Update of the Nuclear Criticality Slide Rule Calculations: Plutonium systems – Delayed Fission Gamma

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ABSTRACT

IRSN (France), LLNL (USA) and ORNL (USA) began a long-term collaboration effort in 2015 to update the nuclear criticality Slide Rule for the emergency response to a nuclear criticality accident. The Slide Rule permits the estimation of neutron and gamma dose rates and integrated doses based upon estimated fission yields, as a function of distance from the fission source, and time after criticality accidents for different critical systems.

This paper presents results from the fourth phase of the current update of the Slide Rule project [1], in which delayed fission-product gamma (DFG) dose rates of unreflected plutonium critical systems were compared by several modern 3D radiation transport codes (MCNP, COG, SCALE), using updated flux-to-dose conversion factors. Dose rates are calculated for fissile material at five moderation ratios (H/Pu), at 1 m above the ground as a function of distance (between 30 cm and 1,200 m) from the external surface of the source to the center of the detector, and for periods between 1 s and 1,000 min after the critical instantaneous event.

Further efforts have been devoted to the determination of the delayed gamma source, by comparing the time-dependent energy spectra obtained from several methods. Overall, DFG dose rates calculated by each participant led to consistent results. Extra effort is under way to identify the cause of the remaining differences, by comparing precisely the gamma source, and particularly nuclides inventories.

Key Words: nuclear criticality accident, slide-rule, plutonium, delayed gamma, dose

1 INTRODUCTION

In the framework of the US DOE Nuclear Criticality Safety Program [1] a long-term collaboration effort between AWE (UK), IRSN (France), LLNL (USA) and ORNL (USA) began in 2015 to update the Nuclear Criticality Slide Rule as a tool for emergency response to nuclear criticality accident. This document published in 1997 [2-3] gives order of magnitude estimates of key parameters for five unreflected spherical uranium systems, such as number of fissions and doses (neutron and gamma), useful for emergency response teams and public authorities. As a result, the complete work is envisioned to spread over many years and is divided into several steps:

1. The first phase [4] repeated simulations from initial reports with modern radiation transport codes and nuclear data,
2. The second phase introduced plutonium systems and additional configurations that combine new source geometries and reflectors [5],
3. The third phase was dedicated to sensitivity studies for uranium systems. The impact of shielding material on the initial configurations was evaluated. The contribution of humidity on the unshielded configurations in the first step was determined, as well as the effects of changing the ground composition used in the first step from concrete to dry soil [6].

This paper presents results from the fourth phase of the current update of the Slide Rule project, in which delayed fission-product gamma (DFG) dose rates of unreflected plutonium critical systems were compared by several modern 3D radiation transport codes, using updated flux-to-dose conversion factors.

2 CONFIGURATION OVERVIEW

2.1 Geometry

The geometry used for the additional configurations, derived from the initial configuration of the Slide Rule, is presented hereafter. It consists of a simple air-over-ground configuration with a source located at the center of a right-circular cylinder. The radius and the height of the air cylinder are both equal to 1,530 m. The ground is modeled as 50 cm layer of concrete. The Figure 1 presents the model to be calculated. Additional information is given in the following paragraphs.

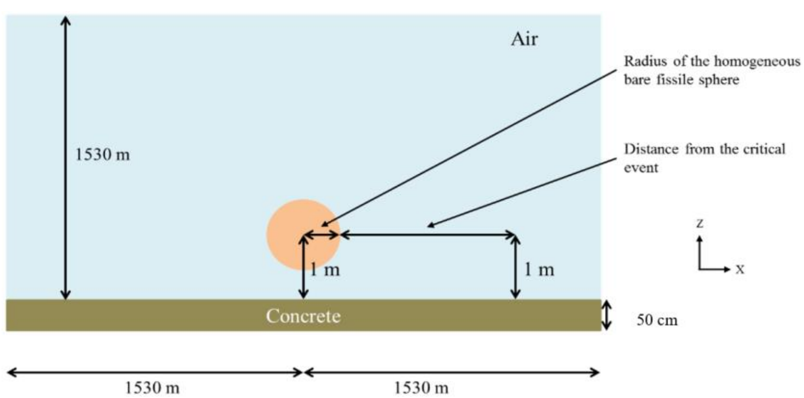


Figure 1. X-Z plan view of the configuration

2.2 Plutonium systems

For the “step 4” configuration, a bare critical sphere with plutonium is considered. This theoretical material is only composed of Pu-239 metal homogeneously mixed with water. Five moderation ratios (H/Pu) are considered: 0, 10, 100, 900 and 2,000. These values roughly cover the range of representative sizes (sphere radii), neutron/gamma leakages and spectra for this plutonium material. For a given H/Pu ratio, this material minimizes the critical size of the source compared to other plutonium material (oxide, fluoride, etc.). The Table I provides additional information regarding the five cases.

