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Calculation of neutron emission from spent LWR fuel assemblies: SNF method and validation

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Abstract

The SNF program calculates the isotopic inventories and radiation source terms of spent LWR fuel assemblies based on the multi-dimensional methods employed in in-core fuel management (ICFM), thus utilizing models with precise neutron spectra, detailed power histories and a nodal representation of the fuel assemblies. It is applicable to all fuel types that may be represented by the ICFM system.

Acceptability for loading of spent fuel assemblies into dry casks depends on both gamma- and neutron source rates, as well as on burnup, decay heat power, etc. Conservatism in the applied calculation model may lead to overestimation of the cooling times required to reduce the radiation to acceptable levels and thus there is considerable incentive to apply high accuracy methods for the source terms calculations. The aim of this paper is to review the SNF method with emphasis on prediction of the neutron emission rates from spent LWR fuel assemblies and to present the underlying, experimental validation.

1. Introduction

The SNF program (Børresen, 2004) is routinely used to improve the utilization of fuel storage casks and for calculation of the core or fuel pool decay heat (Becker, et al., 2007). Acceptability for loading of spent fuel assemblies into dry casks depends on both gamma- and neutron source rates, as well as on burnup, decay heat power, etc. Conservatism in the applied calculation model may lead to overestimation of the cooling times required to reduce the radiation to acceptable levels. Due to the relatively slow decay rate of the total neutron source, an overestimation by only 10% will result in more than 30 months additional waiting time. Avoiding such additional waiting time becomes increasingly important as the on-site fuel pool capacities become exhausted.

The neutron release rates due to spontaneous fissions and (α, n)-reactions increase strongly for increased burnup and hence the neutron emission rate may become a limiting factor w.r.t. cask loading, especially for high-burnup fuel. Thus there is considerable incentive to utilize a precise,

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qualified calculation model for the total neutron source of spent fuel assemblies.

The aim of this paper is to review the SNF method for prediction of the neutron emission rates from spent LWR fuel assemblies and to present the underlying, experimental validation.

2. Method of calculation

SNF is based on the isotopic summation method using nodal, isotopic concentrations based on standard 2-D/3-D CASMO/SIMULATE (Edenius, et al., 1995) or HELIOS/PRESTO-2 (J. J. Casal, et al., 1991) core-tracking calculations. This assures consistency between in-core and back-end methods. The nuclear libraries of CASMO or HELIOS are based on either JEF-2.2 or ENDF/B-VI. The required 2-D/3-D calculations are performed as part of the normal ICFM activity and thus very little additional effort and quality assurance is required. The ICFM results are usually continuously verified against in-core detector systems, etc., and may thus be applied with confidence.

The nodal isotopic concentrations at the time of discharge are calculated by a special integration method where the nodal power densities of all time-steps of the 3-D simulation are taken into account. Many fission products and also many actinides, e.g. ^{238}Pu, ^{241}Am and ^{242}Cm, are quite sensitive to the detailed power history.

The SNF decay chains have been extended, relative to those of the ICFM codes, to include e.g. all lower actinides of importance for decay times up to 100,000 years. The decay chains have been verified by comparison with ORIGEN-S. The final radioactivity, gamma-power, total power, and photon spectrum are obtained by multiplication with SNF isotopic library data, for the most part derived from ENDF/B-VI.

2.1. Neutron yield data

The neutron emission due to spontaneous fission and (α, n)-reactions, of a given isotope, is calculated by multiplication of the calculated isotopic concentration with the respective neutron yield data. The neutron yield data of the SNF library are consistent with the new ORIGEN-S data (Gauld et al., 2002), supplemented by Japanese data (Matsunobu et al.,1992) for the lower actinides of e.g. the ^{235}U and the ^{233}U decay series. The spontaneous fission yields were compared with data calculated using ENDF/B-VI decay constants and alpha-branching factors. The resulting yields were in good agreement for all isotopes except ^{217}At for which the JAERI yield had to be corrected by a factor of 10. The library contains neutron yield data for a total of 43 actinides.

The uncertainty in the basic library data has a non-negligible influence on the calculated neutron yields. As an example, the influence of the new library data, relative to the previous ORIGEN-S version data is shown in Table 1. The effect on the calculated spontaneous fission, alpha-n and total neutron source rates for a BWR-LEU lattice and a PWR-MOX lattice are shown. As may be seen, the new data gives a reduction of 8.2 - 8.4% in the alpha-n source and a reduction of 2.8 - 3.0% in the total neutron source in the most important cooling time range, from 3 to 30 years. This reduction gives a ‘saving’ of 8-9 months cooling time to get the same neutron source level as before. It is also seen that there is only little difference between the results for the LEU and MOX lattices.

