Analysis of Fission Gas Release-to-Birth Ratio Data from the AGR Irradiations

Jeffrey J. Einerson, Binh T. Pham, Dawn M. Scates, John T. Maki, and David A. Petti
Idaho National Laboratory
P.O. Box 1625, Idaho Falls, Idaho, 83415, United States of America
phone: 208-526-9798, jeffrey.einerson@inl.gov

Abstract – A series of Advanced Gas Reactor (AGR) irradiation tests is being conducted in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL) in support of development and qualification of tristructural isotropic (TRISO) fuel used in the High Temperature Gas-cooled Reactor (HTGR). Each AGR test consists of multiple independent capsules containing fuel compacts placed in a graphite cylinder shrouded by a steel shell. These capsules are instrumented with thermocouples (TC) embedded in the graphite enabling temperature control. For AGR-1, the first US irradiation of modern TRISO fuel completed in 2009, there were no particle failures detected. For AGR-2, a few exposed kernels existed in the fuel compacts based upon quality control data. For the AGR-3/4 experiment, particle failures in all capsules were expected because of the use of designed-to-fail (DTF) fuel particles whose kernels are identical to the driver fuel kernels and whose coatings are designed to fail under irradiation. The release-rate-to-birth-rate ratio (R/B) for each of krypton and xenon isotopes is calculated from release rates measured by the germanium detectors used in the AGR Fission Product Monitoring (FPM) System installed downstream from each irradiated capsule. Birth rates are calculated based on the fission power in the experiment and fission product generation models. Thus, this R/B is a measure of the ability of fuel particle coating layers and compact matrix to retain fission gas atoms preventing their release into the sweep gas flow. The major factors that govern gaseous diffusion and release processes are found to be fuel material diffusion coefficient, temperature, and isotopic decay constant. To compare the release behavior among the AGR capsules and historic experiments, the R/B per failed particle is used. HTGR designers use this parameter in their fission product behavior models. For the U.S. TRISO fuel, a regression analysis is performed to establish functional relationships between R/B per failed particle of selected noble gas isotopes and temperature. The effect of isotopic half-life is also obtained from the data. To reduce measurement uncertainty of the release rate, the krypton and xenon isotopes selected for regression analysis have a short enough half-life to achieve equilibrium in the capsule, but are also long enough to provide a measureable signal in the FPM detector. The impact of uncertainty of the estimated number of failed particles in each capsule is also examined. This study found that R/B values for AGR test fuel are comparable to R/B obtained in historic tests. The correlation with isotopic half-life is very stable with time indicating no strong influence of burnup on release. The correlation can be used by reactor designers to estimate fission gas release from postulated failed fuel in HTGR cores.

I. INTRODUCTION

The fission product behavior of the tristructural isotropic (TRISO) coated particle fuel is the key safety factor for performance assessment of the high temperature gas-cooled reactors (HTGR). The activity of gaseous fission products, such as krypton and xenon isotopes, in the coolant is a direct indicator of fuel performance. The main sources of release are from the heavy metal contamination in the fuel graphite and from defective or failed coated particles [1]. Also, a few historical irradiations of low enriched uranium carbide/oxide (UCO) TRISO fuel were conducted in the 1980s that had either designed-to-fail (DTF) particles or in-pile failures to study fission gas releases from UCO kernels. Since 2006, three Advanced Gas Reactor (AGR) irradiation tests have been conducted in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL) in support of the development and qualification of the U.S. TRISO fuel used in the HTGR. Each AGR test consisted of multiple independent capsules containing cylindrical fuel compacts placed in a graphite cylinder shrouded by a steel shell and instrumented with thermocouples (TC) embedded in the graphite enabling temperature control. AGR configuration and irradiation conditions are based on prismatic HTGR technology distinguished primarily by the use of helium coolant, a low-power-density ceramic core capable of withstanding very high temperatures, and coated particle fuel [2]. Thus, these tests provide valuable irradiation performance data to support fuel process development, qualify fuel for normal operating conditions, and support development and validation of fuel performance and fission product transport models and codes.

The release-rate-to-birth-rate ratio (R/B) is a measure of the ability of fuel kernels, particle coating layers and compact matrix material to retain fission gas species preventing their release into the sweep gas flow. In the absence of particle failure, this ratio is expected to be very low because standard particles within the specification limits are not expected to contribute to the release of fission products under normal operating conditions. The R/B for each of the krypton and xenon isotopes is calculated from release rates measured by the germanium detectors used in the AGR Fission Product Monitoring (FPM) system installed downstream from each irradiated capsule. Birth rates are calculated based on the fission power in the experiment and fission product generation models. To compare the release behavior among the AGR

capsules and between different experiments, the R/B per failed particle is used for the analysis. This parameter is also used in the fission product behavior models within the HTGR research and development program. This paper describes the regression analysis performed to establish functional relationships between R/B per failed particle of selected noble gas isotopes and temperature for the U.S. TRISO fuel. The effect of isotopic half-life is also obtained from the data.

