LACKING SPENT NUCLEAR FUEL CRITICAL BENCHMARKS? - GOT REACTOR CRITICALS?

William J. Anderson
Framatome ANP, Inc.

ABSTRACT

With increased interest in the use of burnup credit (BUC) for spent nuclear fuel (SNF) storage, transportation and disposal, the scarcity of SNF critical experiment's is a serious obstacle to successful licensing of BUC applications. Historically, laboratory critical experiments (LCE) have been used for criticality benchmarks. However, for SNF applications, no directly applicable LCEs are available. What is available is an enormous supply of critical measurements used in operations already licensed by the U.S. Nuclear Regulatory Commission (NRC) (i.e., commercial reactors).

While commercial reactor criticals (CRCs) are not perfect matches to the system of interest, it is important that the fundamental parameters of the benchmarks be similar. Currently, CRCs are the only SNF critical benchmarks available. Their materials, geometry, and spectrum compare favorably to those in BUC applications. Reactor criticals are the answer to the benchmark obstacle for BUC.

INTRODUCTION

With increased interest in the use of burnup credit (BUC) for spent nuclear fuel (SNF) storage, transportation and disposal, the scarcity of SNF critical experiment's is a serious obstacle to successful licensing of BUC applications. Historically, laboratory critical experiments (LCE) have been used for criticality benchmarks. However, for SNF applications, no directly applicable LCEs are available. What is available is an enormous supply of critical measurements used in operations already licensed by the U.S. Nuclear Regulatory Commission (NRC) (i.e., commercial reactors).

A commercial reactor critical (CRC) is a zero-power critical measurement used to verify the license conditions of the reactor. Various parameters are measured and recorded as part of the critical. The data are used to define the critical configuration at the time of the critical measurement. In addition to the critical configuration definition, these measurements are used to verify the shutdown capability of the reactor (i.e., control rod worth) and moderator temperature coefficient of reactivity. These are license specifications that must be confirmed prior to power ascension.

Core follow/operating history data are available for use in computer simulations of depletion of the fuel. The simulation allows the utility to predict the isotopic composition of the nuclear fuel in the reactor resulting from the specific history. Using these predicted isotopic concentrations, the user can calculate the expected reactivity of the reactor core for the various zero-power critical configurations. These predicted values can then be compared to the actual measured values from the CRC.

Therefore, modeling these CRCs relies on real, measured data obtained for measurement techniques that are already accepted by the NRC. But is a CRC really acceptable as a criticality benchmark?

According to Lichtenwalter (1), "The calculation method used to establish the criticality safety of transportation and storage packages needs to be validated against measured data that have been shown to be applicable to the package under consideration." Lichtenwalter goes on to say that there are three fundamental parameters that should be considered in selecting experiments: materials (fissionable, nonfissionable, reflector, moderator, form, temperature, and ratio of nonfissionable to fissionable); geometry (positions, size, homogeneity -vs- heterogeneity, shape, lattice pattern spacing and interstitial materials, reflection); and neutron spectrum (leakage, absorption, production and flux).

While no benchmark perfectly matches the system of interest, the fundamental parameters of the benchmarks must be similar. A benchmark must be compared to the expected system to ensure that sufficient similarity exists. This paper compares CRCs to the expected SNF environment of a waste package (WP) to demonstrate that sufficient similarity exists between the two and to justify the use of CRCs for critical benchmarks of BUC application environments.

MATERIALS

The first fundamental parameter for consideration is "Materials." This includes consideration of the fuel materials, the moderator, the reflector, and any neutron-absorbing poisons.

Fuel Materials

For the purposes of this paper, the fuel materials include all of the actual fuel material, including any integral burnable absorbers, the grid spacers and assembly lattice support materials, and the cladding. It is clear that the materials comprising the assembly and the cladding do not change between the reactor and the WP. However, due to the nature of the irradiated fuel, the actual composition of the fuel material is affected by the decay of radioactive isotopes, and thus changes with time after shutdown.

Table I lists the "major" isotopes and their half-lives. The table also displays the percentage of the total neutron absorption represented by the neutron absorption in each isotope and the time of peak absorption rate, as a fraction of the total, for each isotope. This list of isotopes is based on work performed by the Yucca Mountain Project (2) and Oak Ridge National Laboratory (3). The absorption rate data represents a hypothetical pressurized water reactor (PWR) assembly with an initial enrichment of 4.5 wt% ^{235}U and a burnup of 50 GWd/mtU.

