the jaws of a micrometer. The average R/B for Kr-85m at 1100°C was approximately 0.02 for the cracked samples (Ref. 4-1).

In another study, fission gas release measurements were performed on samples of loose fuel particles that had been irradiated to mostly low burn-ups in the P13N capsule. These particle samples were first visually examined using a stereomicroscope to determine the number of failed particles in the sample. After examination, the samples were tested in the TRIGA King furnace facility (see Section 3). The R/B values thus obtained were divided by the fraction failed to obtain $(R/B)_f$ values. The data, consisting of nine R/B values, are given in Ref. 4-1. The average R/B value is 0.023, which is essentially identical to that obtained for the unirradiated mechanically cracked particles.

4.1.3.3. Concept of Constrained Failed Particles. The data presented above show that R/B values for loose failed fuel particles are appreciably higher than values for failed particles in fuel rods and failed particles in loop and reactor experiments. This led Burnette et al. (Ref. 4-1) to the concept of the constrained failed particle, defined as a failed particle with a cracked coating in a fuel rod where the crack is constrained from opening by the matrix material and adjacent particles.

It seems reasonable that constrained failed particles should exhibit lower R/B values than loose failed particles. A closed crack will offer more resistance to release than an open crack. Also, the exposed kernel surface area will be smaller for the constrained failed particles.

The failed particles represented by the data in Tables 4-1 and 4-2 are considered to be constrained.

4.1.3.4. Fission Gas Release from Laser-Failed Particles. The technique of laser-drilling, described in Section 3.1.3, may be used to simulate failed constrained particles. The laser beam intensity is adjusted to drill pinholes, approximately 10 μm in diameter, which extend from the particle surface to the kernel surface. The pinholes cannot open up, and thus laser-failed particles simulate failed particles constrained in fuel rods.

Fission gas release (R/B) data derived from tests on laser-failed particles (Refs. 4-1, 4-23) are given in Tables 4-3 and 4-4. These data supplement and are in accord with the data given in Tables 4-1 and 4-2 and, of particular importance, provide R/B values for failed particles irradiated up to 63% FIMA burnup (see Section 4.6.2).

4.1.3.5. Discussion of Fission Gas Release from Failed Particles. Comparison of the R/B data given above for (1) failed particles in reactor, loop, and capsule tests, (2) loose failed particles, and (3) laser-failed particles indicates that the constrained failed particle is representative of failed particles in fuel rods. The average R/B for all the constrained failed particle data (Tables 4-1 through 4-4), assuming equal weight for each value, is 0.0053 for Kr-85m at 1100°C. The associated standard deviation is 0.0027. On this basis, the reference R/B value for the release of Kr-85m at 1100°C from constrained failed particles (both fissile and fertile) is taken to be 0.005 ± 0.003 .

The $(R/B)_f$ value may be greater than 0.005 if the constrained failed particle configuration is not representative of failed particles in fuel rods. Under temperature transients during HTGR operation, more severe types of failure, such as pressure vessel failure, where the coating breaks into many pieces are possible. The extent of such types of failure has to be established; and if the extent is significant, corresponding fractional release (R/B) values must be determined.

TABLE 4-3
R/B VALUES DERIVED FROM TESTS ON LASER-FAILED PARTICLES
WITH DENSE UC₂ KERNELS (a)

Coating Type	Burnup (% FIMA)	Irradiation Test	(R/B) _f (b)
TRISO (c)	0	--	0.0048
	63 (d)	P13P	0.0056
BISO	8	FTE-2 (e)	0.0066
BISO	21	FTE-1 (e)	0.0042
BISO	30	FTE-1 (e)	0.0057

(a) Data were taken from Ref. 4-23.

(b) Kr-85m value at 1100°C.

(c) Two particle samples from the same batch of reference-type fissile particles.

(d) This burnup value replaces the preliminary value of 58% FIMA given in Ref. 4-23.

(e) Fuel test element irradiated in the Peach Bottom HTGR.

TABLE 4-4
R/B VALUES DERIVED FROM TESTS ON LASER-FAILED PARTICLES^(a)

Kernel Type	Coating Type	Burnup (% FIMA)	Irradiation Test	(R/B) _f ^(b)
UO ₂	BISO	0	FTE-1 ^(c) and -2	0.0025
		10		0.011
		15		0.011
		30		0.0055
UC ₂	BISO	25	FTE-1 ^(c) and -2	0.0026
		30		0.0038
		54		0.0072

(a) Data were taken from Ref. 4-1.

(b) Kr-85m value at 1100°C.

(c) Fuel test elements irradiated in the Peach Bottom HTGR.

The product of $(R/B)_f$ and f (see Eq. 2-1) must be consistent with observed in-pile fission gas release behavior for reference-type fuel rods. The predicted failure fraction, as calculated by particle failure models (Ref. 4-24) and core averaging procedures (Ref. 4-25), defines the fraction of particles with exposed kernels. Defining the failure fraction and f in this way is important for at least one reason. The fraction of fissions occurring in particles with exposed kernels must be known in order to predict the effect on fission gas release of the chemical interaction of fuel kernels with coolant impurities (see Section 4.8).

The apparent practice in the Dragon program is to use an equivalent uncoated particle fraction, termed ϕ . This coefficient is defined (Ref. 4-26) as the fraction of uncoated kernels which would give the same steady-state gas release rate as the particle assembly in question. In other words, the adopted reference failed-fuel configuration is a bare kernel; $(R/B)_f$ is based on fission gas release from bare kernels; and ϕ is made consistent with this $(R/B)_f$ so that calculated release is consistent with observed release. This practice, although somewhat empirical, is satisfactory as long as calculated and observed releases are consistent; however, it has the disadvantage that ϕ does not accurately define the fraction of particles with exposed kernels.

4.1.4. Fission Gas Release from Bare Fuel Kernels

Bare fuel kernels are fairly retentive of fission gases. Burnette et al. (Ref. 4-1) found Kr-85m R/B values at 1100°C for bare kernels to be in the range of 0.002 to 0.04, depending on the material environment surrounding each kernel. (See Table 4-5.) The addition of loose carbon (graphite type) matrix to a packed bed of oxide fuel kernels increases R/B by a factor of about five. (Similar behavior was observed for coated particles; see Section 4.1.1.) Further R/B increases are attained when the carbon matrix is replaced with fuel rod matrix. (See Section 4.1.5 for more detail on these phenomena.)

TABLE 4-5
EFFECT ON R/B FOR BARE KERNELS OF THE MATERIAL ENVIRONMENT AROUND THE KERNEL ^(a)

Failed Fuel Configuration	Density of Material Around Kernel	(R/B) _f ^(b)
1. Packed bed of bare UO ₂ kernels	Dense UO ₂	0.002
2. Packed bed of UO ₂ kernels with carbon matrix ^(c)	Combination of dense UO ₂ and low-density matrix ^(c)	0.01
3. Bare UO ₂ kernels in fuel rod	Mostly low-density matrix	0.03-0.04

^(a) Information was obtained from Ref. 4-1 for cases 1 and 2. Information for case 3 is presented in Section 4-3. In each case the kernel diameter was around 200 μm.

^(b) R/B for Kr-85m at 1100°C for failed fuel configurations.

^(c) Same as case 1, except that the void space between the kernels was filled with matrix powder. The kernels were in contact with each other.

Zumwalt et al. (Ref. 4-27) extensively studied fission gas release behavior in spherical monogranular UC_2 particles. They found that, for the most part, the release of fission gases followed the kinetics and temperature dependence of an activated diffusional process, although there was evidence for a fast release component.

Flowers (Ref. 4-28) describes the release of fission product nuclides from UO_2 kernels into the internal porosity of coated particles. From his description one can derive a Kr-85m R/B value of 0.041 at 1100°C for a kernel surrounded by a buffer coating. This value is in reasonable agreement with the R/B value for bare kernels in a fuel rod (see Table 4-5), suggesting that the environment for a kernel surrounded by a buffer coating layer is similar to that for a bare kernel in a fuel rod.

4.1.5. Release Mechanisms for Failed Particles

The data in Table 4-5, show that fission gas release from kernels increases with a decrease in the density of the material around the kernel. This is consistent with a mechanism of release from the kernel involving recoil. If diffusion of fission products from the kernel were the predominant mechanism of fission gas release, one would expect that the density of the material surrounding the kernel would have little effect on release. If diffusion from the kernel were dominant, one would expect, for example, that cases 1, 2, and 3 of Table 4-5 would show the same $(R/B)_f$ values.

As indicated above (Section 4.1.1) and in Ref. 4-29, fission gas release increases when coated fuel particles are fabricated into fuel rods. This is also consistent with a mechanism involving recoils.

The relatively low activation energies observed for R/B, around 70 kJ/mol (17 kcal/mol) for krypton (see Section 4.3), are consistent with

recoil-related release from the kernel. If diffusion from the kernel were predominant, one would expect an activation energy of around 146 kJ/mol (35 kcal/mol) for R/B (see Section 4.3.2). (Note that, on the basis of Eq. 2-4, the activation energy for R/B is one-half the activation energy for D, the diffusion coefficient.) In fact, the low activation energy, which applies to all fuel configurations at temperatures up to 1300°C at least, suggests that the predominant release mechanism involves recoil for all fuel configurations of interest.

The fraction of recoils is sufficient to account for the observed fractional releases, which range up to about 0.03 for failed particles and bare kernels. The recoil fraction for 200- μ m-diameter UO_2 kernels was experimentally determined to be 0.08 (Ref. 4-1).

The conclusion is reached that the predominant mode of release from fuel kernels is by a mechanism involving recoil at least for temperatures up to 1300°C and for Kr-85m and the shorter-lived isotopes. The recoil mechanism for fission gas migration in fuel material can involve direct recoil, surface ejection (or knockout), and fission-induced sorption (or knockon).

Carroll (Ref. 4-30) discusses models involving the concept of trapping gas at irradiation-induced defects within the fuel structure. Fission gas collecting at defects will form bubbles (or clusters), and these bubbles can move by diffusion. Models for bubble growth have been derived. During irradiation there is competition between bubble growth and bubble shrinkage.

4.1.6. Retention of Fission Gases by the Graphite Web

The fission product gases which are released from the fuel rods are transported through the graphite web by two mechanisms: (1) transverse flow of the helium coolant and (2) diffusion. The time delay in this transport governs the decay of fission gas nuclides during passage through the graphite web.

Transverse flow is the process in which helium coolant flows through the graphite web surrounding the upper part of the fuel rod stack, into and along the gap between the fuel rods and graphite, and back through the graphite web surrounding the lower part of the fuel rod stack to the main helium coolant stream. The fission gases in the gap and the graphite are entrained by the helium undergoing transverse flow and are swept into the main helium coolant stream. In unpublished work at GA, Mysels and Rovner have estimated the possible effects of transverse flow on the transport of fission products through the graphite web. They concluded that in the absence of adsorption, the gaseous fission products entrained by the helium in the gap will be delivered to the circulating mainstream in times of the order of 1 s.

The time delay resulting from only diffusion of the fission gases through the graphite web can be simply estimated from the relationship

$$t_d = a^2/2D \quad , \quad (4-1)$$

where t_d is the delay time, a is the thickness of the graphite web, and D is the gas phase diffusion coefficient in the graphite web. The diffusion coefficients of krypton and xenon in the graphite web have been calculated to be 0.044 and 0.037 mm²/s, respectively, at the representative reactor conditions of 800°C graphite temperature and 5.2 MPa helium pressure. These diffusion coefficient values are based on (1) diffusion coefficients for krypton and xenon in helium of 64.4 and 54.6 mm²/s, respectively, at 298 K and 0.1 MPa (see Ref. 4-31) and (2) a tortuosity factor of 0.005 derived from graphite-helium permeability measurements performed at GA. The diffusion coefficients were assumed to vary as $[T(K)]^{3/2}/P(\text{MPa})$. For a typical graphite web thickness of 4.7 mm, the calculated decay times were between 250 and 300 s.

Thus, the conclusion is reached that under normal operating conditions, transport of fission product gases through the graphite in the absence of adsorption will be controlled by transverse helium flow and that depletion

of isotopes of krypton and xenon by radioactive decay will be negligible except for the very short-lived nuclides. This conclusion is in agreement with that of Faircloth (Ref. 4-32).

Retention by the graphite web of fission product gases such as the halogens and related elements is discussed in Section 4.7.

4.1.7. Summation on Fuel Configuration Effects on Fission Gas Release

The information on the effect of fuel configuration on fission gas release may be summarized as follows:

1. Fission gas release from HTGR fuel elements is governed by the amount of uranium and thorium contamination (including particles with missing, permeable, or broken coatings) in as-manufactured fuel rods and by the number of failed particles resulting from coating failure during irradiation.
2. Values of the fraction of fissions occurring in failed fuel particles, f , and the fractional fission gas release for failed particles, $(R/B)_f$, are required in order to calculate, using Eq. 2-1, the core average R/B during irradiation.
3. Failed fuel particles in a constrained configuration (where the coating is constrained from opening) appear to be representative of failed fuel particles in fuel rods. Evidence for this is that loose (unconstrained) failed particles exhibit higher R/B values than failed particles in fuel rods and that laser-failed particles and failed particles in fuel rods show similar R/B values.
4. The value of $(R/B)_f$ for Kr-85m at 1100°C is 0.005 ± 0.003 for constrained failed fuel particles (both fissile and fertile) in fuel rods. This value is based on observed R/B data obtained from TRIGA tests on laser-failed particles and data derived from loop, reactor, and capsule experiments.

5. The graphite web has a negligible effect on the release of noble gas fission products.
6. Verification that the constrained failed particle represents failed particles in fuel rods is needed.
7. The Kr-85m R/B value at 1100°C for uranium and thorium contamination in fuel rod matrix material is 0.30.
8. The predominant mode of release from fuel kernels is by a mechanism involving recoils, at least for temperatures up to 1300°C and for Kr-85m and the shorter-lived isotopes.

4.2. DEPENDENCE OF FISSION GAS RELEASE ON NUCLIDE HALF-LIFE

The dependence of the fractional release, R/B, on half-life is indicative of the delay that occurs between birth and detection of the fission product. This delay is related to the mechanism of fission gas release as well as to the structure and composition of the material through which the gases pass.

In this section, data are presented for (1) short-lived isotopes with half-lives between 8 s and 5.3 d and (2) long-lived isotopes with half-lives greater than 10 y and stable isotopes.

4.2.1. Half-Life Dependence for Short-Lived Isotopes

For the short-lived isotopes the half-life dependence is given by:

$$(R/B)_{ij} = A_j (\tau_i)^n, \quad (4-2)$$

where, for each temperature, A_j and n are constants, j represents the element, and τ_i is the half-life for isotope i . The relationship between Eq. 4-2 and Eq. 2-4 is developed below.