II. TECHNICAL APPROACH

II.A. Fission Product Gas Release Model

Diffusion of Fission Gas

Diffusion can be defined as the mechanism by which matter is transported into or through matter. *Volume diffusion* describes progressive movement of atoms in a solid with regular lattice structure: by jumping to a neighboring vacancy or interstitial position and by exchanging of lattice positions within the crystal structure [3]. *Grain boundary diffusion* describes movement of atoms along grain surfaces within free spaces between graphite granules of around a few microns in size with voids in between (Figure 1). The rate of diffusion along grain boundaries is much faster than that of diffusion in the bulk crystal. In the case of noble gases (e.g., krypton and xenon) when there is no chemical interaction with the surface of material, an unhindered transport of fission gases occurs from grains to pores. Then a free movement of the diffusing atoms is possible as in a gas. Therefore, pore diffusion can be compared with the diffusion in the gas phase.

For the TRISO coated fuel particle, the fuel kernel itself is the main barrier to fission product releases due to its solid structure (volume diffusion). In fact, under normal operating conditions, the kernel retains more than 95% of the radiologically important short-lived fission gases such as Kr-88. The second barrier to fission product release is the fuel particle coatings. The fuel compact matrix is relatively porous and provides little holdup of the fission gases which are released from the fuel particles (grain boundary diffusion). Therefore, for an intact coated fuel particle, fission product species are contained within its coatings, and releases are insignificant. For a coated fuel particle with a coating failure (e.g., a broken second barrier), some of the fission gas releases from the kernel are able to penetrate through the cracks in coating layers. These fission gases then diffuse through the graphite

matrix with much less obstruction and are released into the sweep gas flow. Therefore, the diffusion coefficient depends mainly on the fuel kernel properties (size and density) and temperatures.

The temperature dependence of the diffusion coefficient is usually expressed by the Arrhenius equation:

$$D = D_0 e^{-\frac{E_a}{RT}}, \quad (1)$$

where D_0 is pre-exponential factor [m^2/s], E_a is diffusion process activation energy [J/mol], R is the universal gas constant [8.3143 J/(mol K)], T is the absolute temperature [K].

To characterize release resulting from the diffusion process within a solid spherical material, such as a fuel kernel, the reduced diffusion coefficient D' (s^{-1}) is used. It is defined as the diffusion coefficient D (m^2/s) divided by the squared “equivalent sphere” diffusion radius, a (m):

$$D' = \frac{D}{a^2} \quad (2)$$

Fig. 1: The porous structure of graphite.

Radioactive Decay of Fission Gas

Right after birth in a fuel kernel, the isotopic atoms begin depleting over time due to the radioactive decay process. This process allows the kernel to retain most of the fission product species preventing their releases into the sweep gas flow. The radioactive gaseous depletion rate (or decay constant) is characterized by isotope half-life ($\tau_{1/2}$), during which half of the activated atoms are depleted. The decay constant is given by:

$$\lambda = \frac{\ln 2}{\tau_{1/2}}, \quad (3)$$

R/B Model

Two governing processes, diffusion and decay, for fission product species transportation from the kernel to sweep gas flow are described in the previous two subsections. R/B for a bare kernel is a function of the kernel diffusion coefficient, the decay constants of the fission product isotopes, and

the surface to volume ratio, $(3/a)$, of a sphere equivalent to the representative grain of fuel with radius a (or kernel radius assuming solid sphere kernel). For the equilibrium condition when isotope production in and release from the kernel reach a steady state, this function is expressed as [1]:

$$R/B = \frac{3}{x} \left(\coth x - \frac{1}{x} \right), \quad (4)$$

where $x = \left(\frac{\lambda}{D'} \right)^{0.5}$; λ is decay constant (s^{-1}); D' is the reduced diffusion coefficient (s^{-1}).

For short-lived isotopes when $x \gg 1$ then:

$$\frac{R}{B} \approx \frac{3}{x} = 3 \sqrt{\frac{D}{\lambda a^2}} \quad (5)$$

Given experiment specific diffusion coefficient D and radius a , Equation 5 shows the characteristic feature of the $1/\sqrt{\lambda}$ dependence of release for short-lived isotopes. However, when the kernel is surrounded by fuel particle coatings, the second barrier to fission product release, some of the releases from the kernel actually terminate in the remaining coatings. This is known as the recoil effect, which will lower the power of D/λ to less than 0.5. In previous experiments, these values are in the range between 0.1 and 0.5.

