# ACTIVITIES OF THE OECD/NEA EXPERT GROUP ON ASSAY DATA FOR SPENT NUCLEAR FUEL

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*Management of spent nuclear fuel is a key issue for many NEA member countries. In nuclear criticality safety, the decision of many countries to advance burnup credit as part of their licensing strategy has heightened recent interest in experimental data needed to validate computer codes used in burnup credit calculations. This paper discusses recent activities of an Expert Group on assay data, formed under the OECD/NEA/NSC/WPNCS (Working Party on Nuclear Criticality Safety) to help coordinate isotopic assay data activities and facilitate international collaboration between NEA member countries developing or implementing burnup credit methodologies. Recent activities of the Expert Group are described, focusing on the planned expansion of the Spent Fuel Isotopic Composition Database (SFCOMPO), and preparation of a state-of-the-art report on assay data that includes sections on recommended radiochemical analysis methods, techniques, and lessons learned from previous experiments.*

## I. INTRODUCTION

Management of spent nuclear fuel is a key issue for many Nuclear Energy Agency (NEA) member countries. As interim storage facilities in many countries reach their design capacities, the need to optimize spent fuel storage is becoming an increasingly important issue to managing fuel cycle costs while reducing associated risks. In nuclear criticality safety, the decision by many countries to advance burnup credit as part of their licensing strategy has heightened recent interest in measurement data that are needed to validate computer code calculations for a burnup credit methodology.

The code validation process is extremely important for safety and licensing evaluations to demonstrate that margins for safety account for code bias and uncertainty. An essential component of computational modeling and simulation is the availability of a qualified experimental benchmark against which calculation systems can be validated and the bias and uncertainty associated with the codes and data can be evaluated. For safety analyses that require irradiated fuel data, the spent nuclear isotopic compositions and resulting activities are generally calculated using depletion and decay codes. The isotopic concentrations predicted by the codes should be compared with corresponding spent fuel measurement data as a fundamental part of any code performance evaluation. To obtain high accuracy isotopic composition data, spent fuel is generally destructively examined by means of radiochemical analysis. The quantitative determination of the radionuclide content requires a series of complex methods for sample preparation, chemical separation of the various elements, and finally isotopic and elemental measurements. In recent years there has been growing interest by many countries in acquiring high quality assay data that can be used to validate code calculations for modern fuel designs and higher burnup fuel. This is particularly true for those countries that have advanced the technical basis for burnup credit in criticality safety to the point that it is being implemented in a regulatory and licensing environment, and for those countries actively developing facilities for long-term waste disposal. Measurements of spent nuclear fuel isotopic composition are essential to establishing code accuracy for burnup credit calculations used in transportation and spent fuel storage facilities, as well as to criticality and radiological safety studies involving fuel reprocessing and geologic repositories.

To further consolidate cooperation and interest in this area, an OECD/NEA Workshop on “The Need for Post Irradiation Experiments to Validate Fuel Depletion Calculation Methodologies” was held in 2006 [1]. In response to this interest, and moreover the general concern about the lack of an adequate publicly available spent fuel isotopic database for validation, an Expert Group on Assay Data for Spent Nuclear Fuel (EGADSNF) was formed in 2007 as a working group of the WPNCS under the leadership of K. Suyama (formerly of JAEA). The objectives of the group are to help coordinate isotopic assay data activities and foster collaboration between countries developing or implementing burnup credit methodologies, with the aim of making optimal use of resources and experimental data. This collaboration is particularly beneficial considering the very high cost of initiating new experimental assay programs (requiring fuel transportation, hot cell facilities, radiochemical analysis, and waste management capability), limited resources, and the limited experience of many countries in conducting high-precision isotopic measurement programs. The EGADSNF aims to take maximum advantage of existing experimental data through better dissemination, improved documentation, and enhanced peer review of data, and by providing a central repository for public assay data.

The EGADSNF membership is composed of criticality safety practitioners as well as experts from radiochemical analysis laboratories, waste management, reactor physics. The expert group currently has active members from Belgium, Czech Republic, Finland, France, Germany, Hungary, Japan, Slovak Republic, Spain, Sweden, Switzerland, United Kingdom, and the United States. The most recent meeting, held in June 2009, was attended by 27 participants representing 12 countries.

Assay data are necessary to validate 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. In addition to applications of nuclear criticality safety involving burnup credit, spent fuel compositions are the basis for calculations of radioactivity, neutron and gamma ray source terms, and decay heat. Because of the importance of the assay data to many different areas of spent fuel management, the expert group consists not only of the WPNCS members representing nuclear criticality safety, but also members of the standing technical Committee on the Safety of Nuclear Installations (CSNI) and the Integration Group for the Safety Case (IGSC). The IGSC is the main technical advisory body to the NEA Radioactive Waste Management Committee on the deep geological disposal of long-lived and high-level radioactive waste.