Table II shows the same data as Table I but sorted by percentage of total absorption. The top four neutron absorbers are ^{239}Pu, ^{238}U, ^{235}U, and ^{240}Pu. The contribution of these four isotopes to the total neutron absorption represents about 77% of the total neutron absorption in the fuel. The contributions of each of the isotopes to the total neutron absorption in the fuel do not appreciably change over the first 200 years of cooling. This is also true for 17 of the other 23 isotopes in the list.

Of the 27 major isotopes, only 6 (^{241}Am, ^{241}Pu, ^{155}Gd, ^{151}Sm, ^{151}Eu, and ^{234}U) show a significant change in their neutron absorption worth over the first 200 years. Two of these isotopes (^{241}Pu and ^{151}Sm) "peak" within two years of discharge. The other four peak later in time.

Although Reference 3 shows both ^{241}Pu and ^{151}Sm peaking at two years after discharge (the beginning point for the data reported), their actual peaks occur before two years. The ^{241}Pu isotope decays faster than its parent isotopes. Its peak concentration occurs at discharge. It is therefore "bounded" by the available CRC information.

The ^{151}Sm peaks about 10 days after discharge. To bound the peak worth of this isotope, the database of CRCs should include a range of times between the shutdown of the reactor and the CRC. By including a range of cooling times, the benchmark database can be certain to consider a range of ^{151}Sm concentrations/worths.

The ^{155}Gd, although not adequately represented in the CRC case presented here, can easily be benchmarked using CRCs from reactors that use gadolinia as a burnable absorber. These data can also be supplemented with the 20 Urania-Gadolinia experiments performed by Babcock & Wilcox involving a variety of enrichments and absorbers (4). These experiments were designed to simulate a PWR fuel assembly lattice containing fresh fuel.

The remaining three isotopes (^{241}Am, ^{151}Eu and ^{234}U) represent a maximum total neutron absorption worth of approximately 5.5% of the neutron absorption in the fuel; 4.8% of that is represented by the ^{241}Am alone. The ^{151}Eu and the ^{234}U "peaks" occur at 200 years and are 0.7% and 0.3%, respectively. The impact of ^{151}Eu on the k_{eff} of a system is further minimized, because it is a non-fissionable isotope. Its impact is only in the loss factor of the k_{eff} calculation. On the other hand, ^{241}Am and ^{234}U are fissionable, but they only account for approximately 0.01% - 0.05% of the production. The production term is dominated by just four isotopes (^{235}U, ^{238}U, ^{239}Pu, and ^{241}Pu), which account for more than 99.5% of the total neutron production.

Table I: Major Isotopes

Isotope	$T_{1/2}$	Approximate Range of Percentage of Absorption Rate[a]	Time of Peak Fraction of Absorption Rate[a]
Molybdenum-95	Stable	0.29%	Flat
Technitium-99	2.13×10^5 years	0.61%	Flat
Ruthenium-101	Stable	0.21%	Flat
Rhodium-103	Stable	1.5%	Flat
Silver-109	Stable	0.27%	Flat
Cesium-133	Stable	0.78%	Flat
Neodymium-143	Stable	1.7% - 1.8%	200 years
Neodymium-145	Stable	0.4%	Flat
Samarium-147	Stable	0.22% - 0.34%	200 years
Samarium-149	Stable	1.6% - 1.7%	200 years
Samarium-150	Stable	0.3%	Flat
Samarium-151	90 a	0.23% - 1%	2 years
Samarium-152	Stable	0.52%	Flat
Europium-151	Stable	0 - 0.65%	200 years
Europium-153	Stable	0.45%	Flat
Gadolinium-155	Stable	0.39% - 1.7%	200 years
Uranium-234	2.46×10^5 years	0.12% - 0.3%	200 years
Uranium-235	7.04×10^8 years	18%	Flat
Uranium-236	2.34×10^7 years	1%	Flat
Uranium-238	4.47×10^9 years	25%	Flat
Neptumium-237	2.14×10^6 years	0.8% - 1.1%	200 years
Plutonium-239	2.4×10^4 years	27%	Flat
Plutonium-240	6560 a	7%	Flat
Plutonium-241	14.3 a	0% - 7%	2 years
Plutonium-242	3.8×10^5 years	0.6%	Flat
Americium-241	432 years	0.6% - 4.8%	50 - 100 years
Americium-243	7370 years	0.3%	Flat

a Fractional absorption rate information is taken from Reference 3 (pp. 13 and 14) and is considered for the first 2 to 200 years only.