Substituting the diffusion coefficient of Equation 1 into Equation 5, the R/B for a TRISO coated fuel particle can be expressed as a function of fuel temperature and decay constants as:

$$\frac{R}{B} \approx \frac{3}{x} = 3 \sqrt[n]{\frac{D_0 e^{-\frac{E_a}{RT}}}{\lambda a^2}}, \quad (6)$$

where instead of using the theoretical value of 0.5, a variable n is introduced in the power of D/λ to account for the dependence of release on particle coatings.

Regression Fitting Function

Equation 6 will be used as a basis for the regression analysis to establish the functional relationships of R/B per failed particle for krypton and xenon isotopes with decay constants and temperature for the TRISO coated fuel. The fission product release model can be derived for R/B per failed particle (R_p) by taking the natural logarithm of Equation 6 and replacing R/B with R_p as:

$$\ln R_p = n \ln \frac{1}{\lambda} + \frac{B}{T} + C, \quad (7)$$

where B is a fuel particle specific constant representing diffusion coefficient dependency on

temperature, and C is an irradiation specific constant. This revision helps transform non-linear Equation 6 into the linear form in Equation 7. The regression analysis is performed to best fit this equation to R/B data obtained from AGR irradiations to estimate parameters n , B , and C .

II.B. Fuel Performance Irradiation Data

The R/B for a noble gas isotope is used to characterize the performance of the modern U.S. fabricated TRISO coated fuel particles. The summary of fission product monitoring system and release data determination for the AGR-1 experiment is given in Scates' 2010 conference paper [4]. This procedure is also used for all AGR irradiations. Each AGR capsule is continuously and independently monitored by a gross radiation detector and a spectrometer detector in the FPM system installed downstream from the capsule sweep gas line as shown in Figure 2. This system provides fission gas release rate in individual capsules at 8-hour intervals. It also helps detect particle failure under irradiation.

Fig. 2: Gross radiation monitor and spectrometer detector for one AGR capsule.

R/B per failed particle

The R/B per failed particle is used for this analysis to compare the release behavior among the AGR capsules and historical experiments. For each AGR capsule, the R/B per failed particle for a radionuclide isotope can be calculated as:

$$R_p = \frac{R}{B \cdot N_f} \quad , \quad (8)$$

where: R_p is R/B per failed particle; R is the isotope release rate (atom/sec) measured separately for each capsule; B is the isotope birthrate (atom/sec) calculated for each capsule; and N_f is the total number of failed particles in the capsule. The necessary data for determination of R/B per failed particle are birthrate, release rate, and number of failed particles, which will be discussed below.

Birthrate

By definition, birthrate of an isotope is the rate of entire production for a specific isotopic atom, even if it is immediately lost to transmutation or decay. The isotope birthrates in each capsule are calculated from inventory data supplied using JMOCUP code that links neutronic data computed in MCNP (for Monte Carlo N-Particle) to inventory data computed in ORIGEN2.2 [5]. The birthrates calculation used an ORIGEN2.2 depletion model assuming no transmutation and decay for krypton and xenon isotopes. The JMOCUP simulation code uses daily averaged values of the ATR operating parameter inputs, such as lobe powers and control shim cylinder positions to compute daily birthrates.

Release rate

Sweep gas carries released fission product gases from the capsules to the detector system under normal conditions with a transit time of about 150 seconds. Gas flow passes through a high purity germanium (HPGe) detector gamma-ray spectrometer system. The continuous gamma-ray spectrum measurements from the HPGe detectors were used to compute the release activities of several isotopes of krypton and xenon. These measured activities are converted to capsule fission release rates by appropriate correction to account for decay that occurred during transport from the capsules to the detectors. Actual transport times were calculated from outlet gas flow rates and the capsule-specific volumes through which samples flow to reach the respective monitoring detector. This conversion formula was derived under the assumption that the equilibrium release conditions were established [4].

To reduce measurement uncertainty of the release rate, the krypton and xenon isotopes selected for regression analysis have a short enough half-life to reach equilibrium in the capsule, but are also long enough to provide a measureable signal in the FPM system detector. These isotopes are Kr-85m, Kr-87, Kr-88, Xe-135, Xe-137, and Xe-138. In addition, R/B data obtained during ATR start-ups and the first 10 days of an irradiation cycle are excluded to assure only equilibrium data are used.

Number of failed particles

For AGR capsules, the total number of failed particles has three sources: (1) defective exposed

kernels, (2) heavy metal contamination, and (3) particle coating failures under irradiation (in-pile failures). The number of initially exposed kernels and heavy metal contamination can be estimated based on failure fractions provided from the fabrication quality control data. The in-pile failure among qualified fuel particles is not likely.

The in-pile particle failures in a capsule are detected using the independent capsule-specific FPM system. The sweep gas carrying fission gaseous species passes in front of the sodium iodide [NaI(Tl)] scintillation detector-based gross radiation monitor. This gross radiation detector has a capacity of detecting every fuel particle failure up to and including the first 250 failures from each capsule. These fuel particle failures are indicated by a rapid rise and drop, or spike, in the temporal profile of the measured gross gamma count rate as shown in Figure 3. Such spikes are a result of a sudden release of stored fission product inventory inside a just-failed particle. The visual interpretation of the gross gamma data was used to count the number and to record the time of particle failures.