2.2. 3-D vs. 0-D neutron source calculation

The nodal representation of the fuel assemblies, which is the same as that of the 3-D simulator is essential for proper calculation of the neutron source but somewhat less important for other source terms, such as activity and decay heat. This is mostly due to the non-linear burnup dependence of the curium isotopic concentrations. Fuel assemblies with axial zoning, such as BWR assemblies with axial enrichment zones, part length fuel rods, etc. obviously require special attention and would automatically be properly represented using SNF with a 3-D nodalization.

An example of source terms differences, in percent, between calculations with and without a nodal representation is shown in Fig. 1. The total neutron source, which is the sum of the spontaneous fission and alpha-n-source contributions, show differences of 47-49% for cooling times from 3 to 30 years in this example. This difference remains very high up to about 3000 years. It is gradually reduced to 10-12% for higher cooling times. As may be seen in Fig. 1, the 3-D vs. 0-D differences of the decay heat and the activity are much less (<6%).

Table 1
%-Change in calculated neutron source rates due to new SNF library data.

Tcool (years)	BWR-LEU 30,000 MWd/t			PWR-MOX 30,000 MWd/t		
	Sp.fiss.	Alpha-n	Tot.n	Sp.fiss.	Alpha-n	Tot.n
1	-1.5	-6.5	-1.8	-1.7	-6.4	-1.9
3	-2.7	-8.2	-2.8	-2.7	-8.2	-2.8
10	-2.7	-8.4	-2.9	-2.7	-8.4	-2.8
30	-2.6	-8.4	-3.0	-2.6	-8.4	-2.8
100	-1.4	-8.4	-3.8	-1.3	-8.4	-2.8
300	0.6	-8.4	-3.6	1.9	-8.4	-2.0
1000	0.6	-8.5	-2.0	2.0	-8.5	-0.3

This example illustrates the necessity of a nodal (3-D) calculation for evaluation of the total neutron emission rates of spent fuel assemblies. Without such a model, the neutron source may be seriously underestimated.

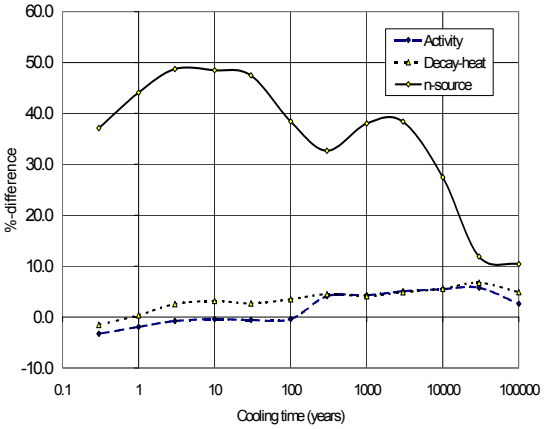

Fig. 1 %-difference in activity, decay heat and neutron source between 3-D and 0-D SNF calculations (BWR fuel at 41 GWd/t).

3. Neutron source validation

The experimental validation of the neutron emission rates calculated by the SNF program consists of two parts: 1) Isotopic measurement comparisons of isotopes contributing the neutron source, such as ^{242}Cm, ^{244}Cm and ^{246}Cm and 2) comparisons with fuel assembly neutron emission experiments.

Comparisons with fuel assembly decay heat measurements provide additional validation since the neutron generating isotopes also contribute significantly to the decay heat, especially at high burnup. As an example, ^{244}Cm generates 15-20% of the decay heat and about 95% of the neutron source in the cooling time range from 3 to 30 years for a typical spent BWR fuel assembly.

3.1. Decay heat comparisons

The decay heat of large number of BWR and PWR spent fuel assemblies from Swedish reactors was measured by a calorimetric method in the CLAB intermediate fuel storage facility. SNF calculations were carried out for a total of 16 BWR and 33 PWR fuel assemblies. A summary of the results was presented by Becker et al. (2007). A plot of calculated versus measured decay heat for the 49 assemblies is shown in Fig. 2. The average calculation/experiment (C/E) ratio was 0.994 ± 0.019, thus showing excellent agreement. The average C/E for the 8 BWR and 17 PWR fuel assemblies that corresponded with the fuel types included in the neutron emission experiments described below was 0.991±0.041, respectively 0.993±0.011, hence very good agreement was obtained for both BWR and PWR.

