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# Spent Nuclear Fuel Assay Data for Isotopic Validation

State-of-the-art Report

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## *State-of-the-art Report*

Nuclear Science Committee
Working Party on Nuclear Criticality Safety (WPNCS)
Expert Group on Assay Data of Spent Nuclear Fuel (EGADSNF)

**NUCLEAR ENERGY AGENCY**
**Organisation for Economic Co-operation and Development**

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# Foreword

Management of nuclear fuel is a key issue for many NEA member countries with operating or decommissioned nuclear power plants. In the area of nuclear criticality safety, burn-up credit is an accepted practice for more efficient and cost effective management of irradiated fuel which takes into account a decrease in the reactivity of spent fuel due to the burn-up. In many countries burn-up credit has been implemented for spent fuel transportation, dry storage, pool storage, and in areas of spent fuel reprocessing. In many other countries the technical basis for burn-up credit is being actively developed and advanced for use in spent fuel management safety and licensing. The OECD/NEA Nuclear Science Committee (NSC) has been active in the area of criticality safety of irradiated nuclear fuel since 1980. In 1991, the OECD/NEA NSC formed the working group that in 1997 became the Working Party on Nuclear Criticality Safety (WPNCS), and the Expert Group on Burn-up Credit Criticality (EGBUC) was formed to co-ordinate burn-up credit technical activities and benchmarks of the criticality safety for irradiated nuclear fuel.

Within the EGBUC, the importance of experimental assay data for irradiated nuclear fuel compositions to support validation of computer codes and nuclear data libraries used to calculate isotopic compositions used in burn-up credit has been recognised and discussed. In the EGBUC and WPNCS meetings in 2006 at Aix-en-Provence, it was decided that an effort dedicated specially to the collection and analyses of nuclear fuel assay data should be initiated, and the new Expert Group on Assay Data of Spent Nuclear Fuel (EGADSNF) was established. Because of the importance of assay data to a broad range of nuclear safety topics, the new Expert Group consists not only of members from WPNCS but also members from the standing technical Committee on the Safety of Nuclear Installations (CSNI) and the working group for the Integration Group for the Safety Case (IGSC) of the Radioactive Waste Management Committee.

This report discusses the technical activities of the EGADSNF and the associated efforts of many NEA member countries to compile and document a comprehensive database of assay data to support applications related to nuclear energy and irradiated fuel safety and management. The new contributions of assay data are reviewed, with a summary of the existing database, experimental radiochemical assay measurement methods and best practices, and an uncertainty analysis. Based on the review of new assay data compiled by NEA member countries, progress to update the OECD/NEA web-based nuclear fuel nuclide composition database SFCOMPO is discussed.

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| | |
|---|---|
| AEE | Atomic Energy Establishment (Winfrith Newburgh, UK) |
| AERE | Atomic Energy Research Establishment (Harwell, UK) |
| AGR | Advanced gas-cooled reactor |
| ANL | Argonne National Laboratory (USA) |
| AREVA | French multinational industrial conglomerate known mainly for nuclear power; CEA is the major shareholder, followed by Siemens |
| ARIANE | Actinide Research in a Nuclear Element (Belgian programme) |
| ATM | Approved Testing Material (OCRWM programme) |
| BMU | Bundesumweltministerium (Ministry for the Environment, Germany) |
| BN | Belgonucléaire (Belgium) |
| BUC | Burn-up credit |
| BWR | Boiling water reactor |
| CAGR | Civil advanced gas-cooled reactor |
| CANDU | Canada deuterium uranium reactor |
| CEA | Commissariat à l'énergie atomique et aux énergies alternatives (Atomic and Alternative Energy Commission, France) |
| CSNI | Committee on the Safety of Nuclear Installations |
| DC | Direct current |
| EGADSNF | Expert Group on Assay Data of Spent Nuclear Fuel |
| EGBUC | Expert Group on Burn-up Credit Criticality |
| EOL | End of life |
| EU | European Union |
| FIMA | Fissions per initial metal atom |
| FP | Fission product |
| FZK | Forschungszentrum Karlsruhe (Karlsruhe Research Center, Germany) |
| GE | General Electric |
| GRS | Gesellschaft für Anlagen- und Reaktorsicherheit (German scientific-technical expert organisation for nuclear safety and waste management) |
| HLW | High-level waste |
| HM | Heavy metal |

| | |
|---|---|
| HPGe | High purity germanium (detector) |
| HPLC | High performance liquid chromatography |
| IAEA | International Atomic Energy Agency |
| ICP | Inductively coupled plasma |
| ICPMS | Inductively coupled plasma mass spectrometer/spectrometry |
| ICSBEP | International Criticality Safety Benchmark Evaluation Project |
| ID | Isotope dilution |
| IDA | Isotope dilution analysis |
| ID-MS | Isotope dilution mass spectrometry |
| ID-$\alpha$-spec | Isotope dilution alpha spectrometry |
| IGSC | Integration Group for the Safety Case of Radioactive Waste Repositories |
| INRNE | Institute for Nuclear Research and Nuclear Energy (Bulgaria) |
| IRC | Institute of Radiochemistry (Germany) |
| IRPhEP | International Reactor Physics Experiments Evaluation Project |
| ISO/IEC | International Organisation for Standardisation/International Electrotechnical Commission |
| ISTC | International Science and Technology Center |
| ITU | Institute for Transuranium Elements (Germany) |
| JAEA | Japan Atomic Energy Agency |
| JAERI | Japan Atomic Energy Research Institute |
| JAPC | Japan Atomic Power Company |
| JAVYS | Jadrova a Vyradovacia Spolocnost, a.s. (Nuclear and Decommissioning Company, plc, Slovak Republic) |
| JNC | Japan Nuclear Cycle Development Institute (now Japan Atomic Energy Agency) |
| JNES | Japan Nuclear Energy Safety Organization |
| KFKI AERI | Atomic Energy Research Institute (Hungary) |
| KRI | Khlopin Radium Institute (Russia) |
| LANL | Los Alamos National Laboratory (USA) |
| LECA | Laboratoire d'examens des combustibles actifs (a hot laboratory for the study of irradiated fuel, France) |
| LSC | Liquid scintillation counting |
| LWR | Light water reactor |
| MALIBU | Radiochemical Analysis of MOX and UOX LWR Fuels Irradiated to High Burn-up (proprietary programme) |
| MB | Mass balance |
| MC-ICPMS | Multi-collector inductively coupled plasma mass spectrometry |
| MOX | Mixed-oxide |

| | |
|---|---|
| MS | Mass spectrometry |
| MTHM | Metric tonne of heavy metal |
| Nagra | Nationale Genossenschaft für die Lagerung Radioaktiver Abfälle (National Co-operative for the Disposal of Radioactive Waste, Switzerland) |
| NEA | Nuclear Energy Agency |
| NFD | Nuclear Fuel Development (Japan) |
| NIST | National Institute of Standards and Technology (USA) |
| NRG | Nuclear Research and Consultancy Group (Netherlands) |
| NRI | Nuclear Research Institute Rez (Czech Republic) |
| NSC | Nuclear Science Committee |
| NUMO | Nuclear Waste Management Organization (Japan) |
| NUPEC | Nuclear Power Engineering Corporation (Japan) |
| NWMO | Nuclear Waste Management Organization (Canada) |
| OCRWM | Office of Civilian Radioactive Waste Management (of US DOE) |
| OECD | Organisation for Economic Co-operation and Development |
| ONDRAF/NIRAS | Agency for Management of Radioactive Waste and Enriched Fissile Materials (Belgium) |
| ORNL | Oak Ridge National Laboratory (USA) |
| PFR | Prototype Fast Reactor (Japan) |
| PIE | Post-irradiation examination |
| PIPS | Passivated implanted planar silicon (detector) |
| PNNL | Pacific Northwest National Laboratory (USA) |
| Posiva | Radioactive waste manager (Finland) |
| PSI | Paul Scherrer Institute (Switzerland) |
| PTB | Physikalisch-Technische Bundesanstalt (German national metrology institute) |
| PWR | Pressurised water reactor |
| Q-ICPMS | Quadrupole inductively coupled plasma mass spectrometry |
| RAWRA | Radioactive Waste Repository Authority (Czech Republic) |
| RBMK | Reaktor Bolshoy Moschnosti Kanalniy (Soviet Union-designed high-power channel-type reactor) |
| REBUS | Reactivity Tests for a Direct Evaluation of the Burn-up Credit on Selected Irradiated LWR Fuel Bundles (Belgian programme) |
| RF | Radio frequency |
| RIAR | Research Institute of Atomic Reactors (Russia) |
| SCK•CEN | Studiecentrum voor Kernenergie-Centre d'étude de l'Energie Nucléaire (nuclear research centre in Mol, Belgium) |
| SEM | Secondary electron multiplier |

| | |
|---|---|
| SFCOMPO | OECD/NEA web-based spent nuclear fuel isotopic database |
| SF-ICPMS | Sector field inductively coupled plasma mass spectrometry |
| SKB | Svensk Kärnbränslehantering a.b. (Swedish radioactive waste management company) |
| TEPCO | Tokyo Electric Power Company (Japan) |
| TIMS | Thermal ionisation mass spectrometry |
| TMI-1 | Three Mile Island-1 |
| TRU | Transuranic |
| UJD SR | Nuclear Regulatory Authority of the Slovak Republic |
| UK | United Kingdom |
| UOX | Uranium oxide |
| URE | Uranium de retraitement enrichi (re-enriched uranium from reprocessed uranium) |
| URT | Uranium de retraitement (uranium from reprocessed uranium) |
| US DOE | United States Department of Energy |
| US | United States |
| USA | United States of America |
| UV-VIS | Ultraviolet-visible |
| VENUS | Experimental low-powered reactor at SCK•CEN |
| VGB | Association of Major Power Utilities (Germany) |
| VUJE | VUJE, A.S., nuclear company in the Slovak Republic |
| VVER | Vodo-Vodyanoi Energetichesky Reactor (water-water energetic reactor, a Russian type of pressurised water reactor) |
| WAK | Karlsruhe Reprocessing Plant (Germany) |
| WPNCS | Working Party on Nuclear Criticality Safety |
| WPPR | Working Party on the Physics of Plutonium Fuels and Innovative Fuel Cycles (now WPRS) |
| WPRS | Working Party on the Scientific Issues of Reactor Systems |
| YMP | Yucca Mountain Project (project of OCRWM) |

# Chapter 1: Introduction

Management of spent fuel from commercial nuclear reactors is a key issue for many NEA member countries. As interim storage facilities in many countries reach their design capacities, the need to optimise spent fuel storage management is becoming an increasingly important issue to managing fuel cycle costs while reducing associated risks. In nuclear criticality safety studies involving spent fuel, burn-up credit is being pursued and has been implemented in many countries as a means of more accurately and realistically determining the system reactivity by taking into account a decrease in the reactivity of spent fuel during irradiation. Implementation of burn-up credit has gained in world-wide interest during the last 20 years and it represents one of the most technically challenging issues in nuclear criticality safety. To address these challenges and help co-ordinate activities in NEA member countries, the Working Party on Nuclear Criticality Safety (WPNCS) of the OECD/NEA Nuclear Science Committee (NSC) has organised the Expert Group on Burn-up Credit Criticality (EGBUC). The decision of many countries to advance burn-up credit as part of their criticality safety licensing strategy has heightened interest in measurement data needed to validate code calculations for a burn-up credit methodology.

The continuous development of computational methodologies for nuclear reactor fuel analysis and revision of nuclear data has improved the accuracy of neutronics and burn-up calculations. Numerous numerical benchmarks have been undertaken by the OECD/NEA/NSC that involve comparing different code results for reference calculations as a form of code verification and as a means to evaluate code performance relative to other codes and data used by industry and research. The ability to accurately calculate nuclide inventories for spent fuel is vital to many areas of nuclear plant operations including spent fuel storage and transportation, reprocessing, conditioning, disposal, emissions reporting, etc. For safety evaluations of spent nuclear fuel systems, the nuclide compositions and resulting radiation sources used in safety and design assessments often must be calculated using burn-up codes. The accuracy of these models is, therefore, of considerable importance to establishing the safety basis. Comparison of nuclide concentrations calculated by the codes with corresponding experimental assay data for spent fuel is a fundamental part of code validation.

To obtain high accuracy measurements of isotopic compositions, spent fuel samples are destructively examined by means of radiochemical analysis methods that involve a series of complex analytical methods for sample preparation, may involve chemical separations of the various elements, and finally isotopic and elemental measurements. The importance of having available measured nuclide assay data for spent nuclear fuel has been discussed and is recognised by the EGBUC. An important contribution of the EGBUC in this area is an electronic database of spent fuel isotopic compositions (SFCOMPO), originally developed by the Japan Atomic Energy Research Institute (JAERI). After its release, SFCOMPO was transferred to the NEA for further maintenance, and the NEA Data Bank currently operates the database through its website. After several years of experience maintaining SFCOMPO, the members of EGBUC recognised the need to expand the database to include more recent measurements, and the need to perform this activity by a dedicated expert group.

The activity towards establishing a new expert group focused on assay data was initiated during the thirteenth meeting of the EGBUC which was held – concurrently with WPNCS – in Prage, Czech Republic on 2 September 2004. To further consolidate interest in this area, an

OECD/NEA workshop on "The Need for Post-irradiation Experiments to Validate Fuel Depletion Calculation Methodologies" was organised and held in Rez (near Prague), Czech Republic, on 11-12 May 2006 [1]. At the EGBUC and WPNCS meeting in 2006, held in Aix-en-Provence after the workshop at Rez, it was concluded that an effort dedicated specially to the collection and analyses of the measured assay data for nuclear fuel should be emphasised and the new Expert Group on Assay Data for Spent Nuclear Fuel (EGADSNF) was established. The objective of the group is to help co-ordinate assay data activities and facilitate co-operation between countries developing and implementing burn-up credit methodologies. In the area of spent fuel assay data, co-operation is especially beneficial considering the very high cost of initiating new experimental assay programmes (fuel transportation, hot cell facilities, radiochemical analysis capabilities and waste management requirements). The major activities initiated by the EGADSNF for study include:

- Expand the SFCOMPO database of available assay data by increasing the number of fuel samples, include data with higher initial enrichment and burn-up values and expand the reactor types beyond mainly LWR fuel.
- Review and improvement of the formats and structure of the spent fuel database to allow inclusion of more detailed information and measurement uncertainties.
- Provide access to primary experimental reports where available.
- Provide recommended fuel design and operating history information necessary for computational analysis and validation.
- Evaluate potential uncertainties due to missing or incomplete experiment documentation.
- Document recommended radiochemical analysis techniques, typical accuracies, data reduction methods and best practices based on previous laboratory experience.
- Publish a final state-of-the art report on assay data of spent nuclear fuel.

The present report discusses the activities of the EGADSNF and the importance of the assay data for a wide range of spent nuclear fuel storage, nuclear safety and fuel cycle issues. The report attempts to represent the state-of-the-art in spent fuel assay data and analysis methods. Experimental methods cover the full range of the isotopic analysis process including radiochemical and radiometric analysis techniques commonly used to measure nuclide compositions in spent fuel, documentation requirements, requirements for modelling and simulation, and uncertainties associated with modelling approximations and incomplete data. Perhaps one of the most important and ongoing activities of the Expert Group is the expansion of SFCOMPO to include uncertainty information as well as additional spent fuel measurements from previously unpublished experimental programmes and new programmes initiated and completed since the last update of the database in 2002. The new data include high burn-up PWR and BWR data for modern $UO_2$ fuels designs, MOX fuels, VVER fuels and fuel from gas-cooled reactors.

Because of the importance of the assay data to many different areas of spent fuel safety and management, the Expert Group consists not only of the members from WPNCS representing nuclear criticality safety, but also members of the standing technical Committee on the Safety of Nuclear Installations (CSNI) and the working group Integration Group for the Safety Case (IGSC). Spent fuel compositions are the basis for evaluation of radioactivity, neutron and gamma ray source terms and decay heat. Assay data are necessary to validate many design and safety evaluations for the nuclear fuel cycle and back-end nuclear facilities related to fuel handling, dry spent fuel storage installations, pool storage, fuel reprocessing facilities and waste repository studies.

# Chapter 2: Requirements for assay data of spent nuclear fuel

The purpose of assay experiments is to measure the nuclide content of irradiated nuclear fuel. The measurements can include non-destructive gamma-ray scanning of fuel rods (for burn-up distribution measurements), mass spectrometry measurement of fission product gases, and destructive analysis of fuel rod segments for radiochemical analysis and gamma counting. The measured data are used to validate the accuracy of computer code predictions, by comparing measured compositions directly with compositions calculated by codes using well-characterised fuel samples (i.e. fuel with a sufficient level of design and operational detail to enable modelling). The nuclide compositions are also used to experimentally determine the burn-up of the fuel.

The results obtained from the comparison between calculated and measured spent fuel inventories are not only part of the validation process of the codes and their associated libraries, but moreover can represent an important component of nuclear data evaluation. The accuracies of isotopic predictions provide one part of the overall feedback given to the international data evaluation committees through the results of differential and integral experiments.

Requirements for the assay measurements are determined to a large extent by the intended use of the data and technical application area. Early experiments focused mostly on measuring the major actinides for studies of uranium transmutation and plutonium production and nuclides for burn-up determination of the fuel (e.g. $^{137}$Cs and $^{148}$Nd). Later programmes were expanded to include radiological important nuclides such as $^{90}$Sr, $^{137}$Cs and $^{154}$Eu to support nuclear fuel safety analysis, and $^{14}$C, $^{135}$Cs, $^{99}$Tc, $^{126}$Sn, $^{129}$I and other long-lived fission products for waste repository analysis. The requirements of assay data for burn-up credit have broadened the range of nuclides of interest to include stable and long-lived fission products with large capture cross-sections for which very few measurements were previously available, including isotopes of Sm, Eu and Gd. Other stable fission products important to burn-up credit, including $^{95}$Mo, $^{101}$Ru, $^{103}$Rh and $^{109}$Ag, may form partially as metallic particles in the fuel that are difficult to dissolve and present unique challenges for radiochemistry. In this section the requirement for measured nuclide assay data for nuclear fuel is described for several research areas.

## 2.1 Nuclear criticality safety

Burn-up credit is defined as the consideration of a reduction in reactivity of nuclear fuel due to nuclide composition changes associated with fuel irradiation in nuclear criticality safety analyses. The evolution of the nuclide content during fuel irradiation results in reactivity changes that can be characterised by the reduction in the net fissile material content, the build-up of actinides and the increase in the concentration of fission products. In the case of fuel in the presence of burnable absorbers such as gadolinium or other neutron poisons, the reactivity effect associated with the reduction of the absorber during irradiation must also be considered. The application of burn-up credit in criticality calculations requires a demonstrated capability of determining $k_{eff}$ for a system. A methodology specifically relying on assay data experiments involves two distinct steps: the first to determine the nuclide compositions of the spent fuel and their distributions, the second to perform the criticality calculation using the nuclide compositions calculated in the first step. Consequently, validation of this burn-up credit analysis method requires validation of both the burn-up calculation and the criticality calculation.

Selection of nuclides adopted in a burn-up credit criticality calculation depends on the model to be adopted for the safety evaluation. However, the nuclides should be selected on the basis of their importance in the criticality calculation (i.e. large macroscopic neutron capture or fission cross-sections with large reactivity worth), and their nuclear and chemical stability. These nuclides will generally include the major actinides that contribute to significant positive reactivity nuclear fuel such as $^{235}$U, $^{239}$Pu and $^{241}$Pu, stable isotopes or isotopes with long half-lives, and exclude volatile elements such as noble gas xenon and krypton, or caesium in the case of long-term repository analysis.

Another important consideration in selecting nuclides used in burn-up credit is the availability of sufficient experimental data for validating both the depletion calculation and the criticality calculation. Despite wide interest in including and expanding the use of fission products in burn-up credit, the lack of adequate experimental data for fission products has been an obstacle to code and data library validation and the acceptance of fission products in burn-up credit criticality analyses in many countries.

Table 1 lists the actinides and fission products commonly considered in burn-up credit for interim spent fuel storage configurations [2]. Although the relative importance of each nuclide is a function of the fuel configuration, enrichment, burn-up and cooling time, the principal nuclides remain relatively unchanged.

Many actinides are generated through the processes of neutron reactions, mainly neutron capture and (n,2n) reactions, and radioactive decay ($\beta$ and $\alpha$ decay), and play a dominant role in reactor analysis and nuclear criticality safety calculations. Different nuclides may be important for different fuel types. For example, for studies on the fuel cycle using thorium and the long-term storage of spent fuel, $^{232}$Th and $^{233}$U should be considered. Higher actinides such as $^{245}$Cm may become important for MOX fuel studies. In the OECD/NEA benchmarks described in the following section, the twelve nuclides $^{234}$U, $^{235}$U, $^{236}$U, $^{238}$U, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu, $^{241}$Am, $^{243}$Am, and $^{237}$Np were selected as the dominant burn-up credit actinides considered for the benchmark problem.

More than a thousand fission products (FP) are generated by the nuclear fission process. However, from the view point of reactivity effect, the importance of approximately 20 FP is generally emphasised. In the OECD/NEA benchmarks described in the following section, the 15 fission product nuclides $^{95}$Mo, $^{99}$Tc, $^{101}$Ru, $^{103}$Rh, $^{109}$Ag, $^{133}$Cs, $^{143}$Nd, $^{145}$Nd, $^{147}$Sm, $^{149}$Sm, $^{150}$Sm, $^{151}$Sm, $^{152}$Sm, $^{153}$Eu and $^{155}$Gd were selected for the benchmark problem. The aggregate fission product inventory in spent fuel represents typically 25-30% of the net reactivity effect in spent fuel. The contribution from the six most important FP $^{143}$Nd, $^{149}$Sm, $^{103}$Rh, $^{151}$Sm, $^{133}$Cs and $^{155}$Gd, is about 20% of the net reactivity effect for typical UOX fuels.

The following section outlines the international benchmarks organised by the OECD/NEA that are related to nuclear criticality safety involving burn-up credit.

### 2.1.1 Related OECD/NEA benchmark activities

The importance of the accuracy of burn-up codes used to calculate nuclide compositions for burn-up credit and the need for comparisons between calculation and experimental data were recognised from the beginning of the benchmark activities by the Expert Group. The OECD/NEA initiated activities on burn-up credit in 1991 by establishing the EGBUC. The group has organised several code benchmarks dedicated to burn-up credit. Its initial activity was a simple criticality calculation for a PWR fuel pin cell. Since then the EGBUC has performed a large array of code benchmarks related to burn-up credit [3]. Table 2 lists the series of completed and ongoing calculational benchmarks within the WPNCS and related depletion benchmarks organised by the Working Party on the Scientific Issues of Reactor Systems (WPRS).

## Table 1: List of nuclides commonly considered in burn-up credit criticality analyses

| Nuclide | Half-life (years) | Content in spent UOX PWR fuel[a] (g/MTHM) 52 GWd/t at discharge | Thermal neutron capture cross-section[b] (barns) | Thermal neutron fission cross-section[b] (barns) | Relative importance rank for 40 GWd/MTHM PWR fuel – 5 years cooling[c] |
|---|---|---|---|---|---|
| $^{234}$U | $2.446 \times 10^{5}$ | 143 | 99.8 | | 24 |
| $^{235}$U | $7.038 \times 10^{8}$ | 6 050 | 98.8 | 582.6 | 1 |
| $^{236}$U | $2.342 \times 10^{7}$ | 5 650 | 5.09 | | 11 |
| $^{238}$U | $4.468 \times 10^{9}$ | 927 000 | 2.68 | | 3 |
| $^{238}$Pu | 87.74 | 372 | 540 | 17.9 | 22 |
| $^{239}$Pu | $2.411 \times 10^{4}$ | 5 810 | 269.3 | 748 | 2 |
| $^{240}$Pu | 6 550 | 2 840 | 289.5 | | 4 |
| $^{241}$Pu | 14.4 | 1 820 | 362.1 | 1 011 | 5 |
| $^{242}$Pu | $3.763 \times 10^{5}$ | 1 020 | 18.5 | | 19 |
| $^{237}$Np | $2.14 \times 10^{6}$ | 811 | 175.9 | | 14 |
| $^{241}$Am | 432.6 | 228 | 587 | 3.2 | 10 |
| $^{243}$Am | 7 370 | 1.74 | 75.1 | | 11 |
| $^{243}$Cm[d] | 28.5 | 0.624 | 130 | 617 | |
| $^{244}$Cm[d] | 18.11 | 141 | 15.2 | 1.04 | |
| $^{245}$Cm[d] | 8 532 | 11 | 369 | 2 144 | |
| $^{133}$Cs | Stable | 1 630 | 30.3 | | 12 |
| $^{143}$Nd | Stable | 1 070 | 325 | | 7 |
| $^{145}$Nd | Stable | 989 | 50.0 | | 17 |
| $^{147}$Sm | $1.06 \times 10^{11}$ | 196 | 57 | | 20 |
| $^{149}$Sm | $2.0 \times 10^{15}$ | 3.36 | 40 140 | | 6 |
| $^{150}$Sm | Stable | 446 | 100 | | 23 |
| $^{151}$Sm | 93 | 14.7 | 15 170 | | 9 |
| $^{152}$Sm | Stable | 134 | 206 | | 15 |
| $^{153}$Eu | Stable | 184 | 312 | | 18 |
| $^{155}$Gd | Stable | 3.93 | 60 900 | | 13 |
| $^{95}$Mo | Stable | 1 180 | 13.4 | | 21 |
| $^{99}$Tc | $2.1 \times 10^{5}$ | 1 120 | 22.8 | | 16 |
| $^{101}$Ru | Stable | 1 210 | 5.2 | | 26 |
| $^{103}$Rh | Stable | 540 | 243.5 | | 8 |
| $^{109}$Ag | Stable | 119 | 91.0 | | 25 |
| $^{113}$Cd[d] | $9.10 \times 10^{15}$ | | 20 615 | | |

[a] Measured content from ARIANE experimental programme data.

[b] From S.F. Mughabghab, *Atlas of Neutron Resonances – Resonance Parameters and Thermal Cross Sections Z = 1-100*, 5th Edition, Elsevier, Amsterdam (2006).

[c] Based on relative sensitivity coefficients from G. Radulescu, D.E. Mueller, J.C. Wagner, *Sensitivity and Uncertainty Analysis of Commercial Reactor Criticals for Burn-up Credit*, NUREG/CR-6951, Nuclear Regulatory Commission (2008).

[d] Important for MOX fuel only.

**Table 2: OECD/NEA burn-up credit criticality and fuel cycle safety benchmarks**

| Working party | Benchmark | Fuel type | Description |
|---|---|---|---|
| WPNCS | Phase I-A [5] | PWR $UO_2$ | Criticality calculation using the nuclide density data of spent nuclear fuel |
| | Phase I-B [6] | | Burn-up calculation using a single pin cell model based on Calvert Cliffs Unit 1 |
| | Phase II-A [7] | | Criticality calculation for an finite array of fuel pins, with a burn-up distribution specified in the axial direction |
| | Phase II-B [8] | | Three-dimensional transport calculation of a cask configuration, considering a burn-up distribution in the axial direction |
| | Phase II-C [9] | | Assessment of the effect of the asymmetry of burn-up distribution on the neutron multiplication factor |
| | Phase II-D [10] | | Effect of the control rod on burn-up calculation |
| | Phase II-E [11] | | Study of the nuclide inventory changes due to control rod insertion and the impact on the end effect |
| | Phase III-A [12] | BWR $UO_2$ | Three-dimensional criticality calculation with axial burn-up and void distribution |
| | Phase III-B [13] | | Two-dimensional burn-up calculation |
| | Phase IV-A [14] | PWR MOX | Criticality calculation for one-dimensional pin cell geometry |
| | Phase IV-B [15] | | Burn-up calculation for fuel assembly |
| | Phase V [16] | VVER 440 | Simplified burn-up benchmark based on VVER nuclide measurements |
| | Phase VII [17] | PWR $UO_2$ | Study of spent fuel compositions and criticality safety for long-term waste management |
| WPRS | Phase I [18] | PWR $UO_2$ | Depletion benchmark for fuel cycle studies |
| | Phase II [19] | PWR MOX | Depletion benchmark for fuel cycle studies |

WPNCS Phase I-B, Phase III-B and Phase IV-B benchmarks all include comparisons of calculated fuel nuclide compositions at different burn-ups. One of the first benchmark activities of the EGBUC, carried out in the Phase I-B benchmark, evaluates code predictions of nuclide composition data for spent fuel by comparison to radiochemical experimental data obtained for fuel irradiated in the Calvert Cliffs Unit 1 reactor. The geometry specification for the benchmark was an equivalent cell calculation to simplify the assembly calculational model. The nuclides selected by the Expert Group for the comparison included 12 actinides and 15 fission products. Results from 21 different sets of calculations submitted by 16 organisations world wide were compared with the averaged calculation result in order to evaluate the differences between calculation methods and data.

The OECD/NEA/NSC Working Party on the Physics of Plutonium Fuels and Innovative Fuel Cycles (WPPR), now the Working Party on the Scientific Issues of Reactor Systems (WPRS), is active in the area of fuel cycle issues. This working group released a benchmark in 2004 to evaluate code results for calculations of radioactivity, decay heat and the neutron emission rate of nuclear fuel waste to address important fuel cycle issues. The WPRS benchmark problem was based on experimental assay data selected from fuel irradiated in the Takahama-3 PWR. The WPRS benchmark specified a simple cell model and a full fuel assembly model for the model geometries. The results of this benchmark exercise were presented at PHYSOR 2006, the international conference on reactor physics [4].

In 1996 the Czech Republic became the first country of Central and Eastern Europe to join the OECD and NEA. Other countries with operating VVER reactors, Hungary and Slovakia, later became members of the Nuclear Energy Agency and joined the WPNCS activities on burn-up credit benchmarking. They specified and calculated a series of VVER benchmarks named CB1, CB2, CB2-S, CB3, CB3+, CB4 [20] and CB5 [21], similar to the burn-up credit benchmarks for PWR mentioned in Table 2. Although these benchmarks were prepared and carried out in collaboration with the OECD/NEA WPNCS, they were not formally part of the OECD benchmarks. The CB2 benchmark, which was focused on nuclide predictions, was a VVER equivalent of the Phase I-B benchmark for PWR. Due to the lack of available measured assay data for the VVER fuels at the time, the CB benchmarks involved only calculations. Benchmark results were not compared with any experimental data. An experimental VVER benchmark was later carried out within the framework of the OECD/NEA. This Phase V burn-up credit benchmark was based on a simplified representation of a VVER 440 assembly and measured assay data acquired under the International Science and Technology Center (ISTC) Project #2670 experiments [22].

## 2.2 Nuclear waste management

Existing waste management avenues for spent nuclear fuel consist of either storage and disposal of the spent fuel, or reprocessing, encapsulation, storage and disposal of the waste products. Measurement data on the concentration of radionuclides in spent fuel are required to support the waste management and post-closure safety assessments of nuclear fuel repositories. However, prioritised lists of radionuclides of interest and the measurement accuracy for waste disposal are somewhat different from other projects, e.g. criticality safety. In the field of safety assessment of spent fuel and vitrified high-level waste repositories, long-lived fission products are very important radionuclides in addition to long-lived actinides. Also, long-lived activation products from the fuel impurities can represent dominant radiological sources (e.g. $^{14}$C and $^{36}$Cl). Factors influencing the importance (or otherwise) of a radionuclide in post-closure safety assessments under normal evolution scenarios are its half-life, its mobility (which varies depending on the host rock and the disposal concept) and its radiological significance when reaching the biosphere.

For the waste disposal safety assessment, the accuracy of the measured radionuclide inventories may not need to be as high compared to other applications because of the large uncertainties inherent in dose or risk calculations over the long time frames associated with repository safety analyses. Nevertheless, the radioactive inventory of spent fuel represents the initiating source of long-term safety assessment, and activities related to confirming or improving the accuracy of the spent fuel inventory will clearly be of interest and benefit the waste disposal safety assessment. Table 3 is a compiled list of radionuclides of interest to waste repository evaluations based on information provided by member organisations from the Integration Group for the Safety Case of Radioactive Waste Repositories (IGSC) and other organisations. The list is based on results of post-closure safety analyses for normal situations of release through the aqueous pathway in waste disposal programmes of each country. In human intrusion scenarios, however, other less often mentioned radionuclides, such as $^{126}$Sb, $^{126m}$Sb and $^{209}$Tl, also become important for gamma dose calculations over various time frames.

