To the Graduate Council,

I am submitting herewithin a thesis written by Daniel A. Smith entitled "Development of an Improved Model for Predicting Fission Product Release from High Temperature Gas Cooled Reactors." I have examined the final copy of this thesis for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Master of Science, with a major in Nuclear Engineering

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We have read this thesis
and recommend its acceptance

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Accepted for the Council

Associate vice Chancellor
and Dean of the Graduate School

# DEVELOPMENT OF AN IMPROVED MODEL FOR PREDICTING FISSION PRODUCT RELEASE FROM HIGH TEMPERATURE GAS COOLED REACTORS.

A Thesis

Presented for the

Master of Science

Degree

The University of Tennessee

Daniel A. Smith

August, 1994

# DEDICATION

This work is dedicated to my wife Cinder Smith and my daughter Kyla Gabriel Smith. Without their love and devotion, this work would not have been possible.

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# ABSTRACT

Gas Turbine-Modular High Temperature Gas Reactors have the capacity to supply low cost electric power and high temperature process heat for industrial applications. The important factor in keeping the cost of the reactor system low is the high degree of inherent safety of the reactor core under normal operating and extreme accident conditions. The reactor fuel has a enormous capacity to retain fission products in the fuel compact. This work has developed an improved fuel performance model for predicting fission product retention under normal operating and extreme accident conditions. While the model has been developed for Pebble Bed Reactor (PBR) fuel, it also provides information pertinent to the performance of U.S. Modular High Temperature Reactor Fuel.

PBR fuel particles consist of a sphere of Uranium Oxide (radius=250$\mu$m) coated by successive layers of pyrocarbon, silicon carbide and pyrocarbon. These particles are embedded in a sphere of matrix graphite. The improved fuel performance model predicts fission product release by treating diffusion of the fission products out of the fuel kernel, through the various coated particle layers and out of the fuel element. It also includes release due to contamination in the matrix graphite, contamination of the outer PyC layer, and possible fracture of coated particle layers. The emphasis here is on the diffusive release of fission products under normal operation and accident conditions. In general, the primary

mechanism for the release of fission products from the fuel is diffusion. Overall, this work has developed a phenomenally sound model for the release of fission products by numerically solving the Fickian Diffusion Equation under several different conditions. Good agreement was obtainted between calculated and measued data from experimental fuel results for PBR fuel consistant with both normal operation and accident contitions.

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# Chapter 1

# Introduction

## 1.1 Background

Modular High Temperature Gas Reactors (MHTGRs) show promise as a reactor system which can provide electrical power and/or high temperature process heat at a relatively low cost. The best system for this purpose is the Gas Turbine Modular High Temperature Gas Reactors (GT-MHTGR) which operates at very high temperatures and thus has a very high thermal efficiency. The low cost is achieved by utilizing the large inherent and passive safety factor of the reactor fuel. Since the reactor fuel retains almost all of the key fission product even under severe accident conditions (in which temperatures can reach approximately 1600°C), the balance of the plant can be constructed to the standards of a conventional fossil fuel power plant. The fuel considered in this work is Pebble Bed Reactor fuel, since a large set of experimental fuel performance data was availabe for many test of high qualite PBR fuel.

A primary reason for the retention of fission products by the fuel particles under normal and accident conditions is the fact that the uranium fuel for the reactor is coated with various layers that retain fission products. The coated particles consist of a kernel of $UO_2$ surrounded by successive layers of a low density buffer layer of carbon, pyrolitic carbon, silicon carbon, and another layer of pyrolitic carbon.

Several thousand of these coated particles are embedded in a sphere of matrix graphite material in German Pebble Bed Fuel. In addition to the coated particle layers, the matrix graphite also slows the release of fission products and helps keep the fission product in the fuel element.

While extensive work has been done on fuel performance modeling in the U.S, Germany and Japan; those models do not properly predict fuel performance under all pertinent conditions. This thesis has resulted in an improved fuel performance model, utilizing the results of the irradiation and heating tests of German Pebble Bed Reactor fuel. These tests provide the most extensive database currently available for high quality HTGR fuel.

This research was based on the "KFA model" for fuel performance developed at the Nuclear Research Center at Julich, Germany (KFA). Modifications and extensions have been performed and the resulting model is called the UT-KFA model. This model provides a technically sound basis for the estimation of fission product release from PBR fuel elements. To maintain the safety factor required, the release of fission products must be accurately predicted during both normal operation and accident conditions. Although developed for PBR fuel, this model gives pertinent information concerning the conditions that must be met by US fuel in order to have high performance.

There are currently two models in use to predict the

release of fission products. The first is "pressure vessel failure" model[8] as represented in the PANAMA-I code; this code includes failure attributed to SiC dissociation in addition to failure of the SiC due to excessive pressure. However, the SiC dissociation portion of the PANAMA-I code does not have a sound phenomenalogical basis. In the work here, diffusion of fission products through the various coated particle layers takes the place of SiC disassociation.

The other model is the diffusion code FRESCO-II developed at KFA. This model has a good basis for the release of fission products, however the parameter values used to determine this release needed considerable improvement to generate accurate results over a wide range of test conditions.

## 1.2 Motivation

The reason for this work is to better understand PBR fuel performance, i.e. the retention of key fission products within the fuel elements under irradiation and accident conditions. The safety of MHTGRs relies primarily on the capacity of the fuel element to retain key fission products. This capacity to retain a very high fraction of the key fission products is termed high fuel performance. With improved comprehension of the mechanics of the release of fission products from the fuel elements, safety analysis of High Temperature Gas Cooled Reactors can be improved.

## 1.3 Original Work

The original work of this thesis is the development of a new/revised model for fission product release from PBR fuel elements. This model enables good prediction of key fission product release during normal operating and accident conditions. By having a better release model, the safety of the reactors can be more accurately evaluated.

# Chapter 2

# Theory

## 2.1 Introduction

The release of fission products is dominated by diffusion out of the coated particles and through the matrix graphite. However there are other mechanisms that must be considered in calculating the release of fission products. These factors are: particle breakage, recoil release of the fission product, natural uranium contamination in the matrix graphite, fuel contamination in the outer PyC layer and silicon carbide corrosion.

## 2.2 Diffusion

The primary mechanism considered by the UT-KFA model for the release of fission products is diffusion. The diffusion process is controlled by the Fickian Diffusion Equation which is shown in Equation 2.1[4].

$$\frac{\partial c}{\partial t} = D(T)\left(\frac{\partial^2 c}{\partial r^2} + \frac{2}{r}\frac{\partial c}{\partial r}\right) - \lambda c + Q \qquad 2.1$$

where c is the concentration of fission products
D(T) is a temperature dependant diffusion coefficient
r is the distance from the center of the sphere
λ is the decay constant for the fission product
Q is the source term of the fission product

The diffusion coefficient is based on the formula in Equation 2.2[4].

$$D=D_0 e^{(-\frac{A}{RT})} \qquad 2.2$$

where $D_0$ is the diffusion coefficient
A is the activation energy
R is the gas constant
T is the temperature

In addition, the SiC and possibly the PyC may be degraded by the fast neutron fluence (neutrons with energy greater than 0.1 Mev) which increases the diffusion coefficient by an exponential decay factor of[3]

$$e^{\frac{\Gamma}{K}} \qquad 2.3$$

where $\Gamma$ is the fast neutron fluence and
K is a empirically determined constant which varies depending on the fission product and the layer.

In this case, the factor is multiplied by the diffusion coefficient from equation 2.2, so the revised formula for the diffusion coefficient becomes[3]

$$D=D_0 e^{(-\frac{A}{RT})} e^{(\frac{\Gamma}{K})} \qquad 2.4$$

Finally, at higher temperatures, the diffusion coefficient may increase drastically. This is modeled by adding a second term to the diffusion coefficient, so the formula for the diffusion coefficent becomes[3]

$$D=D_{0,1}e^{(-\frac{A_1}{RT})}e^{(\frac{\Gamma}{K})}+D_{0,2}+e^{(-\frac{A_2}{RT})} \qquad 2.5$$

where $D_{0,1}$ $A_1$ and $D_{0,2}$ $A_2$ are two distinct sets of parameters. In general, the value of $A_2$ is sufficiently high so that the second term can be neglected at temperatures below 1450°C.

## 2.3 Crystal Contamination

The dominant source of krypton release at low temperatures is the natural uranium contamination in the graphite matrix. The matrix material consists of graphite crystals or "grains" and a binder material or "pore graphite". A coated particle with an intact SiC or PyC layer will effectively retain all the krypton generated in that particle during irradiation, so any krypton release measured must be from the contamination in the matrix graphite or contamination in the outer PyC layer of the fuel particles.

During irradiation at normal operation temperatures, the release of krypton from the graphite crystals in the matrix graphite is primarily dominated by recoil. The recoil distance of fission products can be approximated as a 4 Mev alpha particle[5]. This distance is given by[6]

$$R=1.2402\frac{A^{\frac{1}{3}}}{\rho} \qquad 2.6$$

where R = the recoil distance,
A = the atomic number of the medium and
$\rho$ = the density of the medium.

To calculate the amount released, first consider what happens when a uranium atom is fissioned in a sphere of radius $R_o$ (source sphere). An atom of krypton is generated that will travel a distance R. This distance may or may not be sufficient to for the atom to leave the source sphere. Since the fission product can recoil in any direction, the total area in which it could be located is given by the surface of a sphere of radius R (destination sphere) around the point where the fission occurs. Figure 2.1 shows a sketch of the source sphere and destination sphere. The probability that this atom can escape from the source sphere is the surface area of the destination sphere which is outside the source sphere divided by the total surface area of the destination sphere. That probability is defined to be[4]

$$P_{release}=\frac{4\pi Rh}{2\pi R^2} \qquad 2.7$$

where R = the recoil distance of the fission product
$R_o$= the radius of the sphere.

$$h=R-\frac{R_o^2-R^2-r^2}{2R} \qquad 2.8$$

r = the distance from the point of fission to the center of the sphere.

Integrating this probability times the fission product generation rate divided by the total source in the sphere gives the fractional release from the sphere due to recoil as

$$F.R. = \left(\frac{R}{R_o} - \frac{1}{12}\frac{R^3}{R_o^3}\right) \qquad 2.9$$

where F.R.= the fractional release
$R_o$=the radius of the graphite crystal

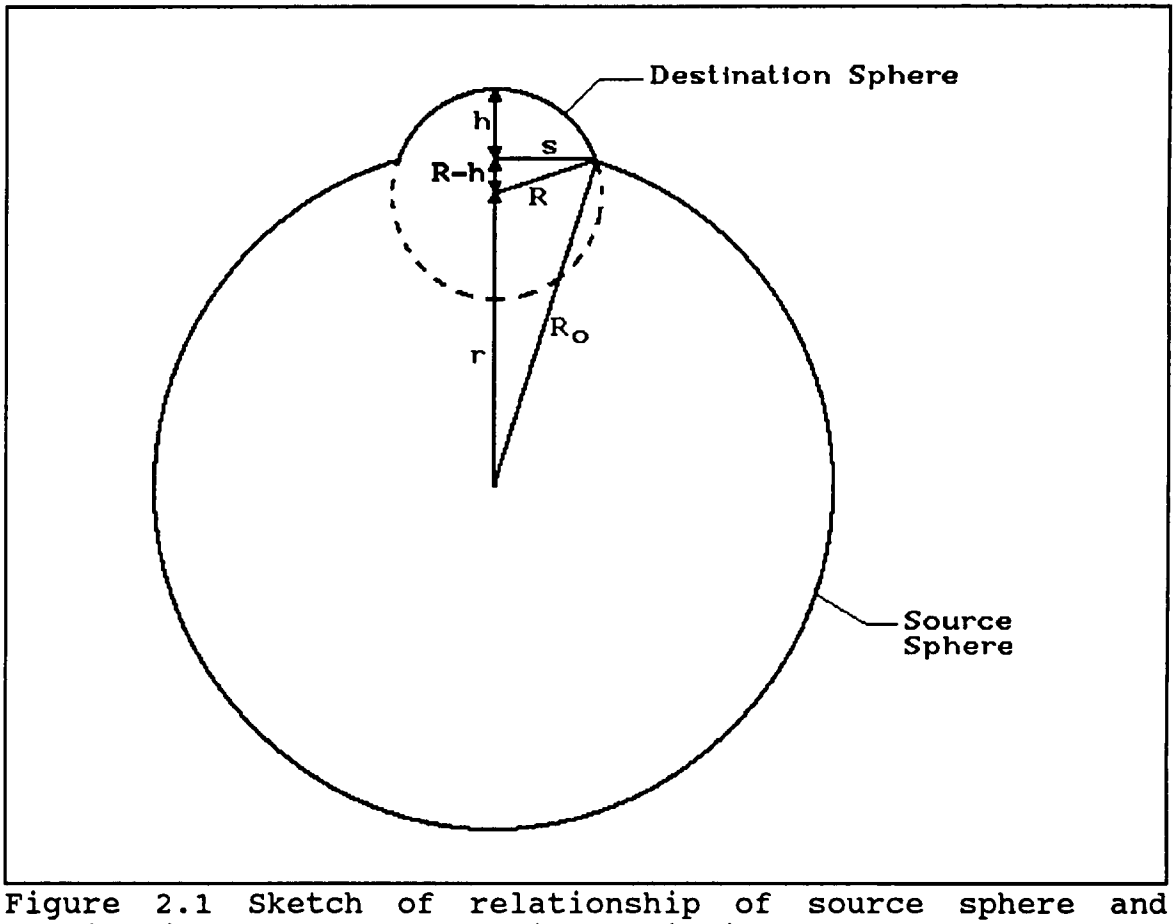



Figure 2.1 Sketch of relationship of source sphere and destination sphere for recoil of fission products.

## 2.4 Corrosion of SiC

The SiC layer can be corroded by attack from the fission product and by thermal decomposition. This corrosion can result in the thinning of the SiC layer. In the UT-KFA code,

the corrosion rate is modeled empirically by using the formula[7]

$$\frac{\partial L}{\partial t} = e^{-6.321-\frac{9.3x10^3}{T}} * 100 \qquad 2.10$$

where L is the thickness of the SiC layer in microns
T is the temperature °K
t is the time (in seconds)

This reduction in SiC thickness is only a small contribution to the diffusional release of the fission products for the cases studied.

## 2.5 Diffusion Trapping Model

In the trapping model, the fission products diffuse through the medium and are trapped in infinitesimally small sites distributed throughout the medium.

The equations defining this model is[4]

$$\frac{\partial C}{\partial t} = D(T)\left(\frac{\partial^2 C_1}{\partial r^2} + \frac{2}{r}\frac{\partial C_1}{\partial r}\right) - \lambda C_1 + \dot{Q}_1 - \mu C_1 + bm \qquad 2.11$$

$$\frac{\partial m}{\partial t} = \mu C_1 + bm - \lambda m \qquad 2.12$$

$$C_{total} = C_1 + m \qquad 2.13$$

where
$C_1$= concentration in the diffusion medium

m = concentration in traps
r = radius
D(T)= Diffusion Coefficient
$\mu$ = transfer rate constant for the exchange from the diffusion medium to the traps.
b = transfer rate constant for the exchange from the traps to the diffusion medium.

The total number of experimental constants for the above system of equations is 3 (D, mu, b). Values for cesium have been experimentally determined and show that above 1300°C, an equilibrium is reached between the traps and the diffusing medium, with simple diffusion holding above 1300°C. All the fuel heating tests were above 1300°C.

Even though the simple diffusion equation applies at fuel heating test temperatures, it is not a simple matter to determine the proper diffusion coefficients because of the number of parameters that apply in determining the diffusion coefficients.

At irradiation temperatures (< 1300°C), the "trapping" diffusion model would be preferred, however, the difference in results between the diffusion trapping model and the simple diffusion model would be small compared with the uncertainty in diffusion coefficients associated with the various diffusing media (kernel, different coating layers, matrix graphite). Further, the diffusion coefficients given for the various materials[3] are inferred values based on interpting experimental results using simple diffusion theory analysis. As a result, simple diffusion theory was used for the "irradiation" and "heating" conditions. It

should also be noted that the "reference" diffusion coefficients as given in Reference 3 are be normalized values based on interpting a number of experimental results. In any given fuel, the "correct" diffusion coefficients may be different than the reference value.

# Chapter 3

# Prior Work

## 3.1 Prior Work

A computer code called FRESCO-II was used to model the release of the fission products. This code was written in Germany and is currently in use there.[3] The FRESCO-II code models the release by numerically solving the Fickian diffusion equation using the tri-diagonal matrix method. The code first calculates the release of fission products from the coated particle by diffusion and recoil, then it uses diffusion to find the release from the fuel element itself. In addition, the program takes into account uranium contamination in the matrix graphite and sorption effects for metallic fission products in the matrix graphite. The program is used to model fission product release for four different nuclides of interest: cesium, strontium, krypton and silver. The model also assumes a coated-particle-layer failure fraction due to the fabrication process, attack of the SiC layer from the fission products and a pressure vessel failure fraction due to excessive pressure within the coated particle.

The FRESCO-II code, as received, severely overestimates the release of the various fission products from the fuel elements as shown in Figure 3.1. Through out the report, the term fractional release referers the the total amount of a

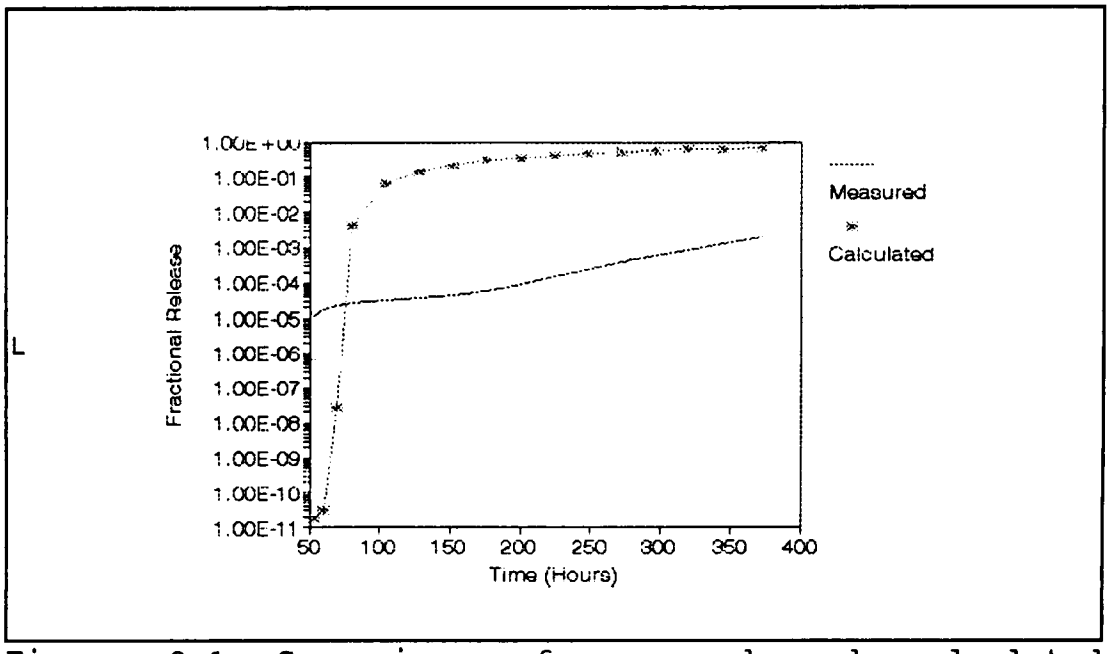



Figure 3.1 Comparison of measured and calculated fractional release of cesium for heating experiment HFR-P4/1/8 heated at 1600°C using the FRESCO-II code as received.

fission product released from the fuel element divided by the total amount of the fission product generated in the fuel element. The high release from the FRESCO-II code is due, in part, to the fact that the code uses diffusion coefficients for earlier reactor fuel designs. In 1980, the graphite used to manufacture fuel elements was changed from the A3-3 matrix graphite to A3-27 matrix graphite. The diffusion coefficients used in the original FRESCO-II code were for the A3-3 matrix graphite which has much higher diffusion coefficients than A3-27 matrix graphite. More importantly, the "reference" diffusion coefficients for fuel coatings did not give a good fit to all the results from PBR fuel tests. Modified diffusion coefficients were developed in this work.

