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Fission Product Monitoring and Release Data for the Advanced Gas Reactor-1 Experiment

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Abstract – *The AGR-1 experiment is a fueled multiple-capsule irradiation experiment that was conducted in the Idaho National Laboratory (INL)'s Advanced Test Reactor (ATR) from December 26, 2006, until November 6, 2009. The experiment was the first in a series of measurements in support of the Next Generation Nuclear Plant (NGNP) Advanced Gas Reactor (AGR) Fuel Development and Qualification program An important measure of the fuel performance of these tests is the quantification of the fission product releases over the duration of the experiment. A fission product monitoring system (FPMS) was developed and implemented to monitor the effluent gasses, from the individual capsule effluents for several inert radioactive species. The FPMS continuously measured the sweep gas from each AGR-1 capsule to provide an indicator of fuel irradiation performance. Spectrometer detector systems measured the concentrations of various krypton and xenon isotopes in the sweep gas from each capsule. Eight-hour counting intervals were used to measure the concentrations of Kr-85m, Kr-87, Kr-88, Kr-89, Kr-90, Xe-131m, Xe-133, Xe-135, Xe-135m, Xe-137, Xe-138, and Xe-139. To determine initial fuel quality and fuel performance data, release activity was calculated and paired with birthrates. Release rates were calculated using sweep gas flow data. The release rates were compared to the calculated birth rates in the form of release-to-birth (R/B) ratios for selected nuclides. R/B values provide indicators of initial fuel quality and fuel performance during irradiation. This paper presents a brief summary of the FPMS, the release to birth ratio data for the AGR-1 experiment and preliminary comparisons of AGR-1 experimental fuels data to fission gas release models.*

I. INTRODUCTION

TRI-Structural-Isotropic (TRISO) coated particle fuel development and irradiation of such fuel is being accomplished to support the development of the next generation of gas reactors in the United States. The Advanced Gas Reactor (AGR) experiment series, which is part of the Next Generation Nuclear Plant (NGNP) Fuel Development and Qualification Program, was established to perform the vital baseline fuel qualification data to support licensing and operation of a high temperature gas-cooled reactor (HTGR).

The AGR series of fuel irradiation experiments are planned for insertion into the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL). The first of these experiments termed AGR-1 was inserted in to the ATR in 2006 with initial irradiation commencing on December 24, 2006, and finishing on November 6, 2009, which represents 620 effective full power days (EFPD).

The intent of the AGR-1 experiment was to serve as a shakedown for a multi-capsule test train using laboratory scale fuel. To monitor fuel performance a Fission Product Monitoring System (FPMS) was used to monitor effluent from the individual test train capsules in near real time. With the use of the FPMS TRISO fuel release activities were measured and release-to-birth (R/B) values were computed.

This paper discusses a brief summary of the AGR-1 experiment, the FPMS and the computed R/B values. The computed R/B values enhance the understanding of the fission product release and potential particle failure mechanisms which can then be used to support future fuel fabrication process development.

II. SUMMARY OF THE AGR-1 EXPERIMENT

The AGR-1 experiment exclusively contained UCO type fuel. The test train incorporated six individual test capsules. Each capsule consisted of 12 compacts that contained on average 4,150 fuel particles with total uranium content of 0.9 grams which were supported within a graphite matrix [1]. All capsules were instrumented with thermocouples and were continuously swept with a custom blend of inert gas during irradiation [2]. The inert gas carried fission products downstream to the individual FPMS as is shown in Figure 1.

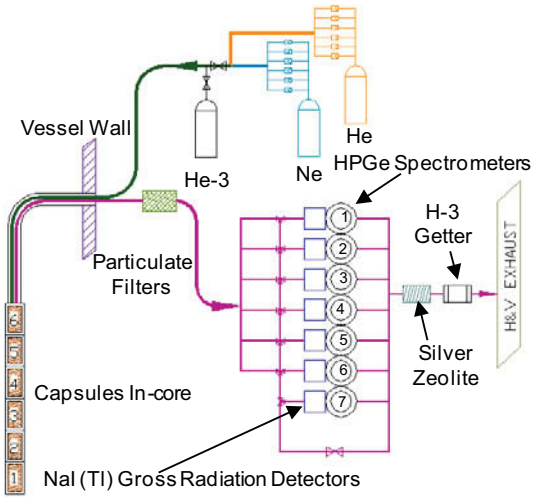

Fig. 1: Simplified gas flow path for the AGR-1 experiment.