Table II: Major Isotopes Sorted Based on Percentage of Total Absorption

Isotope	$T_{1/2}$	Approximate Range of Percentage of Absorption Rate[a]	Time of Peak Fraction of Absorption Rate[a]
Plutonium-239	2.4 $x10^4$ years	27%	Flat
Uranium-238	4.47 x 10^9 years	25%	Flat
Uranium-235	7.04 x 10^8 years	18%	Flat
Plutonium-240	6560 years	7%	Flat
Plutonium-241	14.3 years	0% - 7%	2 years
Americium-241	432 years	0.6% - 4.8%	50 - 100 years
Neodymium-143	Stable	1.7% - 1.8%	200 a
Samarium-149	Stable	1.6% - 1.7%	200 a
Rhodium-103	Stable	1.5%	Flat
Gadolinium-155	Stable	0.39% - 1.7%	200 years
Uranium-236	2.34 x 10^7 years	1%	Flat
Neptumium-237	2.14 x 10^6 years	0.8% - 1.1%	200 years
Samarium-151	90 a	0.23% - 1%	2 years
Cesium-133	Stable	0.78%	Flat
Technitium-99	2.13 $x10^5$ years	0.61%	Flat
Plutonium-242	3.8 x 10^5 years	0.6%	Flat
Samarium-152	Stable	0.52%	Flat
Europium-153	Stable	0.45%	Flat
Europium-151	Stable	0 - 0.65%	200 years
Neodymium-145	Stable	0.4%	Flat
Samarium-147	Stable	0.22% - 0.34%	200 years
Americium-243	7370 years	0.3%	Flat
Samarium-150	Stable	0.3%	Flat
Molybdenum-95	Stable	0.29%	Flat
Silver-109	Stable	0.27%	Flat
Ruthenium-101	Stable	0.21%	Flat
Uranium-234	2.46 x 10^5 years	0.12% - 0.3%	200 years

a Fractional absorption rate information is taken from Reference 3 (pp. 13 and 14) and is considered for the first 2 to 200 years only.

Although the total expected impact of ^{241}Am, ^{151}Eu, and ^{234}U on k_{eff} is expected to be small, their effect must be addressed in validation efforts. The isotopic concentrations are being measured in a series of radiochemical assay measurements that support the Yucca Mountain Project. Consideration may also be given to "worth" measurements for these isotopes. Such experiments are already underway under a current Nuclear Energy Research Initiative (NERI) project to measure the worth of rhodium, samarium, and cesium isotopes.

In general, the stable isotopes and those with peak absorption worths at two years cooling are bounded by the CRC isotopics. The ^{155}Gd can be validated by including the available urania-gadolinia LCEs (4). The remaining three isotopes (^{241}Am, ^{234}U, and ^{151}Eu) are not adequately covered by the CRC isotopics, but neither are they particularly significant to the WP system k_{eff}. Their total reactivity worth is expected to be significantly less than 1%. Overall, the CRCs, when supported with appropriate LCEs (i.e., Reference 4), adequately cover the expected range of isotopics for the BUC application environments.

Ratio of Fissionable to Nonfissionable in Fuel

In addition to the comparison of CRCs to the burnup credit application environments on an isotopic basis, it is also valuable to consider the ratio of fissionable to nonfissionable materials in the fuel. This comparison can be accomplished using the data in Reference 3, Table III shows the estimated fractional absorption rates for the isotopes of interest at several cooling times. Both the fissionable and nonfissionable isotope totals are reported along with the calculated ratio.

Although the ratios do show a downward trend with decay time, the difference between the two-year and the 100-year cases is not large. The decrease in the ratio is due to increased fractional absorption in the nonfissionable isotopes with a coinciding decrease in the fractional absorption in the fissionable isotopes. The effect appears to be primarily related to the changes in ^{241}Pu, ^{241}Am, and ^{155}Gd. The change in the ratio is really a change in just three isotopes with the other isotopes remaining close to constant. This further supports the argument for adequate similarity between the fuel material in the CRCs and that in the potential BUC application environment.

