Determination of the Quantity of I-135 Released from the AGR-1 Test Fuels at the End of ATR Operating Cycle 138B

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ABSTRACT

The AGR-1 experiment is a multiple capsule fuel irradiation experiment being conducted in the Advanced Test Reactor (ATR) in support of the Advanced Gas Reactor (AGR) Fuel Development and Qualification Program. The experiment began irradiation in the ATR with cycle 138B that reached full power on December 26, 2006 and was temporarily shutdown for a brief reactor outage on February 10, 2007 at 0900. The AGR-1 experiment will continue irradiation for about 2.5 years. In order to allow estimation of the amount of radioiodine released during the first cycle, purge gas flow to all capsules continued for about 4 days after reactor shutdown. The Fission Product Monitoring System (FPMS) data acquired during part of that shutdown flow period has been analyzed to determine the level of ¹³⁵I released during the later portions of the operating cycle.

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1. Introduction

The AGR-1 experiment is a multiple capsule fuel irradiation experiment being conducted in the Advanced Test Reactor (ATR) in support of the Advanced Gas Reactor (AGR) Fuel Development and Qualification Program. The experiment began irradiation in the ATR with cycle 138B that reached full power on December 26, 2006 and ended with shutdown of the reactor for a brief outage on February 10, 2007 at 0900. The AGR-1 experiment will continue cyclical irradiation for about 2.5 years. In order to allow determination of the amount of radioiodine released during the first cycle, purge gas flow to all capsules continued for about 4 days after reactor shutdown. The FPMS data acquired during part of that shutdown flow period has been analyzed to elucidate the level of ¹³⁵I released.

The AGR-1 test train (the in-core portion of the experiment) incorporates six individual test capsules. Each capsule contains about 51,000 TRISO (TRIstructural ISOtopic) coated uranium oxicarbide fuel particles supported in a graphite matrix. Each capsule is continuously swept with an inert gas during irradiation. Details on the experiment and its design have been published.²

For intact fuel particles, the TRISO coating provides a barrier to fission product release. However, particles with failed TRISO coatings, either those that fail during irradiation or the tiny percentage of particle that are initially defective, can release fission products directly to the flowing gas stream. Because reactive fission products like the radioiodines and cesiums quickly deposit on test train and piping structures, the only released fission products that move downstream unattenuated are the noble fission gas isotopes of Kr and Xe. One important measure of the fuel performance in these tests is quantification of the fission product releases over the duration of each irradiation experiment. To provide this important data for the inert fission gas releases, a Fission Product Monitoring System (FPMS) has been developed and implemented to monitor the individual capsule effluents for radioactive species.³ There is however, a technical interest in gathering release information for species other than noble gasses.

Due to some operational difficulties with the AGR-1 auxiliary equipment during the cycle 138B irradiation, the utility of a detailed analysis of the FPMS-acquired fission gas release data is being studied. However, we have taken this opportunity to analyze some of the fission gas data taken following reactor shutdown in order to provide insight into releases of ¹³⁵I, a reactive fission product that if released from the fuel particles deposits in the test train and piping and thus never reaches the FPMS. This report details the methods, techniques and results of these measurements.

2. Background

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 135 Xe ($T_{1/2}$ = 9.1 hours). Among the populated states of 135 Xe is an isomeric state – 135m Xe – that decays by isomeric transition to the 135 Xe ground state with a half life of 15.3 minutes and the emission of a 526.6 keV photon. The 15.3 minute isomeric state is populated through a 16.4% branch of the 135 I decays 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 1.⁴

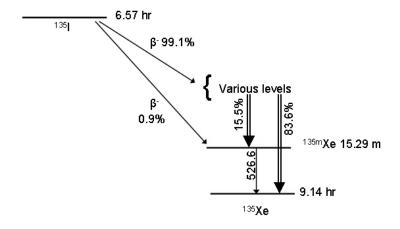


Figure 1 Simplified decay scheme of ¹³⁵I

During the AGR-1 irradiation, ¹³⁵Xe and ^{135m}Xe concentrations detected at the FPMS were generated both from direct fission yield and from the decay of their ¹³⁵I parent; however, by about 2.5 hours (ten 15.3 minute half lives) after the irradiation stops, the only source of ^{135m}Xe is the decay of its 6.6 hour ¹³⁵I parent. Further, decay systematics dictate that the ^{135m}Xe will be in secular equilibrium with its ¹³⁵I parent, its production rate will equal the decay rate of the parent, and its concentration in the flowing stream will appear to decay with the parent half-life.⁵ This equilibrium arrangement enables determination of the amount of ¹³⁵I 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.

