IAEA CRP ADS Research
Subtask: Reactor Fuel Burn-up Qualification / Validation
Application: Isotope Correlation Experiment in NPP Obrigheim

(Updated version April 14, 2010)

C.H.M. Broeders, Y. Cao , Y. Gohar, F. Alvarez-Velarde

1. Introduction

In the definition phase of the IAEA CRP on ADS research it was agreed that in connection with incineration of wastes from the nuclear fuel cycle it is of high importance to have good prediction tools for the characterization of the contents of discharged fuel from nuclear reactors. It was recognized that only few experimental information about this issue is available in open literature. Most of the extensive gathered data base is proprietary and not free accessible. At Research Center Karlsruhe (FZK) a dedicated experiment has been utilized for a long period: the Isotope Correlation Experiment ICE.

2. The ICE-Experiment

The ICE-Experiment was performed in the nuclear power plant at Obrigheim, Germany in the early 1970s. The reactor is a Pressurized Water Reactor (PWR) with 3.1% enriched UO_2 fuels. In this experiment five pre-selected 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. The normalized power history is well documented, as well as the mean soluble boron-10 concentration in the moderator during each step. The power density was 219.6 W/cm at full power. The average fuel rod temperature was 1028K, and the average moderator temperature was 572K at normal operating conditions. After the reactor shut down and one year of cooling time, the five fuel assemblies were chopped into halves, dissolved to form 10 batches of solutions, and were processed by the European Institute of Trans-uranium Elements (EITU), the IAEA Safeguards Analytical Laboratory, the Institute of Radio chemistry (IRCH) and the Karlsruhe Reprocessing Plant, respectively. The fuel burn-up corresponding to each batch was determined by measuring the Nd-148 concentration in the solution. The burn-up uncertainties are in the range of 3 to 4%. 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 UO_2 fuel. 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.

3. Some previous utilization of ICE

In the noncommercial research groups on the nuclear fuel cycle in Germany, analysis of the ICE-Experiments was and is a major approach for the validation of simulation tools for nuclear fuel irradiation. Some examples in the IAEA CRP access area are:

- Validation work at FZK for KORIGEN; KFK3014 (1983).
- Validation work at GRS for OREST; ORNL/TR-88/20 (1984).
- Validation work at FZK for TLLWR; KFK5072 (1992).
- Re-validation work at FZK for KAPROS/KARBUS; Dipl-Thes. (2005)
- Validation work at FZK for SCALE5/TRITON; PHYSOR-2006.

4. Documents for the benchmark

For the IAEA CRP ADS ICE benchmark the following resources were provided:

- L. Koch, and S. Schoof, "The Isotope Correlation Experiment ICE," ESARDA 2/81, EUR7766 EN, KFK3337 (1981).
- U. Hesse, "Verification of the OREST (HMER-ORIGEN) Fuel Assemblies 168, 170, 171 and 176 from the Obrigheim Reactor," ORNL/TR-88/20 (1984).
- C.H.M. Broeders, "Entwicklungsarbeiten für die neutronenphysikalische Auslegung von fortschrittlichen Druckwassereaktoren (FDWR) mit kompakten Dreiecksgittern in hexagonalen Brennelementen," KFK5072 (1992).
- Ph. Oberle, C. H. M. Broeders, and R. Dagan, "Comparison of PWR - Burn-up Calculations with SCALE5.0/TRITON other Burn-up Codes and Experimental Results," PHYSOR-2006, Advances in Nuclear Analysis and Simulation, Vancouver, BC, Canada (2006).
- L. Send, "Investigations for Fuel Recycling in LWRs," Diploma thesis, FZK, Karlsruhe, Germany (2005).

5. Participants

The following IAEA CRP ADS participants prepared simulations of the ICE benchmark:

- **FZK**

  At FZK currently the ICE benchmark is part of the provided test examples in the modular code system KAPROS/KANEXT for nuclear reactor simulations, mainly based on multi-group deterministic solutions. In this way it is also support for a kind of quality assurance for new versions of code and libraries. Solutions based on several 69 and 350 group libraries are available. For the SCALE5 validation also a Monte Carlo solution with MONTEBURNS was considered. The deterministic multi-group solutions are based on the first collision probability method to solve the neutron transport equation in a cylindrical geometry (Wigner-Seitz cell). In KARBUS $\approx$80 fission products are represented explicitly in the transport calculation. The full KORIGEN data base is utilized for activation and decay. As far as available on the multi-group libraries ($\approx$800), the required one-group cross sections are determined by using best estimate many groups spectra, to replace the KORIGEN data. Typical running time for a full simulation is less than half an hour on a modern LINUX PC for a 350 group solution. More details of the KARBUS simulations may be found in the diploma thesis of L. Send.

- **ANL**

At ANL strong efforts are ongoing to validate the burn-up capabilities of the Monte Carlo code MCNPX2.7. MCNPX utilizes a Monte Carlo solution to solve the neutron transport equation and has the CINDER90 computer code integrated as a separate burn-up/depletion module. Directly passing of one-group reaction rates and 63-groups neutron fluxes from the MCNPX transport calculation to the depletion calculation for all the isotopes, including each fission product, is applied. In addition to the code-to-code comparisons, the ICE is considered as valuable verification by experimental results. Very detailed solutions are prepared, based on the same geometry as applied at FZK, and using several nuclear data libraries. Two publications are devoted to these ICE simulations:

  - Y. Cao, Y. Gohar and C.H.M. Broeders, “MCNPX Burn-up Simulation of the Isotope Correlation Experiment,” *PHYSOR* 2010-Advances *in Reactor Physics to Power the Nuclear Renaissance*, Pittsburg, Pennsylvania, USA, May 9-14 (2010)
  - Y. Cao, Y. Gohar, and C.H.M. Broeders, “MCNPX Monte Carlo Burn-up Simulations of the Isotope Correlation Experiments in the NPP Obrigheim”, to be published