The AGR-1 experiment was irradiated for a total of 620 EFPD and achieved a peak fuel burn-up of 19.6% FIMA (fissions per initial heavy metal atoms). During the total irradiation period no particle failures were observed and only small amounts of "tramp" uranium contamination were detected by the FPMS. Table 1 contains key irradiation conditions for the AGR-1 experiment [2].

Table 1: Preliminary AGR-1 Experiment Results.

Capsule	Peak Burn Up (% FIMA)	Peak fast Fluence (10^{25}n/m^2, E>0.18 MeV)	Time Average Peak Temperature (°C)
1	17.2	3.43	1162
2	19.1	4.12	1238
3	19.6	4.39	1211
4	19.5	4.32	1254
5	18.4	3.91	1231
6	15.0	3.08	1183

III. THE FISSION PRODUCT MONITOR SYSTEM

III.A. System Description

The AGR-1FPMS consisted of seven (7) fission product monitor stations, one for each capsule plus a spare. Each station contained a heavily-shielded high-purity germanium (HPGe) gamma-ray spectrometer for determining specific fission gas release concentrations and a sodium iodide [NaI(Tl)] scintillation detector for count rate monitoring of the capsule effluent. The NaI detector, also referred to as the "gross radiation" detector, was used to serve as an "early warning" indicator in the event of a TRISO coating failure.

As is shown in Figure 2, the pipe chase (1) carries the AGR test capsule effluent lines to and from the monitoring stations. Each capsule effluent line (2) branches from the channel, passes through the gross radiation monitoring substation (3) where it is viewed by a shielded NaI(Tl) detector, and then into the HPGe spectrometer shield (4) and through the spectrometer sample chamber viewed by the HPGe detector (5). The effluent line returns to the pipe chase channel and leaves the cubicle to the treatment filters and then to the plant ventilation system [3].

Fig. 2: The seven FPMS stations installed at the Advanced Test Reactor (ATR), which is located at the Idaho National Laboratory (INL).

Each spectrometer was calibrated using a set of simulated gas standards prepared by a source vendor in sample chambers that mimicked the FPMS flow-through, baffled 58±3cm^3 sample chambers.

The FPMS gamma-ray detectors were fitted with standard commercial pulse processing electronics modified to implement the INL pulse injection with subsequent removal technology [4]. This provided quality assurance data in the gamma-ray spectra that the authors believe was essential for unattended operation of the gamma-ray spectrometers.

The NaI(Tl) gross detectors were equipped with multichannel scalar (MCS) units to obtain counts in successive time intervals, while the HPGe spectrometers were processed by multichannel analyzer (MCA) units to obtain counts vs. pulse height. Thus the MCS data details the time for changes in count rate, while the MCA data details the energy of the gamma rays.

III.B. Software Description

The FPM control program monitored the operation of each of the seven stations continually. The operation of each was recorded to a disk file and provided a permanent record of the operation of the system over the course of the fuel irradiation period. Each FPMS station was operated with the MCS and MCA in tandem, starting and stopping at the same time. The usual measurement protocol acquires gamma-ray spectra with counting times of eight hours. This gives adequate measurement sensitivity and provides three sets of results each day. Graphical tools were implemented throughout the course of the experiment that allowed the operator to display and review all seven HPGe spectrometers or all seven gross monitor plots at a glance and to display and investigate any individual acquiring or archived spectral files.

The control program monitors the operation of the data acquisition and at the end of the eight hour real preset measurement time, saves the collected spectra to disk and initiates the online analysis task. An example spectrum is presented in Figure 3.

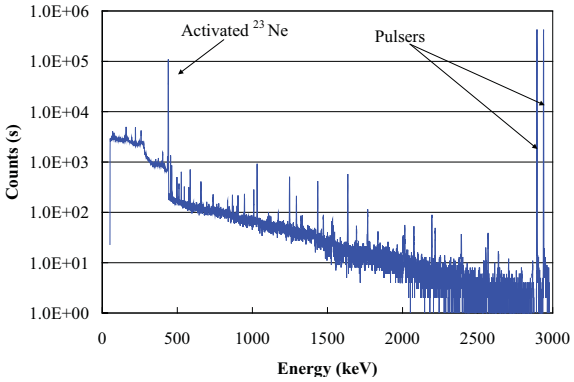

Fig. 3: A sample spectrum of the effluent from an AGR-1 capsule. The large peak located at 439.9 keV is from ^{23}Ne from activation of the inlet gas. The pulser peaks are the result of pulse injection with subsequent removal [4].