Although it is difficult to give a clear priority to assay data needs from the list presented in Table 3, it can be concluded that experimental data for long-lived fission products is limited as compared with the available data for actinides. Based on the experience of the members of the EGADSNF, there is broad interest in the spent fuel and high-level vitrified waste management community as regards $^{14}$C, $^{36}$Cl, $^{79}$Se, $^{99}$Tc, $^{126}$Sn, $^{129}$I and $^{135}$Cs. The development of reliable analytical techniques may be needed for some radionuclides. In addition to the isotopic concentrations, it is also important to know the total elemental concentrations for elements such as Se and Sn since their release and transport behaviour is solubility-controlled. As some of these nuclides/ elements are normally found within inter-metallic particles in spent fuel (i.e. Mo, Tc), experiments need to ensure that these are adequately included in the analyses. In the case of $^{14}$C, the chemical state is also important, as it influences the chemical form (i.e. organic or inorganic form) after release into the water. However, such determinations are presently generally not possible with current spent fuel assay measurements.

In addition to the total radionuclide inventory, the distribution of the inventory associated with different release categories, such as the solid fuel matrix, gap, grain boundaries and metal parts (e.g. surface of Zircaloy cladding), is also important with regard to long-term safety

Table 3: Nuclides of primary interest for the disposal of spent fuel and high-level vitrified waste (shown with the symbol ■)

| Nuclide | Nagra$^{a}$ Switzerland | ONDRAF$^{b}$ Belgium | NWMO$^{c}$ Canada | SKB$^{d}$ Sweden | OCRWM United States$^{e}$ | Posiva$^{f}$ Finland | RAWRA$^{g}$ Czech Republic | GRS-BS$^{h}$ Germany | | | NUMO/JAEA$^{i}$ Japan | NRG $^{j}$ Netherlands | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | Salt | Granite | Clay | | Salt | Clay |
| $^{14}$C | ■ | ■ (20%) | ■ | ■ | ■ | ■ | ■ | ■$^{B}$ | ■$^{A}$ | ■$^{B}$ | ■ | | |
| $^{36}$Cl | ■ | ■ (20%) | ■ | ■ | ■ | ■ | ■ | ■$^{B}$ | ■$^{A}$ | ■$^{A}$ | ■ | ■ | ■ |
| $^{41}$Ca | | | ■ | | | | | | | | | | |
| $^{59}$Ni | | | | | | ■ | | ■$^{B}$ | | | | | |
| $^{79}$Se | ■ | ■ (50%) | ■ | ■ | ■ | ■ | ■ | ■$^{A}$ | ■$^{B}$ | ■$^{A}$ | ■ | ■ | ■ |
| $^{90}$Sr | | | | | ■ | ■ | | | | | | | |
| $^{93}$Zr | | | ■ | | | ■ | | | | | ■ | | |
| $^{93}$Nb | | | | | | ■ | | | | | | | |
| $^{94}$Nb | | | | | | ■ | | | ■$^{B}$ | | | | |
| $^{93}$Mo | | | | | | ■ | | | ■$^{B}$ | | | | |
| $^{99}$Tc | | ■ (20%) | ■ (<50%) | | ■ | | ■ | ■$^{B}$ | | | ■ | | |
| $^{107}$Pd | | ■ (20%) | | | | ■ | | | | | | | |
| $^{126}$Sn | | ■ (20%) | ■ | ■ | ■ | ■ | ■ | ■$^{B}$ | ■$^{B}$ | | ■ | | ■ |
| $^{126}$Sb | | | | | | ■ | | | | | | | |
| $^{129}$I | ■ | ■ (20%) | ■ (<50%) | ■ | ■ | ■ | ■ | ■$^{A}$ | ■$^{A}$ | ■$^{A}$ | ■ | ■ | ■ |
| $^{135}$Cs | | | | | ■ | ■ | | ■$^{A}$ | ■$^{A}$ | | ■ | ■ | |
| $^{137}$Cs | | | ■ (<5%) | | ■ | | | | | | | | |
| $^{210}$Pb | | | | | ■ | | | | | | | | |
| $^{226}$Ra | | | | | ■ | ■ | ■ | ■$^{B}$ | ■$^{B}$ | | | | |
| $^{227}$Ac | | | | | ■ | | ■ | | | | | | |
| $^{228}$Ra | | | | | ■ | | | | | | | | |
| $^{229}$Th | | | | | ■ | ■ | ■ | | | | | | |
| $^{230}$Th | | | | | ■ | | | ■$^{C}$ | ■$^{C}$ | | | | |
| $^{232}$Th | | | | | ■ | | | | | | | ■ | ■ |
| $^{231}$Pa | | | | | ■ | | | | | | | | |
| $^{232}$U | | | | | ■ | | | | | | | | |
| $^{233}$U | | | | | ■ | | | | | | | | |
| $^{234}$U | | | | | ■ | | | | | | | ■ | ■ |
| $^{235}$U | | | | | ■ | | ■ | ■$^{B}$ | ■$^{C}$ | | | | ■ |
| $^{236}$U | | | | | ■ | | ■ | ■$^{B}$ | ■$^{C}$ | | | ■ | ■ |
| $^{238}$U | | | | | ■ | | ■ | ■$^{C}$ | ■$^{C}$ | | | ■ | ■ |
| $^{237}$Np | | | ■ (<20%) | ■ | ■ | ■ | ■ | ■$^{B}$ | ■$^{B}$ | | ■ | ■ | ■ |
| $^{238}$Pu | | | | | ■ | | | | | | | | |
| $^{239}$Pu | | | ■ | | ■ | ■ | ■ | ■ | ■$^{C}$ | | | | |
| $^{240}$Pu | | | ■ | | ■ | | | ■$^{C}$ | ■$^{C}$ | | | | |

**Table 3: Nuclides of primary interest for the disposal of spent fuel and high-level vitrified waste (shown with the symbol ■) (*cont.*)**

| Nuclide | Nagra[a] Switzerland | ONDRAF[b] Belgium | NWMO[c] Canada | SKB[d] Sweden | OCRWM United States[e] | Posiva[f] Finland | RAWRA[g] Czech Republic | GRS-BS[h] Germany | | | NUMO/JAEA[i] Japan | NRG [j] Netherlands | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | Salt | Granite | Clay | | Salt | Clay |
| $^{241}$Pu | | | ■ | ■ | ■ | ■ | | ■$^{C}$ | ■$^{C}$ | | | | |
| $^{242}$Pu | | | | | ■ | | | | | | | | |
| $^{241}$Am | | | ■ | | ■ | ■ | | ■$^{C}$ | ■$^{C}$ | | ■ | | |
| $^{243}$Am | | | | | ■ | | | | | | | | |
| $^{245}$Cm | | | | | ■ | ■ | | | | | ■ | | |
| Mo (total) | | | ■ | | | | | | | | | | |
| Te (total) | | | ■ | | | | | | | | | | |

[a] Nuclides of primary interest based on dose calculations for the reference case of Project Entsorgungsnachweis, Nagra Technical Report NTB 02-05. The order of importance (spent fuel): $^{129}$I > $^{36}$Cl > $^{14}$C > $^{79}$Se.

[b] Dominant radionuclides in case of spent fuel disposal in a clay layer within the Belgian radioactive waste disposal report (NIROND 2001-06E, SAFIR2). The order of importance: $^{129}$I, $^{79}$Se, $^{126}$Sn > $^{36}$Cl, $^{99}$Tc > $^{14}$C, $^{107}$Pd. The desired accuracy of the measurements for each nuclide is shown in parentheses.

[c] Nuclides important to long-term safety assessment of used fuel. The number shown in parentheses is reported accuracy based on a comparison between ORIGEN-S analysis and CANDU fuel assay data [Tait, *et al.*, *J. Nucl. Mat.*, 223, 109-121 (1995)]. Mo (total) and Te (total) are potential chemical risk contributors present as fission products. $^{137}$Cs is important for scaling of difficult-to-measure species. More important species are $^{129}$I, $^{36}$Cl, $^{14}$C, $^{241}$Pu, $^{241}$Am (parent of $^{237}$Np) [Gierszewski, *et al.*, *TCS (Third Case Study) Post Closure Safety Assessment*, Ontario Power Generation, Report 06819-REP-01200-10109-R00 (2004)].

[d] The prioritised list of nuclides of interest for spent fuel management based on recent safety assessment, SKB TR-06-09. Note that also the contents of $^{238}$U and $^{234}$U and their daughter products are crucial for the results of the safety assessment. However, the inventories of the U isotopes follow trivially from the amount of spent fuel and its enrichment and the inventories of the daughter products then follow trivially and with negligible uncertainties from the Bateman equations. These nuclides are thus not listed as prioritised regarding the need to compile additional inventory data.

[e] Nuclides identified as important for the Total System Performance Assessment, Report of the Office of Civilian Radioactive Waste Management, *Initial Radionuclide Inventories*, Sandia National Laboratories, ANL-WIS-MD-00020 REV 01 ADD 01, Las Vegas, Nevada. Sandia National Laboratories, ACC: DOC.20050927.0005; DOC.20070801.0001; LLR.20080408.0246 (2007). The broader interest in nuclides for the OCRWM programme reflects the proposed use of an oxidising repository environment. However, US policy changed in 2009 and this particular environment will not be used for spent fuel disposal.

[f] The most important nuclides based on the recent safety assessments and the two-tiered screening evaluation performed in the ongoing biosphere assessment in POSIVA 2007-07, SKB R-08-38 and POSIVA 2008-06. Note also that the content of the parent uranium nuclides $^{238}$Pu, $^{238}$U and $^{234}$U and their daughter products are important.

[g] The prioritised list of radionuclides for waste management and disposal is based on reference geological repository performance analysis in RAWRA-PR-11.

[h] Nuclides of primary interest based on dose relevance of the radionuclides and derived from the results of previous long-term safety assessment studies (Report GRS-154, 206, 228 and 240) for radioactive waste repositories of rock salt, granite and clay in particular for repository with spent fuel. The nuclides are divided into three categories:

A: Most important nuclides. The accuracy of the inventory should be as high as possible. The maximum error should be below 10%.

B: Important nuclides. The maximum error should be below 50%.

C: Not directly dose relevant radionuclide but their inventories as parent nuclides are important.

[i] Nuclides dominant to the estimated dose in the H12 safety analyses of HLW (vitrified waste) disposal, JNC TN1400 2000-001, and in the safety analyses of TRU waste disposal, JAEA-Review 2007-010.

[j] Dominancy of radionuclides in case of spent fuel or vitrified waste disposal in a salt dome or a clay layer within the Dutch radioactive waste disposal programme was determined based on potential contributions to the effective dose rate. Radionuclides only relevant as decay products are not indicated in the table, as opposed to their determining parent nuclides – even if these are less relevant for the dose rate themselves. For a disposal facility to be acceptable, at all times the maximum dose rate should remain well below acceptability norms. For that reason, accuracy of the dominant radionuclide inventories within a factor of 2 is usually sufficient.

assessment. In particular, the instant release fraction of $^{129}$I is an important parameter for disposal safety assessment of spent fuel. If fuel assay measurements are also able to characterise these parameters, they would provide highly valuable information for waste management applications.

It should be noted that $^{14}$C and $^{36}$Cl are generated not only in spent fuel but also in assembly and reactor structural components such as Zircaloy, steel, graphite, etc. The initial concentration of impurities is often not well known and therefore information on these impurity levels is required in order to perform validation of these activation products. The relative importance of the radionuclides may change as more information becomes available along with the progress of each national disposal programme.

## 2.3 Nuclear fuel and fuel cycle safety

Assay data requirements for other nuclear fuel safety applications are generally different than for nuclear criticality safety and waste management purposes. Nuclear fuel safety, as discussed in this section, covers a broad range of areas including severe accident analysis, spent fuel handling and storage, reprocessing, decay heat, shielding and other radiological applications.

Current reprocessing operations carried out, for example, in France, Japan and the UK use the PUREX process for spent fuel pre-processing. This fuel cycle is well understood and involves operations including the transport of irradiated nuclear fuel, chemical processing of dissolved fuel, storage or reuse of uranium and plutonium, and the disposal of the various waste streams from the cycle. In general, each of these stages has design requirements or operational safety case requirements based upon the composition and radiological properties of the spent fuel. For example, the transport of irradiated nuclear fuel requires a safety case which specifies the radiation shielding to be used and sets a limit on the heat capacity of the transport cask. These of course depend on the nuclide concentrations in the fuel. It is clear that uncertainties in the compositions contribute to the calculated limits of these safety cases and need to be determined.

In postulated design-basis and severe accident analyses involving loss of the fuel integrity during operations or fuel handling, noble gases (Xe and Kr) and volatile fission products (I, Cs, Te, Ru) may be released. Caesium and iodine are important radiological isotopes for the source term evaluation since they are chemically active elements and have relatively large release fractions. Caesium has chemical properties similar to sodium, potassium and iodine, and is known to gather in the thyroid gland. Iodine is a chemically volatile element with high release fractions from fuel at high temperature. Iodine-129 is measurable using gamma spectrometry. However, the half-life of $^{129}$I is very long ($1.57 \times 10^7$ years), making it difficult to measure. Consequently, measurement data for iodine are not abundant.

Dose consequence analysis for severe accidents is typically dominated by many short half-life radionuclides, in particular $^{131}$I ($T_{1/2} = 8.02$ days). From the viewpoint of measured assay data it is difficult to measure the activities of the dominant nuclides in severe accident analysis directly since measurements are usually only carried out several months or years after the irradiation, depending on the capability of the experimental facility. Although direct measurement of the short-lived fission products is often impractical, the measurements of longer-lived daughters of the short-lived nuclides can provide a measure of integral short-lived fission product inventory and activity. In other cases, such as for reprocessing plants and isotope production from irradiated fuel targets, the amount of noble gas is an important parameter for the safety evaluation.

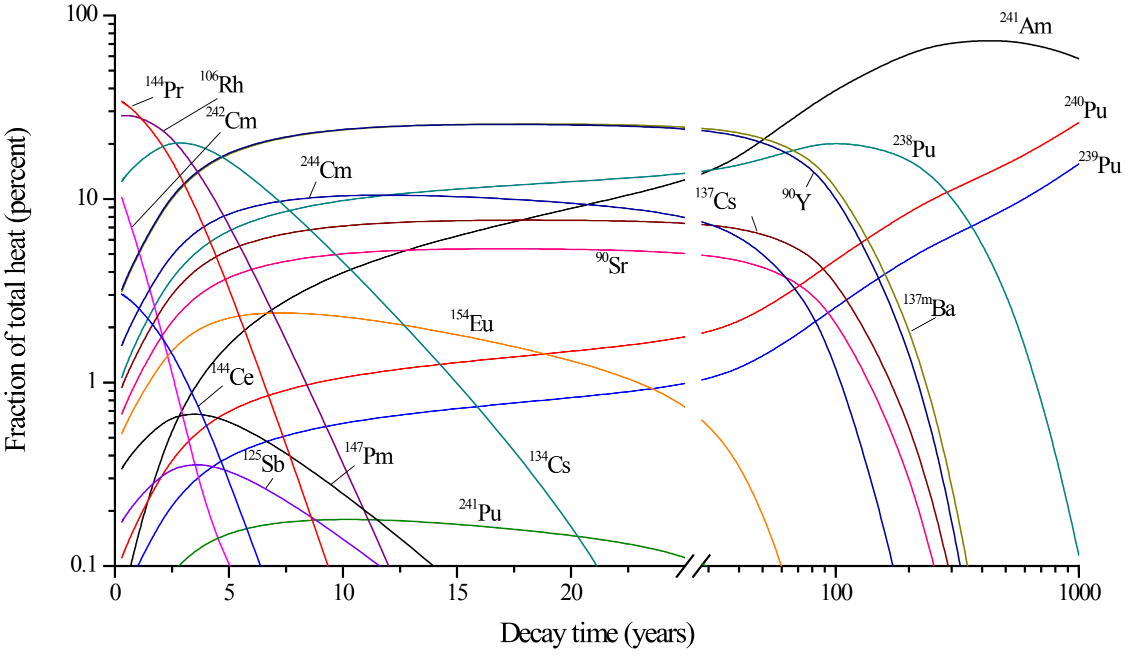
Other safety and licensing activities involving spent nuclear fuel relate to the energy release by radioactive decay (decay heat) for spent fuel handling, interim, and long-term storage, and radiation source terms used in neutron and gamma ray shielding studies. Decay heat in the time frame of severe accident analysis involves many short-lived fission products, and short-lived actinides, primarily $^{239}$U and $^{239}$Np. Because of the large number of nuclides and the short half-lives, validation of code calculations is generally performed using integral measurements of energy release after fission. At longer cooling times, of months to years, the decay heat is dominated by relatively few nuclides, many common to other application areas. The principal fission products at times less than 10 years are $^{134}$Cs, $^{137}$Cs + $^{137m}$Ba progeny, $^{90}$Sr + $^{90}$Y progeny, $^{106}$Rh, $^{154}$Eu, $^{144}$Ce + $^{144}$Pr progeny and $^{147}$Pm. Beyond 20 years the fission product decay heat is generated predominantly by $^{137}$Cs, $^{90}$Sr and their decay daughters. Principal actinides $^{244}$Cm, $^{241}$Am, $^{238}$Pu and lesser contributions from $^{239}$Pu and $^{240}$Pu become the dominant source of decay heat after approximately 50 years cooling.

Radiation source terms (neutron and gamma ray) nuclides are similar to those observed for decay heat. An exception is for high activity nuclides with low emitted radiation energies that are eliminated by source shielding. In many safety-related nuclear design studies, the source inventory is generally shielded, reducing the relative importance of nuclides emitting charged particles and low-energy gamma radiations and enhancing the importance of nuclides with energetic photon emissions. The principal nuclides identified in shielding studies for cooling times less than 100 years include $^{144}$Pr (progeny of $^{144}$Ce), $^{106}$Ru, $^{134}$Cs, $^{137m}$Ba (progeny of $^{137}$Cs), $^{154}$Eu and $^{90}$Y (progeny of $^{90}$Sr). The actinide contribution to dose rate is smaller than the fission product contribution up to 50-80 years, but $^{242}$Cm, $^{244}$Cm, $^{241}$Am and $^{238}$Pu add significantly to the dose rate in this period and for longer times.

The relative nuclide importance in spent fuel decay heat evaluations, as a function of cooling time, is shown in Figure 1. The calculations were performed for an enrichment of 4.5 wt.% and burn-up of 50 GWd/t. The nuclides shown are those with a large importance for cooling times longer than about one year.

A summary of the nuclides commonly measured in modern large experimental programmes, such as Actinide Research in a Nuclear Element (ARIANE) [23,24], is listed in Table 4 showing their respective importance to different applications including burn-up credit, decay heat generation and radiological applications. This list also includes other nuclides that are commonly measured to assess the sample burn-up and other precursors needed to adjust the measured data to a common reference cooling time. These other nuclides are discussed later in the report.

**Figure 1: Important nuclides contributing to decay heat for typical LWR fuel for cooling times from about 1 year to 1 000 years**



### Table 4: Commonly measured nuclides of importance to different safety-related spent fuel applications

| Nuclide | Half-life (years) | Burn-up credit | Radiological safety[b] | Waste management | Comments |
|---|---|---|---|---|---|
| $^{79}$Se | $2.95 \times 10^{5}$ | | | ■ | |
| $^{95}$Mo | Stable | ■ | | | Metallic |
| $^{90}$Sr | 28.9 | | ■ | ■ | Decay precursor of $^{90}$Y |
| $^{99}$Tc | $2.111 \times 10^{5}$ | ■ | | ■ | Metallic |
| $^{101}$Ru | Stable | ■ | | | Metallic |
| $^{106}$Ru | 371.6 days | | ■ | | Metallic |
| $^{103}$Rh | Stable | ■ | | | Metallic |
| $^{109}$Ag | Stable | ■ | | | Metallic |
| $^{125}$Sb | 2.7586 | | ■ | | Metallic |
| $^{129}$I | $1.6 \times 10^{7}$ | | | ■ | Off gas during dissolution |
| $^{133}$Cs | Stable | ■ | | | |
| $^{134}$Cs | 2.065 | | ■ | | |
| $^{135}$Cs | $2.3 \times 10^{6}$ | | | ■ | |
| $^{137}$Cs | 30.0 | | ■ | ■ | Burn-up indicator and precursor of $^{137m}$Ba |
| $^{139}$La | Stable | | | | Burn-up indicator |
| $^{143}$Nd | Stable | ■ | | | |
| $^{145}$Nd | Stable | ■ | | | |
| $^{148}$Nd | Stable | | | | Burn-up indicator |
| $^{144}$Ce | 284.9 days | | ■ | | Decay precursor of $^{144}$Pr |
| $^{147}$Pm | 2.623 | ■[a] | | | Decay precursor of $^{147}$Sm |
| $^{147}$Sm | $1.06 \times 10^{11}$ | ■ | | | |
| $^{149}$Sm | Stable | ■ | | | |
| $^{150}$Sm | Stable | ■ | | | |
| $^{151}$Sm | 90 | ■[a] | | | Decay precursor of $^{151}$Eu |
| $^{152}$Sm | Stable | ■ | | | |
| $^{151}$Eu | Stable | ■ | | | |
| $^{153}$Eu | Stable | ■ | | | |
| $^{154}$Eu | 8.59 | | ■ | | |
| $^{155}$Eu | 4.753 | ■[a] | | | Decay precursor of $^{155}$Gd |
| $^{155}$Gd | Stable | ■ | | | |
| $^{234}$U | $2.455 \times 10^{5}$ | ■ | | ■ | |
| $^{235}$U | $7.037 \times 10^{8}$ | ■ | | ■ | |
| $^{236}$U | $2.342 \times 10^{7}$ | ■ | | ■ | |
| $^{238}$U | $4.468 \times 10^{9}$ | ■ | | ■ | |
| $^{237}$Np | $2.14 \times 10^{6}$ | ■ | | ■ | |
| $^{238}$Pu | 87.71 | ■ | ■ | ■ | |
| $^{239}$Pu | $2.41 \times 10^{4}$ | ■ | ■ | ■ | |
| $^{240}$Pu | $6.56 \times 10^{3}$ | ■ | ■ | ■ | |
| $^{241}$Pu | 14.29 | ■ | | ■ | |
| $^{242}$Pu | $3.75 \times 10^{5}$ | ■ | | ■ | |
| $^{241}$Am | 433 | ■ | ■ | ■ | |
| $^{243}$Am | 7 370 | ■ | | ■ | |
| $^{242}$Cm | 162.8 days | | ■ | | |
| $^{243}$Cm | 29.1 | ■ | | | MOX and high burn-up UOX fuel |
| $^{244}$Cm | 18.1 | | ■ | | |
| $^{245}$Cm | $8.5 \times 10^{3}$ | ■ | | ■ | MOX and high burn-up UOX fuel |

[a] Not directly relevant radionuclide but their inventories as parent nuclides are important.

[b] Radiological safety for a dry spent fuel storage facility for spent fuel cooling times >1 year to 1 000 years.

# Chapter 3: Status of nuclear fuel assay programmes in NEA member countries

Although the importance of nuclear fuel assay data as a critical component in computer code validation and uncertainty analysis is widely recognised, there has not been a concerted or co-ordinated effort in most countries to acquire the necessary data to support validation efforts. Experimental programmes have often been carried out as *ad hoc* activities. Performing radiochemical assay measurements on spent fuel is not an easy or inexpensive task and it requires the collaboration of many institutions, utilities, fuel manufacturers and engineering companies. Experimental programmes involve transportation of spent fuel from nuclear power plants to laboratories, fuel rod sectioning, sample preparation, and radiochemical analysis. Facilities must be able to receive, handle and dispose of relatively large quantities of radioactive materials. Information on the fuel design and reactor operating conditions also requires collaboration with the reactor operators and fuel vendors. Moreover, radiochemical analysis measurements require expertise that is developed over many years of experience. Only a limited number of countries have the necessary facilities and experience to carry out assay measurements on spent nuclear fuel. Due to the complexity of organising and carrying out such large experimental programmes, several recent programmes have involved international co-operation of many organisations with expertise in reactor operations, transportation, assay measurements and other organisations involved with the application of the data such as research, engineering companies and regulatory authorities.

This section provides an overview of the experimental assay data activities in several NEA member countries. Data from several programmes discussed in this section are considered publicly available while other data involve commercial proprietary programmes. As time passes some of the commercial data are being released to the public domain. Therefore this section describes both the publicly available data and several commercial programmes.

## 3.1 Belgium

In Belgium, Belgonucléaire (BN) has been very active in the field of radiochemical analysis of spent fuel and has managed several international programmes to measure assay data. Because BN belongs to the private sector, they carry out experimental programmes not only for their purposes, but also for other organisations in the international community by gathering the budget from participating institutes. In the programmes managed by BN, several radiochemical analysis laboratories in Europe have been used to perform assay measurements including Studiecentrum voor Kernenergie-Centre d'étude de l'Energie Nucléaire (SCK•CEN) in Belgium, the Institute for Transuranium Elements (ITU) in Germany, the Paul Scherrer Institute (PSI) in Switzerland and the Commissariat à l'énergie atomique (CEA) in France. In many of the assay programmes multiple laboratories are involved to provide independent cross-check measurements on fuel samples to reduce uncertainty and independently confirm the accuracy of the data. Most of these programmes include extensive measurements for the nuclides important to burn-up credit, decay heat and radiation source terms listed in Table 4.

One of the most well known experimental programmes co-ordinated by BN is "Actinide Research in a Nuclear Element" (ARIANE) [23], carried out to obtain isotopic composition data of $UO_2$ fuel and MOX fuel. The measurement matrix of ARIANE is summarised in Table 5.

In December 2000, the final ARIANE programme report was issued by BN. Examples of the use of these experimental data for code validation can be found in Refs. [25,26]. The "Radiochemical Analysis of MOX and UOX LWR Fuels Irradiated to High Burn-up" (MALIBU) programme is a successor of ARIANE designed to obtain assay data for high burn-up $UO_2$ and MOX fuel from both PWR and BWR [27]. The measurements performed under the MALIBU programme are summarised in Table 6. Since MALIBU is an ongoing proprietary programme, the details of the programme are not open.

**Table 5: Outline of ARIANE programme**

| Reactor type and assembly design | Sample ID | Fuel type | $^{235}$U/U (wt.%) | (Pu+Am)/HM (wt.%) | Burn-up (GWd/t) | Assembly | Fuel rod |
|---|---|---|---|---|---|---|---|
| Goesgen PWR 15 × 15 | GU1 | $UO_2$ | 3.5 | – | 59.7 | 12-40 | 14H13 |
| | GU3 | | 4.1 | – | 52.5 | 16-01/ 17-01 | 16B05 |
| | GU4 | | 4.1 | – | 29.1 | | |
| Beznau-1 PWR 14 × 14 | BM1 | MOX | 0.2 | 6.0 | 47 | M109 | D3 |
| | BM3 | | | | 48.5 | | B6 |
| | BM5 | | | 5.5 | 58.9 | M308 | K7 |
| | BM6 | | | | 39.6 | | |
| Dodewaard BWR 6 × 6 | DM1 | MOX | 0.2 | 6.4 | 55.6 | Y014 | M09 |
| | DM2 | | | | 33.5 | Y012 | M05 |
| | DM3 | | | | 46.5 | | |
| | DM4 | | | | 38.1 | Y013 | M07 |
| | DU1 | $UO_2$ | | | 54.4 | | U004 |

**Table 6: Outline of MALIBU programme**

| Reactor type and assembly design | Sample ID | Fuel type | $^{235}$U/U (wt.%) | Puf/HM[a] (wt.%) | Burn-up (GWd/t) | Void fraction (%) |
|---|---|---|---|---|---|---|
| Goesgen PWR 15×15 | GGM1 | MOX | 0.2 | 5.5 | ~70 | – |
| | GGU1 | $UO_2$ | 4.3 | – | ~70 | – |
| | GGU2 | | 4.3 | – | ~50 | – |
| Gundremmingen BWR 9×9 | GRM1 | MOX | 0.2 | 5.5 | ~80 | ~20 |
| | GRM2 | | 0.2 | 5.5 | ~75 | ~50 |
| | GRM3 | | 0.2 | 5.5 | ~50 | ~70 |

[a] Puf = $^{239}$Pu + $^{241}$Pu.

Another BN programme is Reactivity Tests for a Direct Evaluation of the Burn-up Credit on Selected Irradiated LWR Fuel Bundles (REBUS), dedicated to the validation of computer codes for criticality calculations involving burn-up credit. The REBUS programme [28] included destructive analysis measurements of the irradiated fuel nuclide concentrations and the measurement of reactivity decrease associated with irradiation as measured in the VENUS critical facility at SCK•CEN.

In 2008, responsibility for the management of the BN experimental programmes was transferred to SCK•CEN in Mol, Belgium.

### 3.2 France

France has a very active experimental programme for assay measurements of the fuels irradiated in French PWR. The experimental information can be divided into two categories:

- Small fuel samples from fuel rods irradiated in French reactors, with positions in the assembly that are well characterised. The data from these measurements are sensitive to local irradiation conditions. These time-consuming and expensive experiments provide very accurate results for a limited number of samples.
- Dissolution samples of entire fuel assemblies; these are numerous and very different as to the type of assemblies measured; however, the information on the irradiation condition is more limited.

The list of French data are summarised in Table 7, as presented at the special session on spent fuel isotopic data at the 2003 International Conference on Nuclear Criticality Safety held in Tokai, Japan [29]. Five main programmes related to spent nuclear fuel analysis are used for the experimental validation of the actinide and fission product inventory. Four programmes are devoted to UOX type fuels: Bugey3, Fessenheim II, Gravelines, Cruas. One programme is dedicated to MOX fuel: Saint Laurent B1.

- Bugey3 uses standard fuel with 3.1% initial enrichment and the assembly consists of 17 × 17 rods with a Zircaloy clad. The maximum burn-up is 40 GWd/t.
- Fessenheim II allows the study of UOX fuel (3.1% $^{235}$U) with high burn-up up to 60 GWd/t.
- Gravelines is devoted to the extension of the calculation scheme validation for high burn-up (five irradiation cycles) with higher enrichment corresponding to 4.5%. This experimental programme on a 900 MWe PWR is the most important being carried out in France today.
- Cruas is devoted to the validation of fuel using enriched reprocessed uranium. It allows the $^{236}$U capture cross-section to be validated.
- Saint Laurent B1 concerns French 17 × 17 MOX assemblies. The initial Pu amount in the central zone is 5.6% and the maximum burn-up 45 GWd/t.

Full assembly dissolutions performed at the AREVA/La Hague reprocessing plants provide chemical analysis data for uranium and plutonium. The assemblies involved are 17 × 17 PWR at 3.1 wt.%, 3.25 wt.% and 3.45 wt.% enrichments, and burn-up values between 25 and 40 GWd/t.

Table 7 does not include assay data measured for other fuel types including:

- the fast reactor Phénix (TRAPU experiment) [30];
- irradiation experiment for pure isotopes (PROFILE experiment) [30];
- recent radiochemical assay experiments carried out in order to extend the current French UOX and MOX fuel database to include higher burn-up in PWR [31,32] fuel rods originating from the Cruas-II (UOX 17 × 17) and Dampierre-II reactors (MOX 17 × 17) with burn-up values up to respectively 70 GWd/t (UOX, six cycles) and 60 GWd/t (MOX, five cycles).

In France, assay data measurements are carried out by CEA. After the irradiated fuel rods are cut at the LECA, a CEA laboratory in Cadarache, the fuel samples are dissolved in nitric acid by the CEA in the COMIR or ATALANTE laboratories. The dissolved solutions are sent to CEA Saclay for radiochemical analysis. Uranium and plutonium are first separated with an ion exchange resin and are then ready for isotopic characterisation using mass spectrometry. Liquid chromatography is used to separate the minor actinides americium and curium (to eliminate interferences between masses 242 and 243), elements neodymium, cerium, promethium and samarium (interferences between masses 142, 144, 147, 148 and 150) and caesium and barium (interaction between masses 134, 135 and 137).