- **CIEMAT**

At CIEMAT since several years a code package for the simulation of the burn-up of a nuclear reactor is being developed, coupling Monte Carlo and deterministic simulations: the current version is EVOLCODE2. This benchmark is an excellent opportunity to validate the qualification of EVOLCODE2. For this exercise MCNPX2.5.0 for neutron transport and ORIGEN2 for depletion calculations have been used. For simulating the ICE experiment a quarter of a fuel assembly was modeled in a first step with very detailed geometry specification, requiring hence large computing resources. 30000 isolethargy energy bins were used to simulate the neutron flux for the calculation of the one-group cross sections of every geometry cell. A preliminary solution could be provided in a late stage of the IAEA CRP ADS ICE benchmark activities. However, the first comparisons of the results with ANL and FZK data indicated problems with this solution. A careful assessment of the investigations showed that library problems had introduced unacceptable deviations. For this reason it was decided to provide a solution for the same geometrical data as in the ANL and FZK solutions. In a next step it is planned to perform sub-assembly analysis for comparisons. The CIEMAT results presented here are based on pin cell burn-up.

6. Selected results

For the present document some typical results of the three solutions are compared in the following. In figures 1-3 the additionally proposed parameter K-infinity, absolute flux and applied boron content are shown for the three solutions. Now the agreement of the results of ANL, CIEMAT and FZK, is very satisfactory. The figures 4-15 show the comparison for heavy metal isotopes, with varying agreements. Error bars are taken from the diploma thesis L. Send. The agreement between ANL, CIEMAT and FZK solutions is good in most cases. For the minor actinides isotopes of americium and curium the agreement is satisfactory with exception of Am-241. Here the experiment evaluation is difficult due to the Am-241 production by Pu-241 decay with relatively short half life. In figure 16 and 17 examples of some reaction rate ratios for fission product isotopes are shown. Both for the ratio Nd-146/Nd-148 and for the ratio Xe-132/Xe-131 the agreement between ANL and CIEMAT solutions and experiment is better than for the FZK result. Analysis of this observation probably will lead to improvements of the FZK simulation tools. Indeed, the fission product yields on the KARBUS libraries are not consistent with JEFF3.1 and should be updated.

7. Summary/outlook

The present document gives a first overview of the activities in the framework of the IAEA CRP ADS related to the prediction of the composition of discharged fuel from nuclear reactor systems. The ICE experiment in the Obrigheim NPP in Germany in the early seventies is a quite well in open literature documented project. Three CRP participants prepared solutions with detailed final results for a representative unit cell model for the experiment, as defined in early analysis of ICE at FZK. The agreement between ANL, CIEMAT and FZK solutions is good in most cases. For some experimental results for fission product isotope ratios the ANL and CIEMAT solutions are clearly better than FZK ones. Here the FZK tools will be checked in the near future.

It may be concluded that this part of the IAEA CRP ADS work was beneficial for the participants. A common publication as outcome of the CRP, preferably in a journal, may be envisaged. Based on the analysis of the results and on discussions between the participants the following issues for further analysis could be considered:

- Impact of the model simplifications, fuel assembly vs. unit cell. Handling of the fuel assembly materials, not belonging to the basic fuel cells in cell calculations. Handling of core materials not belonging to regular fuel assembles
- Handling of the boron-10 in the coolant. Some analysis concerning time dependent boron-10 vs. use of a mean value during a fuel cycle is available in the diploma thesis of L. Send. The current simulation tool

KANEXT/KARBUS at FZK offers powerful options for fast running analysis of this problem.

Finally it should be noted that the ICE project only gives experimental information for LWR burn-up up till 30 GWD/THM. Modern LWR are now operated to achieve considerable higher burn-ups up till 50-60 GWD/THM. Extension of the burn-up range to higher burn-ups in an open literature experimental data base for LWR is very desirable to support qualified judgments by non-commercial parties in this sensible area of the nuclear fuel cycle.

