# OAK RIDGE NATIONAL LABORATORY

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September 13, 2005

13336

RAS

Tamara D. Powell Office of Nuclear Material Safety & Safeguards U.S. Nuclear Regulatory Commission Mail Stop T8H7 Washington, DC 20555

Dear Tamara:

#### Estimated Production of Select Fission Products during a Criticality Accident

Enclosed is a revised letter report by Bryan Broadhead of ORNL documenting his analysis of the estimated consequences of a criticality accident for fuel enriched to 4 wt%, 5 wt%, and 10 wt%. This analysis was performed at the request of you and Chris Tripp as part of ORNL's technical support under JCN J5443. It has been peer reviewed at ORNL and is suitable for public release.

If you have any questions regarding the report, please contact Bryan or me.

Sincerely,

Stephen M. Bouman

S.M. Bowman, SCALE Project Leader Nuclear Science and Technology Division

Enclosure

cc: B. L. Broadhead C. V. Parks C. S. Tripp, NRC File NRC/J5443/LET

SEC4-07

### TEMPLATES SELY-027

### Estimated Production of Select Fission Products During a Criticality Accident

B.L. Broadhead (<u>broadheadbl@ornl.gov</u>) Oak Ridge National Laboratory Oak Ridge, Tennessee 37830-6170 September 13, 2005

In the event of an inadvertent nuclear criticality event, there are primarily two radioactive events of concern to individuals. The first event is the prompt release of a large number of neutrons and photons from single or repetitive criticality excursions. The second primary concern is the release of fission product gases and possibly particulate matter in the general vicinity of the accident resulting from the excursions. The duration of the first event is typically milliseconds, the latter a few minutes to several hours. Exposure to the initial burst is unavoidable for those individuals in the immediate vicinity of the accident. The prompt and effective evacuation of the area can easily mitigate radiation exposures due to subsequent criticality excursions and the releases that accompany them.

This document summarizes the calculation of selected fission products resulting from multiple criticality excursion power pulses. These fission products were selected since they are important components of the fission product release accompanying a criticality accident. The assumptions and methodology of this task follow closely the work described in Ref. 1 and represent an updating of a portion of that study.

The scenario of the fission events is unchanged from Ref. 1, in that  $1.6 \times 10^{18}$  fissions are assumed to occur during the first 30 minutes of the accident, followed by  $8.4 \times 10^{18}$  fissions up to 8 hours after the initial event. The initial pulse contains  $1 \times 10^{18}$  fissions, followed by 47 pulses of  $1.92 \times 10^{17}$  fissions each. Each pulse is assumed to last for 5 seconds with the next pulse following 10 minutes later. The fission product activities reported herein are estimated based on several approximations that should overpredict the actual activities. These approximations include the use of "cumulative" rather than "direct" fission product yields. Also, the fission products are not allowed to decay during the entire assumed criticality event. The degree of the fission-product-activity overprediction is estimated.

In this work, the ORIGEN-S<sup>2</sup> code was used only to estimate the fraction of the total number of fissions that occur in the various fuel solutions considered. The original work assumed a 4 wt%  $^{235}$ U solution; this work evaluates 4, 5, and 10 wt%  $^{235}$ U systems. The fraction of the total number of fissions that occur in the  $^{235}$ U and  $^{238}$ U isotopes is given in Table 1 below for each assumed enrichment.

Table 1. Fraction of total number of fissions								
Isotope	4 wt%	5 wt%	10 wt%					
<sup>235</sup> U	0.935	0.948	0.975					
<sup>238</sup> U	0.065	0.052	0.025					

 Table 1: Fraction of total number of fissions

This information along with fission product yield and decay data from the Evaluated Nuclear Data File<sup>3</sup> (ENDF/B-6) was used to construct an EXCEL spreadsheet to estimate the fission product activities. The formula for estimating the fission-product activity is as follows:

#### FP activity (Bq) = $\lambda$ (# fiss.)[(U-235 fiss. fract.)(FP Yield<sub>235</sub>) + (U-238 fiss. fract.)(FP yield<sub>238</sub>)]

The predicted fission product activities in Bq are given in Tables 2–4 for initial enrichments of 4, 5, and 10 wt%, respectively. The "initial Bq" corresponds to the "0–0.5 hr" results in Ref. 1, the "final Bq" corresponds to the "0.5-8 hr" column, and the "total Bq" values correspond to the "total" values. The first column in Tables 2–4 are the isotope labels, the second and third columns are the fission product yields in percent. The column labeled "lambda" is the decay constant in units of sec<sup>-1</sup>.

