Validation of ORIGEN-S Decay Heat Predictions for LOCA Analysis

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Abstract

Recent developments in the nuclear data libraries used by the ORIGEN-S isotope generation and depletion code have enabled the extension of the code to accurately predict the delayed energy release rates from nuclear decay (decay heat) at very short cooling times of interest to reactor accident analysis. Historically this time domain has required integral methods, such as the ANS-5.1 decay heat standard, because isotopic summation codes could not be reliably applied due to incomplete nuclear data. This paper describes work to validate ORIGEN-S against experimental measurements for decay times that extend down to about 1 second after fission. Benchmarks using measured gamma ray spectra following fission are also included because these results are important to predicting spatial energy deposition from delayed gamma energy release.

KEYWORDS: Nuclear decay heat, gamma-ray spectra, ORIGEN-S, computer simulation, accident analysis

1. Introduction

Delayed energy release (decay heat) from the decay of fission products and actinides in irradiated fuel is an important factor in the analysis of postulated loss-of-coolant accidents (LOCAs) and evaluation of emergency core-cooling system (ECCS) performance. After the fission process ends, the accumulated radionuclides continue to release recoverable energy in the form of beta and gamma rays. This delayed energy represents about 7% of the total recoverable energy from fission. Approximately 25% of this residual energy is released in the first 10 s after fission, and about 50% is released by 100 s after fission. Accident analyses typical consider the decay heat rate for times shorter than about 10^5 s after fission. Calculation of decay heat in this time frame is widely performed within the nuclear industry using decay heat standards, such as ANSI/ANS-5.1-2005 [1].

Integrated accident and consequence analysis of nuclear facilities requires additional information on the fuel elemental inventories and the isotopic inventories and activities as a function of time to allow evaluation of releases and radiological dose to members of the public. Nuclide inventories are generally obtained using isotopic summation codes like ORIGEN. The ORIGEN codes explicitly calculate the time-dependent concentrations for a large set of isotopes, as well as decay heat and radiation emissions (neutrons and gamma rays). Integral quantities such as decay heat are calculated by summing the contributions from each individual

radionuclide. However, application of summation codes to very short decay times has not been widely practical in the past because of incomplete and inadequate nuclear data available for the very short-lived fission products.

Recent upgrades and expansion of the fission product data libraries in ORIGEN-S [2] warrant reevaluation of the code performance at these short times. In this paper decay heat predictions from ORIGEN-S are compared against measurements reported for thermal fission of ^{235}U, ^{239}Pu, and ^{241}Pu, and fast fission of ^{233}U, and ^{238}U, over decay times of importance to LOCA analysis. In addition, benchmark results are reported for gamma energy spectra measured several seconds after thermal fission of ^{235}U and ^{239}Pu. The gamma-ray library used by ORIGEN-S, consisting of discrete line-energy and yield data per disintegration, was also upgraded recently to include photon emission of more than 1200 individual nuclides. This enables the accurate prediction of gamma energy release rates and energy spectra at extremely short decay times and is important to evaluating the spatial deposition of delayed energy in materials within the reactor core.

2. ORIGEN-S Code and Nuclear Data Libraries

The accuracy of the decay heat predictions for fission experiments is dependent primarily on the fission yields, the decay schemes (metastable levels, decay modes, branching fractions, and half lives), and the recoverable energy per decay (Q values) for each fission product. The sources of these data in the ORIGEN-S code are briefly described.

2.1 Fission Product Yields

Independent neutron-induced fission product yields from ENDF/B-VI.2 are implemented in ORIGEN-S for 30 fissionable actinides, including ^{227,228,232}Th, ^{231}Pa, $^{232\text{-}238}$U, $^{238\text{-}242}$Pu, ^{241,242m,243}Am, ^{237,238}Np, $^{242\text{-}246,248}$Cm, ^{249,252}Cf, and ^{254}Es. In addition, ternary fission yields for 21 low-mass nuclides were obtained from JEF-2.2. The fission yield update from ENDF/B-V to ENDF/B-VI increased the total number of fission products explicitly represented in the library from 879 to 1119. The addition of neutron-induced fission yields for most of the fissionable actinides having evaluated yields greatly increases the versatility and range of application of the code, particularly for advanced fuel designs and minor actinide transmutation studies. The fission yields developed for ENDF/B-VI were designed to improve the data for short-term decay heat applications.