Table III: Fractional Absorption Rates for Various Decay Times(3)

	Fractional Absorption Rate (% of Total)			
Isotope	2 years	5 years	19 years	100 years
Molybdenum-95	0.29%	0.29%	0.29%	0.29%
Technitium-99	0.61%	0.61%	0.61%	0.61%
Ruthenium-101	0.21%	0.21%	0.21%	0.21%
Rhodium-103	1.50%	1.50%	1.50%	1.50%
Silver-109	0.27%	0.27%	0.27%	0.27%
Cesium-133	0.78%	0.78%	0.78%	0.78%
Neodymium-143	1.7%	1.7%	1.8%	1.8%
Neodymium-145	0.40%	0.40%	0.40%	0.40%
Samarium-147	0.22%	0.28%	0.32%	0.34%
Samarium-149	1.60%	1.60%	1.70%	1.70%
Samarium-150	0.30%	0.30%	0.30%	0.30%
Samarium-151	1%	1%	0.90%	0.49%
Samarium-152	0.52%	0.52%	0.52%	0.52%
Europium-151	0	0	0	0.5%
Europium-153	0.45%	0.45%	0.45%	0.45%
Gadolinium-155	0.4%	0.8%	1.2%	1.5%
Nonfissionables	10.2%	10.7%	11.3%	11.6%
Uranium-234	0.1%	0.1%	0.1%	0.2%
Uranium-235	18%	18%	18%	18%
Uranium-236	1%	1%	1%	1%
Uranium-238	25%	25%	25%	25%
Neptumium-237	0.80%	0.80%	0.80%	1%
Plutonium-239	27%	27%	27%	27%
Plutonium-240	7%	7%	7%	7%
Plutonium-241	7.0%	6.0%	5.0%	0.0%
Plutonium-242	0.60%	0.60%	0.60%	0.60%
Americium-241	0.6%	1.3%	2.0%	4.8%
Americium-243	0.30%	0.30%	0.30%	0.30%
Fissionables	87.4%	87.1%	86.8%	84.9%
Ratio[a]	8.5	8.2	7.7	7.3

a Ratio is the ratio of the total fraction absorption rates for all fissionable isotopes divided by that of the nonfissionable isotopes.

Fuel Form and Temperature

CRCs and commercial SNF environments involve irradiated uranium dioxide in pellet form. The assemblies in the CRCs and the commercial SNF environments represent the same lattice of fuel rods containing the same spent uranium dioxide fuel pellets. It is clear that the form of the fuel is identical in both applications.

Table IV shows the expected values of the fuel temperatures CRCs and the potential Yucca Mountain waste package (WP) (5). The peak temperature for the WP occurs at about 20 years, which is well within the range of cooling times for SNF in other SNF environments (i.e., storage or transport). The difference between the PWR and the boilng water reactor (BWR) results from the fact that the PWR CRCs are hot, zero-power criticals while the BWR CRCs are cold, zero-power criticals. The data in Table IV demonstrate that a database of CRCs including BWRs and PWRs will adequately cover the entire range of fuel temperatures expected in the SNF environments under consideration.

Table IV: Expected Values of Fuel Temperatures

Data set	T_{fuel} (K)
WP	300 - 475[a]
BWR CRCs	330
PWRs CRCs	550

a Range of fuel temperatures taken from Figure 3-22 of Reference 5.

Moderator

In both the reactor and the burnup credit application, the moderator is water. However, the water will be at various temperatures and densities. Table V shows the moderator temperatures and densities expected in the CRCs and the Yucca Mountain WP (5). As with the fuel temperatures reported in Table IV, the difference between the PWR and the BWR values results from the fact that the PWR CRCs are hot, zero-power criticals while the BWR CRCs are cold, zero-power criticals.

Table V: Expected Values of Important Moderator Parameters

Data set	T_{mod} (K)	ρ_{mod} (g/cm^3)
WP	300 - 475[a]	~1
BWR CRCs	330	~1
PWRs CRCs	550	~0.72

a Range of moderator temperatures taken from Figure 3-24 of Reference 5.

A database of CRCs including BWRs and PWRs will adequately cover the entire range of moderator temperatures. The thermal shift related to the moderator temperature (discussed later, see Figure 4) is well understood and easily accounted for in modern computer codes.

Reflector

The CRCs and the SNF container provide significant reflection in the form of water surrounded by carbon or stainless steel. The CRCs include an effectively infinite water reflector around most of the core, where as the SNF container has less than an infinite water reflector. However, the combination of the water reflector and the steel container equates to a near-infinite reflector combination. The reflectors associated with the CRCs and the SNF containers return a highly thermalized neutron flux to the area of interest, the fuel lattice.