Several attempts have been made in the past to model the fission product release for various heating tests. In each of these experiments, a separate set of diffusion coefficients has been used to explain the observed releases. However, little success has been attained in the direction of combining these results into a comprehensive model with a consistent basis for the calculation of the release. The German FRESCO-II model assumes that when a particle fails, all the layers of the coated particle fail. This instantly releases the fission product in the failed coated particle layers and any additional fission products which diffuse out of the kernel are assumed to go directly into the matrix graphite.

The above assumption resulted in a consistent overestimation of the release of fission products unless one of two options were used: either the diffusion coefficients were altered or the failed fraction of the fuel particles were changed to match the release. To correct the calculated release, it was often necessary to have a fraction of a particle fail, which is an unrealistic case. This approach gives good results for the normalizing case, but not for the overall test matrix. To correct this problem, a new model has been developed. This model assumes that when a particle "fails", the coated particle layers may still remain to partially retard the release of fission products; for example, it permits treating of "cracked" SiC layers within

an intact outer PyC layer. The diffusion coefficients developed for this model are consistent with all experiments examined and allows for a reasonable number of "failed" coatings. It enables the code to predict fission product release over a wide range of test conditions with good accuracy. An example of the prediction capability is shown in Figure 3.2. The data in this figure was calculated without changing any values such as diffusion coefficents except for the physical parameters of the experiment and the number of broken particles. The error bars on the measured values represent uncertainty in the actual measured values of the krypton[9], but do not represent uncertainties in parametric values (such as diffusion coefficients) in a specific fuel test. Over all, agreement within a factor of five over the whole spectrum of experiments is considered very good.

### 3.2 Program Modifications

As first received, the FRESCO-II code used estimations for the diffusion coefficients for earlier reactor designs. The latest estimation of diffusion coefficients were entered into the code[2,3]. The diffusion coefficients used are shown in Table 3.1. When the values in Table 3.1 were used without any particle coating breakage, the FRESCO-II code constantly underestimated

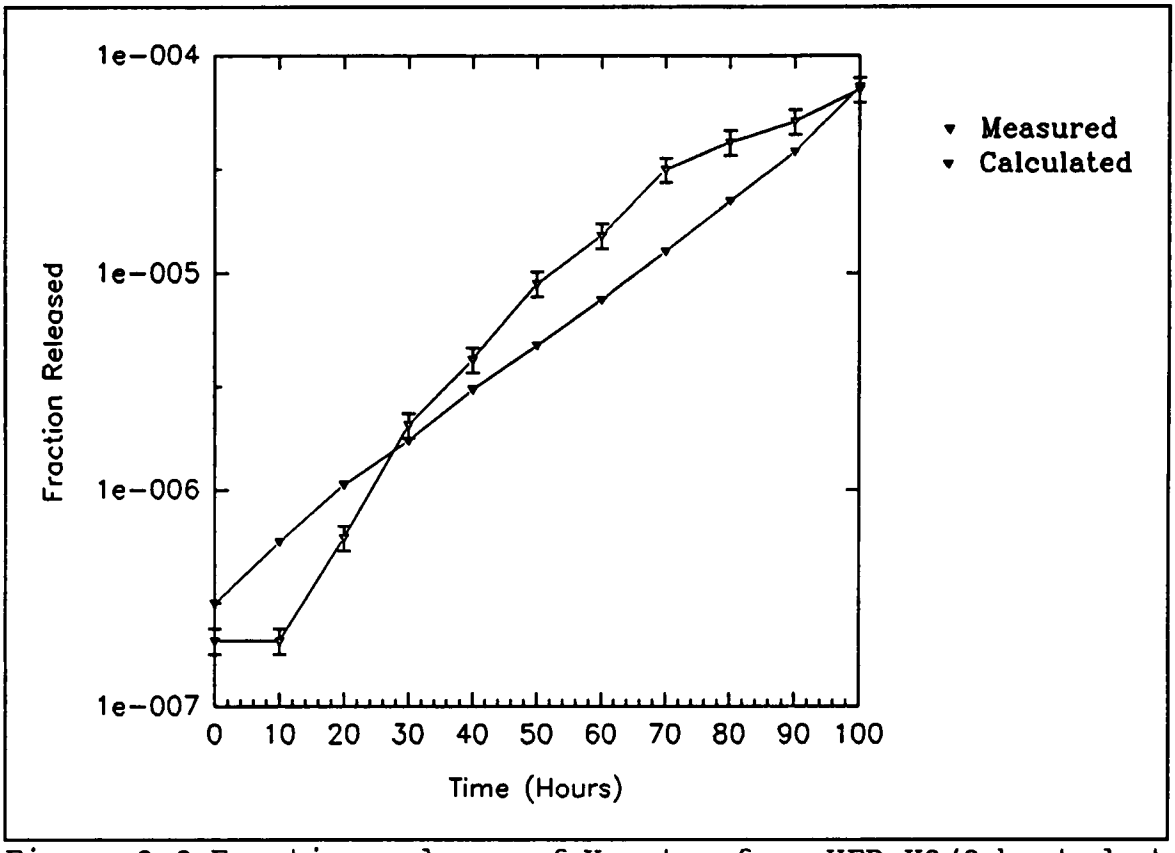



Figure 3.2 Fraction release of Krypton from HFR-K3/3 heated at 1800°C with a fast fluence of $5.9 \times 10^{25}$ n/m$^2$ (E >0.1 MeV)

Table 3.1 Diffusion Coefficients from References 2 and 3.

| Kernel | $D_{01}$ $cm^2/sec$ | $A_1$ J/mol | $D_{02}$ $cm^2/sec$ | $A_2$ J/mol | K |
|---|---|---|---|---|---|
| Cesium | $5.6x10^{-4}$ | $2.09x10^5$ | 5.2 | $3.62x10^5$ | |
| Strontium | $2.2x10^1$ | $4.88x10^5$ | | | |
| Krypton | $1.3x10^{-8}$ | $1.26x10^5$ | | | |
| PyC | | | | | |
| Cesium | $6.3x10^{-4}$ | $2.22x10^5$ | | | |
| Strontium | $2.3x10^{-2}$ | $1.97x10^5$ | | | |
| Krypton | $2.9x10^{-4}$ | $2.91x10^5$ | $2.0x10^9$ | $9.23x10^5$ | |
| SiC | | | | | |
| Cesium | $5.5x10^{-10}$ | $1.25x10^5$ | $1.6x10^2$ | $5.14x10^5$ | 5.5 |
| Strontium | $2x10^{-5}$ | $2.05x10^5$ | $1.8x10^{10}$ | $7.91x10^5$ | |
| Krypton | | | | | |
| Matrix Graphite | | | | | |
| Cesium | $3.6x10^{-1}$ | $1.89x10^5$ | | | |
| Strontium | $1.0x10^2$ | $3.03x10^5$ | | | |
| Krypton | $6x10^{-2}$ | | | | |
| Graphite Grains | | | | | |
| Krypton | $1x10^{-18}$ | $7.1x10^4$ | | | |

the fractional release of fission products from 50% to several orders of magnitude. In general, the recommended procedure to correct the release[3] is to either change the diffusion coefficients or to alter the number of broken particles and the time when the particle breakage occurred. The number of "broken particles" was varied until the calculated release matched the measured release. Unfortunately, the number of broken particles required to match the measured release was less than one (but greater than zero) in most cases. In reality, the number of broken particles is an integral number since a particle either is or is not broken. This assumes the definition of "broken" as having a cracked or otherwise failed coated particle layer.

To obtain better agreement between the calculated and measured values, it was assumed that when a particle coating fails, the layers of the particle do not completely "disappear", but instead remain in place with a higher diffusion coefficient. These higher values allow for an increase in the release of fission products. The diffusion coefficient for a defective layer can be estimated by assuming one particle has failed and changing the value until the calculated release matches the measured release. Initially, only one particle coating failure was assumed since the PANAMA-II code (which estimates "pressure vessel failure" of coatings) indicated that in the operating range of the experiments examined, no particles could be expected

to fail. Also, the "burn-leach" tests performed on fresh fuel indicate that only one or two particles had cracked SiC coatings due to manufacturing. It is important to remember that, if there is more than one "broken" particle, the diffusion coefficients of the defective particle layers can be reduced to find a value which gives good agreement with the measured release. It should be noted that the diffusion coefficients for "intact" layers used in this work were essentially the values found in Table 3.1 .

With the above modification and with an estimation of the number of broken particles, the release of fission products from fuel elements can be calculated to be in agreement with the measurements. In this process, the "failed" or cracked layers have a relatively large diffusion coefficient to allow for the escape of the key fission products.

# Chapter 4

# Experiment and Data

## 4.1 Experiment

The PBR fuel elements are manufactured such that the coated fuel particles are interspersed in a graphite matrix material and the fueled zone is surrounded by a spherical zone containing only matrix graphite. In reference fuel elements, the diameter of the fueled zone is ~5cm and the diameter of the entire fuel element is ~ 6cm. In some fuel tests, the fueled zone is smaller and is surrounded by a cylinder of matrix graphite. The size of this cylinder varies with the fuel test and is termed a "fuel compact". After fabrication, the fuel was irradiated in one of several different test reactors. The parameters studied include: irradiation time, irradiation temperatures as a function of time and different fast fluences.

After irradiation, some of the fuel from the various tests was heated under accident temperature conditions in the furnace as shown in Figure 4.1. The furnace is swept by a constant flow of helium which removes the volatile fission products. The fuel element is heated in four steps. First, it is heated to 300°C to purge the fuel element/compact and the helium of any water vapor. It is then heated to 1050°C and then to 1250°C for short periods to provide equilibrium conditions of the fuel elements and fission products.

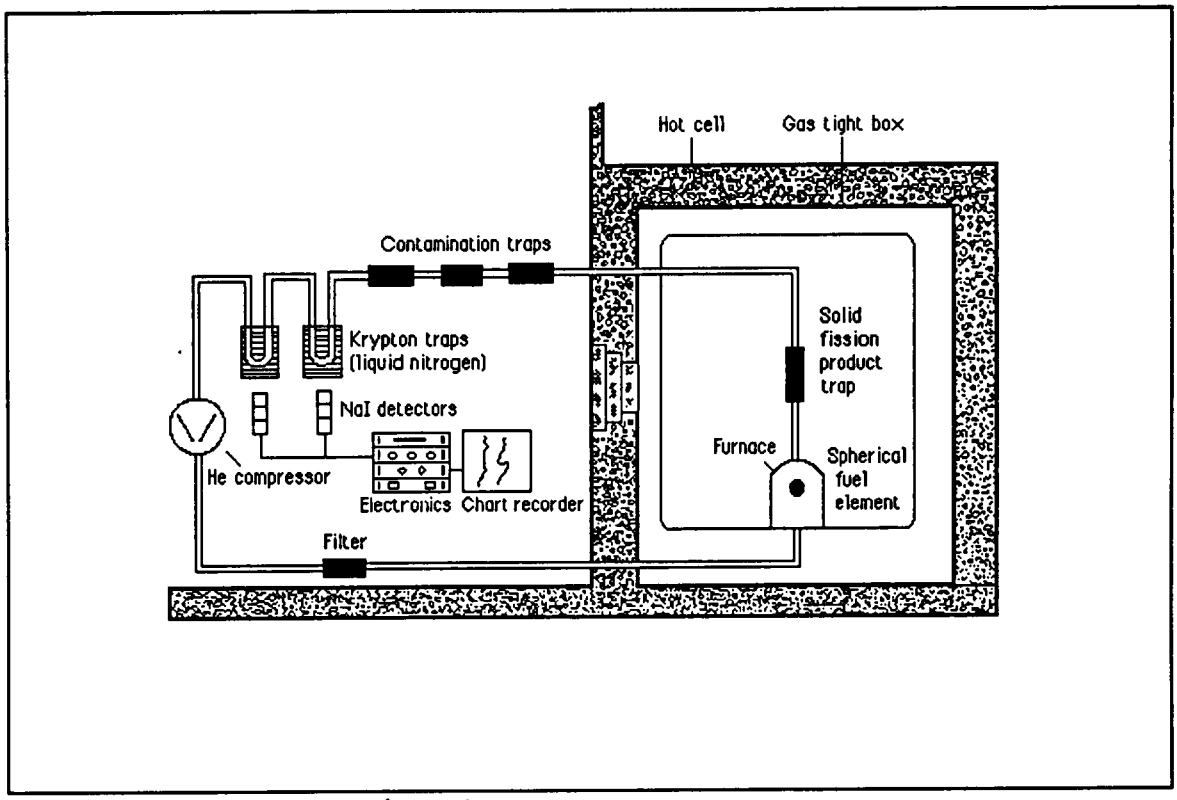



Figure 4.1 The heating furnace

Finally, the fuel is heated to the target temperature (1600°C of higher) at rate of 47°C/hour. A typical heat up graph is shown in Figure 4.2.

The helium carrier gas removes fission products as they diffuse out of the fuel element. The volatile cesium and strontium are condensed on a water cooled "cold finger" located above the fuel, over which the helium carrier gas passes. This causes the metals to plate out of the system. The krypton is trapped by passing the carrier gas through a liquid nitrogen cooled charcoal filter. Calibrated fission product counters are used to determine the release of fission products from the fuel.[9]

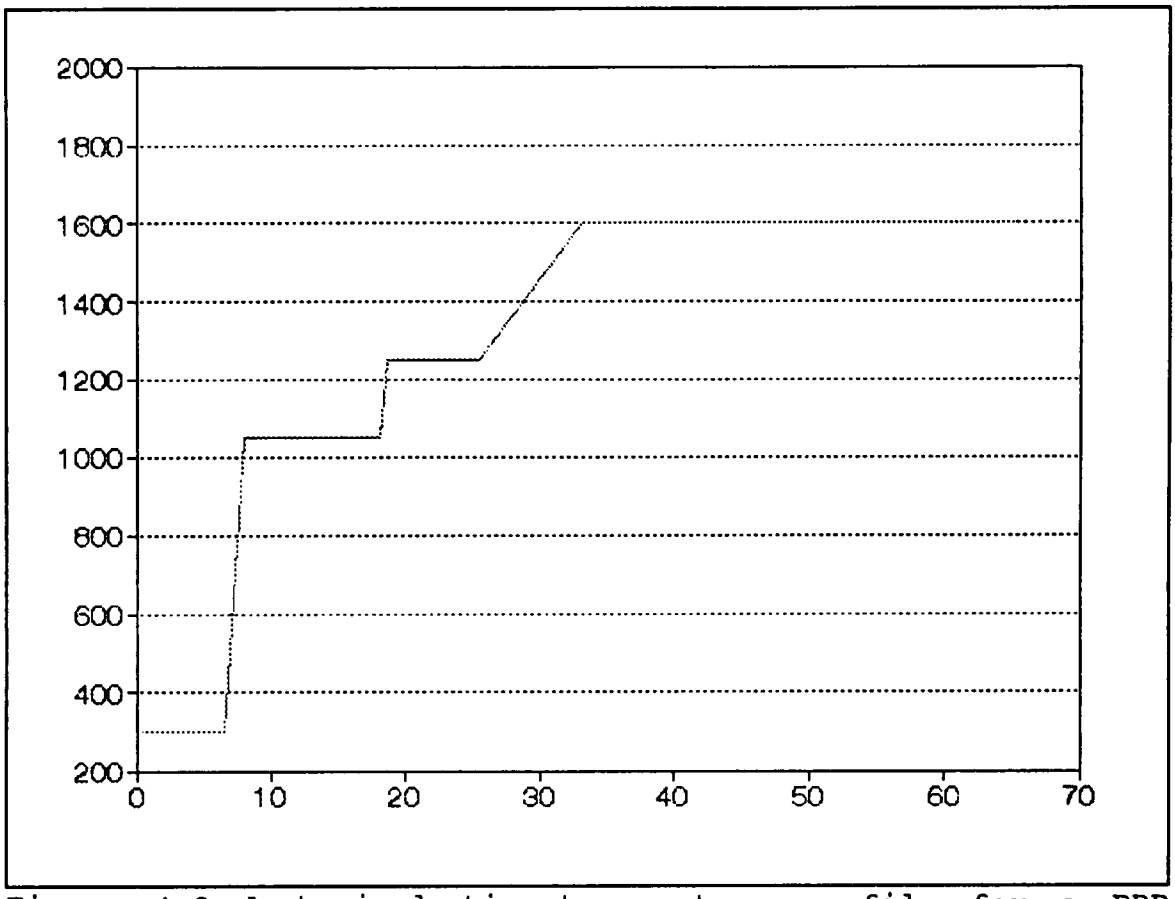


Figure 4.2 A typical time-temperature profile for a PBR heating experiment reaching 1600°C.

## 4.2 Data

The data used in this work covers a total of 16 heating tests. This was supplied from Germany in the form of numerical and graphical data. An example of the numerical data is shown in Table 4.1. For all the experiments, Table 4.2 shows the irradiation and general heating conditions for each of the 16 tests for which data is available. The total statistical and systematic error for the cesium is ±13 for the krypton, ±15% while the error for the strontium measurements is ±50%.[9]

Table 4.1 Data for Fractional Release of Cesium and Strontium from HFR-P4/1/8.

| Time | Measured Cesium Release | Cesium Error | Measured Strontium Release | Strontium Error |
|---|---|---|---|---|
| 60 | 0.000017 | 0.000002 | 0.000002 | 9.55e-07 |
| 69 | 0.000022 | 0.000003 | 0.000017 | 0.000008 |
| 80 | 0.000027 | 0.000004 | 0.000025 | 0.000012 |
| 104 | 0.000033 | 0.000004 | 0.00003 | 0.000015 |
| 128 | 0.000038 | 0.000005 | 0.000038 | 0.000019 |
| 152 | 0.000045 | 0.000006 | 0.000038 | 0.000019 |
| 176 | 0.000058 | 0.000008 | 0.000044 | 0.000022 |
| 200 | 0.000089 | 0.000012 | 0.000046 | 0.000023 |
| 224 | 0.000146 | 0.000019 | 0.00005 | 0.000025 |
| 248 | 0.000234 | 0.00003 | 0.000055 | 0.000027 |
| 272 | 0.000366 | 0.000048 | 0.000061 | 0.00003 |
| 296 | 0.00056 | 0.000073 | 0.00007 | 0.000035 |
| 320 | 0.000842 | 0.000109 | 0.000085 | 0.000043 |
| 344 | 0.00127 | 0.000165 | 0.000107 | 0.000054 |
| 372 | 0.00194 | 0.000252 | 0.000152 | 0.000076 |

Table 4.2 List of all heating experiments examined in this work.

| Experiment | Burnup % Fima | Fast Fluence[1] | Irr. Temp. °C | Irr. Time FPD | Heat. Temp. °C | Heat. Time hours |
|---|---|---|---|---|---|---|
| SL-P1/6 | 10.7 | 6.7 | 790 | 303 | 1600 | 304 |
| SL-P1/9 | 10.3 | 6.3 | 790 | 303 | 1700 | 304 |
| SL-P1/10 | 10.3 | 6.0 | 790 | 303 | 1700 | 304 |
| HFR-P4/1/12 | 11.1 | 5.5 | 900 | 330 | 1600 | 304 |
| HFR-P4/1/8 | 13.8 | 7.2 | 900 | 330 | 1600 | 304 |
| HFR-P4/2/8 | 13.8 | 7.2 | 900 | 330 | 1600 | 304 |
| HFR-P4/3/12 | 9.9 | 5.5 | 1050 | 330 | 1800 | 304 |
| AVR 90/20 | 9.8 | 2.9 | | 1723 | 1620 | 10 |
| AVR 91/31 | 9.0 | 2.6 | | 1500 | 1700 | 10 |
| HFR-K3/1 | 7.7 | 3.5 | 1050 | 359 | 1600 | 500 |
| HFR-K3/3 | 10.2 | 6.0 | 750 | 359 | 1800 | 100 |
| FRJ-K13/2 | 8.0 | 0.1 | 1000 | 396 | 1600 | 138 (168) |
| FRJ-K13/4 | 7.6 | 0.1 | 1000 | 396 | 1600/ 1800 | 138/ 100 |
| AVR 82-20 | 8.6 | 2.4 | 800 | 1405 | 1600 | 100 |
| AVR 76-18 | 7.1 | 1.9 | 800 | 1067 | 1800 | 200 |
| AVR 74-11 | 6.2 | 1.6 | 800 | 891 | 1700 | 184.5 |

[1] The units of fast fluence are $x10^{25}$ $n/m^2$ (E > 0.1 MeV)

### 4.3 Particle Breakage

The layers of a coated particle are susceptible to many kinds of attack. As $UO_2$ fissions, it releases oxygen and generates fission product gases which can build up pressure inside the coating of the fuel particle. This pressure may cause the SiC layer to break, which is generally the limiting material concerning pressure vessel failure. However since there is a distribution of SiC strength in a batch of coated fuel particles, pressure vessel failure of SiC will not always result in failure of the outer PyC coating layer.