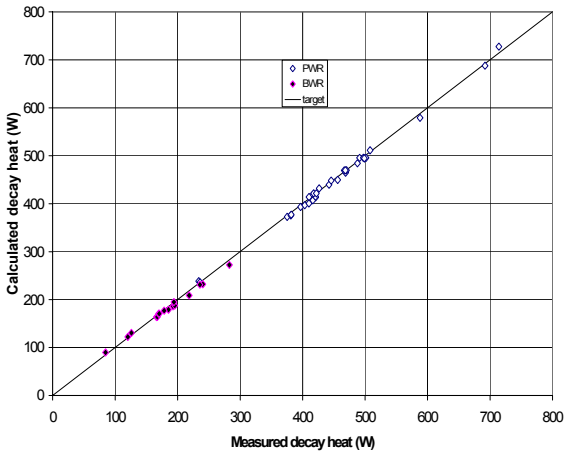

Fig. 2 SNF calculated versus measured decay heat of 49 BWR and PWR spent fuel assemblies

3.2. Isotopic measurement comparisons

Isotopic comparisons with the JAERI benchmark (Nakahara et al., 2002) have been performed with both CASMO-4 and HELIOS using the JEF-2.2 and ENDF/B-VI cross-section libraries. A general trend has been an under-prediction of the curium isotopes, typically by 10-20% for ^{244}Cm and even more for ^{242}Cm and ^{246}Cm. Calculations using a preliminary ENDF/B-VII library have shown an increase of 3-4% in ^{244}Cm and 12-15% in ^{246}Cm relative to the ENDF/B-VI library results, and thus improved agreement.

HELIOS comparisons for both LEU and MOX fuel samples of the ARIANE experiments have been reported by ORNL (Murphy and Primm, 2002). They also found an under-prediction of curium for the LEU fuel, but much better agreement for the MOX samples (very good agreement for ^{244}Cm and 10-20% under-prediction for ^{242}Cm and ^{246}Cm).

3.3. Neutron emission experimental data

Experimental neutron emission rates of spent BWR-8x8 fuel assemblies and also of some PWR 15x15 fuel assemblies, have been reported by Würz (1991). The measurements were performed in FAMOS facility of Kernforschungszentrum Karlsruhe, Germany. The measured data were normalized to 20 months cooling time and the ^{242}Cm contribution was eliminated. The author found that the experimental primary neutron emission, $\overline{ne_p}$ in (n/s)/tU, after correction for self-multiplication, could be fitted as $\overline{ne_p} = a(BU)^b$ where a and b are constants and BU is the assembly burnup in GWd/tU. The constant b was found to be 3.97 for BWR fuel and 3.94 for PWR fuel. The constant a depends on the fuel design and initial enrichment, ε. Using the Würz data for 8x8-BWR fuel in the enrichment range 2.70 to 2.90 % ^{235}U (assumed average of 2.80 %), we found that the corresponding a-value could be fitted as $a = a_{2.8} \cdot e^{-c(\varepsilon - 2.8)}$ with $a_{2.8} = 421$ and c=0.649. This gives an absolute value of 6.00x10^8 n/(sxtHM) at 35.5 GWd/tU, corresponding to the value estimated from the Würz data.

The neutron source at 20 months decay time was calculated with SNF for each of the 16 BWR and 17 PWR-15x15 assemblies of the CLAB decay heat benchmark described above for comparison with the Würz experimental data. The BWR enrichments were in the range from 2.09 to 3.10 % ^{235}U and both regular 8x8-or 9x9 lattice fuel and advanced 'water-cross' (SVEA) fuel types were included. The results, normalized to an enrichment of 2.8 % are shown in Fig. 3, together with the experimental correlation for the enrichment range 2.70 to 2.90 % ^{235}U. The lines marked 'upper limit' and 'lower limit' correspond to the quoted experimental, 1σ uncertainties. As may be seen, the 8x8/9x9 results agree very well with the experimental correlation whereas the 'water-cross' results fall below, close to the lower limit line. The lower neutron source of the water-cross fuel is consistent with the somewhat 'softer' neutron spectrum of such fuel compared with that of the 8x8 fuel used in the experiment. The average calculation/experiment (C/E) ratio of the 8x8/9x9 BWR assemblies is 0.98±0.06. This is considered very good agreement and does not indicate any significant error in the calculated ^{244}Cm concentrations.

The PWR results are shown in Fig. 4. The PWR fuel enrichments were in the range 3.10 to 3.25 % ^{235}U and the calculated values were normalised to 3.2 % for comparison with the experimental data for the enrichment range 3.1 to 3.2 % ^{235}U. The absolute value of the neutron source was, in this case, estimated to 3.00x10^8 n/(sxtHM) at 35 GWd/tU. The calculated points of the 15x15 PWR assemblies fall slightly above the Würz correlation line, but within the experimental uncertainties. The average C/E-ratio is 1.14±0.03 as shown in Table 2. The burnup trend, however, is consistent with the experimental

correlation. It should be remarked that the neutron self-multiplication is larger for PWR than for BWR fuel assemblies. Thus, correction for this effect could possibly make the PWR experimental data somewhat more uncertain. Also, the detailed design (except for type of lattice, e.g. 15x15, and enrichment) and the irradiation histories of the measured assemblies are not specified and may hence differ somewhat from those of the SNF calculations.