The isotopic content and their absolute concentrations are then measured by isotopic dilution mass-spectrometry using a thermo-ionisation-type mass spectrometer (TIMS). For Am, Cm and Cs isotopes a specific method of "total consumption" in TIMS has been developed that

**Table 7: Outline of French experimental programme for PWR fuels**

| Reactor type and assembly design | Fuel type | $^{235}$U/U (wt.%) | Pu/(U+Pu) (wt.%) | Burn-up (GWd/t) | Measured data |
|---|---|---|---|---|---|
| Bugey 3<br>PWR 17 × 17 | UOX | 2.10<br>3.10 | – | 19 to 38 | MjA, MA, Nd, Cs, BUC |
| Fessenheim 2<br>PWR 17 × 17 | UOX | 2.60 | – | 27 to 30 | MjA, MA, Nd |
| Fessenheim 2<br>PWR 17 × 17 | UOX | 3.10 | – | 45 to 60 | MjA, MA, Nd, Cs, |
| Gravelines 3+2<br>PWR 17 × 17 | UOX | 4.50 | – | 25 to 62 | MjA, MA, Nd,<br>Cs, BUC, γ-spec |
| Cruas 4/URE<br>PWR 17 × 17 | UOX<br>(from URT) | 3.61<br>$^{236}$U/$^{238}$U = 1.2 wt.% | – | 11 to 34 | MjA, MA, Nd |
| Saint Laurent B1<br>PWR 17 × 17<br>3 zones | MOX | 0.22% | ~4.5 wt.% | 10 to 45 | MjA, MA, Nd, Cs, BUC |
| Gravelines 4<br>PWR 17 × 17<br>central zone and intermediate zone | MOX | 0.22 wt.% | ~4.5 wt.% | 40 to 50 | MjA, Nd |
| La Hague<br>PWR 17 × 17<br>Fuel dissolution | UOX | 3.1<br>3.25<br>3.45 | – | 25 to 45 | MjA |

MjA = major actinides $^{234}$U, $^{235}$U, $^{236}$U, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu.

MA = minor actinides $^{237}$Np, $^{241}$Am, $^{242m}$ Am, $^{243}$Am, $^{243}$Cm, $^{244}$Cm, $^{245}$Cm, $^{246}$Cm, $^{247}$Cm.

BUC = burn-up credit nuclides $^{147}$Sm, $^{149}$Sm, $^{150}$Sm, $^{151}$Sm, $^{152}$Sm, $^{95}$Mo, $^{99}$Tc, $^{101}$Ru, $^{103}$Rh, $^{153}$Eu, $^{109}$Ag, $^{155}$Gd (plus $^{154}$Eu, $^{155}$Eu and $^{154}$Gd to check $^{155}$Gd build-up).

Nd = neodymium isotopes $^{143}$Nd, $^{144}$Nd, $^{145}$Nd, $^{146}$Nd, $^{148}$Nd, $^{150}$Nd.

Cs = caesium isotopes $^{133}$Cs, $^{134}$Cs, $^{135}$Cs, $^{137}$Cs.

γ-spec = gamma emitting radionuclides $^{154}$Eu/$^{137}$Cs, $^{106}$Ru/$^{137}$Cs, $^{134}$Cs/$^{137}$Cs, $^{144}$Ce/$^{137}$Cs.

URT = uranium de retraitement (uranium from reprocessed U).

URE = uranium de retraitement enrichi (re-enriched uranium from reprocessed U).

yields very accurate results. The isotopic dilution uses double or triple isotopic spikes such as $^{242}$Pu/$^{233}$U/$^{145}$Nd, $^{133}$Cs/$^{233}$U, $^{243}$Am/$^{233}$U, $^{248}$Cm/$^{233}$U (used for the determination of Cm/U ratios). The $^{237}$Np/$^{238}$U ratio is measured by inductively coupled plasma mass spectrometry (ICPMS) and $^{242}$Cm by alpha spectrometry.

Because the French assay measurements are funded by industrial partners such as AREVA, these data are not yet publicly available.

Extensive work has been carried out in France using integral and differential experiments to validate and qualify nuclear data evaluations of important nuclides for fuel cycle applications. This work includes irradiated fuel analysis, as well as measurements of lattice criticality, sample reactivity worth and spectral indices. As an example, $^{236}$U production in PWR fuels has been analysed using the French multi-group transport code APOLLO2 to identify direct trends and bias in the thermal capture cross-section of $^{235}$U (UOX fuels) and of its resonance integral (in MOX spectrum) [33]. Furthermore, experimental validation data have been used with perturbation studies of formation pathways using the French code DARWIN to direct the revisions of the JEF-2.2 evaluated files [34]. Trends in reaction rates and cumulative fission yields have been deduced from the trends in isotopic concentration. For example, the underestimation of the $^{238}$U (n,2n) reaction rate was highlighted by the underestimation of $^{237}$Np, and the underestimation of the $^{143}$Nd capture rate was shown by the overestimation of the $^{143}$Nd content at high burn-up. Also, the $^{148}$Nd fission yield was found to be overestimated by about 2%.

## 3.3 Germany

Post-irradiation nuclide measurement data of samples from the Obrigheim PWR from 1986 have been included in SFCOMPO (see Chapter 4). These data include 23 samples obtained from fuel rods of several assemblies [35,36]. Further review of the Obrigheim samples identified additional data, being mostly publicly unavailable up to now, and these data will be included in SFCOMPO [37-40]. These new data include samples from five uranium dioxide Obrigheim fuel assemblies at the Karlsruhe reprocessing plant with 3.13 wt.% initial enrichment and roughly 28 GWd/tHM average burn-up. The measurements include mostly uranium and plutonium isotopic compositions for fuel assemblies identified as BE 168, 170, 171, 172 and 176. Isotopic determinations were made independently by laboratories of the Institute of Transuranium Elements (ITU), the Institute of Radiochemistry (IRC), the Karlsruhe Reprocessing Plant (WAK), and the International Atomic Energy Agency (IAEA). Additional measurements of MOX samples with 2.0-3.2 wt.% initial fissile plutonium and burn-up between 10-40 GWd/tHM will also be included. The $UO_2$ measurements involve dissolved half assemblies, while the MOX data represent fuel rod samples.

Several of the German reports have been translated into English, and permissions to post these translated reports have kindly been granted by Forschungszentrum Karlsruhe and the German Federal Ministry BMU, who funded the research.

A commercial post-irradiation isotopic experimental programme for high burn-up fuel was performed in 2000 by the Association of Major Power Utilities (VGB) and designated as the "VGB Program". Two samples from 18 × 18-24 fuel assemblies irradiated in the Neckarwestheim II nuclear power plant (GKN II) were analysed by ITU in Karlsruhe, Germany. The two samples had enrichments of 3.5 and 3.8 wt.%, and burn-ups of 58.5 and 54.1 GWd/t, respectively. Extensive radiochemical analyses were performed, including isotopes of uranium, plutonium, neodymium, samarium, caesium, $^{153}Eu$, $^{155}Eu$, $^{144}Ce$, $^{147}Pm$, $^{155}Gd$, and metallic nuclides $^{95}Mo$, $^{99}Tc$, $^{101}Ru$ and $^{103}Rh$. The data from the VGB Program are not currently open to the public.

## 3.4 Japan

The Japan Atomic Energy Research Institute (JAERI), at present Japan Atomic Energy Agency (JAEA), has been conducting experimental research on fuel behaviour during irradiation and reprocessing and on nuclear criticality safety of spent nuclear fuel since the 1960s. Table 8 shows the list of assay data from JAEA, which are accessible to the public. The assay data include measurements of actinides as well as fission products from $UO_2$ and $UO_2$-$Gd_2O_3$ fuel types. Not listed in the table are several experiments that were carried out for the purpose of studying fuel behaviour and radiochemical assay measurements of fuel from the Prototype Fast Reactor (PFR) [41]. The JAEA assay data were summarised in a special session at the 2003 International Conference of Nuclear Criticality Safety held at Tokai, Japan [42].

**Table 8: JAEA experimental programmes in SFCOMPO**

| Reactor | Reactor type | Assembly design | Fuel type | Sample burn-up (GWd/t) |
|---|---|---|---|---|
| JPDR [43,44,45] | BWR | 6 × 6 | $UO_2$ | 0.1-5 |
| Tsuruga 1 [46] | BWR | 7 × 7 | $UO_2$ | 9-28 |
| Mihama 3 [47] | PWR | 15 × 15 | $UO_2$ | 7-31 |
| Genkai 1 [47] | PWR | 14 × 14 | $UO_2$ | 36 |
| Takahama 3 [48] | PWR | 17 × 17 | $UO_2$<br>$UO_2$-$Gd_2O_3$ | 14-47 |
| Fukushima-Daini 2 [48] | BWR | 8 × 8-2 | $UO_2$<br>$UO_2$-$Gd_2O_3$ | 4-44 |

SFCOMPO currently contains the nuclide composition data for BWR fuel from the Japanese JPDR ($UO_2$), Tsuruga-1 ($UO_2$), Fukushima-Daiichi-3 ($UO_2$) and Fukushima-Daini-2 ($UO_2$) [48]. Additional studies on BWR fuel have recently been completed which include isotopic composition measurements of the major heavy metal isotopes and neodymium isotopes [49,50]. Details of these studies are shown in Table 9. The fuel composition data have been compiled in a report [50] with the specifications of the cores and fuel assemblies, burn-up histories, burn-ups of the samples and comparisons with assembly burn-up calculations. These more recent BWR fuel measurements are briefly summarised below.

**Table 9: New assay data in Japan**

| | **Fukushima-Daini-2 [49]** | **Fukushima-Daini-1 [49]** | **Fukushima-Daini-1 [49]** | **Tsuruga-1 [49]** | **Name not released [51]** |
|---|---|---|---|---|---|
| Reactor type | BWR | BWR | BWR | BWR | PWR |
| Fuel type | $UO_2$ | $UO_2$ | $UO_2$ | MOX | $UO_2$-$Gd_2O_3$ |
| Assembly design | 8 × 8-4 | 9 × 9-9 | 9 × 9-7 | 8×8-2 | 17 × 17 |
| Enrichment ($^{235}$U wt.%/U) | 3.4, 4.5 | 3.4 (assembly average) | 3.4 (assembly average) | | |
| MOX (wt.%/HM) | | | | Puf: 4.64, 3.14, 2.25<br>Pu: 6.2, 4.2, 3.0 | |

### 3.4.1 $UO_2$ 8 × 8-4 fuel of Fukushima-Daini-2

Four BWR fuel assemblies, identified as 2F2D1, 2F2D2, 2F2D3 and 2F2D8, were irradiated for one, two, three and five cycles respectively from 1989 through 1997 in Fukushima-Daini-2 (2F-2), one of the BWR operated by Tokyo Electric Power Company (TEPCO) in Japan. The assembly average enrichment of these assemblies is 3.0 wt.% and the burn-ups were in the range of 13-48 GWd/t. The isotopic assay measurements were conducted by Japan Nuclear Fuel Development (NFD) as part of a fuel integrity demonstration programme of the Nuclear Power Engineering Corporation (NUPEC) [52]. A total of 46 samples were measured, the measured isotopes being $^{234,235,236,238}$U, $^{238,239,240,241,242}$Pu, $^{241,243}$Am, $^{242,243,244}$Cm and neodymium isotopes.

### 3.4.2 $UO_2$ 9 × 9-9 fuel of Fukushima-Daini-1

Two 9 × 9-9 type fuel assemblies with $UO_2$ fuel, identified as 2F1ZN2 and 2F1ZN3, were irradiated for three and five cycles respectively from 1996 through 2003 in Fukushima-Daini-1 (2F-1), one of the BWR operated by TEPCO. The average enrichment of these assemblies is 3.4 wt.% and the burn-ups of assemblies 2F1ZN2 and 2F1ZN3 were 35.6 and 53.5 GWd/t respectively. The assay measurements were sponsored by NUPEC [later taken over by the Japan Nuclear Energy Safety Organization (JNES)] and were performed at experimental facilities of the Japan Atomic Energy Agency (JAEA) [53]. A total of 11 samples were measured, and the measured isotopes included $^{234,235,236,238}$U, $^{238,239,240,241,242}$Pu, $^{241}$Am, $^{244}$Cm and neodymium isotopes. In these measurements, isotopic dilution mass spectrometry was applied to neodymium, uranium and plutonium isotopes and alpha spectrometry was used for americium and curium isotopes [54]. The sample burn-ups were evaluated by the $^{148}$Nd method taking into account the effective fission yields of $^{148}$Nd, the effective MeV/fission and the effect of the $^{147}\mathrm{Nd}(n,\gamma)^{148}\mathrm{Nd}$ and $^{148}\mathrm{Nd}(n,\gamma)^{149}\mathrm{Nd}$ reactions on the $^{148}$Nd nuclide concentrations during the irradiation, which were calculated by assembly depletion calculations performed with the SRAC code, and tracking the operating histories such as the nodal powers and the in-channel void fractions.

### 3.4.3 $UO_2$ 9 × 9-7 fuel of Fukushima-Daini-1

Two 9 × 9-7 type fuel assembles with $UO_2$ fuel, identified as 2F1Z3 and 2F1Z2, were irradiated for the same three and five cycles as for the $UO_2$ 9 × 9-9 type fuel above. The assembly average enrichment of these two assemblies is 3.4 wt.% and the burn-ups are 35.0 and 53.0 GWd/t, respectively. Isotopic composition measurements were performed for eight samples from 2F1Z3 and one sample from 2F1Z2. The isotopic composition measurements of seven samples were performed at NFD. In these measurements, isotopic dilution mass spectrometry was applied to neodymium, uranium and plutonium isotopes and alpha spectrometry to americium and curium isotopes. Benchmark calculations indicate that systematic overestimations are seen in total plutonium inventories; therefore, further study is necessary to understand the discrepancies before the data can be used for validation and/or bias determination.

### *3.4.4 MOX 8 × 8-2 fuel of Tsuruga-1*

A fuel assembly of MOX 8×8-2 type fuel, identified as JARX02, was irradiated for three cycles from 1986 through 1990, in Tsuruga-1, a BWR operated by Japan Atomic Power Company (JAPC) [55]. The isotope composition measurements were conducted recently by JNES in the facility of NFD for five MOX samples. Measured isotopes are $^{234,235,236,238}$U, $^{238,239,240,241,242}$Pu, $^{241,243}$Am, $^{242,243,244}$Cm and neodymium isotopes. In this chemical analysis, the isotopic dilution mass spectrometry was applied to neodymium, uranium and plutonium isotopes and the alpha spectrometry to americium and curium isotopes. Benchmark calculations indicate that there are relatively large discrepancies between the calculations and measurements for Pu isotopes; therefore, further study is necessary to understand the source of the discrepancies before the data should be used for code validation.

## 3.5 Spain

The programme "Experimental Measurements of the Isotopic Composition of High-enrichment and High-burn-up PWR Fuel," Spain's first programme of this kind [56], was designed to acquire isotopic assay data for spent nuclear fuel in the burn-up and enrichment ranges where the data are scarce. The programme started in 2002 and was formally finished in 2008. Its scope includes experimental measurements of the isotopic composition of nine samples cut from three different rods and analysed in two campaigns carried out in 2003 and in 2006-2007. Within the framework of a demonstration programme called High Burn-up Fuel Irradiation Programme, fuel rods with an initial $^{235}$U enrichment of 4.5 wt.%, manufactured by ENUSA in Spain, were irradiated up to a rod average burn-up of about 70 MWd/kgU in the Vandellós 2 PWR, during cycles 7-11 between June 1994 and September 2000.

The isotopic composition of the samples with burn-up values ranging from 64 to 78 MWd/kgU was determined. The isotopes measured were selected on the basis of nuclide importance for reactivity, shielding and decay heat calculations, as well as for the sample burn-up determination. The experimental methods applied were developed by Studsvik. They include fission gas analysis by mass spectrometry, gamma scanning and chemical analysis (high performance liquid chromatography, inductively coupled plasma mass spectrometry and isotope dilution analysis) [57].

The final report has recently been published in the open literature [58], and the results have been provided for inclusion in the SFCOMPO database.

A second Spanish experimental programme for the isotopic analysis of a BWR GE-14 rod, manufactured in Spain and irradiated during five cycles in the Swedish Forsmark 3 reactor, has also been performed in the Studsvik laboratory [59]. It started in 2009 and is currently in its final reporting phase. The programme includes the analysis of eight samples, taken over the length of the same mother rod, and a set of measured nuclides, selected with similar criteria but with extended scope regarding to that measured in the PWR programme. The experimental methods developed and applied by Studsvik were essentially the same as those in the PWR programme.

Final results of the programme are not yet available. In the future they will be managed in the same way as the first programme results: published in the open literature and included in the SFCOMPO database.

Table 10 summarises the PWR and BWR Spanish experimental programmes performed in the Studsvik laboratory in Sweden.

**Table 10: Summary of spent fuel PWR and BWR assay measurement in Spain**

| Reactor | Type | Measurement laboratory | Assembly design | Enrichment (wt.% $^{235}$U) | Number of samples | Burn-up (GWd/t) |
|---|---|---|---|---|---|---|
| Vandellós II | PWR | Studsvik Nuclear | 17 × 17 | 4.5 | 9 | 64-78 |
| Forsmark 3 | BWR | Studsvik Nuclear | GE-14 | 3.95 | 8 | 39-53 |

## 3.6 Sweden

In Sweden, Studsvik Nuclear AB is operating facilities for post-irradiation examination and testing of nuclear fuel and other radioactive components. The laboratories are operated by a stock corporation on commercial basis. Most of the results from experimental programmes are proprietary to the respective customers. Studsvik recently joined the MALIBU programme extension as a participating radiochemical analysis laboratory.

Studsvik Nuclear uses a broad set of standard post-irradiation examination techniques, supplemented by some sophisticated special techniques and methods. In co-operation with the Norwegian Institute for Energy Technology, Studsvik Nuclear performs ramp tests in the Halden test reactor with test rodlets refabricated in the Studsvik laboratories from commercially irradiated fuel rods. Post-test examinations of these rodlets are an important part of fuel examination work in Sweden.

During the last decade, Studsvik Nuclear has performed several campaigns of chemical nuclide analyses by means of isotope dilution analysis (IDA). An inductively coupled plasma mass spectrometer (ICPMS) equipped with a quadrupole mass analyser, in combination with a high performance liquid chromatography (HPLC) system for chemical separations, is used. The applied methods were verified by analysing a well characterised sample from a rod irradiated in the Swedish BWR Forsmark 3 [60]. All information on this sample, including fabrication data, irradiation history, nuclide analysis by two independent laboratories and results from two analyses performed by Studsvik Nuclear with two different HPLC-ICPMS systems, has been compiled in support of SFCOMPO.

Nuclide inventories of four high burn-up PWR samples are established by Studsvik Nuclear within a project of the Swedish nuclear fuel and waste management company SKB aiming at extending knowledge on spent fuel corrosion performance to higher burn-up. The results will be published in an SKB technical report.

## 3.7 Switzerland

Switzerland has carried out several well-publicised experimental programmes related to burn-up credit at the Paul Scherrer Institute (PSI). PSI is one of several laboratories that performed analyses for the Belgian ARIANE and MALIBU programmes. Furthermore, they have analysed a number of irradiated fuel sample within bilateral and multilateral commercial programmes. The scope of the isotopic measurements performed under the Swiss programmes, as known by the expert group at this time, is briefly summarised.

The LWR-PROTEUS experimental programme [61] was performed at PSI for the purposes of providing experimental data for validation of computer codes and reactor physics parameters. The isotopic assay phases of the programme have involved extensive measurements on spent fuel from the Leibstadt BWR and Gosgen PWR nuclear power plants operated in Switzerland. Under Phase II of the programme, a total of 13 fuel samples were destructively measured; 11 PWR samples and 2 BWR samples [62]. A total of 17 actinides and 40 fission products were measured in the LWR-PROTEUS experimental programme by PSI. The LWR-PROTEUS programme is commercial and data are not currently open to the public.

### 3.8 United Kingdom

Britain has an active history of performing spent fuel assay experiments and validation using the UK code FISPIN [63]. Recent British-funded work has been performed at the Karlsruhe Research Centre, Forschungszentrum Karlsruhe (FZK), in Germany, although there are plans to restart spent fuel experiments at the UK National Nuclear Laboratory (formerly Nexia Solutions Ltd.).

Within the UK National Nuclear Laboratory a large database of measurements has been collected over the last 30 years for UK nuclear fuels (mostly from Magnox and advanced gas-cooled reactors), and also including some BWR and PWR data. The database has comparisons with calculations using JEF-2.2 based libraries and the WIMS, TRAIL and FISPIN10 code route; see for example Ref. [62]. This report and the underlying UK data have been contributed to the EGADSNF. Table 11 gives details of some isotopic assay experiments performed on AGR, Magnox and PWR fuels.

**Table 11: Parameters of reactors and fuel types for assay data from UK programmes**

| Reactor type | Fuel type | No. of samples | Enrichment (wt.% $^{235}$U) | Sample burn-up (GWd/t) | Measurement data |
|---|---|---|---|---|---|
| AGR (Hinkley R4) | UOX | 15 | 1.542-2.5 | 1.1-22.8 | MjA, Cm, $^{241}$Am, $^{125}$Sb, $^{144}$Ce, Cs, Eu, Nd |
| AGR (Hinkley R3) | UOX | 12 | 2.012-2.55 | 7.8-26.0 | MjA, $^{241}$Am, Cm, $^{95}$Nb, $^{125}$Sb, $^{144}$Ce, Cs, Eu, Nd |
| AGR (Hunterston R3) | UOX | 2 | 2.012 | 13.5-16.3 | MjA, $^{241}$Am, Cm, Nd |
| Magnox (Hunterston A) | U metal | 5 | 0.711 | 3.3-8.9 | MjA, $^{241}$Am, Cm, $^{95}$Nb, $^{95}$Zr, $^{144}$Ce, $^{144}$Pr, Cs, Nd |
| Magnox (Bradwell) | U metal | 1 | 0.711 | 8.9 | MjA, $^{241}$Am, Cm, $^{95}$Nb, $^{95}$Zr, $^{144}$Ce, $^{144}$Pr, Cs, Nd |
| PWR (Beznau M501) | MOX | 4 | 4.019-5.561 (Pu wt.% HM) | 22.4-39.0 | MjA, $^{237}$Np, $^{241,243}$Am, $^{244,245}$Cm, $^{90}$Sr, $^{103}$Rh, $^{101,106}$Ru, $^{129}$I, $^{95}$Mo, $^{99}$Tc, $^{144}$Ce, $^{147}$Pm, $^{155}$Gd, Cs, $^{135}$Cs $^{147-152,154}$Sm, Nd |
| PWR (Beznau M502) | MOX | 2 | 3.72-5.54 (Pu wt.% HM) | 44.4-63.2 | MjA, Xe+Kr, Nd |

MjA = major actinides $^{234}$U, $^{235}$U, $^{236}$U, $^{238}$U, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu.

Cm = curium isotopes $^{242}$Cm and $^{244}$Cm.

Cs = caesium isotopes $^{134}$Cs and $^{137}$Cs.

Nd = neodymium isotopes $^{142-146,148,150}$Nd.

Eu = europium isotopes $^{154}$Eu and $^{155}$Eu.

Xe+Kr = noble gases $^{80,82-86}$Kr and $^{128-132,134,136}$Xe.

In 1991 AEA Technology (UK) and the CEA (France) initiated a programme of experiments and analyses known as the CERES collaboration [64]. The main aim of the programme was to provide experimental data to validate calculation methods and nuclear data used in criticality assessments involving burn-up credit calculations. Phase I of the programme involved reactivity measurements on irradiated PWR fuel samples in the MINERVE reactor at Cadarache and in the DIMPLE reactor at Winfrith. In Phase II reactivity measurements were made on a set of fission product samples to provide validation of the neutron absorption cross-section data for 13 of the major fission products relevant to burn-up credit. Phase III, the final part of the programme extended the collaboration to include the participation of the United States through Sandia National Laboratories. In addition, destructive chemical analysis on selected PWR, BWR, CAGR and MOX fuel samples was carried out at Harwell to determine the concentration of the following fission product nuclides: $^{95}$Mo, $^{99}$Tc, $^{101}$Ru, $^{103}$Rh, $^{109}$Ag, $^{133}$Cs, $^{143}$Nd, $^{145}$Nd, $^{147}$Sm, $^{149}$Sm, $^{150}$Sm, $^{153}$Eu and $^{155}$Gd.

Initial attempts to separate some of the fission products were unsuccessful and new methods were developed during the programme. Tests of the methods on a well characterised sample of fuel from the Beznau reactor confirmed the success of the new approach. Typical accuracy (one standard deviation) of measured fission product concentrations is around the 2% level. Some of the CERES assay measurement data were recently approved for release to the public.

## 3.9 United States

The United States has carried out a number of experimental programmes designed to acquire radiochemical isotopic assay data for spent nuclear fuel. Several of these experiments are included in the SFCOMPO database, as follows:

- Calvert Cliffs 1, 14 × 14 PWR;
- H.B. Robinson 2, 15 × 15 PWR;
- Cooper, 8 × 8 BWR;
- Monticello, 8 × 8 BWR.

The experiments for LWR uranium oxide fuel that have been evaluated are summarised in Table 12. One of the most comprehensive programmes was performed by the US DOE Office of Civilian Radioactive Waste Management (OCRWM), known as the Approved Testing Material (ATM) programme. The programme included measurements of reactor fuel from the Calvert Cliffs [65-67] (designated ATM-103, -104 and -106) and H.B. Robinson [68] (ATM-101) PWR reactors, and Cooper BWR reactor [69,70] (ATM-105 and -108). The Calvert Cliffs ATM-104 data were used as the basis for the EGBUC Phase I-B burn-up credit benchmark [6].

Radiochemical analysis measurements for ATM fuels were performed for the most part at Pacific Northwest National Laboratory (PNNL), with some mass spectrometry cross-check measurements made at LANL. The nuclides measured at PNNL included uranium and plutonium, and the specific nuclides $^{79}$Se, $^{90}$Sr, $^{99}$Tc, $^{126}$Sn, $^{135}$Cs, $^{137}$Cs, $^{237}$Np, $^{241}$Am and $^{242+244}$Cm, with sample burn-up determined by $^{148}$Nd measurements. The Calvert Cliffs ATM-104 data included additional lanthanide measurements to support fission product nuclide validation for burn-up credit. The lanthanide measurements were performed by PNNL without radiochemical separation prior to mass spectrometry measurements, which limited the use of some data for isotopic validation due to isobar interference. Lanthanide measurements were made independently at the Khlopin Radium Institute (KRI), Russia, using radiochemical separations [71]. These later measurements are documented separately and provide isotopic data and absolute concentrations for neodymium, samarium, europium and gadolinium [72]. The addition of the KRI lanthanide measurements for the Calvert Cliffs samples to SFCOMPO is planned.

Other published isotopic measurements performed in the United States that have been used for code validation include Turkey Point reactor fuel measured at Battelle Columbus laboratories in 1980 as part of the US DOE waste management programme [73]. Assay measurements were

**Table 12: List of LWR fuels measured in the United States**

| Reactor | Fuel design | ATM[a] designation | Fuel type | Enrichment (wt.% $^{235}U$) | No. of samples | Burn-up (MWd/kgU) |
|---|---|---|---|---|---|---|
| Cooper[c] | 7 × 7 | 105 | $UO_2$ | 2.94 | 6 | 18-34 |
| | | 108 | $UO_2$-$Gd_2O_2$ | 2.94 | 3 | 12-27 |
| Monticello[c] | 8 × 8 | – | $UO_2$<br>$UO_2$-$Gd_2O_2$ | 1.45, 1.87, 2.14, 2.87 | 25<br>5 | 33-59<br>39-52 |
| Limerick | 9 × 9 | – | $UO_2$-$Gd_2O_2$<br>$UO_2$ | 3.60<br>3.95 | 2<br>6 | 37 and 55<br>57-66 |
| Three Mile Island-1 | 15 × 15 | – | $UO_2$ | 4.013 | 11 | 44.8-55.7 |
| | | – | $UO_2$ | 4.657 | 8 | 22.8-29.9 |
| Calvert Cliffs 1[c] | 14 × 14 | 104 | $UO_2$ | 3.038 | 3 | 27.4-44.3 |
| | | 106 | $UO_2$ | 2.453 | 3 | 31.4-46.5 |
| | | 103 | $UO_2$ | 2.72 | 3 | 18.7-33.2 |
| Robinson 2[c] | 15 × 15 | 101 | $UO_2$ | 2.561 | 6 | 16.0-31.7 |
| Turkey Point | 15 × 15 | – | $UO_2$ | 2.556 | 5[b] | 30.5-31.6 |
| Yankee | 18 × 18 | – | $UO_2$ | 3.4 | 8[b] | 15.9-36.0 |

[a] ATM = Approved Testing Material programme designation.

[b] Additional samples were measured that have not been evaluated recently.

[c] Included in SFCOMPO.

made for five samples obtained from three different fuel rods from assemblies D01 and D04. Measurements were limited to the uranium and plutonium isotopes, and $^{148}$Nd to experimentally determine the sample burn-up. Additional measurements on another Turkey Point fuel assembly have also been reported [74] but have not been widely used for actinide isotopic validation to the knowledge of the authors.

Extensive isotopic measurements for more than 50 samples have been reported for fuel from the Yankee Nuclear Power Station under the Westinghouse Commercial Atomic Power Project. Measurement laboratories included General Electric (GE) Vallecitos, Westinghouse and New Brunswick laboratories [75,76] with some samples having laboratory cross-check measurements. The measurements, performed during the 1960s, included uranium and plutonium isotopes, $^{137}$Cs, $^{90}$Sr and $^{148}$Nd for most samples. An extension programme included additional measurements for $^{232}$U, $^{236}$Pu, $^{237}$Np, $^{242m}$Am, $^{243}$Am, $^{242}$Cm and $^{244}$Cm for a reduced set of samples. Detailed operational data for the assemblies, however, is not available and control rod insertion, used in this core, is not well documented, limiting the application of the data to code validation. Within the United States, use of the data has been limited to evaluation of several samples irradiated for three cycles [77].

The Turkey Point and Yankee reactor measurements are not currently part of the SFCOMPO database.

### 3.9.1 *Three Mile Island-1 PWR measurements*

More recently, assay measurements for more modern fuels from the Three Mile Island-l (TMI-1) B&W 15 × 15 PWR plant were carried out under a programme of the OCRWM Yucca Mountain Project (YMP). The measurements, performed circa 2000, were designed specifically to support burn-up credit validation and therefore included more extensive fission product measurements than in earlier programmes. GE Vallecitos Nuclear Center measured 11 samples from 3 fuel rods of a one-cycle irradiated assembly NJ070G [78,79]. These data include high-precision TIMS measurements for most nuclides of interest to burn-up credit, except $^{133}$Cs and metallic fission products ($^{95}$Mo, $^{99}$Tc, $^{101}$Ru, $^{103}$Rh, $^{109}$Ag). Additional measurements were made for 8 samples from one rod of two-cycle irradiated assembly NJ05YU at Argonne National Laboratory (ANL) [80]. The

ANL measurements were made using lower-precision ICPMS methods and include most nuclides of interest to burn-up credit including the metallic fission products, but also excluded $^{133}$Cs. The TMI-1 data are being considered for addition to SFCOMPO.

### *3.9.2 Limerick BWR measurements*

The Limerick measurements were made on GE 11 9 × 9 BWR fuel under the YMP [81]. Radiochemical analysis measurements were performed at GE Vallecitos Nuclear Center for eight samples from three fuel rods of assembly YJ1433 and include data for metallic fission products. Although these measurements are open to the public, details of the GE 11 assembly design currently remain commercially protected.

### *3.9.3 Calvert Cliffs*

At this time, an experimental programme under OCRWM YMP is planning to perform additional measurements on archived fuel samples from the ATM programme, including ATM-104, ATM-106 and ATM-105. The re-analysis of the samples is designed to provide a comprehensive set of measurements for all actinides and fission products of interest to support burn-up credit for post-closure criticality safety analyses.

## 3.10 Countries using VVER-440

Five EU member countries are operating VVER: Bulgaria, Czech Republic, Finland, Hungary and Slovakia. Spent VVER fuel is also stored in Germany. No country of those operating VVER mentioned above has hot cell laboratories with the capability to cut the samples from spent fuel for the purposes of performing radiochemical assay measurements. As non-proliferation regulations restrict spent fuel from being transported across their borders, the Russian hot cell laboratories are currently the only facilities which can acquire isotopic assay data for VVER fuels. Unfortunately, there is a lack of unrestricted data in countries operating VVER reactors [82]. Due to this, VVER-440 measurements were recently performed at the Research Institute of Atomic Reactors (RIAR) in Dimitrovgrad, Russia, under ISTC project #2670 [22]. This experimental project has become a very important event initiating further activities in this field.

