MCNPX Monte Carlo burnup simulations of the isotope correlation experiments in the NPP Obrigheim

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Abstract
This paper describes the simulation work of the Isotope Correlation Experiment (ICE) using the MCNPX Monte Carlo computer code package. The Monte Carlo simulation results are compared with the ICE-Experimental measurements for burnup up to 30 GWd/t. The comparison shows the good capabilities of the MCNPX computer code package for predicting the depletion of the uranium fuel and the buildup of the plutonium isotopes in a PWR thermal reactor. The Monte Carlo simulation results show also good agreements with the experimental data for calculating several long-lived and stable fission products. However, for the americium and curium actinides, it is difficult to judge the prediction capabilities for these actinides due to the large uncertainties in the ICE-Experimental data. In the MCNPX numerical simulations, a pin cell model is utilized to simulate the fuel lattice of the nuclear power reactor. Temperature dependent libraries based on JEFF3.1 nuclear data files are utilized for the calculations. In addition, temperature dependent libraries based ENDF/B-VII nuclear data files are utilized and the obtained results are very close to the JEFF3.1 results, except for ~10% differences in the prediction of the minor actinide isotopes buildup.

1. Introduction

The MCNPX computer code has the capability to simulate the nuclear fuel irradiation history inside a fission reactor. It is a Monte Carlo transport code with the CINDER90 computer code integrated as a separate burnup/depletion module (Pelowitz, 2008). It eliminates the requirement of linking the transport code and the burnup code for performing a burnup analysis as it was done in other computer code packages (Fensin et al., 2006; Zhong et al., 2009). The numerical simulations with the MCNPX code package should be more accurate than those simply linking the transport code and the CINDER90 code, because the integration allows directly passing one-group reaction rates and 63-groups neutron flux from the MCNPX transport calculation to the depletion calculation for all the isotopes, including each fission product. In previous efforts, the MCNPX burnup capabilities were benchmarked against other numerical code packages, i.e., Monteburns. Our previous work did not track and compare all the isotopes of the fuel cycle with the experimental results (Cao et al., 2010). This paper simulates the Isotope Correlation Experiment (ICE) using the MCNPX code package version 2.7, and directly compares the analytical results with the ICE-Experimental data for all the isotopes (Cao et al., 2010; Koch and Schoof, 1981; Hesse, 1984; Broeders, 1992).

The ICE-Experiment was performed in the nuclear power plant at Obrigheim, Germany in the early 1970s (Koch and Schoof, 1981). The reactor is a Pressurized Water Reactor (PWR) with 3.1% enriched UO2 fuels. Five pre-chosen fuel assemblies were irradiated in the first two fuel cycles, unloaded in the following cycle, and then reloaded and irradiated for one more fuel cycle in the experiment. The normalized power history is shown in Table 1, with a power density of 219.6 W/cm at full power. The average fuel rod temperature was 1028 K, the average clad temperature was 605 K, and the average moderator temperature was 572 K at normal operating conditions. The soluble boron-10 concentration in the moderator was adjusted as listed in Table 1 (Broeders, 1992).

In the ICE-Experiment, after the reactor shut down and 1 year of cooling, 90 of 180 fuel rods corresponding to a geometrical half of each of the five fuel assemblies were chopped and dissolved to form 10 batches of solutions, and were processed by the European Institute of Transuranium Elements (EITU), the IAEA Safeguards Analytical Laboratory, the Institute of Radiochemistry (IRCH) and the Karlsruhe Reprocessing Plant, respectively. The fuel burnup corresponding to each batch was determined by measuring the Nd-148 concentration in the solution. The burnup uncertainties...
are in the range of 3–4% (Koch and Schoof, 1981). The measured atom density of each isotope after the fuel irradiation was determined by averaging the duplicated measurements, and was normalized by the Initial number of Metal Atoms (IMAs) in the UO₂ fuel (Koch and Schoof, 1981). Due to the short half-life of Pu-241 ($T_{1/2} = 14.4$ y), the measured Pu-241 atom density was corrected for β-decay back to the date when the reactor was shut down. Accordingly, the Pu-238 concentrations were also corrected due to the α-decay of Cm-242 with a half-life $T_{1/2} = 162.8$ d. Similar corrections were also made for several fission products in the ICE-data (Koch and Schoof, 1981).