Fission product yields for thermal-neutron induced fission were implemented for all materials evaluated in this study except ^{238}U, for which fast-fission yields are applied. Automated methods have been developed at ORNL to readily generate libraries containing yields for any incident neutron energy from thermal to 14-MeV.

Independent fission yields for the nuclides ^{74m}As, ^{85m}Se, ^{86m}Br, and ^{162m}Tb were combined with the yields to their ground states. A review of current decay schemes indicates that these metastable states have been eliminated from modern evaluations. Furthermore, yields to the second metastable states of ^{116n}In, ^{118n}In, ^{120n}In, ^{122n}In, ^{124n}Sb, ^{126n}Sb, ^{130n}In, ^{152n}Pm, and ^{152n}Eu were combined with their decay progeny because ORIGEN-S currently does not support nuclide identifiers for excited levels beyond the first metastable state. The decay energy was assigned to the daughter in such a way as to mitigate the effects of this approximation on the decay heat predictions.

2.2 Nuclear Decay Data

The nuclear decay data and decay schemes are primarily from ENDF/B-VI. Decay data were compiled for 1093 individual fission products having direct fission yields from ENDF/B-VI. Approximately 230 fission products with yields were not included for lack of evaluated decay schemes. Most of these were low-yield isotopes located far from stability. Nuclear decay data not available from ENDF/B-VI were obtained from ENSDF and JEF-2.2 where available. The ENSDF decay evaluation for ^{79}Se was used in place of the ENDF/B-VI data to correct the half life evaluation (for a description see http://www-rsicc.ornl.gov/rsic-cgi-bin/enote.pl?nb=scale&action=view&page=206).

The sources of nuclear decay data in the ORIGEN-S library are listed in Table 1. This library is distributed with ORIGEN-S in the SCALE 5 code system.

Table 1: Summary of fission product decay data sources

Number of nuclides	Stability, status, and data source
146	Stable nuclides
764	Radioactive, ENDF/B-VI decay data
172	Radioactive, ENSDF decay data
11	Radioactive, JEF-2.2 decay data
26	Ternary fission products and decay progeny (JEF-2.2)
1119	Total fission product library size

2.3 Gamma Emission Libraries

The photon library released with versions of ORIGEN-S prior to SCALE 5 package contained data for 418 mostly longer-lived nuclides. The photon library was developed in the late 1970's from the Evaluated Nuclear Data Structure File (ENSDF) and is relatively unchanged since its initial release. The photon data used to develop the libraries was implemented in both the ORIGEN-S and the ORIGEN2 codes. The library has been shown to be adequate to calculate gamma emission and gamma heating at times longer than approximately 1 d, which was sufficient for many applications involving analysis of transportation and storage casks for spent nuclear fuel. However, improvements in nuclear data evaluations motivated an update of the photon library, using more recent and more comprehensive photon evaluations.

The revised photon library, released with SCALE 5, dramatically increased the number of nuclides with explicit photon yield data, from 418 to 1132, and increased the number of discrete photon lines from 12,000 to 115,000. The photon data were derived from the evaluations of ENDF/B-VI, ENSDF, and JEF-2.2. Both discrete (line-energy) and continuous-energy emission spectra are represented. To quantify the accuracy of the photon database, calculated gamma spectra were compared to spectra measured for times between 2.7 and 14,000 s after fission of ^{235}U, ^{239}Pu, and ^{241}Pu. These results are relevant to accident analysis because the gamma-ray component of decay heat can be transported out of the fuel and deposited in structural components or other regions of the core. Therefore, the ability to accurately predict both the magnitude and the energy distribution of the emitted gamma radiation are of importance to evaluating safety system performance.

3. Fission Benchmarks and Results

The fission experiments used to benchmark the decay heat calculations are listed in Table 2, and include measurements performed at ORNL [3,4], the University of Tokyo YAYOI fast reactor facility [5,6], Karlsruhe [7], Studsvik [8], and the University of Massachusetts, Lowell [9]. All of these experiments provide measurements of total energy release following fission, were performed after circa 1980, and were carried out predominantly using spectroscopic techniques. These techniques involve measuring the gamma and beta radiation spectra using detectors, unfolding the spectra to obtain the emitted energy distributions and intensities, and deriving the decay heat from the individual gamma and beta energy components.