In the VVER #2670 ISTC project completed in 2005, the following nuclides were measured: $^{234,235,236,238}$U, $^{238,239,240,241,242}$Pu, $^{241,242m,243}$Am, $^{244,245,246}$Cm, $^{142,143,144,145,146,148,150}$Nd, $^{133,134,135,137}$Cs, $^{140,142,144}$Ce, $^{147,148,149,150,151,152,154}$Sm, $^{151,153,154,155}$Eu, $^{95}$Mo, $^{99}$Tc, $^{101}$Ru, $^{105,108}$Pd, $^{109}$Ag and $^{155}$Gd. Eight samples of four fuel pins taken from a VVER-440 fuel assembly of the Russian KOLA nuclear power plant had enrichment of 4.4 wt.% and burn-up between 23 and 46 MWd/kgU. The measured data are available to be added to SFCOMPO. Based on the #2670 data an international benchmark has been specified [16] and the results are being evaluated [83].

In 2007, 10 institutions of nuclear industry, nuclear regulation and nuclear research from countries operating VVER-440 reactors in the EU established an international consortium to fund the radiochemical assay of VVER-440 fuel which should be performed in a hot cell laboratory in Russia. The consortium members are:

- Bulgaria: Institute for Nuclear Research and Nuclear Energy (INRNE) and Kozloduy nuclear power plant;
- Czech Republic: Nuclear Research Institute Rez (NRI) and Skoda JS, a nuclear company;
- Finland: Fortum Corporation and VTT Technical Research Centre;
- Hungary: Atomic Energy Research Institute (KFKI AERI);
- Slovak Republic: Nuclear Regulatory Authority of the Slovak Republic (UJD SR); and JAVYS a.s. and VUJE a.s., two nuclear companies.

The selected hot cell laboratory is at the Research Institute of Atomic Reactors (RIAR) in Dimitrovgrad; for more details the RIAR website (*www.niiar.ru*) or the IAEA webpage "Integrated Nuclear Fuel Cycle Information Systems" (*www-nfcis.iaea.org*) can be consulted. The measurements will be made on 12 samples cut out from 3 pins of a VVER-440 fuel assembly with flat 4.4% $^{235}U$ enrichment. The samples have a range of burn-ups from about 30-60 MWd/kgU. The measured nuclides will be the actinides and fission products with high importance to burn-up credit; the list of the other nuclides is currently under negotiation. The new VVER isotopic assay measurements will be performed under a new ISTC project #3958 in 2009-2010 [84]. The latest news concerning the project can be found on the VVER-400 consortium webpage (*www.nri.cz*).

A comprehensive summary of earlier VVER measurements performed in Russia at KRI has recently been published by Makarova, *et al*. [85]. These measurements involve older VVER programmes carried out from 1977 to 2001 and include isotopic data mainly for the actinides. Data are included for 41 samples from VVER-440 and VVER-1000 reactors.

### 3.11 Countries using RBMK reactors

RBMK reactors are currently operated in Russia, the Ukraine and Lithuania, all non-OECD member countries. Consequently measured isotopic assay data for RBMK fuels are not included in the present spent fuel database. However, many countries have an interest in validating their computer codes and models for graphite-moderated systems being considered for next-generation reactor designs. Therefore, it is hoped in the future that these data will be integrated into the database for wider dissemination. A report documenting VVER and RBMK spent fuel assay measurements performed at the Khlopin Radium Institute and published by Makarova, *et al*. [84] includes measurements for the actinides and burn-up indicator fission products for 41 RBMK samples. Benchmark analyses using these assay data for 15 samples have been reported [86].

# Chapter 4: SFCOMPO – a database for spent fuel isotopic compositions

The OECD/NEA currently maintains an extensive electronic database of measured spent fuel composition (SFCOMPO) data that are available to the public. The first version of SFCOMPO was established in the late 1990s by the Japan Atomic Energy Agency (JAEA) to compile and make nuclide validation data accessible for use by the criticality safety community [87-90]. The Expert Group on Burn-up Credit Criticality Safety (EGBUC) recognised the importance of having a centralised framework for the collection and dissemination of assay data from NEA member countries, and in 2002, SFCOMPO was transferred to the NEA Data Bank where it is currently maintained.

## 4.1 Current status of the SFCOMPO database

Table 13 summarises the current status of the database. It contains measured composition data from 14 reactors (7 PWR and 7 BWR) operated in 4 countries. Data from 246 samples are available, including 30 samples from fuels that included burnable absorbers ($UO_2$-$Gd_2O_3$). The database includes measurements of absolute nuclide content and isotopic ratios.

**Table 13: Spent fuel isotopic assay data in SFCOMPO**

| Reactor | Country | Reactor type | Assembly design | Fuel type | Total number of samples |
|---|---|---|---|---|---|
| Obrigheim | Germany | PWR | 14 × 14 | $UO_2$ | 23 |
| Gundremmingen | Germany | BWR | 6 × 6 | $UO_2$ | 12 |
| Trino Vercellese | Italy | PWR | 15 × 15 | $UO_2$ | 39 |
| JPDR | Japan | BWR | 6 × 6 | $UO_2$ | 30 |
| Tsuruga-1 | Japan | BWR | 7 × 7 | $UO_2$ | 10 |
| Fukushima-Daiichi-3 | Japan | BWR | 8 × 8 | $UO_2$,$UO_2$-$Gd_2O_3$ | 36 (10)[a] |
| Fukushima-Daini-2 | Japan | BWR | 8 × 8 | $UO_2$,$UO_2$-$Gd_2O_3$ | 18 (10)[a] |
| Mihama-3 | Japan | PWR | 15 × 15 | $UO_2$ | 9 |
| Genakai-1 | Japan | PWR | 14 × 14 | $UO_2$ | 2 |
| Takahama-3 | Japan | PWR | 17 × 17 | $UO_2$,$UO_2$-$Gd_2O_3$ | 16 (5)[a] |
| Cooper | USA | BWR | 7 × 7 | $UO_2$ | 6 |
| Monticello | USA | BWR | 8 × 8 | $UO_2$,$UO_2$-$Gd_2O_3$ | 30 (5)[a] |
| Calvert Cliffs No.1 | USA | PWR | 14 × 14 | $UO_2$ | 9 |
| H.B. Robinson Unit 2 | USA | PWR | 15 × 15 | $UO_2$ | 6 |

[a] The number in parentheses is the number of samples of $UO_2$-$Gd_2O_3$.

In general the measurement data for the fission products are limited. This limitation is especially true for the large absorber fission products important to burn-up credit. The database provides isotopic data for mainly the uranium, plutonium, americium, curium actinides and several fission products (neodymium, caesium, strontium) used predominantly to determine the burn-up. The experiments with the most complete fission product measurements currently in SFCOMPO are for Takahama PWR fuel samples, and the samples measured as part of the Approved Test Materials (ATM) programme.

Other limitations of the current database include: *i)* relatively few data available for high enrichment and high burn-up fuels, and no data above 60 GWd/t; *ii)* no MOX fuel data or data for non-LWR reactor types; *iii)* lack of data for many very long-lived radioactive fission products; *iv)* incomplete documentation of irradiation conditions, particularly for BWR fuels which have incomplete void history information; *v)* lack of measurement uncertainty information in the database; *vi)* difficulty accessing primary experimental reports to obtain additional information. These areas have been identified by the EGADSNF for future improvement.

### 4.2 New assay data available to the NEA database

One of the main objectives of the EGADSNF is to obtain new assay data that are not yet open to the public from NEA member countries, or data that are published but not widely available. Additional data, as listed in Table 14, have been compiled through the voluntary contribution from expert group members and will be added to the SFCOMPO database. Details of the contributed data are provided in the NEA member country reports with references to the experimental programmes.

The addition of new data to SFCOMPO has not been initiated at this time in order to first update the formats of the electronic database to include more complete information and expanded capability. In the interim, the experimental reports that have been compiled have been made available to users through the EGADSNF webpage [91].

### 4.3 Revision of database

Improvements to the electronic database are currently being studied, including modifications to the formats to allow a more detailed design and operating history data to be included, adding information on the experimental uncertainties which is not now included, and improving database searching options. Providing measurement uncertainties with the data is seen as a high priority task for the Expert Group for use in evaluating differences between the measurement data and code calculations. Efforts to improve the database formats are ongoing.

Another important improvement to the database will be providing users direct access to the primary references for the experimental programmes. Because the experimental data currently in SFCOMPO have not been independently evaluated or peer reviewed to date, it is often necessary for the user to access the primary experimental reports to identify the measurement techniques, accuracies, and other details of the experiment and irradiation history in order to properly interpret and apply the data. These reports can be difficult to access due to the age of some of the experiments. The EGADSNF is currently working to compile the primary references and make these reports available through the Data Bank as part of SFCOMPO, subject to receiving approval from the publishers. To date, the primary references for many of the experiments in SFCOMPO have been scanned and compiled as electronic documents. All future additions to the database will include electronic versions of the supporting references that can be accessed by users directly from the EGADSNF website.

### 4.4 Evaluation of experimental data

Currently, data published by the laboratories are included in the database as reported, without peer review. There are plans within the Expert Group to implement independent peer review of the data to assess the quality of the measurements, identify potential problems with data as determined through inconsistencies in the data (e.g. using parent-daughter relationships) and through comparisons of measurement data for other similar fuel types. In addition, the review

**Table 14: List of new isotopic assay data to be included in SFCOMPO**

| Country of reactor | Reactor name | Reactor type | Measurement laboratory | Assembly design | No. of samples | $^{235}$U enrichment (wt.%) | Burn-up (GWd/t) |
|---|---|---|---|---|---|---|---|
| Germany | Obrigheim | PWR | ITU/IAEA/WAK/IRC | 14 × 14 | 5[a] | 3.13 | 27-29 |
| | Obrigheim | PWR | ITU | 14 × 14 | 1 | 3.2 Pu-fis[b] | 37 |
| Japan | Fukushima-Daini-2 | BWR | NFD | 8 × 8-4 | 46 | 3.0 (assembly average) | 9-64 |
| | Fukushima-Daini-1 | BWR | JAEA | 9 × 9-9 | 11 | 3.4 (assembly average) | 36-68 |
| | | | NFD | 9 × 9-7 | 6 | 3.4 (assembly average) | 46-69 |
| | Tsuruga-1 | BWR | NFD | 8 × 8-2 | 5 | 3.1/4.6 Pu[c] | 30-39 |
| | Not yet public | PWR | JAEA | 17 × 17 | 5 | 3.2 | 22-39 |
| Netherlands | Dodewaard (ARIANE) | BWR | SCK•CEN/PSI | 6 × 6 | 5 | 4.9/6.4 Pu[c] | 34-56 |
| Spain | Vandellós | PWR | Studsvik Nuclear | 17 × 17 | 9 | 4.5 | 43-74 |
| Sweden | Forsmark 3 | BWR | Studsvik Nuclear, Dimitrovgrad, Harwell | SVEA-100 | 1 | 4 | 60 |
| Switzerland | Beznau-1 (ARIANE) | PWR | SCK•CEN/PSI | 14 × 14 | 3 | 4.3-6.01 Pu[d] | 40-59 |
| | Goesgen (ARIANE) | | ITU/SCK•CEN | 15 × 15 | 3 | 3.4/4.1 | 29-60 |
| | Beznau-1 (UK) | | ITU/ Karlsruhe | 14 × 14 | 6 | 3.7-5.5 Pu | 22-63 |
| UK | Hinkley R3/R4 | AGR[e] | AERE Harwell/ AEE Winfrith | Annular | 27 | 1.54-2.55 | 1-26 |
| | Hunterston | AGR | AERE Harwell/ AEE Winfrith | Annular | 2 | 2.0 | 13-16 |
| | Hunterston | Magnox | AERE Harwell | $^{nat}$U metal fuel rod | 5 | 0.711 | 3-9 |
| | Bradwell | Magnox | AERE Harwell | $^{nat}$U metal fuel rod | 1 | 0.711 | 9 |
| USA | TMI-1 | PWR | GE-Vallecitos | 15×15 | 8 | 4.7 | 23-30 |
| | | | Argonne | | 11 | 4 | 45-56 |
| Russia | Novovoronezh (ISTC-2670) | VVER-440 | RIAR | Hexagonal | 8 | 3.6 | 39 |

[a] Five reprocessed fuel assemblies, measured in two batches each (batches averaged).

[b] Pu fissile.

[c] Pu/(U+Pu).

[d] (Pu+Am)/(U+Pu+Am).

[e] The AGR assembly consists of 3 rings of pins with 6 (inner), 12 (middle) and 18 (outer) in each ring.

would evaluate the completeness of the documentation and identify sources of data and methodologies that may be appropriate to provide supplemental data when data are not available from the experimental reports.

The task of actually compiling and applying the experimental data for code validation can be very time consuming and frequently requires the user to search the primary reports to confirm details of the experiment and verify reported data. In some cases data reported in primary references have been identified as being suspect due to large or unrealistically small measurement uncertainties, possible measurement errors, data corrections performed by the laboratory or other reporting errors. The task of reviewing the experimental data could be done by an evaluation working group, so that any organisation using the data would be assured that the data have already been reviewed and vetted for accuracy, rather than having each organisation using the data perform such a review independently.

The outcome of the evaluation process would be a benchmark report containing consensus data recommended for use in validating computational systems and nuclear data. As part of this process, the reported experimental uncertainties would be reviewed and more realistic values would be assigned if the reported values were unrealistically small or otherwise inconsistent with the observed variance of the measurements. The process would also likely result in some data sets not being recommended for the purpose of code validation due to experimental problems or incomplete documentation. The peer review of experiments has been identified by the EGADSNF as a priority task for the future development and improvement of SFCOMPO. However, this would require significant efforts on the part of countries with experience in radiochemical analysis, computational modelling and simulation, and data analysis. Initial efforts to evaluate several experimental data sets have started. This work will also provide a roadmap for performing future data review activities and also identify the scope of the effort.

# Chapter 5: Analytical methods for spent fuel assay data

The aim of this chapter is to give a concise overview of state-of-the-art methods and techniques used in the (destructive) post-irradiation examinations (PIE) for the analysis of the isotopic composition and concentrations in spent nuclear fuel samples. This section is designed to familiarise a non-analytical expert with the analytical work involved in spent fuel radiochemical analysis and the results to be expected in terms of accuracy, sensitivity and reliability using current techniques to measure different nuclides. The overview of the complete analytical process includes the following steps:

- sampling and sample dissolution;
- separations;
- measurement techniques;
- additional experimental information.

Each step is briefly discussed, explaining its purpose, the different possible methodologies available, the basic principles of the techniques, the uncertainty one can expect from each step, and the main sources of these uncertainties. The text is the joint effort of several radiochemical laboratories with world-renowned experience in radiochemical analysis research. However, it is impossible to treat in this report all possible isotopes of interest and all possible experimental techniques, particularly considering the advances in instrumental technology each year. In that sense, no claim of completeness is made.

At the end of the section, a concise table listing the available measurement techniques for the most important nuclides of interest to support typical spent fuel research is given. Although this section concentrates on the analysis of $UO_2$ and MOX fuels, the general aspects of the different methodologies and measurement techniques are in most cases also applicable to other fuel types.

Before starting any analysis the requirements for the data (measured nuclides and precision of measured content) should be clearly established so that one can be sure that the results will meet the objectives of the project or application. Unfortunately, this is not the only criterion that will determine the analytical procedure to follow; as in most cases budgetary constraints need to be considered. In general, it is the experience of this group that costs rise exponentially, rather than linearly, as the accuracy and precision of the measurement data improves.

Some more detailed information on the topic of radiochemical analysis methodologies for spent fuel, sources of uncertainty and best practices based on previous experience can be found in open literature [92].

## 5.1 Analytical techniques and procedures

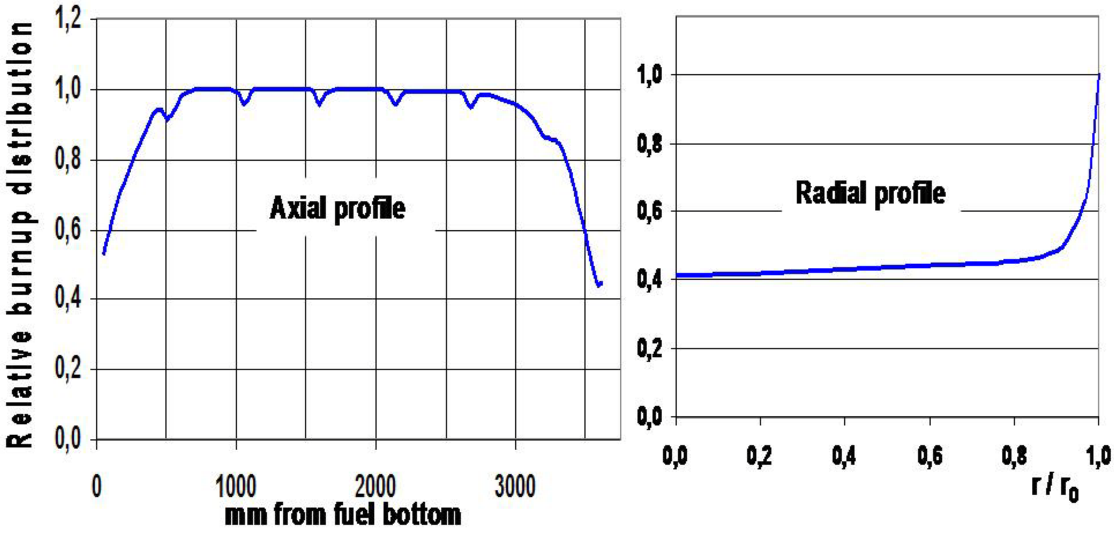
Destructive chemical and radiochemical analysis remains the most reliable analytical approach for the creation of a nuclear database on the isotope vectors and nuclide concentrations in irradiated fuels. This type of analysis can only be performed at specialised laboratories with access to specialised hot cell infrastructure for chemical sample preparation and to specialised radio-analytical and mass-spectrometric techniques for the analysis.

The present section gives a concise description of the analytical methodologies applied for spent fuel characterisation through radiochemistry, which includes sampling procedures, dissolution of the irradiated fuel in a hot cell, chemical separations methods and analytical and radio-analytical measurement procedures.

### 5.2 Sampling and dissolution of the fuel sample

Every successful analysis starts with the selection and sampling of a representative specimen. When selecting spent fuel samples for destructive chemical and radiochemical analyses there are a few considerations that should be made. Irradiated commercial reactor fuel rods have a burn-up gradient over the total length of the rod. Also internally within the pellet itself, the burn-up shows a significant radial gradient (Figure 2) that depends on the fuel design and reactor operating history. For LWR fuel rods, smaller burn-up changes are observed at locations corresponding to the position of the assembly grid structures. In addition, part of the volatile fission products tend to migrate in the fuel pellet during irradiation due to the high temperature gradient in the fuel and can accumulate in the fuel-cladding gap and in the inter-pellet zones. Therefore the fuel has to be considered as an inhomogeneous matrix. This should be kept in mind when developing a sampling strategy for burn-up determination or for a representative spent fuel characterisation.

**Figure 2: Example of axial and radial burn-up distributions**



Before the selection of a representative sample for analysis, in general a radiometric gamma-scanning is performed on the "mother" fuel rod to determine the axial activity distribution, as well as the fuel pellet boundaries and the assembly grids positioning. Note that the total activity profile only approximates the burn-up profile (depending on the time elapsed from reactor discharge); the burn-up profile is closely represented by $^{137}$Cs spectrometry. The actual sampling procedure is generally based on this activity and local burn-up distribution.

Ideally, a large enough sample is taken that includes pellets as well as pellet gaps. For example for the ARIANE programme, three pellet specimens were taken. These samples were cut at mid-pellet, thus including not only two complete and two halves of pellets but also three inter-pellet zones. If it is not possible to take such a large sample, special care has to be taken to obtain a representative sample, taking into account the possible impact of the inhomogeneous nature of the fuel.

Cladding should be included in the dissolution of the fuel sample, as cladding will contain some relevant fraction of fission products. Mechanical de-cladding is generally not recommended, though it would be of reduced impact for low burn-up samples operated at low power (open fuel-cladding gap during in-reactor operation).

There are different methods available for the dissolution of irradiated uranium and plutonium oxide fuels. However, for analysis purposes the most commonly applied procedures use nitric acid and mixtures of nitric acid and hydrofluoric acid. Whenever possible, dissolution of the cladding should be avoided as this will only put a burden on the actual radiochemistry and analysis. Therefore for the dissolution of commercial LWR fuels, in general, a multi-step procedure is preferred. In a first step, fuel is dissolved in nitric acid (8 to 10 *M*) heated near boiling point. In this medium the uranium matrix dissolves readily while the cladding material itself remains unaffected and can be easily removed from the solution afterwards. Besides the uranium fuel matrix, any fuel remnants on the cladding wall and the majority of the fission products and minor actinides will go into solution. The nitric acid solution is then filtered and the residue that is left is treated in a second step using a mixture of nitric acid (~8 *M*) and hydrofluoric acid (0.1 *M*) to ensure that all plutonium oxide is dissolved. Both resulting solutions are recombined into one solution for analysis purposes.

During the first dissolution step, the majority of the iodine species present in the fuel is volatilised in the acidic medium and is thus lost for analysis. In case an analysis of $^{129}$I is required, the off-gasses should be fed through an alkaline solution such as for instance sodium hydroxide solution to collect the iodine. Carrier iodide (KI or NaI) is added to the fuel solution before the dissolution is started to facilitate the complete volatilisation of iodine.

For a straightforward burn-up determination the dissolution procedure stops here as the major actinides and suitable fission product monitors such as Nd and La isotopes or other lanthanides, $^{137}$Cs and $^{144}$Ce, will be dissolved. However after the second dissolution step, there is still part of the metallic fission products, such as Mo, Tc, Ru, Rh, Pd, Ag and Sb isotopes, which remain as an insoluble residue. For a more extensive fuel characterisation this residue needs to be further treated by either dissolution of the residue in a suitably strong acid mixture in a closed vessel system or by a fusion method using liquid salts at elevated temperatures. In general, the solutions obtained from the metallic residue dissolution are analysed separately from the spent fuel solution and the results from both solutions are combined for reporting.

In some MOX fuels, plutonium rich clumps can be formed during fuel manufacture that are highly insoluble and have significantly higher burn-ups that the surrounding fuel. To properly assay such fuel it is therefore important to fully dissolve these nodules.

After completion of the dissolution process of the irradiated fuel, an aliquot of the obtained solution is further diluted in the hot cell, typically with 1 *M* nitric acid, to reduce the dose rate of the fuel solution to a level that a small aliquot can be transferred to the laboratory for sample preparation and analysis. Dilutions are preferentially performed by means of weighing as this will have a lower related uncertainty than volumetric dilutions.

The stability of the solutions obtained with the described methodology has shown to be very acceptable. When the fuel concentration is not too high (≤0.1 g/mL) and the solutions are stored in tightly sealed glass containers, the solutions can be kept over several months.

The sample preparation performed in the hot cell may have an important contribution to the overall uncertainty of the final results reported for the different isotopes of actinides and fission products. There are the uncertainties in the hot cell from the applied dissolution strategy, from incidental losses of material during manipulations, from weighing and so on. Traditionally in chemical analysis an uncertainty of an analysis methodology will be tested using a reference material, thus allowing a validation of the uncertainty of the sample preparation and measurement. However no such reference material exists for spent fuels. As a result, the uncertainties associated with operations performed in the hot cell are not easy to evaluate. A best estimate of uncertainty will have to be made by the expert of the analysis laboratory, based on knowledge and experience. Alternatively, cross-check measurements on replicate samples by independent laboratories have been used by some large international programmes to confirm the measurement accuracies.

### 5.3 Separation techniques and procedures

Chromatographic separations are essential to determine the isotope compositions and concentrations of the actinides and main fission products (Nd, Cs, Sm, Eu, Gd). Separation procedures can be performed off-line or on-line. Off-line separations allow the recovery of pure fractions of the elements to be analysed by mass spectrometric or radiometric techniques. In on-line separation procedures, the chromatographic technique is directly coupled to the measurement technique, which is usually a mass spectrometric technique like inductively coupled plasma mass spectrometry (ICPMS). Measuring on-line allows a rapid detection and determination of the nuclides of concern, but as transient and not perfectly stable signals are being measured, this method is normally less precise as compared to off-line measurements.

For most off-line separations, it is advisable to first isolate uranium and plutonium because of their high concentrations. This is usually done by gravitational chromatography in an open column packed with ion exchange resin in a nitric acid medium. The uranium and plutonium ions bind to the resin in the form of anionic complexes, while the other elements are eluted. The uranium is then separated from the plutonium by elution with nitric acid of different molarity. The fraction containing uranium and the fraction containing plutonium are then free of isobaric interference at mass 238 and ready for analysis by mass spectrometry. In general, several resins can be used in different media (HCl, $HNO_3$, etc.). This has been extensively studied for nuclear matrices [93-101].

The fraction not retained by the resin (fission product and minor actinide fraction) can be used to separate all the fission products and Am and Cm either again by gravitational chromatography or by high performance liquid chromatography (HPLC), illustrated in Figure 3. The separation is performed under gradient elution using different eluents, as described in literature [102,103]. In the example of HPLC separations, Cs can be detected with a scintillation detector. The lanthanides can be detected by UV-visible detection after post column derivatisation and Am and Cm can be detected by both detectors. An example of the possible chromatogram is shown in Figure 4. Each fraction obtained after separation is ready to be analysed by mass spectrometry.

HPLC, or other chromatographic techniques such as ion chromatography or capillary electrophoresis, can also be coupled on-line to an isotopic measurement technique like ICPMS. This approach simplifies the sample preparation procedure (no different sample fractions to be collected), is faster, and so limits the dose rate for the lab technicians and limits the radioactive waste. This approach is thus more economical. The main drawback is that the measurements are performed on transient signals which are inherently less precise than measurements on stable signals.

### 5.4 Radiometric measurement techniques

Irradiated fuel samples contain a multitude of gamma-, beta- and alpha-emitting radionuclides that can be analysed by a radioanalytical measurement technique. Radiometric techniques that are applied for spent fuel characterisation, such as gamma and alpha spectrometry and liquid scintillation counting, are mature and well-established methods. A number of prominent alpha- and gamma-emitting radionuclides in a dissolved fuel sample can be readily measured

**Figure 3: Experimental set-up used for uranium and FP separation in spent fuel solutions [104]**

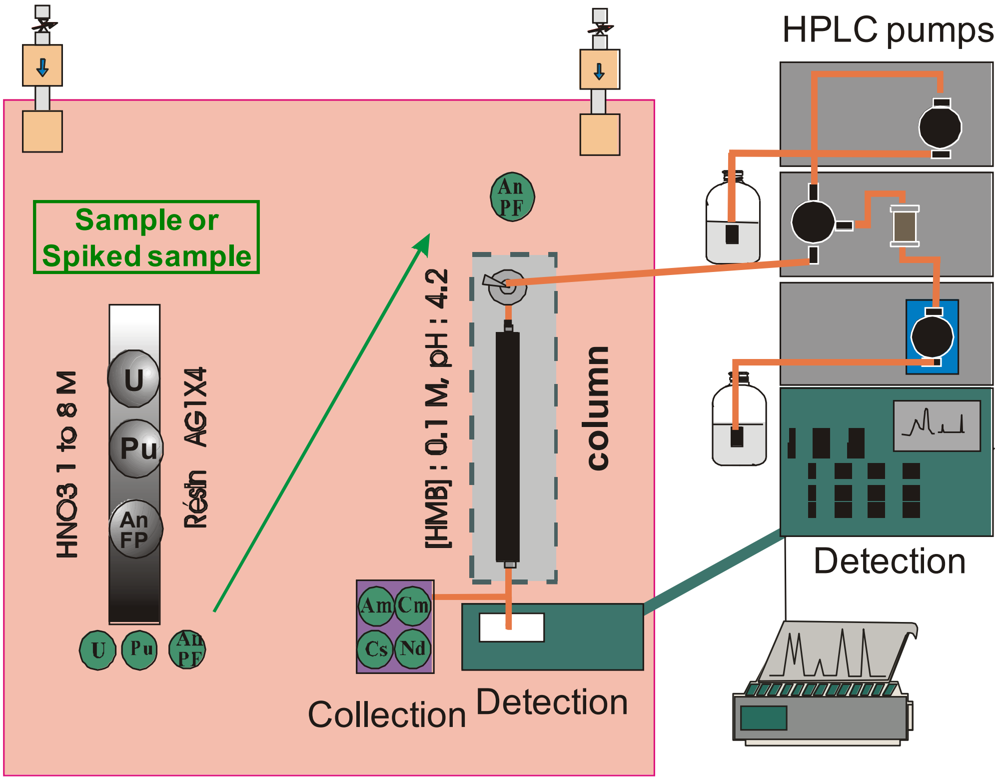
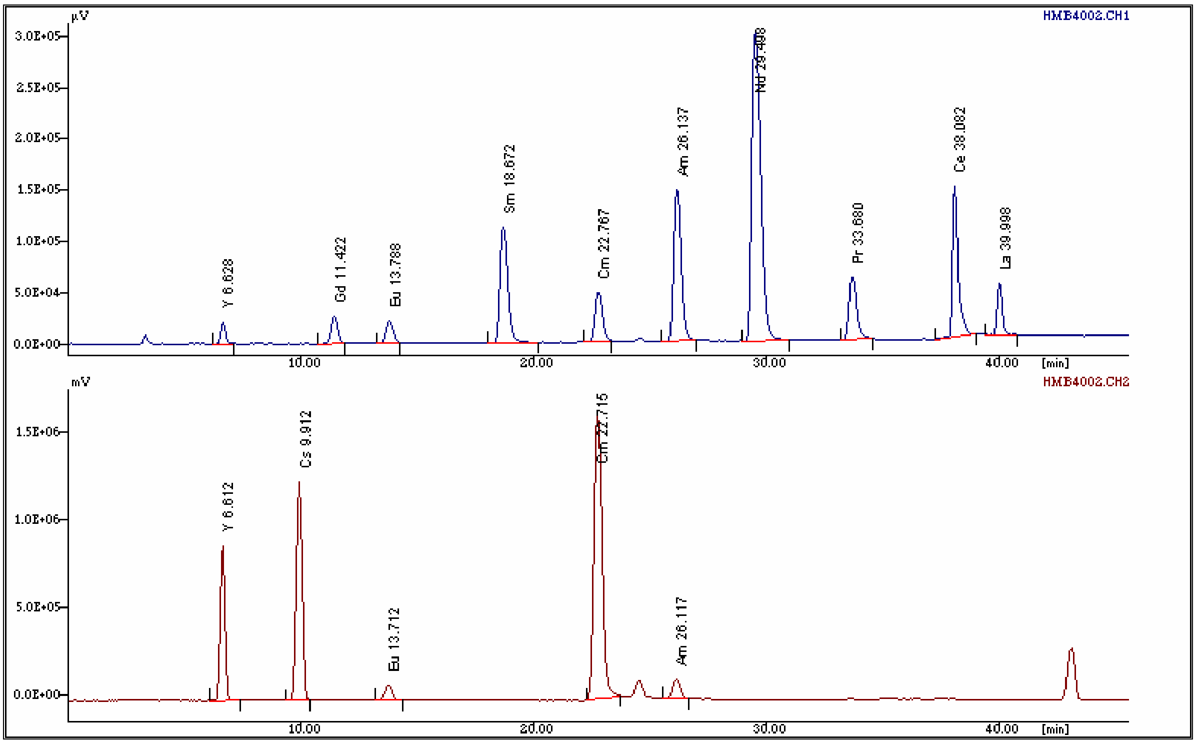


**Figure 4: Separation chromatogram for lanthanides, Am, Cm and Cs [103]**

UV-VIS detector (top), scintillation detector (bottom)



with alpha- and gamma-spectrometry without the need of complex sample preparation. However, for pure beta-emitters and some alpha- and gamma-emitting radionuclides, separation prior to measurement is required to eliminate radionuclides emitting interfering radiation and/or to reduce the background of the measurement source and consequently to reduce the detection limit.

