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Determination of the Quantity of I-135 Released from the AGR Experiment Series

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Abstract – *A series of three Advanced Gas Reactor (AGR) experiments have been conducted in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL). From 2006 through 2014, these experiments supported the development and qualification of the new U.S. tri-structural isotropic (TRISO) particle fuel for Very High Temperature Reactors (VHTR). Each AGR experiment consisted of multiple fueled capsules, each plumbed for independent temperature control using a mix of helium and neon gases. The gas leaving a capsule was routed to individual Fission Product Monitor (FPM) detectors. For intact fuel particles, the TRISO particle coatings provide a substantial barrier to fission product release. However, particles with failed coatings, whether because of a minute percentage of initially defective particles, those which fail during irradiation, or those designed-to-fail (DTF) particles, can release fission products to the flowing gas stream. Because reactive fission product elements like iodine and cesium quickly deposit on cooler capsule components and piping structures as the effluent gas leaves the reactor core, only the noble fission gas isotopes of Kr and Xe tend to reach FPM detectors. The FPM system utilizes High Purity Germanium (HPGe) detectors coupled with a thallium activated sodium iodide NaI(Tl) scintillator. The germanium detector provides individual isotopic information, while the NaI(Tl) scintillator is used as a gross count rate meter. During irradiation, the ^{135m}Xe concentration reaching the FPM detectors is from both direct fission and by decay of the accumulated ^{135}I. About ~2.5 hours after irradiation (ten 15.3 minute ^{135m}Xe half-lives) the directly produced ^{135m}Xe has decayed and only the longer lived ^{135}I remains as a source. Decay systematics dictate that ^{135m}Xe will be in secular equilibrium with its ^{135}I parent, such that it's production rate very nearly equals the decay rate of the parent, and its concentration in the flowing gas stream will appear to decay with the parent half-life. This equilibrium condition enables the determination of the amount of ^{135}I released from the fuel particles by measurement of the ^{135m}Xe at the FPM following reactor shutdown. In this paper, the ^{135}I released will be reported and compared to similar releases for noble gases as well as the unexpected finding of ^{131}I deposition from intentional impure gas injection into capsule 11 of experiment AGR-3/4.*

I. INTRODUCTION

AGR-1, 2 and 3/4 are the first of the Advanced Gas Reactor (AGR) series of in-core, tri-structural isotropic (TRISO) coated fuel experiments irradiated within the Advanced Test Reactor (ATR), located at Idaho National Laboratory (INL). Each one of these three experiments had multi-year irradiations and will provide researchers with decades worth of information to analyze.

Each capsule of every AGR experiment test train was plumbed for temperature control with an

independent mix of helium and neon gas. The effluent gas leaving an experiment capsule was routed to individual Fission Product Monitor (FPM) detectors, where capsule specific isotopic information was collected and analyzed. For intact fuel particles, the TRISO particle coatings provide a substantial barrier to fission product release. However, particles with failed coatings, whether those which fail during irradiation or a minute percentage of particles that was initially defective, can release fission products to the flowing effluent gas stream. Because reactive fission product elements like iodine and cesium readily deposited onto the relatively cooler capsule components and piping surfaces, only the noble fission gas isotopes of krypton and xenon tend to reach FPM detectors.

This paper examines the released ^{135}I from these three experiments and discusses how ^{135}I concentrations are determined from measured ^{135m}Xe concentrations. Once the ^{135}I is determined, it is then compared to the end of cycle (EOC) computed ^{135}I inventories. From these comparisons one can infer fuel integrity. This paper also briefly discusses the surprise finding of ^{131}I in three of the FPM sample chambers.

II. BACKGROUND

Iodine-135 is a radioactive fission product that decays by beta particle emission with a 6.57-hour half-life. It decays to excited states of ^{135}Xe 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 with 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. This decay scheme is depicted in simplified form in Figure 1[1].

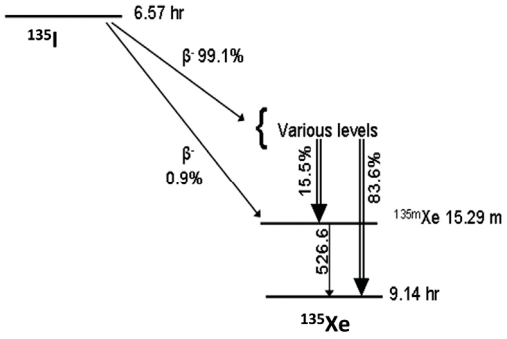

Fig. 1: Simplified decay scheme of ^{135}I.

During irradiation, ^{135}Xe and ^{135m}Xe concentrations at the FPM system 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 rate 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 [2].

III. TECHNIQUES AND METHODS

III.A. Equipment and Measurements

The FPM System (FPMS) is a collection of seven FPM detectors, one for each experiment capsule, plus a spare. The first experiments, AGR-1 and AGR-2 consisted of 6 capsules per experiment, with individual FPM for each capsule with one spare for a total of 7 FPM stations. AGR-3/4 consisted of 12 capsules with an individual FPM for each capsule with two spares for a total of 14 FPM stations (Fig. 2). Each FPM consisted of a NaI(Tl) scintillation detector for gross radiation count rate information and a High Purity Germanium (HPGe) gamma-ray spectrometer to measure fission product decay activities as the effluent gas flowed through a baffled, 58 ml sample chamber located within the lead shielding of the FPM system. In concert with effluent flow rates and transport volumes for the respective capsules, these activities can be related back to release rates at the respective capsule for the observed fission product isotopes. In this paper we discuss ratios of these to modeled release rates, or similarly of measured to modeled capsule inventories. The NaI detector is used to monitor the gross gamma counting rate from the effluent gas flowing through the lines before they reach the sample chamber. The digitized results from the HPGe and NaI detectors are histogrammed by a Multi-Channel Analyzer (MCA) (provided by the Acquisition Interface Module (AIM)) and Multi-channel Scalar (MCS) (provided by the MultiPort II), respectively. The accumulated data is transmitted to the host computer under the control of the FPMS software. The FPMS control program monitors the operation of each of the seven FPM stations continually. 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 which are used to compute daily capsule release activity values [3,4,5].

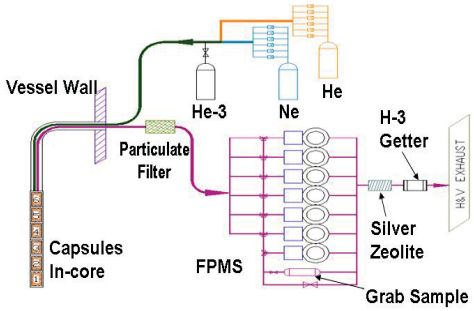

Fig. 2: Simplified gas flow path for the AGR-1 and AGR-2 experiment. The AGR-3/4 experiment had 12 capsules in core and 14 dedicated FPM's. Each AGR experiment had one on-line spare FPM per six test train capsules.