The fission products generated in the fuel can attack the SiC and cause thinning of the layer. This thinning reduces the strength of the SiC and may lead to a pressure vessel failure. The effective "thinning process" includes uniform corrosion and "grain-boundary" attack of the SiC. In addition to the thinning process, the fission product attack also causes cracking and pitting to appear in the SiC layer. These defects offer a preferential pathway for the fission product to escape the coated particle.

There is also a neutron degradation effect on both the material strength and the diffusion coefficient. When a neutron with a high energy ($E > 0.1$ MeV) collides with a carbon or silicon atom, it may remove the atom from the normal lattice. This can degrade the material and reduce its

strength, increasing pressure vessel failure, and creating more pathways for fission product escape.

Finally there is a chance that a coated particle has a defect from manufacturing. This may result in fuel particles with a "thin" buffer layer, or a defective SiC or PyC layer. These defects would allow for the faster escape of fission products. Also, a bare fuel kernel might occur during the later stages of the coating process with the result that the fuel can be crushed and distributed onto surface of the the other intact coated particles. This would result in some fuel contamination of the outer PyC layer. While an intact "bare kernel" would be removed from the fuel during the sizing and checking process during fuel fabrication, fuel contamination of the outer PyC layer would not be detectyed prior or fuel irradiation. product release.

In a larger view, there are at least four possible modes of particle failure. The first is a complete failure of the SiC and PyC layers due to pressure vessel failure. This is characterized by a sudden and immediate increase in the krypton release as all the krypton in the coated particle layers is released into the matrix graphite where it rapidly diffuses out of the fuel element. Cesium and strontium releases are also significantly increased.

The second kind of "failure" is a "crack" in the SiC layer as applied to the krypton release. In this mode of failure, the SiC layer fails in such a way as to allow the

rapid diffusion of krypton, but still slows the diffusion of cesium and strontium. The outer PyC layer remains intact and so slows the release of krypton from the coated particle, but still allows for the release of cesium and strontium.

The third kind of particle breakage is similar to the second, but only applies to the cesium and strontium. This occurs when the SiC "cracks" enough to allow cesium and strontium to transport readily through the SiC layer but the krypton is still retained within the SiC layer.

In both the second an third kind of particle breakage, the diffusion parameters of ceisum and strontium within the SiC layer are the same. The difference between these two lie in how the krypton diffuses through the SiC layer. In the second kind of breakage, the krypton rapidly diffues through the SiC. In the third kind of breakage, the krypton is still retained by the SiC.

Another type of particle breakage is "cracking" or failure of the outer PyC layer. This type of failure primarily influences krypton release if the SiC "cracks" at a later time. Table 4.3 shows the diffusion cofficents for the inner PyC , SiC and outer PyC layers for the various types of particle failure

A primary indication for coating failure is the fractional release of cesium measured at the end of fuel irradiation or during the initial period of the heat up experiments. In every case, the fractional release of cesium

Table 4.3 Diffusion coefficents for the layers of the 4 kinds of defective particles.

| | $D_{01}$ $cm^2/s$ | $A_1$ j/mol | K |
|---|---|---|---|
| Type 1 | | | |
| Cs in SiC | $3x10^{-2}$ | | |
| Sr in SiC | $3x10^{-2}$ | | |
| Kr In SiC | $3x10^{-2}$ | | |
| Type 2 | | | |
| Cs in SiC | $6x10^{-10}$ | $4.69x10^4$ | |
| Sr in SiC | $6x10^{-3}$ | $4.69x10^4$ | |
| Kr in SiC | $3x10^{-2}$ | | |
| Type 3 | | | |
| Cs in SiC | $6x10^{-10}$ | $4.69x10^4$ | |
| Sr in SiC | $6x10^{-3}$ | $4.69X10^4$ | |
| Kr in SiC | $2.66X10^{-8}$ | $2.66X10^5$ | 1 |

is much greater than can be accounted for by diffusion from intact coated particles using reference estimates for the diffusion coefficients. An attempt was made to increase the diffusion coefficient to match the release without any broken particles. This resulted in a drastic overestimation of the release during the later part of the heating experiment. It may be possible that the diffusion coefficients are decreasing with time at a high temperature due to annealing, however, there is no indication of this.

The "broken particle" theory is further supported by the burn leach test of irradiated fuel elements. In this

test, the fuel elements are oxidized in air at 800 to 1000°C for an extended period of time. This removes the matrix graphite and the outer PyC layers. Then an acid is used to leach out uranium exposed by the defective SiC. In almost every instance, the amount of uranium found is significantly greater than can be accounted for by natural-uranium contamination in the graphite matrix.

# Chapter 5

# Results

## 5.1 Introduction

Since the UT-KFA code can handle only one type of cracking at a time for a given coating layer, several tests that exhibited more than one type of cracking could not be analyzed at this time.

The cesium release is governed by two different sources. During heat up and for the first part of the heating experiment, the release is due to defective coated particles. After about 50-100 hours of exposing a particle to high temperatures (at or above 1600°C), diffusion out of intact coated particles then contributes significantly to the cesium release. Figure 5.1 shows the release from intact coated particles only and Figure 5.2 shows the release with one defective SiC coating in a batch of 1630 particles.

## 5.2 Cesium Results

The primary barrier to cesium release is the SiC layer. Depending on the fast fluence, the diffusion coefficient of cesium in PyC may be up to 30 times the diffusion coefficient for cesium in SiC.

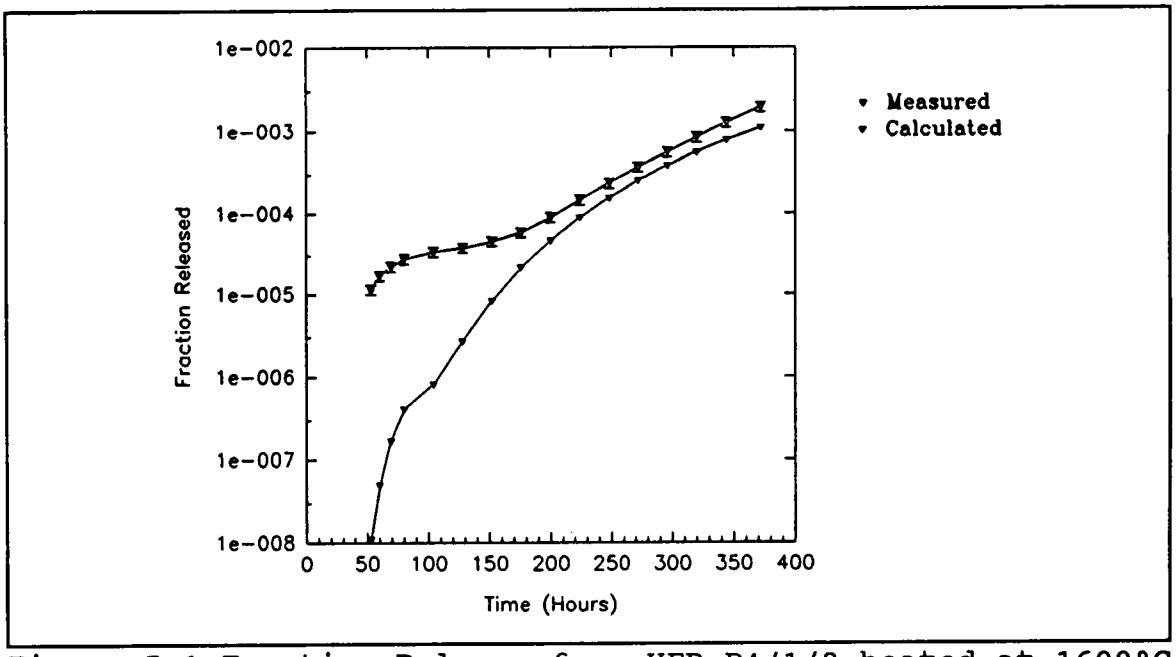



Figure 5.1 Fraction Release from HFR-P4/1/8 heated at 1600°C with no particle breakage.

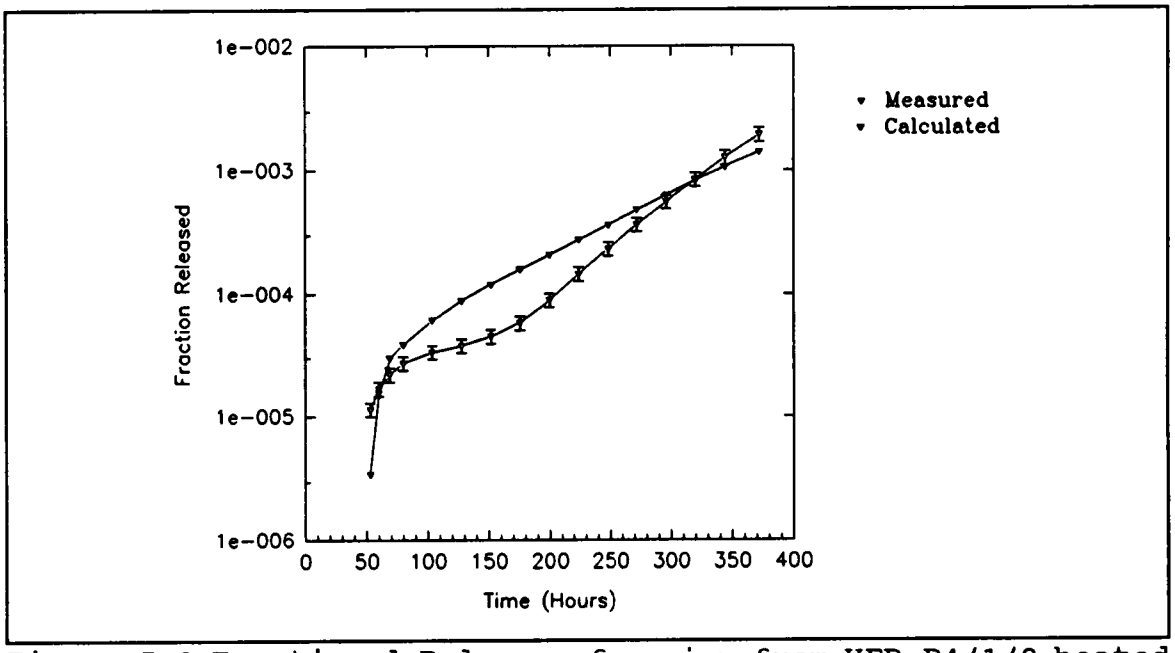



Figure 5.2 Fractional Release of cesium from HFR-P4/1/8 heated at 1600°C with one broken particle.

The release of cesium during the heat up period prior to the actual heating test was modeled to account for the release at the "start" of steady high temperature heating. The first thing that is notable is the fact the intact coated particles do not release enough cesium to account for the release from the fuel element. In HFR-P4/1/12 during the heatup period, the measured release from the fuel element was around $1.5 \times 10^{-5}$ while the intact coated particles had a calculated fractional release of around $8 \times 10^{-8}$. This indicated that the early release was due to defective particles instead of intact particles. However, the release at the later part of heating is accounted for by diffusion through the intact coated particle layers.

To model the early release, it was assumed that the SiC layer in one particle is either defective from manufacturing or failed during irradiation and that this particle could account for the release. Initial calculations have shown that about 40% of the cesium released from the coated particles, would be released from the fuel element at the start of heating. Using this information, a diffusion coefficient for defective SiC was found. The release was then calculated and compared to the measured release. The diffusion coefficents were then corrected and the release calculated again. Using this itterative process, the diffusion coefficent was found to be

$$D=D_o*e^{(-\frac{A}{RT})} \qquad 5.1$$

where $D_o = 6x10^{-10}$ cm$^2$/sec and
$A = 46900$ j/mol

The diffusion coefficient for the defective SiC layer is shown in Figure 5.3.

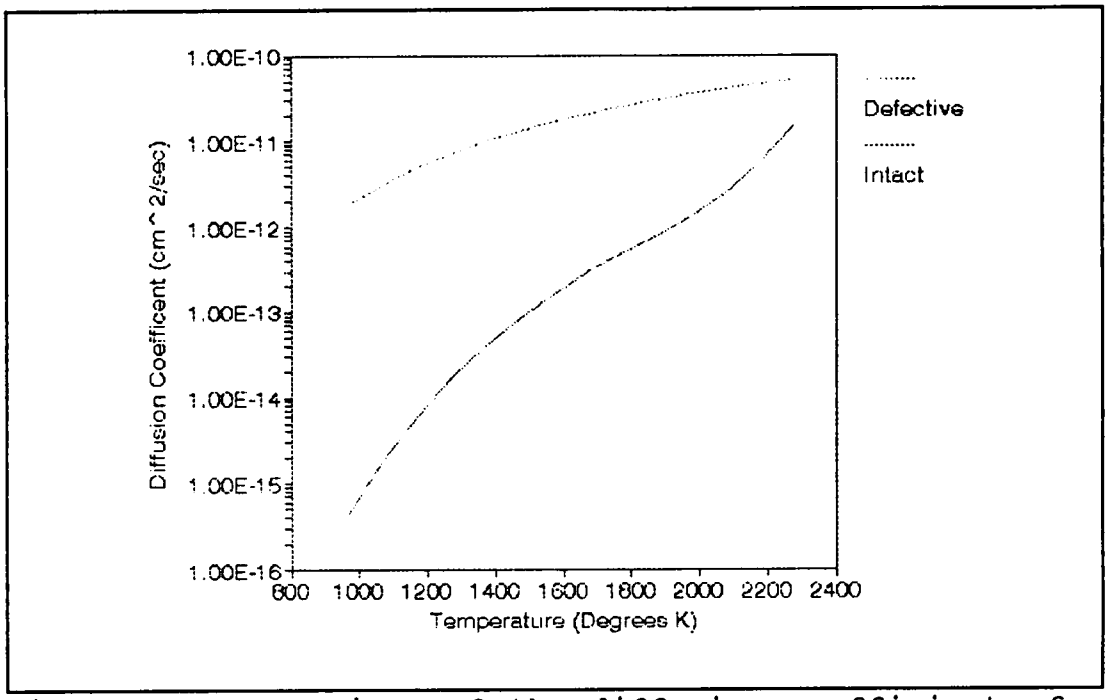



Figure 5.3 Comparison of the diffusion coefficients for intact and cracked SiC layers.

Once this was accomplished, the "reference" diffusion coefficient for the matrix graphite was modified to give the measured amount of cesium released from the fuel element. The new diffusion coefficient is shown in Figure 5.4. The revised diffusion coefficient is found to be

$$D=D_o*e^{(-\frac{A}{RT})} \qquad 5.2$$

where $D_o$ = 0.06 $cm^2/sec$ and

$A$ = 125000 j/mol

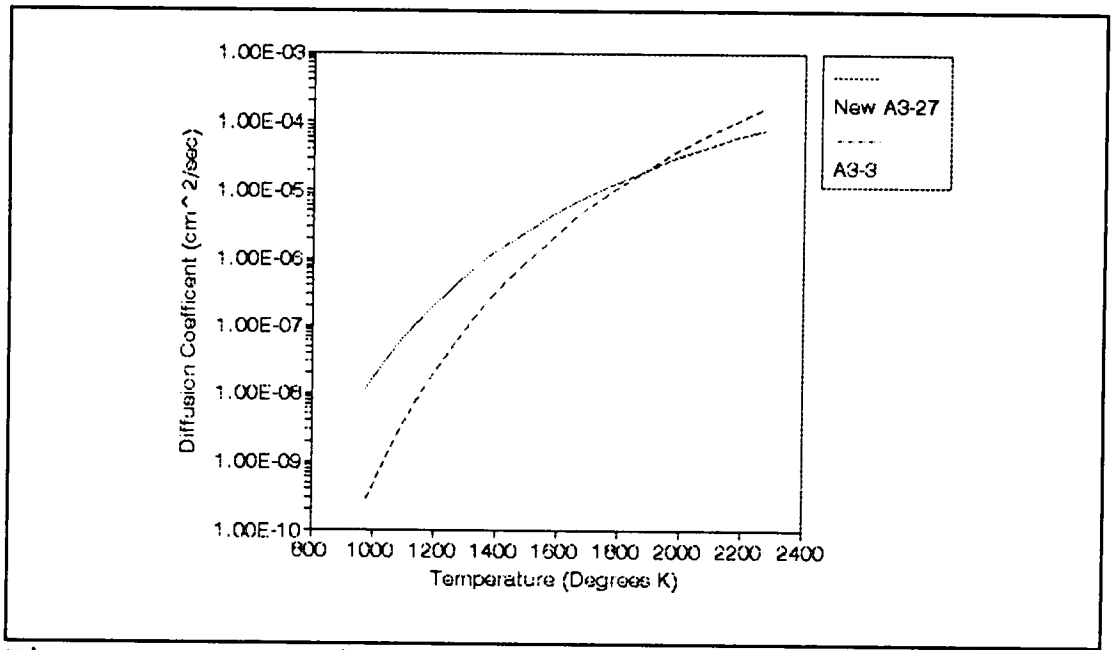



Figure 5.4 Comparison of Diffusion coefficient for Matrix Graphite.

Currently, there is no information about the sorption effects for reference A3-27 matrix graphite. To account for this lack of information, the sorption coefficients for A3-3 matrix graphite were used and an "effective" diffusion coefficient was developed to take into account the difference in sorption between the two matrix types. The new diffusion coefficient for matrix graphite is higher than the previous reference value. The new value takes into account the effects of sorption of cesium on matrix graphite.

Once the two diffusion coefficients for SiC and matrix graphite were determined, fission product release/birth values were calculated for 12 different heating tests and compared with measurement data. In calculating the results, the diffusion parameters were maintained constant in all tests, and only the test parameters such as the neutron fluences, irradiation time and temperature, heating time and temperature, the number of broken particles and the time the breakage occurred were changed. An example of the results is shown in Figure 5.5.