The following major activities have been initiated by the EGADSNF.

- Expand the SFCOMPO database of available isotopic assay data by increasing the number of fuel samples, include data with higher initial enrichment and burnup values, and expand the reactor types beyond mainly light water reactor (LWR) fuel.
- Review and improve 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.

- Develop recommended fuel design and operating history information needed 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.

This paper describes recent activities of the Expert Group, focusing on the planned expansion of the database of spent fuel measurements and a description of the new fuel samples and isotopic assay data they provide, recommended radiochemistry techniques, and future activities.

## II. REQUIREMENTS FOR ASSAY DATA

Requirements for isotopic data are determined in large measure by the intended use of the data and technical application area. Early experiments focused mostly on measurements of major actinides for studies of uranium transmutation and plutonium production, and nuclides required for burnup determination of the fuel (e.g., $^{137}$Cs and $^{148}$Nd). Later programs were expanded to include radiologically important isotopes 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 burnup credit has broadened the range of isotopes of interest to include many stable and long-lived fission products with large capture cross sections for which very few measurements were previously available, such as isotopes of Sm, Eu, and Gd. Moreover, several stable fission products including $^{95}$Mo, $^{101}$Ru, $^{103}$Rh, and $^{109}$Ag, which are large neutron absorbers in spent fuel, form as metallic particles that are difficult to dissolve and present unique challenges for accurate radiochemistry. The application areas currently considered by the expert group include (1) Nuclear Criticality Safety, (2) Nuclear Waste Management, and (3) Nuclear Fuel Safety.

The nuclides of highest importance to criticality calculations involving burnup credit have been widely studied. The actinides identified for benchmark problems coordinated by the OECD/NEA Expert Group on Burnup Credit (EGBUC) typically include up to 12 isotopes: $^{234}$U, $^{235}$U, $^{236}$U, $^{238}$U, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu, $^{241}$Am, $^{243}$Am, and $^{237}$Np. Other actinides such as $^{245}$Cm may become important for mixed oxide (MOX) fuel studies. The most important fission product isotopes include $^{95}$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. The contribution from the most important 20 fission products to the reactivity, excluding the contribution of Xe noble gas isotopes, is about 25% of the net reactivity effect. The largest six fission product contributors, $^{143}$Nd, $^{149}$Sm, $^{103}$Rh, $^{151}$Sm, $^{133}$Cs, and $^{155}$Gd, represent about 75% of the total fission product reactivity effect for typical fuel. The current lack of sufficient experimental data for fission products is seen as one of the main impediments to expanding the use of fission products in burnup credit analyses.

Measurement data on the concentration of isotopes in spent fuel is also required to support the waste management and safety assessments for spent fuel repositories. However, prioritized lists of isotopes of interest and the measurement accuracy for waste disposal are somewhat different from other application areas (e.g., criticality safety). For the waste disposal safety assessment, the required accuracy of the calculated radionuclide inventory may not be as high as for other applications because of the large uncertainties inherent in predicting release of migration of radionuclides over long time frames. Nevertheless, the radioactive inventory of spent fuel represents the initiating source of long-term dose assessment, and activities related to confirming or improving the accuracy of the spent fuel inventory will clearly be of interest and will benefit the waste disposal safety assessment. For the 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, the activation of long-lived activation products from the fuel impurities can be dominant radiological sources (e.g., $^{14}C$ and $^{36}Cl$). Common radionuclides of interest based on information provided by the IGSC member organizations include $^{14}C$, $^{36}Cl$, $^{79}Se$, $^{99}Tc$, $^{126}Sn$, $^{129}I$, and $^{135}Cs$.

The data application area of nuclear fuel safety covers a broad range of radiological applications, including severe accident analysis, spent fuel handling and storage, reprocessing, decay heat, and shielding analysis. In postulated severe accident analyses involving breach of the fuel during operation, noble gases (Xe and Kr) and volatile fission products (I, Cs, Te, Ru) are released. Cesium and iodine are important radiological isotopes for the source term evaluation since they are active elements chemically and have relatively large release fractions. For decay heat analysis involving 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, $^{106}Rh$, $^{154}Eu$, $^{144}Ce$+$^{144}Pr$, 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 sources of decay heat after approximately 50 years of cooling. Radiation source terms (neutron and gamma ray) nuclides are similar to those observed for decay heat. The principal nuclides identified in shielding studies for cooling times less than 100 years include $^{144}Pr$, $^{106}Rh$, $^{134}Cs$, $^{137m}Ba$, $^{154}Eu$, and $^{90}Y$. The nuclide $^{244}Cm$ represents the largest actinide contribution to dose rate.