Figure 3 captures a perfect picture of a single particle failure that occurred in AGR-3/4 Capsule 10 on January 19, 2012. Before the failure occurs, the baseline count rate (B_{bf}) is equal to 3,239 counts per second (cps). At time of failure, the rapid rise (peaking at 13,374 cps) and fall in activity were clearly observed for the event. After the failure, the new baseline count rate (B_{af}) increases to 3,788 cps (549 cps increase relative to before failure), which represents additional release from this failed particle. The release stabilizes after a particle failure in about 4 minutes, which is consistent with what was observed during the NPR-1A experiment [6].

Fig. 3: "Classic" particle failure.

III. TRISO FUEL IRRADIATION DATA

The analysis presented here includes data from the AGR-2 and AGR-3/4 irradiations and a few historical irradiations of low-enriched UCO TRISO fuel conducted in the 1980s. These tests had either DTF particles or in-pile failures from which R/B

data on fission gas releases from UCO kernels could be obtained.

III.A. AGR-2 Irradiation

AGR-2 is the second in the series of planned experiments to test TRISO coated, low-enriched uranium (LEU) fuel [7]. This experiment is intended to demonstrate the performance of uranium oxycarbide (UCO) and uranium dioxide (UO_2) fuel produced in a large (15 cm/6-inch) coater [2]. AGR-2 irradiation was completed in October 2013 for 559.2 effective full power days (EFPDs). Figure 4 shows the sketch of a capsule in the AGR-2 irradiation test train (the same as for AGR-1). Each UCO capsule contains about 38,000 particles and each UO_2 capsule contains approximately 18,500 particles.

Even without DTF fuel particles included in capsules, the AGR-2 irradiation can also provide data for R/B per failed particle because exposed kernels existed in the test fuel that was irradiated. Quality control data [8] indicate that the 95% confidence defect fraction for an exposed kernel is $2.5E-05$ for UCO fuel and $3.2E-5$ for UO_2 fuel. Thus, on a statistical basis there should be one exposed kernel in each UCO capsule and about one-half exposed kernel in each UO_2 capsule. Because exposed kernels should be a whole number, this failure in UO_2 capsules is problematic. Therefore, only R/B data from three UCO capsules (2, 5, and 6) are used in this analysis.

Fig. 4: Sketch of an AGR-2 capsule.

Calculated fuel temperatures

A daily as-run thermal analysis was performed for AGR-2 capsules using the commercial finite element heat transfer code ABAQUS [9]. This thermal model was improved by incorporating gas gaps changing with irradiation time due to graphite shrinkage caused by neutron damage. The gas gap

changes were estimated based on dimensional measurements of AGR-1 irradiated compacts and graphite holders during post-irradiation examination (PIE). The calculated volume average fuel temperature in each AGR-2 capsule is used for this analysis because the locations of failed particles are unknown. Figure 5 shows the calculated fuel temperature in three UCO capsules as a function of EFPDs for time periods when the R/B data were used in the analysis.

Fig. 5: Volume-average fuel temperatures for AGR-2 UCO capsules.

Measured R/B data

Figure 6 shows the temporal profile of R/B data for Kr-85m, Kr-88, and Xe-138 measured from UCO Capsules 2, 5 and 6 during Cycles 148A and 148B. Beyond Cycle 148B, no R/B data were used because of flow issues during the next two cycles, 149A and 149B, and then cross-talk between capsules beginning in Cycle 150B. The cross-talk was probably caused by failure of the refractory gas lines in the experiment during experiment handling between ATR reactor cycles. Cross-talk allows the fission gas atoms from one capsule to get into the FPM detector for another capsule. Therefore, R/B calculated using measurements from one detector are not necessarily representative of the release from the corresponding capsule. Also, R/B data from the first cycle, 147A, were excluded to allow time for releases to reach their equilibrium state.

Fig. 6: R/B for AGR-2 UCO capsules.

In general, the R/B data (Figure 6) in all three capsules agreed well with calculated fuel temperatures (Figure 5) during both Cycles 148A and 148B, so most of the data from these two cycles were used. For Capsules 5 and 6, R/B data and calculated fuel temperatures are fairly stable and correlate well with each other.

For Capsule 2, decreases were observed during Cycle 148B (bottom panel in Figure 6) when the capsule was put on all helium to deal with operational problems at ATR. Then, R/B in Capsule 2 gradually increased for the rest of Cycle 148B in response to increasing fuel temperature (red dots in Figure 5). The broad range of Capsule 2 fuel temperatures between 1000°C and 1400°C during cycle 148B provides valuable data for establishing the correlation between R/B per failed particle and fuel temperature.