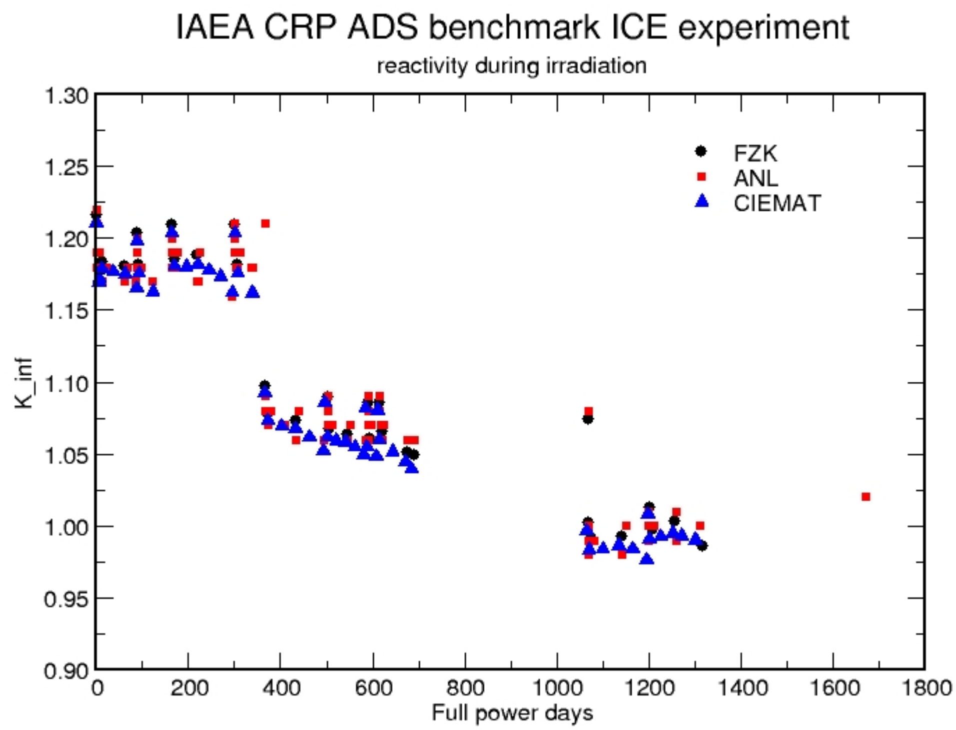

Figure 1: K-infinity during ICE irradiation

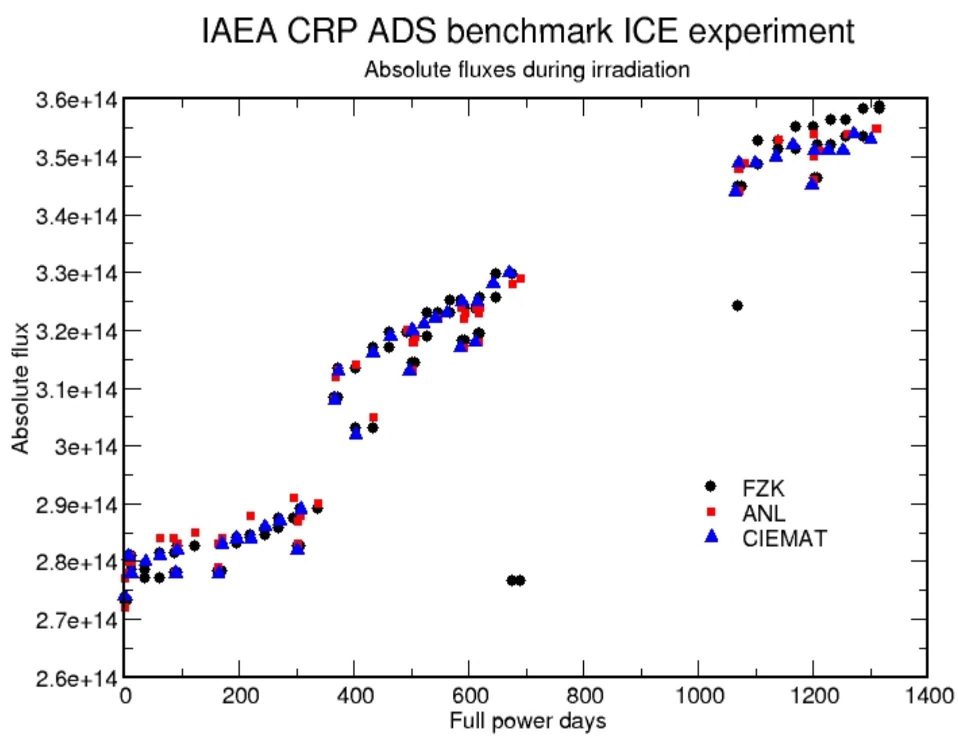

Figure 2: Absolute fluxes during ICE irradiation

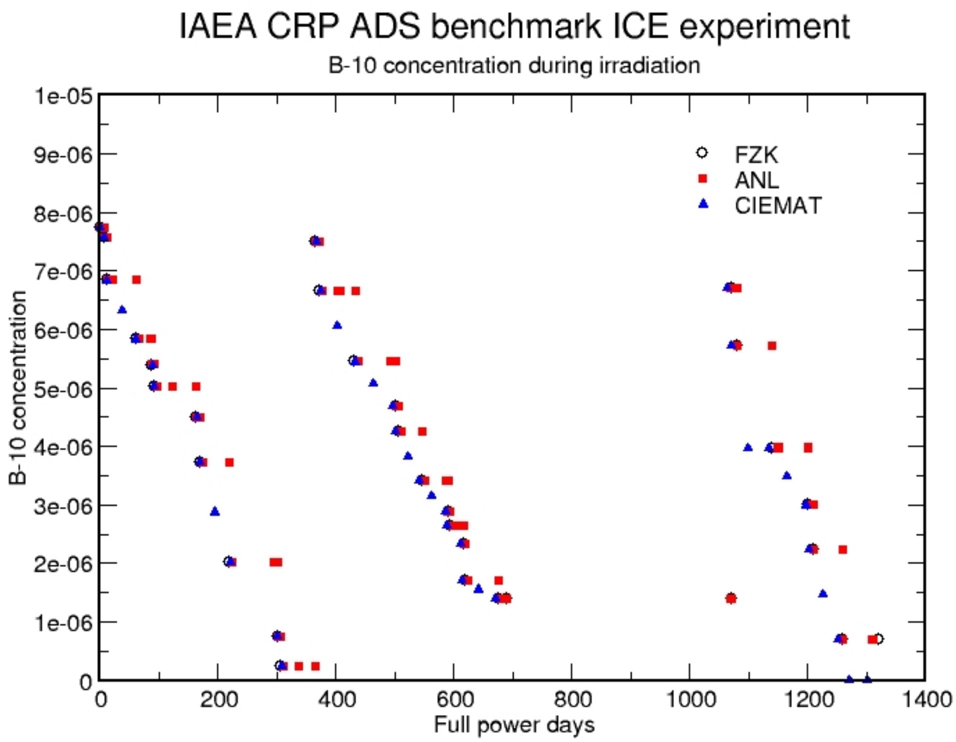

Figure 3: Applied boron-10 concentration during ICE irradiation