During the almost three-year irradiation period the operation of the control software was monitored by a separate supervisory process. This process monitored the control program and if the control program became non-responsive for any reason, the supervisor process would halt and restart the primary control software. Upon re-startup the control program would resume monitoring of any ongoing acquisition and therefore no data was lost. The supervisor process periodically reported, via electronic mail to designated personnel, the status of the control program and any problems requiring a restart of the control program. Any failure in the computer that hosts the control software was reported to an ATR-based operator by an alarm activated by a network-controllable power switch attached to the host computer's private ethernet subnet. The switch monitors a CPU "heartbeat." If the heartbeat stops, the alarm activates.

III.C. Data Analysis

The performance of a nuclear fuel test is typically evaluated using Release-to-Birth Ratios (R/B) that measure the released activity (R) of a certain isotope from the fuel compared to the predicted activity of the isotope in the fuel due to irradiation conditions (i.e., birth activity (B)). The gamma-ray spectrum measurements from the HPGe detectors in each FPMS were used to find the release activities of several different isotopes of Kr and Xe shown in Table 2. The acquired spectra were analyzed automatically using the INL-developed PCGAP gamma-ray spectral analysis code [5,6] and were stored electronically. At the end of each irradiation cycle the FPMS measured activities were corrected to account for decay that occurred during transport from the capsules to the detectors. Transport times were calculated from outlet gas flow rates recorded by the automated experiment data control system (DCS) of the ATR and the capsule-specific volumes through which samples flow to reach the respective monitoring stations [7].

The activities measured from the spectra collected by the FPMS must be corrected to account for decay during transport from the in core capsule to the sample volume and converted to released atoms per second. The proper correction for the measured activity is calculated for equilibrium conditions for the different components illustrated in the simplified flow system shown in Figure 4.

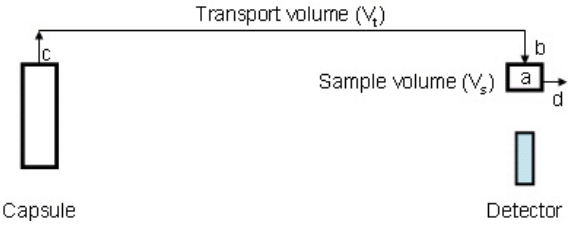

Fig. 4: A simplified single capsule flow graphic to aid in flow correction calculations.

At equilibrium, when the rate of change of the number of atoms of nuclide, 'i,' in the sample volume, 'a,' is zero, one can derive:

$$R_{ci} = \frac{A_{a_i} \cdot e^{\lambda_i \cdot V_t / \dot{f}}}{\left(1 - e^{-\lambda_i \cdot V_s / \dot{f}}\right)} \qquad \text{(eq. 1)}$$

where:

R_{c_i} = Release Activity (atom/sec) of nuclide i at the capsule exit (c)

A_{a_i} = Activity (Bq) of nuclide i measured in sample volume a

λ_i = Decay constant of nuclide i (sec^{-1})

V_t = Transport volume (cm^3) between (c) and (b)

V_s = Sample volume (cm^3)(a)

$\dot{f}$ = Capsule specific flow (cm^3 / sec).

The estimated uncertainty in R_{c_i} can be determined from standard error propagation techniques to be:

$$\sigma^2_{R_{ci}} = \left(\frac{R_{ci}}{A_{a_i}}\right)^2 \cdot (\sigma^2_{A_{a_i}}) + \left(\frac{R_{ci} \cdot \lambda_i}{\dot{f}}\right)^2 \cdot (\sigma^2_{V_t}) + \left(\frac{R_{ci} \cdot e^{-\lambda_i \cdot V_t / \dot{f}} \cdot \lambda_i}{\dot{f} \cdot \left(1 - e^{-\lambda_i \cdot V_t / \dot{f}}\right)}\right)^2 \cdot (\sigma^2_{V_s}) \quad \text{(eq. 2)}$$

or in terms of relative errors:

$$\left(\frac{\sigma_{R_{ci}}}{R_{ci}}\right)^2 = \left(\frac{\sigma_{A_{a_i}}}{A_{a_i}}\right)^2 + \left(\frac{\lambda_i \cdot V_t}{\dot{f}}\right)^2 \left(\frac{\sigma_{V_t}}{V_t}\right)^2 + \left[\frac{V_s \cdot e^{-\lambda_i \cdot V_t / \dot{f}} \cdot \lambda_i}{\dot{f} \cdot \left(1 - e^{-\lambda_i \cdot V_t / \dot{f}}\right)}\right]^2 \left(\frac{\sigma_{V_s}}{V_s}\right)^2 \quad \text{(eq. 3)}$$