A review of the dominant nuclides indicates that many nuclides are common to multiple application areas. Many other elements have multiple isotopes common to different applications. For example, the cesium isotopes $^{133}Cs$, $^{134}Cs$, $^{135}Cs$, and $^{137}Cs$ are dominant in many different applications. By careful planning of the experimental measurements, it may be possible to obtain additional isotopes with little added effort/cost to validate a broader range of applications.

## III. SFCOMPO DATABASE

The importance of having measured isotopic assay data from irradiated fuel experiments available to validate computer code predictions of spent fuel composition used in safety-related studies has long been recognized by members of the EGBUC and the Working Party on Nuclear Criticality Safety (WPNCS). An important activity of the WPNCS in this area has been the adoption and development of an electronic open-source database for spent nuclear fuel isotopic composition data, SFCOMPO http://www.nea.fr/sfcompo/. The first version of SFCOMPO was developed in the late 1990s by the (current) Japan Atomic Energy Agency (JAEA) to compile available isotopic validation data and make it accessible for use by the criticality safety community [2-4]. In 2002, the database was transferred to the OECD/NEA to maintain and coordinate distribution of data to the nuclear engineering community under the framework of an international organization.

SFCOMPO is a public database that contains fuel assembly design information, reactor operating histories, and measured isotopic contents for 246 spent fuel samples from reactors operating with $UO_2$ fuel. Pressurized water reactors (PWRs) include Obrigheim, Trino Vercellese, Takahama-3, H. B. Robinson 2, Calvert Cliffs 1, Mihama-3, and Genkai-1. Boiling water reactors (BWRs) include Gundremmingen, JPDR, Fukushima-Daini-2, Tsuruga-1, Fukushima-Daiichi-3, Cooper, and Monticello. The current datasets in SFCOMPO are summarized in Table I.

Table I. Spent fuel assay data currently in SFCOMPO

| Reactor | Country | Type | Assembly Design | Fuel Type | No. 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-Daiich-3 | Japan | BWR | 8×8 | $UO_2$,$UO_2$-$Gd_2O_3$ | 36 |
| Fukushima-Daini-2 | Japan | BWR | 8×8 | $UO_2$,$UO_2$-$Gd_2O_3$ | 18 |
| 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 |
| Cooper | USA | BWR | 7× 7 | $UO_2$ | 6 |
| Monticello | USA | BWR | 8×8 | $UO_2$,$UO_2$-$Gd_2O_3$ | 30 |
| Calvert Cliffs No.1 | USA | PWR | 14×14 | $UO_2$ | 9 |
| H.B. Robinson Unit 2 | USA | PWR | 15×15 | $UO_2$ | 6 |

In general, the existing data for the fission products are very limited. This is especially true for the large absorber fission products important to burnup credit. The experiments with the most complete fission product measurements are the Takahama PWR fuel data and the samples measured as part of the Approved Test Materials (ATM) program. Other limitations of the current database include:

- relatively few data available for high enrichment and high burnup fuels (no data above 60 GWd/t),
- no data for non-LWR reactor types or MOX fuel data,
- lack of data for many very long-lived radioactive fission products,
- incomplete documentation of irradiation conditions, particularly for BWR fuels that have incomplete void history information, and
- limited access to the primary experimental reports to obtain additional information.

Since the transfer of SFCOMPO to the NEA, no new assay data of spent nuclear fuel has been made available to update the database. Upgrading and expanding the data in SFCOMPO is a priority activity for the EGADSNF.

## IV. NEW EXPERIMENTAL DATA

During the last year members of the expert group have compiled measurement data from a number of additional spent fuel programs and have contributed this data to the NEA. Some data are collected from previously unpublished measurements and data that are not widely known beyond the institute that performed or analyzed the program data. Other data have been added from more recent experimental programs. In particular, new measurement data include several non-LWR designs including Magnox and Advanced Gas-Cooled Reactor (AGR) fuels, VVER fuels, high burnup $UO_2$ fuel, and several MOX fuel measurements. A summary of planned additions includes:

- New measurements for VVER 440 Novovorenezh reactor fuel are being performed under the US DOE/ISTC program [5].