Number of failed particles

Fission gas release in an AGR-2 capsule comes from only two sources: exposed kernels and high heavy metal contamination. (There were no in-pile particle failures detected.) The number of exposed kernels can be estimated from the defect fraction for exposed kernels of $2.5E-05$ for a UCO capsule, which corresponds to one defective particle per capsule. Because fission gas from heavy metal contamination also contributes to the measured release, this effect must be factored out to establish a true estimate of the release from exposed kernels. The heavy metal contamination is $3.9E-06$ based on the fraction of free uranium from the UCO fuel batch estimated from compacts that had no exposed kernels.

Thus, the total measured R/B for a capsule without in-pile particle failure is given by:

$$R_{Total} = N * (R_{HMC} * f_{HMC} + R_p * f_{EK}) , (9)$$

where: R_{Total} is total R/B measured for a capsule, N is total number of fuel particles in a capsule, R_{HMC} is R/B for release from heavy metal contamination, f_{HMC} is heavy metal contamination fraction without exposed kernels (g leached U/g U in compact), R_p is R/B release from one exposed kernel (or R/B per failed particle), and f_{EK} is the exposed kernel fraction (g exposed U/g U in compact).

In addition, prior measurements at General Atomics [10] have indicated that the release from finely dispersed heavy metal contamination is ten times that from an exposed dense kernel for the same amount of uranium ($R_{HMC} = 10R_p$). Then Equation 8 can be expressed as:

$$R_{Total} = R_p * N * (10 * f_{HMC} + f_{EK}) \quad (10)$$

As a result, the total number of exposed kernels (N_{CF}) can be expressed as the number of exposed kernels from the quality control data and an “equivalent” number of exposed kernels due to heavy metal contamination:

$$N_{CF} = N * (10 * f_{HMC} + f_{EK}) \quad (11)$$

Equation 11 corresponds effectively to about 2.5 equivalent exposed kernels in each UCO capsule. Detailed examination of the R/B data suggest that the changes observed in capsules 2, 5 and 6 are related to temperature and no additional in-pile failures were observed. The corresponding R/B per failed particle (R_p) was calculated to account for an exposed kernel and heavy metal contamination in three AGR-2 UCO capsules as:

$$R_p = R_{Total}/2.5 \quad (12)$$

III.B. AGR-3/4 Irradiation

AGR-3/4 is the third and fourth in this series of planned experiments to test TRISO coated LEU fuel and the irradiation was completed in April 2014 for 369.1 EFPDs. This experiment is intended to support the refinement of fission product transport models and to assess the effects of sweep gas impurities on fuel performance and fission product transport [11]. Therefore, together with driver-coated fuel particles, a number of UCO DTF fuel particles were embedded along the vertical centerline in each compact to provide data on fission product diffusivity in fuel kernels and sorptivity and diffusivity in compact matrix and graphite materials. To accomplish the experiment objectives, this irradiation was located in the best ATR position to achieve significant end-of-irradiation conditions in terms of peak burnup (15.3%) and maximum fast neutron fluence accumulation (5.3×10^{21} n/cm²) for a larger amount of irradiated fuel contained in 12 capsules and for minimal irradiation time. Four fuel compacts in

each independently controlled and monitored capsule contain about 7,500 conventional UCO driver fuel coated particles similar to the baseline fuel used in the AGR-1 experiment and 80 DTF UCO fuel particles. The design of the AGR-3/4 test is shown in Figure 7.

Fig. 7: Axial schematic of AGR-3/4 capsules.

Calculated fuel temperatures

A daily as-run thermal analysis was also performed for AGR-3/4 capsules using the commercial finite element heat transfer code ABAQUS [12]. This thermal model incorporates the gas gaps changing with irradiation time due to graphite shrinkage caused by neutron damage. The model of gas gap change was rigorously calibrated to achieve the best fit between predicted temperatures at TC locations and TC readings over the entire irradiation. Contrary to AGR-2, the calculated peak fuel temperature in each AGR-3/4 capsule is used for R/B data analysis because the DTF fuel particles are located along the axial centerline of fuel compacts, which are exposed to the highest temperature in each capsule. Figure 8 shows calculated peak fuel temperature in 12 capsules as a function of EFPDs for the entire irradiation (filtered data not used in analysis are not included). The calculated peak fuel temperatures range broadly from 850°C (lowest temperature in Capsule 12) to 1425°C (highest temperature in Capsule 7). This range provides adequate data to establish the relationship between R/B per failed particle and temperature.

Fig. 8: AGR-3/4 capsule peak fuel temperatures.