One of the measurement sets (Karlsruhe) was made using calorimetric methods. It has been observed that results from calorimeter methods tend to be systematically higher at short cooling times and have generally larger uncertainties than those from spectroscopic methods. However, this difference is not significant at the 95% confidence level [10].

Results are presented as the instantaneous total energy release rate after a fission event $f(t)$ (MeV/s/fission) multiplied by the time after fission t(s), to obtain the units of MeV/fission shown in the figures. This is done since $f(t)$ varies approximately as $1/t$ in this time range.

Table 2: Selected decay heat fission experiments

Data set	Isotopes	Method[a]	Author(s)	Institute	Year (circa)
1	^{235}U, ^{239}Pu, ^{241}Pu	γ, β	Dickens et al.	Oak Ridge National Laboratory	1980
2	^{235}U	calorimeter	Baumung	Karlsruhe	1981
3	^{233}U, ^{235}U, ^{238}U, ^{239}Pu	γ, β	Akiyama et al.	Tokyo University YAYOI fast reactor	1982
4	^{235}U	γ, β	Johansson	Uppsala University / Studsvik	1987
5	^{235}U, ^{238}U, ^{239}Pu	γ, β	Schier and Couchell et al.	University of Massachusetts, Lowell	1997

[a] γ, β = spectroscopic method

3.1 U-235

The comparisons of ORIGEN-S calculations and the representative benchmark data for ^{235}U and thermal fission are shown in Fig. 1. The Karlsruhe and Studsvik measurements are plotted separately because the time range of these measurements is more restricted than that of the other experiments considered. As illustrated in the figures, the calculations are generally within the range of measurement uncertainty (2–5%) over most time periods of importance to LOCA analysis. The Lowell data extend to the shortest decay times available, about 0.4 s after fission. However, the Lowell data suffer from loss of noble gas fission products from the counting samples at the longer cooling times, and an absolute determination of the number of fissions in the samples was not made. Normalization was done using computations, and an attempt was made to account for this loss using calculations; however it is apparent the procedure was not entirely successful for this case.

The Karlsruhe results are the only measurements reported here that were made using calorimetric methods. As noted previously, calorimeter measurements made circa pre-1980

exhibit systematically larger values of decay heat than spectroscopic methods. The Karlsruhe and Studsvik results shown here yield slightly larger decay heat results than the other experiments, but the calculations are observed to be within the range of experimental uncertainty over the entire range of the measurements. Note that these experiments were performed for a 200-s irradiation, time which was long with respect to the measurement times after fission. Consequently, the experimental data were adjusted to account for the long irradiation time relative to the other experiments.

3.2 Pu-239

Energy release rates following ^{239}Pu thermal fission were measured at ORNL and Lowell. Results for fast fission of ^{239}Pu were also measured at YAYOI. The differences between fast and thermal fission were expected to be small (< 1%), and therefore the YAYOI data are included with the other results for thermal fission for the purposes of comparison with calculations. The results shown in Fig. 1 indicate good agreement of the ORIGEN-S calculations with the experiments over the range of the data. The calculations underpredict the decay heat near 10^3 s, but are generally within two standard deviations of experimental error.

3.3 U-238

For low-enriched systems, the fast fission of ^{238}U represents about 5% of the total fission rate. Measurements of residual decay energy from fast fission of ^{238}U have been performed at the YAYOI fast reactor facility and at the University of Massachusetts, Lowell. The calculations by ORIGEN-S, performed using fast fission yields from ENDF/B-VI are compared to the experimental data in Fig. 1. Again, the predictions are seen to be in generally good agreement with experiment over the range of the data. The results of the calculations are slightly lower than those of the measurements near the time periods around 200-s and 10^3 s after fission, but are typically within two standard deviations experimental error.