3. Techniques and Methods

3.1 Equipment and Measurements

The FPMS is a collection of on-line gross radiation monitors and gamma-ray spectrometers that quantify radionuclide content of the test effluent from each of the six test capsules in the AGR-1 experiment. This system has been described elsewhere.³ Each spectrometer views a 58 cm³ volume of the gas effluent from its associated test train capsule. Each spectrum when analyzed provides the activity of fission gases in 58 cm³ of the flowing gas stream from the associated capsule.

Estimated transport times from each capsule to its measurement point are computed using the capsule-specific transport volumes determined from data acquired during a set of tests documented elsewhere, and the output flow rates registered by the capsule-specific outlet flowmeters. Although the outlet flowmeters were not calibrated until later in this outage, there is agreement between the total inlet and total measured outlet flows, and the small differences between these two sums allow estimation of an uncertainty in the flow rate readings. It is necessary to use the capsule *outlet* flows rather than the nominal 15 cm³/min capsule *inlet* flow, since during this measurement, the gas supply to the *leadout* was 50 cm³/min, and this entire supply of gas must exit through the collective capsules. ^{2,6}

Spectra with counting durations (Real Time) of 8 hours were recorded and analyzed prior to, during, and following the reactor scram at 0900 10-FEB-2007. Here we consider the spectra acquired with nominal start times of 1300, and 2100 on 10-FEB-2007, and 0500 on 11-FEB-2007 for the effluent of each of capsules 2 through 6. Capsule 1 was compromised by a failed valve early in the irradiation and is excluded here. These 8-hour-duration spectra started at nominal delay times of 4, 12, and 20 hours following reactor shutdown. Xe-135m, clearly the decay daughter of ¹³⁵I, was detected in these spectra. The measured results are presented in Table 1.

Table 1 Post-shutdown ^{135M}Xe activities measured in the capsule effluents

		Decay time ^a	^{135m} Xe activity	
Capsule	Spectral ID	(hours)	$(Bq/58cm^3)$	%Error
2	G2702101300	4.017	23.28	7.8%
	G2702102100	12.033	7.74	13.2%
	G2702110500	20.033	2.52	36.2%
3	G3702101300	4.017	19.48	7.5%
	G3702102100	12.033	6.57	16.8%
	G3702110500	20.033	3.38	25.4%
4	G4702101300	4.017	17.67	9.4%
	G4702102100	12.017	8.17	17.9%
	G4702110500	20.017	3.46	39.8%
5	G5702101300	4.017	18.76	11.6%
	G5702102100	12.017	7.56	14.5%
	G5702110500	20.017	2.97	36.9%
6	G6702101300	4.017	8.81	15.5%
-	G6702102100	12.017	4.42	22.5%
	G6702110500	20.017	1.73	50.9%

^a The listed decay time is the duration in hours from reactor scram to the start of the listed spectrum.

3.2 Calculations and Corrections

The measured results require correction for a number of decay periods in order to calculate the ^{135m}Xe activity at the test capsule (which is the presumed point of equilibrium with its ¹³⁵I parent.) Figure 2 is provided to facilitate the following discussion.

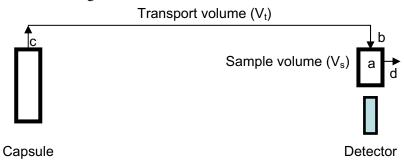


Figure 2 Single capsule flow diagram

Released 135 I is presumed to be firmly deposited in the capsule and does not move downstream. Daughter $^{135\text{m}}$ Xe is in decay 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 flowmeter, moves the $^{135\text{m}}$ Xe through the transport line volume (V_t) to the entrance of the sample counting chamber (b), the gas then sweeps through the sample volume (V_s) filling the sample chamber (a), and then flowing out of the sample chamber at (d). As the sample continuously flows, the detector acquires a spectrum that starts at some time of day and continues for a count time of RT seconds.

3.2.1 Understanding Secular Equilibrium

It is important to understand the effect of the secular equilibrium relationship between ¹³⁵I and its ^{135m}Xe daughter in order to formulate the required corrections properly. When the ^{135m}Xe (15.3 minutes) is collocated with its ¹³⁵I (6.6 hour) parent at the capsule (c), secular equilibrium is established, the ^{135m}Xe and ¹³⁵I activities are nearly equal, and the ^{135m}Xe activity decays with an apparent half life equal to that of its long lived ¹³⁵I 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 online measurements such as those reported here, 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 (like b) the measured ^{135m}Xe activity changes in time according to the 6.6 hour ¹³⁵I 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 ¹³⁵I 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.

3.2.2 Formulation of the Required 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:

$$C_{Cap}(t_r) = \frac{A(t_r)}{V_s} * f_1 * f_2 * f_3$$

Where:

 $C_{can}(t_r)$ = the desired Xe-135m concentration (Bq/cm³) at the capsule exit at a time t_r after

 $A(t_r)$ = the activity (Bq) of Xe-135m calculated from a spectrum that started at time t_r after reactor scram

 V_s = the sample volume viewed by the spectrometer

 f_1 = a factor to correct the activity reported by the spectral analysis code to the start of the spectral acquisition

 f_2 = a factor to correct for decay of the Xe-135m during transport through volume V_t between the capsule outlet and the sample chamber inlet

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

The derivations of the decay correction equations are presented in Appendix A.