In the following sections half-life dependence data for the short-lived isotopes are derived from in-pile (reactor, loop, and capsule) measurements and from laboratory (TRIGA test) measurements. On the basis of in-pile data, the value of n is shown to vary from about 0.2 near 300°C to about 0.5 at 1100°C. Comparison of values of n derived from fuel rod samples in TRIGA tests with those from in-pile measurements is important in establishing a contribution to fission gas release from gas phase diffusion.

4.2.1.1. In-Pile Half-Life Dependence Data. There are a considerable number of in-pile data from which a half-life dependence of R/B may be obtained. These data, which are from capsule, loop, and reactor measurements, are given in Table 4-6. The data represent a variety of materials and configurations, a range of the extent of particle failure and coating contamination, and, of course, different experimental arrangements.

The temperatures given in Table 4-6 for the loop, capsule, and reactor measurements (excepting FSV data) were estimated by using data (Refs 4-36, 4-37) on temperature distributions. The estimated temperatures range from 850° to 1300°C. The uncertainty of the estimated temperatures could be more than 100°C.

The values of n derived from higher-temperature data of Table 4-6 are shown in Table 4-7. These values were obtained from least-squares fits of log-log plots of R/B versus half-life (τ), in accordance with Eq. 4-2. Each datum was given equal weight. Alternate calculations in which longer-lived isotopes were weighted more heavily, possible errors in Xe-133 R/B values were accounted for, and an effective half-life was used for Xe-135 did not yield n values significantly different from those in Table 4-7. The average values of n for both krypton and xenon are close to 0.5 with an uncertainty of ± 0.1 . This average value is indicative of release dominated by a diffusion process, which apparently involves bulk diffusion of recoiled fission gases.

The low-temperature in-pile data were obtained from the Fort St. Vrain reactor during testing at very low power levels between about 1% and 2%.

TABLE 4-6
R/B DATA FOR VARIOUS LOOP, CAPSULE, AND REACTOR SYSTEMS

Isotope	τ (s)	λ (s ⁻¹)	R/B				
			Capsule 6F2 _I (Ref. 4-16)	Capsule 6F2 _{II} (Ref. 4-16)	Capsule 6F2 _{III} (Ref. 4-16)	GAIL III-B Loop (Ref. 4-18)	GAIL IV Loop (Ref. 4-13)
	(a)						
Kr-85m	1.58(4)	4.38(-5)	1.7(-3)	7.0(-3)	1.1(-2)	4.9(-3)	3.3(-5)
Kr-87	4.57(3)	1.52(-4)	5.4(-4)	2.8(-3)	3.5(-3)	2.1(-3)	1.7(-5)
Kr-88	1.00(4)	6.90(-5)	7.7(-4)	3.4(-3)	4.5(-3)	2.3(-3)	2.2(-5)
Kr-89	1.91(2)	3.63(-3)	1.8(-4)	1.5(-3)	1.65(-3)	8.6(-4)	5.3(-6)
Kr-90	3.23(1)	2.15(-2)	1.3(-4)	1.4(-4)	4.0(-3)	2.2(-4)	
Kr-91	8.60(0)	8.05(-2)	—	—	—	—	
Xe-133	4.55(5)	1.52(-6)	4.8(-3)	3.0(-2)	5.0(-2)	9.1(-3)	2.2(-5)
Xe-135	3.30(4)	2.10(-5)	4.5(-4)	2.2(-3)	4.5(-3)	1.4(-3)	6.7(-6)
Xe-135m	9.43(2)	7.35(-4)	—	—	—	—	
Xe-137	2.29(2)	3.02(-3)	3.9(-5)	2.2(-4)	4.8(-4)	2.9(-4)	
Xe-138	8.53(2)	8.12(-4)	7.2(-5)	6.5(-4)	6.5(-4)	2.5(-4)	1.8(-6)
Xe-139	4.00(1)	1.73(-2)	—	—	—	—	
Xe-140	1.36(1)	5.09(-2)	5.8(-6)	7.9(-6)	1.6(-5)	1.9(-5)	3.1(-7)
Estimated temperature (°C)			1300	1300	1300	1134	990
Pressure (MPa)			0.17	0.17	0.17	2.31-2.41	2.38 ^(b)

TABLE 4-6 (Continued)

Isotope	R/B					
	Peach Bottom Core 1, 30% Power (Ref. 4-33)	Peach Bottom Core 1, Full Power (Ref. 4-33)	Peach Bottom Core 1, EOL (Ref. 4-33)	Pluto A Loop Charge 8 (Ref. 4-34)	Pluto IIIa Loop (Ref. 4-14)	Peach Bottom Core 2, (Ref. 4-1)
Kr-85m	2.7(-4)	3.2(-4)	3.3(-3)	1.0(-5)	7.6(-4)	2.1(-4)
Kr-87	1.1(-4)	1.1(-4)	1.5(-3)		5.4(-4)	7.0(-5)
Kr-88	1.2(-4)	1.4(-4)	1.8(-3)	6.6(-6)	6.2(-4)	1.1(-4)
Kr-89	3.6(-5)	2.6(-5)	2.5(-4)	9.6(-7)	1.9(-4)	9.0(-6)
Kr-90					5.4(-5)	
Kr-91					2.1(-5)	
Xe-133	7.5(-4)	1.2(-3)	5.0(-3)	1.5(-5)		4.9(-4)
Xe-135	1.1(-4)	1.9(-4)	9.1(-4)	3.3(-6)		1.4(-4)
Xe-135m						
Xe-137				5.1(-7)		
Xe-138	3.7(-5)	2.5(-5)	2.6(-4)			1.4(-5)
Xe-139						
Xe-140				5.0(-7)		
Estimated Temperature (°C)	<850	850	850	1129	~1100	--
Pressure (MPa)	2.41	2.41	2.41	~2.0 ^(c)	~2.0 ^(c)	2.41

TABLE 4-6 (Continued)

Isotope	R/B	
	Capsule F-30 Cell 5 EOL Ref. (4-3)	Fort St. Vrain, 1%-2% Power
Kr-85m	2.4(-5)	4.7(-6)
Kr-87	2.0(-5)	2.9(-6)
Kr-88	1.4(-5)	2.9(-6)
Kr-89	3.4(-6)	
Kr-90		
Kr-91		
Xe-133		
Xe-135	9.6(-6)	3.6(-6)
Xe-135m		
Xe-137		
Xe-138	2.2(-6)	2.4(-6)
Xe-139		
Xe-140		
Estimated temperature (°C)	1100	250-350
Pressure (MPa)	~0.1	2.84

(a) $1.58(4) \equiv 1.58 \cdot 10^4$.

(b) Ref. 4-19.

(c) Ref. 4-35.

TABLE 4-7
SUMMARY OF VALUES OF EXPONENT $n^{(a)}$

Source and Reference	n	
	Kr Isotopes	Xe Isotopes
Capsule 6F2 _I (Ref. 4-16)	0.375	0.590
Capsule 6F2 _{II} (Ref. 4-16)	0.513	0.688
Capsule 6F2 _{III} (Ref. 4-16)	0.439	0.681
GAIL III-B Loop (Ref. 4-18)	0.424	0.520
GAIL IV Loop (Ref. 4-13)	0.391	0.388
Peach Bottom, Core 1, 30% power (Ref. 4-33)	0.394	0.468
Peach Bottom, Core 1, full power (Ref. 4-33)	0.508	0.613
Peach Bottom, Core 1, EOL (Ref. 4-33)	0.552	0.463
Pluto A Loop, Charge 8 (Ref. 4-34)	0.513	0.334
Pluto IIIa Loop (Ref. 4-14)	0.456	--
Peach Bottom, Core 2 (Ref. 4-1)	0.679	0.570
Capsule F-30, Cell 5 (Ref. 4-3)	0.421	0.406
Average	0.472	0.520
Standard deviation	0.087	0.119

^(a) For temperatures ranging from 850° to 1300°C.

The temperature was between 250° and 350°C and the value of n was found to be about 0.2 ± 0.1 for both krypton and xenon. Such a low value of n is indicative of rapid release from the fuel bodies.

4.2.1.2. TRIGA Test Data on Half-Life Dependence. The values of n for a fuel rod sample (CR-12, see Section 4.3) derived from temperature dependence data measured in TRIGA tests are given in Table 4-8. A comparison of these values with the n values derived from the in-pile data presented in Section 4.2.1.1 is shown in Fig. 4-2 for the case of krypton. The comparison demonstrates that the n values derived from the TRIGA test data are consistently smaller than those derived from the in-pile data. The prominent difference between the measurements from which the data were derived is the gas pressure; for TRIGA tests, the pressure was only 0.24 MPa, whereas for the in-pile measurements, the pressure was between 2.0 and 2.84 MPa except as noted in Fig. 4-2. On this basis, a contribution to the measured fractional release of fission gas from gas phase diffusion is postulated. The data of Fig. 4-2 have a large associated uncertainty, and there are in-pile data at low pressure but high temperature which have n values comparable to those of the high-pressure data. Nevertheless, a gas phase contribution is consistent with the data of Fig. 4-2. The gas phase contribution would be expected to be larger at low temperatures, since the temperature dependence of bulk diffusion is much greater than that of gas phase diffusion (see Section 4.3).

A comparison of the n values from in-pile and TRIGA test data on fuel rod CR-12 for xenon leads to the same conclusions as presented above for the krypton data. Furthermore, the same conclusions are reached using TRIGA test data (see Section 4.3) for other fuel rods and for loose compacts.

4.2.1.3. Gas Phase Diffusion. To conclusively establish the contribution of gas phase diffusion, fission gas release measurements were performed in the TRIGA test facility at a series of pressures and reduced masses. The

TABLE 4-8
VALUES OF THE EXPONENT n FOR FUEL ROD (CR-12)
DATA DERIVED FROM TRIGA TESTS

Temperature (°C)	n_{Kr}	n_{Xe}
40.9	0.128	0.162
98	0.083	0.088
215	0.074	0.088
280	0.090	0.135
416	0.107	0.150
525	0.045	0.135
650	0.025	0.119
713	0.108	0.122
814	0.080	0.153
1015	0.245	0.259
1080	0.253	0.319
1140	0.254	--
1278	0.258	0.283

Fig. 4-2. Comparison of n values of Eq. 4-2 derived from in-pile and TRIGA test data for krypton

gas phase diffusion coefficient is inversely proportional to the pressure and to the square root of the reduced mass (Ref. 4-38). Consequently, the fractional release should be a function of the product of pressure and square root of reduced mass if gas phase diffusion contributes significantly to the fractional release of fission gases; furthermore, the fractional release should decrease as the product of pressure and square root of reduced mass increases.

The results of the gas phase diffusion tests are shown in Fig. 4-3 for the case of krypton; these results conclusively establish a significant contribution to the fractional release by gas phase diffusion under the conditions of the tests. The curves in Fig. 4-3 are calculated with a function developed in Section 4.3 to account quantitatively for the contribution of gas phase diffusion to the total fractional release; these curves indicate the expected dependence of R/B on $P\sqrt{m^*}$, the product of pressure and square root of reduced mass. Curves for other isotopes of krypton (Kr-85m, Kr-88) and for isotopes of xenon (Xe-138, Xe-139) show similar trends, although the extent of change in R/B is a function of the half-life of the isotope.

The gas phase diffusion tests were conducted in the temperature range of 20° to 33°C, since the detection of the effects of gas phase diffusion was expected to be more likely at low temperature. The tests were performed on a mixture of bare UC_2 kernels and matrix material. That loose compacts and fuel rods have similar fission gas release behavior is indicated by the similarity in n values, as shown in Table 4-9.

The UC_2 kernels used in the gas phase diffusion tests were 200 μm in diameter and had been doped with 1.5% thorium for stabilization purposes. The uranium was fully enriched. The matrix material had been fully processed as a fuel rod (but without fuel particles) and was ground before use in the TRIGA tests. The range of the values of the product $P\sqrt{m^*}$ accessible in the TRIGA test facility was limited by comparison with the range of interest, corresponding to a factor of 50 change. Thus, the use of bare kernels was appropriate to ensure large fractional releases and provide for

Fig. 4-3. Fractional release (R/B) of krypton isotopes as a function of pressure and reduced mass

TABLE 4-9
MEAN VALUES OF THE EXPONENT n FOR FUEL ROD AND COMPACT DATA
DERIVED FROM TRIGA TESTS
(See Tables 4-11 Through 4-15)

Configuration	Sample	Temp. Range (°C)	$\bar{n}_{Kr}$ (a)	$\bar{n}_{Xe}$
Fuel rod	CR-12	40 - 814	0.083	0.14
Fuel rod	XXK40	40 - 814	--	--
Fuel rod	CR-11	39 - 823	0.14	0.12
Fuel rod	2B-20	40 - 805	0.14	--
Loose compact	Bare UO ₂ kernels plus matrix	41 - 810	0.11	0.12
Loose compact	Bare (Th,U)C ₂ kernels plus matrix	25	0.19	--
Fuel rod	Cr-12	1015 - 1278	0.25	0.29
Fuel rod	XXK40	1018 - 1502	0.21	--
Fuel rod	CR-11	958 - 1520	0.17	0.32
Fuel rod	2B-20	1087 - 1412	0.29	--
Loose compact	Bare UO ₂ kernels plus matrix	1060 - 1265	0.17	0.32
Loose compact	Bare (Th,U)C ₂ kernels plus matrix	1200 - 1400	0.32	--
		1600 - 2000	~0.14	--

(a) $\bar{n}_j$ = average value of n for element j in the indicated temperature range.

the largest possible absolute change in R/B over the accessible $P\sqrt{m^*}$ range. In the TRIGA test facility, the pressure range was between 0.18 and 0.30 MPa; the range in $\sqrt{m^*}$ was between 2.0 and 4.2, which was accomplished by changing the sweep gas from helium to neon. The use of other rare gases was prevented either by licensing restrictions on the operation of the facility or by restrictions based on the procedures of measurement. Thus, the range of values of $P\sqrt{m^*}$ was between 0.35 and 1.27 for experiments on the release of either krypton or xenon isotopes.

4.2.1.4. Discussion of Half-Life Dependence for Short-Lived Isotopes.

The in-pile and TRIGA test data indicate that the dependence of the fractional release on nuclide half-life is a function of temperature; this implies that the temperature dependence of the fractional release is a function of the isotope identity. The quantitative relationship between the temperature dependence of the fractional release and of the exponent n of Eq. 4-2 is developed in Section 4.4.