Table I. Fissile material specifications

Case	H/Pu	Critical radius (cm)	Pu-239 mass (kg)	Number density (atom/barn-cm)		
				Pu-239	O-16	H-1
1	0	4.93	9.97	5.00305E-02	0	0
2	10	12.53	19.26	5.88706E-03	2.94353E-02	5.88706E-02
3	100	15.36	3.97	6.58436E-04	3.29218E-02	6.58436E-02
4	900	19.5	0.91	7.40255E-05	3.33115E-02	6.66230E-02
5	2,000	29.17	1.38	3.33386E-05	3.33386E-02	6.66772E-02

2.3 Delayed fission gamma dose rate

The time-dependent DFG dose rates presented in this paper are given in rad/min assuming a criticality accident that generates 10^{17} fissions in 1 µs. The expected dose rates for periods of 1 s, 5 s, 10 s, 1 min, 5 min, 10 min, 50 min, 100 min, 500 min and 1,000 min after the event are calculated for all five critical systems. For these configurations, only the delayed gamma doses are calculated.

Doses are calculated at 1 m above the ground as a function of distance (between 30 cm and 1,200 m) from the external surface of the source to the center of the detector (see Figure 1). By default, the detector geometry is in the shape of a cylindrical shell with a square cross-section of 5 cm x 5 cm, to take advantage of the symmetry of the problem. The center of the detector is also at a height of 1 m above the ground.

Flux-to-dose conversion factors have a significant impact on the final dose and are likely to change in the future. That is why all participants involved used the same response functions to convert the simulated photon fluxes-to-doses, namely Henderson [7], ICRU Report 57 [8], ANSI/HPS N13.3 [9].

3 COMPUTATIONAL METHODOLOGY

This section describes the different codes and computational methods used by the laboratories contributing to this work.

3.1 ORNL

SCALE [10] is a comprehensive modeling and simulation suite developed and maintained by ORNL. SCALE 6.3 beta was used with the ENDF/B-VII.1 cross section data. The CAAS analysis capability, coupling KENO and MAVRIC, was used for this purpose. The libraries used with KENO-VI and MAVRIC both used a continuous-energy representation of the cross sections. First, KENO was run with a Cartesian mesh tally of the fission neutron production, which captured the asymmetry due to the ground 1 m below the center of each fissile assembly. Then MAVRIC used the KENO fission source distribution as a fixed source, generated variance reduction parameters, and simulated mesh tallies of fluxes or dose rates. For the prompt dose calculations, total nu-bar was used. The plutonium sphere is discretized by introducing 100 equivolume rings to calculate neutron flux and activation sources in order to capture self-shielding of the fissile assembly. The activation sources, generated by ORIGEN, can only use multi-group activation libraries, hence depend on the group structure. The new very-fine AMPX 1597-group structure (v7.1-1597n) was used for the last iteration. Once the delayed gamma sources were obtained from the ORIGEN calculations, MAVRIC is run again to calculate the doses from the activation and fission product gammas to detector in a gamma-only simulation.

3.2 LLNL

COG [11] is a full-featured Monte Carlo radiation transport code developed by LLNL that provides accurate simulation results to complex shielding, criticality and activation problems. COG 11.2 was used with ENDF/B-VII.1 cross-section library data. A feature in COG can generate, track and score DFG rays born between two given times. Point-wise continuous cross-sections are used in COG and a full range of biasing options are available for speeding up solutions for deep penetration problems. A direct one-step criticality/detector calculation method was applied for all neutrons and prompt and delayed gamma ray dose calculations. All neutron and gamma particles are tracked from birth due to fission within the spherical fissile volume to absorption in or leakage from the system in one single, massively parallel, COG supercomputer run with no variance reduction biasing applied. To activate the DFG option, DELAYEDPHOTONS (and associated time interval values) and DGLIB are input in the BASIC and MIX blocks, respectively. A 1 cm high cylindrical boundary-crossing detector was used to score the dose calculations.