The required isotopic birth rates (B) for the isotopes listed in Table 2 are computed from inventory data supplied using a code termed MCWO (for Monte Carlo With Origen) developed at the INL that links neutronic data computed in MCNP to inventory data computed in ORIGEN2 [8].

Table 2: Release activities were measured with the FPMS and fission product birth rates were estimated using MCWO for these 12 isotopes.

Isotope		
Kr-85m	Xe-131m	Xe-135m
Kr-87	Xe-133	Xe-137
Kr-88	Xe-135	Xe-138
Kr-89		Xe-139
Kr-90		

The desired release-to-birth ratio for nuclide, 'i,' is then calculated as:

$$\left(\frac{R}{B}\right)_i(t) = \frac{R_{c_i}(t)}{B_i(t)} \quad \text{(eq. 4)}$$

with a percent uncertainty (%σi) given by:

$$\%\sigma_i = \sqrt{\left(\frac{\sigma_{R_{ci}}}{R_{ci}}\right)^2 + \left(\frac{\sigma_{B_i}}{B_i}\right)^2} \times 100 \quad \text{(eq. 5)}$$

To speed the calculation of these ratios and to ensure accuracy, a semi-automatic processing code was developed that accessed the results of each spectral analysis to extract the required results for activity at the detector at a given time, extracted from the ATR DCS the required capsule specific outlet flow rates, performed the release rate calculations and then correlated these release activities with the appropriate provided and interpolated birth rates to compute release-to-birth ratios.

IV. AGR-1 RELEASE-TO-BIRTH RATIO RESULTS

The gross monitoring plots and the spectrometer data acquired on AGR-1 were routinely examined for evidence of irradiation test-induced TRISO fuel particle failures. Typically a fuel particle failure would be indicated on the gross monitoring system by an observed "puff" release. By November 2009, the test fuels have exceeded peak burnups of 19% FIMA in 3 of the 6 capsules with a minimum peak of 15% FIMA with no particle failure evidence [2]. All measured releases as indicated by the spectrometer data are consistent with and are assumed to come from heavy metal contamination in the graphite of the capsule.

The measured R/B values indicated excellent fuel performance when compared to both historical U.S. TRISO fuel experience [9] and modern German TRISO fuel tests [10]. Figures 5-10 show the R/B values for ^{85m}Kr, ^{88}Kr, and ^{135}Xe computed for Capsules 1-6 during the entire irradiation period of the AGR-1 experiment. The half-lives of these three isotopes make them well suited to observe R/B. The isotopes are long enough lived that they do not significantly decay during transport to the detector, but they are short lived enough that they quickly reach their equilibrium activity during each cycle.

Data from the first irradiation cycle starting on December 24, 2006, is not included in Figures 5-10 because the experiment was undergoing initial experiment shakedown testing.

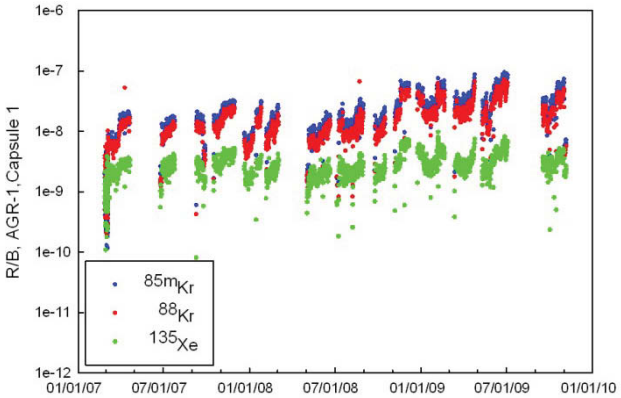

Fig. 5: R/B ratios for AGR-1 Capsule 1, 12/24/06 through 11/06/09.