- New $UO_2$ and MOX fuel measurement from the ARIANE International program are made publicly available by Oak Ridge National Laboratory (ORNL) [6].
- Swedish experts have contributed experimental data from the BWR Swedish Formark-3 reactor measured at Harwell (Sweden) and Dimitrovgrad (Russia) [7].
- Experimental data from AGR and Magnox fuels have been provided by the UK National Nuclear Laboratory (UKNNL) [8].
- The Spanish regulatory authority, Consejo de Seguridad Nuclear (CSN), is providing data for seven high burnup samples from the Vandellos PWR reactor with a burnup of up to 75 GWd/t [9].
- The Japanese Nuclear Energy Safety Organisation (JNES) is contributing data from a new measurement program in Japan. Fuel from a BWR (9×9) with a burnup of 55 GWd/t has been analysed. A new measurement program that includes high burnup BWR fuel (35–70 GWd/t) is under way. The assay data includes fission product compositions [10].
- Measurements of Three Mile Island-1 (TMI-1) reactor fuel made at GE-Vallecitos and Argonne National Laboratory (ANL) that include extensive fission product data (including all fission product isotopes considered in burnup credit) will be added to the database in the near future [11].
- German experts are contributing isotopic measurements on full-length Obrigheim spent fuel assemblies made at the Karlsruhe Reprocessing Plant (WAK). This particular set of measurements is currently not included in SFCOMPO [12].
- Isotopic measurements for the irradiation $UO_2$ fuel used in the REBUS international program [13] are being contributed by ORNL.

Experimental data from newer programs typically have much more extensive isotopic measurements and include isotopes important to burnup credit, decay heat, and radiation source terms. The samples also include more high burnup fuel measured to address safety and licensing issues. The experiments and the operating history data also tend to be more complete and uniformly documented. The new isotopic data sets are summarized in Table II.

At this time the new experimental data have not been uploaded to the electronic database SFCOMPO, and the formats and content of the database are currently being reviewed and improved. To provide users with access to the data, the original experimental reports and, in some cases, the computational analysis reports have been uploaded to the NEA Expert Group website. Note that in some cases information such as operating history data may be documented in several different reports and may not have been consolidated or independently reviewed at this time. The report archival area is currently being compiled and is organized by country contribution. It can be accessed from the following link and subdirectories:

http://www.nea.fr/html/science/wpncs/ADSNF/reports/

- F3F6-Sweden/
- JAEA/
- JNES/
- ORNL/
- UK/

To improve the value and usefulness of the existing data in SFCOMPO, many of the original experimental reports for the data are being made available directly, or indirectly, through the NEA website. The reports are prepared by institutions around the world, and many can be difficult to obtain due to their age. Providing centralized access to the reports can be particularly valuable for users of the data as they often provide details on the measurement methods, uncertainties, and other evaluations of the data that cannot be easily captured in an electronic database alone. Moreover, we are collecting secondary data references used in the development of operating histories or measurements, and other reports that have been translated into English.

Table II. List of new isotopic assay data to be included in SFCOMPO

| Country of Reactor | Reactor Name | Reactor Type | Measurement Facility | Assembly Design | No. Samples | $^{235}U$ Enrichment [wt%] | Burnup [GWd/t] |
|---|---|---|---|---|---|---|---|
| Germany | Obrigheim | PWR | ITU/IAEA/WAC / IRCh | 14×14 | 5* | 3.13 | 27–29 |
| | | | ITU | MOX 14×14 | 1 | 3.2 Pu fissile | 37 |
| Japan | Fukushima-Daini-2 | BWR | NFD | 8×8-4 | 46 | 3.0 (assembly average) | 9–62 |
| | Fukushima-Daini-1 | | JAEA | 9×9-9 | 11 | 3.4 (assembly average) | 36–69 |
| | | | NFD | 9×9-7 | 6 | 3.4 (assembly average) | 61–78 |
| | Tsuruga-1 | | NFD | MOX 8×8-2 | 5 | 3.1/4.6 Pu/(U+Pu) | 31–39 |
| | Not yet opened | PWR | JAEA | 17×17 | 5 | 3.2 | 22–39 |
| Netherlands | Dodewaard (ARIANE) | BWR | SCK•CEN/PSI | MOX 6×6 | 5 | 4.9/6.4 Pu/(U+Pu) | 34–56 |
| Spain | Vandellós | PWR | Studsvik | 17×17 | 9 | 4.5 | 43–74 |
| Sweden | Forsmark 3 | BWR | Studsvik, Dimitrovgrad, Harwell | SVEA-100 | 1 | 4 | 60 |
| Switzerland | Beznau-1 (ARIANE) | PWR | SCK•CEN/PSI | MOX 14×14 | 3 | 4.3–6.01 (Pu+Am)/ (U+Pu+Am) | 40–59 |
| | Beznau-1 (UK) | | ITU/ Karlsruhe | MOX 14×14 | 6 | 3.7–5.5 Pu | 22–63 |
| | Gosgen (ARIANE) | | ITU/SCK•CEN | 15×15 | 3 | 3.4/4.1 | 29–60 |
| UK | Hinkley R3/R4 | AGR | AERE Harwell/ AEE Winfrith | Annular | 27 | 1.54–2.55 | 1–26 |
| | Hunterston | AGR | AERE Harwell/ AEE Winfrith | Annular | 2 | 2.0 | 13–16 |
| | | Magnox | AERE Harwell | Fuel rod | 5 | 0.711 | 3–9 |
| | Bradwell | Magnox | AERE Harwell | 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 (Russia) | Hexagonal | 8 | 3.6 | 39 |

* Five reprocessed fuel assemblies, measured in two batches each.