Transport times from each capsule to its respective FPM are estimated using capsule-specific effluent flow rates and transport volumes, the volumes through which the effluent flows between the capsule and the FPM. These were determined early in each AGR experiment from data acquired in a series of neon-injection test measurements. Capsule specific outlet flow meters are located upstream of the fission product monitors and measure the flow of effluent as it leaves the specified capsule. Later in the experiment series a set of flow meters was added on the outlet line of the FPM to be used as verification of outlet flow through the capsule and through the FPM. The capsule specific outlet flow values and transport volumes are used to compute the releases and hence the release ratios discussed in this paper.

At the end of each Advanced Test Reactor (ATR) irradiation cycle, the FPMS was kept running for a minimum of 48 hours which is long enough to observe ^{135m}Xe generated from the decay of the accumulated ^{135}I inventory. Eight-hour (real time) spectra were recorded and analyzed for this time duration following reactor shutdown. By 2.5 hours following reactor shutdown, ten ^{135m}Xe half-lives, any measurable ^{135m}Xe produced by fission has decayed away. Any ^{135m}Xe detected in the spectra acquired after that is clearly from decay of ^{135}I. ^{135m}Xe continued to be detected 40 hours following reactor shutdown and in some instances longer, as shown in Fig. 3.

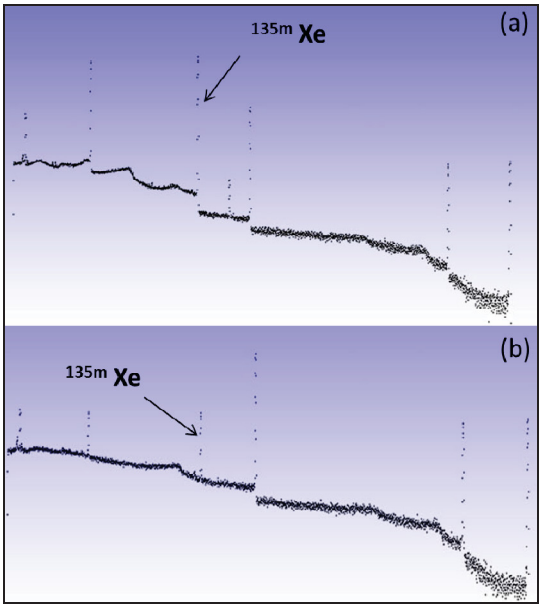

Fig. 3: ^{135m}Xe clearly visible at the FGM chamber for AGR-3/4, Capsule 6, 11 hours (a) and 47 hours (b) after reactor shutdown.

III.B. Calculations and Corrections

Computing end-of-cycle ^{135}I from the measured ^{135m}Xe activities requires correction for decay of both species, of ^{135}I following shutdown and of ^{135m}Xe as it was transported by the gas from its source near the test capsule, the presumed point of equilibrium with its ^{135}I parent. Figure 4 is provided to facilitate the following discussion.

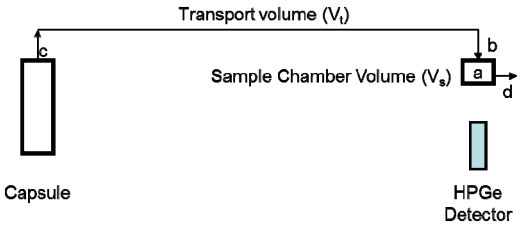

Fig. 4: Single capsule flow diagram.

Released ^{135}I is presumed to be firmly deposited near the capsule and does not move downstream. Daughter ^{135m}Xe is in secular equilibrium with its ^{135}I parent at the exit of the fuel capsule (c). The sweep gas, flowing at a rate given by the capsule outlet flow meter, carries the ^{135m}Xe through the transport line volume (V_t) to the entrance of the sample counting chamber (b), then through the sample volume (V_s) filling the baffled sample chamber (a), and then flowing out of the sample chamber at (d). As the sweep gas continuously flows, the FPM detector acquires repeated 8 hour spectra without interruption until manually stopped.

Immediately after reactor shutdown, helium gas flows through the AGR test train capsules, carrying capsule specific effluent to the respective FPM for up to 72 hours or as directed by research staff.

III.C. Secular Equilibrium

It is important to understand the effect of the secular equilibrium relationship between ^{135}I and its ^{135m}Xe daughter in order to formulate the required corrections properly. Collocated the ^{135m}Xe (half-life equal to 15.3 minutes) is assumed to be [after 2.5 hours following reactor shutdown] in secular equilibrium with its ^{135}I (half-life equal to 6.6 hours) parent in the capsule. Secular equilibrium is established such that the ^{135m}Xe and ^{135}I activities are nearly equal, and the ^{135m}Xe activity decays with an apparent half-life equal to that of its long lived ^{135}I parent. However, if the two were separated, as would be the case if one took a grab sample of the noble gases, then the ^{135m}Xe decays with its own 15.3 minute half-life. In the case of on-line measurements, the situation is somewhat different since the activity at any downstream point is renewed by fresh releases from the upstream equilibrium parent. In practice this means that at a downstream point, position b in Fig. 4, the measured ^{135m}Xe activity changes in time according to the 6.6 hour ^{135}I half-life, but the absolute activity of ^{135m}Xe at any downstream point will be a function of the transport time from the capsule and a decay constant dominated by the 15.3 minute ^{135m}Xe half-life. In other words, picking two positions along the downstream sample line separated in transport time by 15.3 minutes, the ^{135m}Xe concentrations at both points will vary in time with the ^{135}I parent half-life of 6.6 hours; however, at any given moment, the ^{135m}Xe concentration at the second point would be very close to half of the concentration measured at the first point.

III.D. Extrapolation and Corrections

The corrections that need to be applied to the measured ^{135m}Xe data in order to correct its measured activities at the detector to concentrations at the capsule exit can be specified as:

$$C_{Cap}(t_r) = \frac{A(t_r)}{V_s} * f_1 * f_2 * f_3 \qquad \text{(eq. 1)}$$

where:

$C_{cap}(t_r)$ = the desired ^{135m}Xe concentration (Bq/cm^3) at the capsule exit at a time t_r after reactor shutdown.

$A(t_r)$ = the activity (Bq) of ^{135m}Xe calculated from a spectrum that started at time t_r after reactor shutdown.