3.3 IRSN

MCNP [12] is a general-purpose, Monte Carlo radiation transport code designed to track many particle types over broad ranges of energies developed by LANL. MCNP 6.2 was used with the continuous energy ENDF/B-VII.1 cross section library. For the delayed gamma doses computation, a “rigorous two-step” (R2S) methodology was followed to generate the source term and calculate the doses.

The first step is a static calculation (KCODE mode) to determine the spatial distribution of fission neutron production inside the fissile assembly. A Watt spectrum was used for the energy distribution. The MCNP results presented were obtained with 20 meshes (SMESH for a sphere), having the same volume, in which the fission reaction rate was tallied.

The second step uses the results of the first step to describe a fixed source (SDEF mode) of delayed gamma from neutron-induced fissions. Gamma spectrum and intensity were obtained by linking the same SCALE 6.1.2 sequences/modules as ORNL used with the 200 neutron/47 gamma group structure based on ENDF/B-VII.1 cross section library (v7.1-200n47g). KENO calculated the distribution of fission neutron production inside the sphere. MAVRIC/Monaco used the KENO results to calculate the neutron flux inside the sphere. COUPLE used the resulting MAVRIC/Monaco neutron flux and created problem dependent flux weighted cross sections to produce reaction rates. Finally, ORIGEN used these reaction rates and performed the depletion and decay of the plutonium systems. For comparative purposes, the FISPACT-II 5.0 inventory and source-term code [13] was also used independently of SCALE to generate fission product gamma source.

Once the delayed gamma source was obtained either from ORIGEN or FISPACT calculations, the fission mesh source was updated to represent delayed gammas instead of fission neutrons by changing particle type and energy spectrum through a new MCNP calculation. The fission neutron production was turned off (treated as absorption), all gammas were simulated. The spatial distributions of the fission neutron mesh source and delayed gamma mesh source are the same. A F4 tally (*i.e.* track length estimate of cell flux) paired with an EM card (*i.e.* energy multiplier) was used to score the delayed gamma dose rates at the desired distances. Contrary to the previous step which included neutrons and strong attenuation, ADVANTG automated tool for generating variance reduction parameters was not used for this kind of calculations. Instead, Weight Windows generator in space and energy was well enough to be used.

4 RESULTS AND DISCUSSION

This section presents and discusses the simulation results obtained for the cases described in the previous sections as well as the comparison with the results of the first phase of the Slide Rule.

4.1 Delayed fission gamma dose rates results

At least 750 results (15 distances x 10 decay times x 5 cases) are needed to cover all the cases for one conversion factor. Figure 2 shows the DFG dose rates calculated with FISPACT-MCNP only using the ANSI/HPS N13.3 flux-to-dose conversion factor. Calculations have also been performed with the two other conversion factors. In ascending order, ANSI/HPS N13.3 flux-to-dose conversion factor is more penalizing than Henderson one, followed by ICRU-57 (calculated doses about 10 % and 20 % higher respectively).

Whatever the case, dose rates decrease by more than 9 orders of magnitude between 30 cm and 1,200 m from the external surface of the source, and by 5 orders of magnitude between 1 s and 1,000 min after the critical event. The difference between the extreme cases goes from a factor 9.3 for short distances to a factor 3.4 for long distances. The lowest dose rate is obtained for the metal plutonium system whereas the highest dose rate is obtained for an intermediate moderation ratio (H/Pu=100 or 900).