Neutron-Absorbing Poisons

In a PWR, the water is borated. The boron concentration will vary in the PWR CRCs based on the burnup and operation of the individual plants. Typical boron concentrations for an operating PWR range from nearly 0 ppm to about 1,800 ppm. The water in a burnup credit application will most likely not contain soluble boron, but in most instances, there will be boron matrix plates (aluminum or stainless steel). Although soluble boron criticals do not bound the possible range of fixed neutron-absorbing boron for the various burnup credit applications, they can be supported with available laboratory criticals that include various neutron absorbers, including fixed boron (4,6,7,8,9).

GEOMETRY

Lichtenwalter (1) lists position, size, homogeneity versus heterogeneity, shape, lattice pattern, spacing interstitial materials and reflection as the "important items to consider regarding geometry of construction." In these categories, CRCs agree well with the SNF container designs.

The geometry of a CRC fuel assembly is identical to that of a fuel assembly in a container used for storage, transportation or disposal of SNF. However, the assemblies in the SNF container are typically on a different pitch than those in a CRC. In both cases, the fuel is heterogeneous and the shape and dimensions of the fuel rods and the lattice pattern are identical for fuel assemblies of the same type. The shapes (cylindrical systems containing assemblies on a square pitch) of the systems are the same, although containers used for storage, transportation or disposal of SNF are smaller than those for the CRC. As discussed above, the reflectors are also very similar.

The one difference, other than scale, between the reactor and the burnup credit application environment is the assembly pitch. The assemblies in a reactor are typically

on a smaller pitch than the assemblies in a BUC application environment. This difference is primarily a result of the addition of borated metal plates that form the "basket" for holding the assemblies in place. This difference is covered in the previous discussion of "Neutron-Absorbing Poisons."

SPECTRUM

Under the fundamental parameter called "Spectrum," Lichtenwalter (1) includes neutron leakage, absorption and fission reaction rates, and flux spectrum. The following addresses each of these "sub-parameters."

Leakage

Because of the size difference between a reactor and a container used for storage, transportation or disposal of SNF, a CRC has less leakage than an SNF container, but this is a difference between a neutron non-leakage probability for a CRC of 96.5% and for a WP of 94%. These results are based on calculations for a Crystal River, Unit 3 CRC and for a 21-PWR assembly WP. For an SNF container that holds more fuel (e.g., 32-PWR assembly shipping cask) the leakage comparison will be even more favorable.

For comparison, a typical laboratory critical experiment will have a neutron non-leakage probability on the order of 77%. This value obviously represents a much higher leakage than a CRC or a WP.

Absorption Reaction Rates and Fission Reaction Rates

Absorption and fission reaction rates are also available for the WP and CRC systems (10). Figures 1 and 2 compare the absorption reaction rate and the fission reaction rate, respectively, for the same fuel assemblies in the two different environments. Although the reaction rates are not identical, they are clearly very similar.