V_s = the sample volume viewed by the spectrometer.

f_1 = Factor to correct the activity reported by the spectral analysis code to the start of the spectral acquisition.

f_2 = Factor to correct for decay of the ^{135m}Xe during transport through volume V_t between the capsule outlet and the sample chamber inlet.

f_3 = Factor to correct for decay during hold-up in the sample chamber volume V_s.

Correction for activity *(f_1)*:

Because of the on-line nature of the FPMS data, the decay correction in the automatic analysis software is turned off. The reported activity is the average activity over the specified counting interval. If the half-life of the species under study is not long relative to the duration of the spectral acquisition, then a correction must be applied for this decay. The correction factor *(f_1)* is computed as:

$$f_1 = \frac{\lambda_I * RT}{(1 - e^{-\lambda_I * RT})} \qquad \text{(eq. 2)}$$

where:

λ_I = the ^{135}I decay constant of 2.931x10^{-5} s^{-1}

RT = the spectral acquisition time (real time) in seconds

Note that since the activity at the detector varies with the ^{135}I half-life, the ^{135}I decay constant is appropriate for this correction. The spectral acquisition times (RT) for all AGR experiments were 8 hours or 2.88x10^4 s, thus f_1 equals 1.481 and any uncertainties are negligible.

Correction for decay during transport *(f_2)*:

As the ^{135m}Xe is swept from the capsule and transported to the sample chamber inlet it appears to decay with a decay constant equal to the difference between the ^{135m}Xe decay constant and the ^{135}I decay constant. This accounts for the decay of the source when the daughter was swept from it. The transport

time is determined by the capsule outlet flow rate and the transport volume (V_t). Capsule and experiment specific outlet flow rates are continuously recorded by the ATR distributive control system (DCS) and then are transferred to the Nuclear Data Management and Analysis System (NDMAS) database. Capsule-specific transport volumes were determined by neon injection testing at the beginning of each AGR experiment. AGR-1, 2 and 3/4 each has specific transport volumes.

The correction factor f_2 can be computed as:

$$f_2 = e^{\frac{(\lambda_{Xe}-\lambda_I)*V_t}{Q}} \quad \text{(eq. 3)}$$

where:

λ_{Xe} = ^{135m}Xe decay constant

λ_I = ^{135}I decay constant

V_t = capsule-specific transport volume (cm^3)

Q = average capsule outlet flow rate during the spectral measurement.

The relative variance of f_2 can be estimated by normal error propagation techniques.

The decay correction procedure outlined above presumes that the ^{135}I producing the ^{135m}Xe is located in the capsule or its immediate surroundings. While this is a convenient assumption, due to the volatility of certain iodine species (for example I_2), deposition might occur well downstream from the capsule. Scoping calculations [6] imply that the drop in temperature when the effluent line leaves the reactor vessel and the relatively warm (50°C to 70°C) primary coolant will cause deposition of the elemental iodine still being carried in the effluent stream. At a nominal flow rate of 0.5cm^3/s this location is about 8 to 12 seconds downstream of the average capsule. For the 15.3 minute ^{135m}Xe half-life, an 8 to 12 seconds difference in decay time alters the concentration by less than 1%. Thus, if all of the previously released ^{135}I were deposited at the reactor vessel exit, some 8 to 12 seconds closer to the spectrometers than was assumed, then the f_2 correction factor would have less than a 1% bias. Because of the somewhat speculative nature of this uncertainty it has not been propagated to the tabular results in this paper.

Correction for holdup in the sample chamber *(f_3)*:

The sample chamber (a) in Fig. 4, viewed by the detector, has a volume V_s. If either V_s is very small, or the half-life of the radioisotope of interest is very long, then the mean concentration measured by the detector is nearly identical to that entering the chamber at point b in Fig. 4. However, if the sample volume is large and/or the species half-life is short, then the mean concentration measured by the detector will be lower than that entering due to decay of the species in the sample chamber.

The correction factor f_3 that corrects for this effect can be formulated as:

$$f_3 = \frac{(\lambda_{Xe}-\lambda_I)*V_s}{Q*\left(1-e^{\frac{-(\lambda_{Xe}-\lambda_I)*V_S}{Q}}\right)} \quad \text{(eq. 4)}$$

For the measurements performed here, $V_S = 58 \pm 2$ cm^3. As with the previous correction, the effective decay constant is the difference at which the measured ^{135m}Xe appears to be decaying.

Calculation of the daughter ^{135m}Xe concentrations at the capsule release point:

Having computed the needed correction factors, the daughter ^{135m}Xe concentrations at the capsules can be calculated using equation 1.

Since all of the operands in this equation are multiplicative, and assuming the parameters are non-correlated (a reasonable assumption), the relative uncertainty in the value can be computed by quadrature propagation of the individual relative uncertainties.

Thus the ^{135m}Xe activity was determined for each spectrum which started at least 2.5 hours after reactor shutdown. Activities at the capsule source at the respective start of acquisition times were calculated for each spectrum using equation 1. These values are examined to verify they follow the expected ^{135}I decay as a function of time following reactor shutdown. Each value is decay corrected to reactor shutdown, providing several values for each capsule at the end of each irradiation cycle. An uncertainty is also calculated for each value using normal error propagation methods and estimates of uncertainties in the transport volumes, the spectrum activities, and the flow rates. Because the activities decay with time after shutdown, the uncertainties grow for later and later spectra. Thus activity at

shutdown is computed as the weighted average with each value weighted by the inverse square of its uncertainty. This automatically devalues less accurate data and is equivalent to the more laborious initial determination.

To expedite the delivery of the iodine data to the project at the end of each AGR irradiation cycle these calculations were incorporated into a semi-automatic processing code called Analyze_I135 (Fig. 5). This code reads in the isotopic information files and the capsule specific flow information from the NDMAS database to compute the extrapolated ^{135}I values.

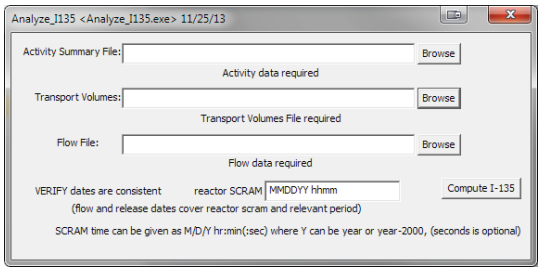

Fig. 5: Screen capture of Analyze_I135, a semi-automatic, post irradiation cycle data processing software**.**

III. E. End Of Cycle ^{135}I Calculational Methodology

The physics methodology used to model the TRISO-particle fuel depletion for the AGR experiment series is a Monte Carlo depletion methodology. The specific method employs the INL JMOCUP (Jim Sterbentz's MCNP-ORIGEN2 Coupled Utility Program) [7,8] utility code which links the standard and well-known MCNP5 [9] and ORIGEN2.2 (Oak Ridge Isotope Generation) [10] computer codes. The MCNP5 code performs the neutron and gamma transport calculation and the ORIGEN2.2 code solves the coupled time-dependent ordinary differential equations governing the nuclide buildup, depletion, and decay. The JMOCUP utility links the MCNP and ORIGEN2.2 inputs and outputs in a back and forth manner as the depletion calculation steps through each irradiation cycle.