In Germany, simulating the ICE-Experiments was and is the major approach to validate the deterministic code KAPROS/KARBUS (Broeders, 1992; Oberle, 2006; Send, 2005). For the ICE-Experimental simulation, KAPROS applies the first collision probability method to solve the neutron transport equation in a cylindrical geometry (Wigner–Seitz cell). Overall, very good agreements have been achieved with the KAPROS code for simulating the uranium fuel depletion and plutonium buildup. Therefore, in this paper, we will also use the KAPROS numerical results to compare with the Monte Carlo simulations in addition to the experimental measurements. The Monte Carlo code is also expected to be at least as accurate as or more accurate than the KAPROS code, because the MCNPX code introduces less approximation in solving the neutron transport equation compared with the KAPROS deterministic code.

The rest of the paper is organized as follows: In Section 2, the numerical model utilized in the Monte Carlo simulation is described. The calculated atom densities are compared with the ICE-data for each fuel isotope and fission products in Section 3. In addition, the KAPROS numerical results serve also as a reference if large deviations are observed in the comparisons (Send, 2005). The differences between the results obtained with nuclear data libraries based on the use of ENDF/B-VII and JEFF3.1 nuclear data files in the calculations are examined in Section 4. Finally, our conclusions and the possible future work are discussed in Section 5.

2. ICE numerical model

To simulate the ICE-Experiment, a Wigner–Seitz pin cell model is used for the fuel lattice in the power reactor. The fuel pellet has a radius of 0.465 cm, and it is surrounded by a zirconium cladding with outer radius of 0.535 cm. The moderator to the fuel rod (including zirconium cladding) volume ratio is 1.494. Reflecting boundary conditions are applied on all the surfaces of the outermost cell, and also on the top and bottom surfaces in the axial directions of the pin cell. In this numerical model, the boron-10 atom fraction dissolved in the moderator is changed instantly at the beginning of the time steps and remains constant within the time step as described in Table 1. The reactor shut down periods and fuel cool down periods are also explicitly modeled in the simulation as shown in the table.

In the numerical simulations, the statistical errors of the one-group reaction rates calculated by the MCNPX transport code are small and are only ~0.1% at each time step. To compare with the ICE-data, the JEFF3.1 temperature libraries at 1000 K, 600 K, and 600 K are used for the fuel, the clad, and the moderator regions, respectively. To examine the differences due to the use of different nuclear data files, cross section libraries based on ENDF/B-VII were utilized for the simulations. Two different fuel cross section sets with 900 K and 1200 K temperatures were used for the fuel region in separate simulations and the results are compared with the simulation results obtained with cross section libraries based on JEFF3.1 nuclear data files.

In this work, the Monte Carlo simulations are performed with the MCNPX computer code, in which the CINDER90 interface code has been parallelized. To simulate the buildup of the americium and curium isotopes correctly, especially to take account of the Am-242 isomeric and ground states, the MCNPX computer code calculates the production rates $R$ of the Am-242 m isomeric state approximately (Pelowitz et al., 2009):

$$\text{Am-242}^m R = K_{\text{Am-242}}^{\text{MCNPX}}$$

where the 63-group capture cross sections are taken from the CINDER data library, and the multi-group neutron fluxes are tallied from the MCNPX transport calculations. Therefore, the production rate of the Am-242 at ground state is equal to

$$\text{Am-242}^m R = K_{\text{Am-242}}^{\text{MCNPX}} R$$

so that the total capture reaction rate of Am-241 tallied in the MCNPX transport calculation is preserved.

3. Comparison of the Monte Carlo simulations with ICE-Experiments

3.1. The multiplication factor $k_{\text{inf}}$

The infinite multiplication factor $k_{\text{inf}}$ calculated by MCNPX and the KAPROS result as reported (Send, 2005), are compared in Fig. 1. At the beginning of the experiment, the $k_{\text{inf}}$ calculated with MCNPX is 1.21731 with the standard deviation of 49 pcm. The corresponding value from KAPROS calculation is 1.21786, which agrees with the MCNPX calculations within the statistical errors.

Because the KAPROS simulation for reactivity did not simulate the details of the reactor operating record, for example, the reactor shut down and cool down periods, the reactivity transient given by the KAPROS code is different from the MCNPX simulations as shown in Fig. 1. In the MCNPX simulations, the $k_{\text{inf}}$ increases at each time step when the B-10 concentration in the moderator is reduced. In reactor operations before starting a new fuel cycle, the boron-10 concentration was also adjusted back to a high value in order to compensate the inserted reactivity from fresh fuel as shown in Table 1. The high B-10 concentrations in the moderator leads to the deduction of $k_{\text{inf}}$ in the MCNPX simulation, as shown as the sharp decrease at the time of 366 d and 1070 d, respectively. In Fig. 1, the $k_{\text{inf}}$ also slightly increases while the reactor is shut down or the fuel rods left out of the reactor for a period of time because of the decay of the fission products. In KAPROS simulation, the time steps with zero reactor power and the time step of boron increments are combined, so the intermediate $k_{\text{inf}}$ value show-
ing the $k_{\text{eff}}$ increase is not observed in the KAPROS calculation. The differences of the calculated $k_{\text{eff}}$s can be seen in Fig. 1 for the period from 690 to 1070 days. Subsequent KAPROS simulations with a refined irradiation specification also confirm this observation.