Measured R/B data

Figure 9 shows daily averaged R/B per failed particle for Kr-85m, Kr-88, and Xe-138 measured in Capsules 1 through 4 as a function of EFPDs throughout AGR-3/4 irradiation (filtered data not used in analysis are not included). The uncertainties of R/B data for selected krypton and xenon isotopes are around 6%. It is essential that equilibrium is achieved for the measured release rates at the downstream detector to reflect the true release rate in the capsule. Thus, only R/B data during fission product release equilibrium are used for this analysis. The R/B data excluded from regression analysis are: (1) the first cycle, (2) the first 10 days after each reactor startup following a long cycle outage, and (3) the first 3 days after reactor restart following a short scram. Additionally, R/B data with uncertainty greater than 50% and R/B data acquired for too short intervals (20 minute instead of 8 hour interval) are also excluded. In spite of this R/B data filtering, there remain more than enough data for use in this analysis.

Fig. 9: R/B per failed particle for AGR-3/4 Capsules 1 through 4.

During the first cycle, 151A, about 60% of all DTF fuel particles failed by the cycle end. The uncertainty of daily failed particle counts is large because failure counts were made on a weekly basis. In addition, equilibrium may not have been achieved due to multiple particle failures throughout the cycle and rapid changes of material thermal properties due to fast fluence exposure. This causes large variation of R/B per failed particle during the first cycle in all capsules. At the same time, the calculated peak fuel temperatures are fairly constant. Consequently, R/B data during the first cycle are excluded from analysis.

During reactor outages, test fuel temperature drops to around 30°C. Subsequently, the diffusion coefficients of the fuel kernel and surrounding material also substantially decrease. Thus, fission product releases during this time will accumulate within fuel particles. After reactor startup, fuel temperature rapidly increases to more than 1000°C allowing accumulated fission products to diffuse out of failed particles into the sweep gas flow. The longer outage leads to larger fission product accumulation; therefore, it leads to longer time for the capsule to reach release equilibrium. As a result, 10 days after long outages between reactor cycles are excluded and 3 days after short outages within a reactor cycle are excluded.

Number of failed particles

In AGR-3/4 there were no exposed kernels and the heavy metal contamination was very low. Also, based on AGR-1 results, it is reasonable to assume that there is no in-pile particle failure among the qualified driver fuel particles. Thus, the total fuel particle failures in each AGR-3/4 capsule is capped at a maximum of 80 failures.

The in-pile particle failures in a capsule are detected using the independent capsule-specific NaI(Tl) total radiation detector. Even though the detector is sensitive to each fuel particle failure, visually counting the exact number of failed particles during AGR-3/4 irradiation was a challenging task due to multiple failures of DTF fuel particles occurring at the same time, partial failures, and high background activity of releases from already failed particles. Figure 10 shows examples of multiple failure gross gamma spectra. The top plot shows an example of a “clean” multiple failure gross gamma spectra. This spectra output is from Capsule 7 collected on December 29, 2011, only 16 days after the start of AGR-3/4 irradiation when DTF particles began to fail. Each failure event is clearly defined by a rapid rise/fall activity with a distinct increase in baseline count rate after each event. The area in the red oval and its

blow-up plot on the top left present one such instance where one failure occurred right after another. This double failure is further confirmed by the large step increase in baseline count rate after the event. During this 8 hour period 5 particle failures clearly occurred.

Fig. 10: Examples of multiple failure gross gamma spectra: (a) – “clean” multiple failure and (b) – “unclean” multiple failure.

The bottom plot in Figure 10 shows an example of an “unclean” multiple failure gross gamma spectra. This spectra output is from Capsule 9 collected on December 10, 2013, near the end of AGR-3/4 irradiations (only one more cycle left), when most DTF particles had already failed. There are multiple “peaks” present in this output profile. As the baseline increases over time and more fuel has failed it becomes difficult to distinguish a failure from a “burp.” By closely reviewing the baseline of the 8-hour period here, it can be argued that there are between one and three failure events. Note that it is also possible that a partial failure occurred here or that a small pocket of trapped gas was released due to kernel structuring with burnup at high temperature.

The challenges in the failure counting process lead to high uncertainty of total number of particle failures in some capsules. Therefore, each inspection period provides three estimates of failure counts: best-estimate, maximum, and minimum as shown in Figure 11. For Capsule 1 (plots on the top of Figure 11) the three failure estimates are quite different from each other indicating high counting uncertainty. By contrast, for Capsule 9 (plots on the bottom of Figure 11) the three failure estimates are very similar indicating low counting uncertainty or high confidence about the number particle failures.

Fig. 11: Particle failures in Capsules 1 and 9.

Figure 12 shows the best estimated number of failed fuel particles as a function of EFPDs for 12 AGR-3/4 capsules. For most capsules, fuel failures occurred soon after start of irradiation in the first cycle (e.g., first 55 EFPDs). For a few other capsules (e.g., Capsules 2 and 3), fuel failures occurred throughout irradiation. In three capsules (2, 3, and 9) the total failure counts are higher than the number of DTFs, and are capped at 80.