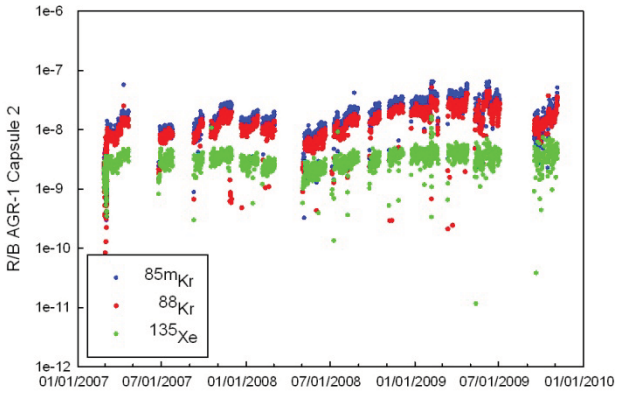

Fig. 6: R/B ratios for AGR-1 Capsule 2, 12/24/06 through 11/06/09.

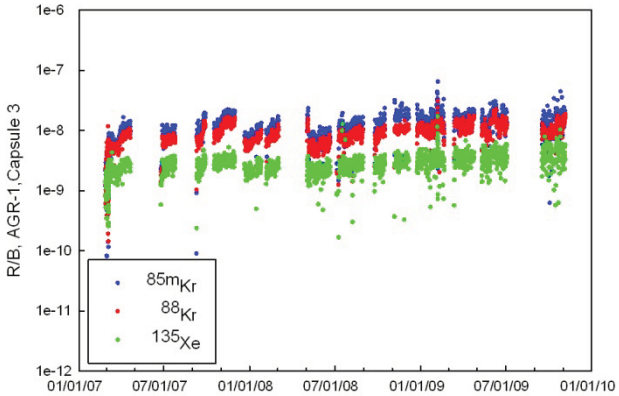

Fig. 7: R/B ratios for AGR-1 Capsule 3, 12/24/06 through 11/06/09.

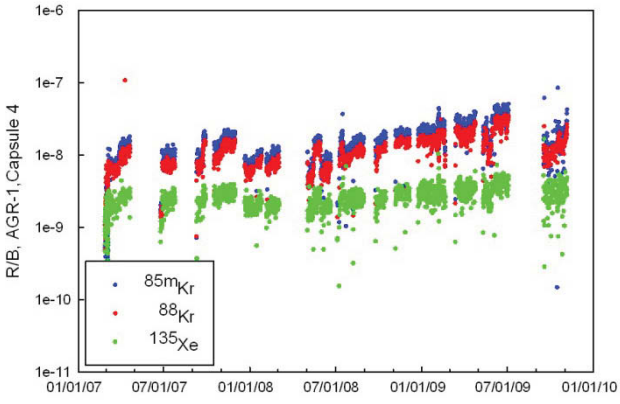

Fig. 8: R/B ratios for AGR-1 Capsule 4, 12/24/06 through 11/06/09.

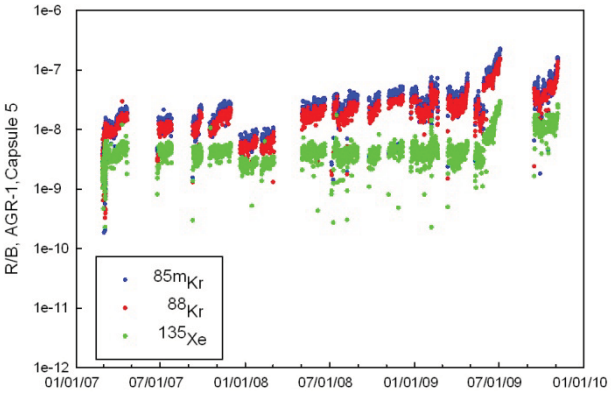

Fig. 9: R/B ratios for AGR-1 Capsule 5, 12/24/06 through 11/06/09.