The calculated MCNPX $k_{\text{eff}}$ at the end of the experiment is 0.9975. The KAPROS calculation shows a larger reactivity swing as shown in Fig. 1. In particular, the $k_{\text{eff}}$ obtained from KAPROS is 1539 pcm smaller than the MCNPX calculation at the end of the experiment. Despite of the different time steps used in these two simulations, the different calculation methods and nuclear data libraries used to solve the transport equations in these two codes may also explain some of the differences observed in Fig. 1.

### 3.2. The uranium isotopes

In the ICE-Experiment, the burnup of the uranium fuels is determined by the Nd-148 method, which has estimated errors of about 3–4% (Hesse, 1984). Thus, to compare the experiment with the numerical simulations, a 3% error bar is shown for the measured burnup values of each experimental data point in the figures, which display the experimental results.

The comparison of the results in Figs. 2 and 4 shows that the MCNPX code can predict accurately the depletion of the uranium fuels. The MCNPX burnup simulation gives excellent agreement with the ICE-data for depleting the U-238 atoms as shown in Fig. 4, where the maximum experimental error bar is 0.00%. Although the Monte Carlo simulation depleted slightly more of the U-235 atoms compared with the ICE-data, the difference between the analytical results and the experimental measurements is within the experimental error bars, which are all less than 1% except it is ~2% at the burnup point of 26 GWD/t.

The MCNPX results do not agree well with the ICE-Experimental measurements for producing the U-236 atoms. Fig. 3 shows that the simulation underestimates the atom density at each burnup point by about 4%, while the maximum experimental error bar is only about 1.5%. Similar discrepancies were also found in the past comparisons of the ICE-Experiment with other numerical simulations using burnup codes such as KORIGEN (Hesse, 1984), KAPROS (Send, 2005), etc. The half-life of the isotope Pu-240 is about 6561 y, which would only lead to a correction much less 0.1% of the total U-236 production. The U-236 atoms are mainly produced from the U-235 neutron capture reactions during the fuel irradiation. The ENDF/B-VII cross section libraries at 900 K and 1200 K are utilized in the fuel region respectively and the results show exactly the same amount of U-236 production. The U-236 atoms are mainly produced from the U-235 neutron capture reactions during the fuel irradiation.

### Table 1

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<th>Time step</th>
<th>Duration (d)</th>
<th>$B^{10}$ (10$^{-6}$)</th>
<th>Power (%)</th>
<th>Time step</th>
<th>Duration (d)</th>
<th>$B^{10}$ (10$^{-6}$)</th>
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* $\dagger$ 1/barn-cm.
** Normalized by the maximum power density 219.6 W/cm$^3$ [6].
inconsistency and inaccuracy of U-235 capture cross sections in this energy range might be one of the possible factors inducing this discrepancy.

3.3. The plutonium isotopes

The buildup of Pu-239 isotope is calculated as a function of the burnup. The comparison with the experimental data in Fig. 5 shows an excellent agreement. Almost all the calculated values match the measurements within the experimental error bars of 1.5–5.0%.

In the ICE-Experiment, the atom density of each individual plutonium isotope has also been measured. Figs. 6 and 7 compare the experimental data from the MCNPX simulations. The MCNPX calculates the atom densities of Pu-240 and Pu-241 produced due to the fuel irradiation very accurately. The ICE-data experimental error bars in these two figures are all less than 6% at each data point. The sharp decrease of the MCNPX simulation curve at burnup of 21.5 GWD/t in Fig. 7 corresponds to the radioactive decay of Pu-241 during the 1 year cooling period after the fuel irradiated for two fuel cycles. The comparison of Pu-242 atom density as a function of burnup is shown in Fig. 8, which shows that the MCNPX simulation produces slightly less Pu-242 atoms than the experimental data at each burnup point. The same amount of differences was also observed in other FZK simulations of the ICE-data (Koch and Schoof, 1981; Oberle, 2006).

In addition, because the technique used to measure the Pu-238 atom densities involves large uncertainties in the ICE-Experiment (Hesse, 1984), a 10–20% large error bar is placed at each of the ICE-Experimental points in Fig. 9. The MCNPX simulation curve is clearly below the ICE-data points out of the experimental error bars as shown in Fig. 9. Similar discrepancies were observed in the FZK simulations with KAPROS (Send, 2005), and the KORIGEN benchmark simulations (Hesse, 1984).