## V. DATA FOR ISOTOPIC BENCHMARK ANALYSIS

Based on experience in the development of calculation methodologies for burnup credit, it is clear that more detailed data related to both operating history and measurement uncertainties are desirable in the database. A study to develop requirements for the types of data and level of detail required in order to apply the data to benchmark analyses has been initiated by EGADSNF members. The results of these studies will be used to help define data needs and recommended formats for the next version of the SFCOMPO database.

The format of the current database includes the experimental data as reported in the open literature and basic information about the fuel type, reactor type, initial enrichment, and burnup (usually an assembly-average value). In many cases, the original reports from which the data were obtained contain little information about the irradiation history or parameters such as the void fraction history, in the case of BWRs. These data deficiencies require the user of the data to supply or derive approximate data for missing parameters, thereby reducing the applicability and quality of the data for benchmarking.

The recommended design and operating data developed by the EG, listed in Table III, provide sufficient detail to allow a rigorous computational analysis of the irradiated fuel and isotopic composition data using most 2-D codes. The data will be used as a data template as new datasets are added to SFCOMPO in the future. This format could also be used as a reference to identify data needs for future experimental programs. New programs should ensure that the necessary fuel design and plant operating data will be available before initiating costly fuel isotopic measurements.

One of the goals of the revised format and content of SFCOMPO is to address the need for more consistent modeling data with a level of detail that is appropriate with the measured sample. The level of detail required for data, however, has evolved and increased as the computational analysis capabilities have increased. One of the challenges, particularly with documentation of older experimental programs, is that some detailed information was not documented because the computational tools available at the time were not capable of using the data. The requirement for more complete and detailed information is driven by increased computational capabilities and the desire to reduce computational bias and uncertainties that can be introduced by missing or incomplete information. In the longer term, the goal of the EGADSNF is to include additional details using revised database formats. A near-term objective is to more clearly identify deficiencies in current documentation and include experimental uncertainties in the SFCOMPO database.

As part of an assessment of the effects associated with incomplete documentation and uncertainties in design and operating data, a sensitivity/uncertainty study was performed. The Takahama-3 experiments were used as an example for the uncertainty analysis study to be carried out when information is missing. Calculations were performed to determine the sensitivity of the final isotopic compositions, actinides and fission products, to input data uncertainty. The studies included the effects of uncertainty in:

- Power history simulation
- Void fraction
- Fuel temperature
- Initial $^{235}U$
- Moderator temperature
- Burnup of the sample
- Water gap between the assemblies
- Surrounding (neighbor) assemblies on peripheral and internal rods

In the analyses performed by the EG, the parameters with the largest sensitivities in the depletion calculation include the initial $^{235}U$ enrichment, the fuel and moderator temperature at the sample position, the local burnup of the sample as deduced from the analysis results of burnup indicators like $^{145\text{-}146}Nd/^{238}U$ and $^{148}Nd/^{238}U$, and the operating core follow history.

Table III. Summary of recommended design and operating data requirements

| Categories | Data |
| --- | --- |
| Reactor | Name of reactor |
| | Type of reactor |
| | Nominal thermal power |
| | Rating (MW/t) and tonnes heavy metal in core |
| | Coolant type (light water, heavy water, $CO_2$, graphite, etc.) |
| | Nominal pressure of the primary system |
| | Nominal core flow |
| | Nominal coolant inlet, outlet, and average temperature |
| | Number of assemblies |
| | Control rods/cruciform assemblies locations |
| | Core positions and orientation during irradiation of the sample mother assembly |
| Assembly | Manufacturer and design of the assembly |
| | Schematic with detailed dimensional description (assembly, channel, and/or gap) |
| | Structural information: spacer grids, braces (position, composition, and dimensions) |
| | Location of the measured rod as well as the type ($UO_2$, MOX, Gd fuel) and individual enrichment of all the fuel rods of the mother assembly |
| | Characteristics of the adjacent assemblies, including geometry (example: 15×15), fuel type ($UO_2$, MOX, Gd fuel), assembly average initial fissile content |
| Fuel Rod | Dimension of fuel/clad/gap |
| | Details of fuel pellet shape/dimensions, dishing etc. |
| | Average density, or linear weight |
| | Active fuel length |
| | Initial composition of all the fuel rods of the mother assembly; for $UO_2$ fuel, U isotopic composition must include $^{234}U$ and $^{236}U$ |
| | Impurities in the fuel (N, Li, etc.) and cladding (Co in stainless steel for example) |
| | Metal/oxygen wt % |
| | Axial position and length of the sample (number of included pellets) |
| Guide Tube | Material (composition) and configuration (locations) |
| | Inner and outer diameters |
| Irradiation Data | Number of irradiation cycles of the mother assembly |
| | Start-up (BOC) and shutdown (EOC) dates for each cycle |
| | Reactor power history for each cycle |
| | Assembly average burnup at each BOC/EOC |
| | Time variation for the following parameters:<br>• Coolant temperature (at the axial level of the sample)<br>• Clad temperature (at the axial level of the sample)<br>• Boron concentration in the coolant<br>• Fuel sample temperature (define as average, centerline, effective, etc.)<br>• Void fraction at the level of the sample (for BWR fuels)<br>• Position of control rods or burnable poison rods in the assembly<br>• Sample burnup |