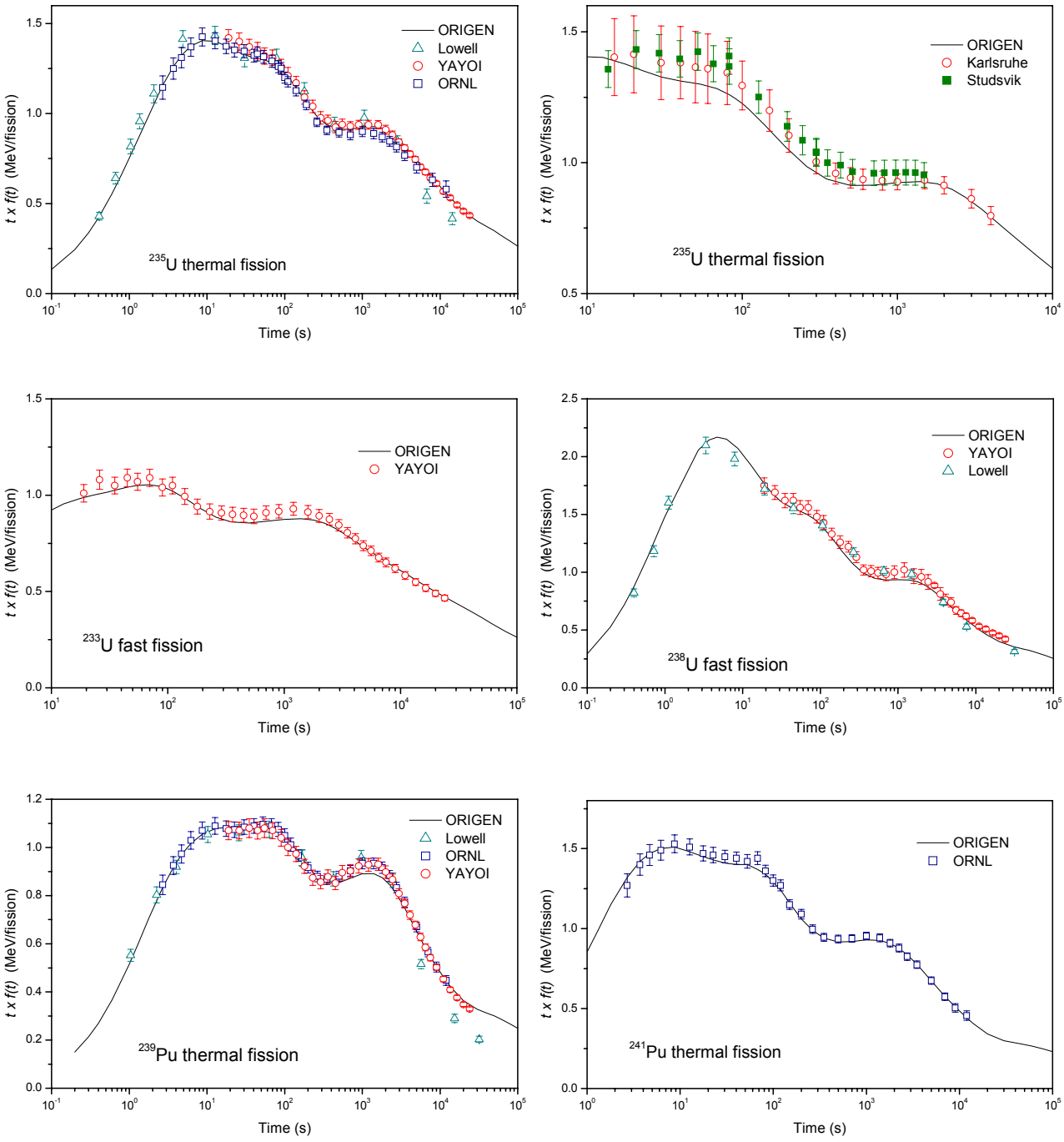
3.4 Pu-241

The fission of ^{241}Pu can represent up to about 10 % of the total fission rate at the end of life for typical commercial light-water reactor (LWR) fuels. The predicted decay heat following thermal fission of ^{241}Pu is compared to the ORNL measurements in Fig. 1. The results are seen to be within the assigned uncertainty of the measurements over the full range of the data.

3.5 U-233

The decay heat from fast fission of ^{233}U was benchmarked against measurements made at the YAYOI fast reactor facility. The calculations for ^{233}U, shown in Fig. 1, are within the assigned error of the measurements over most of the range. There is a trend to underpredict the decay heat near 10^3 s, similar to that seen for ^{239}Pu and ^{238}U. No other independent published measurements for ^{233}U were identified.