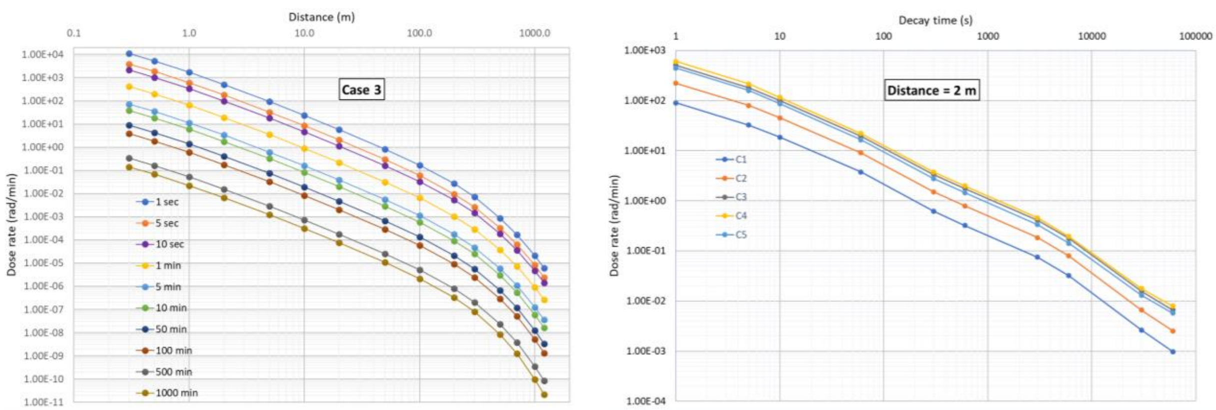


Figure 2. IRSN (FISPACT-MCNP) results obtained with HPS N13.3 conversion factor

Figure 3 presents the DFG dose rate ratio between plutonium metal (case 1) and 93.2 % U-235 enriched uranium metal (obtained from the first phase of the update of the Slide Rule) calculated with MCNP and the Henderson flux-to-dose conversion factor. Results show a slight increase trend at short distances, which stabilizes as the distance increases. It can also be seen that the plutonium metal configuration generates dose rates up to twice higher than the uranium metal configuration (the higher the decay time, the higher the Pu/U ratio). Observed variations of the dose rates from the plutonium configurations than the uranium ones are driven by the differences in half-lives of the fission products and show the interest to update the original slide rule with plutonium systems to adapt the emergency response to a nuclear criticality accident.

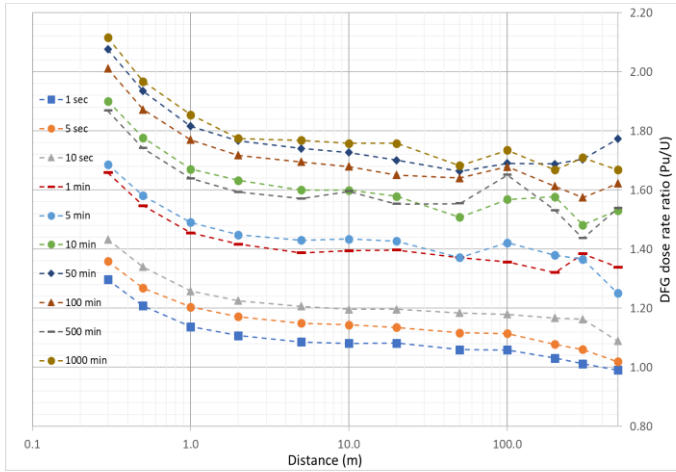
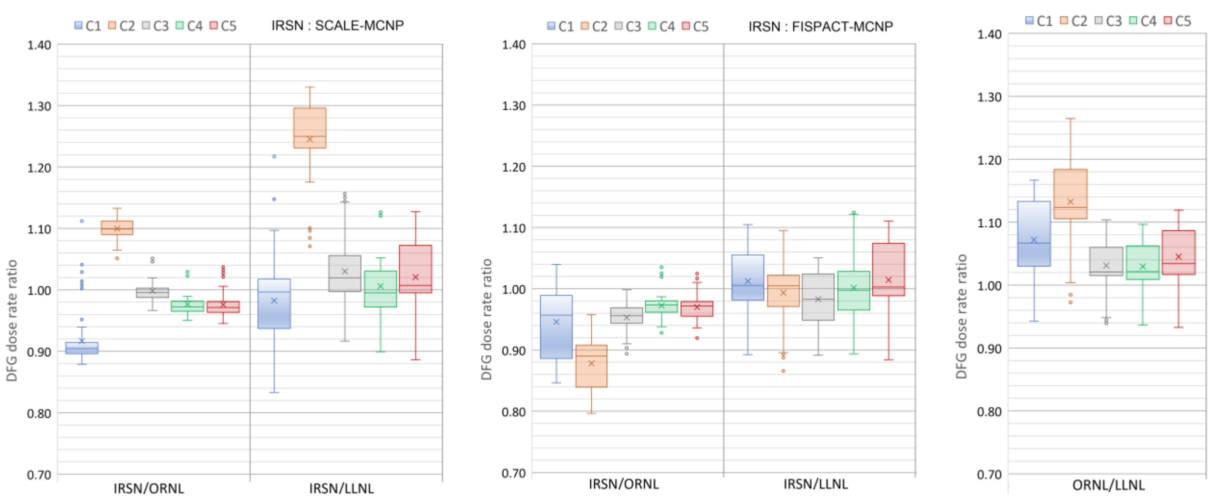