3.2.2.1 Correction to the start of the spectral acquisition (f_1)

Because of the on-line nature of the FPMS data, the automatic analysis software applies no decay correction. The reported activity is the average activity over the 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:

$$f_1 = \frac{\lambda_I * RT}{(1 - e^{-\lambda_I * RT})}$$

Where:

 λ_I = the ¹³⁵I decay constant of 2.931 X 10⁻⁵ s⁻¹ RT = the spectral acquisition time (real time) in s

Note that since the activity at the detector varies with the ¹³⁵I half life the ¹³⁵I decay constant is appropriate for this correction. During this work the spectral acquisition times (RT) were 8 hours or 2.88 $X10^4$ s, thus f_1 equals 1.481 and any uncertainties are negligible.

3.2.2.2 Correction for decay during transport (f₂)

When the ^{135m}Xe is swept from the capsule and transported to the sample chamber inlet it decays during the transport time with a decay constant equal to the difference between the 135m Xe decay constant (7.551E-04 s⁻¹) and the 135 I decay constant (2.931E-05 s⁻¹.) The transport time is determined by the capsule outlet flow rate and the transport volume (Vt). The capsule outlet flow rates are available from flowmeter data logged during the experiment. Capsule-specific transport volumes have been determined in another work. 6

The correction factor f_2 can be computed as:

$$f_2 = e^{\frac{(\lambda_{Xe} - \lambda_I) * V_t}{\dot{f}}}$$

where:

 $\begin{array}{lll} \lambda_{Xe} &=& the^{~135m}Xe~decay~constant~(7.556~X10^{-4}~s^{-1})\\ \lambda_{I} &=& the^{~135}I~decay~constant~(2.931E-05~s^{-1})\\ V_{t} &=& the~capsule-specific~transport~volume~(cm^{3})^{~6} \end{array}$

 \dot{f} = the average capsule outlet flow rate during the spectral measurement.

The relative variance of f₂ can be estimated by normal error propagation techniques as:

$$\left(\frac{\sigma_{f_2}}{f_2}\right)^2 = \left(\frac{(\lambda_{Xe} - \lambda_I) * V_t}{\dot{f}}\right)^2 * \left[\left(\frac{\sigma_{V_t}}{V_t}\right)^2 + \left(\frac{\sigma_{\dot{f}}}{\dot{f}}\right)^2\right]$$

Table 2 presents the average capsule outlet flows computed from the flowmeter data for the measurement times of interest, the capsule-specific transport volumes, and the computed value of the f₂ correction along with estimated uncertainties on these values.

Table 2 The f_2 correction and its input parameters

	1 ransport		
	volume		
	V_{t}	Mean outlet flowrate	e Correction factor
Caps	ule (cm ³)	(cm^3/s)	f_2
2	138 ± 7	0.515 ± 0.014	1.21 ± 0.01
3	128 ± 9	0.502 ± 0.014	1.20 ± 0.02
4	128 ± 9	0.288 ± 0.008	1.38 ± 0.03
5	120 ± 8	0.68 ± 0.02	1.14 ± 0.01
6	128 ± 9	0.410 ± 0.013	1.25 ± 0.02

The decay correction procedure outlined above presumes that the ¹³⁵I 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 I₂), deposition might occur well downstream from the capsule. Scoping calculations⁷ 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 0.5 cm³/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 ¹³⁵I were deposited at the reactor vessel exit, some 8 to 12 seconds closer to the spectrometers than was assumed, then the f₂ correction factors reported in Table 2 would have less than a 1% bias. Due to the somewhat speculative nature of this uncertainty it has not been propagated to the tabular results in this work.

3.2.2.3 Correction for holdup in the sample chamber (f_3)

The sample chamber (a) viewed by the detector has a volume of V_s . Note that 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 (Fig. 2). 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:

$$f_3 = \frac{(\lambda_{Xe} - \lambda_I) * V_s}{\dot{f} * \left(1 - e^{\frac{-(\lambda_{Xe} - \lambda_I) * V_s}{\dot{f}}}\right)}$$

For the measurements performed here, $V_S = 58 \pm 2$ cm³. The other parameter values have been specified earlier. Table 3 presents the results for correction factor f_3 .