3.4. The americium and curium isotopes

Due to the limitations on the experimental technique, the ICE-Experiment did not give the associated experimental error bars for the measured americium and curium isotope atom densities. Similar to the measurements of the Pu-238 atom densities, large error bars are expected in the ICE-data for the minor actinides. Nonetheless, Figs. 10–13 indicate how the MCNPX simulation results deviate from the experimental measurements. The increase of Am-241 in the MCNPX simulation curve at burnup of 21.5 GWD/t is due to the Pu-241 β-decay when the fuel rods were cooled outside of the reactor for one fuel cycle. The ICE-Experimental data points for both Am-241 and Am-243 are scattered in a large range, and the MCNPX simulation results differ from them by more than 100% at most of points for Am-241, and by more than 30% for Am-243. For curium isotopes, the discrepancies between the analytical results and the experimental measurements are around 20%, which are much better agreements than for the
The short half-life of Cm-242 led to a decrease of Cm-242 isotope concentration and an increase of Pu-238 isotope concentration at burnup of 21.5 GWd/t, which are accurately simulated by MCNPX as shown in Figs. 9 and 12.

Other numerical simulations with the KORIGEN code (Hesse, 1984) or the KAPROS code (Send, 2005) gave also very similar results to the MCNPX simulations. The calculated Am-243 atom densities by the numerical methods are always larger than the ICE measurements, and the calculated Cm-242 and Cm-244 atom densities are less than the ICE-Experimental values. In spite of the large differences existing in the comparisons, due to the large uncertainties of the experimental data, the accuracy of the numerical simulations for calculating the buildup of americium and curium isotopes cannot be judged based on the ICE-Experimental results.

Additionally, as described in the Section 2, the MCNPX calculation of the Am-242m production rates requires adopting the 63-group cross sections of Am-241(n,γ) in the CINDER data library directly. Another possible way is to calculate the branching ratio of the Am-242 isomeric state in the MCNPX simulations. In this approach, the multi-group cross sections in CINDER are used to estimate the isomer-state branching ratio $Br$:

$$Br = \frac{\sum_{g=1}^{63} C^{CINDEN}_{g} \cdot Am-241 \rightarrow Am-242m}{\sum_{g=1}^{63} C^{CINDEN}_{g} \cdot Am-241 \rightarrow Am-242g + C^{CINDEN}_{g} \cdot Am-241 \rightarrow Am-242m \cdot MCNPX}_{MCNPX}.$$  \hspace{1cm} (3)

The main advantage of using the branching ratio is to reduce the systematic errors in the CINDER multi-group cross sections by calculating the ratio of two reaction rates. The Am-242m and Am-242g production rates can then be determined by the estimated $Br$ and the total capture rates $^{214}AmR$ tallied from the MCNPX transport calculations:

$$^{214}AmR_{Am-242m} = ^{214}AmR \times Br,$$

$$^{214}AmR_{Am-242g} = ^{214}AmR \times (1 - Br).$$  \hspace{1cm} (4)
3.5. The fission products

The isotopic ratios of several fission products, i.e., Kr, Xe, Cs and Nd, have also been measured in the ICE-Experiment. The comparison of the MCNPX simulation results with the experimental measurements is shown in Figs. 14–18. The fission products concentrations have been calculated reasonably accurate by the MCNPX code. Excellent agreements can be found for the xenon isotopes as shown in Fig. 15, for cesium isotopes in Fig. 16, and for neodymium isotope ratios Nd-146/Nd-145 and Nd-145/Nd-148 in Fig. 18. However, the MCNPX under-predicts the Kr-84/Kr-83 atom ratio by about 6%, and the Kr-84/Kr-86 atom ratio by 15%. The discrepancy between the Monte Carlo simulations and the experimental measurements is also more than 20% for the Nd-144/Nd-148 atom ratio, and is about 6% for the Nd-143/Nd-148 atom ratio. Similarly, in Figs. 16 and 17, the MCNPX simulation accurately reflects the isotope concentration change at burnup of 21.5 GWd/t due to the radioactive decay of some isotopes, i.e., Cs-134, Cs-137, Ce-143, Pr-143 and Pr-145.