Figure 3. Comparison of DFG dose rates between Pu and U metal

4.2 Code-to-code comparison

Figure 4 presents code-to-code comparison of DFG dose rates calculated with the ANSI/HPS N13.3 flux-to-dose conversion factors (results should be equivalent with other conversion factors). For a compact visualization, the results' dispersion is displayed on a box-and-whisker graph. The box shows the interquartile range (IQR) which depicts the middle half of the data set. The median is represented by a horizontal bar and the mean by a cross in the box. The whiskers represent the range between the lowest and the highest value, with dots placed past the line edges to indicate outliers. Ratios presented herein are calculated for distances below 300 m, where the relative error due to the codes is lower than 2 %. Each case contains 110 values (11 distances x 10 decay times).



IRSN: SCALE-MCNP *IRSN: FISPACT-MCNP*

Figure 4. DFG dose rate code-to-code comparison

The left-hand side of Figure 4 presents a code-to-code comparison, with IRSN results obtained using SCALE. The following observations can be made:

- IRSN/ORNL: the overall distribution is narrowly spread around the mean for all cases. IRSN tends to underestimate ORNL results by approximately 10 % for case 1 and conversely overestimate by 10 % for case 2. For cases 3, 4 and 5 results are in reasonably good agreement, with ratios close to one.
- IRSN/LLNL: except for case 2, a relatively good agreement is observed, although ratios variance is slightly higher. For case 2, IRSN clearly overestimates LLNL results by 20 % to more than 30 %.

The middle graph of Figure 4 illustrates the similar comparison but with IRSN results obtained using the FISPACT code:

- IRSN/ORNL: for cases 1 and 2, the distribution is stretched with a trend for IRSN results to underestimate the dose rates calculated by ORNL, with a more pronounced effect for case 2. A relatively good agreement is observed for cases 3, 4 and 5 with a squeezed distribution;
- IRSN/LLNL: for all five cases, a good agreement is observed with discrepancies generally lower than 10 % ($0.95 \leq IQR \leq 1.07$) and ratios are symmetrically distributed around one.

Concerning ORNL results, compared to LLNL (right-hand side of Fig. 4), a relatively good agreement is observed for cases 3, 4 and 5. For cases 1 and 2, ORNL tends to overestimate LLNL dose rates with a pronounced tendency for case 2 ($1.0 \leq$ range ≤ 1.27 and $1.11 \leq IQR \leq 1.18$).

The coarse mesh of the multi-group library used by ORIGEN (v7.1-200n47g) to perform the depletion and decay may lack of accuracy in the intermediate energy region where the 239-Pu fission cross section is sensitive for case 2. The new very-fine AMPX 1597-group structure (v7.1-1597n) used by ORNL in SCALE 6.3 allows to reduce the gap for this case.

5 CONCLUSIONS AND PERSPECTIVES

This report presents the calculations performed by IRSN, LLNL and ORNL to update the Nuclear Criticality Slide Rule for the emergency response to a nuclear criticality accident. This report gives:

- a calculation scheme for DFG doses and its application to five plutonium material,
- comparisons between radiation transport and depletion codes such as MCNP, SCALE, COG and FISPACT,
- comparisons between different flux-to-dose conversion factors.

Overall, DFG dose rates calculated by each laboratory led to consistent results. Extra effort is under way to identify the cause of the remaining differences, especially for case 2, by comparing precisely the gamma source, and particularly nuclides inventories. Prospects could be dedicated to sensitivity studies in the same way as those made for uranium systems in step 3. Once the reasons of these discrepancies will be identified, the overall work produced for the update of the Slide Rule project should lead to the creation of a future new operational document, whose format is still to be discussed. Another consequence of this collaborative effort might be the creation of “computational benchmarks” in order to test and verify the various variance reduction methods and to establish best practices when dealing with this kind of problem.

6 ACKNOWLEDGMENTS

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