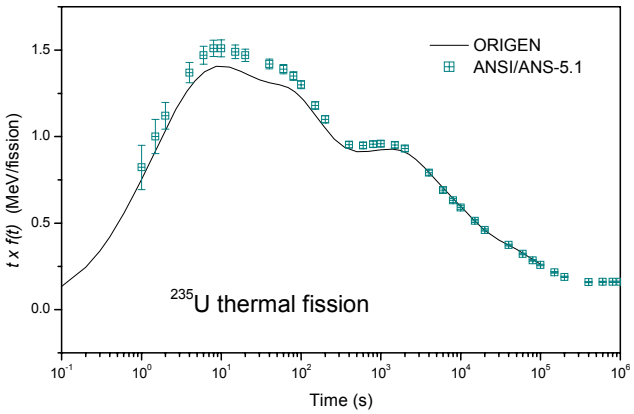
Figure 1: Comparison of total decay heat calculated using ORIGEN-S against selected fission experiments for ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu.



3.6 ANS-5.1 decay heat standard

The ORIGEN-S calculations for ^{235}U fission are compared to the ANSI/ANS-5.1-2005 Standard in Fig. 2. The results of the calculations are lower than those of the standard in the timeframe < 10^3 s. This result reflects the fact that the standard conservatively accounts for deviations observed between different experimental data; particularly with the use of pre-1980 calorimetric experiments (that yield systematically greater values of decay heat than more recent measurements) in developing the standard. The values in the standard are not based on any single set of measurements or even a least-squares best-estimate evaluation of multiple experiments – it is intentionally developed to be conservative and is biased toward agreement with those experiments yielding the highest values for decay heat. The ORIGEN-S results however represent best-estimate values without any uncertainty assignment. Margins for uncertainty in the ORIGEN-S calculations have not been evaluated in this work. Such margins however will be influenced by the selection of experiments used for validation.

Figure 2: Comparison of total decay heat for ^{235}U thermal fission calculated by ORIGEN-S and the ANSI/ANS-5.1 decay heat standard.



3.7 Gamma Energy Spectra

Predicted gamma ray intensities and energy spectra following the fission of ^{235}U and ^{239}Pu are compared to measurements performed at ORNL [11,12] in Fig. 3. The decay times shown here are 1.7 and 19.7 s after fission, although the measurements included longer times. The calculations, performed using the revised photon library (SCALE 5) and the previous version as used in SCALE 4.4 and in ORIGEN2, represent un-normalized results (i.e., they have not been adjusted for energy conservation using the decay Q values). The calculated spectra were generated in an ultra-fine energy group structure and then broadened to the approximate energy resolution of the NaI(Tl) detector used in the experiments. The results indicate that the updated photon library yields spectra in good agreement with experiment. The use of the original library in SCALE 4.4 resulted in a significant underprediction in the spectra caused by missing photon emission data for many of the short-lived fission products. The predicted spectra for ^{239}Pu are observed to be not as good as for ^{235}U, particularly in the energy range above 1.5 MeV where the spectra are underpredicted at the times studied. Although the predicted spectra are

significantly improved over previous calculations, some data deficiencies remain. There are ongoing research activities to evaluate and integrate more recent decay data measurements made using a total absorption gamma-ray spectrometer (TAGS) that have been shown to improve the decay predictions [13]. These data should be further evaluated against these benchmarks and integrated into the ORIGEN-S decay library to improve the accuracy of calculations.

4. Conclusion

It is important to emphasize that the present benchmarks involve mostly spectroscopic measurements of decay heat (with the exception of the Karlsruhe calorimeter experiment) made after about 1980. Inclusion of pre-1980 calorimeter measurements would have indicated larger deviations between calculations and experiment than observed in this work because the earlier calorimeter measurements yielded systematically larger estimates of decay heat.