In the MCNPX burnup calculations, the fission products are represented explicitly in the transport calculation. Their one-group reaction rates are calculated by MCNPX tallies. In KAPROS, the fission products are also represented explicitly in the transport calculation. One-group cross sections are determined by using best estimate of many group spectra. The large discrepancies of the isotopic ratios for xenon and cesium in the KAPROS calculations have been eliminated in the MCNPX calculations. A better agreement has also been found for the Nd-146/Nd-148 atom ratio in the MCNPX calculations. The improvements in the MCNPX simulation can be utilized to analyze the KAPROS results in more details, e.g., checking the fission yields.

4. Burnup simulations with different cross section libraries

The transport cross section library based on ENDF/B-VII was processed with different temperatures, i.e., 293.6 K, 600 K, 900 K, 1200 K and 2500 K. According to Talou et al. (2007), the minor actinide cross sections in the ENDF/B-VII library are greatly improved,
i.e., the values of the Am-241 capture cross sections in the keV energy region and its isomer-to-ground-state branching ratio are increased significantly. Therefore, to investigate the effects of using different libraries, the MCNPX simulations were performed with the ENDF/B-VII library at 900 K, and the simulation results are compared with the simulations done with the JEFF3.1 library at 900 K.

**Fig. 19** shows the multiplication factors calculated by using both libraries. The calculated $k_{inf}$ is always larger with the ENDF/B-VII library than with the JEFF3.1 library, i.e., 328 pcm larger at the beginning of the experiment, and about 320 pcm larger at the end. For uranium isotopes and plutonium isotopes, the numerical results differ by $\sim 2\%$ between the two libraries. Figs. 20–23 compare the numerical simulations for americium and curium isotopes. With the ENDF/B-VII library, the MCNPX gives $\sim 10\%$ more Am-241, $\sim 11\%$ more Am-243, $\sim 8\%$ less of Cm-242, and $\sim 13\%$ more of Cm-244 than with the JEFF3.1 library. A comparison of the Am-241 capture cross sections shows that the first resonance of Am-241 $(n, \gamma)$ at 0.3 eV is about 10\% smaller in the ENDF/B-VII library than in the JEFF3.1 library. Therefore, the MCNPX simulation indicates that the large discrepancies and uncertainties in existing libraries for the minor actinides require further investigations to improve the prediction of the burnup results (Chadwick et al., 2006).

5. Conclusions

The analyses of the ICE-Experimental data show that the MCNPX code can predict accurately the uranium fuel depletion, the plutonium production and the buildup of most of the fission products in a thermal reactor. The simulation results agree with the ICE measurements within the experimental uncertainties. The only exceptions are for U-236 and Pu-238, the largest deviations from the experimental data are $\sim 4\%$, and 20\%, respectively.
It should be noted that the experimental data have very large error bars for the Pu-238 isotope. The Monte Carlo simulations also produced accurate predictions for calculating the atom ratios of most of the long-lived or stable fission products, except that there is 6% difference for the Kr-84/Kr-83 isotopic ratio, 15% for Kr-84/Kr-86, 20% Nd-144/Nd-148 and 6% for Nd-143/Nd-148 from the ICExperiments. The atom densities of americium isotopes calculated by MCNPX departed significantly from the scattered experimental measurements. The accuracy of the calculated atom densities of the americium and curium isotopes cannot be judged, due to the unknown error bars in the ICE-Experimental measurements and the large uncertainties in the current cross sections of minor actinides. 

In general, the MCNPX burnup simulations of the JEFF3.1 libraries for the ICE-Experiment are satisfactory. The comparison of the numerical results based on ENDF/B-VII and JEFF3.1 nuclear data files shows negligible differences in predicting the reactivity swing, the amount of uranium depletion and the plutonium inventories. However, about 10% differences with the two libraries are also observed in calculating the minor actinide atom densities. In ENDF/B-VII nuclear data files, the neutron cross section data for minor actinides have been greatly improved (Talou et al., 2007). Most of the adjustments or corrections are made in the epithermal or fast neutron energy regions. Comparing the Am-241 neutron capture cross sections from ENDF/B-VII, JEFF3.1, and JENDL-3.3 nuclear data files shows significant differences in the low energy resolved resonance region. Nakamura et al., showed that the Am-241 thermal-neutron capture cross sections are significantly overestimated due to an improper cutoff energy imposed in the calculations of JENDL-3.3 nuclear data files (Nakamura et al., 2007). Further evaluations of the current nuclear data files are required to obtain accurate burnup analyses for calculating the buildup and destruction of minor actinides in fission reactors. 

In the ICE-Experiment, the fuel burnup of each fuel sample is limited to 30 GWd/t and the experiment was also performed in a thermal nuclear power plant. Therefore, other experiments are required for deep-burnup benchmarks, in which more plutonium isotopes, minor actinides, and fission products will be produced with different neutron energy spectra.

References

Los Alamos National Laboratory. 