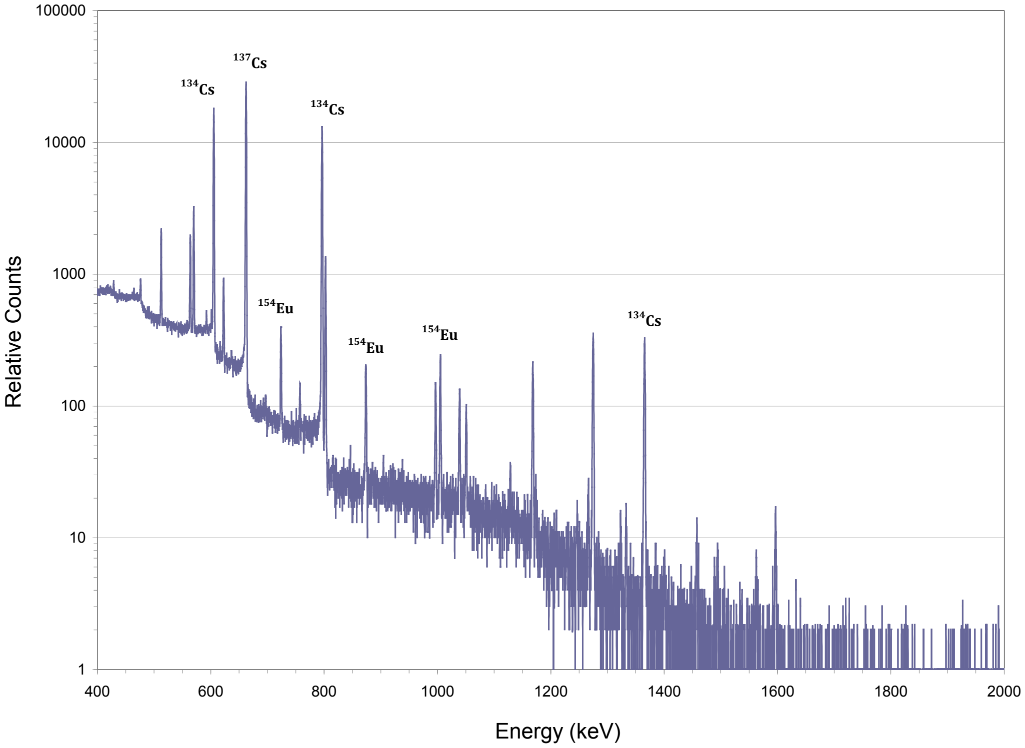
In the last two decades the mass spectrometric technique ICPMS has come into competition with these radiometric techniques for the measurement of radionuclides. ICPMS offers an attractive alternative, especially for the analysis of longer-lived radionuclides, due to its high sensitivity and the fact that for many of these radionuclides no or only limited sample preparation is needed. However, for shorter-lived radionuclides or when direct analysis with mass spectrometry is not possible due to the presence of isobaric or molecular interferences, radiometric techniques will remain the method of choice.

### *5.4.1 Gamma spectrometry (γ-spec)*

Gamma-ray spectrometry is a basic tool in a radioanalytical laboratory. It allows the measurement of gamma-emitting radionuclides without the need of extensive or complex sample preparation. For the analysis of spent fuel solutions it is used to measure prominent gamma-emitting fission products (e.g. $^{106}$Ru, $^{125}$Sb, $^{137}$Cs, $^{144}$Ce, $^{154}$Eu, $^{155}$Eu) and of minor actinides ($^{241}$Am and $^{243}$Cm). Analyses can be performed directly on a small aliquot of the spent fuel solution or on separated fractions. For particular radionuclides, such as $^{243}$Cm or $^{237}$Np, separation prior to the measurement may be needed to eliminate gamma-emitters that overlap with the gamma-rays of the analyte or to decrease the background of the spectrum substantially.

The detector best suited for identifying and quantifying gamma-emitting radionuclides in spent fuels is a high purity germanium (HPGe) detectors, which is a type of semiconductor diode. The energy resolution of an HPGe detector is high and allows excellent peak resolution for the complex spectrum of a spent fuel solution (Figure 5).

**Figure 5: Gamma spectrum of a diluted spent fuel solution**



Different configurations and geometries of HPGe detectors are commercially available. They all consist of a detector crystal, equipped with the necessary electronic connections and preamplifier, which is mounted under vacuum into a metal cap, usually constructed of aluminium. For the measurement of low-energy gamma-rays, this cap will have a thin entrance window of a different material such as beryllium. The detector is always cooled close to -196°C with either a Dewar vessel filled with liquid nitrogen or with an electrically powered cryogenic refrigerator. This is necessary to suppress electronic noise during the measurement and to obtain the high resolutions required. The detector is almost always installed in a shielding to reduce the natural background radiation from the environment and from cosmic radiation. The detector is also equipped with the necessary electronics and software to convert the electrical charges induced in the detector crystal after absorption of the gamma-ray energies according to their energy into a gamma spectrum that can be stored and analysed. Different electronics and software are commercially available.

A crucial step towards a reliable and accurate gamma spectrometry measurement is the calibration of the detector for energy, peak width and efficiency. Calibrations should be performed using a mixed radionuclide standard source or a combination of standard sources, traceable to a national standard (e.g. NIST, PTB). Calibration sources are preferably prepared with identical shape and density as the samples to be analysed. For efficiency calibration of the detector, these sources are placed in a standardised configuration versus the detector head. For some years computer software has become available to calculate an efficiency calibration curve for the gamma spectrometry detector without the use of "real" standard sources. Although reliability of such software has markedly increased, this approach is still not often applied for routine laboratory applications.

Energy calibration results in a linear relationship between channel number and energy of the gamma-ray. Typically, for the analysis of spent fuel samples, the detector will be calibrated over an energy range of approximately 50 to 2 000 keV and over a range of 4 096 channels. Efficiency calibration of the gamma detector varies non-linearly with the gamma-ray energy. It is usually not possible to fit a simple curve over the total energy range of 50 to 2 000 keV for efficiency; therefore in most cases the calibration data are split into two parts and two separate efficiency equations are calculated by the software.

Sample preparation for gamma spectrometry with liquid sources is in general straightforward and only requires the weighing and/or dilution of an aliquot in a calibrated geometry and volume. Typically, for a spent fuel analysis, measurement sources of 1 to 20 mL are prepared. At the high count rates of spent fuel samples coincidence summing of X- or gamma-rays may become a problem. However, if the distance to the detector is sufficiently high, this problem can be substantially reduced. The activity of the sample should be such that dead time of the detector is well below 5%. If this is not the case, the samples need further dilution or the distance to the detector has to be increased.

For routine gamma-ray spectrometry of spent fuel samples that have relatively high count rates, the overall uncertainty of the measurement is mainly dominated by the uncertainty of the standard used for efficiency calibration, the counting statistics, and to a lesser extent the sample preparation. In general, the overall uncertainty of the gamma-measurement will be 3% or more at a 95% confidence level.

### 5.4.2 Alpha spectrometry ($\alpha$-spec)

Alpha-ray spectrometry is a very useful tool for the detection and quantification of alpha-emitting radionuclides. It is routinely used for the measurement of alpha-emitting nuclides of uranium, plutonium, americium and curium in different types of nuclear and environmental samples. However, in the field of spent fuel analyses, mass spectrometry is the technique of choice for many of these actinides. The main reason is that the uncertainties obtained with isotope dilution mass spectrometry (ID-MS) are, in most cases, markedly lower than with isotope dilution alpha spectrometry, i.e. 0.2% for ID-MS versus 2% for ID-$\alpha$ spec.

In addition, mass spectrometry does not suffer from the overlap of isotopes of the same element in separated samples. For example, in alpha spectrometry $^{239}$Pu and $^{240}$Pu have overlapping alpha peaks in the spectrum that cannot be distinguished from one another. Despite the many advantages of using mass spectrometry, alpha spectrometry remains the best option when shorter-lived $\alpha$-emitting radionuclides have to be measured or when it is difficult to completely eliminate isobaric interferences. For example, it may not be possible to completely separate plutonium from the more abundant uranium in the fuel in one separation step. This means that there will be an isobaric interference of $^{238}$U on the $^{238}$Pu during the measurement of the separated Pu fraction with TIMS. Therefore instead of increasing the number of separation steps, some laboratories will prefer to measure $^{238}$Pu in the separated plutonium fraction using alpha spectrometry. Also for the measurement of $^{242}$Cm, with a relatively short half-life of 163 days, alpha spectrometry is the instrument of choice because of the lower detection limits that can be obtained compared to mass spectrometry.

A typical alpha spectrometer consists of a semi-conductor detector placed in a vacuum chamber and connected to the necessary electronics and software to produce an alpha spectrum. Commonly used PIPS detectors (passivated implanted planar silicon) are very suitable to convert the relatively high energy of the alpha particle (2-8 MeV) that is absorbed in the detector into an electronic signal and ultimately into a spectrum. In this spectrum the accumulated energies from an alpha source will be plotted as counts versus peak channel number. Typically for alpha spectrometry 1 024 channels are used.

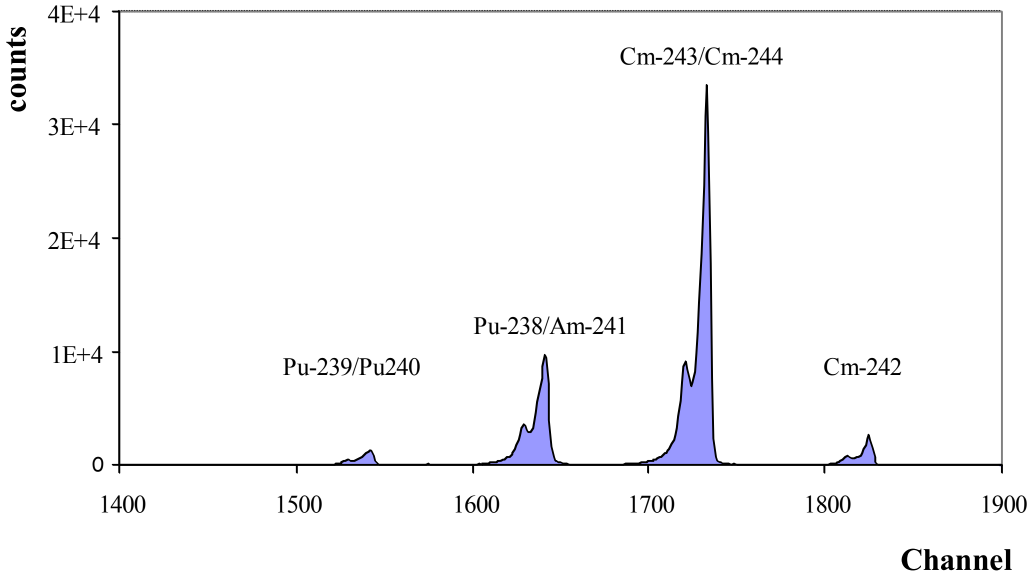
In general PIPS detectors are energy calibrated for a range of 0 to 10 MeV using standard sources of alpha-emitting nuclides with known energies or using a pulser which electronically simulates a signal at a specific energy. Efficiency calibration of the detector is performed using a standard source with known activity and prepared with the same geometry as the actual measurement sample. Because the counting efficiency of a PIPS detector is independent of the energy of the alpha particle, efficiency calibration can be done using a standard with a radionuclide emitting one distinct alpha peak, for instance $^{241}$Am. For detectors with a surface area of 450 mm$^2$ and alpha sources with a 1 cm diameter placed at a distance of ~1 cm of the detector, typically an efficiency of about 20-25% is obtained.

To more precisely determine the gross or total alpha activity of an unknown sample, a scintillation detector, such as an efficiency calibrated zinc sulphide detector, can be used in addition to the alpha spectrometry. These ZnS-detectors do not allow energy discrimination, but are excellent tools to measure the total alpha activity of a source with efficiencies up to 40%.

The resolution of the peaks that are obtained with alpha spectrometry will be influenced by the quality of the measurement source. As an alpha particle loses its energy very quickly through interaction with surrounding material, thin measurement sources have to be produced to minimise self-absorption. This is not a problem for the analysis of a spent fuel sample with high alpha count rates. There are different approaches possible for the preparation of alpha sources, namely electrodeposition, co-precipitation and evaporation. But for the analysis of spent fuel solution measurements sources are usually prepared by simple evaporation.

In general an alpha spectrum is not very complex and interpretation is much more straightforward than with gamma spectrometry. An example of an alpha spectrum of a diluted spent fuel solution of a measurement source prepared through evaporation is given in Figure 6. In case of the higher activities of the spent fuel alpha sources a low energy tail may be present on the peaks that could require corrections due to overlap of the low energy tail of one peak into a peak at lower energy. These corrections can be done manually or by using a computer code.

**Figure 6: Alpha spectrum prepared by evaporation of a diluted spent fuel solution**



In alpha spectrometry, the main contributions to the overall uncertainty of the measurement are the sample preparation, the counting statistics and the standard used for calibration. The overall uncertainty of alpha spectrometry is 2% or more at a 95% confidence interval.

### *5.4.3 Liquid scintillation counting (LSC)*

Liquid scintillation counting is a technique to measure beta- and alpha-emitting radionuclides. In this technique the sample and the scintillator material are incorporated in a homogeneous liquid medium by adding an aliquot of a separated radionuclide to a scintillation cocktail. These LSC cocktails contain a combination of different organic molecules that can convert the kinetic energy of a beta or alpha particle into light energy with a wavelength that is compatible with a photomultiplier tube. In the majority of the commercial LSC instruments, two detectors are placed in coincidence to reduce the background of the measurement. The detector system is usually lead shielded to reduce the effect of background radiation from surrounding materials on the measurement. The detected counts by the instrument for a sample are divided into channels according to their energy and computed into a continuous spectrum. Most systems are equipped with software that allows visualisation of the spectrum and calculation of the count rates.

The pure beta-emitting radionuclides $^{147}$Pm, $^{99}$Tc and $^{90}$Sr are good candidates for measurement with liquid scintillation counting. For LSC analysis, pure fractions of these beta-emitting radionuclides have to be prepared to eliminate interfering radiation from decay of other radionuclides. For spent fuel samples with a multitude of radionuclides with high to low activity levels, this means that a high decontamination factor has to be obtained.

Calibration of a LSC instrument is more complex than with a PIPS or HPGe-detector as the nature of the sample can have a high impact on the measurement. Indeed, the efficiency of LSC will not only depend on the nature of the scintillator, the radiation and applied sample preparation method, but is also strongly influenced by processes such as chemical and colour quenching and luminescence. Although for studies using a single radionuclide or for environmental monitoring, the common method of calibration is the external standard method using an established quench calibration curve, for spent fuel analysis internal standardisation is the method of choice. In this approach a known amount of a standard of the radionuclide to be analysed (for example $^{147}$Pm) is added to an unknown sample. Three measurements are performed, namely a blank sample to evaluate the background, an unknown sample and a sample with the internal standard added. This method allows to fully negate any effects of quenching and to obtain a more reliable measurement result.

In liquid scintillation counting, the main contributions to the overall uncertainty of the measurement are the counting statistics, the uncertainty on the certified value of the reference material used for internal standardisation and the uncertainty on volumetric sampling tools used (e.g. pipette). The overall uncertainty of liquid scintillation counting for a moderately active sample (good counting statistics) approaches the uncertainty of the certified reference material used as internal standard (typically 2% or more at a 95% confidence interval).

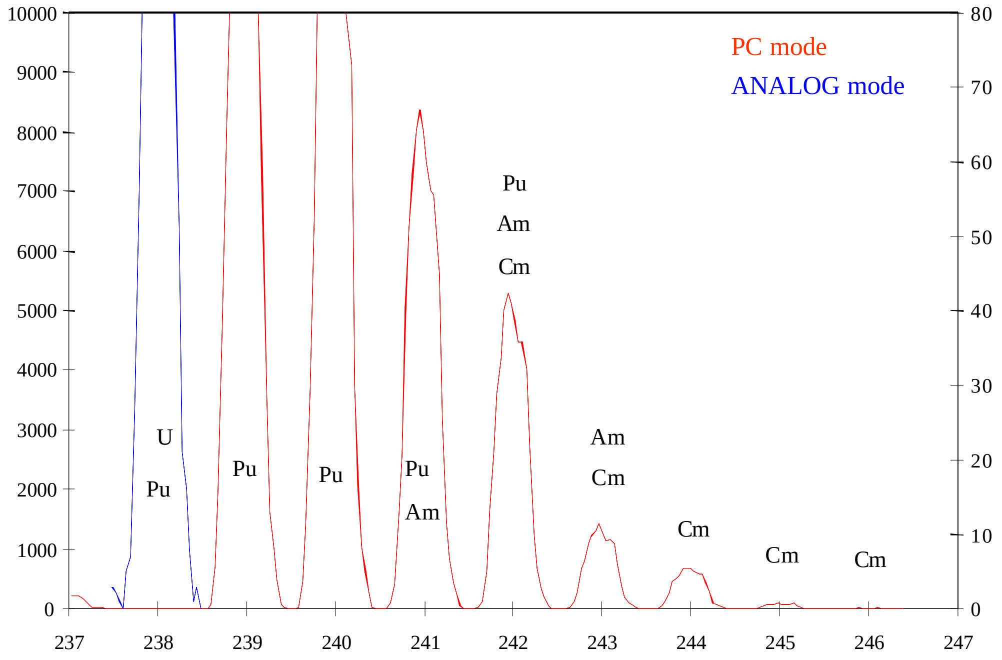
### 5.5 Mass spectrometry techniques

Mass spectrometry techniques permit the precise isotopic characterisation of the elements when applied to separated fractions of the various elements in order to avoid some possible isobaric interferences and matrix effects. Mass spectrometry requires specific and quite complex equipment.

The best performances in terms of accuracy and precision are obtained by thermal ionisation mass spectrometry (TIMS) and by multi-collector inductively coupled plasma mass spectrometry (MC-ICPMS) [105-110]. In case of elemental concentration analysis, accuracies of about 0.1-0.4% can be reached by using the isotope dilution technique after separation of the pure elemental fractions out of the sample.

An example of an un-separated mass spectrum is shown in Figure 7. There is an overlap between some isotopes of the actinides: U and Pu at mass 238, Pu and Am at mass 241, Pu, Am and Cm at mass 242, and Am and Cm at mass 243. It is not possible under these conditions to perform very accurate isotopic analysis because of these overlaps. So it is necessary to separate the different elements in order to obtain pure elemental fractions before their isotopic measurements by mass spectrometric techniques (TIMS or MC-ICPMS).

**Figure 7: Mass spectrum of spent fuel sample showing overlap between some isotopes [103]**



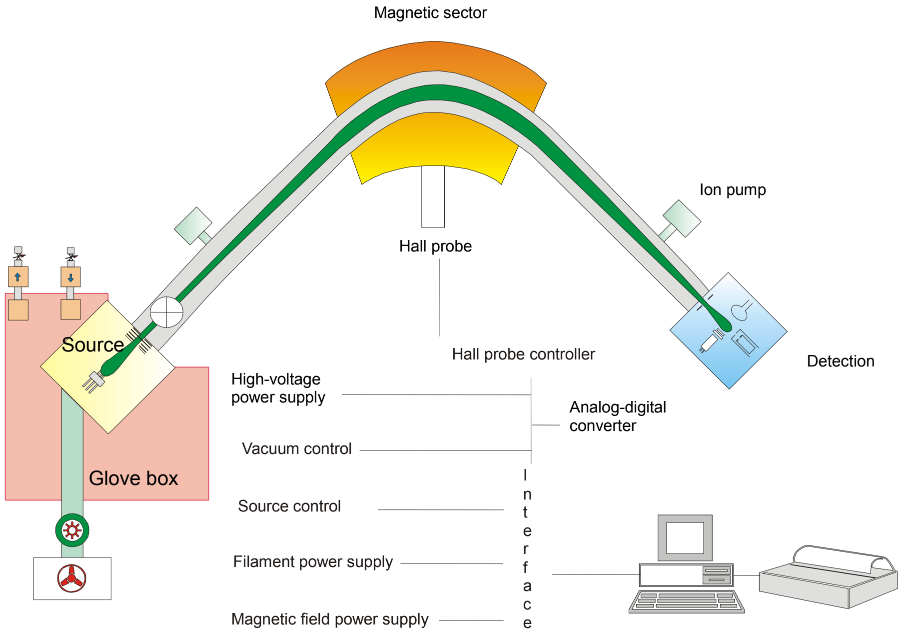
The atomic ratio of the different isotopes of an element relative to $^{238}$U can also be obtained. When only one isotope has to be determined or in case of mono-isotopic elements, ICPMS can be used to determine the concentration of the isotope of interest without separation [111-113]. Concentration of $^{238}$U is then determined by simple isotopic dilution with a $^{233}$U spike and the ratio X/$^{238}$U can be obtained with an accuracy of a few per cent level at the 95% confidence level.

### *5.5.1 Thermal ionisation mass spectrometry (TIMS)*

An amount of the (separated) element of interest (about a few tens of nanograms to several micrograms) is deposited on a filament. By sending an electrical current through this filament under vacuum, the element is thermally atomised and ionised. With the right instrumental conditions, a stable ion beam can be generated for about 1-2 hours. There also exist TIMS instruments for which the sample material should be in the gaseous form but the ionisation is done the same way. An extraction lens followed by focalisation lenses extract and lead the ions to the entrance of the mass spectrometer. In the magnetic field of the magnetic sector, the ions are separated according to their mass/charge ratio. Ion currents corresponding to each isotope present are then detected in a sequential way (single collector) or in a simultaneous way (multi-collector) by the use of judiciously positioned collectors.

Two different types of detector are present on most TIMS instruments: Faraday cup(s) and a more sensitive Daly ion counting system able to measure low signals. For the measurement of isotope ratios, very good precisions can be reached with TIMS, of the order of 0.1% or even better for multi-collector instruments. Figure 8 displays a schematic view of a nuclear TIMS system.

**Figure 8: Schematic of a nuclearised TIMS [103]**



Corrections are needed for the time-dependent effect of instrumental mass fractionation (of the order of per mil per atomic mass unit) by normalisation of the measured ratio to an invariant isotope ratio of the same element.

One of the unique challenges for high precision isotopic ratio measurements in nuclear applications is that internal normalisation using a natural isotopic ratio cannot be considered because irradiation of nuclear fuel samples alters the natural abundances. Nevertheless the problem of isotopic fractionation can be resolved in two different ways: *i)* use isotopic standards to adapt the analytical procedure to obtain highly precise and accurate ratios; *ii)* the flash evaporation method. With the flash evaporation method, the sample is entirely volatilised while the signal from each isotope is integrated. Thus, the effect of isotope fractionation is eliminated. However, the first method is more precise than the second. Furthermore the quality of the TIMS analysis also depends on the mass deposited on the side filament, on the chemical form of the element as well as its chemical purity and on the nature of the filament.

TIMS analysis thus implies sample purification and numerous chromatographic separations (as it is a mono-elemental analysis technique), making the sample preparation time-consuming and the measurements very expensive relative to the ICPMS method.

### 5.5.2 Inductively coupled plasma mass spectrometry (ICPMS)

In ICPMS, the ionisation source is an inductively coupled plasma (ICP). A liquid sample is continuously introduced as an aerosol into an argon plasma produced by application of an RF field (the applied power is typically 1 200-1 400 W) around a quartz torch. The temperature within the plasma being above 5 000 K, the spray is vaporised, desolvated, atomised and atoms are ionised. The efficiency of ionisation is >90% for most elements, making ICP a very interesting ion source for MS.

The ionisation takes place at atmospheric pressure, but for MS a high vacuum is needed. The so-called interface provides that transition. It consists of two topped-off cones, sampler and skimmer, placed in series just in front of the plasma. Between sampler and skimmer, an intermediate vacuum is created and behind the skimmer, a high vacuum. The ions undergo a supersonic expansion twice by passing through sampler and skimmer and are so “frozen” in their ionic state. An extraction lens and some ion optics just behind the skimmer gather as many ions as possible and shape them into an ion beam. Different types of mass spectrometers can be coupled to an ICP. For nuclear applications, quadrupole and sector field mass spectrometers are being used.

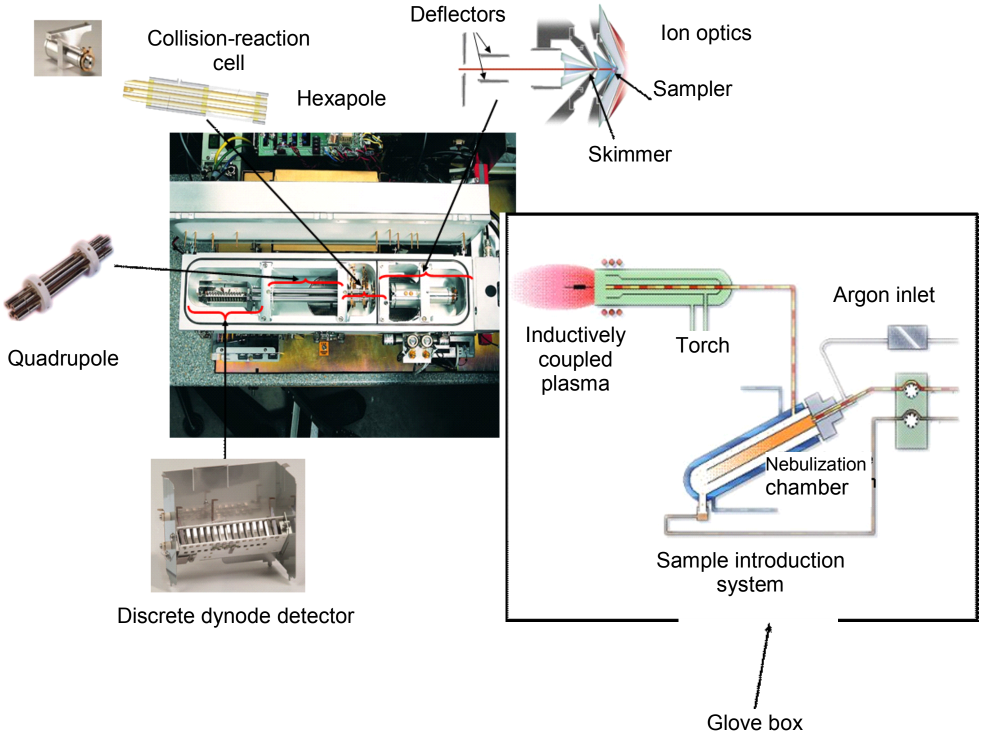
### 5.5.3 Quadrupole ICPMS (Q-ICPMS)

A quadrupole mass spectrometer consists of four cylindrical rods, placed symmetrically around a central beam. The diagonally opposite rods are electrically connected. To both sets of rods, an opposite voltage is applied, consisting of both a direct current ($V_{DC}$) and a radiofrequency ($V_{RF}$) component. This generates a unique oscillating field. At a specified frequency and amplitude, combined with a specified $V_{DC}/V_{RF}$ ratio, only ions of one specific mass-to-charge ratio (m/z) undergo stable oscillation and can reach the detector at the end of the quadrupole region. Ions of higher or lower mass-to-charge ratio undergo oscillation of increasing amplitude and are lost for detection. So, a quadrupole mass spectrometer acts as a mass-to-charge filter. By changing the $V_{DC}$ and $V_{RF}$ potentials of the quadrupole rods, keeping the ratio between them constant, the mass-to-charge ratio that can pass this filter is altered. In this way, the mass region of the isotopes of an element of interest can be scanned. Changing the $V_{DC}$ and $V_{RF}$ potentials is computer-controlled and can be done very quickly.

The separated ions are then detected with a secondary electron multiplier. The sequential measurement of the various ion currents produced allows, after calibration with standards, a quantitative analysis of the various isotopes.

A schematic of a Q-ICPMS is shown in Figure 9. More recent Q-ICPMS instruments can be equipped with a “collision-reaction cell”, a technology allowing the suppression of some poly-atomic interference.

**Figure 9: Schematic of a quadrupole ICPMS including a "collision-reaction cell"**



### 5.5.4 *Sector field inductively coupled plasma mass spectrometry (SF-ICPMS)*

Sector field ICPMS consists of a standard ICP source and a sector field mass spectrometer as for TIMS. But as the energy spread of the ions of an ICP is much broader than for the ions in a TIMS, an electrostatic sector is also added to the magnetic sector. With a correct set-up and tuning of the magnetic and the electric field, together they focus both ion angles (first focusing) and ion energies (double focusing), while being dispersive for the mass to charge ratio (m/z).

With SF-ICPMS, the (separated) ions can be detected sequentially (single detector) or simultaneously (multi-collector). In a single detector instrument, a secondary electron multiplier (SEM) is used as detector. In a multi-collector instrument, the SEM is combined with a series of Faraday cups. For the measurement of isotopic ratios, the advantages of a multi-collector instrument are obvious as the ion signals of different isotopes can be measured simultaneously, resulting in a precision comparable to TIMS.

A significant feature of plasma source mass spectrometry is the large instrumental mass bias, which is related to the enhanced extraction and transmission of the heaviest ions [114]. This phenomenon can be corrected for, using correction factors determined via isotope standards or solutions of natural elements. The analysis of each sample is then bracketed by analyses of an isotope standard before and after the sample of unknown isotopic composition.

## 5.6 Calibration methods in mass spectrometry

Different calibration procedures can be applied to measure isotopic concentrations in spent nuclear fuel samples. The most accurate, without contest, is the isotope dilution technique which is based on the precise measurement of isotopic ratios. When used with multi-collection instruments, it allows a precision of better than 0.5% to be obtained in the majority of cases.

When only one isotope has to be quantified, an external calibration method can be used to measure the concentration of this isotope. The methodology for these methods is described here very briefly.

### *5.6.1 Simple isotope dilution technique*

The principle of isotope dilution (ID) is straightforward: to the unknown amount of an element with known (or measurable) isotopic composition, a known amount of the same element with known (or measurable) but different isotopic composition (the so-called "spike") is added. As a result of this mixing, the isotopic composition of that element is altered. The new isotopic ratios are measured in this blend. Knowing the isotope ratios in the sample (measured), spike (certified or measured) and blend (measured), knowing the concentration of the spike (certified or measured) and knowing the amount of sample and spike added in the blend (usually weighed), the unknown concentration of the element in the sample can be calculated.

For example, the $^{238}U$ concentration can be measured in solutions by simple isotope dilution with TIMS or ICPMS and using a $^{235}U$ spike with a fully characterised isotope composition and concentration and after chromatographic separation from plutonium and fission products in a gravity column. General uncertainty depends on the measurement technique and quality of the spike used. It can be better than 0.5% for MC-ICPMS and TIMS and better than 2-3% for Q-ICPMS.

As ID is only based on isotope ratio measurements of the same element, the determination is highly selective with very few interferences possible (isobaric interferences in the mass spectrum). These can be avoided by, for example, chemical separation or correction (if the isotopic composition of the interfering element is known). Sample treatment (like separation) does not need to be quantitative once the spike has been added and mixed homogeneously to the sample. Whether the separation process is quantitative or not, the ID ratio in the blend is fixed so that each sample of any size can be measured for the required ratio.

Nuclear samples have the additional advantage that the isotopic composition of the fission products differs from the natural isotopic composition of these elements. So, very often, natural material can be used as spike instead of expensive enriched isotopic standard materials.

### *5.6.2 Double isotope dilution technique*

The double isotope dilution technique allows determining directly and in a very accurate way the concentration ratio of two elements present in a sample. Since this technique is also only based on isotope ratio measurements, quantitative or constant elemental recovery is not required and accurate measurements of the amount of sample and spike solutions are not necessary. Given the precise isotope composition and concentration of the spike and the isotope abundance of the element in the sample, the sample-spike mixture isotope composition is used to obtain the element-to-uranium ratio by application of isotope inventory equations. The resulting accuracy depends on that of the isotope ratio measurements, i.e. a few tenths of a per cent by TIMS or MC-ICPMS. Another advantage of this technique is that the chemical separation efficiency does not need to be constant.

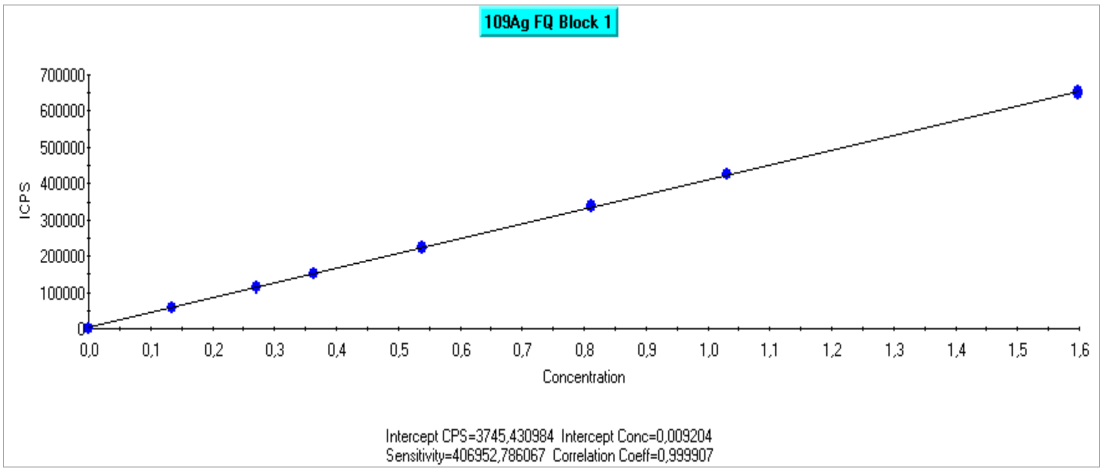
For these element/U determinations, first a mixing of spike and sample is performed and then the different steps of chemical separation as described earlier, depending on the methodology used.