Table 3	The values	of the	correction	factor	fз

Capsule	Correction factor f ₃
2	1.041 ± 0.003
3	1.043 ± 0.003
4	1.075 ± 0.005
5	1.031 ± 0.002
6	1.052 ± 0.004

3.2.3 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. Again, the relevant equation is:

$$C_{Cap}(t_r) = \frac{A(t_r)}{V_s} * f_1 * f_2 * f_3$$

All the relevant parameters have been presented. Note that since the all of the operands in this equation are multiplicative, that if the parameters are non-correlated (a reasonable assumption), the relative uncertainty in the value can be computed by straight forward quadrature propagation of the individual relative uncertainties. The ^{135m}Xe release concentrations are presented in Table 4.

In order to ensure that the $^{135\text{m}}$ Xe release concentrations were decaying with an apparent 6.6 hour 135 I half-life, and to determine the $^{135\text{m}}$ Xe releases at reactor shutdown ($t_D = 0$), release concentrations for each

Table 4 Xe-135m release concentrations

			^{135m} Xe activity	
		Decay time ^a	Bq/cm^3	
Capsule	Spectral ID	(hours)	@capsule	%Error
2	G2702101300	4.017	0.752	7.9%
	G2702102100	12.033	0.250	13.3%
	G2702110500	20.033	0.081	36.2%
3	G3702101300	4.017	0.624	7.6%
	G3702102100	12.033	0.210	16.8%
	G3702110500	20.033	0.108	25.4%
4	G4702101300	4.017	0.669	9.8%
	G4702102100	12.017	0.309	18.1%
	G4702110500	20.017	0.131	39.9%
5	G5702101300	4.017	0.561	11.6%
	G5702102100	12.017	0.226	14.6%
	G5702110500	20.017	0.089	36.9%
6	G6702101300	4.017	0.297	15.6%
	G6702102100	12.017	0.149	22.5%
	G6702110500	20.017	0.058	50.9%

^a The listed decay time is the duration in hours from reactor scram to the start of the listed spectrum

capsule were plotted and fit, using a variance-weighted, nonlinear least squares fit to a linear equation of the form:

$$y(t) = y_0 + A * \exp(-0.1055 * t_D)$$

where:

 $y(t) = {}^{135m}Xe$ activity concentration (Bq/cm³) at decay time t

 y_0 = the fitted intercept

A =the fitted slope

0.1055 = the decay constant of 135 I in hr⁻¹

 t_D = the decay time (hr) from reactor scram

The points should show good correlation with the fitted form, and when the fit equation is evaluated at t_D = 0 it yields the 135m Xe release concentration (Bq/cm³) at reactor shutdown. Dividing this value by the known outlet flow rate then provides the 135m Xe release rate (Bq/s) from which the desired deposited 135 I parent activity can be determined.

4. Results

The corrected ^{135m}Xe concentrations have been plotted and fit as described in the previous section. Plots and details of these fits are presented in the Appendix B. All of the measured ^{135m}Xe results are well fit by a line that assumes the ¹³⁵I half life. An example plot of the capsule 4 data (points) and the fitted line is presented in Figure 3. The fit parameters for capsules 2 through 6 are presented in Table 5

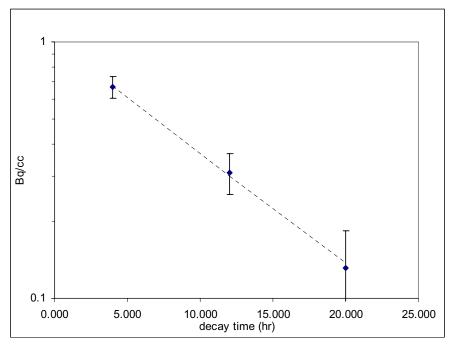


Figure 3 Comparison of the calculated ^{135m}Xe release concentration (Bq/cm³) data for capsule 4 with a line fitted presuming the ¹³⁵I half life of 6.57 hours. The x axis is time since reactor scram in hours.

 Table 5 Parameters for the fitted results by capsule

						Correlation Coefficient
Capsule	y_0	$\pm 1 \sigma$	A	$\pm 1 \sigma$	$Covar(y_0,A)$	R
2	-0.0793	0.0278	1.241	0.093	-2.137E-03	0.9972
3	-0.0229	0.0413	0.959	0.126	-4.210E-03	0.9914
4	0.0159	0.0139	1.005	0.037	-4.211E-04	0.9993
5	-0.0196	0.0045	0.882	0.015	-5.601E-05	0.9999
6	0.0105	0.0160	0.449	0.047	-6.181E-04	0.9945

Substituting t = 0 into each fitted equation yields the 135m Xe release concentration in (Bq/cm 3) at the time of reactor scram. Following standard error propagation techniques 8 the uncertainty on each t = 0 concentration can be estimated as:

$$\sigma_{v(0)}^2 = \sigma_{v_0}^2 + \sigma_A^2 + 2[covar(y_0, A)].$$

Given the release concentration (Bq/cm³) of ^{135m}Xe at the capsule at reactor shutdown, the measured capsule outlet flow rates, and the relevant nuclear data; the number of previously released atoms of ¹³⁵I required to produce the noted activity of ^{135m}Xe can be computed in a straightforward manner. The results of these calculations are presented in Table 6.