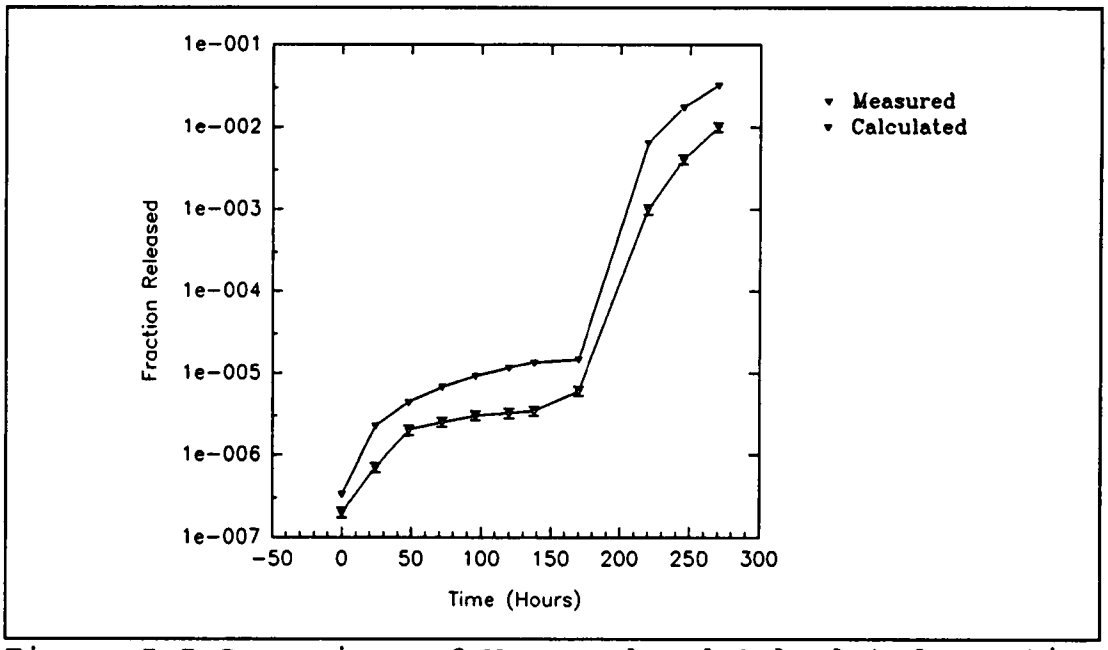



Figure 5.5 Comparison of Measured and Calculated Fraction Release of Cesium from FRJ2-K13/4 heated at 1600°C for 150 hours then heated at 1800°C for 100 hours

Overall, the results based on a consistent set of parameters were very good. All of the calculated releases were in good agreement with the experimental values.

When the measured results of heating experiments where the temperature remained over 1600°C for an extended period of time were examined, a high release of cesium was noted. This release is significantly greater than would be expected using the reference diffusion coefficient. Since the PANAMA code did not show a large particle breakage occurring and the krypton release did not show an increase, the release of cesium must be the result of diffusion out of the intact coated particles. To account for this, a third branch of the diffusion coefficient was added that increases the release at high heating temperatures. The formula for the diffusion coefficient of cesium in SiC becomes

$$D=D_{01}e^{\frac{\Gamma}{5}}e^{\frac{-A_1}{RT}}+D_{02}e^{\frac{-A_2}{RT}}+D_{03}e^{\frac{-A_3}{RT}} \qquad 5.2$$

An example of this is shown in Figure 5.5 where the heating temperature was at 1800°C for the last 100 hours of heating. The calculated fractional release was based on a SiC diffusion coefficent obtained from equation 5.2.

The model can be further verified by comparing the calculated and measured fractional release of Cs-137 at the end of irradiation. This is shown in Table 5.1. It is important to note that in several of the tests, the Cs-137/Cs-134 ratio was found not to be the value expected. This may indicate that some contamination was introduced during the post irradiation testing. The measured values

Table 5.1 Comparison of overall measured and calculated fractional release at the end of irradiation.

| Experiment | Measured Frac Rel. | Calculated Frac. Rel. |
|---|---|---|
| HFR-K3/1 | $9x10^{-6}$ | $4x10^{-7}$ |
| HFR-K3/3 | $2x10^{-5}$ | $2.5x10^{-10}$ |
| FRJ2-K3/2 | $2x10^{-5}$ | $3x10^{-6}$ |
| FRJ2-K3/4 | $6x10^{-6}$ | $1x10^{-6}$ |
| HFR-P4/1 | $5x10^{-6}$ | $3x10^{-6}$ |
| HFR-P4/2 | $5x10^{-6}$ | $1.17x10^{-5}$ |
| HFR-P4/3 | $2x10^{-5}$ | $2.1x10^{-4}$ |
| SL-P1 | $5x10^{-6}$ | $3.77x10^{-6}$ |

Note: The measured values encompass all the fuel/elements and compacts in each test while the calculated values are for only a few elements or compacts in each test.

should be considered upper limits and may not represent the actual values of cesium release from the fuel elements. Additionally in the HFR-P4/1,2,3 and SL-P1 test, the measured values are from all the 12 fuel compacts in a given test while the calculated values are from only one or two fuel compacts. Since failed particles are the main contributors to the cesium release, the number of broken particles in each fuel loading can be estimated, but not their distribution among the compacts, since the measured release is averaged over all 12 fuel compacts.

In general, the calculated release to birth in Table 5.1 is less than the measured fractional release which may reflect that measured release to brith ratios are upper

limits. This follows since in some of the tests particle failures occured during the PIE test in which case the measeured release would be too high. This explains mos of the difference, but HFR-P4/2/8 is too high[1].

Figure 5.6 shows the results of the heating test of compact SL-P1/6. In this test, it was assumed that two SiC layers failed 4200 hours into the irradiation cycle. This failure seemed to be due to a crack in the SiC layer as the krypton was readily released.

Figure 5.7 shows the fractional release from SL-P1/9. This compact had a significantly higher release than almost any other fuel compact. To account for this, it was assumed that 20 particles were broken after the end of irradiation, but before the start of heating. The SiC layers of these particles allowed krypton to pass through them.

SL-P1/10 had significantly higher release at the start of the heat up then did any other compact or fuel element except for SL-P1/9. This release is shown in Figure 5.8. To account for this release, it was assumed that 20 particles were defective from manufacturing. These broken particles may also have been due to fracture when the gas probe was placed into the reactor prior to the start of irradiation.

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[1] This may be do to the fact that several particles broke in HFR-P4/2/8 and did not break in any other test, so the release from HFR-P4/2/8 would be much greater than the other releases.

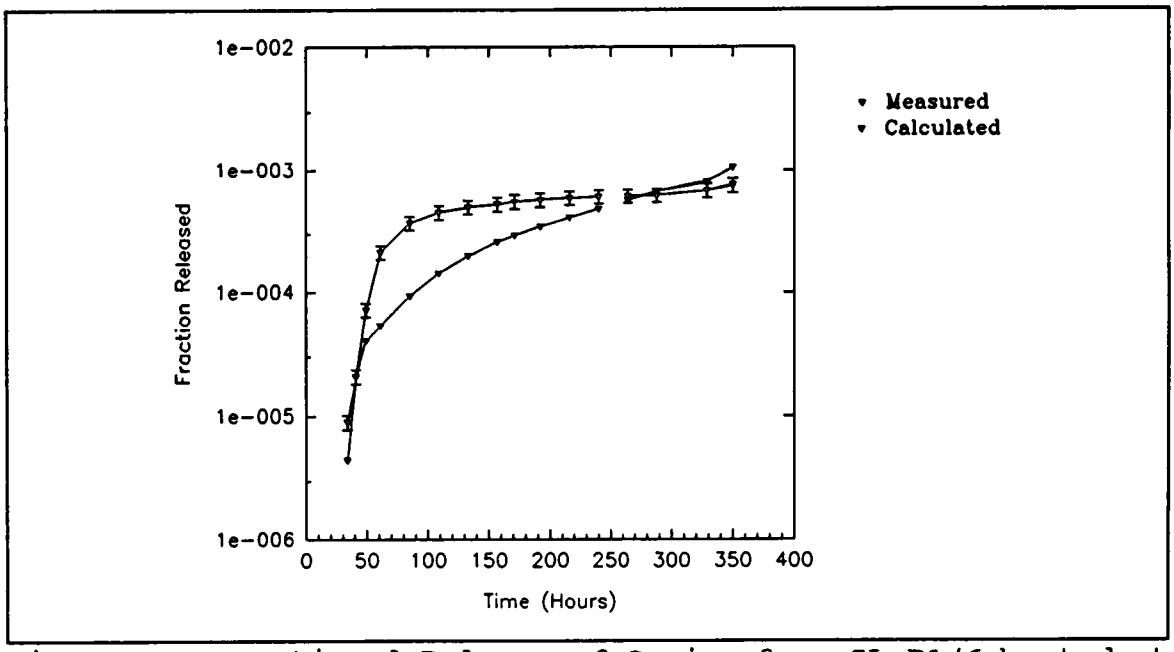



Figure 5.6 Fractional Release of Cesium from SL-P1/6 heated at 1600°C.

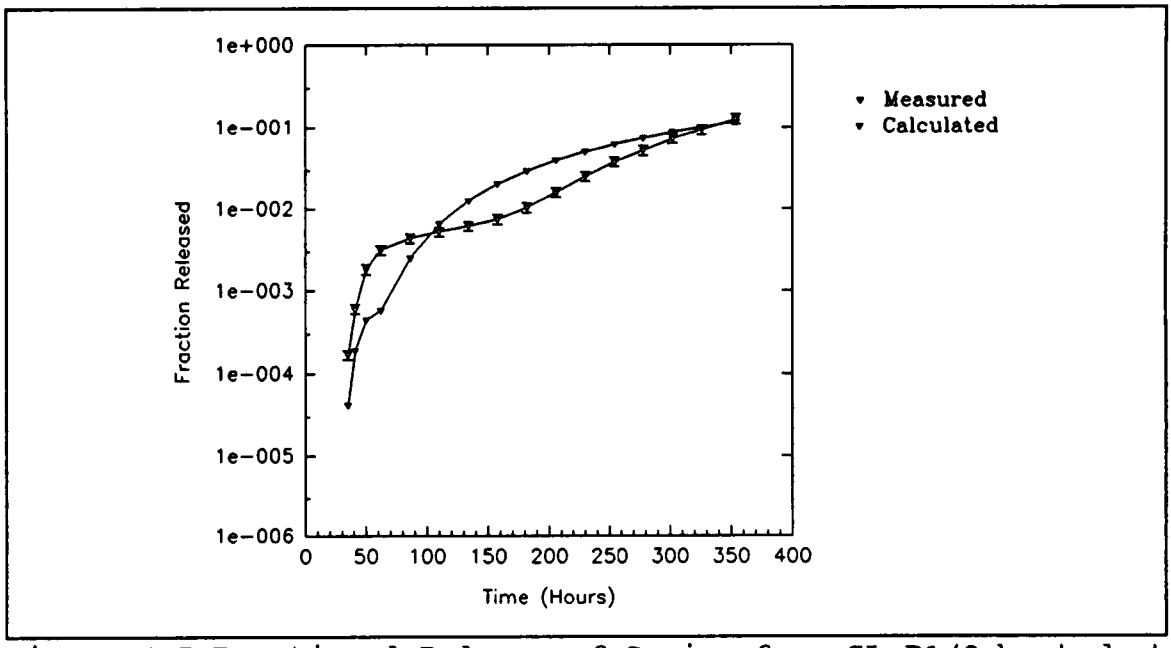



Figure 5.7 Fractional Release of Cesium from SL-P1/9 heated at 1600°C.

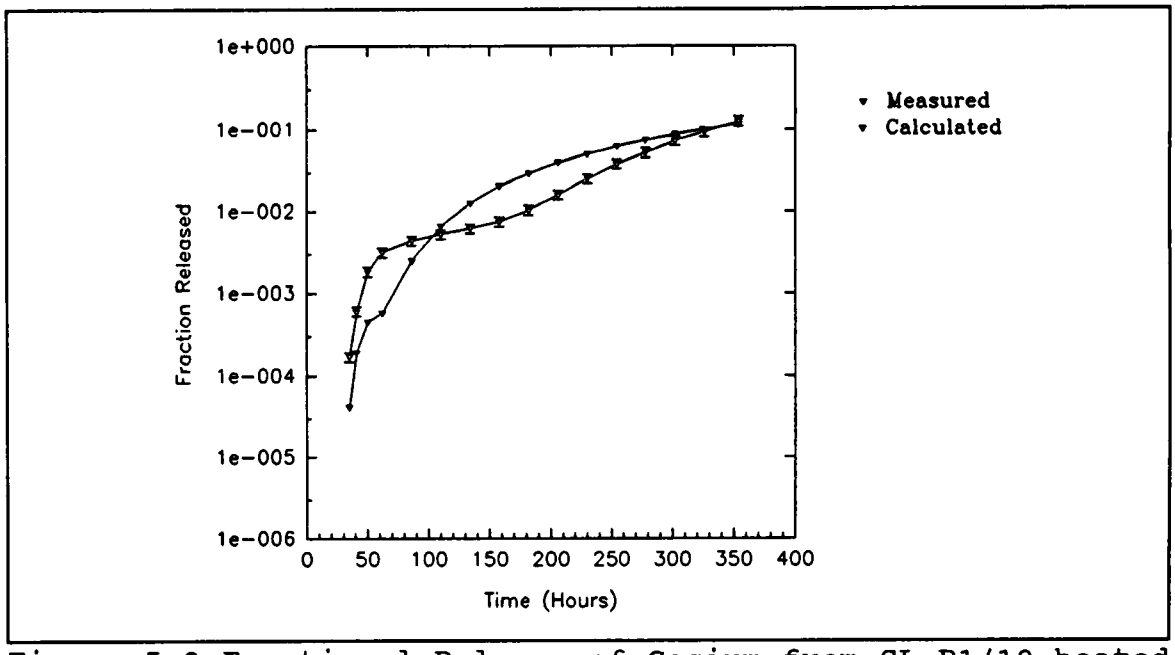



Figure 5.8 Fractional Release of Cesium from SL-P1/10 heated at 1700°C.

Figure 5.9 shows the calculated and measured release from HFR-P4/1/12. This fuel compact had the lowest fast fluence of all the HFR-P4 tests. Here, a particle was assumed to have failed approximately 7700 hours into the irradiation time. The SiC layer in the defective particle appears to have cracked enough to allow for the cesium to escape, however, it still retains the krypton.

HFR-P4/1/8 has a very high fast fluence and a fuel burnup. The release from this heating test is shown in Figure 5.10. In this test, one particle was assumed to have failed at 7700 hours into the irradiation test. This breakage allowed for the rapid release of krypton.

Figure 5.11 shows the release from heating test HFR-P4/2/8. The fuel particles in the HFR-P4/2 compacts had a thicker coating layer (51 $\mu$m) them the rest of the tests. However even with this thicker layer, the cesium release from HFR-P4/2/8 matched the release of other tests at the end of heating. In order to match the calculated release to the measured release, two particles were assumed to break around 7750 hours into the irradiation period. These particles release the krypton at a rapid rate.

The heating test of HFR-P4/3/12 went to 1800°C in two separate stages. The release, as shown in Figure 5.12, at the end of heating was around 30%. To match this release in the calculations, it was necessary to assume that 500 particles had failed at 350 hours into the heating test.

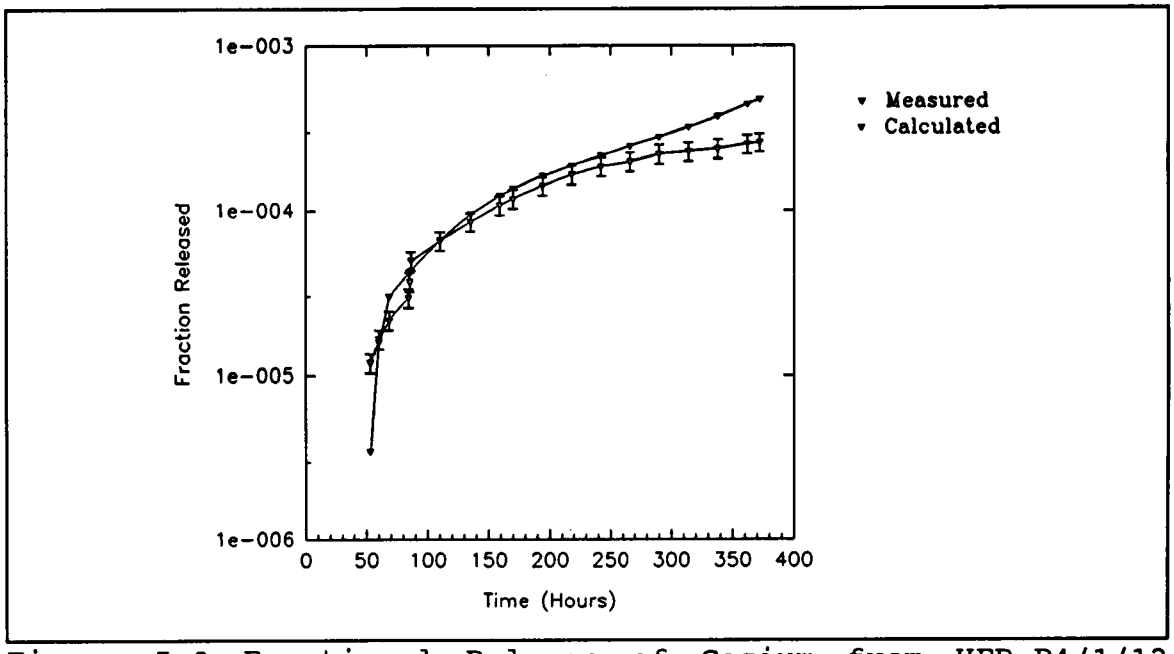



Figure 5.9 Fractional Release of Cesium from HFR-P4/1/12 heated at 1600°C.

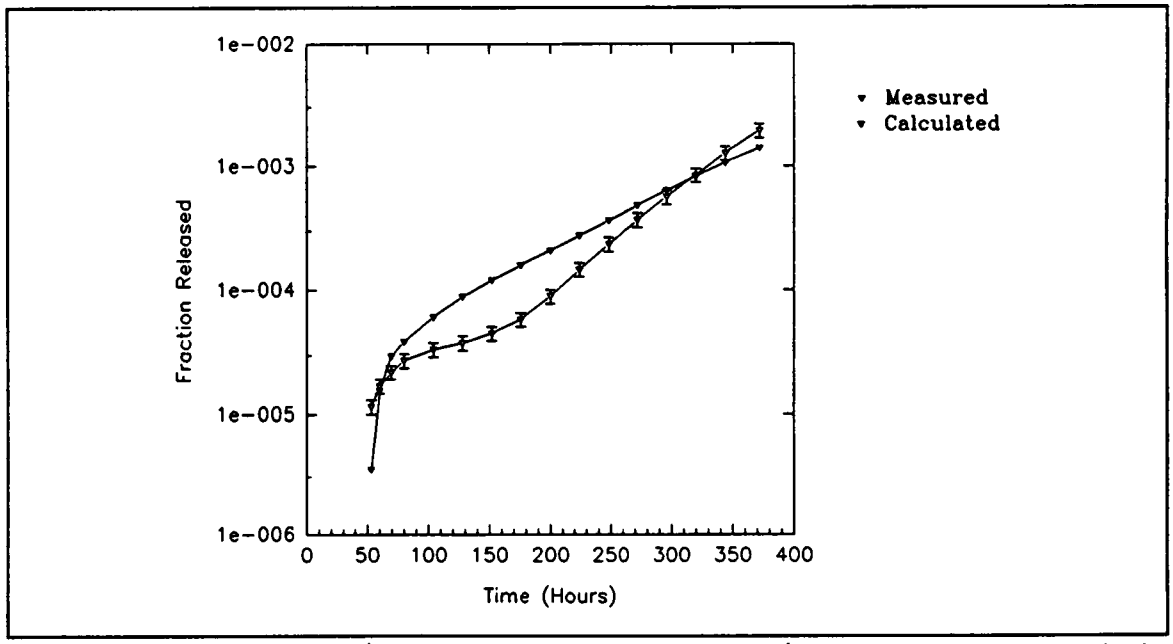



Figure 5.10 Fractional Release of Cesium from HFR-P4/1/8 heated at 1600°C.