### *5.6.3 Determination of concentrations by external calibration*

If no isobaric interferences are present, the measurements can be performed without preliminary chemical separations. The associated uncertainties are at a level of a few per cent.

Typically, a calibration curve is obtained with a minimum of six standards of different concentrations. For each standard, a minimum of 20 replicates is measured in order to verify and guarantee a good repeatability of the measurements. Internal standards are used (e.g. indium and bismuth) in order to correct for instrumental drift over the complete measurement sequence. An example of a calibration curve obtained with Q-ICPMS is shown in Figure 10.

**Figure 10: Calibration curve obtained with Q-ICPMS for $^{109}$Ag between 0.1 and 2 ng/mL [103]**

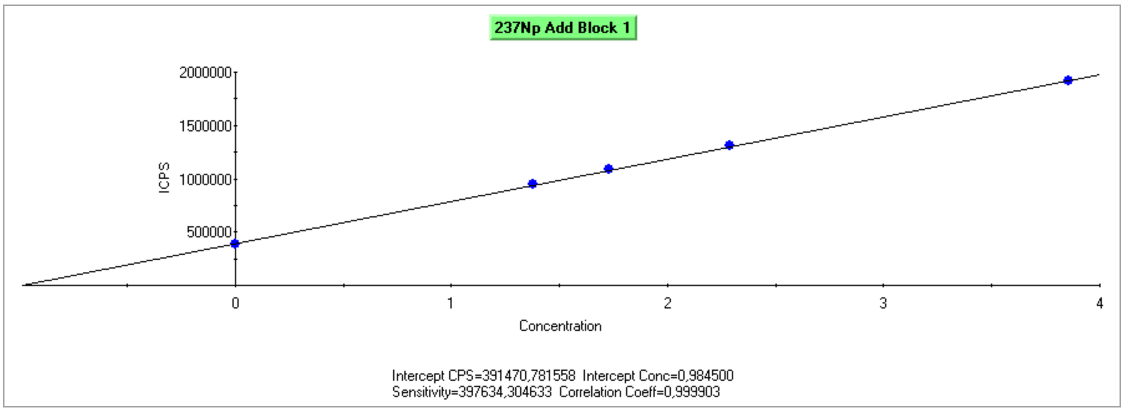


### 5.6.4 *Determination of concentrations by standard addition*

If no isobaric interferences are present, the measurements can be performed without preliminary chemical separations. The associated uncertainties are at a level of a few per cent. This technique, well described in literature [115,116] allows a measurement independent of any matrix effect and is of primary interest in the nuclear field due to the high concentration of uranium (possibly causing matrix effects) in fuel dissolution solutions. This technique is used when no preliminary separation is performed in order to eliminate the major component, uranium in our case.

Typically, a calibration curve is obtained with a minimum of three standard additions to the sample and a non-added sample. For each sample, a minimum of 20 replicates is realised to verify and guarantee a good repeatability of the measurements. Internal standards are used (e.g. indium and bismuth) to correct for instrumental drift over the complete measurement sequence. An example of a calibration curve obtained in Q-ICPMS is shown in Figure 11. The concentration is determined via the intercept of the linear calibration curve with the concentration axis.

**Figure 11: Standard addition calibration curve obtained in Q-ICPMS for $^{237}$Np**



## 5.7 Measurement time

The results of the isotopic assay analysis of spent nuclear fuel always have a date associated to them, most often the date of analysis. This is very important information required to calculate the full set of analytical data back to one reference date, usually at the end of life (EOL), corresponding to the time of discharge from the reactor when the fuel was last irradiated.

Besides its impact on the residual content of short-lived nuclides, the time elapsed from reactor discharge is also important for nuclides with significant production (compared to in-reactor production) after EOL (for example, $^{241}$Am). For these nuclides, the uncertainty on the calculated EOL content (i.e. the in-reactor production) may increase to such an extent that it becomes unsuitable for use in validating calculations. Any corrections of the data to a reference date must account not only for the decay of the nuclide but also production from decay precursors.

Other important information is the "separation date" as mother-daughter relationships in radioactive decay schemes have to be carefully considered. Definitely when off-line separations are being used, the difference between "separation date" and "analysis date" can become significant for the decay calculations. Examples of mother-daughter couples are $^{241}$Pu-$^{241}$Am, $^{144}$Ce-$^{144}$Nd, $^{147}$Pm-$^{147}$Sm and $^{151}$Sm-$^{151}$Eu.

## 5.8 Experimental uncertainty

As described above, the experimental determination of the nuclide content in a spent fuel sample is the result of a series of "steps" starting with the weighing of the fuel sample after cutting and including various handling steps, up to the measurement itself.

The ARIANE experimental programme has shown (based on laboratory cross-checks) that the measurement methodology used for a given nuclide should be qualified against (an)other methodology(ies) (using other standards and/or spikes, and/or separation medium, and/or measurement devices), both to avoid systematic errors (e.g. problems with a standard) and to identify unreliable methodology.

A first, straightforward and relevant check of the global quality of the measurements performed on a spent fuel sample is referred to as the mass balance (MB). A mass balance is obtained, for a given sample, by dividing the as-measured sum of heavy metal atoms plus the heavy metal mass loss due to fissions (based on $^{148}$Nd measurement) by the number of heavy metal atoms initially present in the un-irradiated fuel sample.

Deviations from the balance (i.e. from MB = 1.00) should not exceed the level of uncertainty on major actinide measurements (generally close to 1% at the 95% level). Deviations larger than 3% are an indication that at least one step of the measurement process is questionable (material loss, inefficient separation, weighing mistake, etc.). Depending on the step where the error occurred, this error can affect the as-measured content for all the different nuclides, and not only the actinides and Nd isotopes often used for burn-up determination.

The measured content of a given nuclide should be reported as an absolute content in the dissolved fuel sample (weight/weight dissolved fuel) separately from the contents measured for other nuclides (to avoid biases); each measurement will have its associated uncertainty to which a specified level of confidence can be assigned independently of the other measured contents (e.g. 68%, 95% or another defined value of confidence), cf. Ref. [117], Sections 9.1.2 and 9.1.3. However, in addition to the uncertainties arising from the respective empirical variances, the measured concentrations have covariances that define correlations in the measurement results. The related empirical covariance matrix has recently been shown to enable more realistic uncertainty evaluations in depletion calculation for burn-up credit validation, cf. Refs. [118] and [119]. Analytical results become much more valuable when the associated total uncertainty is reported. Often, only the uncertainty related to the measurement itself is given. Although this uncertainty might be an important contributor, the total uncertainty should include all sources of uncertainty along the complete analytical process.

A list of uncertainty sources in the analysis of spent fuel (without claiming completeness) should include:

- the overall efficiency of dissolution (element-dependent; for most important elements in spent fuel analysis contributing negligibly to the total uncertainty);
- uncertainties on manipulations in hot cell and glove-boxes (e.g. weighing the sample, fuel, cladding, residue and performing dilutions, etc.);
- uncertainty on separation yields (not applicable when using isotopic dilution techniques);
- uncertainty of the standard(s) and/or spike(s);
- uncertainty of the measurements (including precision and accuracy of the measurement technique);
- uncertainties on nuclear data (e.g. for EOL calculations).

The following reference documents contain all detailed information related to this important topic including for example, the definitions and correct terminology to be used, the way to quantify, calculate, combine and report uncertainties:

- *Uncertainty of Measurement – Part 3: Guide to the Expression of Uncertainty in Measurement (GUM:1995)*, ISO/IEC Guide 98-3:2008, Geneva, Switzerland (2008).
- *Quantifying Uncertainty in Analytical Measurement*, EURACHEM/Citac Guide CG4, 2nd edition (2000), ISBN 0 948926 15 5.

The ISO/IEC Guide is intended to be applicable to a very broad spectrum of measurements, whereas the EURACHEM/Citac Guide (based on the ISO/IEC Guide) focuses on analytical chemistry measurements, giving also worked-out examples of total uncertainty calculations.

As a general remark, it would be of interest that laboratories attempt to harmonise their methods/principles for the evaluation of the uncertainties and their confidence level; this would allow a more discerning use of the results.

### 5.9 Recommended techniques for nuclides of interest

Table 15 lists nuclides measured in the framework of many large experimental programmes (like the ARIANE programme) and the applied measurement methodologies and techniques. These nuclides are of importance in various areas related to the use of nuclear energy (e.g. licensing, safety analysis, safeguards) and include major and minor actinides, burn-up indicators, burn-up credit nuclides, as well as major heat-emitting and gamma-emitting nuclides and long-lived fission products.

The list could be enlarged, for example to include several nuclides of interest in the field of long-term spent fuel management. The most important radiological nuclides would be (this list is non-exhaustive) $^{14}$C, $^{36}$Cl, $^{79}$Se, $^{93}$Zr, $^{107}$Pd and $^{126}$Sn, present in spent fuel either as fission or activation products, or both. One notes that for some of them (e.g. $^{14}$C, $^{79}$Se and $^{36}$Cl), measurement methods applying to spent fuel samples have been developed or are being investigated.

An additional consideration for nuclear data evaluation is the availability of isotopic data for important precursors of the nuclide of interest. These data can provide valuable insight into the cause of potential discrepancies as they relate to nuclear data. Measurements of decay precursors of the nuclide of interest are also required in order to adjust any measurements to account for differences in the decay time. This is frequently done in order to provide measurement data for a single reference measurement date. However, in general such corrections should be done using a date as close as possible to the actual measurement date in order to minimise the uncertainties. Lastly, measurements of the important burn-up indicators are required. These nuclides can include $^{137}$Cs, $^{139}$La, $^{145}$Nd, $^{146}$Nd, $^{148}$Nd, $^{149}$Nd and $^{150}$Nd.

**Table 15: Nuclides and recommended measurement techniques and methods**

| Nuclide | Method for measuring isotopic composition | Method for measuring absolute isotopic content | Alternative methods and remarks |
|---|---|---|---|
| $^{234}$U | TIMS/ICPMS | IDMS | |
| $^{235}$U | TIMS/ICPMS | IDMS | |
| $^{236}$U | TIMS/ICPMS | IDMS | |
| $^{238}$U | TIMS/ICPMS | IDMS | U/Pu separation needed to avoid $^{238}$U/$^{238}$Pu isobaric interference |
| $^{237}$Np | Mono-isotopic | ICPMS (a) | Alternatively, via $\gamma$-spec via daughter $^{233}$Pa |
| $^{238}$Pu | TIMS/ICPMS | IDMS (b) | For MS, U/Pu separation needed to avoid $^{238}$U/$^{238}$Pu isobaric interference |
| $^{239}$Pu | TIMS/ICPMS | IDMS (c) | |
| $^{240}$Pu | TIMS/ICPMS | IDMS (c) | |
| $^{241}$Pu | TIMS/ICPMS | IDMS | Pu/Am separation needed to avoid $^{241}$Pu/$^{241}$Am isobaric interference |
| $^{242}$Pu | TIMS/ICPMS | IDMS | Pu/Am/Cm separation needed to avoid $^{242}$Pu/$^{242}$Am/$^{242}$Cm isobaric interference |
| $^{244}$Pu | TIMS/ICPMS | IDMS | Pu/Cm separation needed to avoid $^{244}$Pu/$^{244}$Cm isobaric interference |
| $^{241}$Am | TIMS/ICPMS | IDMS (d) | Pu/Am separation needed to avoid $^{241}$Pu/$^{241}$Am isobaric interference |
| $^{242}$Am | TIMS/ICPMS | IDMS | Pu/Am/Cm separation needed to avoid $^{242}$Pu/$^{242}$Am/$^{242}$Cm isobaric interference |
| $^{243}$Am | TIMS/ICPMS | IDMS | Am/Cm separation needed to avoid $^{243}$Am/$^{243}$Cm isobaric interference |
| $^{242}$Cm | TIMS/ICPMS | $\alpha$-spec | In most cases, concentration too low for MS |
| $^{243}$Cm | TIMS/ICPMS | $\gamma$-spec (on separated Am-Cm fraction) | In most cases, concentration too low for MS; in $\gamma$-spec, correction needed for $^{239}$Np (daughter of $^{243}$Am) interference |
| $^{244}$Cm | TIMS/ICPMS | $\alpha$-spec | |
| $^{245}$Cm | TIMS/ICPMS (e) | | Concentration based on isotopic MS data + concentration of $^{244}$Cm via $\alpha$-spec |
| $^{246}$Cm | TIMS/ICPMS (e) | | Concentration based on isotopic MS data + concentration of $^{244}$Cm via $\alpha$-spec |
| $^{90}$Sr | | $\beta$-LSC *or* ICPMS | $\beta$-LSC measurement requires a pure separated Sr-fraction; ICPMS requires a Sr/Zr separation in order to avoid $^{90}$Sr/$^{90}$Zr isobaric interference |
| $^{95}$Mo | | ICPMS (a) | For full inventory, dissolution of the residue (after standard dissolution procedure) is needed; measurement does not need preceding separation |
| $^{99}$Tc | | ICPMS (a) | |
| $^{101}$Ru | | ICPMS (a) | |
| $^{103}$Rh | | ICPMS (a) | |
| $^{106}$Ru | | $\gamma$-spec of Ru/$^{106}$Rh | |
| $^{109}$Ag | | ICPMS (a) | |
| $^{125}$Sb | | $\gamma$-spec | |
| $^{107}$Pd | | ICPMS (a) | As above (presuming no “natural Ag” contamination) |
| $^{126}$Sn | | ICPMS (a) | As above (presuming no “natural Te” contamination) |
| $^{129}$I | | ICPMS (f) | $^{129}$I requires trapping of the off-gases in alkaline solution during the acid dissolution of the fuel (I is volatile in acid medium) |

(a) External calibration; (b) alternative $\alpha$-spectrometry; (c) alternative $^{239}$Pu+$^{240}$Pu sum-peak in $\alpha$-spectrometry; (d) alternative method $\alpha$- and/or $\gamma$-spectrometry; (e) alternative method in ICPMS is to determine concentration via Cm/U ratio; (f) external calibration; alternative $\gamma$-spec after separation.

## Table 15: Nuclides and recommended measurement techniques and methods (*cont.*)

| Nuclide | Method for measuring isotopic composition | Method for measuring absolute isotopic content | Alternative methods and remarks |
|---|---|---|---|
| $^{133}$Cs | TIMS/ICPMS (+ $\gamma$-spec for $^{134}$Cs and $^{137}$Cs) | IDMS (+ $\gamma$-spec for $^{134}$Cs and $^{137}$Cs) | |
| $^{134}$Cs | | | For MS, Cs/Ba separation needed to avoid $^{134}$Cs/$^{134}$Ba isobaric interference |
| $^{135}$Cs | | | Cs/Ba separation needed to avoid $^{135}$Cs/$^{135}$Ba isobaric interference |
| $^{137}$Cs | | | For MS, Cs/Ba separation needed to avoid $^{137}$Cs/$^{137}$Ba isobaric interference |
| $^{142}$Nd | TIMS/ICPMS | IDMS | Ce/Nd separation needed to avoid $^{142}$Ce/$^{142}$Nd isobaric interference |
| $^{143}$Nd | | | |
| $^{144}$Nd | | | Ce/Nd separation needed to avoid $^{144}$Ce/$^{144}$Nd isobaric interference |
| $^{145}$Nd | | | |
| $^{146}$Nd | | | |
| $^{148}$Nd | | | Nd/Sm separation needed to avoid $^{148}$Nd/$^{148}$Sm isobaric interference |
| $^{150}$Nd | | | Nd/Sm separation needed to avoid $^{150}$Nd/$^{150}$Sm isobaric interference |
| $^{144}$Ce | | $\gamma$-spec *or* ICPMS | For MS, Ce/Nd separation needed to avoid $^{144}$Ce/$^{144}$Nd isobaric interference |
| $^{147}$Pm | | ICPMS *or* $\beta$-metric measurement by liquid scintillation counting | ICPMS requires a Pm/Sm separation to avoid $^{147}$Pm/$^{147}$Sm isobaric interference; $\beta$-metric measurement requires a pure separated Pm-fraction |
| $^{147}$Sm | TIMS/ICPMS | IDMS | Pm/Sm separation needed to avoid $^{147}$Pm/$^{147}$Sm isobaric interference |
| $^{148}$Sm | | | Nd/Sm separation needed to avoid Nd$^{148}$/$^{148}$Sm isobaric interference |
| $^{149}$Sm | | | |
| $^{150}$Sm | | | Nd/Sm separation needed to avoid $^{150}$Nd/$^{150}$Sm isobaric interference |
| $^{151}$Sm | | | Sm/Eu separation needed to avoid $^{151}$Sm/$^{151}$Eu isobaric interference |
| $^{152}$Sm | | | |
| $^{154}$Sm | | | Sm/Eu/Gd separation needed to avoid $^{154}$Sm/$^{154}$Eu/$^{154}$Gd isobaric interferences |
| $^{151}$Eu | TIMS/ICPMS (+ $\gamma$-spec for $^{154}$Eu and $^{155}$Eu) | IDMS (+ $\gamma$-spec for $^{154}$Eu and $^{155}$Eu) | Sm/Eu separation needed to avoid $^{151}$Sm/$^{151}$Eu isobaric interference |
| $^{153}$Eu | | | |
| $^{154}$Eu | | | Sm/Eu/Gd separation needed to avoid $^{154}$Sm/$^{154}$Eu/$^{154}$Gd isobaric interferences |
| $^{155}$Eu | | | Eu/Gd separation needed to avoid $^{155}$Eu/$^{155}$Gd isobaric interferences |
| $^{154}$Gd | TIMS/ICPMS | IDMS | Sm/Eu/Gd separation needed to avoid Sm$^{154}$/$^{154}$Eu/$^{154}$Gd isobaric interferences |
| $^{155}$Gd | | | Eu/Gd separation needed to avoid $^{155}$Eu/$^{155}$Gd isobaric interferences |
| $^{156}$Gd | | | |
| $^{157}$Gd | | | |
| $^{158}$Gd | | | |
| $^{160}$Gd | | | Gd/Dy separation needed to avoid $^{160}$Gd/$^{160}$Dy isobaric interferences |

(a) External calibration; (b) alternative $\alpha$-spectrometry; (c) alternative $^{239}$Pu+$^{240}$Pu sum-peak in $\alpha$-spectrometry; (d) alternative method $\alpha$- and/or $\gamma$-spectrometry; (e) alternative method in ICPMS is to determine concentration via Cm/U ratio; (f) external calibration; alternative $\gamma$-spec after separation.

# Chapter 6: Validation calculations using isotopic assay data

The spent fuel isotopic assay measurements described in Chapter 5 provide the basic experimental data used for code validation calculations. However, isotopic assay data measured in an experimental programme is only one component of the global data needed to validate computer models and data. In addition to the measured isotopic assay data, there are other data essential for performing the computer code analyses. A significant amount of design and reactor operation data are also required, including for example information on the initial (un-irradiated) fuel composition, fuel assembly design, irradiation history, irradiation environment (temperatures, void, etc.), neighbouring assembly effects and exposure to potentially perturbing influences such as burnable poison rods or control blades.

The level of detail required for data has evolved and increased as computational capabilities have increased. Early experimental programmes are generally not as well documented as more modern programmes. Requirements for more detailed design and operating data have been driven largely by the development of increased computational capabilities and increased accuracy of modern computer code systems, and the need to further reduce uncertainties in the calculations that are applied to support safety and licensing studies. Missing critical information on design or operating data can severely reduce the value of the experimental data for validation. In such cases, approximate information must be applied. However, this introduces additional uncertainties in the experimental description, resulting in increased uncertainty in comparisons between calculated and measured quantities.

This chapter discusses the requirements for design and operating information necessary for state-of-the-art computational modelling and simulation. The second part of the chapter addresses the sensitivity of calculations to key input data uncertainties and the typical impact of some of the uncertainties introduced by using commonly approximated data when experimental data are not available.

## 6.1 Required data for validation calculations

Special attention is needed to compile and verify that the necessary information is available for the computational analysts. Compiling and documenting the data required for validation calculations have been recurring challenges for many past experimental programmes. Missing, uncertain or inadequately documented information has frequently limited the value of experimental measurements. Compiling the complete input data necessary to compare measured with computed values requires information from several different organisations including the reactor operator (utility), the fuel manufacturer and engineering companies, and the radiochemical analysis laboratory. Considering the past difficulty in obtaining core management and operating data, these data should be obtained from utilities in parallel or even before the experimental campaign begins. Experience has shown that the earlier the necessary data can be identified and requested, the better the chance of obtaining it. Attempting to reconcile experimental discrepancies and missing operating history data after the programme has ended can frequently have limited success.

There are three general categories of fuel design and reactor operating history data considered in terms of importance for benchmark modelling, as shown in Table 16:

- *Category A – Required data*. Examples of required data include the physical dimensions of the fuel rod and assembly, the locations of water rod and poison rods positions in the assembly, the location of the measured rod in the assembly and axial position of the fuel sample in the fuel rod, and the time-dependent power history for the sample.
- *Category B – Data that can generally be derived from other information*. This category includes data which, if not provided, may be derived or approximated based on other available information or data. These derived data have a low sensitivity on the outcome of the calculations such that the impact of uncertainties associated with the approximations is small. The temperature of the coolant is a typical example of the data in this category. It is required data for the calculation, however, if it is not provided by the reactor operator, it can be accurately estimated, in the case of PWR, based on the inlet and outlet coolant temperature and an assumption that the temperature at the sample position is proportional to the integral of the power from the bottom of the active length to the sample position (i.e. the integral heat deposition). For BWR fuel, thermal energy goes into generating void, making the estimate of coolant density using simple approximations usually unreliable.
- *Category C – Data not necessary for typical analyses*. This type of data will depend on the purpose and type of application. For example, the impurities of the fuel pellet and cladding are not necessary for neutronics calculations. On the other hand, impurity information is required to evaluate the generation of $^{14}C$ which is one of the important radiological source terms for reprocessing plants, and also important in the context of final repository safety.

## 6.2 Sensitivity of modelling to input data uncertainty

In many experimental programmes there will be some physical data identified in Table 16 that are not available, or not documented in sufficient detail for the simulation and thereby influence comparisons of measurements and calculations. This information can include basic design data, or more frequently, it is information related to the reactor operating data and other derived parameters related to plant operation. The notable reactor operating data are power history, operating temperatures of the fuel, boron concentration (PWR), coolant temperature and void fraction (BWR) of the coolant. Other information, such as the use of burnable poison rods in an assembly and insertion of control absorber rods that can be varied during operation and influence the local flux spectrum in the vicinity of the measured sample, are frequently documented with inadequate detail.

**Table 16: Data used in the benchmark description**

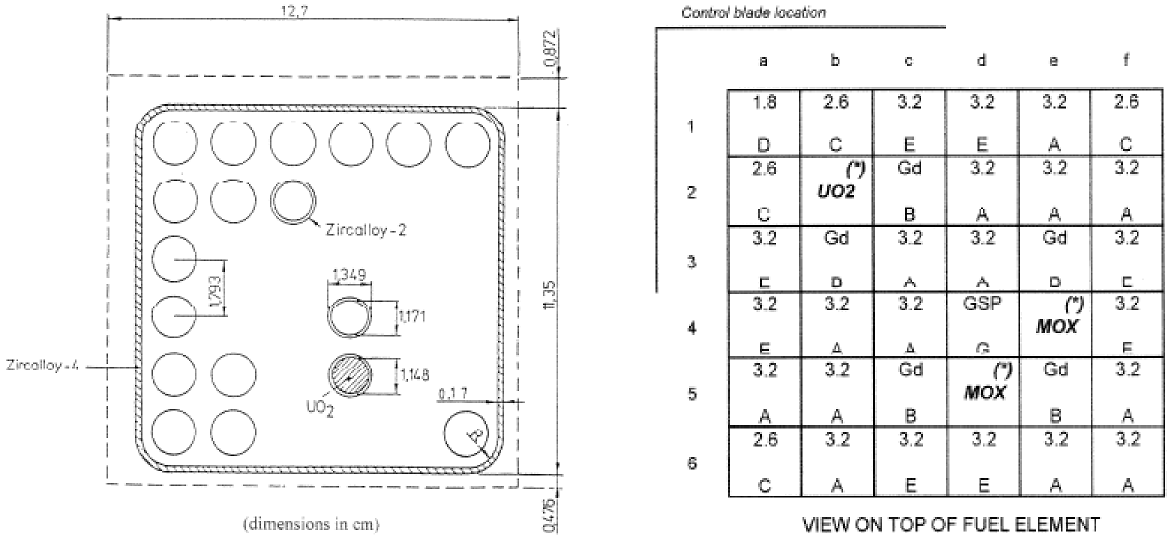
| Type | Category | Parameter |
|---|---|---|
| Reactor | A | Coolant type. |
| | | Composition of control rods (if in proximity of sample). |
| | | Sample burn-up (this value is generally determined from measured assay data). |
| | | Composition of control blades (if in proximity of sample). |
| | B | Nominal system pressure. |
| | | Nominal coolant inlet temperature. |
| | | Nominal coolant outlet temperature. |
| | | Nominal core flow (may be required to calculate fuel temperature). |
| | C | Nominal coolant average temperature. |
| | | Number of assemblies. |

## Table 16: Data used in the benchmark description (*cont.*)

| Type | Category | Parameter |
|---|---|---|
| Assembly design specifications | A | Assembly pitch (wide and narrow water gaps for BWR assemblies). |
| | | Lattice geometry (e.g. 10 × 10). |
| | | Fuel rod pitch. |
| | | Fuel pellet diameter. |
| | | Cladding outside diameter. |
| | | Cladding inside diameter. |
| | | Number of fuel rods (for each type). |
| | | Number of guide tubes/water rods. |
| | | Guide tubes/water rod dimensions. |
| | | Outer flow channel dimensions (for BWR). |
| | | Active rod length (full and partial length rods). |
| | | Configuration of all rod types in the assembly (see example in Figure 12). |
| | | Fuel density – pellet end dishing and chamfer. |
| | | Grid spacer information – if sample in proximity of grid. |
| | B | Fill gas of the fuel rod (pressure and composition). |
| | | Linear fuel weight. |
| | | Length between fuel stack and end plug. |
| Assembly material compositions | A | Fuel material types and initial compositions: includes U/Pu/Am/Gd contents and isotopic composition (uranium fuel should include $^{234}$U and $^{236}$U). |
| | B | Guide tube/water rod/flow tube compositions. |
| | | Clad composition. |
| | C | Impurities in the fuel (N, Li, etc.). For activation product analysis only. |
| | | Impurities in cladding (cobalt in stainless steel for example). Required only for activation and source analysis. |
| Fuel sample data | A | Location of measured fuel rod in assembly. |
| | A | Axial position of sample in rod. |
| Irradiation data (determined at the axial height of the fuel sample)[a] | A | Length of the sample (number of included pellets). |
| | | Cycle start and end dates (or times of irradiation and shutdown). |
| | | Power history of the sample. Frequently derived from assembly burn-up at each end of cycle, or if available the burn-up at the axial height of the sample to account for shifts in the axial power profile. It must reproduce the measured sample burn-up determined from measurements. |
| | | Fuel temperature (e.g. effective resonance temperature, peak centreline temperature, average temperature). It should be requested with operating data, but may be derived analytically or by codes using moderator temperature and fuel power history. |
| | | Coolant temperature and density (at the axial level of the sample). Values in assembly and guide tubes and density outside the flow tubes for BWR. |
| | | Void fraction history at the axial level of the sample. Necessary for BWR analyses. |
| | | Boron concentration variation in the coolant, starting from the beginning of cycle. It should be obtained from utility. |
| | | Assembly exposure to control rod insertion (if present), with the locations (relative to sample), insertion depth, and duration of exposure specified. |
| | B | Clad temperature (at the axial level of the sample). |
| | | Shuffling of assembly and orientation during irradiation. If samples are from a peripheral region, this information is important (Category A). |
| | | Characteristics of adjacent assemblies for each cycle, including design (e.g. 15 × 15), fuel type (UOX, MOX, Gd fuel), average initial fissile content, burn-up. If samples are from a peripheral region, this information is important, particularly for MOX fuel (Category A). |

[a] These values are frequently reported by utilities as time-dependent values.

**Figure 12: Typical BWR assembly design drawings useful in defining physical dimensions (left) and fuel rod configuration and enrichment zoning (right)**



| | a | b | c | d | e | f |
|---|---|---|---|---|---|---|
| 1 | 1.8<br>D | 2.6<br>C | 3.2<br>E | 3.2<br>E | 3.2<br>A | 2.6<br>C |
| 2 | 2.6<br>C | (*)<br>UO2 | Gd<br>B | 3.2<br>A | 3.2<br>A | 3.2<br>A |
| 3 | 3.2<br>E | Gd<br>D | 3.2<br>A | 3.2<br>A | Gd<br>D | 3.2<br>E |
| 4 | 3.2<br>E | 3.2<br>A | 3.2<br>A | GSP<br>G | (*)<br>MOX | 3.2<br>E |
| 5 | 3.2<br>A | 3.2<br>A | Gd<br>B | (*)<br>MOX | Gd<br>B | 3.2<br>A |
| 6 | 2.6<br>C | 3.2<br>A | 3.2<br>E | 3.2<br>E | 3.2<br>A | 3.2<br>A |

This section discusses the potential impact of modelling approximations caused by missing or uncertain data on computed results. The sensitivities of calculated isotopic content to variations in several key design and operating parameters have been studied using a reference model based on the Takahama PWR 17 × 17 assembly. This assembly had an initial $^{235}$U enrichment of 4.1 wt.% and used burnable poison $UO_2$-$Gd_2O_3$ rods. The Takahama assembly was selected because it is a widely used assembly design and has enrichment and burn-up characteristics that are typical of modern fuel design. In addition, some design information (e.g. water gap and neighbour assembly properties) were not reported. Therefore these studies provide general information on the impact of design and operating data uncertainties in addition to addressing the impact of missing data specifically for the Takahama experiments. This section discusses the impact of uncertainties in the following parameters:

- power history;
- moderator temperature (density);
- moderator soluble boron;
- moderator void;
- fuel temperature;
- sample burn-up;
- assembly pitch (defining the gap between assemblies);
- influence of the surrounding assemblies.

There are also physical changes in the fuel dimensions and assembly configuration that occur during irradiation that introduce other uncertainties that are difficult to quantify. These changes are time-dependent and can include: changes in the fuel density (initial reduction in fuel diameter followed by swelling of the fuel), fission gas production and changes in fuel conductivity, and by cladding yield (collapsing) due to coolant pressure.

### *6.2.1 Power history*

The effect of the power history is observed for isotopes which are generated or depleted through radioactive decay characterised by relatively short half-life compared to typical fuel exposure times, such as $^{241}$Am and $^{155}$Gd. Although the period of the reactor start-up and shutdown dates is often available in the open literature, the power history is usually prepared by the utilities using core power measurements and fuel management codes. The ability to perform calculations using detailed power history data therefore depends on the level of detail that can be provided by the utilities.

The importance of using a detailed power history was analysed by JAEA [48] for the Takahama-3 benchmark. In the analysis, a comparison of the results obtained using a detailed power history and constant power history was carried out.

Table 17 summarises the impact of using a detailed power history and a constant power history on the isotopic concentrations as calculated for samples from two fuel rods identified as SF95 and SF97, rods located at the periphery corner and side of the assembly, respectively (see configuration in Figure 14). The results are obtained from the average of five different samples obtained from each rod. The results show that the effect of power history is not large for most actinides and fission products, in general less than 1%, except for $^{144}$Ce, $^{149}$Sm, $^{241}$Am, $^{242m}$Am and $^{243}$Cm. Note that the nuclide concentrations were compared at the time of discharge. At longer cooling times the effect for $^{241}$Am is reduced considerably due to the in-growth of $^{241}$Am from $^{241}$Pu decay.