Table 6 Calculation of the number of ¹³⁵I atoms outside of the fuel and upstream of the FPMS at reactor shutdown from the 138B operating cycle

	Daughter Xe-135m at Capsule at Shutdown	Ave flow	Xe- 135m release rate in	Corresponding Atoms	Activity of released I-135 at end of
Capsule	(Bq/cc)	rate in cc/s	atoms/s	I-135 ^{a,b}	irradiation (Bq)
2	1.16 ± 0.10	0.515 ± 0.014	792±70	$(1.65\pm0.14)E+08$	$(4.8\pm0.4)E+03$
3	0.94 ± 0.13	$0.502 \pm .014$	622 ± 90	$(1.29\pm0.19)E+08$	$(3.8\pm0.6)E+03$
4	1.02 ± 0.04	$0.288 \pm .008$	390±18	$(8.1\pm0.4)E+07$	$(2.38\pm0.11)E+03$
5	0.863 ± 0.016	$0.68 \pm .02$	777±27	$(1.62\pm0.06)E+08$	$(4.74\pm0.16)E+03$
6	0.46 ± 0.06	0.410 ± 0.013	249 ± 28	$(5.2\pm0.6)E+07$	$(1.52\pm0.17)E+03$

^aIncludes correction for 16.4% decay branch to ^{135m}Xe.

^bRead (1.65 ± 0.14) E+08 as 1.65E+08 ± 0.14 E+08.

5. Conclusion

A methodology has been demonstrated for determining the amount of previously released ¹³⁵I from measurement of the ^{135m}Xe concentrations in the capsule effluents noted at the AGR FPMS following shutdown of the ATR. At the conclusion of ATR operating cycle 138B the activity of released ¹³⁵I for each test fuel capsule was determined to be less than 0.2 microCuries.

6. References

1. Advanced Gas Reactor Team, M. A. Feltus, Program Manager, D. A. Petti, R. R. Hobbins, J. M. Kendall, and J. J. Saurwein, Editors, "Technical program plan for the Advanced Gas Reactor fuel development and qualification program," Idaho National Laboratory, Idaho Falls, ID, Tech. Rep. *INL/EXT-05-00465 Revision 1*, Aug. 2005.

- 2. S. B. Grover, "Completing the Design of the Advanced Gas Reactor Fuel Development and Qualification Experiments for Irradiation in the Advanced Test Reactor," INL Report *INL/CON-07-12136*, presented at *ICAPP 2007*, the *2007 International Congress on Advances in Nuclear Power Plants*, May 13-18, 2007, Nice Acropolis, France.
- 3. J. K. Hartwell, D. M. Scates, M. W. Drigert, and J. B. Walter, "Installation and Final Testing of an On-Line, Multi-Spectrometer Fission Product Monitoring System (FPMS) to Support Advanced Gas Reactor (AGR) Fuel Testing and Qualification in the Advanced Test Reactor," *IEEE 2006 Nuclear Science Symposium Conference Record*, Oct. 29 Nov. 4, 2006, San Diego, CA. Also, to be published in *IEEE Trans on Nuclear Science*, Vol. 54, No. 3, June 2007, pages TBD.
- 4. All of the nuclear decay data is from: R. B. Firestone, *Table of Isotopes, 8th Ed.*, V. S. Shirley, Editor, John Wiley & Sons, NY, 1996.
- 5. See for example: G. Friedlander, J. W. Kennedy, and J. M. Miller, *Nuclear and Radiochemistry*, John Wiley & Sons, NY, 1949.
- 6. J. K. Hartwell, J. B. Walter, D. M. Scates, and M. W. Drigert, "Determination of the AGR-1 capsule to FPMS spectrometer transport volumes from leadout flow test data," INL Technical Report *INL/EXT-07-12494*, May 2007.
- R. A. Laviolette, e-mail communication to J. K. Hartwell, "Final Report: Programs PVAP and DENSITY," 9/29/2004.
- 8. See for example, W. E. Deming, Statistical Adjustment of Data, John Wiley and Sons, London, 1948.