The JMOCUP method fully simulates the ATR under as-run operating conditions. For the AGR experiment series, the JMOCUP utility depletes the ATR driver core fuel elements, the AGR TRISO-particle fuel compacts, and the hafnium shroud surrounding the AGR capsules. In addition, the utility modifies the MCNP input model at each time-step to simulate the daily core and lobe power changes as well as rotations in the ATR outer shim control cylinders and neck shim rod configuration in order to maintain a critical reactor configuration and thereby simulate the actual as-run ATR operating conditions.

The depletion calculation is somewhat overburdened in that daily time steps are used to achieve a high-resolution depletion calculation. In general, reactor core depletion calculations do not require daily time steps to achieve desired computational accuracy for burnup estimates. However, the daily heat rates provided by the physics calculation plus the measured daily helium-neon gas mixtures used to control the AGR experiment capsule temperatures are needed as input for the AGR experiment thermal model and the daily temperature predictions. The thermal model is used to predict compact centerline, capsule component, and thermocouple temperatures on a daily basis, since TRISO particle temperature is an important variable to know and control in the evaluation of the particle fuel performance. All other calculated physics parameters (R/B, %FIMA burnup, fission product and actinide concentrations, and fast fluence estimates) simply benefit from the high-resolution calculation.

End-of-cycle ^{135}I concentrations are calculated as part of the JMOCUP depletion calculation. Of particular interest are the specific ^{135}I concentrations in each TRISO-particle compact prior to the end of each ATR power cycle. The ^{135}I tends to build up and reach a maximum concentration at the end of each cycle; the concentrations are readily extracted from the ORIGEN2.2 output in units of moles/compact.

IV. ^{135}I RESULTS

The end of cycle release fractions of ^{135}I as a function of reciprocal fuel temperature are presented in Figure 6 for AGR-1 and Figure 7 for AGR-2 and AGR-3/4. In addition, the release to birth ratio for ^{135m}Xe just prior to EOC is also plotted in Figures 6 and 7. The ^{135}I and ^{135m}Xe exhibit similar behavior with respect to temperature. The low values indicate there was no fuel particle failure. The effective activation energy (Q_A) for the ^{135m}Xe R/B and of the ^{135}I release fraction are similar, Q=32kJ/mole and Q_A=41kJ/mole respectively for AGR-1.

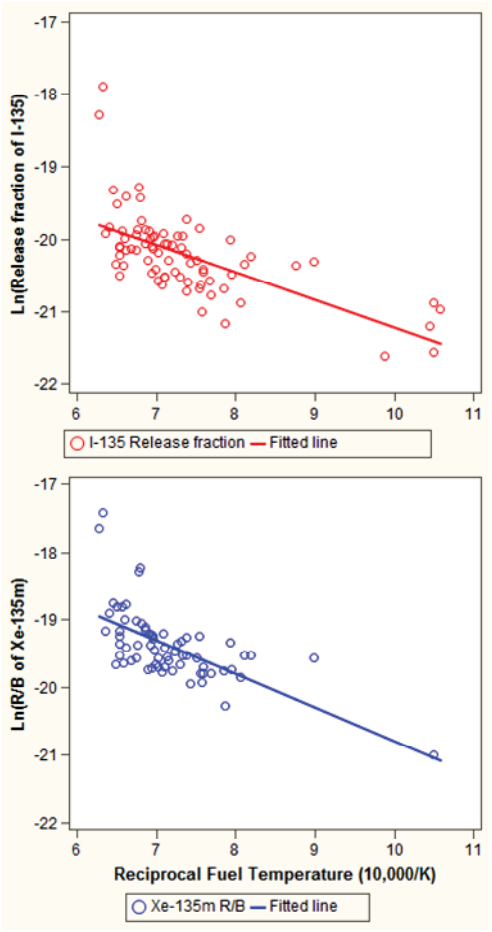

Fig. 6: Release fraction of ^{135}I for AGR-1 and R/B for ^{135m}Xe for AGR-1 are low, indicative of heavy metal contamination outside of the intact silicon carbide layer of TRISO particles.

For AGR-2 and AGR-3/4, the data trends with the preliminary capsule temperature data as well in Figure 7. It is believed that fuel in the AGR-2 experiment had an exposed fuel kernel in each capsule thus the higher release fraction and R/B ratio than that of AGR-1. Because of experiment flow issues during AGR-2, only the first three irradiation cycles (June 2010 – January 2011) of AGR-2 data are presented in Figure 7. AGR-3/4 contained approximately one percent of designed-to-fail (DTF) fuel particles, or approximately 960 DTF in approximately 91,000 driver particles. The first observable failures occurred within the first two weeks of irradiation starting in December of 2011. DTF fuel failures were observed until the end of the experiment in April of 2014. Figure 7, captures the logarithm of the per failed particle quantity for ^{135m}Xe and ^{135}I, for AGR-3/4 and AGR-2 fuel with the exposed kernel in AGR-2 treated as a pre-experiment failure. The slope of the fitted line for ^{135m}Xe R/B per failure and the slope of the ^{135}I release fraction per failure are again similar, with an effective activation energy, Q_A=78kJ/mole and Q_A=103kJ/mole, respectively. This indicates that the measurement of ^{135m}Xe is a good indicator of ^{135}I presence and thus of fuel performance.

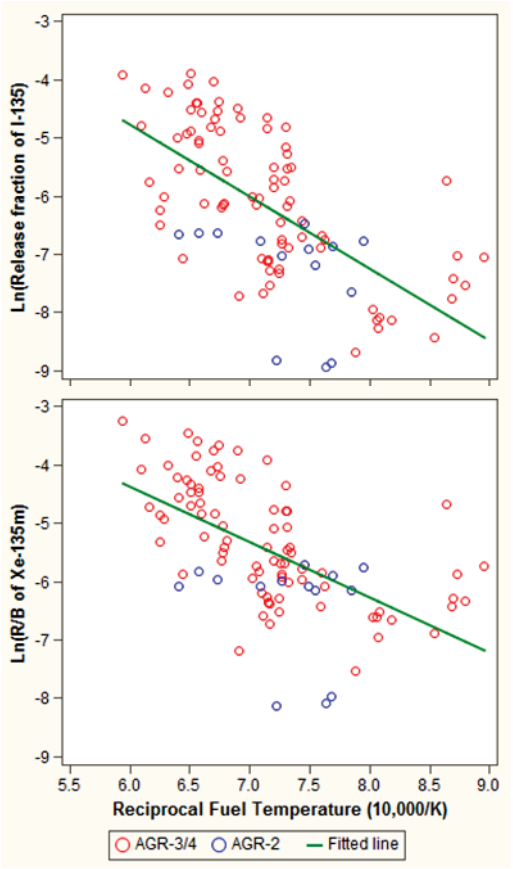

Fig. 7. Release fraction of ^{135}I and R/B per failed particle for ^{135m}Xe for AGR-2 and AGR-3/4.