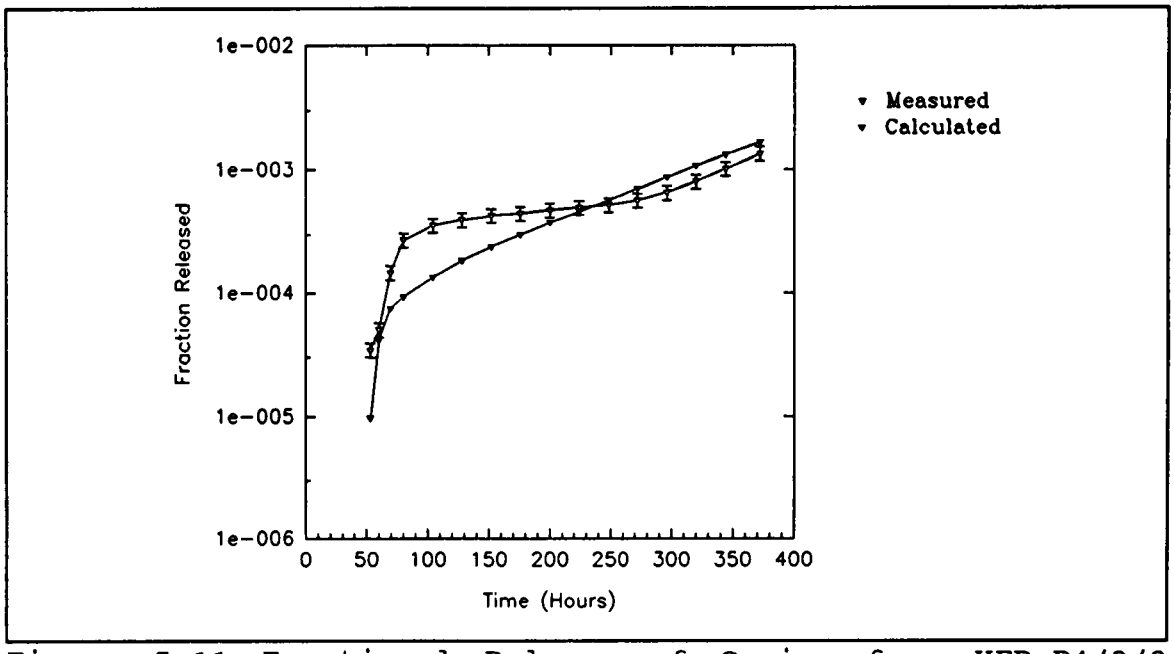



Figure 5.11 Fractional Release of Cesium from HFR-P4/2/8 heated at 1600°C.

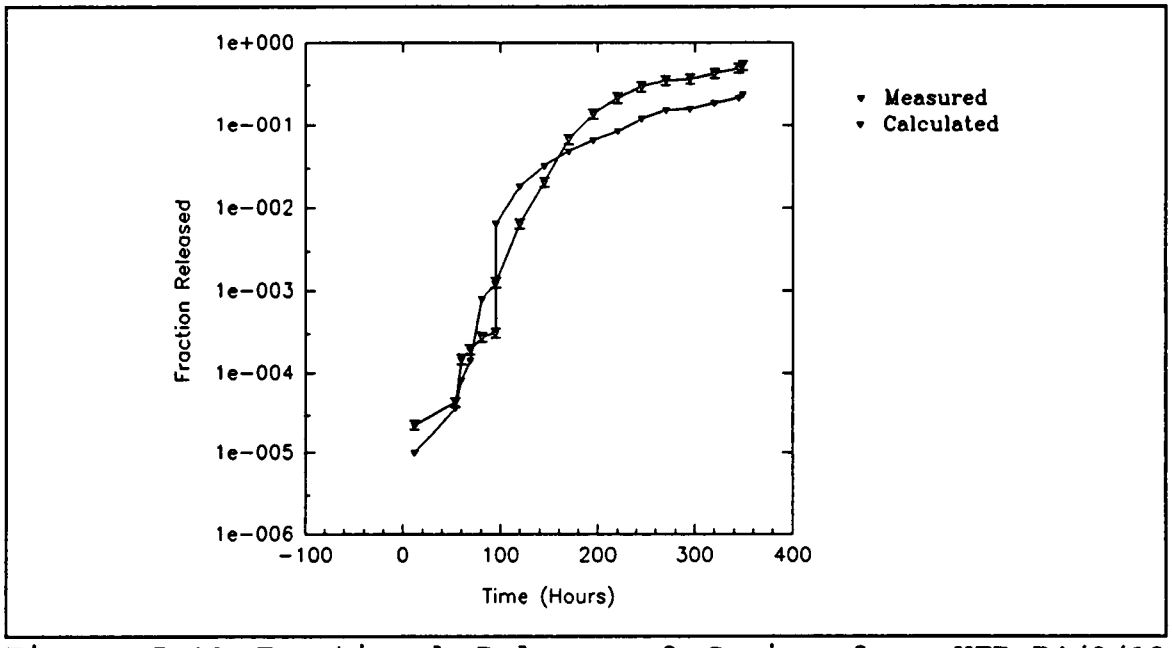



Figure 5.12 Fractional Release of Cesium from HFR-P4/3/12 heated at 1800°C.

Figure 5.13 shows the release from FRJ2-K13/2. It was assumed that this fuel element had four particles which failed at 8000 hours into the irradiation period. The large underestimation at the beginning of the heating period by the model may be due to the assumed sorption coefficents.

In HFR-K3/1, no particles were broken but the calculated release still greatly exceeds the measured release as shown in Figure 5.14. The reason for this is not known.

The fraction release of cesium from FRJ2-K13/4 is shown in Figure 5.15. FRJ2-K13/4 had two different heating tests, the first was at 1600°C and the second at 1800°C which accounts for the last 100 hours on the graph. It was assumed that only one particle failed between the end of the irradiation period and the start of the heating period. The high release during the second heating test was due to the third branch of the diffusion coefficient and the release that branch produced.

HFR-K3/3 was heated for two separate times at 1800°C. The release from this fuel element is shown in Figure 5.16. It was assumed that the only two broken particles occurred between at the end of irradiation and the start of heating, when two SiC layers failed. These failed in such a way to allow for the krypton to be easily released from the SiC layer.

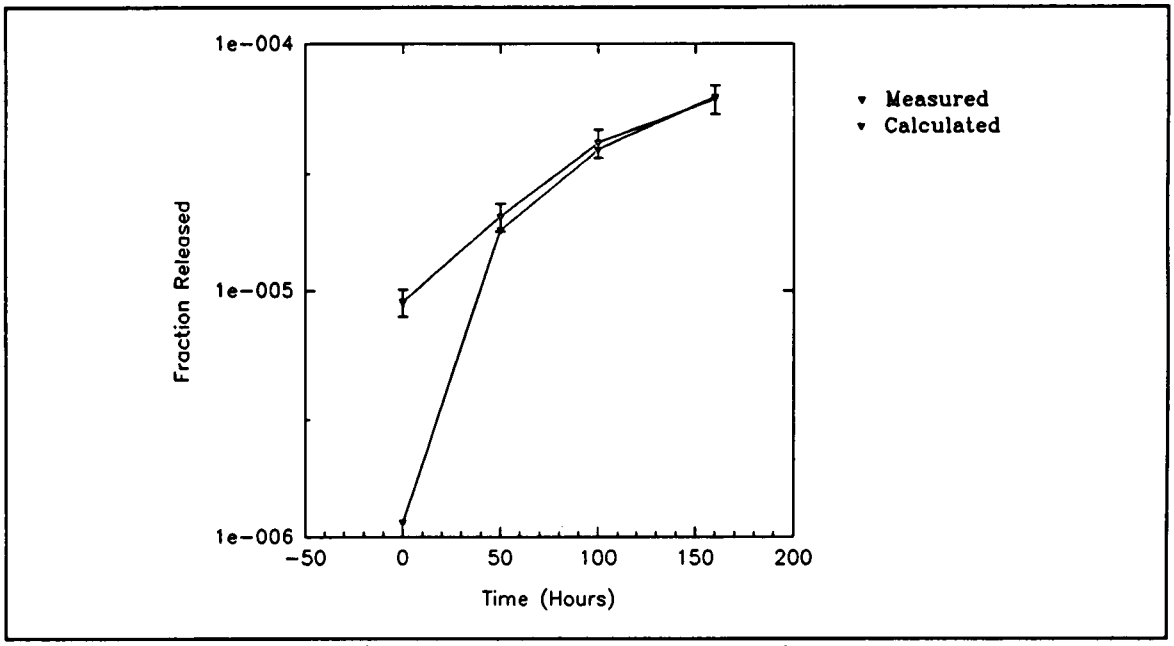



Figure 5.13 Fractional Release of Cesium from FRJ2-K13/2 heated at 1600°C.

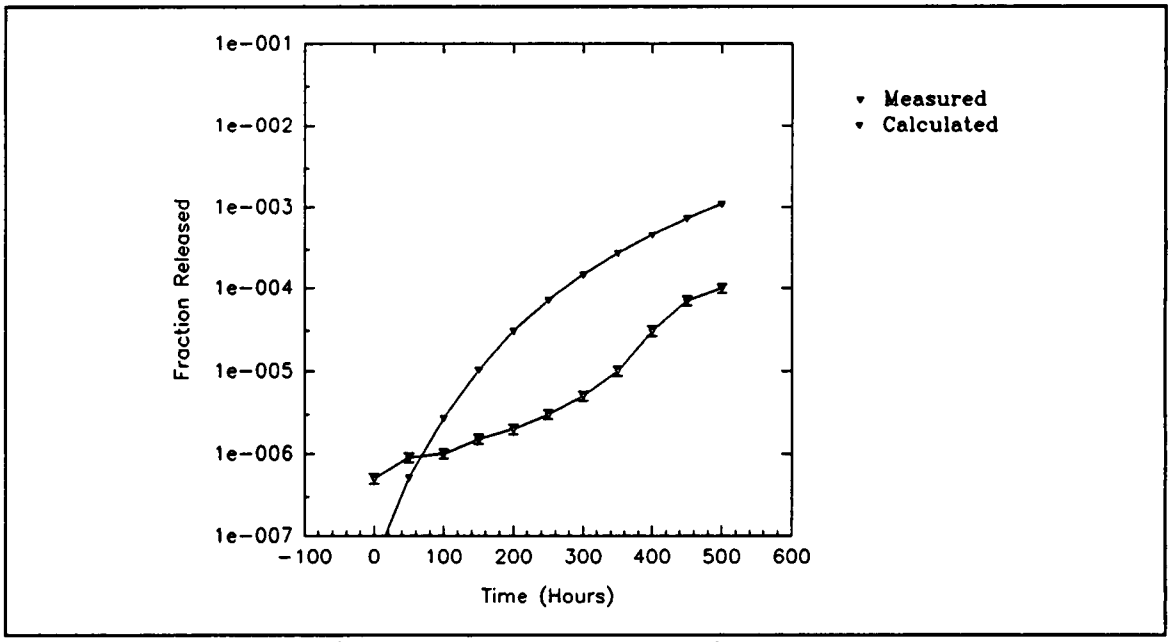



Figure 5.14 Fractional Release of Cesium from HFR-K3/1 heated at 1600°C.

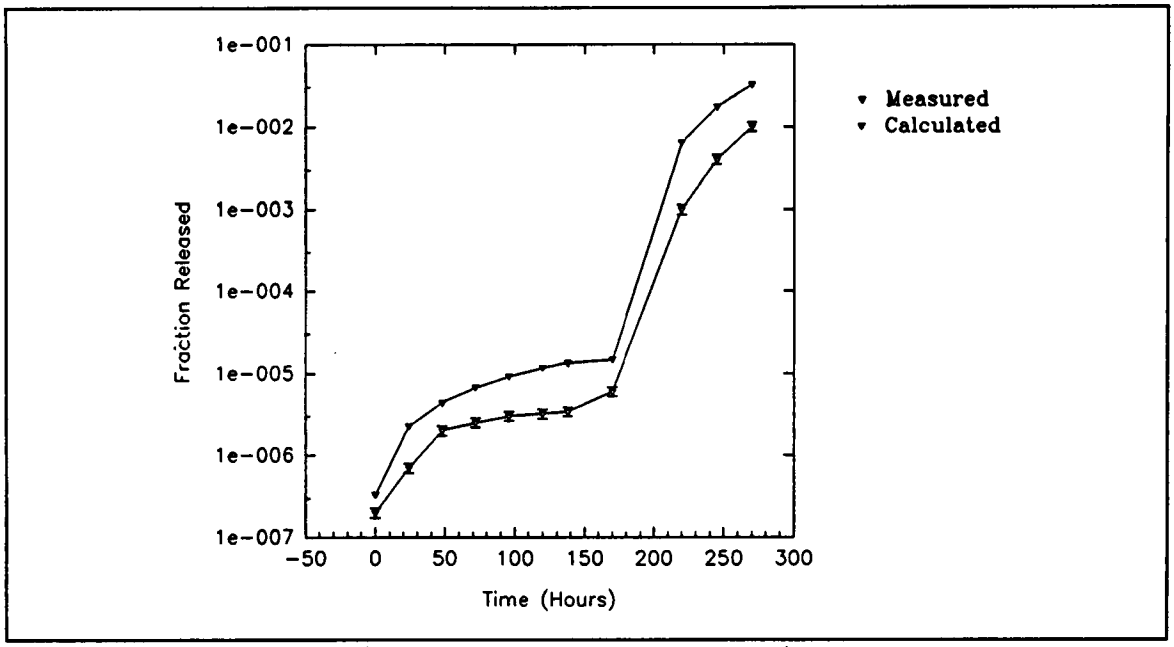



Figure 5.15 Fractional Release of Cesium from FRJ2-K13/4 heated at 1600°C and 1800°C.

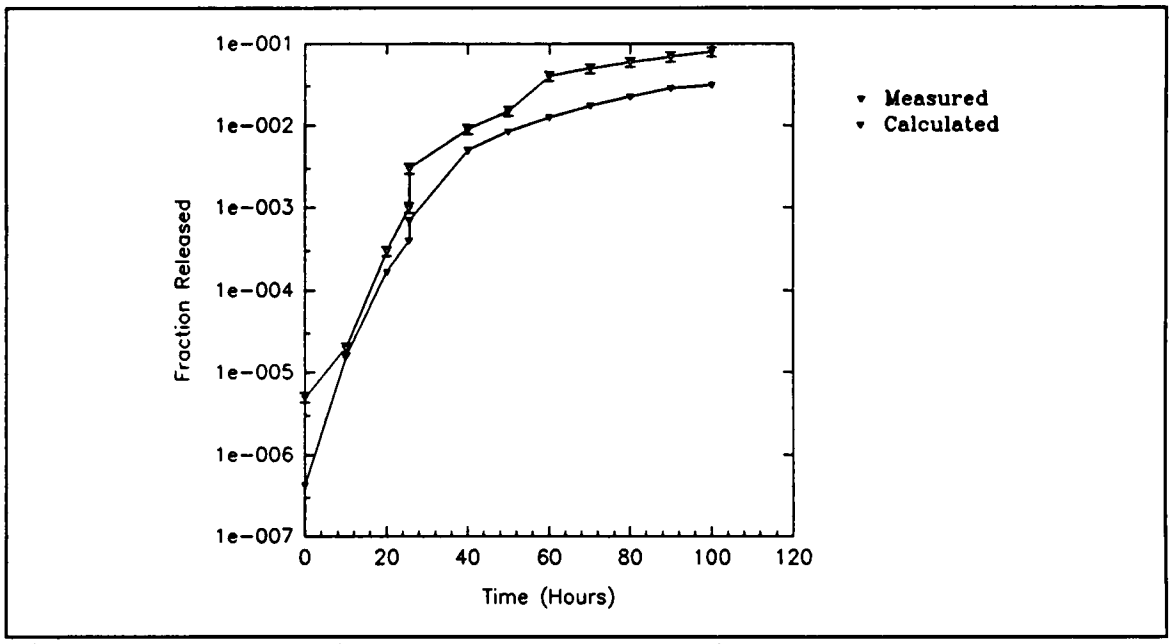



Figure 5.16 Fractional Release of Cesium from HFR-K3/3 heated at 1800°C.

Several tests of AVR fuel were run, however, in the beginning of operation of the AVR reactor, a large number of particles failed and contaminated the reactor with a significant quantity of metallic fission products. Over a period of time, the surface contamination could diffuse into the fuel elements and be released during the heating tests. This makes results of those heating tests suspect and thus, they have not been included in this report.

In general, the overall calculated results for the fractional release of cesium in good agreement with the measured release at the end of irradiation, but underestimates the release at the early part of the heating. This may be due to the sorption coefficients utilized whcih were estimated and not measured. These could be improved with additional work.

### 5.3 Krypton Results

The irradiation data was analyzed and it was observed that there are two distinct release profiles of $Kr^{88}$ during irradiation. The first is a gradual increase with irradiation time in the release rate to birth rate ratio (R/B) for $Kr^{88}$. In the second case, the R/B remains constant. This difference may be due to one of several reasons: surface contamination of the coated particles, particles whose coatings have cracked during irradiation, particles that are defective from manufacturing, or natural uranium contamination in the graphite matrix.

The first kind of release profile is appears to be due to krypton generated by the fissioning of natural uranium contamination in the graphitematrix material. As discussed in section 2.3, graphite consists of graphite grains and pore graphite. Natrual uranium is only in the grains. Krypton that "recoils" into the pore graphite is released immediately from the fuel element or compact. If the SiC or the PyC layers of the fuel particle remain intact, krypton is not release from the coated particles during irradiation. In all the fuel irradiation tests, $Kr^{88}$ R/B values were much greater then can be accounted for by diffusion through intact coated particles. As a result, the R/B ratio measurements for $Kr^{88}$ must be due to matrix graphite contamination, or to contamination in the outer PyC layer.

If the $Kr^{88}$ birth is very low over and raises significantly over irradiation time, the contaimiantion is in the grains. If the fractional release is relatively high and constant during irradiation time that implies surface contaimantion of the fuel particles. Calculations have been preformed for HFR-P4/1/8 to indicate the ratio of natural uranium in the matrix graphite to $U^{235}$ in the kernels should be around $2.8 \times 10^{-8}$, based on assuming uniform contimination in the matrix graphite and that the matris graphite is 100% pore graphite.

Arround 40% of the volume of the matrix graphite is crystals and the remaining 60% is pores, with natural

uranium contamination occurring only in the crystals. Because of the very low diffusion coefficient of Krypton in graphite crystals ($5.5 \times 10^{-19} cm^2/sec$ at 800°C) very little krypton diffuses out of the crystal during fuel irradiation, but a large fraction can be released after a short time at 1600°C or higher.

On the above basis and recoil release, use can be made of the $Kr^{88}$ fractional release rate to calculated the natural uranium in the graphite grains for an assumed effective grain diameter. This is discussed in detail in Reference 10. Given the intial contaminatin in the graphite grains, the release of $Kr^{85}$ during irradiation and heating times can be calculated. The $Kr^{85}$ fractional release is primarily due to "recoil" during irradiation and due to "diffusion" during heating. Based on the theory section, the recoil distance in graphite is R=12$\mu$m and the fractional release from a graphite crystal of diameter 100$\mu$m is about 9%. As a result, nearly all the $Kr^{85}$ generated during irradiation can be released during heating. The diffusion parameters developed for this behavior during heating are shown below.

$D_0 = 2.9X10^{-8}$ $cm^2/sec$
A = 210300 J/mol
K = 14

The calculated fraction of $Kr^{88}$ generated in the graphite grain relative to the amount generated in the fuel kernels can be estimated by multiplying the initial release to birth ratio by the ratio of fission products generated in

the matrix graphite over the irradiation period to the number of fission products generated in the coated fuel particles and by the fraction of krypton that is not release by recoil[10].

For test HFR-P4, the pore contamination level that which gave an initial fractional release of $Kr^{88}$ is $2x10^{-8}$. For HFR-P4/1, the ratio of fission products generated in the matrix graphite over the irradiation period to the number generated in the coated fuel particle is around 17. For a grain diameter of 100$\mu$m, the $Kr^{88}$ concnetration is about 28 times that in the pore graphite as calculated above. If the above information is combined with the initial R/B values, a fraction of about $9.5x10^{-7}$ of the krypton generated in the fuel element is available for release from the graphite grains by diffusion. It was observed that in most cases during the heating tests, the measured fractional release of krypton stayed around $5x10^{-7}$ during the initial heating time. The above implies that the effective grain diameter of less than 100$\mu$m can give a fractional release of $Kr^{85}$ during the heating period that agrees with measued values. More information of this calculation and other results are given by Reference 10. The net result is that the initial rise in krypton release during the heating and the subsequent level can be attributed to the diffusion of $Kr^{85}$ out of the grains with a diameter of 100$\mu$m and that was done here. The above explains the initial $Kr^{85}$ fractional release duing heating.