Appendix A

Derivation of the Decay Corrections Systematics for the On-Line Determination of I-135 From Measured Xe-135m Activities

Postulate an activity of $\lambda_I N_I$ in or near the capsule with an effluent flow rate \dot{f} , transport volume V_t , detection volume V_s , and count time Δ . What is the measured activity of 135m Xe for a count started at t_i given a branching ratio of B_r ? The 135 I is fixed, and the decay products are carried away with the effluent gas. For N_{I0} present at time t=0, we have at time t

$$N_I(t) = N_{I0}e^{-\lambda_I t}$$

This will result in

$$R_c(t) = B_r \lambda_I N_{I0} e^{-\lambda_I t}$$

atoms/s of 135m Xe leaving the vicinity of the 135 I deposition. Designating the number/s arriving at and leaving the sample volume as R_b and R_d respectively, and N_{Xe} the number in the sample volume, we have

$$\begin{split} R_{b}(t) &= R_{c}(t - V_{t} / \dot{f}) e^{-\lambda_{Xe}V_{t} / \dot{f}} = B_{r} \lambda_{I} N_{I0} e^{-\lambda_{I} \dot{t}} e^{-(\lambda_{Xe} - \lambda_{I})V_{t} / \dot{f}} \\ R_{d}(t) &= R_{b}(t - V_{s} / \dot{f}) e^{-\lambda_{Xe}V_{s} / \dot{f}} \\ &= B_{r} \lambda_{I} N_{I0} e^{-\lambda_{I} \dot{t}} e^{-(\lambda_{Xe} - \lambda_{I})V_{t} / \dot{f}} e^{\lambda_{I}V_{s} / \dot{f}} e^{-\lambda_{Xe}V_{s} / \dot{f}} \\ &= B_{r} \lambda_{I} N_{I0} e^{-\lambda_{I} \dot{t}} e^{-(\lambda_{Xe} - \lambda_{I})V_{t} / \dot{f}} e^{-(\lambda_{Xe} - \lambda_{I})V_{s} / \dot{f}} \\ &= B_{r} \lambda_{I} N_{I0} e^{-\lambda_{I} \dot{t}} e^{-(\lambda_{Xe} - \lambda_{I})(V_{t} + V_{s}) / \dot{f}} \\ N_{Xe}'(t) &= R_{b}(t) - R_{d}(t) - \lambda_{Xe} N_{Xe}(t) = -\lambda_{I} N_{Xe}(t) \\ N_{Xe}(t) &= \frac{R_{b}(t) - R_{d}(t)}{\lambda_{Xe} - \lambda_{I}} \\ &= B_{r} \frac{\lambda_{I}}{\lambda_{Xe} - \lambda_{I}} N_{I0} e^{-\lambda_{I} \dot{t}} \left[e^{-(\lambda_{Xe} - \lambda_{I})V_{t} / \dot{f}} - e^{-(\lambda_{Xe} - \lambda_{I})(V_{t} + V_{s}) / \dot{f}} \right] \\ &= B_{r} \frac{\lambda_{I}}{\lambda_{Xe} - \lambda_{I}} N_{I0} e^{-\lambda_{I} \dot{t}} e^{-(\lambda_{Xe} - \lambda_{I})V_{t} / \dot{f}} \left[1 - e^{-(\lambda_{Xe} - \lambda_{I})V_{s} / \dot{f}} \right] \end{split}$$

The measured activity is then (ignoring detector efficiency)

$$A_{Xe}(t_i) = \frac{1}{\Delta} \int_{t_i}^{t_i + \Delta} \lambda_{Xe} N_{Xe}(t) dt$$

$$A_{Xe}(t_{i}) = \frac{\lambda_{I}B_{r}N_{I0}}{\lambda_{Xe} - \lambda_{I}} e^{-(\lambda_{Xe} - \lambda_{I})V_{t}/f} \left[1 - e^{-(\lambda_{Xe} - \lambda_{I})V_{s}/f} \right] \times \frac{1}{\Delta} \int_{i}^{t_{i}+\Delta} \lambda_{Xe} e^{-\lambda_{I}t} dt$$

$$= \frac{\lambda_{I}B_{r}N_{I0}}{\lambda_{Xe} - \lambda_{I}} e^{-(\lambda_{Xe} - \lambda_{I})V_{t}/f} \left[1 - e^{-(\lambda_{Xe} - \lambda_{I})V_{s}/f} \right] \times \lambda_{Xe} e^{-\lambda_{I}t_{i}} \frac{1 - e^{-\lambda_{I}\Delta}}{\lambda_{I}\Delta}$$