Because of plate-out issues, ^{135}I cannot easily be measured, however ^{135m}Xe is easily measured with the FPMS. In both Fig. 6 and Fig. 7, the ^{135}I release fraction is a factor of 2 less than the ^{135m}Xe R/B. These similarities indicate that ^{135m}Xe may be a good surrogate for ^{135}I for all three experiments, AGR-1, AGR-2 and AGR-3/4.

V. UNEXPECTED ^{131}I

An additional goal of the AGR-3/4 experiment was to study the effect of an impure gas mixture on failed TRISO fuel. The design called for injecting 0.5 sccm of impure gas with 30 sccm of existing blended neon/helium gas mixture. The desired amounts of impurities in the flow to capsule 11 were: 50 ppmv hydrogen, 50 ppmv carbon monoxide, and 10 ppmv of water, with 29.5sccm

helium/neon. The mixing ratio of impure gas to neon/helium gas was approximately 1 to 60 (0.5 sccm to 30 sccm). Investigation began in irradiation cycle 5. Injection was carried out briefly to establish design functionality starting on August 26, 2013 and ending on September 5, 2013. The injection resumed in earnest, officially starting on September 9, 2013.

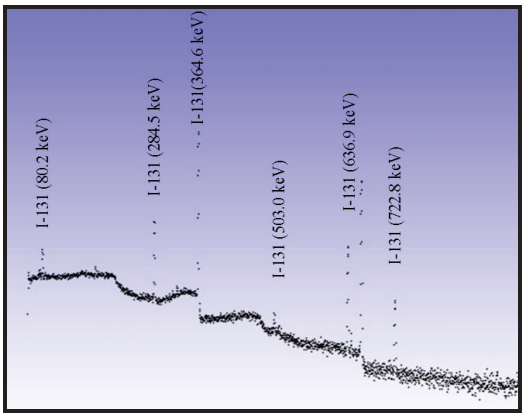

Fig. 8: Iodine–131 from AGR-3/4 Capsule 11. ^{131}I is measured at the FGM sample chamber. This eight hour measurement was acquired on November 2, 2013; sixteen days post irradiation of cycle 6 and seven days prior to the start of irradiation cycle 7, and clearly shows the 6 strongest lines.

Iodine-131 was first identified in spectra from the capsule 11 outlet flow several days after the end of irradiation cycle 5. Spectra observed this long after reactor shutdown typically only show the usual background lines and the long lived ^{137}Cs which accumulates in the FPM sample chamber from the decay of ^{137}Xe. Interestingly, viewing six spectral lines from ^{131}I shown in Figure 8, and at appropriate relative intensities, was entirely unexpected. During irradiation these lines are mostly obscured by the gamma lines from the mobile krypton and xenon isotopes. At this activity level, these lines soon become apparent since most of the krypton and xenon isotopes decay away. Thus, even weaker activities readily show the strongest ^{131}I line at 364.6 keV, once the nearby lines from the short lived ^{89}Kr and ^{138}Cs decay away.

V.A. Detection of ^{131}I in FPM 11 for the AGR3/4 Experiment[1]

Review of all capsule 11 data for evidence of ^{131}I revealed another surprise seen in Figures 9 and 10. ^{131}I was present in the sample chamber of FPM 11 as early as irradiation cycle 2. An activity level of approximately 0.10 μCi was measured when the reactor was scrammed around mid-cycle on March 22 and again on March 27, 2012. Also, an activity level of approximately 0.14 μCi was measured at the end of the cycle on May 5, 2012.

The question arises, is this iodine specie flowing through the chamber like the krypton and xenon isotopes or is it stuck to the surface of the chamber? Figure 10 shows a small excess decrease in the ^{131}I activity in October of 2013 during the shutdown following irradiation cycle 5. Over the course of 16 days during shutdown, the activity values shift from the upper decay line down by an extra 15% to the lower decay line. This means that while 75% decayed in two half-lives, only some 15% was removed from the source by desorption into the effluent gas. If the ^{131}I is 5 times more likely to decay as to desorb, then it's mostly fixed to the surface somewhere. If the iodine is adhered at some upstream location, it seems likely to be similarly adhered at all points between the capsule and the FPM.

During irradiation cycle 5 the ^{131}I inventory in the FPM 11 sample chamber peaked at about 6.2 μCi, some 50 times the activity observed at the end of the 2nd irradiation cycle. This value can seem large, but in comparison to the expected inventory of ^{131}I it represents only 0.6 ppm reaching the FPM in this added impurity situation. When compared to the inventory just from failed fuel particles it is about 60 ppm.

The phenomenal increase in ^{131}I in FPM 11 with the addition of the impurities in irradiation cycle 5 strongly suggested that the impurities were helping to mobilize some of the ^{131}I capsule inventory. As a check, impurity addition was halted on Feb 21, 2014, 8 days into irradiation cycle 7. When the reactor was shut down mid cycle on March 21, the FPM 11 inventory was about 0.014 μCi. Decay corrected back to shutoff of the impurities on Feb 21, this would be about 0.17 μCi, consistent with the activity calculated from the 364.6 keV line after correction for ^{89}Kr and ^{138}Cs. The impurities were restored on March 28, 2014 when the reactor was restarted. When cycle 7 concluded on Apr 12, the inventory was at 0.078 μCi showing that iodine was again being mobilized, though at a lower level than at the end of cycle 6, for which mobilization was lower than for cycle 5.