Examination of the half-life dependence of R/B has led to the postulation and confirmation in TRIGA tests of a gas phase contribution to the fractional release of fission gases. The gas phase contribution dominates the release at lower temperatures and bulk diffusion dominates at higher temperatures; thus, a change in the mechanism occurs with increasing temperature. This is consistent with the change in the half-life dependence of R/B with temperature as illustrated in Fig. 4-2. The quantitative relationships developed in Section 4.4 account for this change in mechanism.

At all temperatures and for the nuclides considered, Eq. 4-2 has been used. Under certain conditions, Eq. 4-2 can be regarded as a simplified version of Eq. 2-4. This results when (1) the exponent n of Eq. 4-2 has the value of 0.5 and (2) the factor $[\coth(\lambda/D')^{1/2} - (D'/\lambda)^{1/2}]$ has the value one; the latter obtains for the isotopes for which data on half-life dependence were presented. Accordingly, the relation between the constant A_j of Eq. 4-2 and the reduced diffusion coefficient of Eq. 2-4 is given by $A_j = 3.6 \sqrt{D'}$.

The in-pile data which have been used to evaluate the temperature dependence of the exponent n of Eq. 4-2 extend only to about 1300°C. The qualitative behavior of values of n derived from compacts (Ref. 4-15) indicates a decline in the value of n above 1500°C. Thus, extrapolation of the in-pile data to temperatures above 1500°C should be restricted until in-pile data are available for higher temperatures. At higher temperatures, a decline in the values of n might be expected on the basis of increased speed of diffusional transport of the gaseous fission products through the barriers to release.

4.2.2. Release of Long-Lived and Stable Isotopes

4.2.2.1. Experimental Data and Analysis. The dependence of fractional release on nuclide half-life has been treated above for isotopes with half-lives between 8 s and 5.3 d. For long-lived ($\tau_i > 10$ y) and stable isotopes, two additional factors must be considered: (1) radioactive decay will not significantly reduce the inventory of the long-lived nuclides in intact coated particles, and (2) the steady-state condition of release is attained only slowly -- thus, Eqs. 4-2 and 2-4 do not apply.

The calculation of the release of long-lived and stable isotopes can be made from knowledge of the accumulation of these isotopes in the free volume of intact particles; upon particle failure, the accumulated gases are released. This knowledge can also be used to calculate the release of the long-lived and stable isotopes from the failed particle.

Data on the fractional release of the long-lived and stable isotopes are limited. The release of stable isotopes of xenon and krypton as a function of temperature, time, and burnup (Refs. 4-39, 4-40) and of Kr-85 as a function of burnup (Ref. 4-41) has been determined for oxide kernels at temperatures above 900°C; releases have also been determined at a single temperature, time, and burnup for both oxide and carbide kernels (Ref. 4-42). These experiments were performed on intact coated particles, and the long-lived and stable isotopes accumulating in the free volume of the particles were measured at the end of the irradiation period.

The experiments (Refs. 4-39, 4-40, and 4-41) indicate that the accumulated fractional release is strongly dependent on the extent of burnup and relatively weakly dependent on temperature. Experiments conducted on TRISO-coated UO_2 kernels (Ref. 4-40) demonstrated that releases from higher-porosity kernels ($\sim 20\%$ porosity) are always greater than those from lower-porosity kernels ($\sim 5\%$ porosity). This is also found when appropriate comparisons are made between additional experiments (Ref. 4-41) on TRISO-coated UO_2 kernels with porosities in the range 10% to 20% and on BISO-coated ThO_2 kernels with porosities less than 2% (Ref. 4-39). Since the reference fertile particles for HTGRs have kernels of dense ThO_2 , the data on the BISO-coated ThO_2 kernels (Ref. 4-39) will be considered in detail.

The experiments (Ref. 4-39) on the release of long-lived and stable isotopes from ThO_2 kernels achieved a specific burnup after a continuous irradiation of the particles over a fixed time period. Comparison of different experiments corresponding to different burnups demonstrates the strong dependence of release (which far exceeds the recoil fraction) on burnup. Consequently, in a single experiment, the effective diffusion coefficient is increasing during the time of the experiment. There is, at present, no suitable model which may be used to quantitatively account for the increasing effective diffusion coefficient. Therefore, the data are analyzed here in terms of the equivalent sphere model (Ref. 2-12) to obtain an "average" effective reduced diffusion coefficient, $\bar{D}'$. This average value is defined as that constant value of the effective reduced diffusion coefficient which, applied to the equivalent sphere model equation, yields the observed release.

The data and the results of the analysis are presented in Table 4-10. The data are based on BISO-coated ThO_2 particles irradiated in the HT-12 through HT-15, P13L, and HRB-6 experiments. The weighted average temperatures given in Table 4-10 were calculated using the temperature profiles for the experiments (Refs. 4-39, 4-43) and a weighting factor $e^{-Q/RT}$.

TABLE 4-10
RELEASE DATA AND EFFECTIVE REDUCED DIFFUSION COEFFICIENTS FOR
ORNL EXPERIMENTS (Ref. 4-39)

Capsule	Location or Holder	Kernel	Average % FIMA	Irrad. Time (d)	Average T(°C) (a)	Percent ^(b) Release	τ (c)	$\bar{D}'$ (d) (s ⁻¹)
HT-12	26	ThO ₂	1.6	38.6	1481	18.2	7.1(-3)	2.1(-9)
P13L	C4T6	ThO ₂	3.0	423.5	1324	52.2	7.5(-2)	2.0(-9)
HT-13	8	ThO ₂	3.6	85.5	1204	21.4	1.0(-2)	1.4(-9)
HT-13	21	ThO ₂	6.0	85.5	1488	71.4	2.1(-1)	2.8(-8)
HT-13	26	ThO ₂	6.6	85.5	1548	76.9	2.9(-1)	3.9(-8)
HT-14	13	ThO ₂	9.5	137.0	1374	72.4	2.3(-1)	1.9(-8)
HT-14	21	ThO ₂	13.2	137.0	1559	84.4	4.8(-1)	4.1(-8)
HT-14	26	ThO ₂	14.7	137.0	1607	86.0	5.4(-1)	4.6(-8)
HT-15	26	ThO ₂	17.7	160.0	1607	98.1	1.3(0)	9.4(-8)
HRB-6	1C-2	Th _{0.81} U _{0.19} O ₂	21.8	184.0	1150	90.6	8.8(-1)	5.5(-8)
HRB-6	1C-1	Th _{0.81} U _{0.19} O ₂	23.1	184.0	1150	89.3	7.1(-1)	4.5(-8)

(a) Weighted average value of temperature (see text).

(b) Percent release of long-lived and stable isotopes of krypton plus xenon.

(c) From Fig. 1 of Ref. 2-12 ($\tau = \bar{D}'t$).

(d) Average effective reduced diffusion coefficient.

To establish a value of the activation energy, Q various values of Q were tested and a value of 105 kJ/mol (25 kcal/mol) was selected. This value yields effective diffusion coefficients which are consistent with (1) release data on short-lived isotopes as discussed below and (2) the known higher release for the higher-porosity kernels. However, the actual value of the activation energy could be larger than 105 kJ/mol (25 kcal/mol). This would be anticipated on the basis of data on the diffusional release of krypton and xenon isotopes in uncoated (Th,U)C₂ particles (Ref. 4-15). In these studies (Ref. 4-15), values of 242 and 326 kJ/mol (58 and 78 kcal/mol) were found for the average activation energy of krypton and xenon isotopes, respectively. Larger activation energies are also consistent with items (1) and (2) above.

The choice of the relatively low activation energy for diffusion of 105 kJ/mol (25 kcal/mol) is conservative. The experiments from which this activation energy was derived were conducted at 1400°C on the average; extrapolation of these data to the normal operating conditions of the HTGR at lower temperature results in an overestimate of the effective diffusion coefficient (and hence the release) for $Q = 105$ kJ/mol (25 kcal/mol) as compared with the estimate based on $Q > 105$ kJ/mol.

The average effective reduced diffusion coefficients of Table 4-10, normalized to a temperature of 1400°C, are shown in Fig. 4-4 together with curves obtained from the following fit functions:

$$\bar{D}' = 1.14 \times 10^6 e^{-105\,000/[RT(1+0.09F)]} \quad , \quad (4-3)$$

and

$$\bar{D}' = 1.14 \times 10^{-6} e^{-(105\,000/RT)+[7.47/(1+11F^{-1})]} \quad , \quad (4-4)$$

where F is the percent FIMA and R is the gas constant (8.314 J/mol•K).

Both Eqs. 4-3 and 4-4 fit the data equally well. In the first representation,

Fig. 4-4. Average effective reduced diffusion coefficient in ThO₂ kernels at 1400°C versus percent FIMA

Eq. 4-3, the activation energy may be considered as dependent on FIMA; i.e., $Q_{\text{eff}} = Q/(1+CF)$, where Q_{eff} is the effective activation energy. In the second representation, Eq. 4-4, the pre-exponential factor may be considered as dependent on FIMA; i.e., $\bar{D}'_{o,\text{eff}} = \bar{D}'_o e^{7.47/(1+11/F)}$. Clearly, no choice can be made between these representations on the basis of the data shown in Fig. 4.4. It should be noted that the data from Ref. 4-42 on particles with ThO_2 kernels are in agreement with the foregoing analysis.

Since the time periods of the ThO_2 experiments (Ref. 4-39) were small compared with the time periods over which the same burnup is achieved in a reactor under normal operating conditions, the question of the applicability of the derived reduced diffusion coefficients to HTGR conditions must be considered. The effective reduced diffusion coefficients applying to the stable and long-lived isotopes apply also to the short-lived isotopes of the same element. Thus, if the effective reduced diffusion coefficients were larger than those given by Eq. 4-3 or 4-4, the short-lived isotopes would be able to escape from the kernels predominantly by a diffusive or diffusive-related mechanism at temperatures below 1300°C , contrary to the interpretation of the data in Section 4.1, indicating dominance of recoil-related release. Conversely, the observation of the dominance of recoil-related release from the kernels for short-lived isotopes at temperatures below 1300°C sets a limit to the effective reduced diffusion coefficient for release of fission gases from the kernels. On the basis of these arguments, the average effective reduced diffusion coefficients presented above for the release of stable and long-lived isotopes are regarded as satisfactory for calculating release from ThO_2 kernels.

The experiment (Ref. 4-42) conducted at a single temperature, time, and burnup yielded data on particles with UC_2 kernels at 74% FIMA. The derived value of D' , corrected to 1400°C , is $4 \cdot 10^{-8} \text{ s}^{-1}$. The data for carbide kernels are insufficient to derive temperature and burnup dependencies.

4.2.2.2. Description of Release for Long-Lived and Stable Isotopes. The accumulated fractional release of long-lived and stable isotopes, G, is given by (Ref. 2-12)

$$G = 1 - (1/15\bar{D}'t) + (6/\pi^4\bar{D}'t) \sum_{n=1}^{\infty} (1/n^4) e^{-n^2\pi^2\bar{D}'t} \quad , \quad (4-5)$$

and the instantaneous fractional release, H, is given by (Ref. 2-12)

$$H = 1 - (6/\pi^2) \sum_{n=1}^{\infty} (1/n^2) e^{-n^2\pi^2\bar{D}'t} \quad , \quad (4-6)$$

where $\bar{D}'$ (s^{-1}) is given by either Eq. 4-3 or 4-4. These equations are based on the equivalent sphere model. Equation 4-5 can be used to calculate the accumulation of long-lived and stable isotopes in the free volume of intact particles and thus to calculate the quantity of fission gas released upon coating failure. Equation 4-6 can be used to calculate the instantaneous fractional release from failed particles. By using $\bar{D}'$ values obtained from Eq. 4-3 or 4-4, it is believed that adequate account will be taken for the recoil contribution to the release of long-lived and stable isotopes, since in the experiments from which the diffusion parameters were derived, recoiled atoms were, of necessity, included in the measurement of the accumulated release of gases from the kernels.

The accumulated fractional release, G, and the instantaneous fractional release, H, for the long-lived and stable isotopes from carbide kernels can be conservatively calculated using the equations given for release from ThO_2 kernels (Eqs. 4-3 through 4-6). This is based on (1) the above-mentioned finding of a value of $\bar{D}'$ ($4 \cdot 10^{-8} s^{-1}$) for UC_2 kernels at near peak fissile burnup of 74% FIMA, which is smaller than predicted by Eq. 4-3 or 4-4, and (2) the argument, given above, that larger values of the effective reduced diffusion coefficient found for the ThO_2 data would not be consistent with the data on the release of the short-lived isotopes as considered in Section 4.1.

4.2.3. Summation on Half-Life Dependence of Fission Gas Release

For short-lived isotopes ($\tau_i \leq 5.3$ d):

1. The half-life dependence of fission gas release is a function of temperature.
2. The exponent of the half-life, in the relationship between half-life and fractional release, Eq. 4-2, varies from 0.2 ± 0.1 at temperatures between 250° and 350°C to 0.5 ± 0.1 at temperatures between 850° and 1300°C, based on in-pile data.
3. Release occurs from the fissile material primarily by a mechanism involving recoil followed by gas phase and bulk diffusion in the surrounding material at temperatures up to 1300°C at least.

For long-lived ($\tau_i > 10$ y) and stable isotopes:

1. The release from kernels at higher temperatures can be described by an average effective reduced diffusion coefficient which is a function of burnup and which is derived on the basis of the equivalent sphere model.
2. The derived effective reduced diffusion coefficients are consistent with the observed release of short-lived isotopes at temperatures below 1300°C.

4.3. TEMPERATURE DEPENDENCE OF FISSION GAS RELEASE

In this section, the temperature dependence of fission gas release is discussed.

4.3.1. Data on Temperature Dependence of Fission Gas Release

Data on the temperature dependence of R/B for the following samples are considered in this section; these data apply to isotopes with half-lives not exceeding 5.3 d:

1. Three Fort St. Vrain type fuel rods.
2. A fuel rod containing bare UO_2 kernels.
3. A loose mixture of bare UO_2 kernels plus matrix.
4. Pellets containing UO_2 powder and natural graphite powder.
5. A compact consisting of bare kernels of $(\text{Th},\text{U})\text{C}_2$ dispersed in a graphite matrix.

The fuel rods, items 1 and 2, are considered to be the most representative of the fuel configuration used in HTGR systems. Also, the fuel rod data represent the most extensive, relevant data on the temperature dependence of fission gas release for the range of temperatures encountered under HTGR normal operating conditions. Accordingly, the fuel rod data are used as the basis for deriving parameters to describe the temperature dependence of fission gas release. Data for the other samples are presented for comparison.