Fig. 12: AGR-3/4 best-estimate failure counts.

III.C. Historical Irradiations

There are a few historical irradiations of low enriched UCO TRISO fuel that had either DTF particles or had in-pile failures that can provide additional data for comparison to the AGR-3/4 and AGR-2 data. In many cases, these experiments were moisture injection experiments to study hydrolysis effects on fission gas release from UCO kernels, but the R/B prior to moisture injection provides valuable data for comparison here. Table 1 summarizes the results from four key irradiations: HRB-17/18 [13], COMEDIE-BD1 [14], HFR-B1 [15], and HRB-21 [16]. Results are plotted using the average fuel temperature unless noted otherwise.

Table 1: R/B per failed particle for Kr-85m from four key historic irradiations.

Experiment	Fuel Temperature (°C)	R/B per Failed Particle
HRB-17 & 18 ^a	840	0.004
COMEDIE	1160	0.01
HFR-B1	1140	0.008
Capsule B ^b	1015	0.003
	912	0.002
HRB-21 ^c	950 (max) / 932	0.009
		0.007

- a. Results were identical for two independent capsules run at the same time in different locations in the reactor [13].
b. 125 °C added to TC temperatures to get fuel temperatures as recommended in report (see Table 6.4.33 in [15]).
c. First R/B value represents value for first particle failure (assumed to be at the maximum temperature) and second value is an average based on total number of estimated failures [16].

IV. ANALYSIS RESULTS AND DISCUSSION

IV.A. R/B correlation with decay constants

As stated in Section II.B, the variable n for TRISO coated fuel particles can range between 0.1 and 0.5 due to the recoil effect of the particle coatings. Also, a constant fuel temperature leads to a constant reciprocal fuel temperature term, the second term in Equation 7, and so Equation 7 can be expressed as:

$$\ln R_p = n \ln \frac{1}{\lambda} + C' \quad , \quad (13)$$

where C' is a new constant including the reciprocal fuel temperature term. Therefore, the n value in Equation 7 for AGR test fuel can be estimated for each day of irradiation when the fuel temperature is considered to be constant. According to Equation 13, n represents the slope between $\ln R_p$ and $\ln \frac{1}{\lambda}$ for isotopes of each gas element.

Figure 13 shows daily n values for krypton and xenon isotopes for Capsules 9 through 12 as a function of EFPDs over the entire irradiation (they

are similar for all other capsules). In general, n values for krypton isotopes (blue dots) and for xenon isotopes (red dots) are comparable and equal about 0.3. Except for the first cycle, the n values are fairly constant over the entire irradiation indicating insignificant burnup influence on fission product release from AGR-3/4 fuel particles, which achieved a peak burnup of 15.3%. Similar behavior of n values is also observed for AGR-2 irradiation.

Fig. 13: Krypton and xenon n values as function of EFPD for AGR-3/4 Capsules 1 through 4.

IV.B. Regression Analysis Results

Regression is used to fit Equation 7 to daily averaged R/B per failed particle data obtained from 12 AGR-3/4 capsules during the entire irradiation. To study the impact of the uncertainty of the failure estimates on the release relationship with fuel temperature and decay constants, the regressions were performed for three sets of R/B per failed particle: best-estimate, maximum, and minimum failure estimates. Figure 14 shows AGR-3/4 R/B per failed particle and their fitted function of reciprocal peak fuel temperature for Kr-85m using best-estimated (blue), maximum (red) and minimum (green) failure counts. Table 2 presents parameter estimates of regression functions separately for krypton and xenon isotopes. The n value for AGR fuel particles is equal to approximately 0.3 for both krypton and xenon isotopes, which is consistent with the calculated values plotted in Figure 13. As expected, the uncertainty of the failure counts does not affect n because it is defined mainly by fuel particle material properties. Additionally, estimates for the B parameter are similar for krypton and xenon isotopes. However, estimates for Parameter C are much lower for xenon isotopes indicating their lower release.

Table 2: Parameter estimates for AGR-3/4 R/B data.

Isotopes	n	B	C
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Best-estimate number of failures			
Kr-85m, Kr-87, Kr-88	0.327	-9118	-1.11
Xe-135, Xe-137, Xe-138	0.302	-8657	-2.19
Maximum number of failures			
Kr-85m, Kr-87, Kr-88	0.327	-9580	-1.02
Xe-135, Xe-137, Xe-138	0.302	-9119	-2.10
Minimum number of failures			
Kr-85m, Kr-87, Kr-88	0.326	-9548	-0.531
Xe-135, Xe-137, Xe-138	0.302	-9086	-1.61

Fig. 14: AGR-3/4 R/B per failed particle and their fitted function of reciprocal peak fuel temperature for Kr-85m using best-estimated (blue), maximum (red) and minimum (green) failure counts.