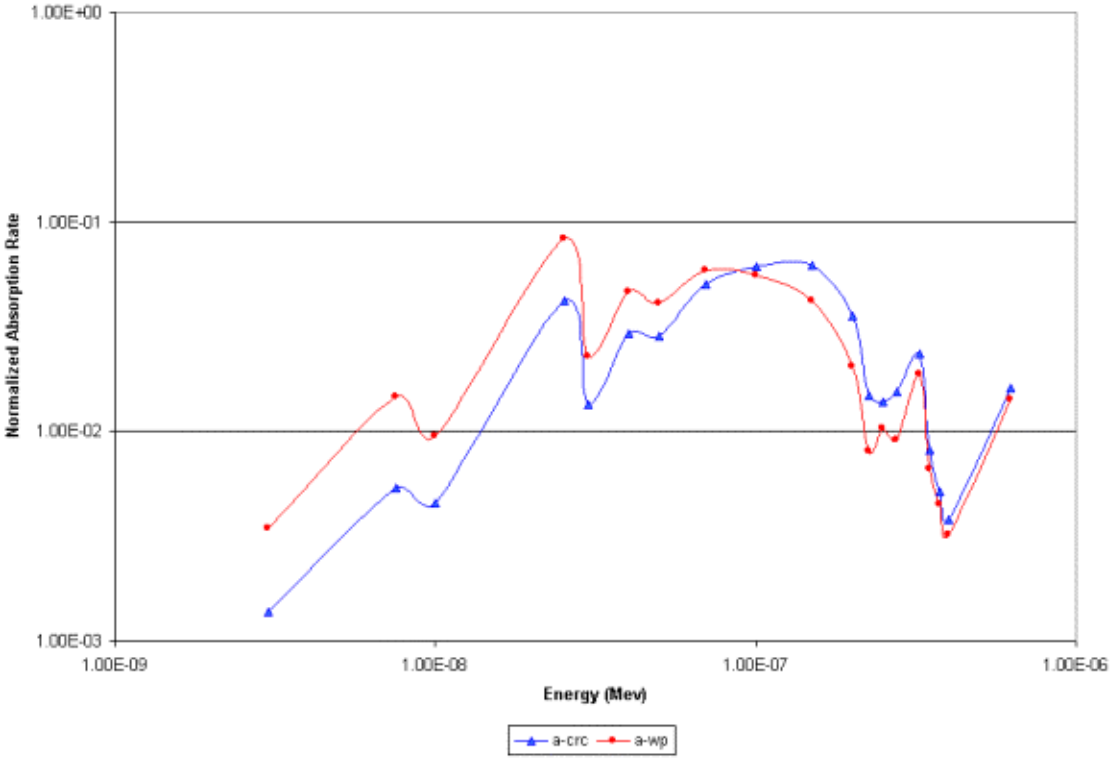

Fig. 1. Absorption Reaction Rate in the Fuel Region of the WP and CRC

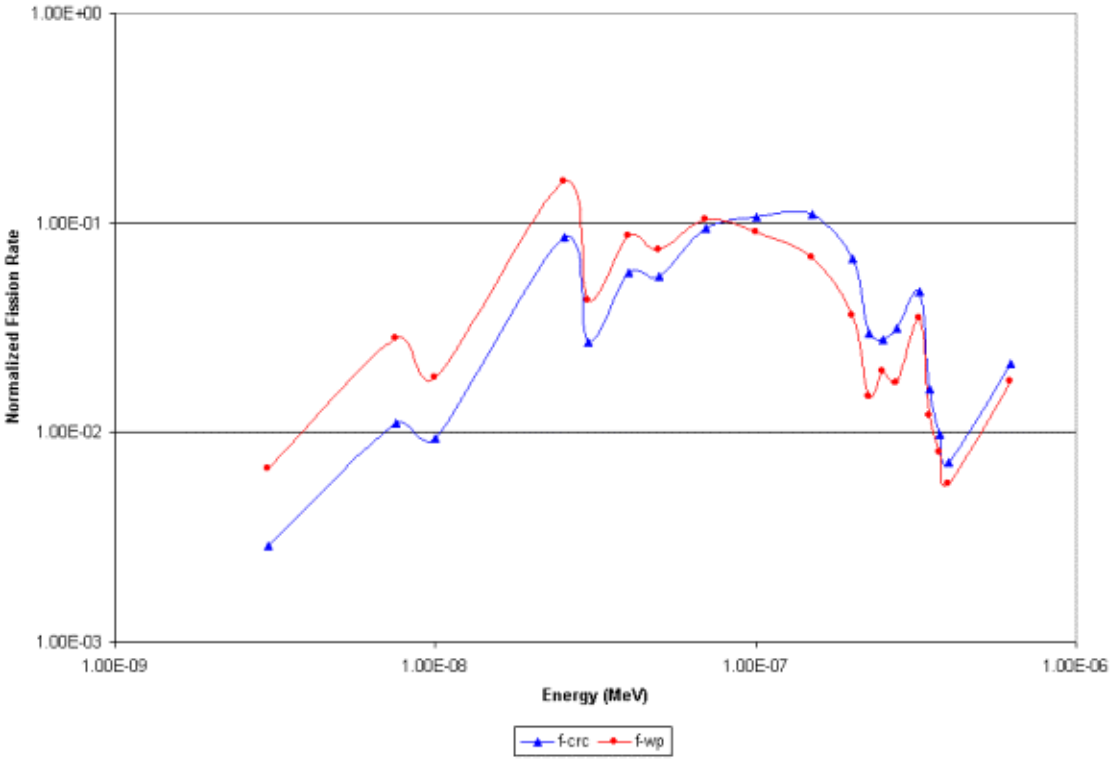

Fig. 2. Fission Reaction Rate in the Fuel Region of the WP and CRC

Neutron Flux

Reference 10 reports the results of criticality evaluations for a selection of benchmark experiments and a 21-PWR WP. The experiments include a Crystal River, Unit 3 CRC, two mixed-oxide (MOX) LCEs, and one low-enriched uranium (LEU) LCE. In each benchmark case, the average neutron flux fraction versus energy was calculated across the system. Figure 3 shows the neutron flux fraction spectra of the five models.