Uncertainties of the sample surrounding geometry are particularly relevant to the Takahama measurements, because two of the fuel rods were obtained from the edge of the assembly, and adjacent assembly information from the core during irradiation was not available. Studies compared the results obtained using a single assembly with reflective boundary conditions (see ¼ model in Fig. I) and a more detailed model that included the neighbor assemblies. In the latter model (see Fig. II), the isotopic composition of the neighbor assemblies was varied for several typical core loading patterns.

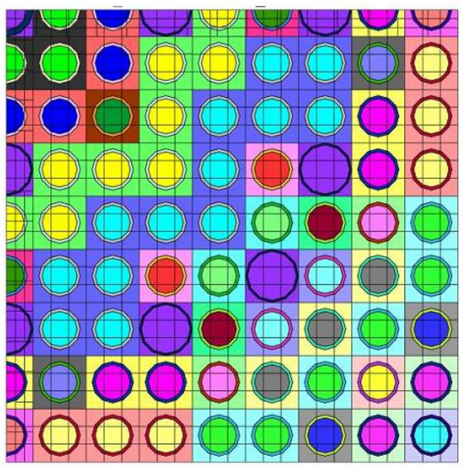

Figure I. Takahama single assembly model (1/4 assembly).

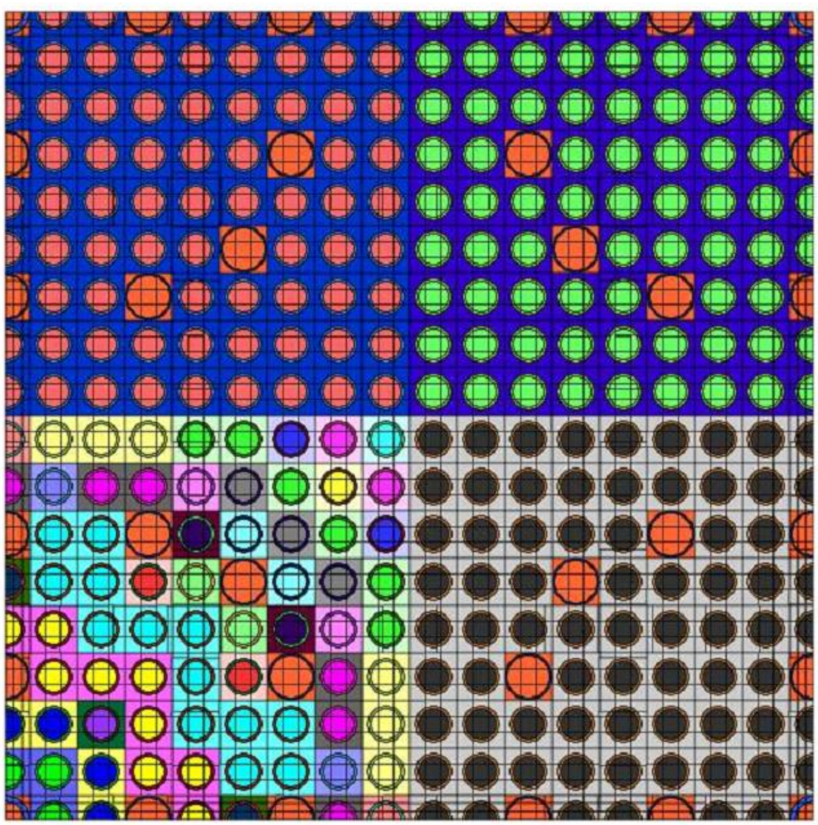

Figure II. Takahama model with neighbor assemblies (1/4 assemblies).