The fuel rod data, which were measured at GA, represent samples which include both intact particles and bare kernels and thus represent the possible range of fuel configurations. The methods of measuring R/B are described in Section 3. The fuel rod samples were maintained at each measurement temperature for about 1 to 2 d to assure that steady-state conditions were attained and to allow several determinations at each temperature.

Data on the fuel rods containing intact particles are given in Tables 4-11 through 4-13. The particles in these fuel rods were TRISO-coated particles containing ThC_2/UC_2 or ThC_2 fuel kernels. The matrix material was formulated from natural flake graphite flour and coal tar pitch, the latter constituting about 70 wt % of the matrix material. The fuel rods were formed by the hot injection method and cured at 1800°C and were production-type Fort St. Vrain stock. Since these fuel rods contain intact TRISO-coated particles, the source of the released fission gas is attributable to contamination of the fuel rod matrix and outer pyrocarbon coatings of the particles.

Data on the fuel rod containing bare kernels are given in Table 4-14. This fuel rod was fabricated by using (1) bare kernels of UO_2 , (2) pyrocarbon-coated carbon particles (without fuel), and (3) the matrix material described above.

The data shown in Table 4-15 were obtained from samples made solely by a physical mixing of bare kernels of UO_2 and matrix. The matrix material was the same as described above.

Two additional experiments (Refs. 4-15, 4-44) in which fissionable material was dispersed in graphite-containing matrix can be compared with the above data. For one case (Ref. 4-44), UO_2 and natural graphite powder were formed into pellets and irradiated. The particle size of the powder was about $1\ \mu\text{m}$; thus, fission products escaped from the UO_2 particles primarily by recoil. About 90% of the fission products were found to be captured in the graphite.

In the second experiment (Ref. 4-15), bare kernels of $(\text{Th},\text{U})\text{C}_2$ were dispersed in a graphite matrix and R/B measurements were made under steady-state conditions during photofission. These experiments were performed at temperatures as high as 2000°C and involved R/B values as large as 30%, so mechanisms of release from the kernel other than recoil-related release would be expected to contribute.

TABLE 4-11
DATA ON FISSION GAS RELEASE FROM FORT ST. VRAIN TYPE FUEL ROD (NO. CR-12)

Temp. (°C)	$10^4/T(K)$	N ^(a)	R/B					
			Kr-85m	Kr-87	Kr-88	Kr-89	Kr-90	Kr-91
40.9	31.85	>6	2.32(-6)	1.99(-6)	1.81(-6)	1.23(-6)		
98	26.95	2	2.23(-6)	2.09(-6)	1.82(-6)	1.46(-6)		
215	20.49	2	3.08(-6)	2.81(-6)	2.56(-6)	2.10(-6)		
280	18.08	2	3.88(-6)	3.39(-6)	2.96(-6)	2.39(-6)		
416	14.51	2	6.80(-6)	5.61(-6)	5.19(-6)	3.88(-6)		
525	12.53	2	6.52(-6)	6.63(-6)	5.39(-6)	4.61(-6)		4.62(-6)
650	10.83	2	1.04(-5)	1.04(-5)	8.60(-6)	7.03(-6)	6.85(-6)	1.00(-6)
713	10.14	2	1.02(-5)	1.00(-5)	8.80(-6)	6.81(-6)		4.36(-6)
814	9.20	3	1.68(-5)	1.70(-5)	1.38(-5)	9.83(-6)	8.19(-6)	1.06(-5)
1015	7.76	2	3.95(-5)	2.93(-5)	3.17(-5)	1.58(-5)	3.94(-6)	9.59(-6)
1080	7.39	2	6.66(-5)	5.19(-5)	5.38(-5)	2.10(-5)	1.05(-5)	1.17(-5)
1140	7.08	2	8.41(-5)	6.38(-5)	6.77(-5)	2.71(-5)	1.04(-5)	1.70(-5)
1278	6.45	2	1.37(-4)	1.03(-4)	1.15(-4)	4.75(-5)	2.5(-5)	2.03(-5)
			Xe-133	Xe-135	Xe-135m	Xe-137	Xe-138	Xe-139
40.9	31.85	6	1.31(-5)	2.38(-6)	1.69(-6)	1.36(-6)	1.19(-6)	7.41(-7)
98	26.95	2	2.36(-6)	1.69(-6)	1.11(-6)	1.47(-6)	1.09(-6)	9.55(-7)
215	20.49	2		2.30(-6)	1.73(-6)	1.69(-6)	1.80(-6)	1.22(-6)
280	18.08	2	5.35(-6)	2.54(-6)	2.02(-6)	1.89(-6)	1.56(-6)	1.32(-6)
416	14.51	2	2.65(-5)	5.75(-6)	3.04(-6)	2.75(-6)	2.14(-6)	2.04(-6)
525	12.53	2	1.20(-5)	4.66(-6)	4.03(-6)	4.02(-6)	2.91(-6)	2.85(-6)
650	10.83	2	1.34(-5)	7.49(-6)	6.38(-6)	5.31(-6)	4.21(-6)	4.12(-6)
713	10.14	2	1.19(-5)	7.13(-6)	5.36(-6)	5.90(-6)	3.93(-6)	3.21(-6)
814	9.20	3	2.16(-5)	9.04(-6)	7.47(-6)	6.13(-6)	4.39(-6)	4.74(-6)
1015	7.76	2	5.21(-5)	1.67(-5)	1.03(-5)	5.24(-6)	5.60(-6)	4.79(-6)
1080	7.39	2	1.04(-4)	2.77(-5)	1.31(-5)	7.17(-6)	7.27(-6)	5.28(-6)
1278	6.45	2	1.44(-4)	8.02(-5)	4.51(-5)	1.81(-5)	1.91(-5)	1.03(-5)

(a) N = number of determinations of R/B for which the listed R/B values represent an average.

TABLE 4-12
DATA ON FISSION GAS RELEASE FROM FORT ST. VRAIN TYPE FUEL ROD
(No. XXX40)

Temp. (K)	$10^4/T(K)$	$N^{(a)}$	$10^5 R/B$				
			Kr-85m	Kr-87	Kr-88	Kr-89	Xe-138
1343	7.45	13	9.21	6.12	5.54	3.83	1.62
1291	7.75	1	11.4	7.08	6.71	3.86	1.06
1333	7.50	1	10.3	7.06	6.22	4.39	1.60
1364	7.33	1	8.82	5.93	5.32	5.3	2.1
1375	7.27	1	14.9	9.47	8.98	5.93	2.04
1447	6.91	5	18.7	11.8	11.9	7.1	3.15
1437	6.96	1	15.3	10.5	9.61	5.68	2.11
1427	7.00	3	17.2	11.3	10.5	6.19	2.32
1468	6.81	1	22.2	13.2	15.0	6.6	1.8
1547	6.46	1	30.9	18.0	19.8	10.1	5.1
1572	6.36	1	26.2	18.2	18.1	11.6	6.8
1562	6.40	3 ^(b)	23.9	16.0	16.7	8.37	3.6
1577	6.34	1	13.7	9.5	8.6	6.6	2.3
1463	6.84	1	18.2	12.8	12.7	6.6	3.3
1551	6.45	3	21.8	14.6	15.6	7.2	3.2
1541	6.49	1	30.9	19.9	21.0	10.4	4.1
1531	6.53	1	25.1	17.6	17.8	8.4	3.2
1557	6.42	1	19.6	14.3	14.3	7.2	2.5
1650	6.06	1	46.1	25.5	30.2	13.1	6.2
1656	6.04	3 ^(c)	34.4	22.6	22.1	11.8	5.0
1645	6.08	1	36.1	23.4	24.4	4.6	5.7
1676	5.97	1	35.8	24.4	24.5	12.8	--
1624	6.16	1	30.0	20.2	22.0	10.4	3.8
1635	6.12	(d)	24.8	9.3	9.6	5.8	3.6
1640	6.10	1	61.1	30.3	37.4	11.2	4.3
1750	5.71	5	44.2	26.0	29.8	14.5	8.0
1760	5.68		41.6	26.8	28.0	12.8	5.6
1739	5.75	2 ^(e)	41.4	27.4	27.4	14.3	7.2
1771	5.65	2	24.4	16.2	16.4	9.7	3.8
1781	5.61	1	41.7	28.4	28.9	14.0	6.2
1343	7.45	5	17.9	9.7	10.9	4.0	1.3
1353	7.39	3	25.6	12.3	15.9	4.0	2.3
1348	7.42	2 ^(f)	28.9	13.7	16.7	5.0	1.4
1323	7.56	1	11.7	8.1	7.3	5.6	2.7
1336	7.49	2	13.3	9.2	8.4	5.4	2.1
1362	7.34	2	14.7	10.0	9.0	6.2	2.2
1388	7.20	1	17.2	10.8	11.1	5.7	1.9
1358	7.36	5	14.9	9.3	8.1	5.0	2.2

(a) N = number of determinations of R/B for which the listed R/B values represent an average.

(b) N = 2 for Kr-88.

(c) N = 2 for Kr-89 and Xe-138.

(d) N = 6, 7, 5, 7, 6 for Kr-85m, Kr-87, Kr-88, Kr-89, and Xe-138, respectively.

(e) N = 1 for Kr-88.

(f) N = 1 for Xe-138.

TABLE 4-13
DATA ON FISSION GAS RELEASE FROM FORT ST. VRAIN TYPE FUEL ROD (No. CR-11)

Temp. (°C)	$10^4/T(K)$	N ^(a)	R/B				
			Kr-85m	Kr-87	Kr-88	Kr-89	Kr-91
38.6	32.09	4	2.21(-6)	2.53(-6)	1.95(-6)	1.21(-6)	4.96(-7)
823	9.12	4	3.45(-6)	3.26(-6)	2.84(-6)	1.78(-6)	1.88(-6)
958	8.12	4	1.94(-5)	1.69(-5)	1.42(-5)	8.16(-6)	3.21(-6)
1089	7.34	5	3.71(-5)	3.14(-5)	2.36(-5)	1.21(-5)	4.84(-6)
1399	5.98	8	5.74(-5)	5.73(-5)	4.89(-5)	3.06(-5)	2.53(-5)
1520	5.58	4	7.49(-5)	7.65(-5)	6.60(-5)	4.32(-5)	4.24(-5)
			Xe-133	Xe-135	Xe-138	Xe-139	
38.6	32.09	4	1.84(-5)	5.93(-6)	1.70(-6)	--	
823	9.12	4	9.18(-5)	2.91(-5)	1.10(-6)	4.94(-6)	
958	8.12	4	4.16(-5)	2.01(-5)	6.00(-6)	4.94(-6)	
1089	7.34	5	8.02(-5)	3.44(-5)	6.19(-6)	2.56(-6)	
1399	5.98	8	9.11(-5)	5.01(-5)	1.54(-5)	3.94(-6)	
1520	5.58	4	1.73(-4)	8.60(-5)	3.01(-5)	1.46(-5)	

(a) N = number of determinations of R/B for which the listed R/B values represent an average.

TABLE 4-14
DATA ON FISSION GAS RELEASE FROM FABRICATED BARE KERNEL ROD (No. 2B-20)

Temp. (°C)	$10^4/T(K)$	N ^(a)	R/B			
			Kr-85m	Kr-87	Kr-88	Kr-89
40	31.95	3	1.06(-3)	1.15(-3)	8.56(-4)	1.01(-3)
805	9.28	3	5.51(-3)	4.56(-3)	4.03(-3)	2.76(-3)
1087	7.35	4	4.85(-2)	4.16(-2)	3.86(-2)	1.38(-2)
1092	7.33	3	2.57(-2)	2.37(-2)	2.06(-2)	8.17(-3)
1412	5.94	3	1.59(-1)	1.19(-1)	1.14(-1)	5.45(-2)

(a) N = number of determinations of R/B for which the listed R/B values represent an average.

TABLE 4-15
DATA ON FISSION GAS RELEASE FROM BARE KERNELS OF UO_2 PLUS MATRIX

Temp. (°C)	$10^4/T(K)$	$N^{(a)}$	R/B					
			Kr-85m	Kr-87	Kr-88	Kr-89	Kr-90	Kr-91
41	31.8	3	8.70(-4)	8.49(-4)	7.18(-4)	4.35(-4)		
100	26.8	4	1.46(-3)	1.14(-3)	1.07(-3)	6.56(-4)		
310	17.2	4	1.76(-3)	1.72(-3)	1.40(-3)	8.57(-4)		1.29(-3)
435	14.12	2	3.19(-3)	3.00(-3)	2.52(-3)	1.77(-3)	1.43(-3)	1.92(-3)
603	11.42	3	4.16(-3)	4.05(-3)	3.46(-3)	2.62(-3)	1.67(-3)	2.98(-3)
715	10.12	2	6.87(-3)	6.28(-3)	5.81(-3)	4.20(-3)	2.92(-3)	4.31(-3)
810	9.23	2	1.04(-2)	9.75(-3)	8.50(-3)	5.04(-3)	3.71(-3)	5.53(-3)
950	8.18	3	1.52(-2)	1.30(-2)	1.23(-2)	6.73(-3)	4.83(-3)	6.68(-3)
1060	7.50	2	2.71(-2)	2.18(-2)	2.35(-2)	1.07(-2)	1.08(-2)	9.89(-3)
1080	7.39	1	2.71(-2)	2.16(-2)	2.34(-2)	1.06(-2)	1.36(-2)	5.17(-3)
1165	6.95	2	3.83(-2)	3.18(-2)	3.33(-2)	1.59(-2)	1.14(-2)	1.28(-2)
1265	6.50	2	5.53(-2)	4.20(-2)	4.70(-2)	1.93(-2)	1.59(-2)	1.70(-2)
			Xe-133	Xe-135	Xe-135m	Xe-137	Xe-138	Xe-139
41	31.8	2		5.81(-4)	5.70(-4)	5.86(-4)	4.36(-4)	2.76(-4)
100	26.8	3		7.91(-4)	6.67(-4)	4.78(-4)	5.06(-4)	2.06(-4)
310	17.2	3	1.48(-3)	1.16(-3)	1.21(-3)	1.33(-3)	8.64(-4)	7.14(-4)
435	14.12	2	4.13(-3)	2.33(-3)	1.40(-3)	1.18(-3)	1.01(-3)	8.52(-4)
603	11.42	3	3.23(-3)	2.06(-3)	2.12(-3)	1.76(-3)	1.40(-3)	1.36(-3)
715	10.12	2	5.15(-3)	2.88(-3)	2.50(-3)	2.15(-3)	1.74(-3)	1.42(-3)
810	9.23	2	1.05(-2)	3.54(-3)	3.03(-3)	2.33(-3)	2.10(-3)	2.22(-3)
950	8.18	3	6.37(-3)	3.83(-3)	3.04(-3)	2.49(-3)	2.13(-3)	2.14(-3)
1060	7.50	2	2.17(-2)	8.12(-3)	4.45(-3)	3.90(-3)	3.86(-3)	2.68(-3)
1080	7.39	1	4.61(-2)	1.36(-2)	1.00(-2)	6.35(-3)	3.58(-3)	2.08(-3)
1165	6.95	2	4.22(-2)	1.28(-2)	6.95(-3)	5.74(-3)	5.30(-3)	3.10(-3)
1265	6.50	2	4.12(-1)	6.33(-2)	3.04(-2)	6.13(-3)	8.18(-3)	4.36(-3)

(a) N = number of determinations of R/B for which the listed R/B values represent an average.