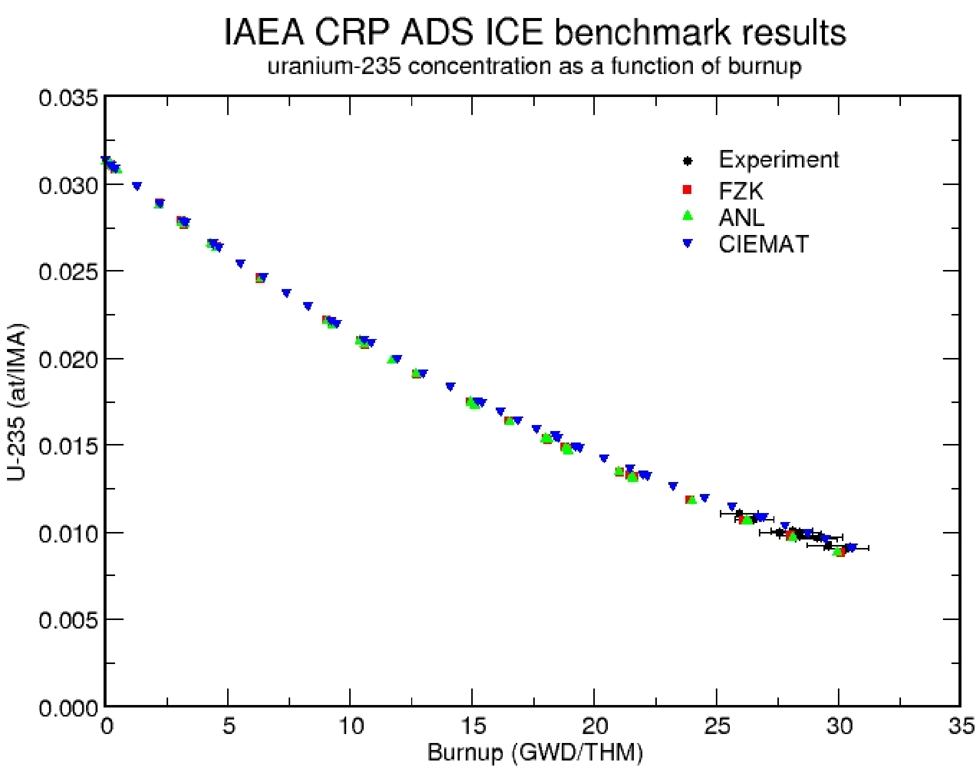

Figure 4: U-235 concentration during ICE irradiation

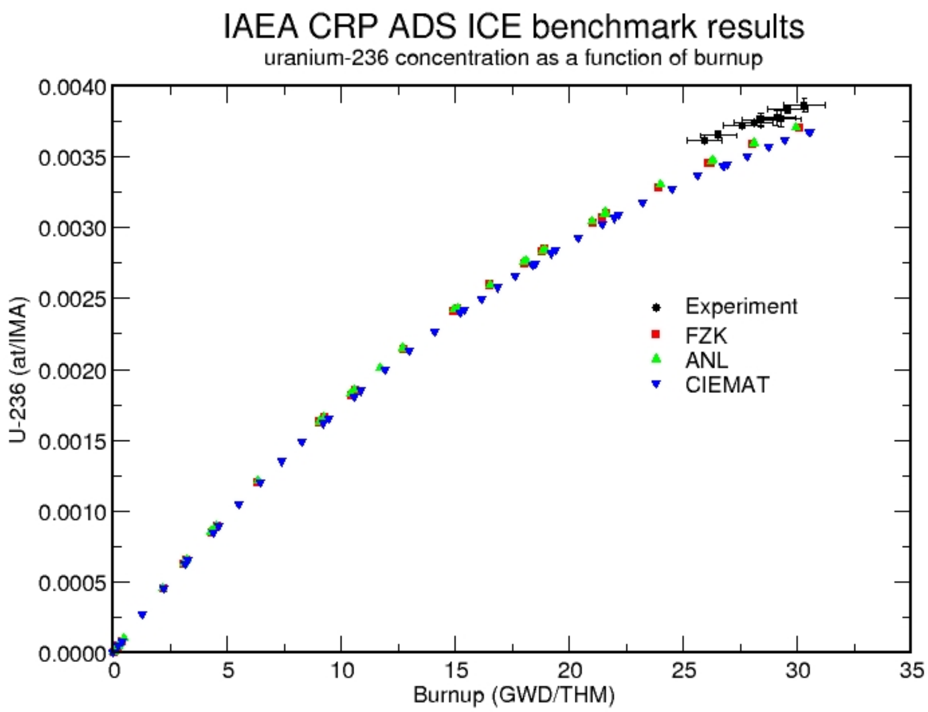

Figure 5: U-236 concentration during ICE irradiation

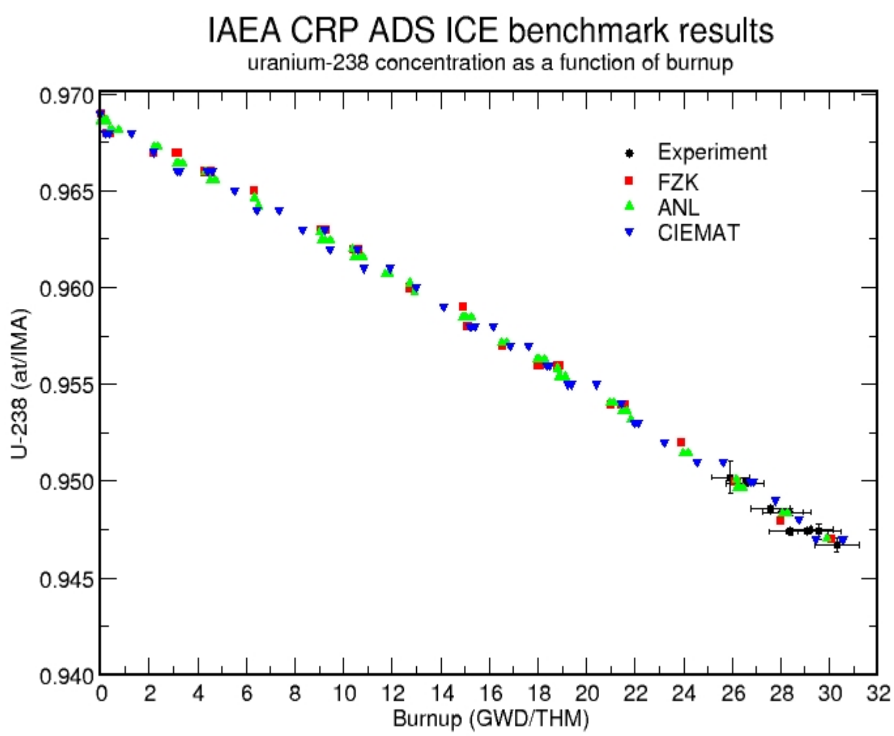

Figure 6: U-238 concentration during ICE irradiation