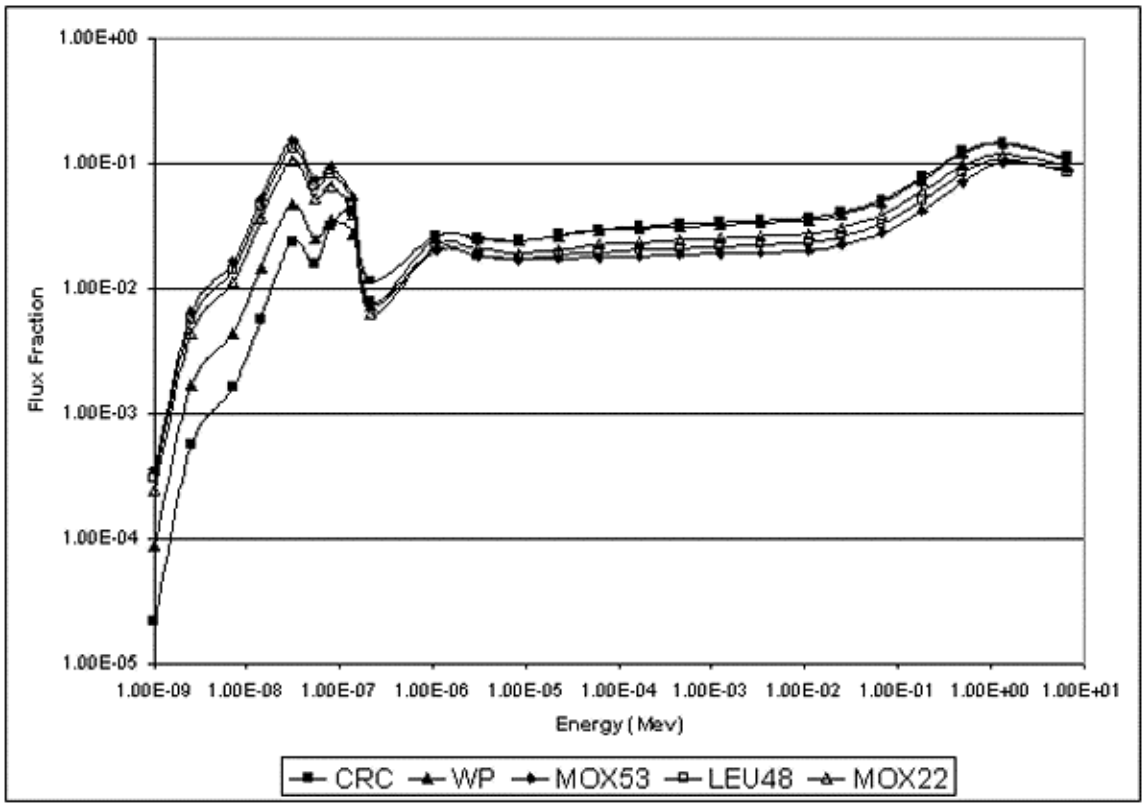

Fig. 3. Neutron Energy Spectrum of WP and Critical Benchmarks

For the CRC and WP, the flux spectra in the source and slowing-down regions are almost identical. A shift in the thermal energy range between the CRC and WP can be noted in the plot with the WP clearly being more thermalized. The three LCE spectra show shifts in the source and slowing-down regions as well as in the thermal energy range. The spectral shifts seen in the LCEs are primarily the result of material and geometry (H/X) difference between the LCEs and the burnup credit application environments. The magnitude of the difference between the LCEs and the WP compared to that of the difference between the CRC and the WP highlights the need for CRC in a BUC application benchmark database.

The shift in the CRC spectra is primarily a result of the temperature difference between the hot, zero-power CRC and the WP. This shift is magnified in Figure 3 by the fact that

the modeled WP was assumed to be at room temperature (300 K). In reality, the WP will go through a range of temperatures from a high of about 425 K to a low of 300 K. The PWR CRC shown in Figure 3 is at a temperature of 550 K. To account for this range, the BWR CRCs must be included in the benchmark database. A BWR CRC is a cold, zero-power critical with a temperature of ~330 K. Including a sufficient number of PWRs and BWRs in the benchmark database allows the analyst to validate the range of expected temperatures in a burnup credit application environment.