4.3.2. Interpretation of Data on Temperature Dependence of Fission Gas Release

Examination of all available temperature dependence data for isotopes with half-lives not exceeding 5.3 d shows that the temperature dependence of fission gas release is weak at low temperatures and relatively stronger at high temperatures. This general behavior applies to fuel configurations of interest. The characteristic temperature dependence in the temperature range between 40° and 1300°C is illustrated in Fig. 4-5 by the temperature profile of R/B for Kr-85m. (The data in Fig. 4-5 were taken from Table 4-11.)

In the temperature range between 40° and 1300°C, the observed temperature dependence can be represented by two contributions, a low-temperature contribution and a high-temperature contribution. This is illustrated by the asymptotes in Fig. 4-5. These two contributions apparently correspond to the two mechanisms of release considered in Section 4.2; the low-temperature portion of the R/B profile of Fig. 4-5 is governed by gas phase diffusion and the high-temperature portion by bulk diffusion.

The temperature dependence in the two temperature regions can be empirically evaluated from the R/B data by determining the activation energy, $-R[\partial \ln(R/B)/\partial(1/T)]$, derived from the function $R/B = A \exp(-q/RT)$. In the high-temperature region where bulk diffusion dominates release, the activation energy for R/B is one-half the activation energy for diffusion in accordance with Eq. 2-4. In the case of gas phase diffusion, the activation energy is not constant but is weakly dependent on the temperature. The exact dependence of $-R[\partial \ln(R/B)/\partial(1/T)]$ on temperature for gas phase diffusion can be found from the model of gas phase diffusion introduced in Section 4.4. For present purposes, the activation energy at low temperature will be regarded as constant; this can be justified on the basis of the uncertainty in R/B measurements being larger than the expected changes in the activation energy at low temperatures. For convenience, the activation energies for R/B data will be designated by q in the following discussion.

Fig. 4-5. Temperature profile of R/B for Kr-85m release from fuel rod CR-12

Activation energies derived from the fuel rod data of Tables 4-11 through 4-13 are given in Table 4-16. The method used in deriving the activation energies involved a visual fit of the function $R/B = A \exp(-q/RT)$ to a plot of the low-temperature data followed by a visual fit of the same function to the high-temperature data corrected for the contribution of the low-temperature process.

On the basis of Table 4-16, the value of q is chosen as the mean of the activation energies for fuel rods CR-11 and CR-12 in the high-temperature region (1000° to 1500°C); thus, for krypton the activation energy is 69.6 kJ/mol (16.6 kcal/mol), and for xenon it is 56.1 kJ/mol (13.4 kcal/mol). The activation energy in the low-temperature region (40° to 400°C) is taken as 2.9 kJ/mol (0.7 kcal/mol) for both krypton and xenon. The standard deviations of the activation energies for both krypton and xenon are about ± 17 kJ/mol (± 4 kcal/mol) in the high-temperature region and about ± 1.3 kJ/mol (± 0.3 kcal/mol) in the low-temperature region. These activation energies and standard deviations are considered to be the best values for use in reactor calculations because the Fort St. Vrain type fuel rods are considered to be more representative of the reference HTGR fuel rods than are the other fuel samples for which temperature dependence data exist.

Table 4-17 lists activation energies derived from the data in Table 4-14 for the fuel rod containing bare kernels. The method of deriving the activation energies was similar to that described above with an exception; since only one datum was available at low temperatures, the high-temperature data had to be corrected with this datum rather than by using a low-temperature curve of the form $R/B = A \exp(q/RT)$.

The average activation energy, 78.8 kJ/mol (18.8 kcal/mol), given in Table 4-17 agrees reasonably well with the average activation energy given above (69.6 kJ/mol) for the fuel rods containing intact particles. Thus, the switch from intact particles to bare kernels in the fuel rod had little effect on the release process, and this is consistent with release from the fissile material in both cases by recoil or recoil-related mechanisms.

TABLE 4-16
ACTIVATION ENERGIES FOR FRACTIONAL FISSION GAS RELEASE FROM
FUEL RODS CONTAINING INTACT PARTICLES

Element	Isotope	q (kJ/mol); 1000 ≤ T(°C) ≤ 1500			q (kJ/mol); 40 ≤ T(°C) ≤ 400		
		CR-12	XXK40	CR-11 (a)	CR-12	XXK40	CR-11
Krypton	85m	67.3	61.5	74.4	3.1	--	1.7
	87	56.5	58.2	78.2	3.5		1.3
	88	61.1	66.5	77.8	3.1		1.7
	89	51.9	52.3	66.9	4.1		1.7
	91			102.1	--		--
	Average	59.2	59.6	79.9	3.5		1.6
Xenon	133	68.6	--	66.5 ^(b)	3.9	--	(c)
	135	57.7	--	56.5 ^(b)	3.5	--	
	135m	59.8	--	--	3.1	--	
	137	29.7	--	--	1.2	--	
	138	37.7	--	69.0	2.3	--	
	139	26.8	--	69.5	3.8	--	
	Average	46.7	--	65.4	3.0		

(a) For xenon isotopes, the values of q for the high-temperature region were based on high-temperature data corrected by using the 39°C value of R/B, since there were insufficient data at low temperatures to follow the procedure outlined in the text.

(b) Datum at $10^4/T(K) = 9.12$ excluded in obtaining these values.

(c) For xenon there are insufficient data to obtain a value of q.

TABLE 4-17
ACTIVATION ENERGIES FOR FRACTIONAL FISSION GAS RELEASE
FROM FUEL ROD (2B-20) CONTAINING BARE KERNELS

Element	Isotope	$q(\text{kJ/mol}); 1000 < T(^{\circ}\text{C}) \lesssim 1400$
Krypton	85m	78.7
	87	74.1
	88	83.7
	89	78.7
	Average	78.8

Comparison of the activation energies for the above fuel rod data and the activation energies for bare kernels in loose matrix material suggests that release of fission gas from the kernel is by a mechanism involving recoils. Activation energies for the bare kernels in loose matrix material are listed in Table 4-18. For krypton isotopes, the average activation energy (for high temperatures) is smaller than any of the average activation energies for the fuel rods containing bare kernels or intact particles. Comparison of the activation energies for the bare kernels plus loose matrix and the bare kernels in fuel rod suggests dominance of the matrix material and its type in governing fission product release at temperatures between 1000° and 1300°C; if non-recoil-related release of fission gas from the kernel were rate controlling, activation energies for the two cases would be expected to be the same and to be higher in magnitude than found above. For xenon isotopes, the larger uncertainties associated with the average activation energies prevent reaching the conclusions found for krypton data. The average activation energies for krypton and xenon in the low-temperature region for the bare kernels plus loose matrix are not significantly different from those found for the fuel rod data, suggesting an independence of the low-temperature release mechanism on the matrix material.

Analyzing the data for the pellets of UO_2 and natural graphite powder (Ref. 4-44) yields an average activation energy for R/B at temperatures between 700° and 950°C of about 46 kJ/mol (11 kcal/mol) for krypton; this value is similar to that found for the data on bare kernels plus matrix in Table 4-18. For xenon, the temperature range of the experiments reported in Ref. 4-44 ($T_{\text{max}} = 950^\circ\text{C}$) was too small to analyze the data as in the case of the krypton isotopes. This difficulty may be attributed to a higher temperature for transition to the high-temperature region of release in the case of xenon; for the data of Table 4-15, the average transition temperature is 600°C for krypton and 800°C for xenon. (The transition temperature is defined as the temperature of the intersection of the two asymptotes in Fig. 4-5.) In the low-temperature region, the experiments (Ref. 4-44) yielded activation energies about 25% to 50% larger, in general, than did the experiments with bare kernels.

TABLE 4-18
ACTIVATION ENERGIES FOR FRACTIONAL FISSION GAS RELEASE FROM
MIXTURES OF BARE UO_2 KERNELS AND MATRIX^(a)

Element	Isotope	q (kJ/mol)	
		$1000 \leq T(^{\circ}\text{C}) < 1265$	$40 \lesssim T(^{\circ}\text{C}) < 400$
Krypton	85m	58.2	3.7
	87	50.2	3.8
	88	55.6	3.3
	89	41.0	3.1
	90	50.2	--
	91	36.0	--
	Average	48.5	3.5
	Standard deviation	8.7	
Xenon	133	95.0	--
	135	82.0	3.3
	135m	72.4	3.1
	137	41.4	3.3
	138	57.3	3.7
	139	38.9	5.2
	Average	64.5	3.7
	Standard deviation	22.5	

^(a) Activation energies were derived from data given in Table 4-15.

In the case of bare kernels of $(\text{Th,U})\text{C}_2$ dispersed in a graphite matrix (Ref. 4-15), the average activation energy for release was found to be 146 kJ/mol (35 kcal/mol) in the temperature range between 1000° and 2000°C after correction of the data given in Ref. 4-15 for the low-temperature contribution. This activation energy is considerably larger than those derived above from the other data. The relationship of the results for bare kernels of $(\text{Th,U})\text{C}_2$ in graphite matrix to those for bare kernels of UO_2 in matrix can be more readily appreciated by comparison, as in Fig. 4-6 where data for Kr-85m are shown. The extrapolation of the UO_2 data to temperatures above 1300°C is consistent with the $(\text{Th,U})\text{C}_2$ data provided allowance is made for slight curvature.

Above about 1600°C, the R/B values for $(\text{Th,U})\text{C}_2$ kernels plus matrix (in Fig. 4-6) exceed 10%; release is clearly dominated by diffusion from the bare kernels, and the activation energy of 146 kJ/mol (35 kcal/mol) is reasonable for this mode of release. This value of the activation energy for R/B is equivalent to a value of about 292 kJ/mol (70 kcal/mol) for the activation energy of diffusion.

In the temperature range 1000° to 1500°C, the R/B values for the $(\text{Th,U})\text{C}_2$ kernels plus matrix are lower than the R/B values for UO_2 kernels plus matrix as shown in Fig. 4-6. Also, the R/B values for $(\text{Th,U})\text{C}_2$ kernels plus matrix are lower than those for the fabricated bare kernel rod given in Table 4-14. Apparently, in the case of the $(\text{Th,U})\text{C}_2$ kernels plus matrix, the matrix, which was highly graphitized, was comparatively retentive of the fission gas nuclides that recoiled from the kernel; consequently, release by diffusion from the kernel was dominant. In the case of the bare UO_2 kernels plus matrix, the matrix, which was largely composed of binder material, was less retentive of the recoiled fission gas nuclides, and recoil-related release was dominant. Note that the $(\text{Th,U})\text{C}_2$ and UO_2 kernels had about the same diameter (around 200 μm) and thus had similar recoil fractions.

Fig. 4-6. Comparison of temperature profiles of R/B for Kr-85m between matrix-dispersed bare kernels of $(\text{Th,U})\text{C}_2$ and UO_2

4.3.3. Summation of Data on Temperature Dependence of Fission Gas Release

The temperature dependence of the fractional release, R/B , in terms of the activation energy, $-R[\partial \ln (R/B)/\partial (1/T)]$, was found to be:

1. 69.6 and 56.1 kJ/mol (16.6 and 13.4 kcal/mol) for krypton and xenon, respectively, in the temperature range 1000° to 1500°C.
2. 2.9 kJ/mol (0.7 kcal/mol) for both krypton and xenon below 400°C.

These values apply to intact and failed particles in fuel rods.

4.4. EQUATIONS DESCRIBING FISSION GAS RELEASE FROM FUEL RODS

On the basis of the data presented in Sections 4.2 and 4.3, a general equation is developed relating fractional release, temperature, pressure, and isotope half-life and taking into account the mechanisms of fission gas release at low and high temperatures.

4.4.1. Equations Describing Release of Short-Lived Isotopes

The data in Sections 4.2 and 4.3 lead to the following model which is the basis of a general equation describing the release of short-lived gaseous nuclides from fuel rods as a function of temperature, pressure, nuclide half-life, and mass. In this model, the release of the fission gases from the fissile material is predominantly by recoil or recoil-activated processes at temperatures below 1300°C. The recoil-released fission gases undergo two subsequent processes: (1) bulk diffusion of the fission gas and (2) gas phase diffusion. The former process occurs for fission gas atoms which recoil into the solid non-fissile material; the latter process occurs for fission gas atoms (1) whose recoil paths terminate at weakly

binding sites, (2) which are released from the fissile material by knock-out, or (3) which have reached the surface of a grain by bulk diffusion and are released into the pore system of the fuel rod. Neutron fluence (burnup), neutron flux, and changes in fuel configuration are assumed to have a negligible effect. (However, see Sections 4.5.2 and 4.6.2.)

The quantitative description of fission gas release, based on the above model, is given by the following equations:

$$(R/B)_{ij} = \frac{C_j}{\left[\cosh \sqrt{\lambda_i \rho / \mathcal{D}'_j} \right]^\theta} + 3(1-C_j) \sqrt{\mathcal{D}'_j / \lambda_i} [\coth \sqrt{\lambda_i / \mathcal{D}'_j} - \sqrt{\mathcal{D}'_j / \lambda_i}], \quad (4-7)$$

$$\mathcal{D}'_j = \mathcal{D}'_{o,j} T^{3/2} / P \sqrt{m_j^*}, \quad (4-8)$$

$$1/m_j^* = 1/m_j + 1/m_{\text{He}}, \quad (4-9)$$

$$\mathcal{D}'_j = \mathcal{D}'_{o,j} e^{-Q_j/RT}, \quad (4-10)$$

where $(R/B)_{ij}$ = fractional release for isotope i of element j ,

C_j = dimensionless constant for element j ,

λ_i = decay constant of isotope i (s^{-1}),

$\mathcal{D}'_j$ = reduced gas phase diffusion coefficient for element j (s^{-1}),

ρ, θ = constants,

$\mathcal{D}'_j$ = reduced bulk diffusion coefficient for element j (s^{-1}),

$\mathcal{D}'_{o,j}$ = reduced gas phase diffusion constant for element j
 $[\text{MPa}/(\text{K})^{3/2} \cdot s^\rho]$,

T = temperature (K),

P = pressure (MPa),

m_j = atomic weight of element j ,

$D'_{o,j}$ = reduced bulk diffusion constant for element j (s^{-1}),

Q_j = activation energy for bulk diffusion of element j
(J/mol),

R = gas constant (= 8.314 J/mol·K).