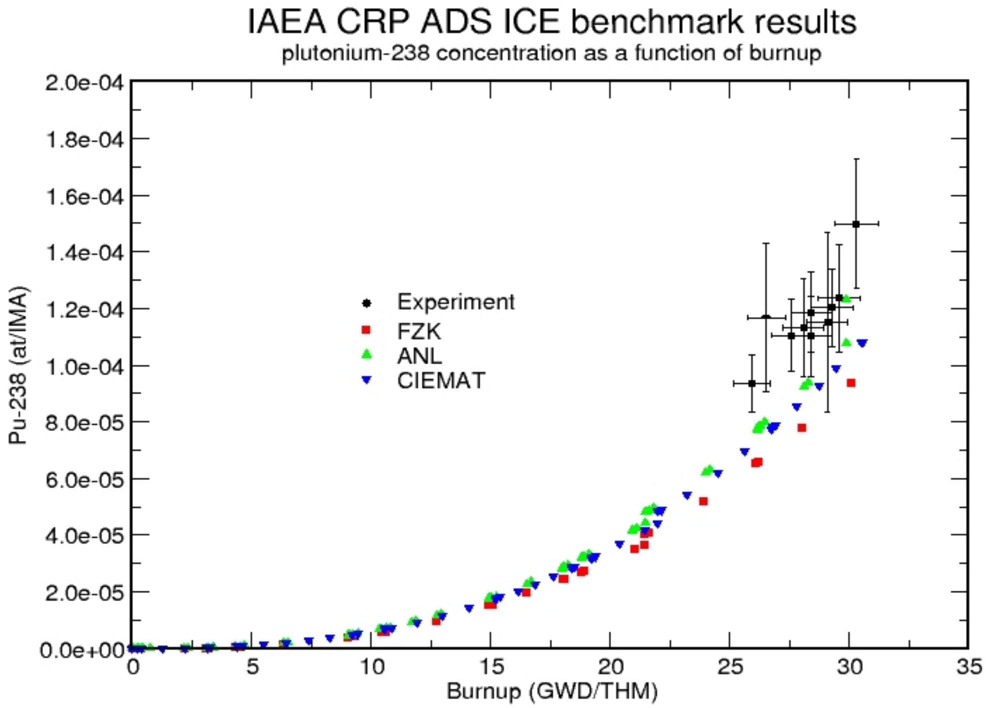

Figure 7: Pu-238 concentration during ICE irradiation

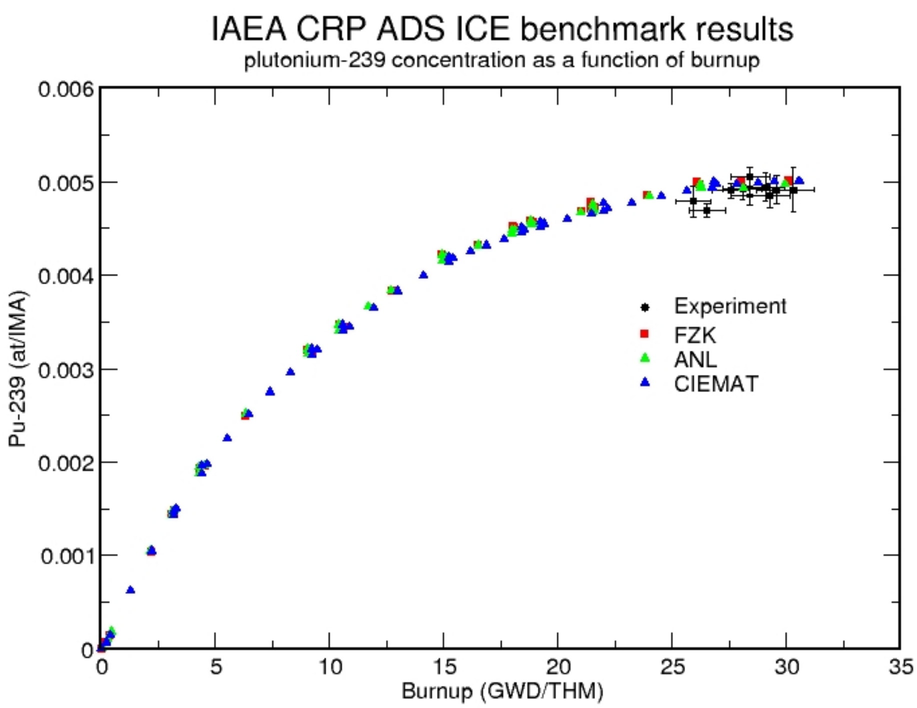

Figure 8: Pu-239 concentration during ICE irradiation

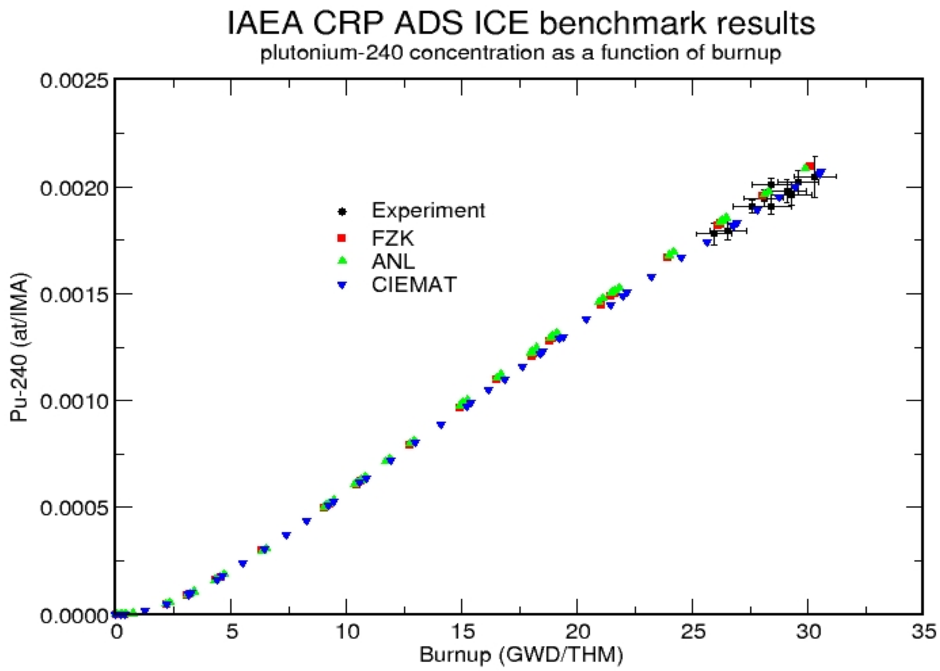