A comparison of the total activities predicted in this study for 4, 5, and 10 wt% enrichments is shown in Figure. 1, where the ratios of the 5 wt% to 4 wt% and 10 wt% to 4 wt% are given. The maximum difference of 15% is seen for the  $^{106}$ Ru isotope; however the 4% values are larger than those of either the 5 wt% or 10 wt%. The maximum difference seen for the remaining isotopes is only 3%.

ENDF/B-6 yields (%)			Fissions-> 1.58E+18 8.45E+18			
Isotope	235 yield	238 yield	lambda	Initial Bq	Final Bq	Total Bq
Kr-83m	0.5362	0.3959	1.05E-04	8.74E+	-11 4.68E+12	5.56E+12
Kr-85m	1.291	0.7429	4.30E-05	8.50E+	-11 4.56E+12	5.41E+12
Kr-85	0.2834	0.1486	2.05E-09	8.87E+	·06 4.75E+07	5.64E+07
Kr-87	2.558	1.625	1.51E-04	5.96E+	-12 3.19E+13	3.79E+13
Kr-88	3.552	2.026	6.78E-05	3.69E+	-12 1.98E+13	2.35E+13
Kr-89	4.511	2.672	3.64E-03	2.52E+	-14 1.35E+15	1.60E+15
Sr-91	5.827	4.039	2.02E-05	1.82E+	-12 9.76E+12	1.16E+13
Sr-92	5.938	4.312	7.10E-05	6.53E+	12 3.50E+13	4.15E+13
Ru-106	0.4015	2.49	2.16E-08	1.83E+	-08 9.81E+08	1.16E+09
Cs-137	6.188	6.053	7.32E-10	7.13E-	-07 3.82E+08	4.53E+08
Ba-139	6.413	5.67	1.37E-04	1.37E-	+13 7.34E+13	8.71E+13
Ba-140	6.214	5.815	6.29E-07	6.14E+	-10 3.29E+11	3.90E+11
Ce-143	5.956	4.622	5.83E-06	5.40E-	-11 2.89E+12	3.43E+12
Xe-131m	0.04047	0.04607	6.74E-07	4.34E+	-08 2.33E+09	2.76E+09
Xe-133	6.699	6.761	1.53E-06	1.62E+	-11 8.66E+11	1.03E+12
Xe-133m	0.1947	0.1959	3.66E-06	1.12E+	-10 6.03E+10	7.15E+10
Xe-135	6.538	6.968	2.11E-05	2.18E+	-12 1.17E+13	1.39E+13
Xe-135m	1.102	1.036	7.55E-04	1.31E+	-13 7.01E+13	8.31E+13
Xe-137	6.128	6.041	3.03E-03	2.92E+	-14 1.56E+15	1.86E+15
Xe-138	6.297	5.702	8.20E-04	8.09E+	-13 4.34E+14	5.15E+14
I-131	2.891	3.291	9.98E-07	4.59E+	-10 2.46E+11	2.92E+11
I-132	4.312	5.146	8.43E-05	5.80E+	-12 3.11E+13	3.69E+13
I-133	6.697	6.759	9.25E-06	9.77E+	-11 5.24E+12	6.22E+12
I-134	7.83	7.6	2.20E-04	2.70E+	13 1.45E+14	1.72E+14
I-135	6.282	6.941	2.93E-05	2.92E+	-12 1.57E+13	1.86E+13
Br-83	0.5362	0.3959	8.02E-05	6.66E+	-11 3.57E+12	4.24E+12
Br-84	0.9849	0.8224	3.63E-04	5.57E+	-12 2.99E+13	3.55E+13
Br-84m	0.01672	0.003189	1.93E-03	4.82E+	-11 2.58E+12	3.06E+12
Br-85	1.285	0.7428	4.02E-03	7.92E+	-13 4.24E+14	5.04E+14
Br-86	1.595	1.161	1.26E-02	3.11E+	-14 1.67E+15	1.98E+15
Br-87	2.035	1.537	1.24E-02	3.91E+	-14 2.10E+15	2.49E+15