[1] 1μCi = 37,000 Bq

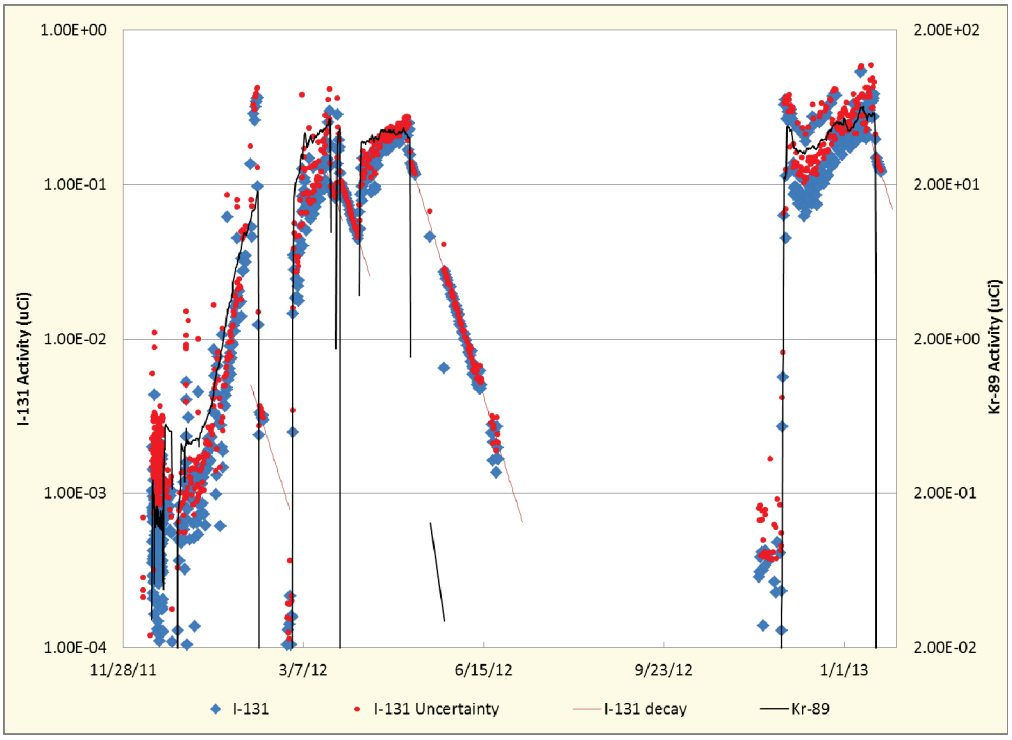

Fig. 9: ^{131}I accumulation in FPM 11 for the first three irradiation cycles of AGR-3/4. The ^{131}I uncertainty is extremely small when fitted with the measured ^{131}I values and trend with the decay rate of ^{131}I at the end of cycle 2. Kr-89 is plotted to serve as visual aid in regards to when the reactor is running.

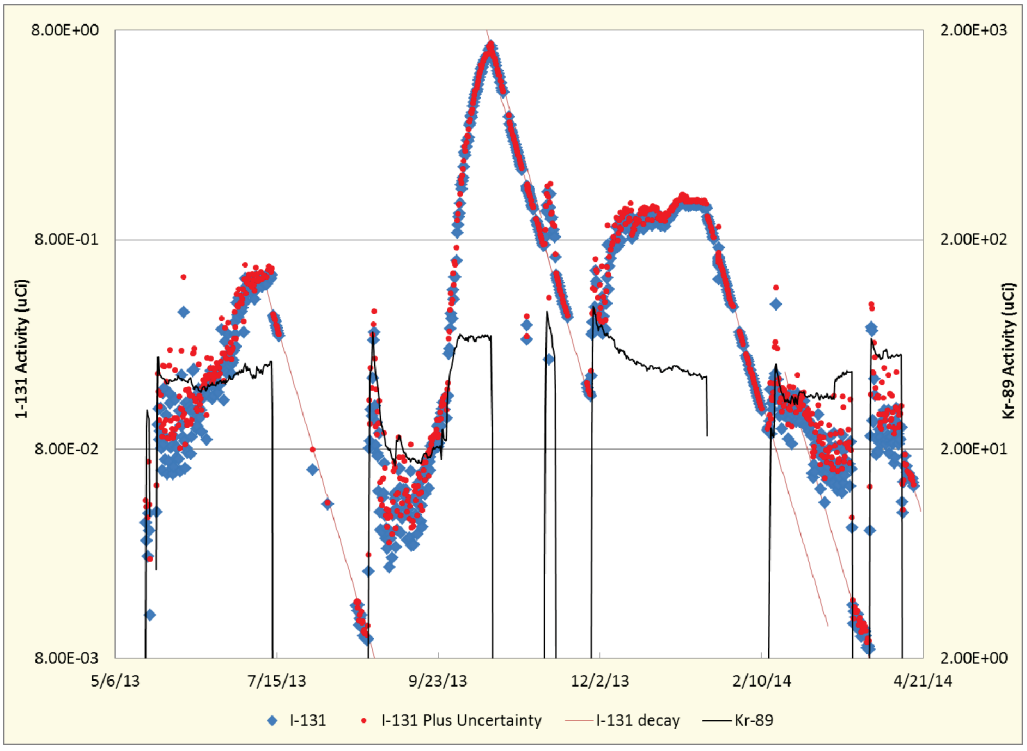

Fig. 10: ^{131}I accumulation in FPM 11 of AGR-3/4 capsule 11 over the last 4 irradiation cycles. ^{131}I maximum plate-out within the FGMS sample chamber occurred during cycle 5.

V.B. Detection of ^{131}I in other FPM's for the AGR3/4 Experiment

The detection of ^{131}I at FPM 11 in irradiation cycle 2 prior to impure gas injection prompted review of the data for all of the other capsules. Of the 11 other capsules only capsules 7 and 8 showed ^{131}I in the FPM sample chamber. FPM 7 and FPM 8 respectively had inventories of 1.9 and 1.8 µCi at the end of the first irradiation cycle. Both had significantly diminished activity afterwards as depicted in Figures 11 and 12 for FPM 7 and Figures 13 and 14 for FPM 8. For FPM 7, there was evidence for small additions to the sample chamber inventory early in cycle 2, at the mid cycle shutdowns on March 22 and March 27, 2012, about 0.06 and 0.02 µCi respectively. There was no convincing evidence for later additions to the FPM 7 inventory. For FPM 8, there were inventory additions totaling 0.31 µCi in irradiation cycle 2 and 0.05 µCi in irradiation cycle 6. The FPM 8 inventory at the end of cycle 6 amounts to 4 ppb of the capsule inventory and 0.4 ppm of the failed particle inventory.

Iodine-131 was clearly present in both FPM 7 and 8 after the first irradiation cycle, yet an impure gas mixture was never injected into those capsules. Our assumption is that some moisture was initially present in the graphite of these two capsules and off-gassed during the first few irradiation cycles providing mobility to the iodine inventory.