Figure 9: Pu-240 concentration during ICE irradiation

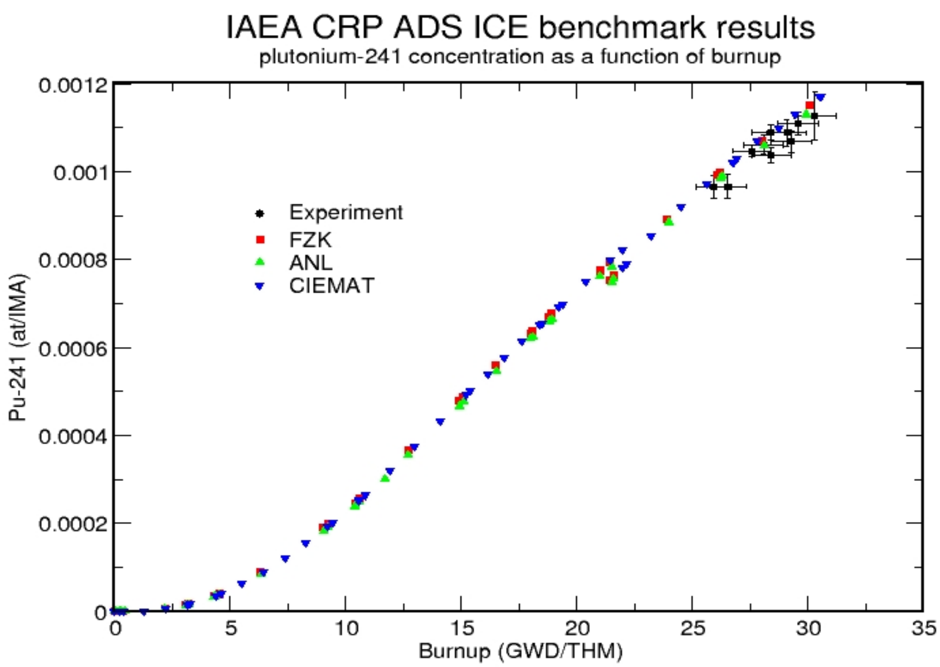

Figure 10: Pu-241 concentration during ICE irradiation

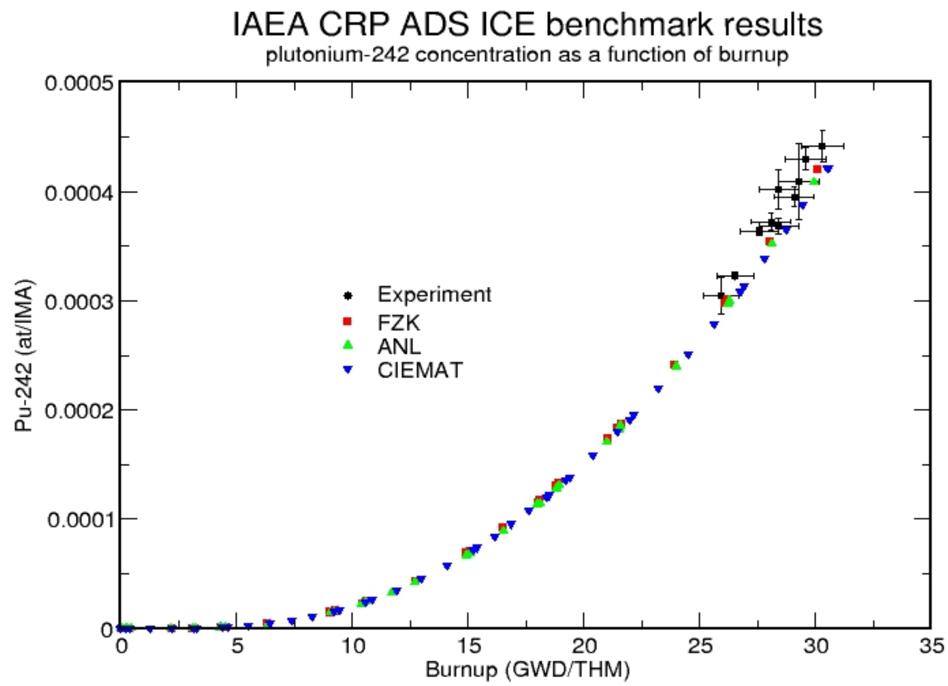

Figure 11: Pu-242 concentration during ICE irradiation

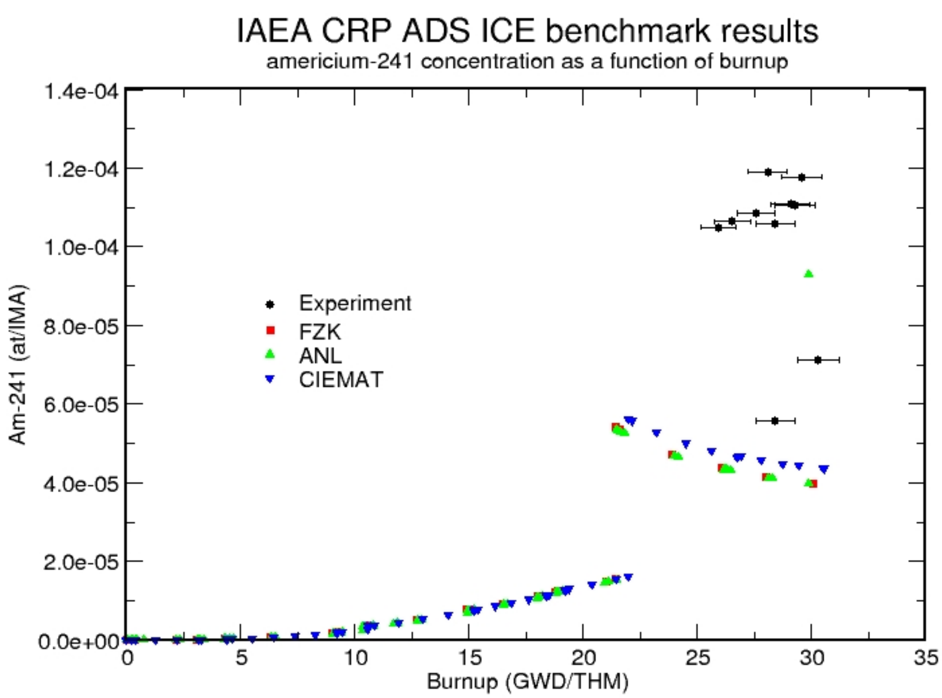