While the level of release is not very significant (since is very low) the above is important in understanding the phenomna that characterize the release.

The values received for the diffusion coefficient of krypton in SiC were empirically derived for different parameters[3] for several fast fluences, but they were not tied together to form a unifying set of diffusion parameters. This was done here and the diffusion parameters used are:

$D_0 = 2.66X10^{-8}$ cm$^2$/sec
A = 266000 J/mol
K = 1

These values gave good agreement in some cases, however they drastically underestimated them in other cases. It was assumed that in these cases, a SiC layer had cracked in at least one particle and the SiC layer was no longer effectively retaining the fission gas. However, since there was not an immediate increase in the krypton release, the outer PyC layer must have remained intact to retain the gas. A fast fluence degradation value was estimated for the PyC to enable the calculated releases to match the measured releases. The parameters used were

$D_{01}$ = $2.9X10^{-4}$ cm$^2$/sec
$A_1$ = 291000 J/mol
K = 100
$D_{02}$ = $2X10^{9}$ cm$^2$/sec
$A_2$ = 923000 J/mol

In general, the failure of the SiC layer was modeled in two distinct manners. Either the particle failed completely in relation to krypton, it which case the diffusion coefficient was set to $3x10^{-2}$ $cm^2/sec$, or the SiC layer was largely undamaged, in which case the diffusion coefficient was the same as for an intact layer.

Figure 5.17 shows the fraction release from fuel compact HFR-P4/1/8. It was assumed that one particle that failed approximately 7700 hours into the irradiation time. This SiC in this particle failed in such a way as to allow the krypton be readily released.

Figure 5.18 shows the fractional release from HFR-K3/3. In this fuel element it was assumed that two particles had failed between the end of irradiation and the start of heating. The SiC failed completely in regards to krypton.

HFR-K3/1 had no broken particles and was heated at 1600°C for 500 hours. The release is shown in Figure 5.19. The underestimation of release at the end of heatin suggest the need for further refinement of the diffusion parameters.

Figure 5.20 shows the release from SL-P1/9. This fuel compact was heated at 1700°C. It was assumed that 20 particles failed after irradiation but before heating. This SiC in this particle failed in such a way as to allow the krypton be readily released.

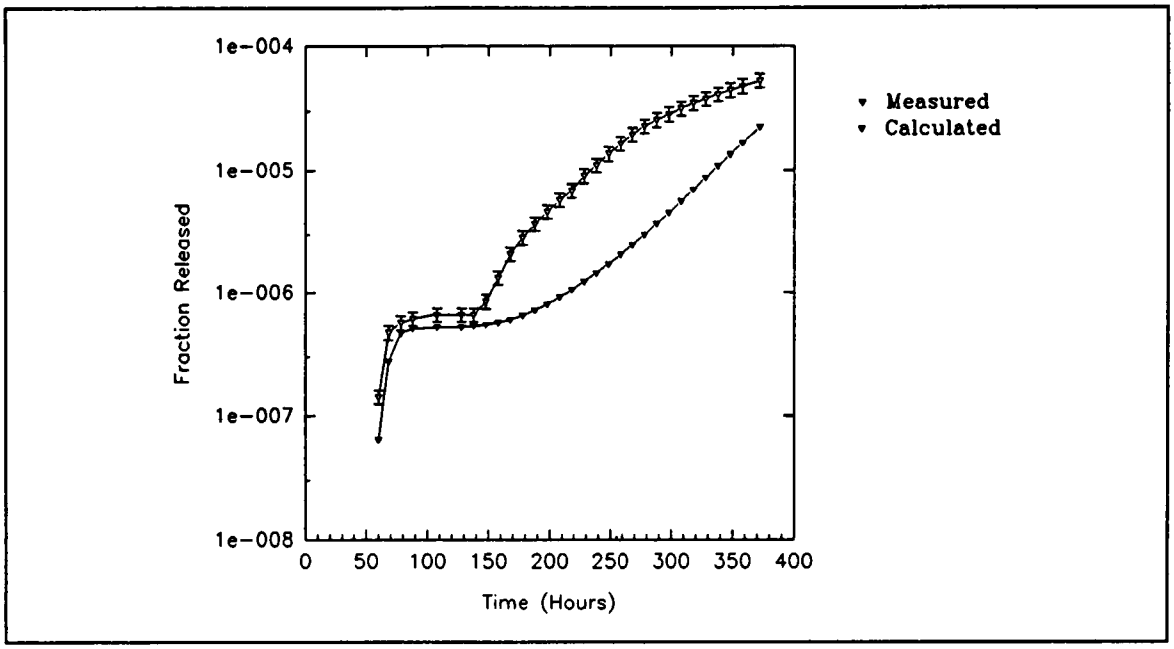



Figure 5.17 Fractional release of Krypton from HFR-P4/1/8 heated at a temperature of 1600°C.

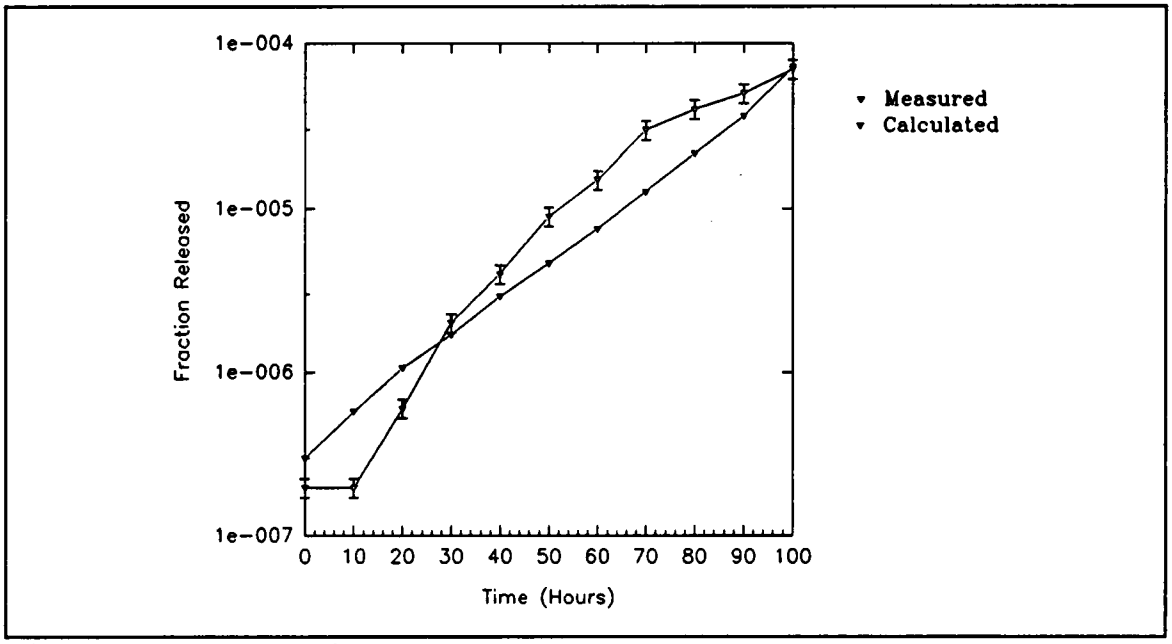



Figure 5.18 Fractional Release of Krypton from HFR-K3/3 heated at 1800°C.

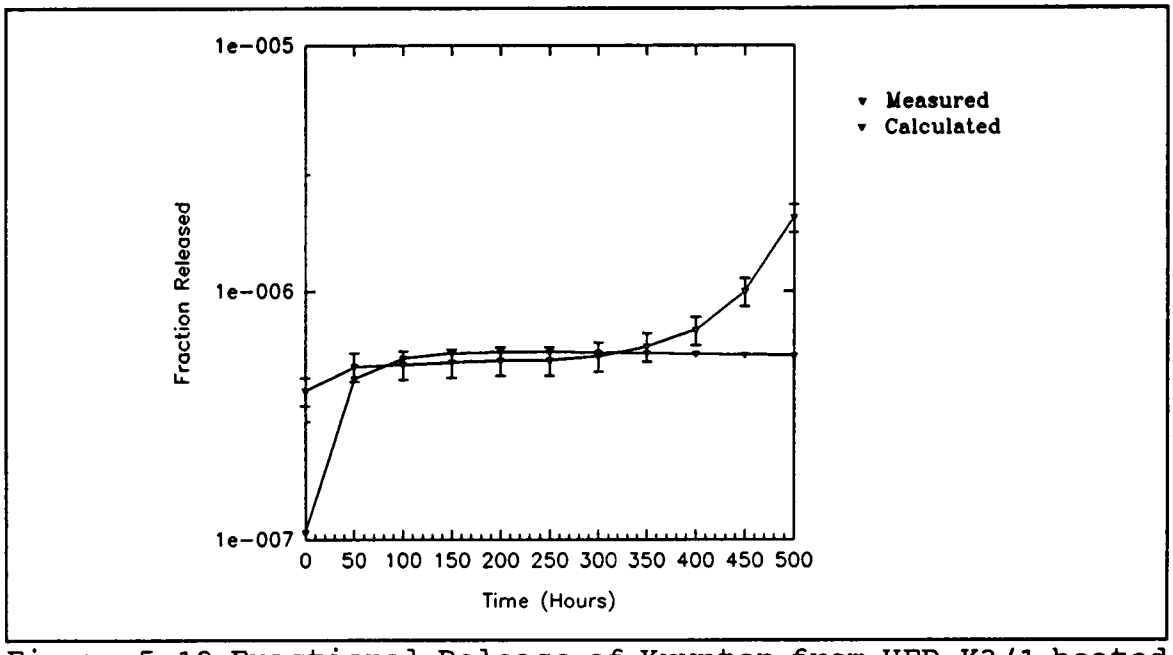



Figure 5.19 Fractional Release of Krypton from HFR-K3/1 heated at 1600°C.

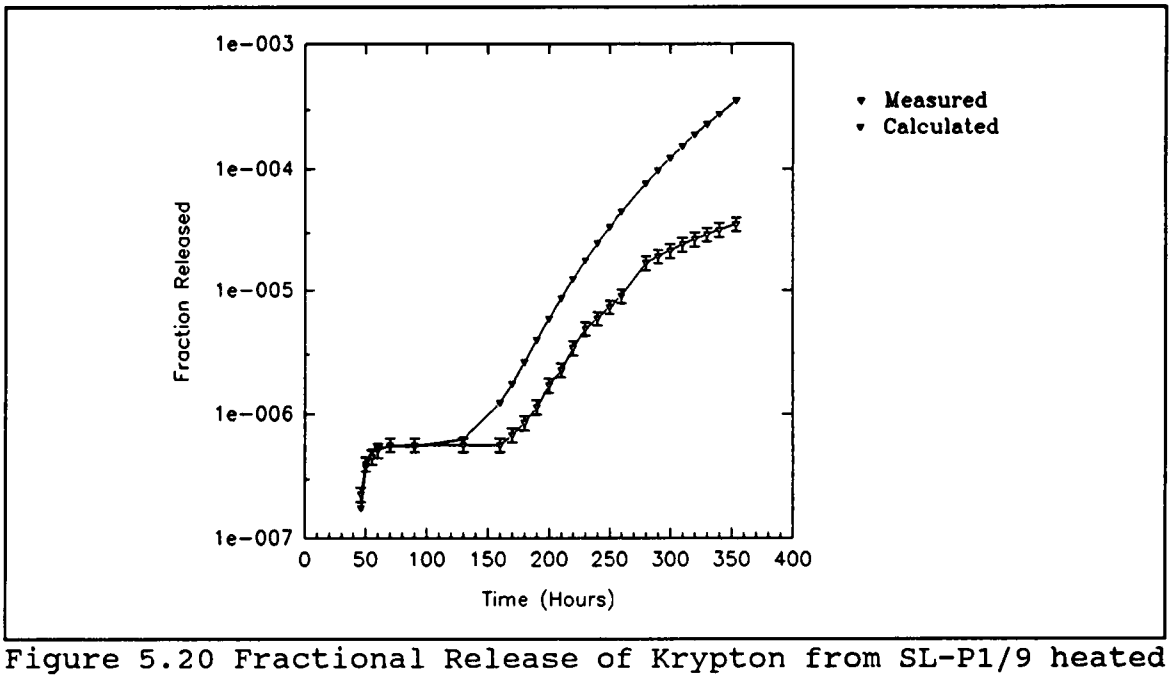



Figure 5.20 Fractional Release of Krypton from SL-P1/9 heated at 1700°C.

Figure 5.21 shows the release from HFR-P4/2/8, which was heated at 1600°C. It was assumed that two particles failed approximately 7700 hours into the irradiation time. This SiC in these particles failed in such a way as to allow the krypton be readily released. The SiC layer in this particle was 51μm however this did not significantly decrease the release.

Figure 5.22 shows the release from SL-P1/10. The fuel compact was heated at 1700°C. It was assumed that 20 particles were defective due to manufacturing or were fractured when the gas probe was inserted. Some of the SiC in this fuel compact failed in such a way as to allow the krypton to be readily released while others managed to retna the krypton. Since UT-KFA code cannot model this release, it was assumed that all the cracked SiC layers eailsy released the krypton, resulting in an overestimation of the release.

The fuel compact SL-P/1/6 was heated at 1600°C. The relese from this compact is shown in Figure 5.23. It was assumed that two particles that failed approximately midway into the irradiation time. This SiC in this particle failed in such a way as to allow the krypton be readily released.

Figure 5.24 shows the release from fuel compact HFR-P4/1/12 which was heated at 1600°C. It was assumed that one particle that failed approximately 7700 hours into the irradiation time. This SiC in this particle still retained the krypton.

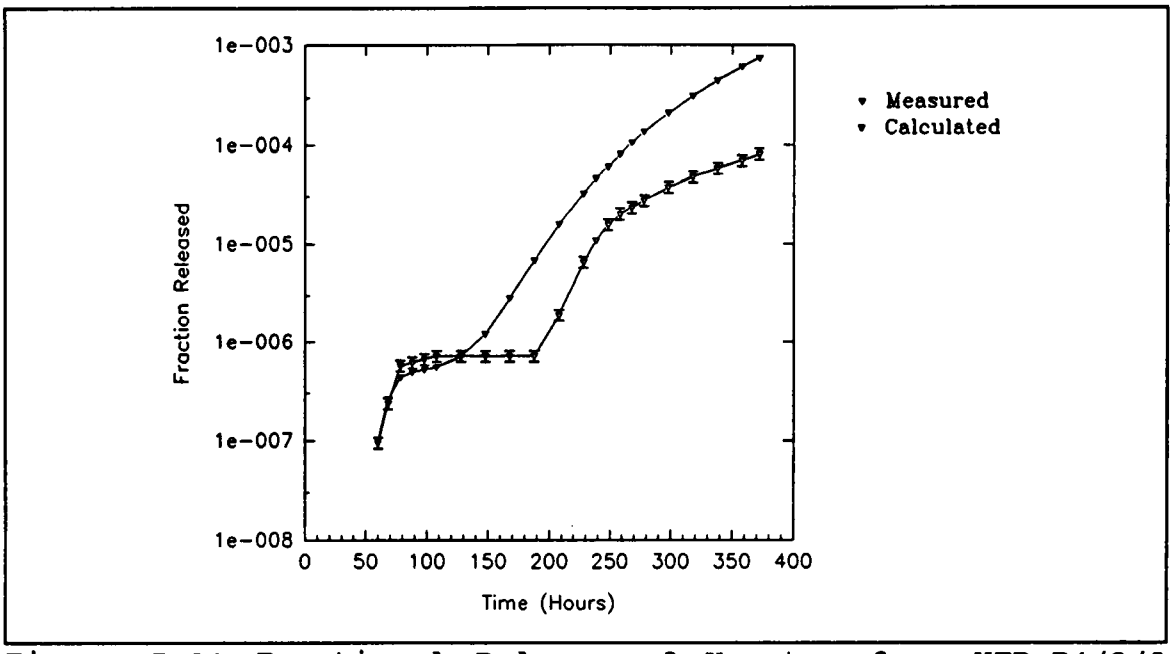



Figure 5.21 Fractional Release of Krypton from HFR-P4/2/8 heated at 1600°C.

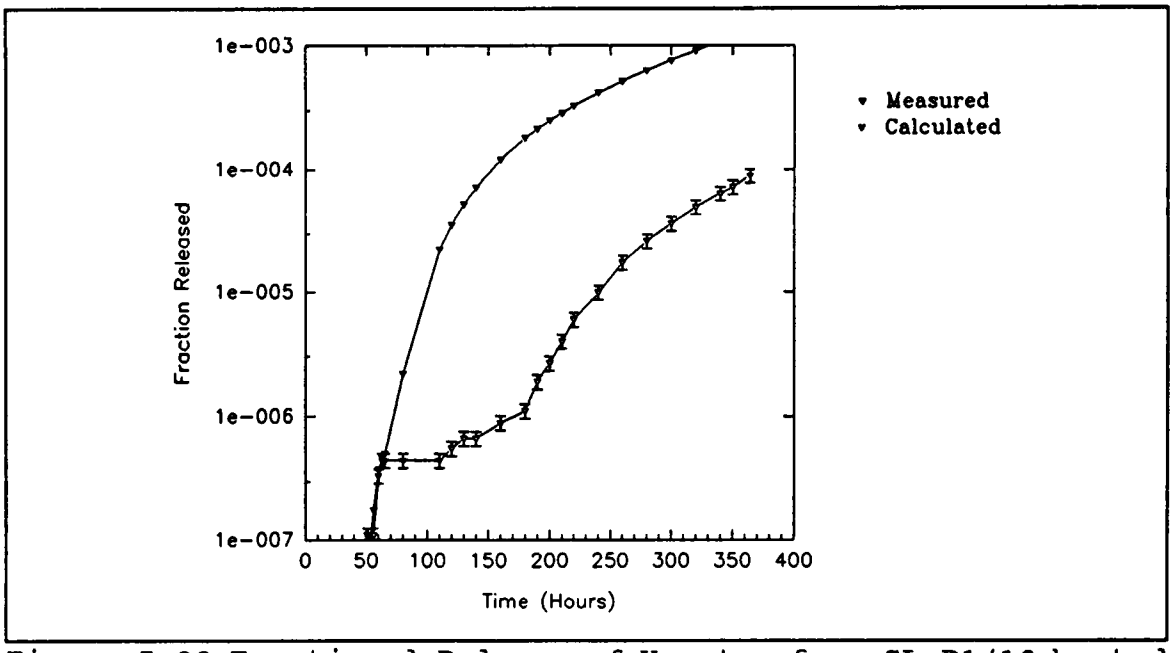



Figure 5.22 Fractional Release of Krypton from SL-P1/10 heated at 1700°C.