Previous work characterizes the temperature-related spectral shift by modeling changes in the various temperature-related parameters (i.e., fuel temperature, uranium temperature, moderator temperature, and moderator density) on an individual basis (11). Figure 4 shows the effect of the changing the various parameters. Although the fuel temperature and the moderator density do affect the total flux in the thermal region, only the moderator temperature actually causes a shift within the thermal region. In fact, the change in the spectrum related to the fuel temperature is barely visible. The effect of moderator density does create in shift in the altitude of the peak. The moderator temperature on the other causes a shift in the altitude of the peak as well as the "position" of the peak. To account for this shift, a benchmark database should include PWR CRCs, BWR CRCs, and room temperature LCEs. This approach will adequately cover the range of expected temperatures in a burnup credit application, as well as "test" the model at a variety of temperature-related spectral shifts.

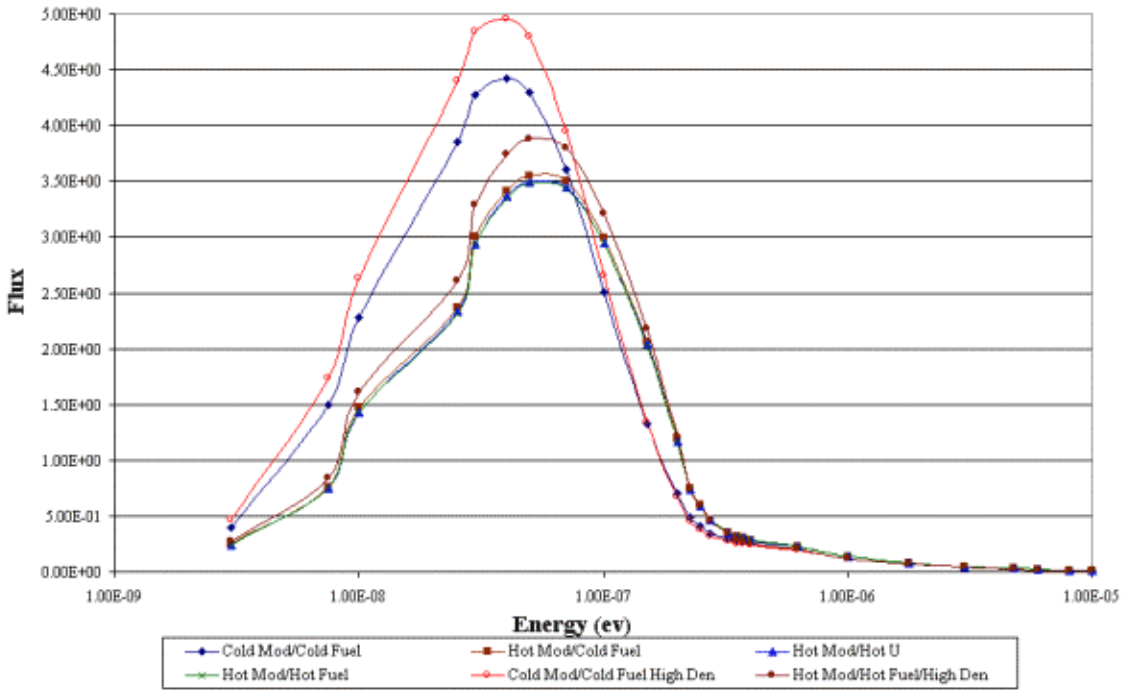

Fig. 4. Spectral Shift Related to Temperature Effects

CONCLUSIONS

It has been shown that CRCs provide important geometric and material composition information that is not currently available in LCEs. It is also clear that LCEs are necessary for filling the gaps in the CRC data. A properly selected benchmark database for BUC applications will include a mix of PWR CRCs, BWR CRCs, and LCEs.

Currently, CRCs are the only SNF critical benchmarks available. Compared to Lichtenwalter's criteria (1) for acceptable benchmarks, CRCs adequately represent the expected range of parameters for a BUC application, with few exceptions. When CRC physical parameters are compared to those of the BUC applications, it is apparent that CRCs can be effectively used to benchmark codes for BUC applications. When supplemented with applicable LCEs, CRCs provide the necessary database for benchmarking BUC SNF models.

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