# VI. METHODS AND TECHNIQUES FOR RADIOCHEMICAL ANALYSIS

Destructive chemical and radiochemical analysis remains the most reliable analytical approach for measuring the isotope vectors and absolute isotope concentrations in irradiated fuels used in the creation of a nuclear isotopic database. This type of analysis can be performed only at specialized laboratories with access to the specialized hot cell infrastructure needed for chemical preparation of samples and to perform specialized radio-analytical and mass-spectrometric techniques. As part of the isotopic database development activity being performed by the Expert Group, a report has been prepared that documents state-of-the-art radiochemical analysis techniques and best-practice methods for isotopic measurements of spent nuclear fuel. The report is a joint effort of several radiochemical laboratories with recognized worldwide experience in spent fuel measurements and research, including SCK•CEN (Belgium), CEA (France), ITU (Germany), and Studsvik Nuclear (Sweden). These laboratories are actively involved in performing spent fuel measurements for domestic and international programs.

The objective of the report, to be published as part of a larger report on the current status of spent fuel assay data being prepared by the expert group, is to give a concise overview of state-of-the-art methods and best-practice techniques used in the destructive post-irradiation fuel analysis of the isotopic composition and concentrations in spent nuclear fuel samples. The information is designed to help the non-analytical expert in getting an idea of the analytical work involved in the measurements and what results can be expected in terms of quality, sensitivity, reliability, etc., from the analysis of nuclides using different techniques. The report provides a concise description of the complete end-to-end analytical methodologies applied for spent fuel radiochemistry, which include sampling procedures, dissolution of irradiated fuel in a hot cell, chemical separations methods, and analytical and radio-analytical measurement procedures, including the following steps:

- Fuel sample selection and dissolution
- Separations techniques essential to measure the actinides and main fission products (Nd, Cs, Sm, Eu, Gd).
- Measurement techniques, including
  - Radiometric techniques
    - Gamma-spectrometry (γ-spec)
    - Alpha-spectrometry (α-spec)
    - Liquid scintillation counting (LSC)
  - Mass spectrometry techniques
    - Thermal ionization mass spectrometry (TIMS)
    - Inductively coupled plasma mass spectrometry (ICPMS)
    - Quadrupole ICPMS (Q-ICPMS)
    - Sector field inductively coupled plasma mass spectrometry (SF-ICPMS)
    - Calibration methods in mass spectrometry
      - Simple isotope dilution technique
      - Technique of double isotope dilution
    - Determination of concentrations by external calibration
    - Determination of concentrations by standard addition
- Measurement time and data adjustments
- Uncertainty on the experimental result
- Nuclides of interest and recommended techniques

Each step is discussed in terms of the 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 uncertainties. The report includes a discussion of different measurement techniques and typical accuracies; including experimental techniques for dissolution, chemical separations, mass measurements (e.g., Fig. III), etc.

Nuclides measured in the framework of large experimental programs (like the ARIANE program) and applied measurement methodologies and techniques are summarized. These nuclides include those of importance in several areas related to nuclear energy, including licensing, safety, safeguards, etc. They include major and minor actinides, burnup indicators, burnup credit nuclides, as well as major heat-emitting and gamma-emitting nuclides and long-lived fission products. While it is impossible to cover all possible isotopes of interest and all possible experimental techniques, particularly with advancements every year in instrumental technology, the report is intended to capture the experience and current practice in radiochemical isotopic analysis methods at leading laboratories around the world. The list will continue to be developed and enlarged, for example, to include nuclides of interest in the field of long-term spent fuel management. The most important radiological nuclides would be (this list is not exhaustive) $^{14}$C, $^{36}$Cl, $^{79}$Se, $^{93}$Zr, $^{107}$Pd, and $^{126}$Sn, present in spent fuel either as fission products or as activation products, or both. For some of these nuclides (such as $^{14}$C, $^{79}$Se, and $^{36}$Cl), measurement methods applied to spent fuel samples have been developed or are being actively investigated.