However, the overall agreement, considering both the BWR and PWR assemblies is satisfactory.

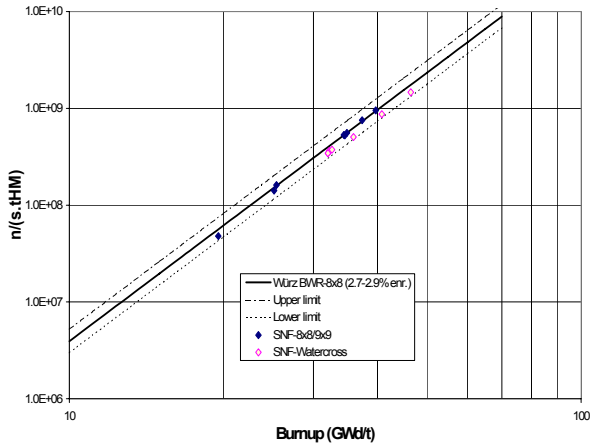

Fig. 3. SNF vs. Würz neutron emission correlation for CLAB BWR assemblies.

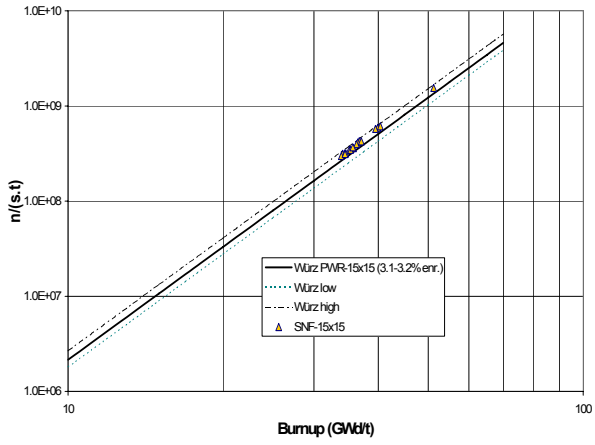

Fig. 4. SNF vs. Würz neutron emission correlation for CLAB PWR fuel assemblies.

A summary of the comparisons of calculated and measured data of both neutron source and decay heat of the same BWR and PWR fuel assemblies is shown in Table 2. The only noticeable deviation is that of the PWR neutron source which is over-predicted by 14 %.

Table 2
Summary of C/E-average and std. deviations, SNF calculation vs. experiment

Fuel assembly type	Number of assemblies	Neutron source	Decay heat
BWR-8x8/9x9	8	0.98±0.06	0.991±0.041
PWR-15x15	17	1.14±0.03	0.993±0.011

4. Summary and conclusions

The SNF program is used to perform spent fuel calculations based on multi-dimensional methods by utilising models and calculation results of the Studsvik ICFM codes CASMO/SIMULATE or HELIOS/PRESTO-2. The focus of this paper is on the ability of the SNF program to predict the neutron emission rates of spent fuel assemblies. Three aspects have been discussed; 1) the influence of the neutron yield library data, 2) the importance of 3-D effects on the assembly neutron emission rates, and 3) the validation of neutron emission rates against experimental data of BWR and PWR fuel assemblies.

It was found that the latest revision of the SNF neutron yield library data reduces the overall neutron emission rates of both LEU and MOX fuel by typically about 3% in the most important cooling time range.

Using a calculation example for a BWR fuel assembly, it was shown that the difference in the neutron source between 3-D and 0-D SNF calculations reached nearly 50 % for cooling times from 3 to 30 years. Thus the importance of 3-D calculations is emphasised.

The experimental validation of the SNF neutron emission rates of spent BWR and PWR fuel assemblies was discussed, both in terms of isotopic data comparisons (curium isotopes) and by comparison with published data on measured emission rates of fuel assemblies. The curium

isotopic comparisons for uranium fuel tend to show some under-prediction, in the order of 10 to 20 %, however, the comparisons with the neutron emission experiments show very good agreement for BWR fuel and even some over-prediction for the PWR fuel. This is not consistent with any under-prediction of the curium contents. Also, comparisons with decay heat measurements of the same set of fuel assemblies that were used for the neutron emission comparison showed excellent agreement.

The deviations between the various experiments related to neutron emission and the corresponding calculations are generally small and may as well reflect experimental uncertainties. It is concluded that SNF may be applied with confidence for calculation of the neutron emission of spent BWR and PWR fuel assemblies.

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