In summary, the ORIGEN-S predictions of decay heat yield results are in good agreement with more recent determinations of decay heat from fission in the time range of about 1 to 3×10^4 s after fission; for example, within the uncertainty assigned to the measurements of typically < 5%. These results indicate that summation codes, like ORIGEN-S, using modern data, may be applied to accurately calculate decay heat for accident analysis evaluation. Use of summation codes also provide a consistent set of elemental and isotopic concentrations, and radionuclide activities necessary for consequence analysis. Margins of safety to account for code bias and uncertainty are necessary for safety-related studies; however, their evaluation was beyond the scope of the present work.

Acknowledgments

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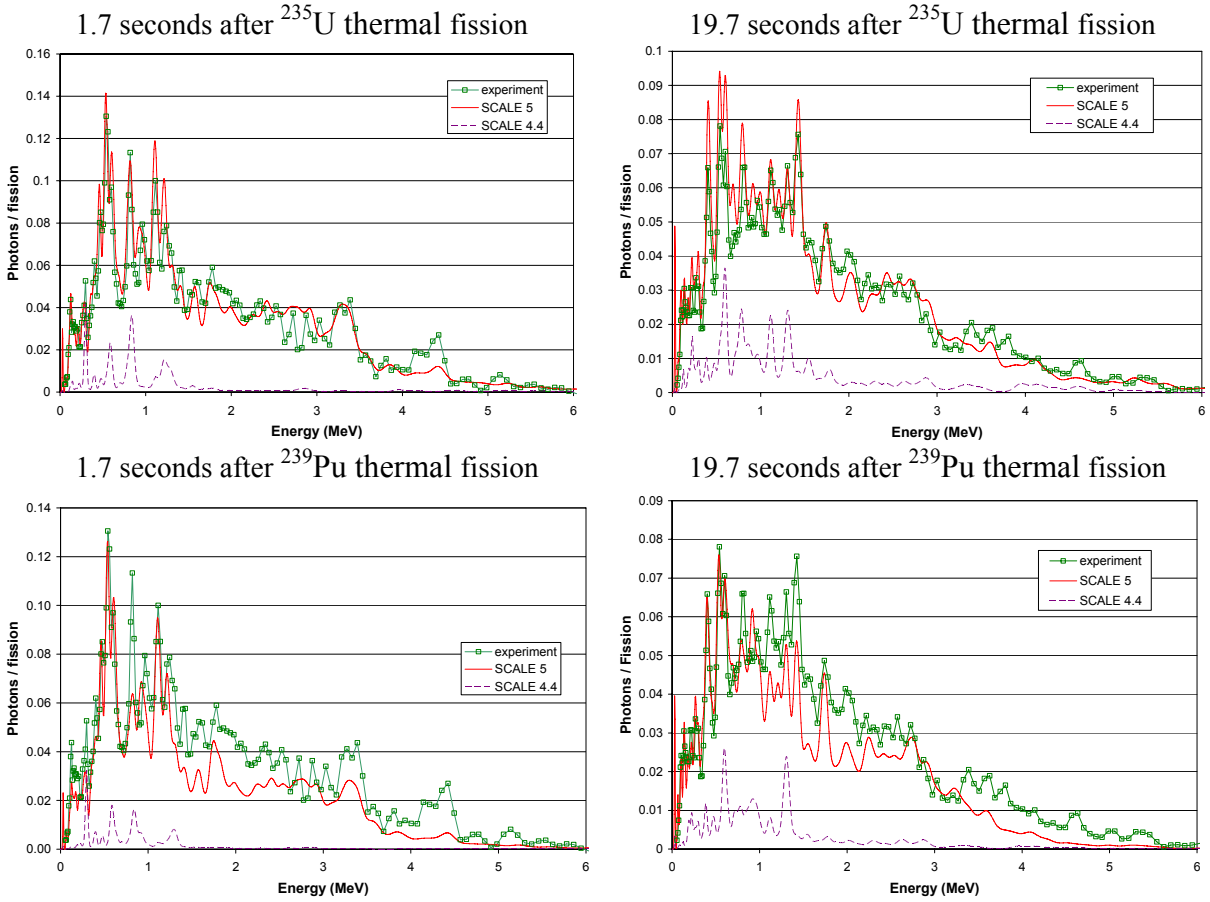
Figure 3: Comparison of photon intensity and energy spectra calculated using ORIGEN-S and experiments performed for ^{235}U and ^{239}Pu thermal fission.



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