A sensitivity study was also performed by CEA for the Takahama-3 fuel measurements [120] and similar results were observed. The concentrations of $^{242m}$Am, $^{242}$Cm and $^{243}$Cm, formed by the capture of $^{241}$Am (produced by $^{241}$Pu decay), are the most sensitive to the irradiation history detail. Other isotopes that exhibit a large sensitivity include $^{147}$Sm, $^{149}$Sm, $^{151}$Sm, $^{238}$Pu and $^{242}$Cm. The burn-up credit nuclides that exhibit sensitivity to the power history detail include: $^{238}$Pu, $^{239}$Pu, $^{241}$Pu, $^{241}$Am, $^{242m}$Am, $^{147}$Sm, $^{149}$Sm, $^{150}$Sm, $^{151}$Sm and $^{155}$Gd.

### *6.2.2 Moderator temperature*

The moderator temperature at the sample position is very important information required for the physics calculation. The temperature impacts the moderator density and therefore the thermal flux spectrum in the fuel. The moderator temperature is usually calculated from other reactor operating data including operating pressure, and the inlet and outlet coolant temperatures, assuming that the temperature increase is proportional to the integral of assembly power from the inlet to the axial location of the fuel sample. As discussed, this method cannot be applied to BWR fuel because of the complex coolant flow management that can lead to large variations in the time-dependent axial coolant void during plant operation.

In is important to note that because moderator temperature varies with axial height in the reactor core, the axial location of the fuel sample in the rod is necessary in order to derive the moderator temperature at the sample position. Consequently, any uncertainty in the axial location assigned to the measured sample will introduce an additional component of uncertainty in the input data used in the calculations.

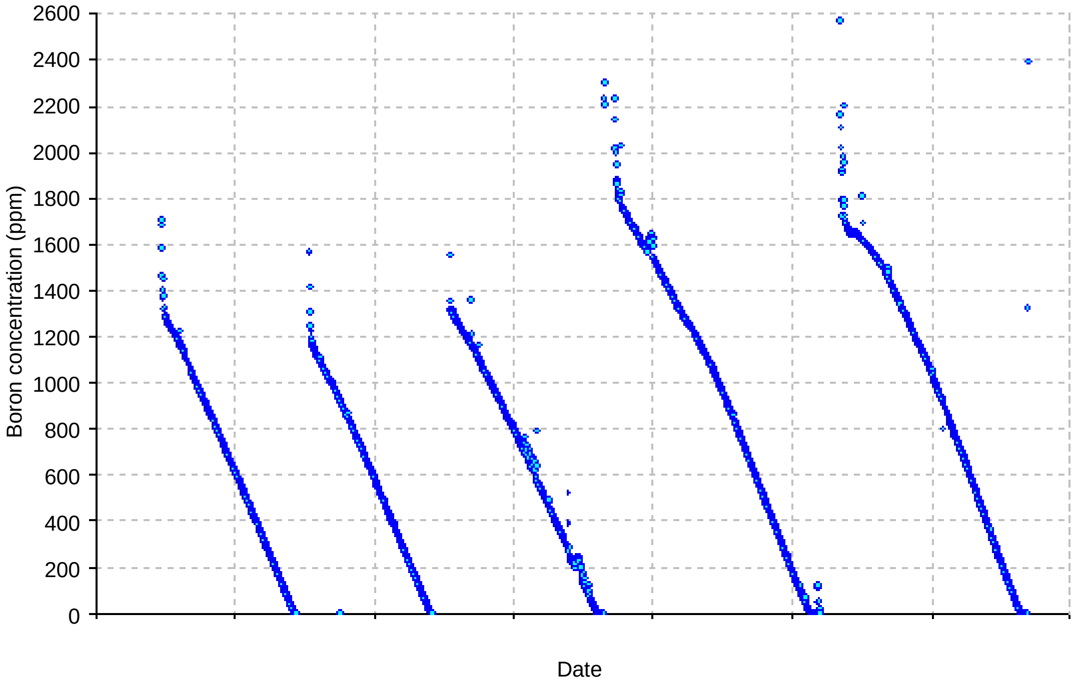
### *6.2.3 Moderator soluble boron*

The soluble boron level in the coolant is an important parameter for the analysis of fuel from PWR plants because of its strong influence on the neutron spectrum in the fuel. The boron concentration typically decreases nearly linearly during reactor operation. The typical variation during several operating cycles is illustrated in Figure 13. The sensitivity of the nuclide results to the level of detail in the boron curve has been studied and it has been shown that using a cycle average concentration gives results that are consistent with a more detailed time-dependent representation.

**Table 17: Comparison of Takahama-3 results for calculations adopting detailed power history and constant power history**

| Isotope | Constant/detailed power history | |
|---|---|---|
| | SF95 average (5 samples) | SF97 average (5 samples) |
| $^{234}$U | 1.00 | 1.00 |
| $^{235}$U | 1.00 | 1.00 |
| $^{236}$U | 1.00 | 1.00 |
| $^{238}$U | 1.00 | 1.00 |
| $^{237}$Np | – | 1.00 |
| $^{238}$Pu | 1.00 | 0.99 |
| $^{239}$Pu | 1.00 | 1.00 |
| $^{240}$Pu | 1.00 | 0.99 |
| $^{241}$Pu | 1.00 | 1.01 |
| $^{242}$Pu | 1.00 | 1.00 |
| $^{241}$Am | 0.96 | 0.97 |
| $^{242m}$Am | 0.96 | 0.97 |
| $^{243}$Am | 1.00 | 1.00 |
| $^{242}$Cm | 0.99 | 1.01 |
| $^{243}$Cm | 1.00 | 1.03 |
| $^{244}$Cm | 1.00 | 1.00 |
| $^{245}$Cm | 1.00 | 1.00 |
| $^{246}$Cm | 1.00 | 1.00 |
| $^{247}$Cm | – | 1.00 |
| $^{134}$Cs | 1.01 | 1.00 |
| $^{137}$Cs | 1.00 | 1.00 |
| $^{144}$Ce | 1.03 | 1.03 |
| $^{142}$Nd | 1.00 | – |
| $^{143}$Nd | 1.00 | 1.00 |
| $^{144}$Nd | 0.99 | 0.99 |
| $^{145}$Nd | 1.00 | 1.00 |
| $^{146}$Nd | 1.00 | 1.00 |
| $^{148}$Nd | 1.00 | 1.00 |
| $^{150}$Nd | 1.00 | 1.00 |
| $^{154}$Eu | 1.00 | 1.00 |
| $^{147}$Sm | – | 1.00 |
| $^{148}$Sm | – | 1.00 |
| $^{149}$Sm | – | 1.02 |
| $^{150}$Sm | – | 1.00 |
| $^{151}$Sm | – | 1.00 |
| $^{152}$Sm | – | 1.00 |
| $^{154}$Sm | – | 1.00 |

Figure 13: Illustration of detailed boron curves during irradiation, shown for five cycles



### 6.2.4 Void fraction

With regard to BWR fuel analyses the value of the void fraction is extremely important for the physics calculation due to the influences on moderator density and thus the hydrogen-to-uranium (H/U) ratio. The general lack of accurate void fraction data for many experiments represents one of the most significant limitations in the ability to accurately model BWR fuel depletion for isotopic validation. Void fraction data are not available for many of the BWR experiments currently included in SFCOMPO. Several analyses using these data have applied derived void data based on other information [121]. Clearly such derived data may have very large uncertainties.

A relatively recent experimental programme in Sweden to measure the decay heat of full-length BWR fuel assemblies using a calorimeter, reported the void profiles provided by the utilities as a function of axial node position. From void data such as that reported in Sweden it may be possible to develop correlations with the assembly type and power of the assembly that may be used to approximate the axial void data for other, similar fuels [122]. As discussed, such extrapolation of data should be done with caution because of the complex coolant flow management that can lead to large variations in the time-dependent axial coolant void during plant operation and can vary for different assemblies in the core.

### 6.2.5 Fuel temperature

Similar to the void information, the fuel temperature is not usually measured directly but it can be evaluated using analytical models and computer codes. Examples of codes used to predict fuel behaviour include FEMAXI, TRANSURANUS, FRAPCOM, FALCON, COSMOS and METEOR. Fuel temperatures are sometimes provided by the utility. In many analyses these values are determined externally and supplied as input to the burn-up analysis code.

Fuel temperature can be reported as peak centreline, physical average or effective temperature for the purposes of calculating Doppler broadening to preserve the resonance absorption in the fuel pin correctly. Note that codes that model the fuel pellet as a single radial material generally require an effective temperature for the purposes of calculating Doppler broadening of the cross-section resonances. An effective temperature is needed since the average volumetric temperature does not correctly account for large variation of radial fuel temperatures and does not preserved the resonance effects. Effective temperatures can also be derived using the centreline and outer fuel temperature [123] or may be available from other calculations.

The fuel temperature is influenced by changes in heat transfer caused by variations in the fuel pellet dimensions and conductivity during irradiation. Changes in fuel characteristics during irradiation have been studied as part of activities of the OECD/NEA Nuclear Science Committee Expert Group on Reactor Plutonium Disposition in close co-operation with the NEA Working Party on Scientific Issues in Reactor Systems (WPRS). The Expert Group activities include fuel benchmark studies (both fuel performance and reactor physics) involving the OECD Halden Reactor Project MOX irradiation experiment and the Belgonucléaire and SCK•CEN PRIMO MOX rod experiment. Benchmarking of computer code predictions against measured fuel temperatures indicates that there is considerable uncertainty in calculated values. Reported benchmarks [124] show that differences in computer model predictions of 100°C or more are not unusual during irradiation.

### 6.2.6 Initial uranium enrichment

The initial $^{235}$U enrichment is usually given with a manufacturing tolerance. The uncertainty in the $^{235}$U enrichment is typically less than 0.05% and this level of uncertainty does not cause a significant uncertainty on the calculated nuclide contents. However, in addition to the $^{235}$U enrichment, it is also important to obtain the enrichments of $^{234}$U and $^{236}$U. These values can vary widely depending on the method of enrichment and country of origin. The final concentration of $^{236}$U does not depend much on the initial $^{236}$U concentration because of the large production from $^{235}$U neutron capture. However, the final $^{234}$U content is very sensitive to the initial $^{234}$U concentration. To a lesser extent, these minor uranium isotopes impact the calculated $^{235}$U concentration.

### 6.2.7 Fuel sample burn-up

The local burn-up of the sample is most frequently derived from the radiochemical analysis results of burn-up indicators like $^{148}$Nd, $^{143+144}$Nd, $^{145+146}$Nd, $^{139}$La and $^{137}$Cs. These nuclides are accurate measures of burn-up since they have relatively large cumulative fission yields and small capture cross-sections, they are stable or long-lived, and the fission yields are similar for $^{235}$U and $^{239}$Pu making the effective fission yield value relatively insensitive to the burn-up. The burn-up determined by $^{148}$Nd is one of the most widely used techniques for fuel burn-up determination [125] because it can be measured accurately using isotopic dilution mass spectrometry methods. However, several independent measures of burn-up are desirable to ensure large errors are not caused by a single measurement problem. Values for sample burn-up determined from operator data are generally not considered sufficiently reliable to be used for code benchmarking because the accuracy is much less than the accuracy of the measurements.

The units of burn-up can also cause problems when comparing the results from different codes. Burn-up is a measure of the integral local energy released by the fuel during irradiation. However, this derived quantity requires values for the recoverable energy per fission for each fissionable nuclide and requires assumptions regarding the definition of local energy deposition and the amount of energy generated by neutron capture on structural materials and fission products and non-fission actinides [126]. Consequently, different codes may produce different estimates of burn-up for the same number of fissions. A more suitable unit for reporting burn-up for radiochemical assay data is the number of fissions relative to the number of heavy metal atoms initially present in the fuel (fissions per initial metal atom, FIMA).

The uncertainty in the derived or estimated sample burn-up can represent one of the largest sources of error in the overall validation process, because the burn-up error influences all of the calculated isotopic results. The practical limit on burn-up accuracy is about 2% due to the measurement accuracy for the burn-up indicators and the nuclear data (e.g. fission yields). The magnitude of the sensitivity for each isotope depends on the burn-up. For example, $^{239}Pu$ is relatively insensitive at high burn-up because the concentration slowly varies, whereas $^{235}U$ is extremely sensitive.

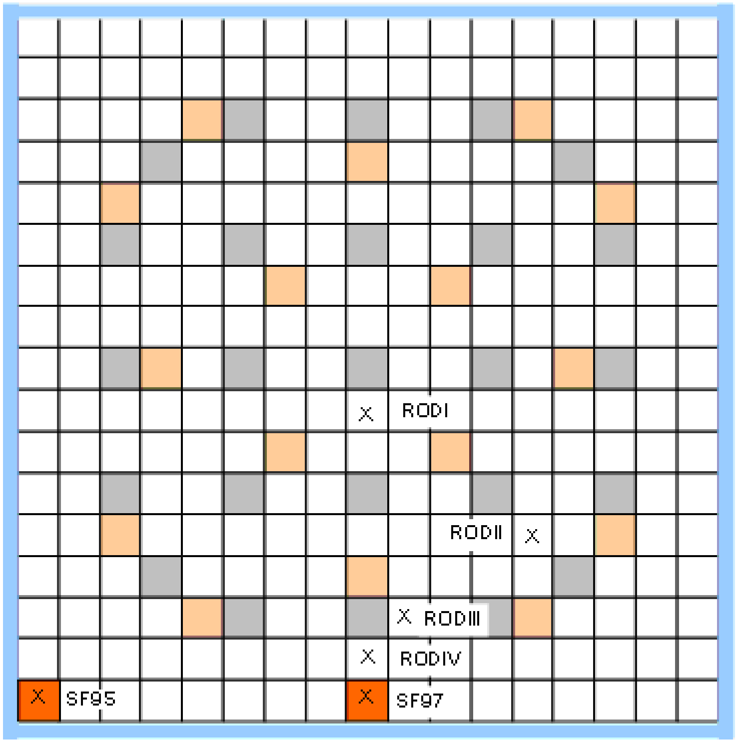
### *6.2.8 Gap between fuel assemblies*

An important assembly design parameter is the water gap between fuel assemblies. This is also defined by the assembly pitch (spacing between the adjacent fuel rods in the assembly). The gap introduces additional water moderator surrounding the assemblies. Modelling studies found that in the case of peripheral rods, the uncertainties in the water gap can represent an important source of modelling and calculation uncertainties for isotopes with a large sensitivity to the neutron spectrum, like $^{239}Pu$. In most experiments the fuel rod pitch is given (although without uncertainty). However, in some experiments, the assembly pitch is not reported, and information available from documents open to the public is not sufficiently precise (±1 mm). It is known that in the case of PWR assemblies the gap between the fuel assemblies is generally very narrow (<1 mm). However, studies have shown that even small variations in the gap can influence the neutron spectrum in the fuel and the calculated compositions.

A sensitivity analysis study performed by the CEA using the Takahama assembly model was used to estimate the uncertainty for several fuel rods positions in the assembly as shown in Figure 14. The rod positions included measured rod SF97, located at the side of the assembly, and several interior rods. The calculations were performed with and without an extra 1 mm water

**Figure 14: Takahama assembly and selected rods used in uncertainty study [4]**

Grey – guide tubes, orange – $Gd_2O_3$ rods, white – $UO_2$ rods, red – measured rods



gap. The gap uncertainty therefore represents an upper limit on the expected uncertainty due to an undocumented assembly pitch value. The results are shown in Table 18. In general there is an observed decrease in $^{235}$U and the plutonium isotopes due to the additional moderation around the assembly that causes a shift of the thermal flux spectrum. The impact is nearly 2% on the concentrations of the main fissile actinides ($^{235}$U, $^{239}$Pu, $^{241}$Pu) at the periphery of the assembly (rod SF97), decreasing below 0.5% for the interior rod (rod I). The results show an enhanced effect for the peripheral rod and reduced impact for the interior rods as expected. The sensitivities of 3% and 5% given at the end of irradiation for the fission products $^{149}$Sm and $^{155}$Gd are reduced to 2% after five years. However, no significant impact from this uncertainty is seen for the metallic fission products or the burn-up indicators since they are not as influenced by the neutron spectrum.

**Table 18: Uncertainties (%) due to water gap (1 mm) for different fuel rod positions in the assembly**

| Isotope | SF97 | Rod IV | Rod III | Rod II | Rod I |
|---|---|---|---|---|---|
| $^{234}$U | 0.3 | 0.2 | 0.1 | 0.2 | 0.1 |
| $^{235}$U | -1.8 | -1.3 | -1.0 | -0.5 | -0.3 |
| $^{236}$U | 0.0 | 0.1 | 0.1 | 0.0 | 0.0 |
| $^{237}$Np | -1.1 | -0.8 | -0.6 | -0.5 | -0.2 |
| $^{238}$Pu | -1.8 | -1.3 | -0.9 | -0.7 | -0.3 |
| $^{239}$Pu | -2.4 | -1.9 | -1.3 | -0.7 | -0.4 |
| $^{240}$Pu | -1.1 | -0.5 | -0.3 | 0.1 | 0.1 |
| $^{241}$Pu | -1.7 | -1.5 | -1.2 | -0.7 | -0.4 |
| $^{242}$Pu | 0.4 | 0.1 | 0.0 | 0.0 | 0.0 |
| $^{241}$Am | -2.3 | -1.9 | -1.4 | -0.8 | -0.4 |
| $^{242m}$Am | -3.5 | -2.9 | -2.2 | -1.3 | -0.7 |
| $^{243}$Am | -0.6 | -0.6 | -0.5 | -0.4 | -0.2 |
| $^{242}$Cm | -1.1 | -0.9 | -0.7 | -0.4 | -0.2 |
| $^{243}$Cm | -1.9 | -1.5 | -1.1 | -0.8 | -0.3 |
| $^{244}$Cm | -1.7 | -1.4 | -1.1 | -0.9 | -0.4 |
| $^{245}$Cm | -4.1 | -3.1 | -2.4 | -1.8 | -0.9 |
| $^{246}$Cm | -2.4 | -1.6 | -1.3 | -1.1 | -0.5 |
| $^{95}$Mo | 0.1 | 0.1 | 0.1 | 0.1 | 0.0 |
| $^{99}$Tc | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 |
| $^{101}$Ru | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| $^{103}$Rh | -0.2 | -0.2 | -0.1 | 0.0 | 0.0 |
| $^{109}$Ag | -0.3 | -0.3 | -0.2 | 0.0 | 0.0 |
| $^{133}$Cs | 0.1 | 0.1 | 0.1 | 0.1 | 0.0 |
| $^{143}$Nd | -0.6 | -0.4 | -0.3 | -0.2 | -0.1 |
| $^{145}$Nd | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 |
| $^{147}$Pm | 0.2 | 0.2 | 0.2 | 0.2 | 0.1 |
| $^{147}$Sm | 0.6 | 0.4 | 0.3 | 0.3 | 0.1 |
| $^{149}$Sm | -2.8 | -2.3 | -1.7 | -1.1 | -0.5 |
| $^{150}$Sm | -0.2 | -0.1 | -0.1 | 0.0 | 0.0 |
| $^{151}$Sm | -2.4 | -1.9 | -1.4 | -0.9 | -0.5 |
| $^{152}$Sm | 0.5 | 0.4 | 0.3 | 0.3 | 0.2 |
| $^{153}$Eu | -0.1 | -0.1 | -0.1 | 0.0 | 0.0 |
| $^{154}$Eu | -1.0 | -0.7 | -0.5 | -0.3 | -0.1 |
| $^{155}$Eu | -2.1 | -1.6 | -1.2 | -0.9 | -0.4 |
| $^{155}$Gd | -4.6 | -3.6 | -2.7 | -1.8 | -1.0 |
| $^{156}$Gd | -0.3 | -0.1 | -0.1 | -0.1 | 0.0 |
| $^{144}$Nd | 0.5 | 0.4 | 0.3 | 0.2 | 0.1 |
| $^{146}$Nd | -0.1 | 0.0 | 0.0 | 0.0 | 0.0 |
| $^{148}$Nd | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| $^{150}$Nd | -0.2 | -0.1 | -0.1 | 0.0 | 0.0 |
| $^{134}$Cs | -0.6 | -0.5 | -0.4 | -0.4 | -0.1 |
| $^{137}$Cs | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |

A similar analysis of the gap uncertainty was performed independently by JAEA. The reference fuel assembly used in the study was also the Takahama PWR 17 × 17 assembly. Burn-up calculations were carried out with and without a water gap between assemblies. The isotopic results were calculated for the average of four samples from rod designated as SF95, taken from the corner rod of the assembly (the CEA studies used a side rod) which is expected to be the most sensitive to the surrounding water gap uncertainty. Again, the nominal value of the gap is less than 1 mm. The calculations were performed using water gap values of 0.0 mm, 0.8 mm and 1.6 mm. Table 19 summarises the results as the ratio of the calculated isotopic concentrations for different values of the water gap, relative to the configuration with no gap. It is shown that $^{239}$Pu is very sensitive to the water gap. When a nominal 0.8 mm gap is included the relative change in the $^{239}$Pu is about 4%. The impact of the gap is larger than that observed in the CEA study (2.4%) for results obtained for the SF97 rod position, which is expected to be less sensitive to the gap than the corner rod used in the JAEA study.

**Table 19: Fraction change in calculated nuclide composition in the SF95 corner rod with assumed water gap dimensions**

| Isotope | Ratio to value with no gap | |
|---|---|---|
| | Gap = 0.8 mm | Gap = 1.6 mm |
| $^{234}$U | 1.01 | 1.02 |
| $^{235}$U | 0.98 | 0.97 |
| $^{236}$U | 1.00 | 1.00 |
| $^{238}$U | 1.00 | 1.00 |
| $^{238}$Pu | 0.96 | 0.92 |
| $^{239}$Pu | 0.96 | 0.93 |
| $^{240}$Pu | 0.98 | 0.98 |
| $^{241}$Pu | 0.97 | 0.94 |
| $^{242}$Pu | 1.00 | 1.00 |
| $^{241}$Am | 0.96 | 0.93 |
| $^{242m}$Am | 0.95 | 0.89 |
| $^{243}$Am | 0.97 | 0.95 |
| $^{242}$Cm | 0.98 | 0.95 |
| $^{243}$Cm | 0.95 | 0.91 |
| $^{244}$Cm | 0.96 | 0.90 |
| $^{245}$Cm | 0.91 | 0.82 |
| $^{246}$Cm | 0.94 | 0.87 |
| $^{142}$Nd | 0.97 | 0.93 |
| $^{143}$Nd | 0.99 | 0.99 |
| $^{144}$Nd | 1.01 | 1.02 |
| $^{145}$Nd | 1.00 | 1.00 |
| $^{146}$Nd | 1.00 | 1.00 |
| $^{148}$Nd | 1.00 | 1.00 |
| $^{150}$Nd | 1.00 | 0.99 |
| $^{106}$Ru | 0.99 | 0.98 |
| $^{125}$Sb | 1.00 | 0.99 |
| $^{134}$Cs | 0.98 | 0.97 |
| $^{137}$Cs | 1.00 | 1.00 |
| $^{144}$Ce | 1.00 | 1.00 |
| $^{154}$Eu | 0.96 | 0.93 |

From the perspective of experiment design and fuel rod selection, these results emphasise the importance of having precise geometry data of fuel assembly design and dimensions of the water gap if fuel rods are selected from locations at the periphery of the assembly, especially the corner locations. Note that uncertainties in the gap may also arise due to changes in the assembly configuration that can occur during irradiation, particularly for high burn-up fuels where deformation and bowing of the rods are known to occur. Selecting fuel rods from the inner region of the assembly, away from the periphery or other perturbing effects, will reduce the influence of some of these uncertainties in the calculational model. However, selecting only unperturbed rods also results in an undersampling of rods in the assembly and may lead to an underestimate of the calculated isotopic uncertainties.

### *6.2.9 Effect of the surrounding fuel assemblies*

In many assay experiments, the surrounding conditions of the measured fuel assembly are not reported and are not available from open sources. Design and operating data are in many cases available only for the host fuel assembly of the measured fuel rod. In the past, this level of information was usually sufficient because with the capabilities of the codes available it was not possible or practical to include the effects of neighbouring assemblies. Hence, detailed information on the surroundings was not needed and therefore generally not documented. However, because of the rapid increase in computing power and progress in the development of the calculational models with two- and three-dimensional capability, and the accumulated experience with fuel depletion analyses, modelling of the surrounding assemblies can be achieved and has been demonstrated to be important in some conditions.

#### *6.2.9.1 PWR UOX fuels*

Advances in computational capability now allow modelling details representing not only the fuel rod and mother assembly, but also the surrounding assemblies. However, commensurate with this ability to represent the detailed environment it is necessary to have large amounts of additional information available for all adjacent assemblies during each cycle that the mother assembly was irradiated. This can rapidly lead to information overload and the need for automated methods to handle the large amount of time-dependent and location-dependent isotopic data.

The impact of the composition of the surrounding assemblies was studied by the CEA, again for the Takahama configuration shown in Figure 14, by performing the depletion calculations using a reference reflective boundary assembly model (infinite pattern of assemblies) and a model with the surrounding assembly fuel compositions corresponding to a burn-up of 24 GWd/t (typical of one-cycle irradiated fuel). The results in Table 20 show the relative differences in the results for the two models. The results show a significant impact on the peripheral rod SF97 (-3% on the $^{239}$Pu concentration and -1% on the $^{235}$U concentration). There is no noticeable effect on the inner rod position, with the sensitivity of the calculated concentrations being generally <1%.

Another more detailed evaluation of the impact of the surrounding assembly was performed by the NRI [127] using the Takahama assembly model. To explore the effect of nearest assembly neighbours under a variety of possible scenarios, four different kinds of boundary conditions were employed: *i)* reflection (reference case); *ii)* non-depleting neighbour assemblies similar to the study performed by the CEA; *iii)* depleting neighbour assemblies; *iv)* refuelling with changing surroundings (as in real core operation). The reference case used reflective boundary conditions (typically used when the surroundings are unknown or not examined, which is often the case).

Figure 15 shows the model with surrounding assemblies included – four quarters of an assembly were modelled. The surrounding assemblies are each approximated for model simplicity as a single fuel mixture. The initial compositions of depleted one-cycle and two-cycle neighbour assemblies were calculated for burn-up values of 15 and 30 GWd/t, respectively.

The simplest case of modelling neighbours is to put the same composition into the neighbour assemblies and either let them deplete or hold them constant. The latter option was not performed using fresh fuel, since it is an unrealistic condition since no equilibrium fission products would be present and the neutron absorption in the assembly would be strongly underestimated. More realistic conditions were simulated by letting the neighbouring assemblies deplete during irradiation and then change (reshuffle) them after each depletion cycle. Although the real shuffling schemes are often not known, a reasonable range of possible scenarios is covered by four shuffling schemes shown in Figure 16.

**Table 20: Impact (%) of using depleted surrounding assemblies compared to a reference model using reflective boundaries on the calculated isotopic contents**

| Isotope | Rod SF97 | | Rod "I" | |
|---|---|---|---|---|
| | EOL | 5 years | EOL | 5 years |
| $^{234}$U | 0 | 0 | 1 | 0 |
| $^{235}$U | -1 | -1 | 1 | 1 |
| $^{236}$U | 0 | 0 | 0 | 0 |
| $^{237}$Np | -1 | -1 | -1 | -1 |
| $^{238}$Pu | -2 | -2 | -1 | -1 |
| $^{239}$Pu | -3 | -3 | 0 | 0 |
| $^{240}$Pu | -2 | -2 | 0 | 0 |
| $^{241}$Pu | -2 | -2 | -1 | -1 |
| $^{242}$Pu | -2 | -2 | -1 | -1 |
| $^{241}$Am | -3 | -2 | 0 | -1 |
| $^{242m}$Am | -4 | -4 | 0 | 0 |
| $^{243}$Am | -3 | -3 | -2 | -2 |
| $^{242}$Cm | -3 | -3 | -1 | -1 |
| $^{243}$Cm | -5 | -5 | -2 | -2 |
| $^{244}$Cm | -5 | -5 | -3 | -3 |
| $^{245}$Cm | -8 | -8 | -4 | -4 |
| $^{246}$Cm | -8 | -8 | -4 | -4 |
| $^{95}$Mo | 0 | 0 | 0 | -1 |
| $^{99}$Tc | 0 | 0 | 0 | 0 |
| $^{101}$Ru | 0 | 0 | -1 | -1 |
| $^{103}$Rh | -1 | -1 | 0 | 0 |
| $^{109}$Ag | -2 | -2 | -1 | -1 |
| $^{133}$Cs | 0 | 0 | 0 | 0 |
| $^{143}$Nd | -1 | -1 | 0 | 0 |
| $^{145}$Nd | 0 | 0 | 0 | 0 |
| $^{147}$Pm | 0 | 0 | 0 | 0 |
| $^{147}$Sm | 1 | 0 | 0 | 0 |
| $^{149}$Sm | -2 | -1 | -1 | -1 |
| $^{150}$Sm | -1 | -1 | -1 | -1 |
| $^{151}$Sm | -2 | -2 | -1 | -1 |
| $^{152}$Sm | 0 | 0 | 0 | 0 |
| $^{153}$Eu | -1 | -1 | -1 | -1 |
| $^{154}$Eu | -2 | -2 | -1 | -1 |
| $^{155}$Eu | -3 | -3 | -1 | -1 |
| $^{155}$Gd | -4 | -3 | -1 | -1 |
| $^{156}$Gd | -2 | -2 | -1 | -1 |
| $^{144}$Nd | 0 | 0 | -1 | -1 |
| $^{146}$Nd | -1 | -1 | -1 | -1 |
| $^{148}$Nd | 0 | 0 | -1 | -1 |
| $^{150}$Nd | -1 | -1 | -1 | -1 |
| $^{134}$Cs | -1 | -1 | -1 | -1 |
| $^{137}$Cs | -1 | -1 | -1 | -1 |

**Figure 15: Model for the case with three nearest-neighbour (1/4) assemblies [124]**

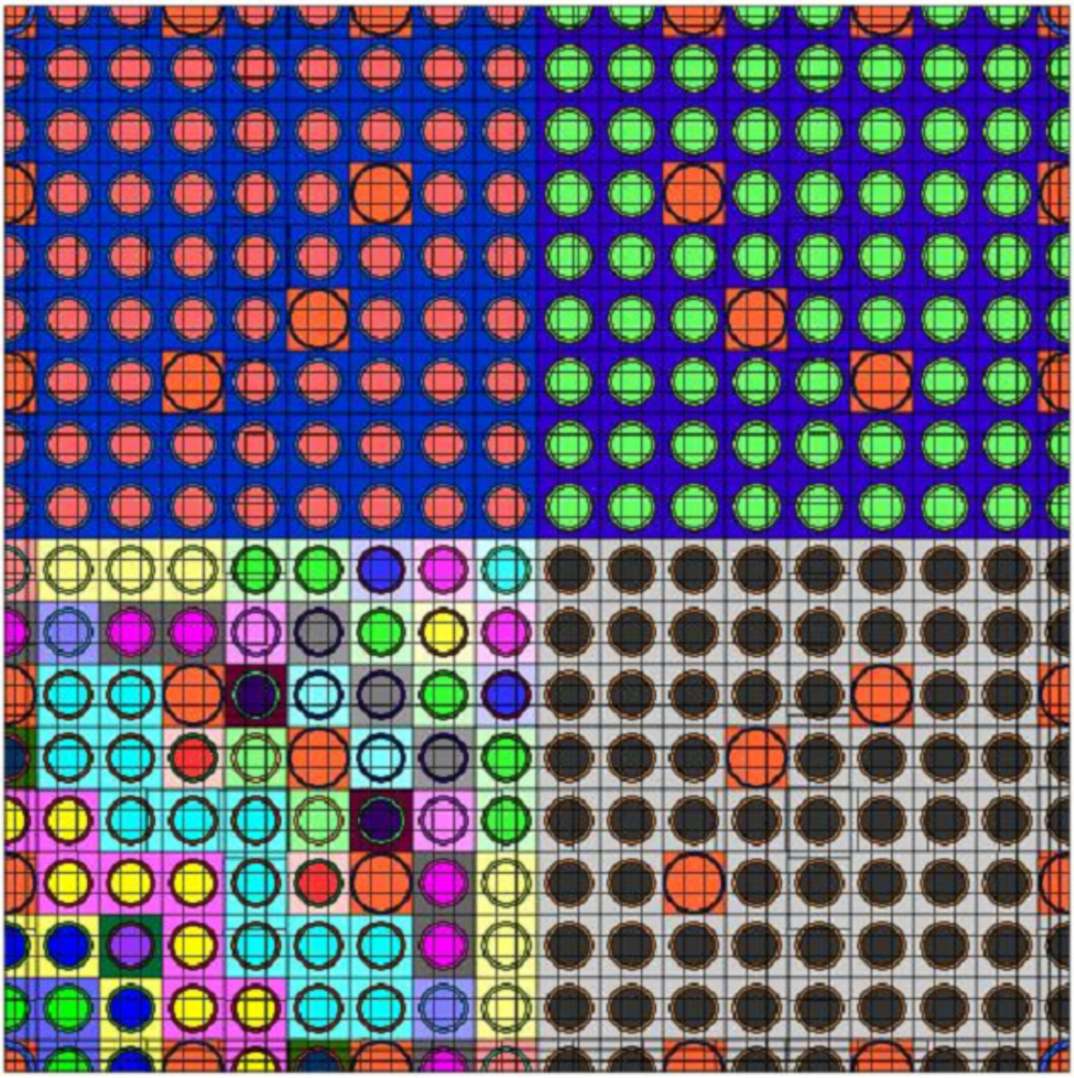
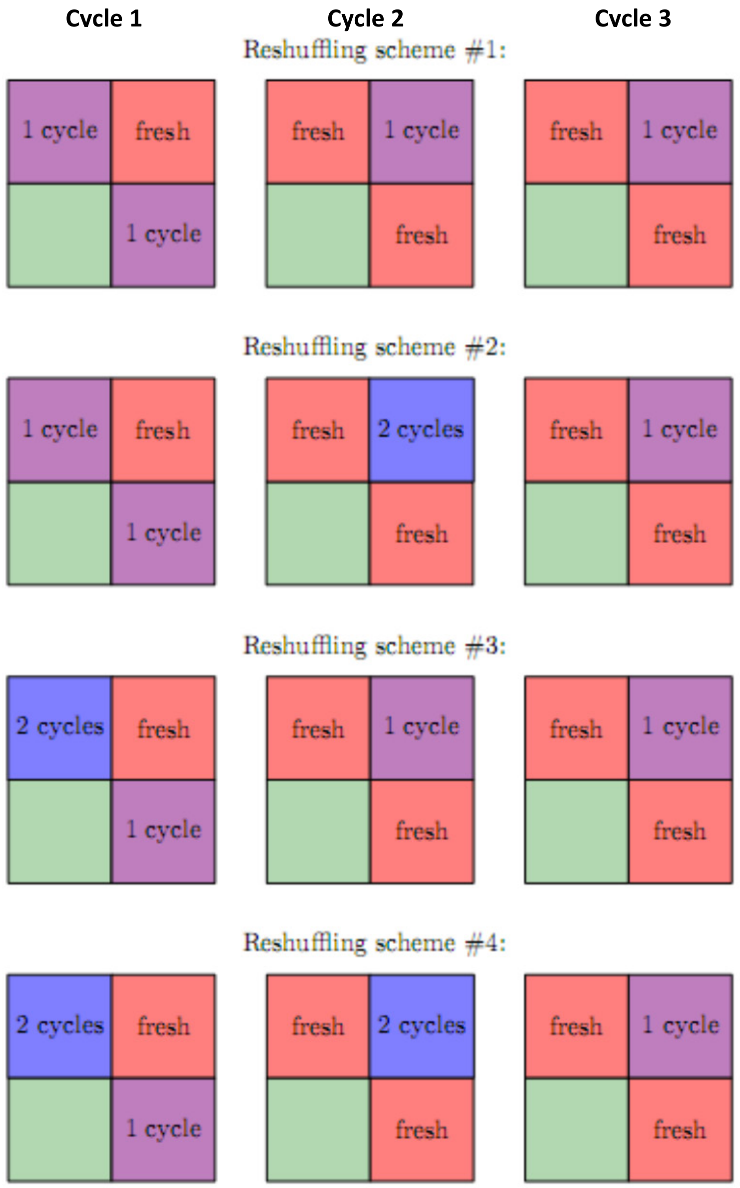


**Figure 16: Assembly shuffling schemes showing neighbour assembly initial cycle compositions [124]**

The neighbour assembly loading pattern for each of three cycles is shown for each scheme



The results shown in Table 21 (corner rod SF95) and Table 22 (periphery side rod SF97) show that the effect of the neighbour assemblies is more pronounced for the corner rod than for the side rod. Also, the largest differences with the reference case (reflection) are seen for the two-cycle assembly surroundings with depletion. This latter situation represents a high burn-up surrounding environment that is heavily absorbing compared to actual conditions.