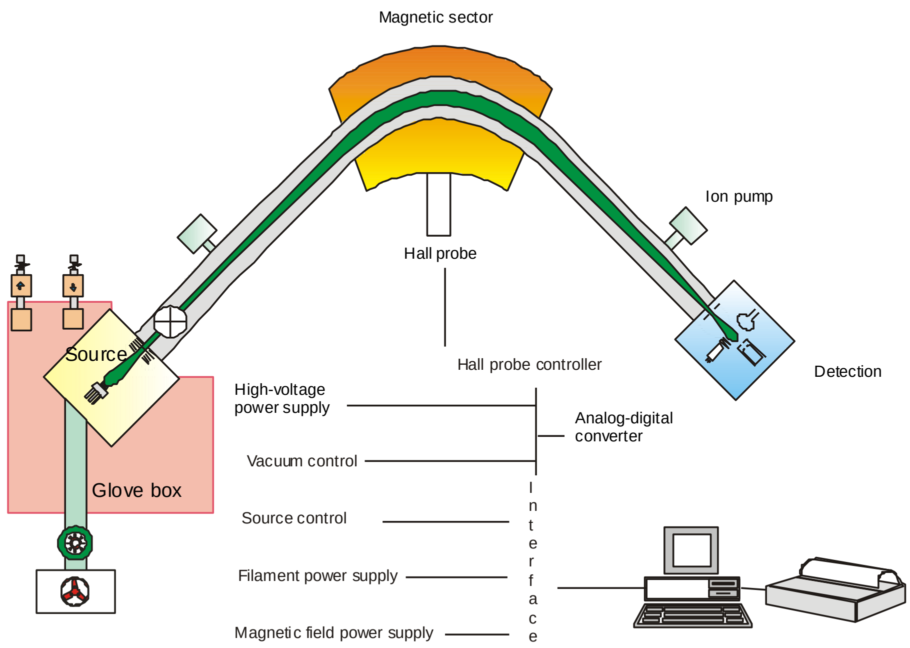


Figure III. Schematic of TIMS measurement setup.

## VII. FUTURE ACTIVITIES OF THE EXPERT GROUP

In addition to the revisions and updates of SFCOMPO, the Expert Group will publish in 2010 a state-of-the-art report on *Assay Data for Spent Nuclear Fuel*. The report will include detailed information on the requirements and need for additional isotopic composition data in areas such as waste management, fuel reprocessing, and spent fuel criticality. Further development and expansion of SFCOMPO will continue as new datasets become available. In particular, primary experimental reports are being compiled and archived on the NEA website for future use and evaluation. As the data are evaluated, they will be uploaded to the electronic SFCOMPO database once they have been recommended for use.

The evaluation of the isotopic data and the associated fuel design and operating history information is seen as a logical next high priority task of the expert group. In many cases, particularly for the older experiments and open isotopic data, experiments are largely unreviewed. Exceptions include some of the more recent international experimental programs, such are ARIANE, that have generally undergone internal review by program participants. Some of the observations based on previous evaluations of data include:

- A generally inconsistent level of detail in the documentation.
- Missing information that can seriously compromise the value of the data for benchmarking.
- Little quality assurance of data or peer review/evaluation of the experimental data.
- Some measurement data containing large errors due to techniques used, adjustment of the data by the laboratory (e.g., to a common time point), and typographical errors in the reports.

Review and processing of the experimental measurements into usable data formats can be very time consuming and currently must be done individually by each organization utilizing the measurements. The next phase of the expert group activity will be focused cooperation to evaluate data sets for completeness and accuracy, and documenting each reviewed dataset in a form that can be readily used directly by institutes for the purpose of code validation. Examples of review procedures have already been developed for benchmarks of the International Criticality Safety Benchmark Evaluation Project (ICSBEP) and the International Reactor Physics Experiments Evaluation Project (IRPhEP), and these procedures will be considered part of the current activity.

Under the support of the Spanish regulatory authority CSN, an evaluation of two relatively recently added data sets has been initiated: (1) the Vandellos PWR high burnup fuel measurements and (2) the Forsmark BWR measurements selected because of the different modeling data requirements for BWR fuel compared to PWR fuel. The scope of the evaluation will include an independent assessment of the experimental data documentation and quality, and review of the design and operating data for completeness and accuracy. The outcome will be peer-reviewed benchmark reports for each dataset, and a preliminary report that includes lessons learned and recommended guidelines for performing such reviews in the future. Another important outcome of this work will be that resource requirements necessary to perform peer reviews and documentation for other datasets can be estimated based on this experience.

## VIII. CONCLUSIONS

The work performed by members of the EGADSNF has (1) developed criteria and procedures to improve the quality of future experiments by identifying required data for fuel characterization and operating history data to qualify samples to be used in isotopic validation, (2) updated the format used in SFCOMPO to include experimental uncertainties, (3) expanded the number of measurements and fuel types included in SFCOMPO, (4) expanded access through the NEA website to original reports on the assay measurements included in the SFCOMPO database, and (5) documented best practices and recommendations for radiochemistry techniques based on experiences of several laboratories that are actively involved in performing state-of-the-art spent fuel measurements. A report summarizing the EGADSNF activities is being drafted by the participants, and final publication is expected in early 2010.

We hope that this expert group will contribute to the progress of nuclear criticality safety and nuclear fuel cycle research programs in NEA member countries. We invite and encourage specialists interested in spent fuel isotopic data in the NEA member countries to participate in this expert group.

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