## Table 2: Yield Predictions (in Bq) for 4 wt% solution

U-235 fiss U-238 fiss fraction fraction 0.935 0.065

	ENDF/B-6	/ields (%)		Fissions-> 1.58E+18	8.45E+18		U-235 fiss	U-238 fiśs
Isotope	235 yield	238 yield	lambda	Initial Bq	Final Bq	Total Bq	fraction	fraction
Kr-83m	0.5362	0.3959	1.05E-04	8.77E+11	4.70E+12	5.58E+12	0.948	0.052
Kr-85m	1.291	0.7429	4.30E-05	8.55E+11	4.58E+12	5.44E+12		
Kr-85	0.2834	0.1486	2.05E-09	8.92E+06	4.78E+07	5.68E+07		
Kr-87	2.558	1.625	1.51E-04	5.99E+12	3.21E+13	3.81E+13		
Kr-88	3.552	2.026	6.78E-05	3.71E+12	1.99E+13	2.36E+13		
Kr-89	4.511	2.672	3.64E-03	2.54E+14	1.36E+15	1.61E+15		
Sr-91	5.827	4.039	2.02E-05	1.83E+12	9.80E+12	1.16E+13		
Sr-92	5.938	4.312	7.10E-05	6.55E+12	3.51E+13	4.17E+13		
Ru-106	0.4015	2.49	2.16E-08	1.74E+08	9.31E+08	1.10E+09		
Cs-137	6.188	6.053	7.32E-10	7.13E+07	3.82E+08	4.54E+08		
Ba-139	6.413	5.67	1.37E-04	1.37E+13	7.35E+13	8.72E+13		
Ba-140	6.214	5.815	6.29E-07	6.14E+10	3.29E+11	3.91E+11		
Ce-143	5.956	4.622	5.83E-06	5.41E+11	2.90E+12	3.44E+12		
Xe-131m	0.04047	0.04607	6.74E-07	4.33E+08	2.32E+09	2.75E+09		
Xe-133	6.699	6.761	1.53E-06	1.62E+11	8.66E+11	1.03E+12		
Xe-133m	0.1947	0.1959	3.66E-06	1.12E+10	6.03E+10	7.15E+10		
Xe-135	6.538	6.968	2.11E-05	2.18E+12	1.17E+13	1.38E+13		
Xe-135m	1.102	1.036	7.55E-04	1.31E+13	7.01E+13	8.32E+13		
Xe-137	6.128	6.041	3.03E-03	2.92E+14	1.56E+15	1.86E+15		
Xe-138	6.297	5.702	8.20E-04	8.10E+13	4.34E+14	5.15E+14		
I-131	2.891	3.291	9.98E-07	4.58E+10	2.45E+11	2.91E+11		
I-132	4.312	5.146	8.43E-05	5.79E+12	3.10E+13	3.68E+13		
I-133	6.697	6.759	9.25E-06	9.77E+11	5.24E+12	6.22E+12		
I-134	7.83	7.6	2.20E-04	2.71E+13	1.45E+14	1.72E+14		
I-135	6.282	6.941	2.93E-05	2.92E+12	1.56E+13	1.86E+13		
Br-83	0.5362	0.3959	8.02E-05	6.69E+11	3.58E+12	4.25E+12		
Br-84	0.9849	0.8224	3.63E-04	5.59E+12	2.99E+13	3.55E+13		
Br-84m	0.01672	0.003189	1.93E-03	4.87E+11	2.61E+12	3.10E+12		
Br-85	1.285	0.7428	4.02E-03	7.96E+13	4.27E+14	5.06E+14		•
Br-86	1.595	1.161	1.26E-02	3.12E+14	1.67E+15	1.99E+15		
Br-87	2.035	1.537	1.24E-02	3.93E+14	2.10E+15	2.50E+15		

