Decay Heat Analyses after Thermal-Neutron Fission of ²³⁵U and ²³⁹Pu by SCALE-6.1.3 with Recently Available Fission Product Yield Data

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1. Introduction

After the Fukushima Daiichi nuclear disaster in 2011, the need for full knowledge and proper handling of the decay heat in reactors and spent-fuel pools has been opened to the public. Although nuclear fission stops immediately after a reactor has been shut down, the fission products in the fuel continue to release decay heat, which initially corresponds to about 6.5% of full reactor power. The heat reaches about 1.5% after one hour and falls to 0.4% after a day. After a week it will be about 0.2%. The reactor, however, still requires further cooling for several years to keep the fuel rods safe.

In general, the decay heat in the reactors can be calculated using a summation calculation method [1], which is simply the sum of the activities of the fission products produced during the fission process and after the reactor shutdown weighted by the mean decay energies. Consequently, the method is strongly dependent on the available nuclear structure data. Nowadays, the method has been implemented in various burnup and depletion programs such as ORIGEN and CINDER.

In this study, the decay heat measurements after thermal-neutron fission of ²³⁵U and ²³⁹Pu have been evaluated by the ORIGEN-S with the decay data and fission product yield libraries included in the SCALE-6.1.3 software package [2]. New fission product yields libraries for ORIGEN have been generated based on the recently available fission product yield data such as ENDF/B-VII.1 [3], JEFF-3.1.1 [4], JENDL/FPY-2011 [5], and JENDL-4.0 [6]. The new libraries were applied to the decay heat calculations, and the results were compared with those by the ORIGEN reference calculation.

2. Decay Heat Measurements

During the 1970s and 1980s, most of the experiment related to decay heat emphasized a short cooling times after fission of between ~1 sec and ~1 day. The impetus for these experiments was a concern regarding consequences of a LOCA coupled with the difficulty in calculating decay heat for very short times because of a lack of experimental decay data on short-lived fission products. In addition, these experimental data were invoked to establish the basis for an American Nuclear Society Standard on decay heat for thermal-neutron fission. Some experiments were performed at Oak Ridge National Laboratory (ORNL), the University of Tokyo YAYOI fast reactor facility, Karlsruhe, Studsvik, and the University of Massachusetts, Lowell [5,7]. Most of these experiments provided measurements of total, beta, and/or gamma energy release following thermal- and/or fast-neutron fission. The actinides involved in the experiments were ²³²Th, ^{233,235,238}U, and ^{239,241}Pu. Since this work concentrated on the analyses after thermal-neutron fission of ²³⁵U and ²³⁹Pu, the measurements by ORNL [8] and Studsvik [9] were chosen for comparison.

3. Generation of Fission Product Yield Library

The fission product yield data plays a major role in determining the initial fission product inventories after a fission event. This implies that the yield data can help us accurately predict the decay heat at very short cooling times of interest to reactor accident analysis. Moreover, an international co-operation has been carried out to improve the fission product yield evaluation methodologies under the auspices of OECD/NEA.

For the generation of the ORIGEN yield library, four up-to-date fission product yield files have been chosen out of the three major nuclear data evaluations, ENDF/B, JEFF, and JENDL. The ENDF/B-VII.1, JENDL/FPY-2011, and JENDL-4.0 provide neutroninduced fission product yields data for 31 fissionable isotopes and spontaneous fission product yields data for 9 fissionable isotopes. These data were originally taken from ENDF/B-VII.0 with some adjustments, which have been the basis for the ORIGEN yield libraries given in the SCALE-6.1.3. The JEFF-3.1.1 provides neutron-induced fission product yields data for 19 fissionable isotopes and spontaneous fission product yields data for 3 fissionable isotopes. These data were originally taken from UKFY-3.6 [4].

The ORIGEN yield library contains the energydependent yields of 1151 fission products for 30 fissionable actinides. The independent fission product yields are obtained from the MF=8/MT=454 of each evaluation. In ORIGEN, energy-dependent yields are available for thermal (0.0235 eV), fast (2.0 MeV), and high-energy (14 MeV) incident neutron energies depending on the actinides. The original yield files, however, supply yield data at 500 keV or 400 keV (in JEFF-3.1.1) for fast fission. The ORIGEN yield library adopts an effective incident neutron energy for fast fission adjusted from 500 keV to 2.0 MeV to more accurately reflect the average fission energy [2]. The effective fast neutron energy for the JEFF-3.1.1-based yield library has been adjusted from 400 keV to 2.0 MeV.

4. Decay Heat Analyses

The summation calculation models used to predict the decay heat for very short cooling times after thermal-neutron fission of ²³⁵U and ²³⁹Pu have been set up for the ORIGEN-S code. All the reference calculations were carried out with the latest decay data library and fission product yield library (known as the Revision 3) in the SCALE-6.1.3 package. For a comparative study among different yield data, the yield library has been replaced with new yield libraries based on ENDF/B-VII.1, JEFF-3.1.1, JENDL/FPY-2011, and JENDL-4.0.

4.1 Comparison with Measurements

The decay heat varies as a function of time after a shutdown and can be computed based on the inventory of fission products created during the fission process and after a reactor shutdown. The results are given as the instantaneous energy release rate after a fission event (f(t), MeV/fission/sec) multiplied by the time after fission (t).



Fig. 1. Comparison of total decay heat for ²³⁵U thermal fission.