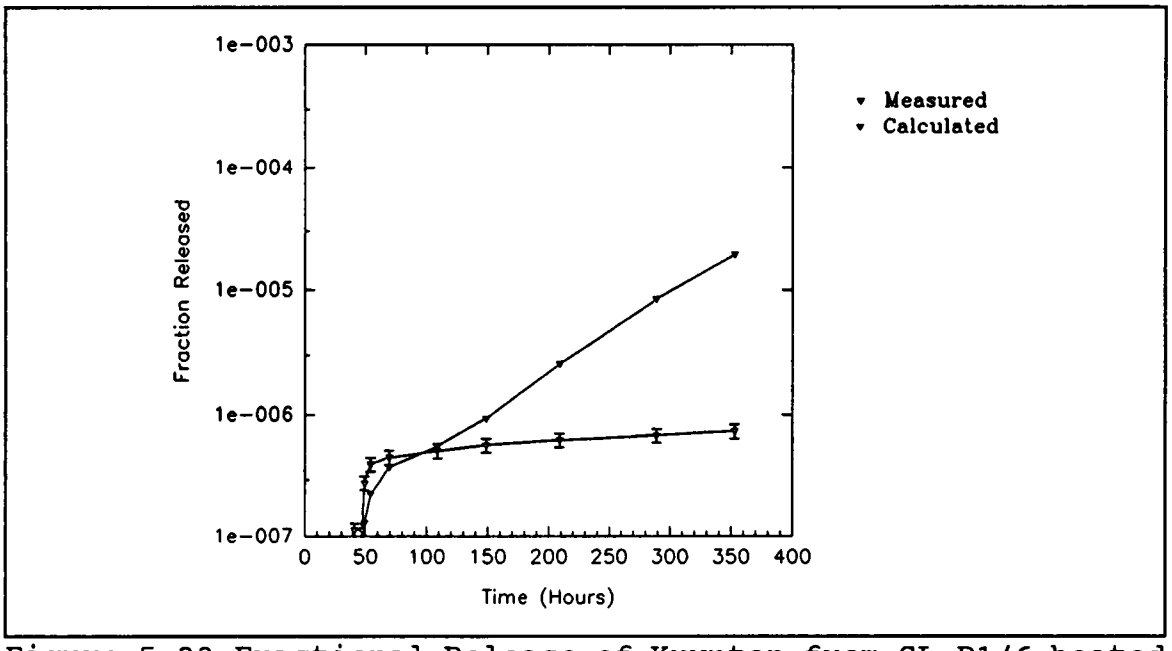



Figure 5.23 Fractional Release of Krypton from SL-P1/6 heated at 1600°C.

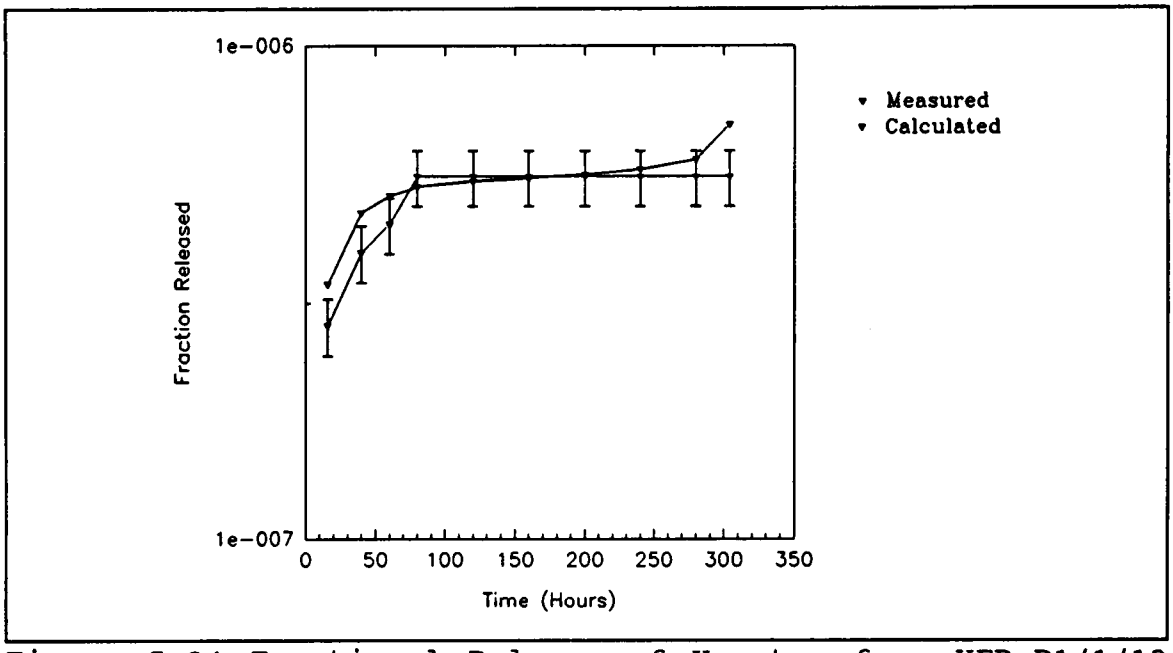



Figure 5.24 Fractional Release of Krypton from HFR-P1/1/12 heated at 1600°C.

In HFR-P4/3/12, the heating temperature reached 1800°C for an extended period of time. The release is shown in Figure 5.25. Due to a furance malfunction, the total heating time was broken up into two sections. The fuel compact was held at 1800°C for around 24 hours when the furance failed. It was repaird and the heating resumed for the remaining time. Even though a large number of particled did break (around 500) the release of krypton was slowed by the SiC layer and the intact outer PyC layers.

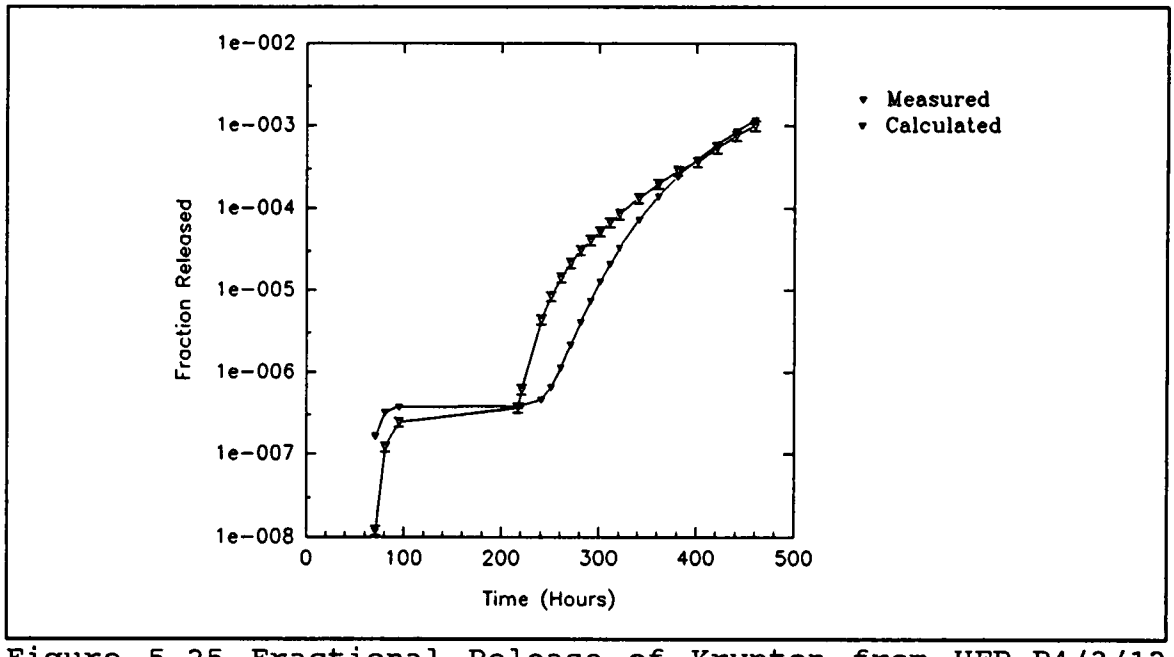



Figure 5.25 Fractional Release of Krypton from HFR-P4/3/12 heated at 1800°C.

## 5.4 Strontium Results

The UT-KFA model uses a diffusion coefficient for strontium in SiC that allows for a fast fluence effect. To find this effect, a version of the code was developed that allowed the SiC diffusion coefficient to be arbitrary changed as a function of time. The diffusion coefficient was adjusted for HFR-P4/1/8 until the measured release matched the calculated release (within a factor of two.) A Arrhenius relationship was then used to solve for the $D_0$ and A. This data was used to show the release of HFR-P4/2/8 and gave excellent agreement. However when it was applied to HFR-P4/3/7, the calculated release was significantly greater than the measured release. To explain this difference, it is important to note that the diffusion coefficient for strontium in matrix graphite is fairly low. This means that any strontium released at the beginning of heating was released from the coated particles early in the irradiation period. Since HFR-P4/2/8 and HFR-P4/1/8 were irradiated at 920°C while HFR-P4/3/7 was irradiated at 1050°C, values for both ranges need to be developed to attain a suitable result. The diffusion coefficient required for HFR-P4/3/7 to calculate the correct release was then found using the modified program. From two low temperature diffusion coefficients (one at 920°C and one at 1100°C) an Arrhenius type relationship was calculated. This underestimated the required diffusion coefficient for the strontium in SiC at

1600°C and greater and a high temperature branch was added to correct the release. The revised diffusion coefficient is shown in Figure 5.24.

The values used are for the low temperature branch

$D_{01}$ = $1x10^{-13}$ $cm^2/sec$
$A_1$ = 10000 j/mol

for the high temperature branch

$D_{02}$ = $1.5x10^{10}$ $cm^2/sec$
$A_1$ = 810000 j/mol

All particle breakages are the same as were used in the same cesium calculations.

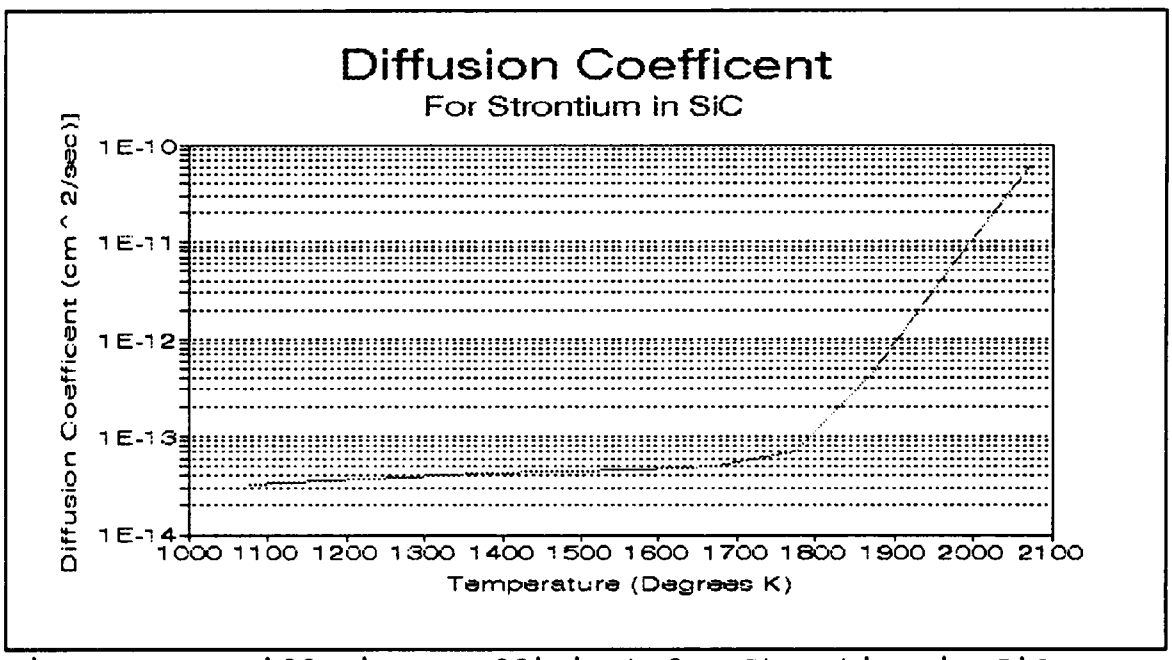



Figure 5.26 Diffusion coefficient for Strontium in SiC.

These values gave good agreement with the strontium release at the early heating stages. Since matrix graphite retains strontium much better than it retains cesium or

krypton, the strontium release is due to diffusive release out of the coated particles. A the defective particle diffusion coefficient was developed using the particle breakage times and numbers from the cesium data.

Since the number of broken particles was already known, it was a simple matter to determine the diffusion coefficients of strontium which are

$D_0$= $6x10^{-3}$ $cm^2/sec$
A = 46900 j/mol

The strontium release is one of the most difficult of releases to model. This is due to the low number of heating tests for which strontium is available.

In addition, since the sorption of strontium on matrix graphite is much stronger than that of cesium, the effects of not having accurate sorption coefficients are much more pronounced.

Figure 5.27 shows the release from SL-P1/6. In this test, it was assumed that there were two broken particles which failed around 4200 hours into the irradiation cycle.

Figure 5.28 shows the fractional release from SL-P1/9. This compact had a significantly higher release than almost any other heating test. To account for this, it was assumed that 20 particles were broken after the end of irradiation, but before the start of heating. The SiC layers of these particles allowed krypton to pass through them.

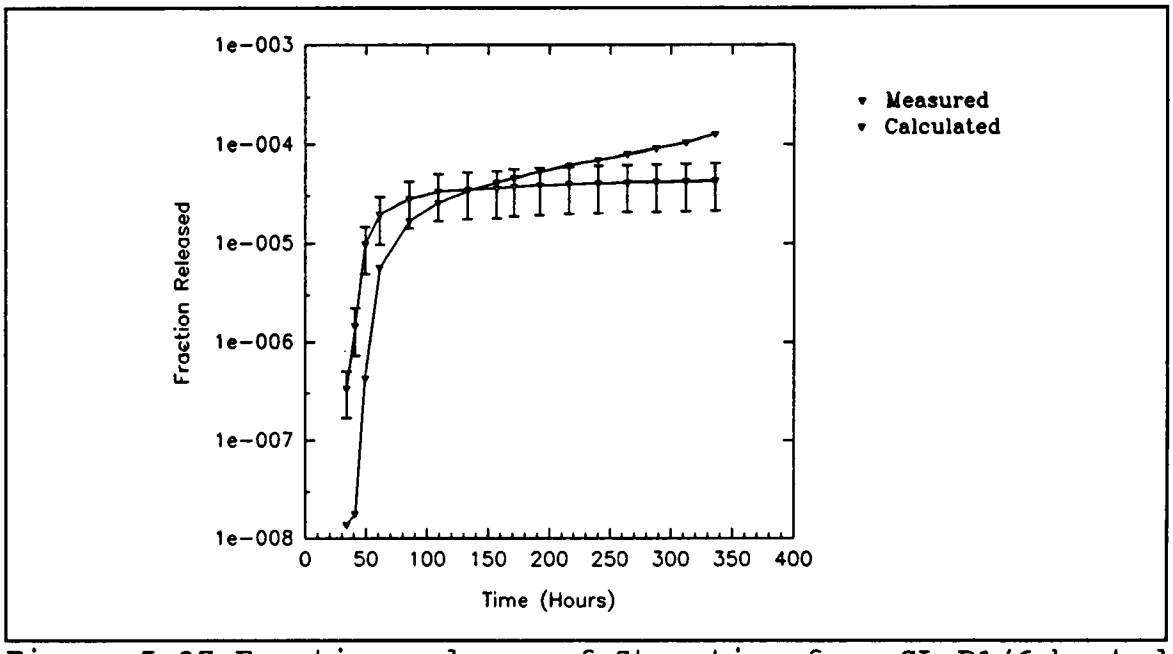



Figure 5.27 Fraction release of Strontium from SL-P1/6 heated at 1600°C.

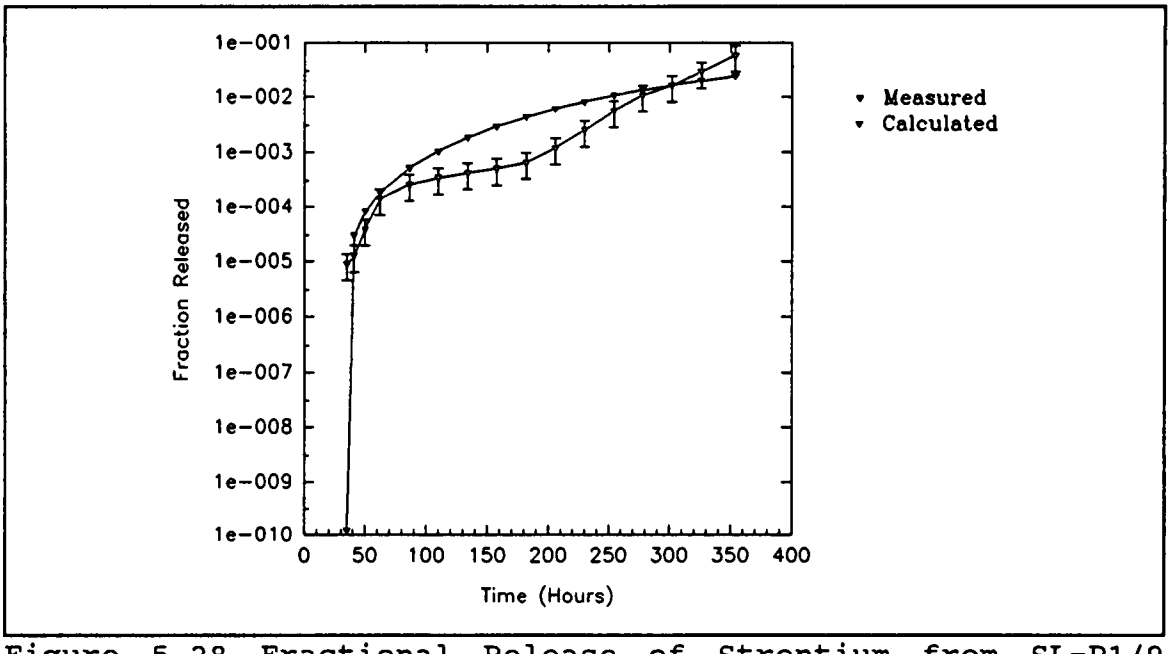



Figure 5.28 Fractional Release of Strontium from SL-P1/9 heated at 1700°C.

Figure 5.29 shows the release from SL-P1/10. This test had significantly higher release at the start of the heat up then did any other element except for SL-P1/9. To account for this release it was assumed that 20 particles were defective from manufacturing. These broken particles may also have been due to fracture when the gas probe was placed into the reactor.

Figure 5.30 shows the release from HFR-P4/1/12. This fuel compact had the lowest burnup of all the HFR-P4 tests. Here, it was assumed that SiC layer in one particle "cracked"e approximately 7700 hours into the irradiation time.

In Figure 5.31, the release from HFR-P4/1/8 is shown. In this test, it was assumed that 1 particle broke at 7700 hours into the irradiation test.

Figure 5.32 shows the release from HFR-P4/2/8. The fuel particles in test HFR-P4/2 had a thicker coating layer (51 $\mu$m) them the rest of the tests. To macth the calculated release to the measured release, it was assumed that two particles had failed around 7750 hours into the irradiation.

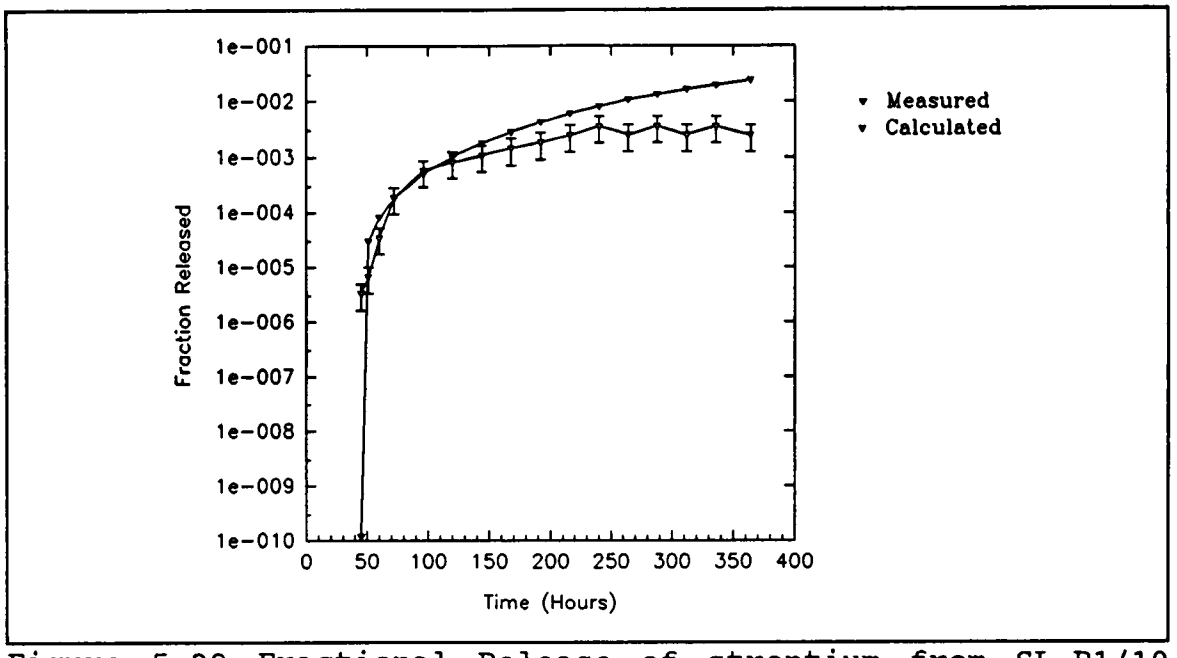



Figure 5.29 Fractional Release of strontium from SL-P1/10 heated at 1700°C.