## Table 3: Yield Predictions (in Bq) for 5 wt% solution

		-							
	ENDF/B-6 yiel	ds (%)		Fissions->	1.58E+18	8.45E+18		U-235 fiss	U-238 fiss
Isotope	U-235 yield	238 yield	lambda		Initial Bq	Final Bq	Total Bq	fraction	fraction
Kr-83m	0.5362	0.3959	1.05E-04		8.83E+11	4.73E+12	5.62E+12	0.975	0.025
Kr-85m	1.291	0.7429	4.30E-05		8.65E+11	4.64E+12	5.50E+12		
Kr-85	0.2834	0.1486	2.05E-09		9.04E+06	4.85E+07	5.75E+07		
Kr-87	2.558	1.625	1.51E-04		6.05E+12	3.24E+13	3.85E+13		
Kr-88	3.552	2.026	6.78E-05		3.75E+12	2.01E+13	2.39E+13		
Kr-89	4,511	2.672	3.64E-03		2.56E+14	1.37E+15	1.63E+15		
Sr-91	5.827	4.039	2.02E-05		1.84E+12	9.88E+12	1.17E+13		
Sr-92	5.938	4.312	7.10E-05		6.60E+12	3.54E+13	4.20E+13		
Ru-106	0.4015	2.49	2.16E-08		1.55E+08	8.28E+08	9.83E+08		
Cs-137	6.188	6.053	7.32E-10		7.13E+07	3.82E+08	4.54E+08		
Ba-139	6.413	5.67	1.37E-04		1.38E+13	7.37E+13	8.75E+13		
Ba-140	6.214	5.815	6.29E-07		6.15E+10	3.30E+11	3.91E+11	•	
Ce-143	5.956	4.622	5.83E-06		5.44E+11	2.92E+12	3.46E+12		
Xe-131m	0.04047	0.04607	6.74E-07		4.31E+08	2.31E+09	2.74E+09		
Xe-133	6.699	6.761	1.53E-06		1.62E+11	8.66E+11	1.03E+12		
Xe-133m	0.1947	0.1959	3.66E-06		1.12E+10	6.03E+10	7.15E+10		
Xe-135	6.538	6.968	2.11E-05		2.17E+12	1.17E+13	1.38E+13		
Xe-135m	1.102	1.036	7.55E-04		1.31E+13	7.02E+13	8.33E+13		
Xe-137	6.128	6.041	3.03E-03		2.92E+14	1.57E+15	1.86E+15		
Xe-138	6.297	5.702	8.20E-04		8.12E+13	4.35E+14	5.17E+14		
I-131	2.891	3.291	9.98E-07		4.56E+10	2.44E+11	2.90E+11		
I-132	4.312	5.146	8.43E-05		5.76E+12	3.09E+13	3.66E+13		
1-133	6.697	6.759	9.25E-06		9.77E+11	5.24E+12	6.21E+12		
I-134	7.83	7.6	2.20E-04		2.71E+13	1.45E+14	1.72E+14		
1-135	6.282	6.941	2.93E-05		2.91E+12	1.56E+13	1.85E+13		
Br-83	0.5362	0.3959	8.02E-05		6.73E+11	3.61E+12	4.28E+12		
Br-84	0.9849	0.8224	3.63E-04		5.61E+12	3.01E+13	3.57E+13		
Br-84m	0.01672	0.003189	1.93E-03		4.98E+11	2.67E+12	3.17E+12		
Br-85	1.285	0.7428	4.02E-03		8.06E+13	4.32E+14	5.12E+14		
Br-86	1.595	1.161	1.26E-02		3.15E+14	1.69E+15	2.00E+15		
Br-87	2.035	1.537	1.24E-02		3.95E+14	2.12E+15	2.51E+15		

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## Table 4: Activity Predictions (in Bq) for 10 wt% solution

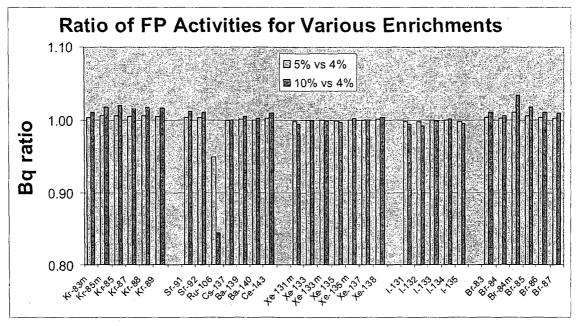


Figure 1: Ratio of fission product activities for various enrichments (5% values divided by 4% values or 10% values divided by 4% values)

An additional task was performed to compare the results of this work with those of the previous study. In Figure 2, the total activities are plotted for the 4 wt% scenario from both studies. The results are largely the same except for the <sup>85</sup>Kr isotope and most of the Xe isotopes. The difference in the <sup>85</sup>Kr results seems to be due to the use of a half life of 10.7 hours in the previous study versus the actual half life of 10.7 years. The differences in the predictions for the Xe isotopes are consistent with the use in Ref. 1 of the direct yields instead of the cumulative yields.

The average decay energies for beta and gamma emissions estimated in this work are entirely consistent with those in Ref. 1 and are not reported herein.

Due to the approximations made, the results shown in Tables 2–4 are known to be overestimates of the expected activities. The ORIGEN-S code was used to estimate the degree of this overprediction by inserting the actual power history of the criticality accident (a total of 48 pulses as described in Ref. 1 and above). The ORIGEN-S predicted activities for the 13 highest activities are shown in Figure 3 for the first 30 minutes of the accident. It is obvious that the decay for the top 3-4 isotopes is significant as compared to the assumed duration of the accident. The degree of overprediction for each fission product is shown in Figure 4 and Table 5 as a ratio of the activity due to the bounding approach divided by the ORIGEN-S prediction. The overprediction factors correspond to the end of 30 minutes and 8 hours, the breakpoints assumed for the initial and final accident times.