Solving for $N_I(t)$, or $N_{I0}e^{-\lambda_I t_i}$

$$\begin{split} N_{I}(t_{i}) &= A_{Xe}(t_{i}) \frac{(\lambda_{Xe} - \lambda_{I})}{\lambda_{Xe} \lambda_{I} B_{r}} \frac{\lambda_{I} \Delta}{1 - e^{-\lambda_{I} \Delta}} e^{(\lambda_{Xe} - \lambda_{I})V_{t} / \dot{f}} \frac{1}{1 - e^{-(\lambda_{Xe} - \lambda_{I})V_{s} / \dot{f}}} \\ &= \frac{A_{Xe}(t_{i})}{\lambda_{Xe} \lambda_{I} B_{r}} \frac{\lambda_{I} \Delta}{1 - e^{-\lambda_{I} \Delta}} e^{(\lambda_{Xe} - \lambda_{I})V_{t} / \dot{f}} \frac{(\lambda_{Xe} - \lambda_{I})V_{s} / \dot{f}}{1 - e^{-(\lambda_{Xe} - \lambda_{I})V_{s} / \dot{f}}} \frac{\dot{f}}{V_{s}} \\ \lambda_{I} N_{I}(t_{i}) &= \frac{A_{Xe}(t_{i}) \dot{f}}{\lambda_{Ye} B_{r} V_{s}} \frac{\lambda_{I} \Delta}{1 - e^{-\lambda_{I} \Delta}} e^{(\lambda_{Xe} - \lambda_{I})V_{t} / \dot{f}} \frac{(\lambda_{Xe} - \lambda_{I})V_{s} / \dot{f}}{1 - e^{-(\lambda_{Xe} - \lambda_{I})V_{s} / \dot{f}}} \end{split}$$

To follow the protocol in the document, we note that the activity concentration of 135m Xe at the capsule exit at time t_i [$C_{Xe}(t_i)$] is given by:

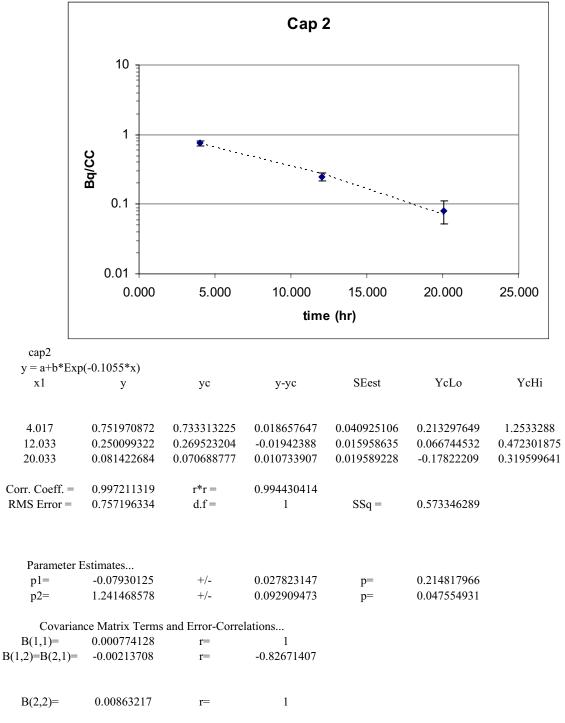
$$C_{Xe}(t_i) = \frac{N_I \lambda_I B_r \lambda_{Xe}}{\dot{f}} = \frac{A_{Xe}(t_i)}{V_S} \cdot f_1 \cdot f_2 \cdot f_3$$

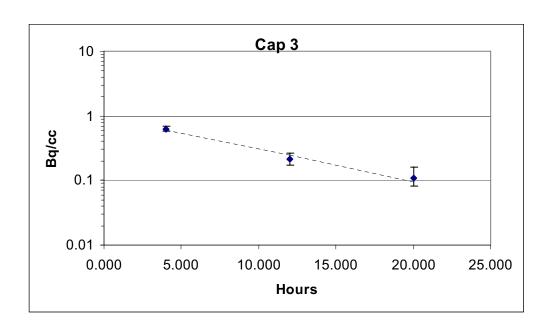
where the correction factors have the formulations used in the report.

Appendix B

Graphs and Fit Parameters for XE-135m Concentration Data

Note: All dashed lines in this Appendix are the results of the fit equations





cap 3 y = a+b*Exp(-0.1055*x)

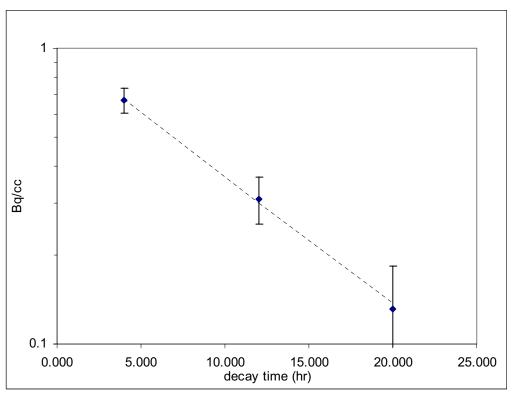
) a = -/-p(a.						
x1	У	ус	у-ус	SEest	YcLo	YcHi
4.017	0.624045213	0.604614715	0.019430498	0.055089635	-0.09538269	1.304612119
12.033	0.210345662	0.246483635	-0.03613797	0.024463827	-0.0643664	0.557333675
20.033	0.108277651	0.092946938	0.015330713	0.030319075	-0.29230292	0.478196797
Corr. Coeff. =	0.991422163	r*r =	0.982917904			
RMS Error =	1.233364964	d.f =	1	SSq =	1.521189135	