The quantitative description of the gas phase diffusion contribution for a distributed source, as represented by the first term of Eq. 4-7, is based, as follows, on analogy with that for a fixed source of diffusant. For a slab of material through which there is gas phase diffusion and for which (1) a constant flux of gas atoms (fixed source) is incident on face A and (2) the concentration of gas is zero on the opposite face, B, the fractional release is given by

$$R/B = [\cosh (\sqrt{\lambda/D'})]^{-1}, \quad (4-11)$$

where λ is the nuclide decay constant and D' is the reduced gas phase diffusion coefficient; $D' = D/a^2$, where a is the dimension of the slab perpendicular to the two faces of interest and D is the diffusion coefficient. The fractional release of Eq. 4-11 represents the ratio of the flux leaving face B to that incident on face A of the slab. By analogy, the fractional release for the distributed source case is taken to be

$$R/B = [\cosh (\sqrt{\lambda^\rho/D'})]^{-\theta}, \quad (4-12)$$

where ρ and θ are constants to be determined from the available data. Equation 4-12 is an empirical equation and its use has to be justified at present on the basis of correlations of the fission gas release data.

The quantitative description of the bulk diffusion contribution, the second term of Eq. 4-7, is based on the familiar equivalent sphere model (Ref. 2-12) as represented by Eq. 2-4.

Equation 4-7 has reasonable limits. As $\lambda \rightarrow 0$, the denominator of the first term approaches one and the first term approaches C; the second term

approaches 1-C and therefore R/B approaches one. As $T \rightarrow \infty$, R/B becomes equal to or less than $f + C(1-f)$, where f is the value of $t_2/(1-C)$, t_2 being the second term of Eq. 4-7. The value of f is always less than one and the value of R/B will remain less than one as $T \rightarrow \infty$ provided $C \leq 1.0$; this condition is satisfied as shown below.

The parameters of Eq. 4-7 have been evaluated, in the case of krypton, using data on fission gas release from the Fort St. Vrain type fuel rods as measured in TRIGA tests, except for the reference values of R/B for the Kr-85m at 1100°C. The latter values were chosen to be 3×10^{-5} for as-manufactured fuel rods on the basis of present design criteria and as 5×10^{-3} for fuel rods with failed-constrained fuel particles. The parameters are listed in Table 4-19a and the estimated uncertainties of the parameters in Table 4-19b. The parameters ρ , θ , $\mathcal{D}'_{o,j}$, m^* , and Q are common to both the as-manufactured and failed-fuel cases. For the parameter $\mathcal{D}'_{o,j}$, a difference is possible owing to the difference in fuel rod configuration brought about by the failed particles; however, there are no data which demonstrate such a difference and for the present, this possibility is ignored. Also, note that there is at present no independent measurement of the reduced gas phase diffusion coefficient, $\mathcal{D}'_j$, so that the value of $\mathcal{D}'_{o,j}$ was derived by means of the fit of Eq. 4-7 to the fission gas release data. In evaluating the parameters in the case of xenon, there are no reference R/B values; this problem is circumvented by using values of the ratios of R/B for krypton to xenon which are available from a variety of sources as presented below (see Table 4-20).

To illustrate the fit to TRIGA test data and the predictions for in-pile data based on Eq. 4-7, two calculations have been made. These calculations, to a large extent, are independent of the absolute, reference R/B values used in deriving the parameters of Eq. 4-7. In one of the calculations, the quantity $-\langle \partial \ln (R/B) / \partial \ln \lambda \rangle$ is determined as a function of temperature. This quantity represents the average value of the slope of the $\ln (R/B)$ versus $\ln \lambda$ curve over the range of λ values represented by the available data. In other words, the quantity calculated is the mean

TABLE 4-19a
VALUES OF PARAMETERS FOR USE IN FISSION GAS RELEASE EQUATION (EQ. 4-7)

Parameter	Values for Kr in Fuel Rods		Values for Xe in Fuel Rods	
	As-Manufactured	Failed Fuel Particles	As-Manufactured	Failed Fuel Particles
ρ	0.18	0.18	0.18	0.18
θ	0.3	0.3	0.3	0.3
C_j	2.11(-6)	3.52(-4)	1.81(-6)	2.98(-4)
$D'_{o,j}$ (MPa/K ^{3/2} ·s ^{ρ})	1.21(-6)	1.21(-6)	1.42(-6)	1.42(-6)
m^*	3.82	3.82	3.88	3.88
$D'_{o,j}$ (s ⁻¹)	6.93(-10)	1.885(-5)	8.80(-12)	2.39(-7)
Q (J/mol)	138 000	138 000	113 000	113 000
Q (cal/mol)	33 000	33 000	27 000	27 000

TABLE 4-19b
ESTIMATED UNCERTAINTIES FOR PARAMETERS IN THE FISSION
GAS RELEASE EQUATION (EQ. 4-7)

Parameter	Equation	Estimated Fractional Standard Deviation
ρ	4-7	+1.0 -0.5
θ	4-7	+1.0 -0.5
C_j	4-7	+6.0 -0.9
D'_j	4-7, 4-8	+2.0 -0.7
$D'_{o,j}$	4-7, 4-10	+2.0 -0.5
Q_j	4-10	±0.3

TABLE 4-20
VALUES OF A_j AND THE RATIOS A_{Kr}/A_{Xe} ^(a)

Source	Temp. (°C)	A_{Kr}	A_{Xe}	A_{Kr}/A_{Xe}
Capsule 6F2 _I	1300	6.36(-4)	2.58(-4)	2.47
Capsule 6F2 _{II}	1300	2.80(-3)	1.18(-3)	2.37
Capsule 6F2 _{III}	1300	3.97(-3)	1.98(-3)	2.00
GAIL III-B Loop	1130	2.05(-3)	8.17(-4)	2.51
GAIL IV Loop	990	1.63(-5)	3.00(-6)	5.43
Peach Bottom, Core 1, 30% power	850	1.08(-4)	6.03(-5)	1.79
Peach Bottom, Core 1, full power	850	1.09(-4)	5.68(-5)	1.92
Peach Bottom, Core 1, EOL	850	1.25(-3)	4.45(-4)	2.81
Pluto A Loop, Charge 8	1130	4.29(-6)	1.88(-6)	2.28
Pluto IIIa Loop	--	4.46(-4)	--	--
Peach Bottom, Core 2	1000	6.38(-5)	3.39(-5)	1.88
Fort St. Vrain	300			1.00
Fuel Rod CR-11	39	1.87(-6)	2.84(-6)	0.66
	823	2.83(-6)	--	--
	958	1.38(-5)	6.31(-6)	2.19
	1089	2.40(-5)	7.63(-6)	3.15
	1399	4.79(-5)	1.23(-5)	3.89
	1520	6.51(-5)	2.80(-5)	2.33
	41	1.80(-6)	2.29(-6)	0.79
	98	1.88(-6)	1.44(-6)	1.30
	215	2.62(-6)	1.97(-6)	1.33
	280	3.12(-6)	2.33(-6)	1.34
Fuel Rod CR-12	416	5.29(-6)	4.81(-6)	1.10
	525	5.80(-6)	4.74(-6)	1.22
	650	9.15(-6)	6.62(-6)	1.38
	713	8.78(-6)	6.05(-6)	1.45
	814	1.42(-5)	8.16(-6)	1.74
	1015	2.61(-5)	1.18(-5)	2.21
	1080	4.44(-5)	1.73(-5)	2.57
	1140	5.51(-5)	--	--
	1278	9.32(-5)	4.04(-5)	2.30

^(a) A_j is defined by Eq. 4-2.

value of the half-life exponent, n , in Eq. 4-2. The results of this calculation are shown in Figs. 4-7 and 4-8. The experimental data shown are from Tables 4-7 and 4-8. The fits of Eq. 4-7 account for the general features of the data, but there is a large scatter in the data.

The other calculation is made in terms of the ratio $A_{\text{Kr}}/A_{\text{Xe}}$, which represents the ratio of the fractional release of krypton to that of xenon for isotopes of equal half-lives (hypothetical) of 1 h. These ratios are given in Table 4-20 for fuel rods, loops, capsules, and reactors. Comparisons between the observations and predictions for the ratio $A_{\text{Kr}}/A_{\text{Xe}}$ are shown in Fig. 4-9 as a function of temperature. As shown, the calculations are in agreement with the data within the scatter and, as expected, the pressure dependence of the ratio is weak.

In Section 4.3.2, the activation energy in the high-temperature region was found to be larger for krypton than for xenon, although the uncertainties in the absolute values of the activation energies made the mean values significantly different at only the 30% confidence level. That the value is larger for krypton than for xenon is clearly shown in Fig. 4-10 by the unreasonable result obtained by repeating the calculations for the ratio $A_{\text{Kr}}/A_{\text{Xe}}$ but using equal activation energies for krypton and xenon.

4.4.2. Equations Describing Release of Long-Lived and Stable Isotopes

The mechanism of fission gas release for long-lived and stable isotopes is different from that for short-lived isotopes in the high-temperature range, at least up to 1300°C. The long-lived and stable isotopes are predominantly released from the kernel by diffusive processes (see Section 4.2.2). A description of the release in the high-temperature range is given in Section 4.2.2.2. As the temperature is lowered, the dominant release mechanism will change and become the same as that for short-lived isotopes, i.e., release from the kernel by recoil-related processes followed by gas phase diffusion.

Fig. 4-7. Comparison of predicted and observed half-life dependence as a function of temperature for krypton nuclides

Fig. 4-8. Comparison of predicted and observed half-life dependence as a function of temperature for xenon nuclides

Fig. 4-9. Comparison of predicted and observed ratio A_{Kr}/A_{Xe} as a function of temperature

Fig. 4-10. Effect of activation energy on the ratio A_{Kr}/A_{Xe} as a function of temperature

In the absence of data on the low-temperature release of long-lived and stable isotopes, a conservative estimate of the low-temperature contribution to release may be made using the recoil fraction. Thus, Eqs. 4-5 and 4-6 become

$$G = 1 + (1-f_R) \left[-(1/15) \bar{D}' t + (6/\pi^4 \bar{D}' t) \sum_{n=1}^{\infty} (1/n^4) e^{-n^2 \pi^2 \bar{D}' t} \right] \quad (4-13)$$

$$H = 1 + (1-f_R) \left[-(6/\pi^2) \sum_{n=1}^{\infty} (1/n^2) e^{-n^2 \pi^2 \bar{D}' t} \right] , \quad (4-14)$$

and

$$f_R = 0.75 (r/a) - 0.0625 (r/a)^3 , \quad (4-15)$$

where f_R is the recoil fraction, r is the recoil range, and a is the kernel radius. Equations 4-13 and 4-14 are analogous to Eqs. 4-5 and 4-6 but with the contribution of recoil fraction included. Equations 4-13, 4-14, and 4-15 plus Eqs. 4-3 and 4-4 describe the release of long-lived and stable isotopes. The values of the parameters for these equations are given in Table 4-21a and estimated uncertainties are given in Table 4-21b. $\bar{D}'$ values are derived by using values of $\bar{D}'_0$ (from Table 4-21a) in Eq. 4-3 or Eq. 4-4.

4.4.3. Summation of Equations Describing Fission Gas Release

A relationship (Eq. 4-7) between the steady-state fractional release, R/B , for short-lived nuclides and the temperature, pressure, and nuclide half-life and mass was developed. This relationship is based on a model for fuel rods in which fission gases (1) escape from fissile material predominantly by recoil or recoil-related processes and (2) subsequently escape from the fuel rod predominantly by (a) gas phase diffusion at temperatures below 800°C and (b) bulk diffusion at temperatures above 900°C. This relationship is valid for temperatures up to 1300°C and for nuclides with half-lives greater than 8 s under steady-state conditions.

TABLE 4-21a
VALUES OF PARAMETERS IN FISSION GAS
RELEASE EQUATIONS (Eqs. 4-3, 4-4, 4-13, 4-14, 4-15)

Parameter	Value for Kr	Value for Xe
$\bar{D}'_o$ (s^{-1})	1.14(-6)	1.14(-6)
$\bar{Q}$ (kJ/mol)	105	105
$\bar{Q}$ (kcal/mol)	25	25
a (μm)		
ThO ₂ kernels	250	250
UC ₂ kernels	100	100
r (μm)		
ThO ₂ kernels	10	5.5
UC ₂ kernels	9.5	5

TABLE 4-21b
ESTIMATED UNCERTAINTIES FOR PARAMETERS IN THE
FISSION GAS RELEASE EQUATIONS

Parameter	Equation	Estimated Fractional Standard Deviation
$\bar{D}'_o$	4-3, 4-4	+1.0 -0.5
f_R	4-15	+1.0 -0.2
$\bar{Q}_j$	4-3, 4-4	+3.0 -0.5

Equations (Eqs. 4-3, 4-4, 4-13, 4-14, and 4-15) were presented for calculating the release of long-lived and stable isotopes.

4.5. EFFECT OF NEUTRON FLUX LEVEL (OR FISSION RATE DENSITY)

The effect of neutron flux level, or fission rate density*, on fission gas release must be considered in predicting in-pile (capsule, loop, and reactor) fission gas release from laboratory (low flux level) test data and in correlating fission gas release data measured under different neutron flux conditions. The effect of the neutron flux level on fission gas release is usually considered to be insignificant. A review of the data, presented below, indicates that the effect is probably not significant for HTGR-type fuel, but this needs to be verified by further work.

The neutron flux level range of interest in HTGR-related studies is 10^{16} to 10^{18} $\text{n/m}^2\cdot\text{s}$. This includes the range of neutron flux levels available in the TRIGA King furnace facility ($1.6\cdot 10^{15}$ to 1.6×10^{17} $\text{n/m}^2\cdot\text{s}$) and the range in the HTGR systems (around 10^{18} $\text{n/m}^2\cdot\text{s}$). The fission rate density for dense fissile fuel kernels in an HTGR is around 2×10^{21} fissions/ $\text{m}^3\cdot\text{s}$.