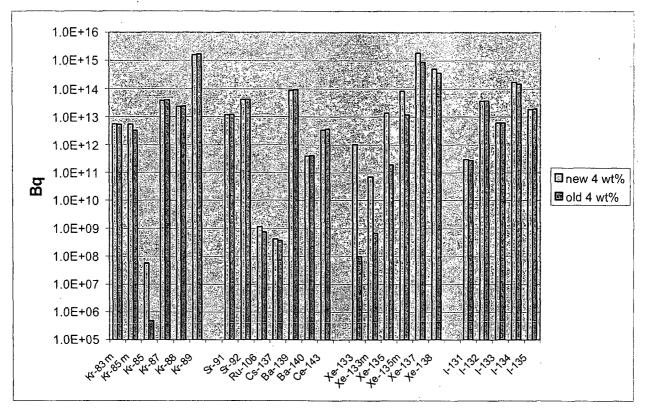


Figure 2: Comparison of activity results from this study (new) and those reported in Ref. 1 (old)

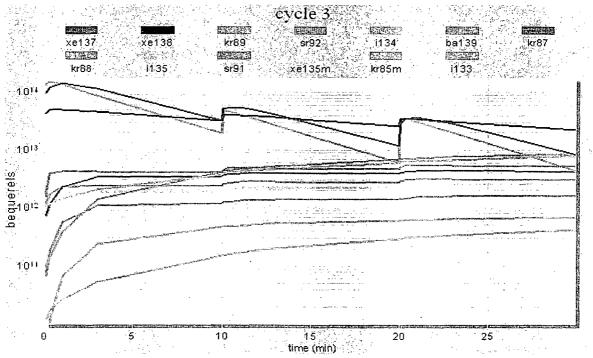


Figure 3: Fission product activities for the 14 highest activity isotopes during the first 30 minutes of the criticality accident

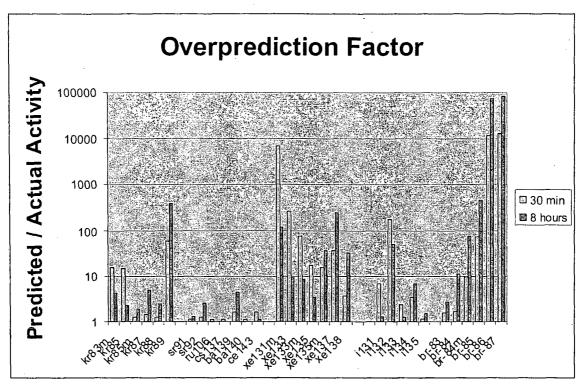


Figure 4: Overprediction factors for each of the fission product activities included in this study. Factors correspond to ratio of bounding activities to those from ORIGEN-S with actual accident power history accounted for

### Table 5: Overprediction factors<sup>+</sup> for each isotope

	<b>I</b>				<b>I I</b>		
		Overprediction factor					
	30 min	8 hours		30 min	8 hours		
kr83m	16	4.2	xe135	17	3.4		
kr85	15	2.3	xe135m	15	36		
kr85m	1.3	1.9	xe137	· 36	250		
kr87	1.4	5.0	xe138	3.7	32		
kr88	1.2	2.4					
kr89	58	390	i131	6.9	1.3		
			i132	180	50		
sr91	1.1	1.3	i133	2.3	1.2		
sr92	1.3	2.5	i134	3.4	6.9		
<b>r</b> u106	1.1	1.0	i135	1.2	1.5		
cs137	1.1	1.0	,				
ba139	1.6	4.3	br-83	, 1.6	2.7		
ba140	1.1	1.0	br-84	1.7	11		
ce143	1.6	1.1	br-84m	9.5	76		
			br-85	73	470		
xe131m	7100	120	br-86	12000	73000		
xe133	260	9.2	br-87	13000	84000		
xe133m	76	8.4					

<sup>+</sup>these factors are the ratio of the predicted bounding activities to the best-estimate results based on an ORIGEN-S calculation with the actual power history included

References:

- 1. Nuclear Fuel Cycle Facility Accident Analysis Handbook, NUREG/CR-6410, U.S. Nuclear Regulatory Commission (March 1998).
- 2. SCALE A Modular Code System for Performing Standardized Computer Analysis for Licensing Evaluation, NUREG/CR-0200, (ORNL/NUREG/CSD-2/R4), Vol. II, Sec. F7, Oak Ridge National Laboratory (March 1995).
- 3. Cross Section Evaluation Working Group, ENDF/B-VI Summary Documentation, Report BNL NCS-17541 (ENDF-201) (1991), edited by P. F. Rose, National Nuclear Data Center, Brookhaven National Laboratory, Upton, NY, USA.