Release fractions of ^{131}I for each capsule at the end of each irradiation cycle are all below 1 ppm. When normalized such that only the inventory from failed particles is considered the release fraction values range from 0.1 to 60 ppm. These low values speak to the outstanding detection capability for the FGMS. ^{131}I plate-out is dominating the behavior but the overall release of ^{131}I is very small.

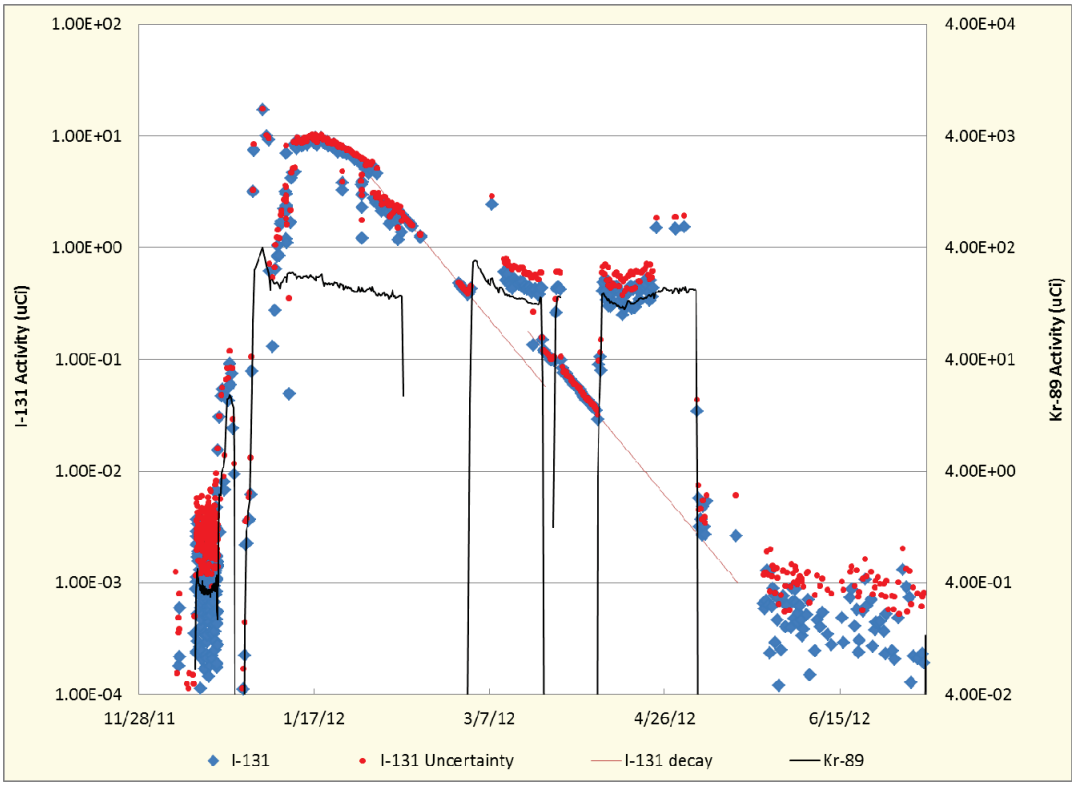

Fig. 11: ^{131}I accumulation in FPM 7 for the first 2 irradiation cycles of AGR-3/4.

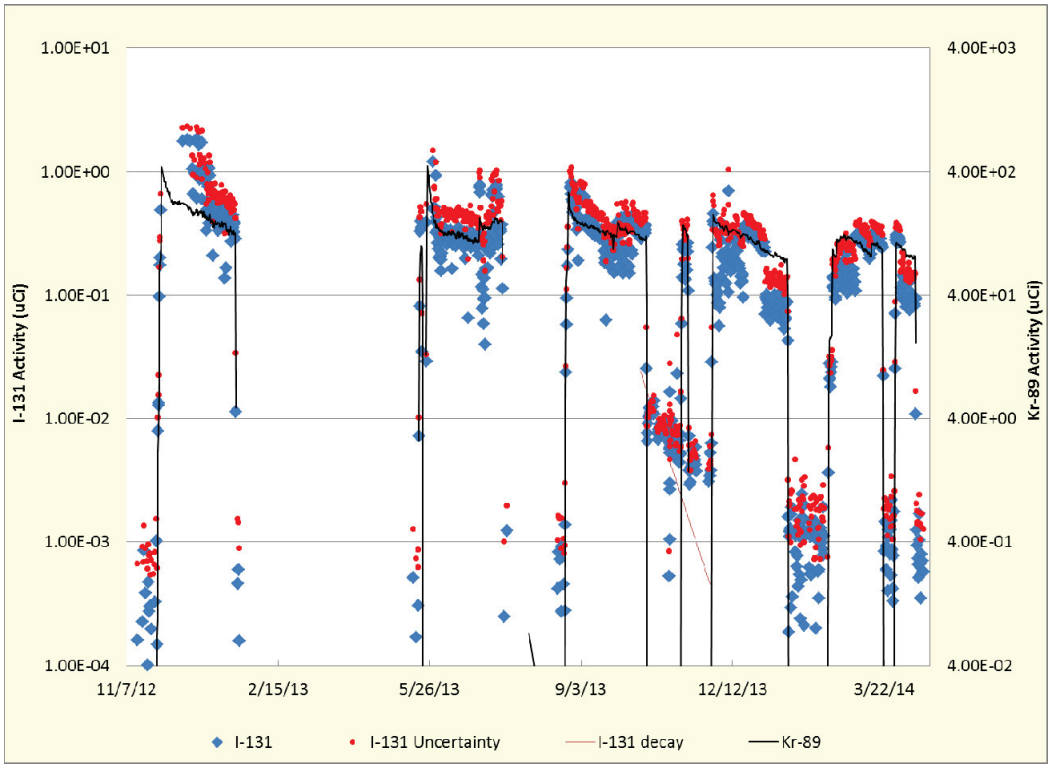

Fig. 12: ^{131}I accumulation in FPM 7 for the last 5 irradiation cycles of AGR-3/4.