4.5.1. Experimental Results on Effect of Neutron Flux Level on R/B

Several investigators have studied the effect of neutron flux level, or fission rate density, on fission gas release (R/B). The results disagree to some extent, but possible explanations exist for the disagreement.

Carroll et al. (Refs. 4-45, 4-46) at ORNL found a significant effect of fission rate density on fission gas release. They used an in-pile loop experiment in which the fission rate density in UO_2 fuel specimens could be varied independently of the temperature. Three specimens were irradiated

*Neutron flux ($\text{n/m}^2\cdot\text{s}$) and fission rate density (fissions/ $\text{m}^3\cdot\text{s}$) are related by the cross section (m^2) and concentration of fissile atoms (atoms/ m^3).

separately: (1) a hollow cylinder specimen of single crystal UO_2 , (2) a hollow cylinder of fine grain UO_2 , and (3) a bed of spheres of fused crystal UO_2 .

The ORNL investigators combined their results with those of Soulhier (Ref. 4-47) in a plot of fission gas release versus fission rate density, shown in Fig. 4-11 (from Ref. 4-46). The results indicate that an increase in fission rate density reduces R/B up to a fission rate density of about 10^{19} fissions/ $\text{m}^3 \cdot \text{s}$, whereas an increase in fission rate density above about 5×10^{19} fissions/ $\text{m}^3 \cdot \text{s}$ increases R/B.

To explain this behavior, Carroll (Ref. 4-45) postulated that the fissioning process which creates fission gas also creates traps to retard escape of the fission gas and that at high fission rate densities (around 10^{20} and above), the probability of fission gas escaping along overlapping fission spikes increases exponentially with fission rate density.

Data obtained at GA on the effect of neutron flux level on R/B are given in Table 4-22. These data were derived from short-time (1 h or less) fission gas release tests at 1100°C performed in the King furnace facility. As shown in Table 4-22, a variety of HTGR-type fuel configurations was studied, including artificially contaminated fuel particles with TRISO-coated ThC_2 kernels, and Triplex-coated (pyrolytic-carbon-coated) fuel particles like those used in the GAIL IV in-pile loop experiment (Ref. 4-19). The GAIL IV particles were tested in order to obtain R/B data for use in correlating laboratory and loop data. The data in Table 4-22 indicate that fission gas release is independent of the fission rate density in the range between $1.5 \cdot 10^{16}$ and $2.2 \cdot 10^{20}$ fissions/ $\text{m}^3 \cdot \text{s}$. The trends in the data for bare UO_2 kernels, both normal and enriched, are not presently regarded as significant since the resulting changes in R/B are less than the uncertainty in the measurement of R/B.

Fig. 4-11. Effect of fission rate density on fractional release of Kr-85m from UO_2 at 1400°C (Ref. 4-46)

TABLE 4-22
GA DATA ON THE EFFECT OF NEUTRON FLUX LEVEL ON FISSION GAS RELEASE FOR
VARIOUS HTGR-TYPE FUEL CONFIGURATIONS^(a)

Sample Configuration	Flux (n/m ² ·s)	Fission Rate Density (f/m ³ ·s)	TRIGA Test Temp. (°C)	R/B			
				Kr-85m	Kr-88	Kr-87	Kr-89
Bare UO ₂ (enriched U) kernels, 200-μm dia., in loose bed	1.6 x 10 ¹⁵	2.2 x 10 ¹⁸	1100	1.34(-2)	1.44(-2)	1.75(-2)	1.76(-2)
	1.6 x 10 ¹⁶	2.2 x 10 ¹⁹	1100	1.45(-2)	1.52(-2)	1.69(-2)	1.49(-2)
	1.6 x 10 ¹⁷	2.2 x 10 ²⁰	1100	1.13(-2)	1.30(-2)	1.42(-2)	1.57(-2)
Bare UO ₂ (normal U) kernels, 250-μm dia., in loose bed	1.6 x 10 ¹⁵	1.5 x 10 ¹⁶	1100	1.55(-2)	1.65(-2)	1.92(-2)	1.83(-2)
	1.6 x 10 ¹⁶	1.5 x 10 ¹⁷	1100	1.40(-2)	1.65(-2)	1.84(-2)	1.60(-2)
	1.6 x 10 ¹⁷	1.5 x 10 ¹⁸	1100	1.30(-2)	1.57(-2)	1.65(-2)	1.44(-2)
Triplex-coated (Th,U)C ₂ ^(b)	1.6 x 10 ¹⁵	(c)	1100	2.88(-5)	3.14(-5)	3.90(-5)	3.97(-5)
	1.6 x 10 ¹⁶	(c)	1100	2.93(-5)	2.87(-5)	3.65(-5)	3.93(-5)
	1.6 x 10 ¹⁷	(c)	1100	3.81(-5)	5.31(-5)	5.38(-5)	4.59(-5)
Contaminated TRISO-coated ThC ₂ ^(d)	8.5 x 10 ¹⁴	(c)	1400	1.6(-1)			
	4.2 x 10 ¹⁶	(c)	1400	1.4(-1)			

(a) The data were obtained from 30-min irradiation tests utilizing the TRIGA King furnace facility at GA.

(b) Triplex-coated particles like those used in the GAIL IV loop experiment. (Triplex coatings consist of three layers: buffer, laminar pyrolytic carbon, and columnar pyrolytic carbon.)

(c) Fission rate density is low but how low is unknown. Release was from uranium contamination and it is not known how the contamination was distributed.

(d) The particles were dipped in a uranium nitrate solution, evaporated to dryness, and then annealed.

Findlay et al. (Ref. 4-48) studied fission gas release from loose beds of uranium-plutonium dioxide particles during irradiation to burnups as high as 12%. The particles were made by crushing pellets to sieve sizes. Two samples of particles were tested, one in the sieve size range -10 to +16 (1190 to 2000 μm) and the other in the range -30 to +100 (149 to 589 μm). Each sample was tested at 550° and 1150°C and at fission rate densities of 2.6×10^{18} and 8.4×10^{19} fissions/ $\text{m}^3 \cdot \text{s}$. Kr-85m values found were in the range of 5×10^{-4} to 1.6×10^{-2} . Findlay et al. (Ref. 4-48) concluded that under their range of conditions, R/B is not significantly affected by fission rate density.

4.5.2. Discussion and Conclusions on Effect of Neutron Flux Level on R/B

Carroll (Ref. 4-46) found a significant effect of fission rate density on fission gas release, whereas GA investigators and Findlay et al. (Ref. 4-48) did not. Considerations that could explain this apparent disagreement are presented below.

One consideration is the difference in the size and configuration of the specimens used in the investigations. Surface-to-volume ratios for the UO_2 specimens used by Carroll et al. (Ref. 4-45) were 6 mm^{-1} , whereas ratios for the UO_2 fuel kernel specimens used in the GA work were $>24 \text{ mm}^{-1}$. It is surmised that different release mechanisms predominate for the different size specimens. Release from the small fuel particle specimens in the GA experiments is predominantly by surface-related processes such as ready release of recoiled fission atoms deposited on or near surfaces and knockout; release from the large specimens of Carroll et al. may be dominated by bulk processes including the creation and destruction of traps by fission events in the fuel. These considerations are consistent with the data, i.e., no effect of neutron flux level for the small specimens and an effect for the large specimens.

A second consideration is the time duration of the experiments. Short-time (30 min) TRIGA experiments were involved in the GA work, whereas

long-time in-pile loop experiments were involved in the ORNL work. It may be that there was not time enough in the GA experiments to establish the mechanisms of release important in the long-time experiments. This could explain why no effect of neutron flux level was found in the GA work.

A third consideration is the temperature difference. The ORNL experiments were performed at 1400°C and the GA experiments at 1100°C. At temperatures below 1300°C, the release of Kr-85m and other short-lived isotopes is predominantly by processes involving recoils (see Section 4-2). At higher temperatures, other mechanisms of release become dominant and the effect of fission rate density may be more pronounced for these mechanisms.

On the basis of the above data and considerations, it is concluded that the effect of neutron flux level on fission gas release is probably not important for HTGR-type fuel under normal operating conditions. However, this should be verified by further investigation.

4.6. EFFECT OF NEUTRON FLUENCE ON FISSION GAS RELEASE

This section describes the effect of neutron fluence (and burnup) on fission gas release.

4.6.1. Effect of Neutron Fluence on Fission Gas Release from Intact Fuel Rods

Examination of R/B data for reference-type fuel rods irradiated in capsule experiments shows cases where R/B values were low at beginning of life and did not change very much after irradiation to high neutron fluence. Examples of such data are given in Tables 4-11 and 4-12 of Ref. 4-49.

It can be deduced from these data that irradiation did not appreciably affect (1) the ability of the particle coatings to retain fission gases, (2) the release of fission gases from the low levels of uranium and thorium contamination, and (3) the fraction of failed particles. Thus, fission gas

release from reference-type fuel rods (i.e., $(R/B)_{am}$, as defined by Eq. 2-1) is not significantly affected by irradiation, outside of a possible increase due to particle coating failure.

It should be noted here that decrease in R/B with irradiation time has been observed in measurements of R/B in the Peach Bottom reactor (Core II) (Ref. 4-50) and the German pebble bed reactor, AVR (Ref. 4-51). This phenomenon is possibly due to a slow sintering process in which the kernel material densifies.

4.6.2. Effect of Burnup on Fission Gas Release from Failed Fuel Configurations

The effect of burnup on fission gas release has been studied mainly for fissile particles of various configurations including (1) in-pile failed particles, (2) loose failed particles, (3) laser-failed particles, and (4) bare kernels. These data have been presented in Tables 4-1, 4-3, and 4-4 for particles with dense UC_2 , $(Th,U)C_2$, and UO_2 kernels. The data cover the burnup range between 0% and 63% FIMA and indicate no dependence on burnup within a factor of two of the average R/B value for Kr-85m at 1100°C. For the UC_2 data alone, the variation in R/B with burnup is within 50% of the average R/B value over the burnup range from 0% to 63% FIMA. Burnette et al. (Ref. 4-1) further treated the laser-drilled UO_2 particles described in Table 4-4 by burning off the coatings in air at 800°C, leaving the bare kernels. Fission gas release measurements performed on the bare kernels yielded an R/B of 0.027 ± 0.0048 for Kr-85m at 1100°C, indicating no dependence on burnup over the range 0% to 30% FIMA within 20% of the average value.

For dense ThO_2 kernels, data (see Table 4-2) have been obtained at General Atomic only up to about 2% FIMA. The burnup dependence cannot be established on the basis of these data.

The release of long-lived and stable isotopes has been shown in Section 4.2.2 to be strongly dependent on burnup for dense ThO_2 kernels.

This dependence suggests a significant change in kernel microstructure leading to increased diffusive release; also, this change could affect the release of short-lived isotopes by the mechanism involving recoils.

At temperatures above 1300°C where diffusive release of the short-lived isotopes becomes important, a burnup dependence is expected.

Further experiments are required to establish the burnup dependence of R/B for particles with dense ThO_2 kernels.

4.7. RELEASE OF HALOGEN AND RELATED ELEMENTS

Information on iodine release from HTGR fuel materials has been derived from laboratory, in-pile loop, and reactor experiments. These results are compared below with data on the release of xenon isotopes. Little information exists on the release of tellurium, bromine, and selenium isotopes.

4.7.1. Iodine Release Data

Iodine release data have been obtained from decay experiments, which involve determination of the xenon daughter activity following a reactor or loop shutdown (see Section 3.3), and from diffusion tube studies (see Section 3.3 and Ref. 4-52) performed in conjunction with GAIL IV, a General Atomic in-pile loop experiment.

Figures 4-12 and 4-13 (taken from Ref. 4-13) include iodine release results obtained from diffusion tube measurements performed in the GAIL IV experiment (Ref. 4-19) and from decay experiments performed in the Peach Bottom HTGR (Ref. 4-33), the GA2-284-6F2 irradiation experiment (Ref. 4-16), and the GAIL IIIB loop experiment (Ref. 4-18). (Note that the R/B data for GAIL IV and 6F2 (Cell 1) are representative of intact fuel particles, and the data for GAIL IIIB and Peach Bottom are representative of particles with failed coatings. The fuel particles used in GAIL IIIB and Peach Bottom Core 1 had an early type of coating applied mainly to protect the carbide fuel kernels during fuel manufacture. These coatings failed, as expected, during irradiation.)

LC 81167

Fig. 4-12. R/B values vs half-life for GAIL IV and Peach Bottom Core 1

LC 81166

Fig. 4-13. R/B values vs half-life for 6F2 (cell 1) and GAIL IIIB

The data from GAIL IV show iodine releases lower than those for xenon. However, since the iodine R/B values were derived from the diffusion tube measurements, these values might actually be larger (see Section 3.3). (This is indicated by the vertical arrows in Fig. 4-12. Note also the horizontal arrows; these are a reminder that the effective half-life of Xe-135 is smaller than the value represented in Fig. 4-12 as a result of the large neutron cross section of Xe-135.)

The data from the Peach Bottom HTGR (Core 1) indicate that iodine and xenon have similar release behavior. However, R/B values for iodine are probably too high, since these values were derived from decay measurements (see Section 3.3).

The R/B values for Xe-133 and I-133 from both the GA2-284-6F2 (Cell 1) and GAIL IIIB tests are regarded as too large because of background activity (Refs. 4-16 and 4-18). However, the values for I-135 are comparable to those expected for xenon. Again, the I-135 values may be too high since these values were derived from the decay method.

One can deduce from the above GA data that under reactor conditions, the steady-state fractional release of iodine into the helium coolant is comparable to, and probably lower than, that of xenon (taking half-life into account).

Dragon project investigators (Ref. 4-53) compared Te-129m, I-131, and Xe-133 fractional release (R/B) values from HTGR fuel during irradiation in DIDO, Studsvik, and PLUTO experiments. In most experiments, the iodine release was found to be 30% to 70% of the xenon release. The fractional release of Te-129m was commonly lower than that of iodine.

Thus, under normal reactor operating conditions, the release of iodine isotopes may be delayed relative to the release of xenon isotopes. Sites for retardation cannot be identified from the in-pile data. The kernels of particles are unlikely to be significantly involved in any retardation since, as stated in Section 4.2, the release from fissile material is primarily by recoil under HTGR normal operating conditions. Furthermore,

studies on the relative release of xenon and iodine from monogranular UC_2 kernels in postirradiation anneals (Ref. 4-27) and from polycrystalline UO_2 under irradiation conditions at steady state (Ref. 4-54) are not conclusive.