Figures 1 - 3 show comparisons of the ORIGEN-S reference calculation results with the measurements for the total, beta, and gamma decay heat after thermalneutron fission of ²³⁵U, respectively. For the total and beta decay heats, the calculation results tend to follow the measurements of ORNL well rather than those of Studsvik. The gamma decay heat calculation shows some discrepancies with the measurements. As shown in Fig. 1, the ANSI/ANS-5.1-2005 Standard decay heat data [10] overestimates the total decay heat below ~1000 seconds when compared with the calculation results and the measurements of ORNL. It is believed that the Standard decay heat data have been intentionally developed to be conservative and biased toward agreement with the experiments yielding the highest values.



Fig. 2. Comparison of beta decay heat for ²³⁵U thermal fission.



Fig. 3. Comparison of gamma decay heat for 235 U thermal fission.

The calculated total, beta, and gamma decay heats after thermal-neutron fission of ²³⁹Pu have been compared with the measurements of ORNL, as shown in Fig. 4. The results indicate good agreement between the calculated and experimental values over the range of the data. The ORIGEN-S calculations underestimate the total decay heat near 1000 seconds, which mainly comes from the discrepancies in the beta decay heat.



Fig. 4. Comparisons of total, beta, and gamma decay heats for 239 Pu thermal fission.

4.2 Decay Heat Analysis for ²³⁵U by Different Yield Data

The initial isotopic inventories obtained from the summation calculation follow exactly the fission product yield distribution given in the yield library. As a result, the differences in the isotopic contents between any two fission product yield libraries result in nearly the same differences in the decay heat estimation of each isotope. This fact can be clarified by examining the total decay heats induced by the individual fission products for different yield libraries.

The decay heat calculation results for the 235 U thermal-neutron fission are shown in Figs. 5 - 8. Figures 5 and 6 show the relative differences of JEFF-3.1.1- and JENDL/FPY-2011-based calculation results to the ORIGEN-S reference results for the 10 most influential isotopes over the whole time interval (0.05 - 100,000 seconds), respectively. Quite big differences in the beginning diminish with the cooling time for most isotopes. The biggest differences of 84.6% for ¹³³I and - 90.7% for ⁹³Y directly come from the differences of yield data between JEFF-3.1.1 and ORIGEN reference library, i.e., 85.3% for ¹³³I and -82.2% for ⁹³Y.



Fig. 5. Relative differences (%) of JEFF-3.1.1-based total decay heats for the 10 most influential isotopes to ORIGEN-S reference results over the whole time interval after 235 U thermal fission.

Figure 7 shows the relative differences of total decay heat calculated with the new fission product yield libraries to the ORIGEN-S reference results. As expected from Fig. 5, the JEFF-3.1.1 yield data brings about the biggest differences with the reference out of all new yield libraries. The ENDF/B-VII.1 shows slight differences of within 1% with the reference yield library based on ENDF/B-VII.0. The maximum difference between two JENDL-based libraries is about 1.3% at very short cooling times. As shown in Fig. 8, the calculated decay heats vary with the yield libraries as much as shown in Fig. 7. In spite of this, all libraries have a tendency to follow the measurements within the experimental error bounds.



Fig. 6. Relative differences (%) of JENDL/FPY-2011-based total decay heats for the 10 most influential isotopes to ORIGEN-S reference results over the whole time interval after ²³⁵U thermal fission.



Fig. 7. Relative differences (%) of total decay heats for different yield data to ORIGEN-S reference results over the whole time interval after 235 U thermal fission.



Fig. 8. Comparison of decay heats for ²³⁵U thermal fission with respect to different yield libraries.

4.3 Decay Heat Analysis for ²³⁹Pu by Different Yield Data

Figures 9 - 11 show the decay heat calculation results for the ²³⁹Pu thermal-neutron fission. Even for the JEFF-3.1.1 showing the biggest differences with the reference, the magnitude of differences for the 10 most influential isotopes shown in Fig. 9 becomes smaller when compared to 235 U.



Fig. 9. Relative differences (%) of JEFF-3.1.1-based total decay heats for the 10 most influential isotopes to ORIGEN-S reference results over the whole time interval after ²³⁹Pu thermal fission.



Fig. 10. Relative differences (%) of total decay heats for different yield data to ORIGEN-S reference results over the whole time interval after ²³⁹Pu thermal fission.



Fig. 11. Comparison of decay heats for ²³⁹Pu thermal fission with respect to different yield libraries.

Figure 10 shows the relative differences of total decay heats calculated with the new fission product yield libraries to the ORIGEN-S reference results. In contrast with the ²³⁵U, the differences by ENDF/B-VII.1 slightly increase up to about 1.2%, while the

maximum difference by JEFF-3.1.1 drastically decreases to about 5.5% at very short cooling times. The maximum difference between two JENDL-based libraries is about 1.5% right after the fission has ceased. Accordingly, the variations of the calculated decay heats by the different yield libraries become small, as shown in Fig. 11.

5. Summary and Future Work

The decay heat measurements for very short cooling times after thermal-neutron fission of ²³⁵U and ²³⁹Pu have been evaluated by the ORIGEN-S summation calculation. The reference calculation results by the latest ORIGEN data libraries of the SCALE-6.1.3 have been validated with the measurements by ORNL and Studsvik. In addition, the generation of the new ORIGEN yield libraries has been completed based on the ENDF/B-VII.1, JEFF-3.1.1, JENDL/FPY-2011, and JENDL-4.0. The new libraries have been successfully applied to the decay heat calculations and comparative analyses have been devoted to verifying the importance of the fission product yield data when estimating the decay heat values for each isotope in a very short time.

The decay data library occupies an important position in the ORIGEN summation calculation along with the fission product yield library. There are some radioactive decay data files available from ENDF/B, JEFF, and JENDL. The decay heat analyses by the different decay data files remain to be conducted in the near future.

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