It should be noted that the case with surrounding fuel assemblies with initial fresh fuel and allowed to burn should give the same results as the reference case (with reflection). The reason for the small discrepancy lies in the approximate modelling of the surroundings which were assumed to have a uniform composition in each pin of the assembly. For better accuracy, the fuel rods in the surrounding assemblies could be depleted independently, thus decoupling the outer rods and inner rods of the assembly.

### Table 21: Relative effect (%) of neighbour assemblies for the assay data evaluation (SF95)

| Isotope | Shuffling schemes* [see Figure 16] | | | | Depleting surrounding | | | Constant surroundings | |
|---|---|---|---|---|---|---|---|---|---|
| | #1 | #2 | #3 | #4 | One-cycle | Two-cycle | Fresh fuel | One-cycle | Two-cycle |
| $^{235}$U | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | -0.1 | 0.0 | 0.1 |
| $^{236}$U | 0.1 | 0.1 | 0.1 | 0.1 | 0.5 | 0.5 | 0.1 | 0.3 | 0.4 |
| $^{238}$U | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| $^{237}$Np | -0.8 | -0.5 | -0.7 | -0.4 | 2.8 | 3.8 | -1.6 | 0.9 | 2.6 |
| $^{238}$Pu | -0.7 | -0.6 | -0.5 | -0.4 | 3.1 | 3.5 | -2.3 | 1.0 | 2.2 |
| $^{239}$Pu | -0.4 | -0.2 | -0.3 | -0.1 | 3.3 | 4.1 | -0.9 | 1.7 | 3.2 |
| $^{240}$Pu | 1.2 | 1.2 | 1.3 | 1.3 | 1.5 | 1.6 | -0.5 | 1.2 | 1.4 |
| $^{241}$Pu | -0.1 | -0.4 | -0.1 | -0.4 | -0.6 | -2.5 | -1.2 | -1.4 | -3.1 |
| $^{242}$Pu | 0.2 | -0.3 | -0.1 | -0.5 | -2.1 | -4.7 | -0.9 | -2.1 | -4.9 |
| $^{241}$Am | -0.3 | -0.5 | -0.5 | -0.7 | -0.1 | -2.1 | -1.3 | -1.0 | -2.7 |
| $^{243}$Am | -1.5 | -1.8 | -1.9 | -2.2 | -0.5 | -3.8 | -1.4 | -2.3 | -4.8 |
| $^{242}$Cm | -0.5 | -1.0 | -0.9 | -1.4 | -2.1 | -5.4 | -1.4 | -2.6 | -5.8 |
| $^{243}$Cm | -2.0 | -2.2 | -2.6 | -2.7 | -0.2 | -3.4 | -1.9 | -2.5 | -4.8 |
| $^{244}$Cm | -3.0 | -3.4 | -3.6 | -4.0 | -1.3 | -6.1 | -2.1 | -3.8 | -7.4 |
| $^{106}$Ru | 0.0 | 0.1 | 0.1 | 0.1 | 0.8 | 0.8 | -0.7 | 0.3 | 0.5 |
| $^{125}$Sb | 0.1 | 0.1 | 0.2 | 0.2 | 0.3 | 0.2 | -0.2 | 0.1 | 0.2 |
| $^{134}$Cs | -1.0 | -0.9 | -1.0 | -0.9 | 1.1 | 0.8 | -0.5 | -0.2 | 0.0 |
| $^{137}$Cs | 0.1 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 |
| $^{143}$Nd | 0.0 | 0.0 | 0.0 | 0.0 | -0.1 | -0.1 | 0.1 | 0.0 | -0.1 |
| $^{144}$Ce | 0.0 | 0.0 | 0.0 | 0.0 | -0.1 | -0.1 | 0.1 | 0.0 | -0.1 |
| $^{144}$Nd | 0.0 | 0.0 | 0.0 | 0.0 | -0.2 | -0.2 | 0.1 | -0.1 | -0.2 |
| $^{145}$Nd | 0.0 | 0.0 | 0.0 | 0.0 | -0.1 | -0.1 | 0.1 | -0.1 | -0.1 |
| $^{146}$Nd | 0.0 | 0.0 | -0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| $^{148}$Nd | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| $^{150}$Nd | -0.1 | -0.1 | -0.1 | 0.0 | 0.1 | 0.2 | -0.2 | 0.0 | 0.1 |
| $^{154}$Eu | -1.1 | -0.9 | -1.0 | -0.8 | 2.5 | 2.8 | -0.8 | 0.7 | 1.9 |
| $^{147}$Sm | 0.1 | 0.1 | 0.1 | 0.1 | -0.3 | -0.3 | 0.1 | -0.1 | -0.1 |
| $^{148}$Sm | -1.5 | -1.4 | -1.5 | -1.4 | 1.2 | 1.0 | -0.6 | 0.0 | 0.3 |
| $^{149}$Sm | -0.8 | -0.8 | -0.8 | -0.7 | 0.8 | 0.4 | -1.0 | -0.8 | -0.5 |
| $^{150}$Sm | 0.1 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | -0.1 | 0.0 | 0.0 |
| $^{151}$Sm | -0.4 | -0.4 | -0.4 | -0.3 | 0.5 | 0.4 | -0.7 | -0.2 | 0.0 |
| $^{152}$Sm | 0.2 | 0.2 | 0.2 | 0.2 | -0.2 | -0.1 | 0.0 | 0.1 | 0.0 |
| $^{154}$Sm | 0.1 | 0.1 | 0.1 | 0.1 | 0.4 | 0.5 | -0.4 | 0.2 | 0.4 |

* Reshuffling schemes #1-#4; cases assume various initial concentrations for surrounding assemblies and allow surrounding assembly compositions to deplete during irradiation; constant surrounding cases hold the surrounding assembly concentrations constant with irradiation.

### Table 22: Relative effect (%) of neighbour assemblies for the assay data evaluation (SF97)

| Isotope | Shuffling schemes* [see Figure 16] | | | | Depleting surrounding | | | Constant surroundings | |
|---|---|---|---|---|---|---|---|---|---|
| | #1 | #2 | #3 | #4 | One-cycle | Two-cycle | Fresh fuel | One-cycle | Two-cycle |
| $^{235}$U | -0.1 | -0.1 | -0.1 | 0.0 | 0.2 | 0.3 | -0.2 | 0.0 | 0.1 |
| $^{236}$U | -0.1 | -0.1 | -0.1 | -0.1 | 0.1 | 0.0 | 0.1 | 0.0 | 0.0 |
| $^{238}$U | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| $^{237}$Np | -1.8 | -1.7 | -1.8 | -1.7 | 1.3 | 1.7 | -0.3 | -0.6 | 0.5 |
| $^{238}$Pu | -1.7 | -1.6 | -1.6 | -1.5 | 0.9 | 0.8 | -0.8 | -0.9 | -0.3 |
| $^{239}$Pu | -2.4 | -2.4 | -2.4 | -2.3 | 1.7 | 2.3 | -0.2 | -0.5 | 1.0 |
| $^{240}$Pu | 0.4 | 0.5 | 0.5 | 0.5 | 1.0 | 1.5 | -0.4 | 0.6 | 1.2 |
| $^{241}$Pu | -0.2 | -0.2 | -0.2 | -0.2 | -0.2 | -1.1 | -0.4 | -0.8 | -1.4 |
| $^{242}$Pu | 1.1 | 1.0 | 1.1 | 1.0 | -1.3 | -2.7 | -0.4 | -0.6 | -2.3 |
| $^{241}$Am | -0.5 | -0.5 | -0.5 | -0.5 | 0.1 | -0.7 | -0.6 | -0.8 | -1.2 |

**Table 22: Relative effect (%) of neighbour assemblies for the assay data evaluation (SF97) (*cont.*)**

| Isotope | Shuffling schemes* [see Figure 16] | | | | Depleting surrounding | | | Constant surroundings | |
|---|---|---|---|---|---|---|---|---|---|
| | #1 | #2 | #3 | #4 | One-cycle | Two-cycle | Fresh fuel | One-cycle | Two-cycle |
| $^{243}$Am | -1.5 | -1.7 | -1.7 | -1.8 | -1.2 | -3.2 | -0.4 | -2.0 | -3.5 |
| $^{242}$Cm | 0.6 | 0.5 | 0.5 | 0.4 | -1.3 | -2.9 | -0.6 | -1.1 | -2.8 |
| $^{243}$Cm | -1.7 | -1.8 | -1.8 | -1.9 | -0.8 | -2.5 | -0.5 | -2.2 | -3.3 |
| $^{244}$Cm | -2.6 | -2.7 | -2.9 | -3.0 | -1.9 | -4.9 | -0.4 | -3.1 | -5.4 |
| $^{106}$Ru | -0.2 | -0.2 | -0.2 | -0.2 | 0.4 | 0.4 | -0.2 | -0.1 | 0.1 |
| $^{125}$Sb | 0.0 | 0.1 | 0.1 | 0.1 | 0.2 | 0.2 | -0.1 | 0.1 | 0.1 |
| $^{134}$Cs | -1.6 | -1.6 | -1.6 | -1.6 | 0.2 | -0.2 | 0.1 | -0.8 | -0.8 |
| $^{137}$Cs | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| $^{143}$Nd | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 |
| $^{144}$Ce | 0.1 | 0.1 | 0.1 | 0.1 | -0.1 | -0.1 | 0.0 | 0.0 | 0.0 |
| $^{144}$Nd | 0.1 | 0.1 | 0.1 | 0.1 | -0.2 | -0.2 | 0.1 | 0.0 | -0.1 |
| $^{145}$Nd | 0.1 | 0.1 | 0.1 | 0.1 | -0.1 | -0.1 | 0.0 | 0.1 | 0.0 |
| $^{146}$Nd | -0.1 | -0.1 | -0.1 | -0.1 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 |
| $^{148}$Nd | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| $^{150}$Nd | -0.1 | -0.1 | -0.1 | -0.1 | 0.1 | 0.2 | -0.1 | 0.0 | 0.1 |
| $^{154}$Eu | -2.6 | -2.5 | -2.5 | -2.5 | 1.0 | 1.0 | 0.0 | -1.0 | 0.0 |
| $^{147}$Sm | 0.4 | 0.4 | 0.4 | 0.4 | -0.2 | 0.0 | 0.0 | 0.2 | 0.2 |
| $^{148}$Sm | -2.0 | -2.0 | -2.0 | -2.0 | 0.3 | 0.2 | -0.1 | -0.7 | -0.4 |
| $^{149}$Sm | -1.2 | -1.1 | -1.1 | -1.1 | 0.8 | 0.8 | 0.0 | -1.2 | -0.5 |
| $^{150}$Sm | 0.2 | 0.2 | 0.2 | 0.2 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| $^{151}$Sm | -1.0 | -1.0 | -1.0 | -0.9 | 0.7 | 0.8 | -0.1 | -0.6 | -0.1 |
| $^{152}$Sm | 0.6 | 0.6 | 0.6 | 0.7 | -0.1 | 0.0 | -0.1 | 0.3 | 0.2 |
| $^{154}$Sm | -0.1 | -0.1 | -0.1 | 0.0 | 0.3 | 0.4 | -0.2 | 0.0 | 0.2 |

* Reshuffling schemes #1-#4; cases assume various initial concentrations for surrounding assemblies and allow surrounding assembly compositions to deplete during irradiation; constant surrounding cases hold the surrounding assembly concentrations constant with irradiation.

Although there is no gap between the assemblies of this simulation model, which increases the effect of interactions of adjacent assemblies, the effect of the neighbours is not large, even for the corner rod under quite extreme and in some cases unrealistic conditions (e.g. surrounding fuel corresponding to constant 30 GWd/t compositions during irradiation). The results suggests that little accuracy is lost by neglecting the neighbour assemblies and using reflective boundary conditions, since the four shuffling cases (which are far closer to the reality than the other cases) yield isotopic concentrations that are generally within 1% of the reference reflection case. The results obtained using all four fuel shuffling schemes are observed to be in very good agreement indicating that the details of the shuffling are not critical since they lead to a similar impact on the isotopic contents. The CEA results are also consistent with the NRI results for the situation of one cycle depleting surrounding assemblies. The effect on $^{239}$Pu in the CEA calculations was 3% compared to 2.3% in the NRI study for similar assumptions.

It is expected where the surrounding assemblies are significantly different from the modelled assembly that greater differences would be seen between the reflected single assembly model and a multi-assembly (supercell) model, for example, where surrounding assemblies have very different enrichments, burn-ups, burnable poison rods or MOX pins. The example of MOX fuel is studied in the following section.

### *6.2.9.2 PWR MOX fuels*

The importance of modelling the surrounding fuel assemblies for analyses of the array of MOX and $UO_2$ fuel assemblies on MOX fuel compositions has been studied. The effect is especially pronounced for samples obtained from fuel rods located at the periphery of the MOX assembly where interactions between MOX and $UO_2$ fuel assemblies results in large changes in the neutron spectrum particularly in the region of the interface. As the location of the measured fuel rods shifts away from the assembly periphery towards the assembly centre, the influence of the surroundings is reduced. Models that consider only one assembly type cannot account for the strong influences of the neighbour assemblies.

For PWR-MOX fuel analyses, it was shown by the CEA [128] that the description of the UOX surrounding was necessary to properly describe the thermal flux spectrum at the boundary of the MOX assembly. The surrounding UOX assemblies define the thermal flux entering the MOX assembly. The errors introduced using an infinite pattern of MOX assemblies (reflective boundary) are large, particularly for the actinides in the peripheral rods. The errors can be estimated from the deviations for the two model cases in the peripheral fuel zone of a MOX assembly from St. Laurent B1 nuclear power plant. To summarise the deviations observed in the calculated isotopic composition for several major actinides between a reflective case and a supercell case modelling the UOX assembly surroundings, the following per cent variations were observed:

| | |
|---|---|
| $^{239}$Pu | +28% |
| $^{235}$U | +12% |
| $^{241}$Pu | +18% |
| $^{241}$Am | +20% |

Notably to the contrary, it has been found that for modelling of the UOX assemblies in a MOX core the influence of the MOX assemblies is relatively minor and models using an infinite pattern of assemblies (reflection) are found to be adequate.

## 6.3 Example sensitivity calculations

The uncertainties in the calculated spent fuel isotope contents resulting from typical input value uncertainties have been calculated via the direct propagation of the uncertainties by comparing the results for a reference calculation [129] and a calculation performed with the perturbed value of the considered parameter. The sensitivity of the fuel inventory was calculated for several of the most important input parameters using the following representative experimental uncertainties ($1\sigma$):

- $^{235}$U enrichment: ±0.05 wt.%.
- Fuel temperature: ±50°C.
- Moderator temperature: ±2°C.
- Fuel sample (local) burn-up: ±2%.

The influence of uncertainties in the initial enrichment, the fuel and the moderator temperature, and the local burn-up are summarised in Table 23. The combined quadratic sum of all these contributions are also given to provide an estimate of the total uncertainty associated with each isotope concentration, assuming the effects are not correlated. Although the estimated uncertainties in each parameter are assumed to be typical, the individual and combined uncertainties are provided for illustrative purposes only. The enrichment variation in particular is considered to be larger than in practice, with the typical $^{235}$U uncertainty generally being about 0.01 to 0.005 wt.%.

The total uncertainties for the major actinides ($^{239}$Pu, $^{240}$Pu, $^{241}$Pu) are of the order of 1 to 2%, except for $^{235}$U whose total uncertainty is about 5% due to uncertainties in the initial $^{235}$U concentration and in the burn-up component. The uncertainties are largest for the isotopes at the end of the transmutation chain, especially the curium isotopes (uncertainties of 10% or more), which are most sensitive to changes in burn-up. Uncertainties are of the order of 2% for

**Table 23: Relative nuclide uncertainties due to the input parameters of initial enrichment, fuel temperature, moderator temperature and sample burn-up, calculated for a fuel burn-up of 46 GWd/t**

| Isotope | End of irradiation (no cooling time) | | | | |
|---|---|---|---|---|---|
| | $^{235}$U +0.05 wt.% | Fuel temp. +50°C | Moderator Temp. +2°C | Burn-up 2% | Total uncertainty |
| $^{234}$U | 0.5% | 0.2% | -0.1% | -1.5% | 1.6% |
| $^{235}$U | 3.1% | 0.6% | 0.5% | -4.0% | 5.1% |
| $^{236}$U | 1.0% | -0.1% | 0.0% | 0.7% | 1.3% |
| $^{237}$Np | 0.4% | 0.3% | 0.3% | 2.2% | 2.3% |
| $^{238}$Pu | -0.2% | 0.2% | 0.6% | 4.3% | 4.3% |
| $^{239}$Pu | 0.5% | 0.7% | 0.6% | -0.1% | 1.0% |
| $^{240}$Pu | -0.3% | 0.2% | 0.3% | 1.6% | 1.6% |
| $^{241}$Pu | 0.0% | 0.7% | 0.6% | 1.2% | 1.5% |
| $^{242}$Pu | -1.3% | 0.2% | 0.0% | 4.4% | 4.6% |
| $^{241}$Am | 0.5% | 0.9% | 0.6% | -0.2% | 1.2% |
| $^{242m}$Am | 0.7% | 1.1% | 1.1% | -0.3% | 1.8% |
| $^{243}$Am | -1.7% | 0.3% | 0.4% | 6.1% | 6.3% |
| $^{242}$Cm | -0.6% | 0.4% | 0.4% | 3.3% | 3.4% |
| $^{243}$Cm | -1.1% | 0.4% | 0.7% | 5.3% | 5.5% |
| $^{244}$Cm | -2.4% | 0.1% | 0.7% | 9.0% | 9.4% |
| $^{245}$Cm | -2.4% | 0.3% | 1.5% | 10.4% | 10.8% |
| $^{246}$Cm | -3.8% | -0.3% | 0.9% | 14.4% | 14.9% |
| $^{95}$Mo | 0.2% | 0.0% | -0.1% | 1.7% | 1.7% |
| $^{99}$Tc | 0.1% | 0.0% | 0.0% | 1.7% | 1.7% |
| $^{101}$Ru | 0.0% | 0.0% | 0.0% | 2.0% | 2.0% |
| $^{103}$Rh | 0.1% | 0.2% | 0.0% | 1.3% | 1.4% |
| $^{109}$Ag | -0.7% | 0.3% | 0.1% | 2.8% | 2.9% |
| $^{133}$Cs | 0.2% | 0.0% | 0.0% | 1.6% | 1.6% |
| $^{143}$Nd | 0.6% | 0.2% | 0.2% | 0.8% | 1.0% |
| $^{145}$Nd | 0.2% | 0.0% | 0.0% | 1.5% | 1.6% |
| $^{147}$Pm | 0.5% | 0.2% | -0.1% | 0.5% | 0.7% |
| $^{147}$Sm | 0.5% | 0.1% | -0.2% | 0.1% | 0.6% |
| $^{149}$Sm | 1.1% | 0.7% | 0.9% | -0.3% | 1.6% |
| $^{150}$Sm | 0.0% | 0.0% | 0.1% | 2.3% | 2.3% |
| $^{151}$Sm | 0.7% | 0.5% | 0.9% | 0.5% | 1.3% |
| $^{152}$Sm | 0.0% | 0.0% | -0.2% | 1.5% | 1.5% |
| $^{153}$Eu | -0.2% | 0.0% | 0.1% | 2.5% | 2.5% |
| $^{154}$Eu | -0.4% | 0.0% | 0.3% | 3.7% | 3.7% |
| $^{155}$Eu | -0.2% | 0.2% | 0.5% | 3.1% | 3.1% |
| $^{155}$Gd | 0.8% | 0.9% | 1.5% | 0.7% | 2.1% |
| $^{156}$Gd | -1.2% | -0.2% | 0.1% | 5.5% | 5.6% |
| $^{144}$Nd | -0.1% | -0.2% | -0.2% | 2.6% | 2.6% |
| $^{146}$Nd | 0.0% | 0.0% | 0.0% | 2.3% | 2.3% |
| $^{148}$Nd | 0.0% | 0.0% | 0.0% | 2.1% | 2.1% |
| $^{150}$Nd | -0.1% | 0.0% | 0.0% | 2.3% | 2.3% |
| $^{134}$Cs | -0.4% | -0.1% | 0.2% | 4.0% | 4.0% |
| $^{137}$Cs | 0.1% | 0.0% | 0.0% | 2.0% | 2.0% |
| ***Cooling time 5 years*** | | | | | |
| $^{242}$Cm | -0.4% | 0.6% | 0.6% | 2.7% | 2.8% |
| $^{149}$Sm | 0.6% | 0.4% | 0.6% | 1.0% | 1.3% |
| $^{155}$Gd | -0.1% | 0.2% | 0.5% | 3.0% | 3.1% |

the metallic fission products and for the neodymium isotopes. The major component of the total uncertainty is due to the burn-up uncertainty for most isotopes, especially for $^{234}$U, $^{235}$U, $^{237}$Np, $^{238}$Pu, $^{242}$Pu, $^{243}$Am, for the curium isotopes, for the metallic fission products ($^{95}$Mo, $^{99}$Tc, $^{101}$Ru, $^{103}$Rh, $^{109}$Ag), the burn-up indicators, as well as $^{150}$Sm, $^{152}$Sm, $^{153}$Eu, $^{154}$Eu and $^{155}$Eu. The uncertainty of $^{235}$U increases with the total burn-up of the sample and reaches 7% at high burn-up (60 GWd/t). This large uncertainty is due to uncertainty in the depletion of the initial $^{235}$U and the small concentration of initial $^{235}$U after irradiation to high burn-up. However, $^{239}$Pu, $^{241}$Pu, $^{241}$Am and $^{242m}$Am are mostly sensitive to the temperature uncertainties. Lastly, $^{236}$U, which is produced by $^{235}$U neutron capture, is sensitive to the initial enrichment.

Other physical changes in the fuel configuration can occur as a result of deformation of the assemblies from axial pressures leading to bowing of the fuel rods and potential changes in the fuel to moderator ratio as compared to the initial design specification used in a model. These changes cannot be accurately quantified in the computational model but can lead to variability in the calculated to measured isotopic contents of the fuel due to uncertainty in the actual assembly configuration. Other changes during irradiation can result from clad yield (collapse) under the coolant pressure, clad corrosion and hydride formation. The impact of a small dimensional change in the fuel rod pitch, from the nominal value of 1.260 cm given in SFCOMPO to a perturbed value of 1.265 cm, is shown in Table 24. The results illustrate the potential impact of even very small changes in the assembly configuration.

**Table 24: Relative uncertainty (%) due to fuel pitch (1.260 cm vs. 1.265 cm)**

| Isotope | Fuel rod SF97 | |
|---|---|---|
| | EOL | 5 years |
| $^{234}$U | -0.2 | -0.1 |
| $^{235}$U | +1.0 | +1.0 |
| $^{236}$U | 0.0 | 0.0 |
| $^{237}$Np | +0.7 | +0.7 |
| $^{238}$Pu | +1.1 | +1.1 |
| $^{239}$Pu | +1.4 | +1.4 |
| $^{240}$Pu | +0.4 | +0.4 |
| $^{241}$Pu | +1.2 | +1.2 |
| $^{242}$Pu | -0.1 | -0.1 |
| $^{241}$Am | +1.4 | +1.2 |
| $^{242m}$Am | +2.2 | +2.2 |
| $^{243}$Am | +0.5 | +0.5 |
| $^{242}$Cm | +0.7 | +1.0 |
| $^{243}$Cm | +1.2 | +1.2 |
| $^{244}$Cm | +1.2 | +1.2 |
| $^{245}$Cm | +2.7 | +2.7 |
| $^{246}$Cm | +1.5 | +1.5 |
| $^{95}$Mo | -0.1 | -0.1 |
| $^{99}$Tc | -0.1 | -0.1 |
| $^{101}$Ru | 0.0 | 0.0 |
| $^{103}$Rh | +0.1 | +0.1 |
| $^{109}$Ag | +0.2 | +0.2 |
| $^{133}$Cs | -0.1 | -0.1 |
| $^{143}$Nd | +0.3 | +0.3 |
| $^{145}$Nd | -0.1 | -0.1 |
| $^{147}$Pm | -0.2 | -0.2 |
| $^{147}$Sm | -0.4 | -0.2 |
| $^{149}$Sm | +1.8 | +1.1 |
| $^{150}$Sm | +0.1 | +0.1 |
| $^{151}$Sm | +1.5 | +1.5 |
| $^{152}$Sm | -0.3 | -0.3 |

**Table 24: Relative uncertainty (%) due to fuel pitch (1.260 cm vs. 1.265 cm) (*cont.*)**

| Isotope | Fuel rod SF97 | |
|---|---|---|
| | EOL | 5 years |
| $^{153}$Eu | +0.1 | +0.1 |
| $^{154}$Eu | +0.6 | +0.6 |
| $^{155}$Eu | +1.3 | +1.3 |
| $^{155}$Gd | +3.0 | +1.4 |
| $^{156}$Gd | +0.2 | +0.2 |
| $^{144}$Nd | -0.3 | -0.2 |
| $^{146}$Nd | 0.0 | 0.0 |
| $^{148}$Nd | 0.0 | 0.0 |
| $^{150}$Nd | +0.1 | +0.1 |
| $^{134}$Cs | +0.4 | +0.4 |
| $^{137}$Cs | 0.0 | 0.0 |

# Chapter 7: Conclusion and recommendations

The importance of measured spent nuclear fuel isotopic compositions for validating computer codes and nuclear data libraries used for burn-up credit applications, and for a broad range of other spent fuel and fuel cycle applications, is recognised by the OECD/NEA/WPNCS. Under the auspices of the WPNCS, an Expert Group on Assay Data has been formed to help facilitate collaborations of NEA member countries in areas of common interest related to the acquisition and analysis of isotopic assay data, computational modeling, evaluation of uncertainties, and to share experience and best-practices in radiochemical analysis measurements. The EGADSNF convenes experts in the areas of nuclear criticality safety, reactor physics and fuel cycle analysis, and nuclear waste management. Through NEA member country collaboration, the database of publicly available spent fuel measurements, SFCOMPO, is being revised and expanded. Addition of new assay data has been a major accomplishment of the EGADSNF. The database now contains more than twice the number of measured fuel samples since the last update in 2003 and it includes many more fuels with modern enrichments and burn-up and many more measured isotopes than were previously available. The new data dramatically increase the number of measurements for fission product isotopes important to burn-up credit. In addition, new measurements for MOX fuels, VVER, AGR and Magnox reactor fuel types are being added.

A continued effort by the EGADSNF will be needed in the future to update the database as new experiments are completed and as other data are made publicly available when restricted commercial data are opened to the public. A significant but challenging objective of future activities of the EGADSNF is the evaluation of experimental data in SFCOMPO. Such an activity would provide independent peer review of experimental assay data and the supporting design and operating data required to perform validation calculations. The review process would vet the data for potential errors and produce recommended data and uncertainties suitable for use in code validation. Procedures for the qualification of benchmark data have already been established by the International Criticality Safety Benchmark Evaluation Project (ICSBE) for criticality experiments, and the International Reactor Physics Experiments Evaluation Project (IRPhEP) for reactor physics benchmarks. The independent process would significantly reduce the potential duplication of effort by different countries when the data are applied to code validation. Such a task is most effectively realised in the framework of an organisation like the EGADSNF that has access to a large amount of experimental data that can be used to evaluate individual experiments and includes experts in radiochemical analysis, computational modelling and simulation and nuclear safety analysis.

Looking forward, the application and propagation of isotopic bias and uncertainties to nuclear criticality safety analyses is recognised as an area where additional study is needed. When a burn-up credit methodology based on the concentration and reactivity of individual nuclides is applied, then the application of uncertainties and bias values obtained from nuclide validation studies is needed in order to obtain realistic margins for safety in criticality safety calculations. However, propagation of these uncertainties is complicated by the complex nature of the data, characterised by sometimes non-normal distributions, limited sampling of fuel types and assembly designs and in some cases low sample statistics. The statistical approaches being investigated by different countries can have a significant effect on the outcome [130-132]. Although the application of nuclide uncertainties was not considered in the scope of the current review, it is recognised as a fundamental need for the full and efficient application of nuclear fuel assay data. Such follow-up activities could be organised within the framework of future WPNCS activities.

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