Results of experiments on the release of xenon and iodine from the contamination of particle coatings (Ref. 4-4) are, within the experimental uncertainty, consistent with similar release behavior for xenon and iodine in particle coatings.

The sorption of iodine on graphite could result in the delay in transit of iodine relative to that of xenon. Iodine is known to be significantly sorbed on graphite at temperatures near $500^\circ C$ (Refs. 4-55, 4-56, 4-57). Unfortunately, the temperature dependence of the sorptivity is quite uncertain, so an estimate of the sorption of iodine on graphite in the core of an HTGR under normal operating conditions is difficult. The temperature of the graphite in the core can vary from about 340° to above $1000^\circ C$. Clearly, some sorption of iodine in the core graphite can occur, but since most of the graphite will be at temperatures of $800^\circ C$ or higher, the sorptivity could generally be quite small.

4.7.2. Conclusions on Release of Halogens and Related Elements

The available evidence suggests that iodine and tellurium isotopes have smaller fractional release values than do xenon isotopes. However, the data are not conclusive and require validation. Therefore, at present, iodine and tellurium isotopes should be conservatively treated as if they were xenon isotopes. Correspondingly, bromine and selenium isotopes should be treated as if they were krypton isotopes.

4.8. EFFECT OF FUEL HYDROLYSIS ON FISSION GAS RELEASE

Hydrolysis of exposed fissile UC_2 kernels will increase fission gas release from the affected particles. The hydrolysis reaction transforms

the normally dense UC_2 kernels to a loose, powdery hydrolysis product according to the reaction

where s denotes a solid product and g a gaseous product.

The solid products are considerably more voluminous than the original dense carbide (up to a factor of 2), thereby causing the particle coating to open. Studies* of the effect of carbide hydrolysis on R/B have been carried out on a number of fuel configurations including:

1. Loose bare UC_2 kernels.
2. Loose laser-drilled coated particles (both irradiated and unirradiated).
3. Specially prepared fuel rods comprising known amounts of unirradiated bare kernels or laser-drilled particles.
4. Irradiated fuel rods having known failure fractions.

Experiments on loose particles (either bare kernels or laser drilled) have shown that hydrolysis transforms the kernel material to a fine powder. The R/B (Kr-85m at 1100°C) of the final hydrolyzed product (in graphite powder) is of the order of 0.5 (Ref. 4-58) or more, indicating virtually total release of fission gases at steady state. This constitutes an increase in R/B by a factor of approximately 10 for loose bare kernels (the R/B of bare intact UC_2 kernels is about 0.05) and a factor of 100 for loose laser-drilled particles (the R/B of non-hydrolyzed laser-drilled UC_2 particles is about 0.005).

*These studies were performed by F. C. Montgomery and D. G. Harmston at GA.

Fission gas release studies on special fuel rods, prepared with either bare UC_2 kernels or laser-drilled particles and exposed to hydrolysis conditions, have shown that the R/B of hydrolyzed particles constrained in fuel rods tends to decrease with continued exposure.

Details and results of these experiments are described below.

4.8.1. Tests on Effect of Fuel Hydrolysis on Fission Gas Release

Several experiments on fuel rods have been completed, including tests on the laboratory-prepared, unirradiated fuel rods (Ref. 4-23), a sample of irradiated Peach Bottom fuel compact material (Ref. 4-23), and an irradiated FTE fuel rod (Ref. 4-59, p. 178).

The tests on the laboratory-prepared rods involved exposure of the rods to water vapor at temperatures from 300° to 900°C. The R/B measurements were performed intermittently during the exposures, which ranged up to 1315 h. The fissionable material in the rods was UC_2 in either laser-drilled TRISO-coated particles or bare uncoated kernels. In the former case, the TRISO coatings were penetrated three times using a laser beam. The three holes were 10 to 20 μm in diameter and were drilled into each particle to a depth of approximately the kernel surface to assure that the UC_2 kernels would be exposed to the hydrolysis conditions. The laser-drilled particles or bare kernels were positioned in the center of green fuel rods prepared from dummy (nonfissionable) pyrolytic-carbon-coated ZrO_2 particles and matrix material. The rods were carbonized at 1000°C and fired to 1800°C in the usual manner. They were then inserted into individual graphite crucibles and measured for Kr-85m release at 1100°C in the TRIGA King furnace facility. The rods were then gamma counted to determine the quantity of U-235 in each rod, which allowed accurate calculation of the birth rate of the noble gas isotopes (i.e., the "B" term in R/B). The test rods, in their graphite crucibles, were placed inside quartz furnace tubes which were continuously swept with helium containing 100 ppm water vapor at 0.1 MPa total pressure (equivalent to 2 ppm H_2O at 5 MPa He).

In two other experiments, the effect of prior irradiation has been examined. In the first test, a small sample of irradiated, fuel compact (D1305-29) material containing essentially 100% failed $(\text{Th,U})\text{C}_2$ particles (burnup = 4% FIMA) from the Peach Bottom HTGR (Core 1) was exposed to 100 ppm water vapor at 900°C for 2012 h. In this test, because of the high radiation level of the previously irradiated sample, the hydrolysis exposure was conducted in the poolside TRIGA King furnace. Thus, R/B measurements on this sample were performed in situ without having to remove the sample from the exposure furnace. Since the exact quantity of U-235 in this specimen was unknown, the Kr-85m R/B values have been normalized to a value of 1.0% for R/B prior to exposure. The value of 1% is consistent with measurements of Peach Bottom Core 1 fuel material, including the end-of-life average R/B measured during the last several weeks of reactor operation.

In the second test, conducted in the poolside TRIGA King furnace facility, a fuel rod obtained from Peach Bottom fuel test element FTE-3 was used. The rod contained UC_2 TRISO-coated particles having experienced approximately 10% FIMA burnup and ThC_2 BISO-coated particles having experienced approximately 1% FIMA burnup. The fuel rod was first annealed in dry helium at 1600°C in order to induce particle failure prior to the hydrolysis exposures. At the end of the 1600°C anneal, a Kr-85m R/B of 2.1×10^{-4} was determined. This value yields a failed fraction of about 0.04 assuming R/B (Kr-85m at 1100°C) for failed fuel in a rod is 5×10^{-3} (see Section 4.1.3.5). This same rod was then exposed to 100 ppm water in the helium purge gas for approximately 240 h at 900°C. The moisture content of the purge gas was then increased to approximately 30,000 ppm H_2O for 24 h, after which the rod was annealed at 1100°C for 150 h in dry helium. R/B measurements were performed several times during each exposure and before and after each change in conditions.

4.8.2. Results and Discussion on Effect of Fuel Hydrolysis on Fission Gas Release

The results of the R/B tests as a function of hydrolysis exposure are given in Table 4-23 for unirradiated fuel rods and Tables 4-24 and 4-25 for irradiated compact material and fuel rods.

TABLE 4-23
R/B^(a) VERSUS HYDROLYSIS TIME FOR UC₂ PARTICLES IN RODS EXPOSED TO 100 PPM H₂O IN HELIUM

Time (h)	300°C		400°C		500°C		700°C		800°C		900°C	
	Laser- Drilled ^(b)	Bare Kernel ^(c)	Laser- Drilled	Bare Kernel	Laser- Drilled	Bare Kernel	Laser- Drilled	Bare Kernel	Laser- Drilled	Bare Kernel	Laser- Drilled	Bare Kernel
0	0.8	1.5	1.5	1.7 ^(d)	0.7	2.2	0.3	2.9	0.2	4.1	0.2	5.6
7											2.4	23.0
10							2.5	24.0	6.7	31.0		
15	1.5	3.2	6.6	16.0	1.5	14.0						
20											1.5	50.0
30							3.1	23.0	9.6	36.0		
50			6.6	21.0	2.4	39.0	2.4	24.0	8.7	43.0		
60	3.2	7.0									11.0	63.0
90								33.0				
100			6.2	25.0	3.6	29.0					11.0	12.0
110	2.7	16.0					2.1		11.0	42.0		
200							6.1	42.0	18.0	41.0	14.0	14.0
300	7.4	39.0	9.9	19.0	6.3	23.0	38.0	35.0	25.0	34.0	5.7	14.0
400							32.0	17.0	27.0	31.0	4.5	26.0
455												
560							25.0		27.0	32.0	8.3	11.0
630	21.0	23.0	12.0	17.0	10.0	22.0						
670								22.0				
780							16.0		30.0	32.0	9.1	21.0
1185								12.0				
1216			6.5 ^(d)	8.6 ^(d)	5.3 ^(d)	9.4 ^(d)						
1226	12.6 ^(d)	7.2 ^(d)										
1292							10.0					
1315									13.0	16.0	4.2	10.0

(a) Kr-85m R/B values (percent) at 1100°C, corrected for steady state (i.e., multiplied by 1.9).

(b) Fuel rods prepared from laser-drilled UC₂ TRISO-coated particles.

(c) Fuel rods prepared from bare kernel UC₂ particles.

(d) Average of two successive measurements.

TABLE 4-24
EFFECT OF HYDROLYSIS ON R/B OF PEACH
BOTTOM COMPACT MATERIAL (a)

Exposure Time ^(b) (h)	R/B ^(c) (%)
0	1.0
10	1.4
20	2.2
30	2.2
70	1.4
300	1.4
520	6.1
730	10.0
940	11.0
1410	22.0
2010	12.0

(a) Core 1 compact material,
burnup = 4% FIMA.

(b) Exposure to 100 ppmv water
vapor at 900°C.

(c) R/B Kr-85m at 1100°C. Data
were normalized to R/B = 1.0% at
time = 0, which is consistent with
the R/B of the Peach Bottom core
at end-of-life.

TABLE 4-25
FISSION GAS RELEASE FROM FAILED HYDROLYZED FTE-3 FUEL ROD 1-7-8

Run No.	Sample Condition	Total Anneal Time (h)	Incremental Anneal Time (h)	H ₂ O Content (ppmv)	Kr-85m R/B at 1100°C
1	As-received	0	0	0	2.1×10^{-5}
2	Annealed at 1600°C	0-354	354	0	1.6×10^{-5} (a)
3	Annealed at 1600°C	354-520	166	0	2.1×10^{-4}
4	Annealed at 900°C	520-760	240	100	3.3×10^{-4} (b)
5	Annealed at 900°C	760-784	24	30,000	1.1×10^{-3}
6	Annealed at 1100°C	784-930	150	0	5.7×10^{-4}

(a) Average of 20 R/B determinations during the anneal.

(b) Average of five R/B determinations.

For the unirradiated rods, the R/B initially increases with increasing exposure to hydrolysis conditions and then slowly declines. The important observation is that on continued exposure for times as long as 1315 h, the R/B of all the fuel samples decreases to approximately the same value ($9.6\% \pm 3.4\%$ based on the last value of R/B listed in Table 4-23 for each sample). The increase in R/B followed by the slow decline is believed to be associated with the formation of a loose, finely divided oxide product, followed by a slow sintering process in which the hydrolyzed kernel material somewhat densifies. (See Section 4.6.1 for evidence of a slow sintering process in non-hydrolyzed fuel.)

The data of Table 4-23 show that hydrolysis of the bare kernel rods caused R/B to increase from the range of 0.015 to 0.056 to the range of 0.25 to 0.63 within 60 to 300 h of exposure. The rods prepared from laser-drilled particles reached their respective R/B maxima in times significantly longer than for the bare kernel rods (200 to 780 h), suggesting that the rate of hydrolysis was partially controlled by the area of the exposed carbide. Also, in general, the average maximum R/B's observed in rods containing laser-drilled particles were one-half of those observed in rods containing bare kernels (average $R/B_{\max} = 0.21$ for rods with laser-drilled particles and 0.42 for rods with bare kernels).

The results of hydrolysis exposures on the irradiated Peach Bottom Core 1 compact material (given in Table 4-24) show that the long-term effect of hydrolysis on the R/B of this fuel sample was virtually identical to the effect on the unirradiated samples. This was an expected result since the burnup of this specimen was only 4% FIMA. It is of interest, however, that this small degree of burnup caused a significant passivation during the early stages of the exposure; that is, the effects of hydrolysis were insignificant until after 400 h of exposure to water vapor. This result suggests that higher burnups could cause an even longer-term passivation effect.

From the results of the hydrolysis tests on the irradiated FTE fuel rod (given in Table 4-25), it is seen that 240 h of exposure to 100 ppmv H_2O at $900^\circ C$ caused the R/B to increase by a factor of 1.6. After 24 h at 30,000

ppmv water vapor, the overall increase in R/B was only a factor of 5.2. The final anneal in dry helium at 1100°C caused the R/B to decrease, as expected. The overall increase in R/B was therefore only a factor of 2.7. Assuming that the R/B of the failed particles in this fuel sample prior to hydrolysis was 5×10^{-3} , then the R/B after hydrolysis would be 1.35×10^{-2} . Nevertheless, the low relative increase in R/B encourages belief in the conjecture that irradiation exposure causes a chemical passivation of the carbide which inhibits oxidation or hydrolysis. It is conjectured that passivation of the irradiated carbide could be due to stable fission product oxides, such as ZrO_2 , which would form protective films over the exposed carbide surface.

4.8.3. Summary and Conclusions on Effect of Fuel Hydrolysis on Fission Gas Release

Hydrolysis tests on unirradiated, exposed carbide particles in fuel rods and slightly irradiated compact material show that R/B initially increases to 20% to 60%, then declines to approximately 10% after about 1200 h of annealing. In one hydrolysis test on an irradiated fuel rod, the apparent R/B of hydrolyzed carbide particles was estimated to be 1.35%.

The recommended R/B for Kr-85m at 1100°C for failed, hydrolyzed carbide particles in fuel rods is $10\% \pm 4\%$. This value is based on data from the unirradiated fuel rods. Use of the R/B for the hydrolyzed irradiated FTE sample should be deferred until the data can be verified by additional experiments.

The large values of R/B encountered during fuel hydrolysis indicate a change in the mechanism of release for the short-lived isotopes as compared with the recoil-type release deduced for the case of unhydrolyzed failed fuel in fuel rods (see Sections 4.1, 4.2, and 4.3). Diffusive release from the

hydrolyzed fuel during the peak release is apparently contributing significantly to release. This can be understood on the basis of a greatly reduced diffusion distance for the hydrolyzed fuel, which is in a powder form; thus, the short-lived isotopes as well as the long-lived and stable isotopes diffusively escape before they decay. Also, as a result of the change in structure of the fuel during hydrolysis, the recoil fraction may be expected to increase. The relative changes in recoil and diffusive release as a function of annealing time are not known. However, with increased annealing time and possibly the onset of a slow sintering process, the fractional release, R/B, decreases.

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