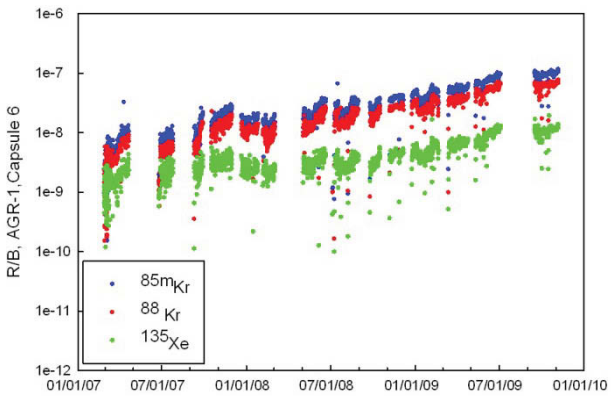

Fig. 10: R/B ratios for AGR-1 Capsule 6, 12/24/06 through 11/06/09.

By the end of final irradiation cycle R/Bs for ^{85m}Kr were mostly below 10^{-7} with the exception of capsule 5 and capsule 6 which neared 2 x $10^{-7.}$ The upward trend is because of increasing fission power and hence, fuel temperature in the experiment.

V. AGR-1 IODINE MEASUREMENTS

Iodine-135 is a fission product radioiodine that decays by beta particle emission with a 6.57-hour half-life. It decays to excited states in Xe-135 with a half-life of 9.1 hours. Among the populated states of ^{135}Xe, the isomeric state of ^{135m}Xe decays to the ^{135}Xe ground state with a half-life of 15.3 minutes and an emission of a 526.6 keV photon. The 15.3-minute isomeric state is populated through a 16.4% branch of ^{135}I and is amenable to determination by gamma-ray spectroscopy using the 526.6 keV gamma line [11].

During irradiation, ^{135}Xe and ^{135m}Xe concentrations at the FPMS result both from direct fission yield and from the decay of ^{135}I. When irradiation stops the only source of ^{135m}Xe is the decay of ^{135}I accumulated during irradiation. The ^{135m}Xe production rate will equal the decay rates of the ^{135}I parent. After several ^{135m}Xe half-lives, it will be in secular equilibrium with its ^{135}I parent. This enables the determination of the amount of ^{135}I released from the fuel particles and deposited in upstream structures from quantification of the ^{135m}Xe concentrations in the flowing gas stream following reactor shutdown.

The ratios of accumulated ^{135}I concentration from ^{135m}Xe measurements after shutdown to end of cycle (EOC) ^{135}I MCWO calculated concentrations are presented in Figure 11 for the six AGR-1 capsules during the 13 irradiation cycles. Currently, the data trends with the preliminary capsule temperature data and indicates that there were no fuel failures. Once the AGR-1 temperature data is finalized the iodine data will be reviewed again.

To expedite the delivery of the Iodine data to the project, extrapolated ^{135}I calculations based on ^{135m}Xe measurements are performed at the end of each AGR irradiation cycle by a semi automatic processing code. This code reads in the isotopic information files and the capsule specific flow information from the NDMAS data base to compute the extrapolated ^{135}I values.

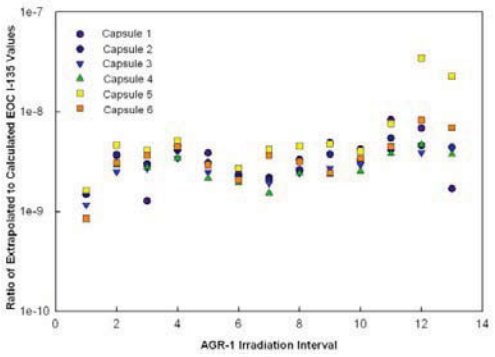

Fig. 11: Ratio of extrapolated to calculated I-135 concentrations. Ratios were computed at the end of each ATR operating cycle for which the AGR-1 experiment was inserted.

VI. COMPARISON TO FISSION GAS RELEASE MODELS

The performance of the AGR-1 experiment can also be evaluated by comparing the measured R/B values to R/B values predicted from semi-empirical R/B models for fission gas release from TRISO fuel. The R/B models include terms for fission product release from catastrophic failure of the TRISO particle, heavy metal contamination in the graphite matrix surrounding the fuel, and the direct recoil of fission fragments. The diffusion pathway is typically modeled using the Booth equivalent sphere model which is dependent on an isotopes diffusion coefficient and half-life [12].