Figure 12: Am-241 concentration during ICE irradiation

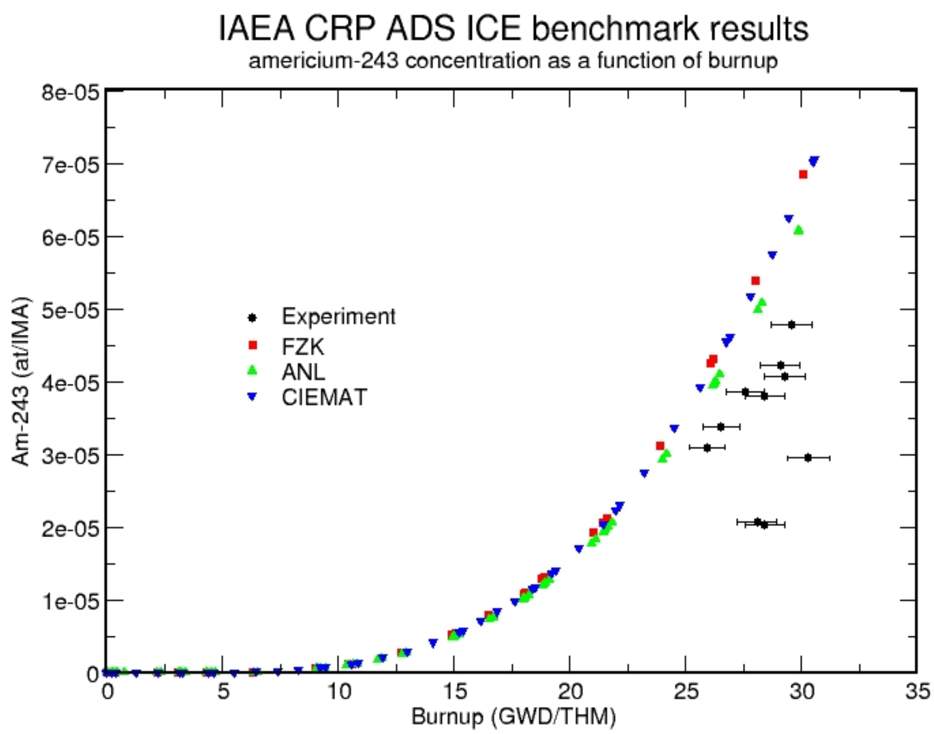

Figure 13: Am–243 concentration during ICE irradiation

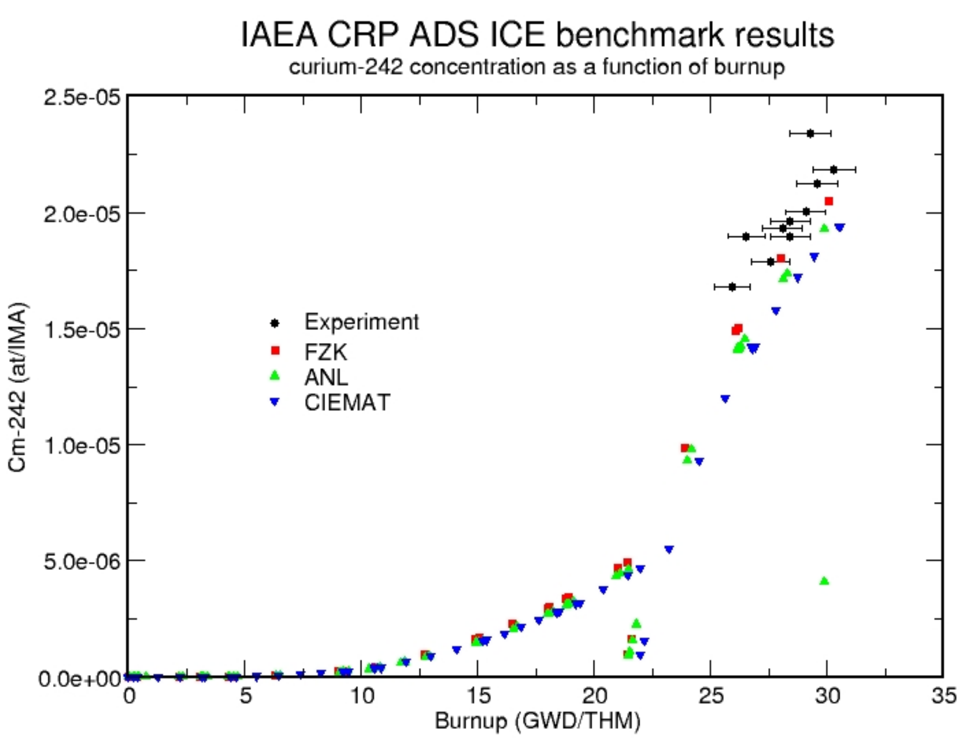

Figure 14: Cm–242 concentration during ICE irradiation

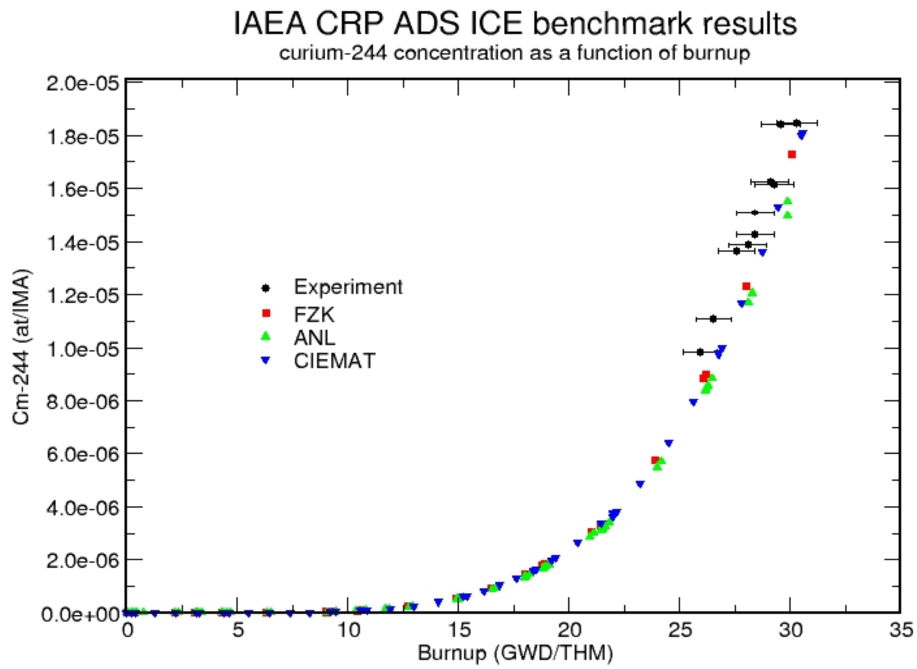