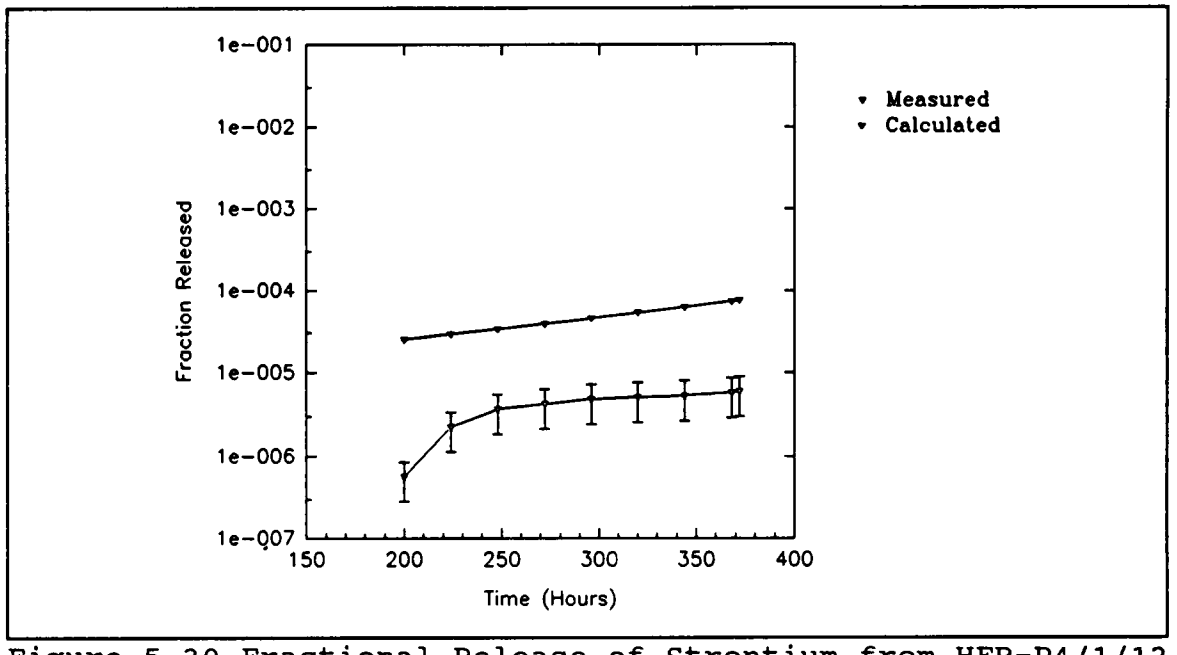



Figure 5.30 Fractional Release of Strontium from HFR-P4/1/12 heated at a 1600°C.

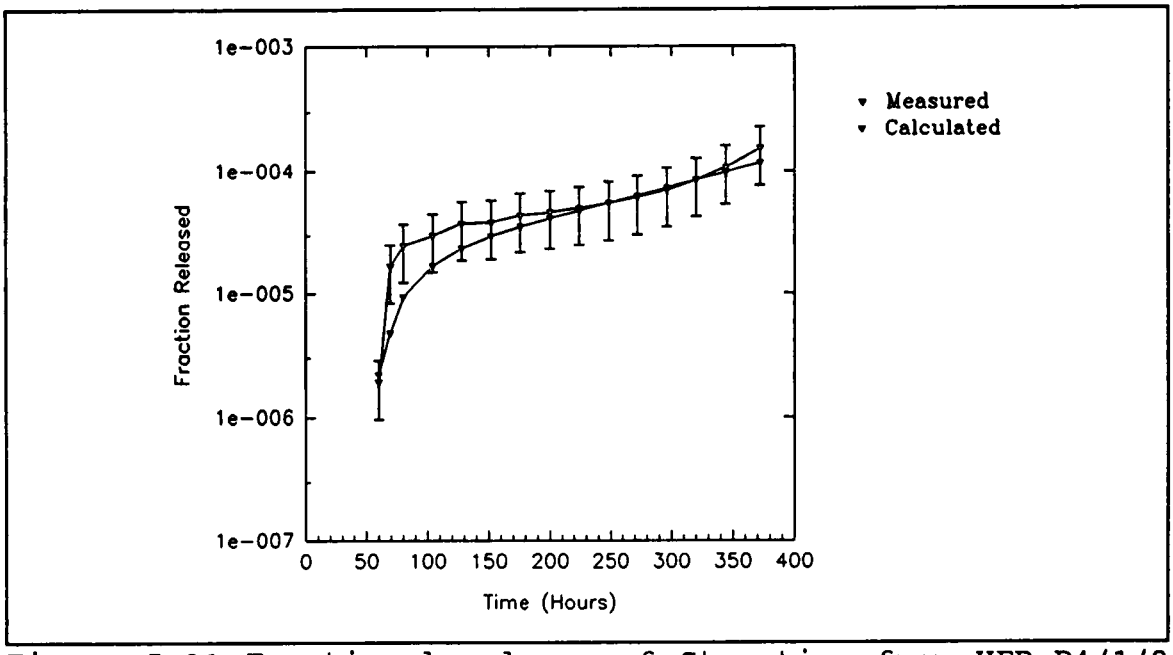



Figure 5.31 Fractional release of Strontium from HFR-P4/1/8 heated at 1600°C.

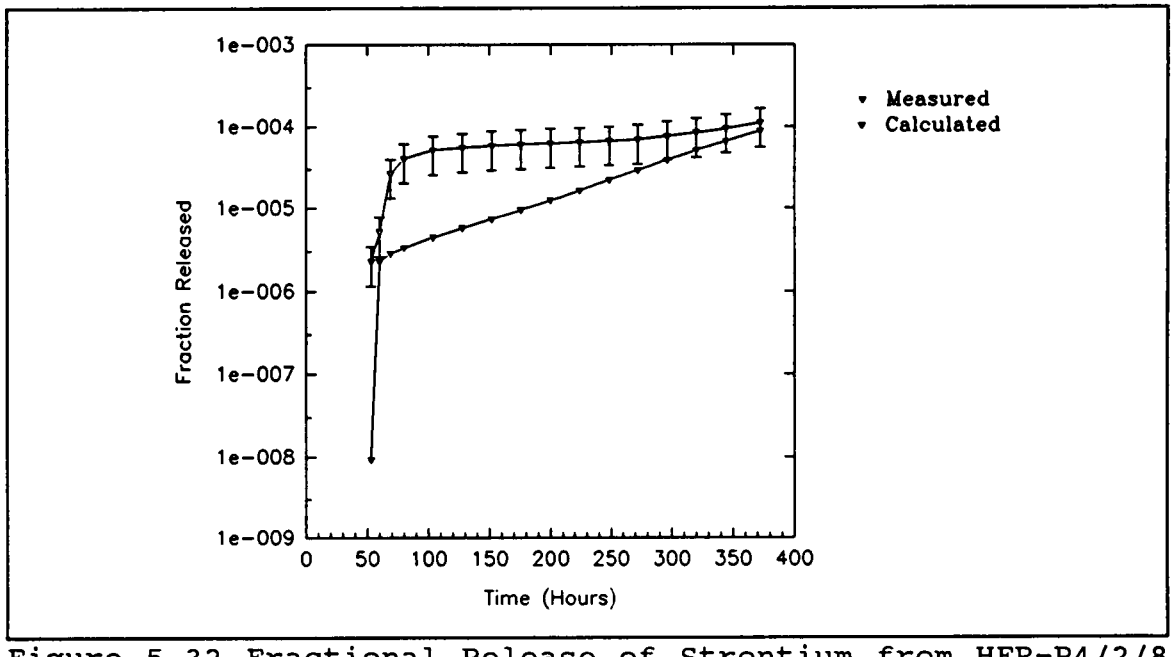



Figure 5.32 Fractional Release of Strontium from HFR-P4/2/8 heated at 1600°C.

# Chapter 6

# Conclusions and Recommendations for Future Work.

## 6.1 Conclusions

With the revised diffusion coefficients and assuming reasonable particle layer breakage, the release of the key fission products can be predicted with good accuracy on a consistant basis. The recommended diffusion coefficients are shown in Table 6.1. For particle breakage, measurements have shown that the assumption of one broken particle is good in most cases. A high assumption of 10 broken particles is sufficient if extreme conservatism is desired.

The releases calculated in this work are in general quite good in comparison with the measured release. The major problem here is the fact that this work attempts to classify the almost infinite way that particles may fail into four distinct categories. This will inevitably leads to errors in the calculated release, however in most cases, however these errors are not beleived to be significant.

## 6.2 Future Work

Important work left to be done is to develop a method to predict the various types of particle layer failure. Since particle failure leads to the predominant amount of release under anticipated accident conditions, it needs to be investigated closely. Failure due to SiC corrosion and

Table 6.1 Recommended Diffusion Parameters

| Kernel | $D_{01}$ $cm^2/sec$ | $A_1$ J/mol | $D_{02}$ $cm^2/sec$ | $A_2$ J/mol | $D_{03}$ $cm^2/sec$ | $A_3$ J/mol | K |
|---|---|---|---|---|---|---|---|
| Cesium | $5.6x10^{-4}$ | $2.09x10^5$ | 5.2 | $3.62x10^5$ | | | |
| Strontium | $2.2x10^1$ | $4.88x10^5$ | | | | | |
| Krypton Irradiation | $1.3x10^{-8}$ | $1.26x10^5$ | | | | | |
| Krypton Accident | $8.75x10^{-11}$ | $5.45x10^3$ | 6002 | $4.8x10^5$ | | | |
| PyC | | | | | | | |
| Cs | $6.3x10^{-4}$ | $2.22x10^5$ | | | | | 0.6/ 1.14 |
| Sr | $2.3x10^{-2}$ | $1.97x10^5$ | | | | | 1.14 /100 |
| Kr | $2.9x10^{-4}$ | $2.91x10^5$ | $2.0x10^9$ | $9.23x10^5$ | | | 0.6 /100 |
| SiC | | | | | | | |
| Cs | $5.5x10^{-10}$ | $1.25x10^5$ | 50 | $5.5x10^5$ | $5.3x10^{-8}$ | $1.14x10^5$ | 5 |
| Sr | $2.5x10^{-15}$ | $1.84x10^5$ | $1.5x10^{10}$ | $8.1x10^5$ | | | 1.51 |
| Kr | $2.66x10^{-8}$ | $2.66x10^5$ | | | | | 1 |
| Matrix Graphite | | | | | | | |
| Cs | $3.6x10^{-1}$ | $1.89x10^5$ | | | | | |
| Strontium | $6.0x10^{-4}$ | $1.x10^5$ | | | | | |
| Krypton | $6x10^{-2}$ | | | | | | |
| Graphite Grains | | | | | | | |
| Krypton | $2.9x10^{-8}$ | $2.1x10^4$ | | | | | 14 |

Note: For Fast Fluence Degradation Constant K in PyC, the first values is used in the inner PyC, the second is used in the outer PyC.

pressure vessel failure may lead to an increase in the release of the fission products. Also, there may be an additional phenomena which could release cesium and strontium from the fuel particle. This could be a result of the fact that the diffusion coefficient for the SiC layer may not be a single value at any one temperature for all particles in a lareg batch of particles at any one time, but instead is a distribution of values. This should be examined.

To check the assumption of the krypton release from the graphite grains, an experiment involving an unfueled fuel element should be conducted. In this experiment, a series of unfueled graphite spheres should be irradiated for around 350 full power days. Heating should then occur following the standard heating pattern up to around 1600°C. The fractional release measured will help validate the fuel model. If this varies significantly, then the krypton released during the heating is coming from another source.

Also, further development needs to be done on the sorption coefficients for A3-27 matrix graphite. The work done here lumps the new sorption coefficient into the diffusion coefficient. Actual values for the diffusion coefficient may be less than the values used here.

Finally, the UT-KFA code should be modified to handle more than one type of failed coating layers in a batch of particles at the same time.

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# APPENDICES

# Code

## 1 Description of Program

The UT-KFA model calculates the release of fission products using the Fickian diffusion equation. The program starts the calculation by spreading a small amount of the fission product throughout the entire fuel element. This keeps the concentration from being zero at any one place and prevents the calculation from "blowing up". The source term is assumed to be proportional to the concentration of $U^{235}$. The source term is a constant and normalized at the end of irradiation. A set of input variables determines the fraction of the $U^{235}$ in the graphite grains, the graphite matrix and the layers of the coated particle. The remainder (generally around >> 99%) is in the coated particle kernel. The kernel and the various layers are divided into several shells and the source term in each layer is divided evenly among each shell. The source is used in the diffusion equation to determine the concentration at each time step through the various layers. The diffusion coefficients for each layer are determined based on the temperature and the fast neutron fluence. Also, the thinning of the SiC layer is determined empirically and the "corroded sections" of the SiC are removed and replace by PyC. Once these special conditions are determined, the fission product concentration is determined by solving the Fickian diffusion equation for all the shells in the coating layers by using Gaussian

elimination for a tri-diagonal matrix. The concentration at the surface of the coated particle is found and used to determine the release into the fuel element matrix material, the release being used as a source term.

For the defective particles, the source term is the same as for the intact coated particles but the diffusion coefficients are significantly different. The calculation for finding the release from the defective particles is the same as for the intact particles.

The diffusive release from the graphite grains is calculated in the same manner. Also, the release due to fission product recoil is calculated for several regions. The recoil release from the kernel is found and added to the source term in the buffer layer. The recoil release from the outer PyC layer is added to the source term in the pore graphite.

Once the total release from the coated particles and the "grains" is found, it is used as a source term to calculate the concentration of the fission products. Again, this is done using Gaussian elimination to solve a tri-diagonal matrix. In addition, the sorption effects in the matrix graphite are considered at this point in the code. The sorption is modeled by either changing the diffusion coefficient of the matrix graphite or by increasing the mass transfer coefficient at the fuel element surface. At this point, the concentration on the fuel element surface is used

to determine the amount released. The overall fractional release of the fission products is then calculated.

## 2 Determination of Concentration

To solve for a radial concentration in a sphere, the Fickian diffusion equation is used. This is

$$\frac{\partial c}{\partial t}=\frac{1}{r^2}\frac{\partial}{\partial}r(Dr^2\frac{\partial c}{\partial r})-\lambda c+\dot{Q} \qquad 1$$

where

D = the diffusion coefficient
c = the concentration
t = time
lambda= the decay constant for the fission product
Q= the source term

The fuel element is divided into N shells, where in each shell the source term and the diffusion coefficient is assumed to be constant in a given time step. So the diffusion equation becomes

$$\frac{\partial c}{\partial t}=D\frac{\partial^2 c}{\partial r^2}+\frac{2D}{r}\frac{\partial c}{\partial r}-\lambda c+\dot{Q} \qquad 2$$

The concentration in the center of the sphere is assumed to represented by a parabola with the formula

$$c=c_N-\frac{2c_{N+1}-c_N}{r_N}(r-r_{N+1})-\frac{c_{N+1}-c_N}{r_N}(r-r_N^2) \qquad 3$$

While the concentration in the outer shells is given by Equation 4

$$c=c_i+\frac{c_{i+1}-c_i}{(r_{i+1}-r_i)}(r-r_i) \qquad 4$$

Substituting this into the diffusion equation gives

$$\frac{\partial c}{\partial t}=\frac{2D}{r}\frac{c_{i+1}-c_i}{r_{i+1}-r_i}-\lambda c+\dot{Q} \qquad 5$$

The time derivative can be approximated by

$$\frac{\partial c}{\partial t}=\frac{1}{\Delta t}(c^{k+1}-c^k) \qquad 6$$

where $c^{k+1}$= the concentration at time k+1

$c^k$= the concentration at time k

Using the above and integrating over the entire volume of the shell gives

$$\frac{1}{\Delta t}\int_{V_i} c^{k+1}-c^k dV=\dot{Q}\int_{V_i} dV-\lambda\int_{V_i} C dV+2D\int_{V_i}\frac{(c_{i+1}-c_i}{(r_{i+1}-r_i)}dV \qquad 7$$

Evaluating the last integral, the equation becomes

$$\frac{1}{\Delta t}\int_{V_i} c^{k+1}-c^k dV=\dot{Q}\int_{V_i} dV-\lambda\int_{V_i} C dV+\frac{4\pi D}{3(r_{i+1}-r_i)}c_{i+1}(r_{i+1}^2-r_i^2)-\frac{4\pi D}{3(r_{i+1}-r_i)}c_i \qquad 8$$

At the surface of the sphere, using convective mass transfer, the boundary condition becomes

$$-D\frac{C_2-C_1}{r_2-r_1}=-b(c_1-c_\infty) \qquad 10$$

where b = the mass transfer coefficient
$c_\infty$= the concentration in the coolant
$c_1$= the concentration in at the fuel element surface

Using these equations, a tri-diagonal matrix can be found and solved using the Gaussian elimination procedure.

## 3 Sorption

The sorption of metallic fission products (cesium and strontium) on graphite has an important effect and must be taken into account. The UT-KFA model takes sorption into account as given below:

At the gas/fuel element interface, the transfer of the fission product is governed by a convective mass transfer equation of the form

$$-D\frac{\partial c}{\partial t}=\beta\,(c_w-c_\infty) \qquad 12$$

$$\beta=\mu\alpha$$

where $\mu$=the mass transfer coefficient
$\alpha$= the sorption factor.
$c_w$= the concentration at the fuel element surface

To model sorption, $\alpha$ is a function of the concentration, when no sorption occurs, $\alpha$ is set equal to 1. If the concentration is low, the Henry regime is used to find $\alpha$

$$\alpha_{Henry} = \frac{1}{T} e^{\left(A_F + \frac{B_F}{T}\right)} \qquad 13$$

where $A_H$ and $B_H$ are the coefficients
T is the temperature

At high concentrations, the sorption enters the Freundlich regime where

$$\alpha_F = \frac{1}{T} e^{\left(A_F + \frac{B_F}{T} + D_F + \frac{E_F}{T}\right) \ln c} \qquad 15$$

where $A_F, B_F, D_F, E_F$ are the sorption coefficients,
c is the concentration at the surface of the fuel element.

Adsorption in the Freudlich regime has the effect of decreasing the mass transfer coefficient at the boundary layer and decreasing the release of fission products from the fuel elements at low temperatures.

If the fission product concentration is high enough that it reaches the Freundlich regime, the diffusion coefficient of the matrix graphite is multiplied by $\alpha_H/\alpha_F$ which decreases the coefficient and retards the release of fission products. In this work, the fission product concentration is generally in the Henry regime.

# VITA

Dan Smith was born in Cleveland, Ohio on August 13, 1965. He graduated from McClintock High School in Tempe, AZ in June of 1983. After a semi-successful stint in the software industry, he received a Bachelor of Science in Nuclear Engineering from Arizona State University in December of 1990. Due to a failing job market, he returned to school, receiving a Masters of Science degree from the University of Tennessee in August of 1994.