The total R/B value for a particular nuclide is found by multiplying the model determined R/B by the available fraction of heavy metal originating from either failure or contamination, and then summing the contributions from the failure and contamination sources. For comparison to the experimentally measured R/B values, the total R/B for isotope, '*i*,' is shown in equation 6:

$$\left(\frac{R}{B}\right)_i^{\text{total}} = f_{fail}\left(\frac{R}{B}\right)_i^{fail} + f_{cont}\left(\frac{R}{B}\right)_i^{contamination} \qquad \text{(eq. 6)}$$

where f_{fail} is the fraction of failed TRISO particles, f_{cont} is the contamination fraction that is defined as the grams of uranium contamination over the grams of uranium in the observed system, and $(R/B)_i$ is calculated from the model being investigated.

Comparison of measured R/B values to predicted R/B values is a metric for gauging fuel performance and the predictive capabilities of future TRISO irradiations. Figure 12 contains a preliminary comparison of calculated R/B values to an average of measured R/B values taken during irradiation cycle 13 for each isotope of interest. The error bars on the experimental data indicate one standard deviation of the measured R/B values. R/B values for two different models are also shown and were calculated for the conditions of the same ATR irradiation cycle. For the models, a contamination fraction of 3.05 x10^{-7} was used [13], and the temperature was set at 1175°C based on thermocouples measurements and computer simulations of the AGR-1 experiment. Model 1 in Figure 12 is based on German experience and accounts for a separate diffusion relationship for Kr and Xe [14]. Model 2 is based on the experience of the fuel testing program and the High Temperature Test Reactor in Japan [15], which uses a single diffusion coefficient relationship for both Kr and Xe isotopes when modeling diffusion release. Additionally, the predicted R/B values for a single failed TRISO particle from Model 1 are also shown in Figure 12.

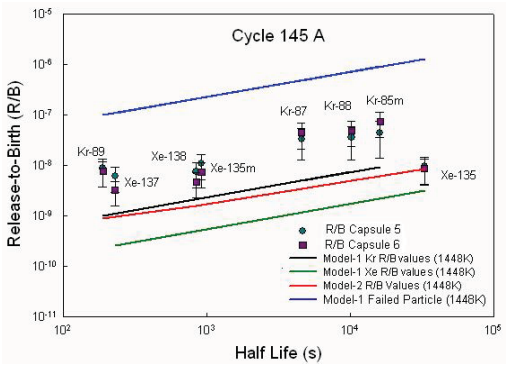

Fig. 12: Preliminary Average R/B's measured for irradiation interval 13 and model dependent R/B ratios for 1175°C and f_{cont} = 3.05 x10^{-7}.

The R/B model for contamination under predicts the observed release to birth ratio from the AGR-1 experiment. Sources of error in the model and observed R/B values include the contamination fraction (f_{cont}), differences between current experiments and the studies the models are based upon, and uncertainty in the isotopic composition of the heavy metal contamination. Any error in the f_{cont} will directly effect a proportional error in the measured R/B values. The density of the fuel compacts for the fuel that the PARFUME (Model 1) R/B model is based on was higher at 1.7 g/cm^3 compared to 1.3 g/cm^3 used in the AGR-1 experiment [3]. A lower density fuel compact will enhance the release from contamination by creating more open porosity in the graphite over-pack for Kr and Xe gas to escape into. The exact isotopic concentration of the contamination Uranium present in the fuel compacts is not well known. The "birth" rate used to calculate R/B is based on the original Uranium content of the six individual capsules, and the birth rate values are updated for the reactor conditions history. The initial enrichment of the uranium in the fuel was 19.7%, however the contamination uranium may not be of the same enrichment. The "birth" activity for contamination calculations is assumed as a fraction of the "birth" being produced from the current fuel uranium and plutonium content that is present from the burn-up of the fuel and breeding. In spite of the under prediction of the R/B models, all release-to-birth ratios are below the predicted R/B value for a single TRISO failure, as seen in Figure 12 for Capsules 5 and 6. This observation provides additional evidence supporting the lack of TRISO failures during the AGR-1 experiment.

VII. CONCLUSIONS

The AGR-1 Fission Product Monitoring System is a collection of gamma-ray rate monitors and gamma ray spectrometers that measured effluent from the AGR-1 experiment which contained 72 cylindrical fuel compacts each containing about 4150 coated particles. During the 620 EFPD irradiation period, zero particle failures were observed based on the measured release-to-birth ratios. Comparisons between measured R/B ratios and predicted R/B ratios from semi-empirical models indicate that the Kr and Xe fission gas detected by the FPMS is diffusing from tramp uranium contamination inside the TRISO coating layers.

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