Despite a large variation of the release data, there is a clear trend of temperature influence on R/B per failed particle (Figure 14). However, the large variation of R/B for reciprocal temperature between 7.0×10^{-4} and 7.5×10^{-4} (R/B in Capsules 2, 4, 5, 6, and 9 with peak fuel temperature around 1100°C) and unexpected higher R/B in Capsule 12 with the lowest peak fuel temperature lead to large 95% bounds (light blue area) around the fitted line.

The regression analysis results presented in Table 2 and Figure 14 indicate insignificant impact of the failure count uncertainty on the regression fitting parameters because of the following observations:

- (1) The parameter estimates are similar for all three data sets of R/B per failed particle,
- (2) Three fitted lines (blue, red and green) are fairly parallel to each other with a small shift
- (3) The shaded area representing 95% confidence bounds on the fitted line using best-estimate failure counts covers most of the three sets of R/B per failed particle.

Thus, the wide variation of the large quantity of R/B data used in the fitting process makes variation due to failure count uncertainty insignificant. However, the actual number of particle failures can be

determined accurately during PIE and will help narrow the model prediction uncertainty.

IV.C. Comparison of R/B data

AGR-2 and AGR-3/4 irradiations

Figure 15 shows the Kr-85m R/B per failed particle data obtained from AGR-2 (red dots) and AGR-3/4 (blue dots) irradiations.

Fig. 15: R/B per failed particle data for AGR-2 (red dots) and AGR-3/4 (blue dots) irradiations.

The fact that AGR-2 R/B data lie completely within the 95% bounds of the fitted line for AGR-3/4 R/B data indicates excellent consistency between these two AGR irradiations. Furthermore, AGR-2 R/B data filled in nicely the gap in reciprocal fuel temperature between 7.4×10^{-4} and 7.9×10^{-4} , where there are no AGR-3/4 R/B data. Consistency is also shown by the decrease in R/B data in the reciprocal temperature range of 6.8×10^{-4} to 7.4×10^{-4} for both AGR-2 and AGR-3/4.

AGR and historical irradiations

Because of the solid agreement of R/B data obtained from AGR-2 and AGR-3/4 irradiations, these data were combined in a regression analysis to best-fit Equation 7. The parameter estimates are presented in Table 3. The n , B , and C estimates are similar to the estimates using only AGR-3/4 R/B data. This confirms the consistency between R/B data obtained from these two AGR irradiations.

Table 3. Parameter estimates for combined AGR-2 and AGR-3/4 R/B data.

Isotopes	n	B	C
Kr-85m, Kr-87, Kr-88	0.324	-9370	-0.904

Figure 16 shows R/B per failed particle data obtained from the two AGR irradiations, the four historical irradiations presented earlier, and two historical models. The German model is discussed in

reference [1]. The model developed by General Atomics (GA) is discussed in Richards' 1994 report [14]. The fact that R/B per failed particle data from all historical irradiations lie well within the 95% bounds of the fitted line for the R/B data of AGR irradiations indicates good performance of AGR experiment fuel particles.

Fig. 16: R/B per failed particle data for AGR irradiations, historical irradiations, and models (the blue shaded area is 95% bounds of the fitted line).

The German model (purple dashed line) is at the upper end of the AGR data range and the slope is very similar. The General Atomics (GA) model (orange dashed line) lies above the AGR fitted line and below the German model.

V. CONCLUSION

With embedded DTF fuel particles, AGR-3/4 irradiation provides invaluable R/B data for studying the fission product behavior of TRISO coated UCO fuel. AGR-3/4 fuel particles were irradiated for 369.1 EFPDs and reached a peak burnup of 15.3%, a maximum fast neutron fluence accumulation of 5.3×10^{21} n/cm², and a peak fuel temperature ranging between 850°C and 1450°C. Even without DTF fuel particles included in capsules, the AGR-2 irradiation provides additional data for R/B per failed particle because exposed kernels existed in the test fuel that was irradiated. The large amount of R/B data obtained for extended irradiation periods allows assessment of the effect of isotopic decay constants on fission product releases. The R/B correlation with isotopic decay constant is very stable with time indicating no evidence of an effect of burnup on release.

AGR irradiations provide sufficient data to perform the regression analyses for establishing functional relationships of R/B per failed particle with noble gas isotope decay constants and

temperature for the U.S. TRISO UCO fuel. This R/B correlation can be used by reactor designers to estimate fission gas release from postulated failed fuel in HTGR cores, which is the key safety factor for fuel performance assessment.

This study found that R/B data for AGR-2 and AGR-3/4 test fuel are consistent with each other and also comparable to R/B obtained in historic tests. The German and GA models are conservative as they predict higher R/B per failed particle than the prediction of the model based on AGR-2 and AGR-3/4 data—the German model for the entire temperature range and the GA model for lower temperatures.

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