Parameter Estimates...

p1=	-0.02287291	+/-	0.041272083	p=	0.677831806
p2=	0.95864176	+/-	0.126377073	p=	0.08344397

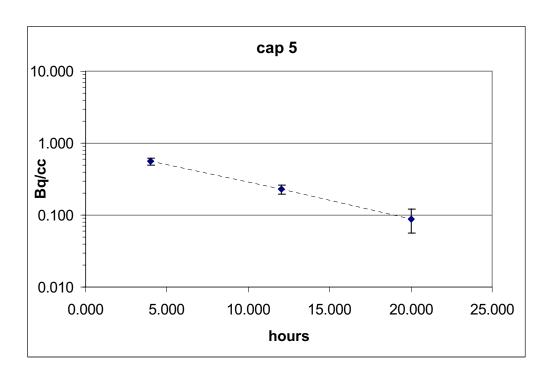
Covariance Matrix Terms and Error-Correlations...

B(1,1)=	0.001703385	r=	1
B(1,2)=B(2,1)=	-0.00420995	r=	-0.8071465
B(2,2)=	0.015971165	r=	1



Cap 4

cap 4						
y = a + b*Exp(-0.1)	055*x)					
x1	у	vc y	-yc S	Eest	YcLo Y	′cHi
4.017	0.669347974	0.673686329	-0.00433835	0.01475442	0.486209033	0.861163624
12.017	0.309307156	0.298735359	0.010571797	0.007844707	0.199056456	0.398414263
20.017	0.131052615	0.137511268	-0.00645865	0.010473161	0.004433859	0.270588677
Corr. Coeff. =	0.999333811r	*r =	0.998668066			
RMS Error =	0.2355028920	d.f =	18	SSq =	0.055461612	
Parameter Estim	ates					
p1=	0.015892463+	-/-	0.013852272p	=	0.456402562	
p2=	1.004942002+	-/-	0.036700476p	=	0.023239023	
Covariance Matri	ix Terms and Err	or-Correlations				
B(1,1)=	0.000191885r	=	1			
B(1,2)=B(2,1)=	-0.00042111r	=	-0.82832368			
B(2,2)=	0.001346925r	=	1			



cap 5
v = a+b*Exp(-0.1055*x)

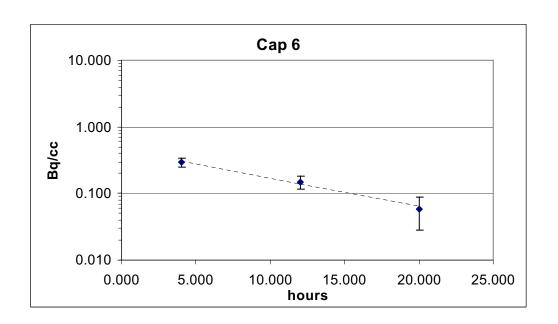
7 1 1	,					
x1	у	ус	у-ус	SEest	YcLo	YcHi
4.017	0.561	0.557832651	0.003167349	0.006470509	0.475615017	0.640050286
12.017	0.226	0.22867465	-0.00267465	0.002454489	0.197486648	0.259862651
20.017	0.089	0.087140945	0.001859055	0.003105195	0.047684727	0.126597163
Corr. Coeff. =	0.999857205	r*r =	0.99971443			
RMS Error =	0.110273022	d.f =	1	SSq =	0.012160139	

Parameter Estimates...

p1=	-0.01962449	+/-	0.004465849	p=	0.142446604
p2=	0.882207884	+/-	0.014910395	p=	0.010758631

Covariance Matrix Terms and Error-Correlations...

B(1,1)=	1.99438E-05	r=	1
B(1,2)=B(2,1)=	-5.6014E-05	r=	-0.84120557
B(2,2)=	0.00022232	r=	1



cap 6

$y = a+b^*Exp(-0.7)$	1055°X)					
x1	У	ус	у-ус	SEest	YcLo	YcHi
4.017	0.297	0.304043974	-0.00704397	0.019965479	0.050352261	0.557735687
12.017	0.149	0.136693055	0.012306945	0.009166502	0.020218737	0.253167373
20.017	0.058	0.064734313	-0.00673431	0.011786122	-0.08502625	0.214494874
Corr. Coeff. =	0.994523961	r*r =	0.989077909			

1 SSq =

0.209458194

Parameter Estimates...

RMS Error =

p1=	0.010452499	+/-	0.015999625	p=	0.631595055
p2=	0.448533225	+/-	0.04713378	p=	0.066654086

Covariance Matrix Terms and Error-Correlations...

0.457666029 d.f =

B(1,1)=	0.000255988	r=	1
B(1,2)=B(2,1)=	-0.00061813	r=	-0.81966616
B(2,2)=	0.002221593	r=	1