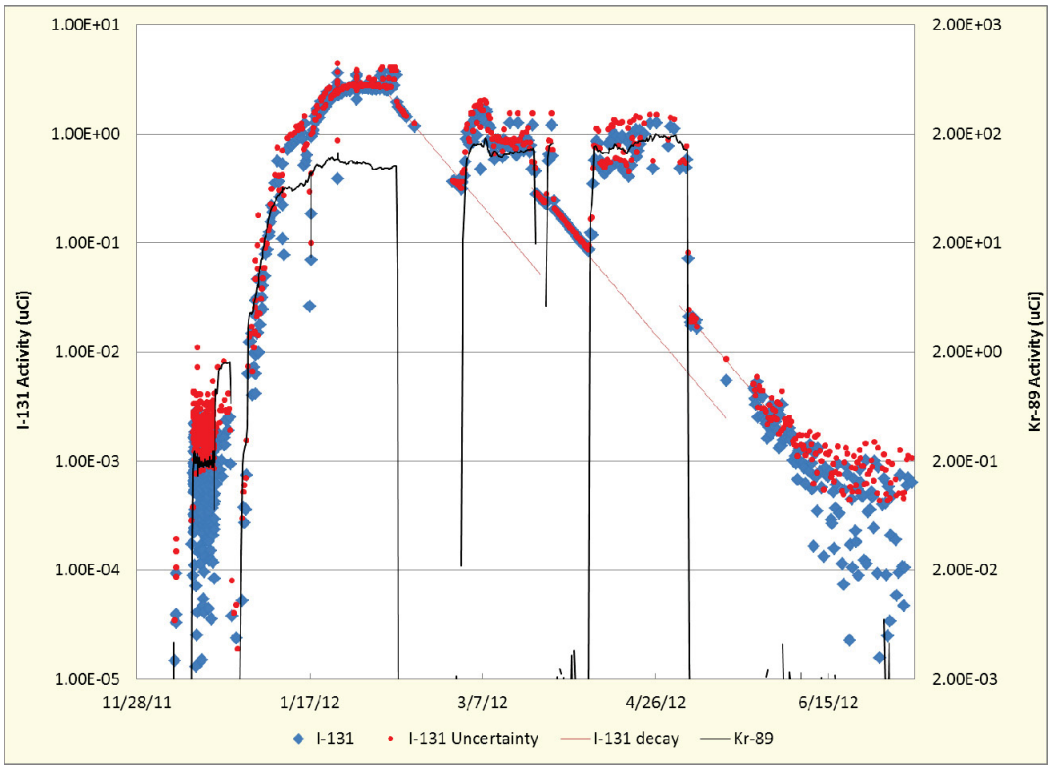

Fig. 13: ^{131}I accumulation in FPM 8 for the first 2 irradiation cycles of AGR-3/4.

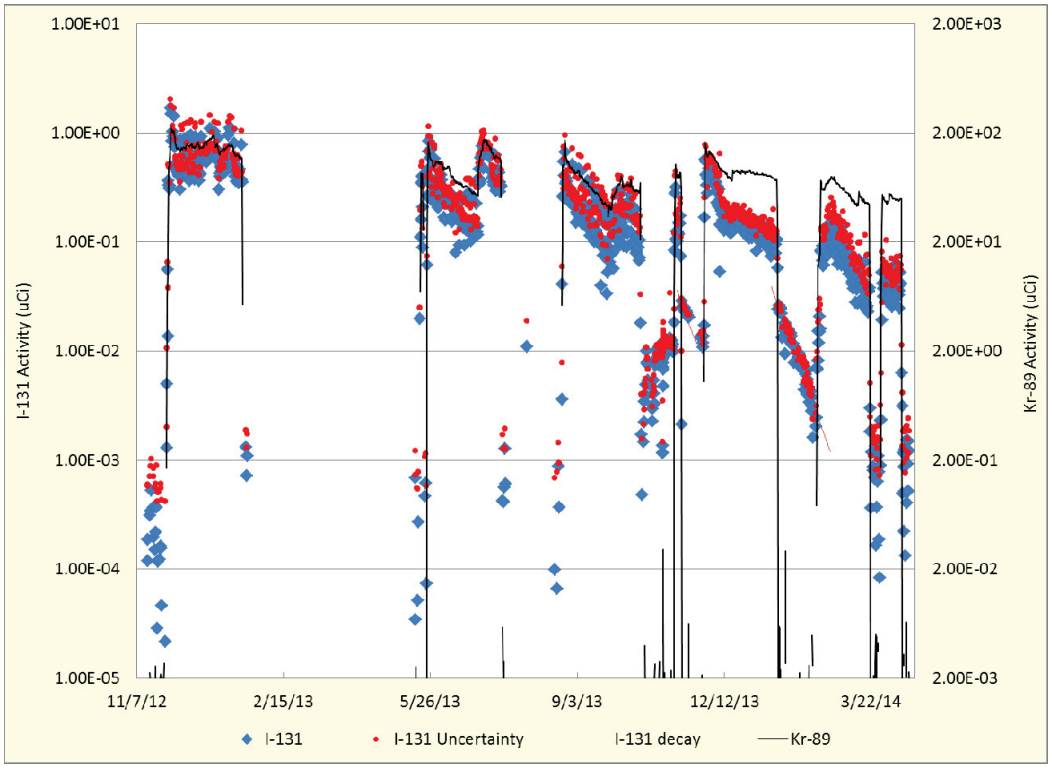

Fig. 14: ^{131}I accumulation in FPM 8 for the last 5 irradiation cycles of AGR-3/4

VI. CONCLUSION

Three Advanced Gas Reactor experiments using TRISO fuel have been conducted in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL). From 2006 through 2014, these experiments supported the development and qualification effort of the new U.S. TRISO particle fuel for Very High Temperature Reactors. During these multi-year experiments, capsule specific activities for xenon and krypton species were measured during irradiation and during reactor shutdown to provide isotopic information that could be used to compute release activities relative to fuel performance.

The concentration of ^{135}I from these three AGR experiments was determined from measured ^{135m}Xe after the reactor shutdown. The amount of ^{135}I present can be an indicator of fuel integrity. For AGR-1, the release fraction for ^{135}I indicated that there were no failed particles and only heavy metal contamination outside of the intact silicon carbide layer of TRISO particles was present. Experiment temperature influenced the amounts of ^{135}I observed. In the case of AGR-2 and AGR-3/4, the release fraction was higher because of initially exposed fuel kernels in AGR-2 and failure of designed-to-fail (DTF) fuel in the AGR-3/4 experiment.

To study the effect of moisture on failed TRISO fuel, an impure gas mixture was injected into capsule 11 in the AGR-3/4 experiment. As moisture began to build up in capsule 11, the failed kernel may have become hydrolyzed and some of the ^{131}I mobilized and carried downstream to the FPM sample chamber. Small amounts of moisture were apparently present at the beginning of irradiation for capsules 7 and 8 in excess of test specifications; however exact moisture concentrations are unknown. The presence of ^{131}I is unmistakably visible in FPM sample chambers for capsules 7, 8 and 11 and there is no evidence of ^{131}I in the other capsule specific FPM sample chambers. ^{131}I release fractions were all below 1 ppm.

ACKNOWLEDGMENT

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