Figure 15: Cm-244 concentration during ICE irradiation

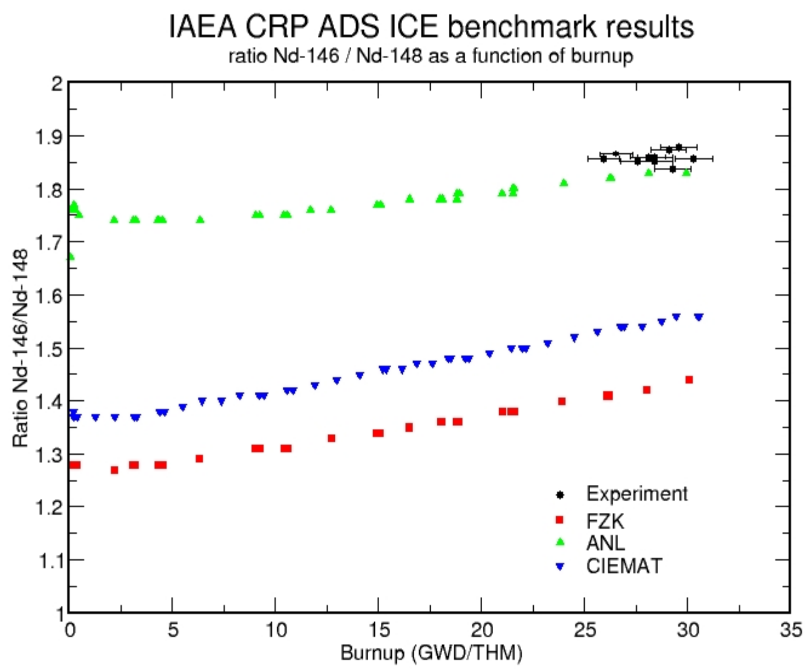

Figure 16: Ratio Nd-146/Nd-148 concentration during ICE irradiation

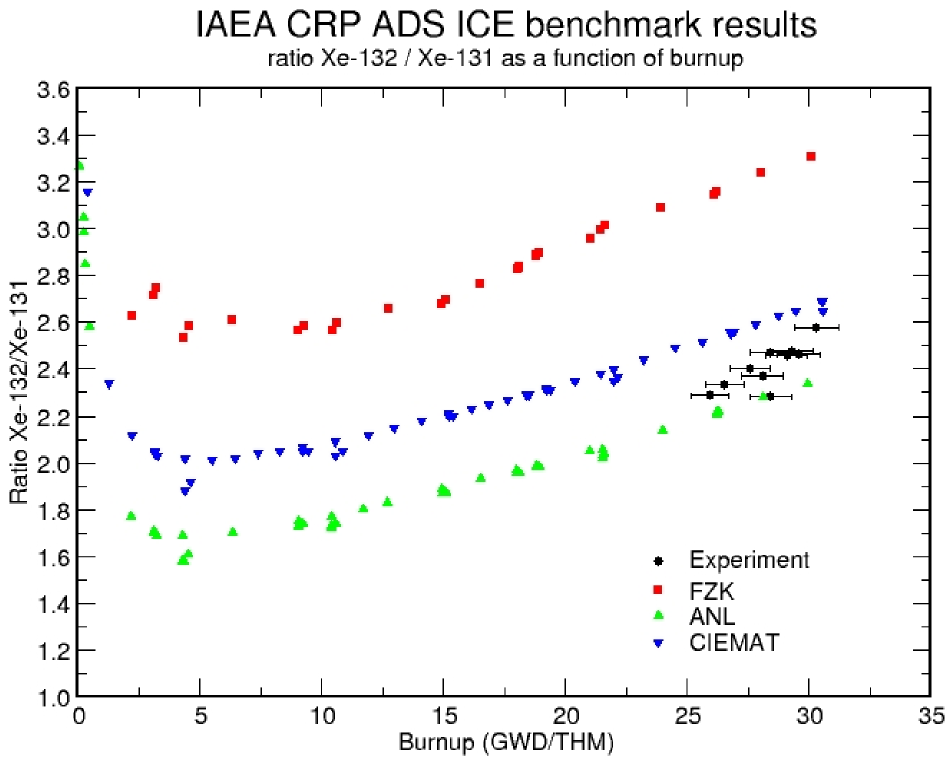

Figure 17: Ratio Xe-132/Xe-131 concentration during ICE irradiation