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NUCLEAR ENERGY AGENCY
NUCLEAR SCIENCE COMMITTEE

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THE NEED FOR POST IRRADIATION EXPERIMENTS TO VALIDATE FUEL DEPLETION CALCULATION METHODOLOGIES

WORKSHOP PROCEEDINGS

11-12 May 2006
Prague, Czech Republic

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English - Or. English

Workshop on the Need for Post-irradiation Experiments to Validate Fuel Depletion Calculation Methodologies

11-12 May 2006
Nuclear Research Institute Rez, plc
Rez, Czech Republic

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The OECD Nuclear Energy Agency (NEA) was established on 1st February 1958 under the name of the OEEC European Nuclear Energy Agency. It received its present designation on 20th April 1972, when Japan became its first non-European full member. NEA membership today consists of 28 OECD member countries: Australia, Austria, Belgium, Canada, the Czech Republic, Denmark, Finland, France, Germany, Greece, Hungary, Iceland, Ireland, Italy, Japan, Luxembourg, Mexico, the Netherlands, Norway, Portugal, Republic of Korea, the Slovak Republic, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The Commission of the European Communities also takes part in the work of the Agency.

The mission of the NEA is:

- to assist its member countries in maintaining and further developing, through international co-operation, the scientific, technological and legal bases required for a safe, environmentally friendly and economical use of nuclear energy for peaceful purposes, as well as
- to provide authoritative assessments and to forge common understandings on key issues, as input to government decisions on nuclear energy policy and to broader OECD policy analyses in areas such as energy and sustainable development.

Specific areas of competence of the NEA include safety and regulation of nuclear activities, radioactive waste management, radiological protection, nuclear science, economic and technical analyses of the nuclear fuel cycle, nuclear law and liability, and public information. The NEA Data Bank provides nuclear data and computer program services for participating countries.

In these and related tasks, the NEA works in close collaboration with the International Atomic Energy Agency in Vienna, with which it has a Co-operation Agreement, as well as with other international organisations in the nuclear field.

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EXECUTIVE SUMMARY

Michaele Brady Raap
Chairperson

The diversity of participation in the *Workshop on the Need for Post Irradiation Experiments to Validate Fuel Depletion Calculation Methodologies* held at the Nuclear Research Institute Rez, Czech Republic, 11-12 May 2006 is testament to the importance of this issue. Thirty-three participants from twelve countries and two international organisations were involved in the proceedings. A thirteenth, Armenia, had registered but the representative did not attend. The workshop was organised by the Organisation for Economic Co-operation and Development (OECD)/Nuclear Energy Agency (NEA) in order to promote a common understanding of the need for experiments for validating the burn-up credit approach to criticality for VVER fuels.

Comparing code results to physical data is an essential step in validating any calculational methodology. The validation process is extremely important when the intent is that those calculations will be used for safety and licensing evaluations. These comparisons to measurement ensure that a computer code model is a correct representation of the process or system for which it is intended to represent. Burn-up credit is a licensing approach which permits some degree of credit for the reduced reactivity of irradiated fuel based on the physics of the transmutation of the fuel isotopics. The results from the depletion codes used to predict these isotopic concentrations should be compared with the corresponding measurements taken as a result of post irradiation experiments (PIE). To obtain the isotopic data, the spent fuel samples are destructively examined using radiochemical analysis. We acknowledge that PIE are of high importance for safety in all stages of spent fuel management and address a much broader scope than simply the isotopic measurements. The impact of burn-up on materials properties such as stress corrosion cracking, and radial hydriding and other fuel performance issues are commonly addressed with PIE. The discussion of PIE at this workshop was limited to the measurement of nuclide concentrations and distribution within spent fuel.

Worldwide there is increasing interest in “full” (accounting for depletion and build-up of both actinide and fission product nuclides) burn-up credit (BUC). As a result of this interest several countries have been developing standards for the implementation of burn-up credit. There is also an effort underway to develop an international standard (ISO) for burn-up credit. These standards address the requirements for establishing and implementing burn-up credit as part of the safety basis and generally conclude that experimental data are required for validation. Therefore, there is increased interest in the availability and integrity of data available for validation. The amount of data that are publicly available for light water reactors is somewhat sparse and covers a limited range of initial fuel enrichments, burn-ups and very few fission product measurements. This is perhaps the driving factor in the broad participation in the meeting. With the exception of France, many countries will need to rely heavily on measurements that are openly available in order to realise the advantages of burn-up credit. France has over the past 10 years undergone a comprehensive and expensive programme to obtain experimental data for the validation of burn-up credit. Several presentations at the meeting were

the results of sensitivity studies to show that if new data were obtained for the VVER fuel, it could potentially be applicable for western PWR fuel designs. The real issue is that limited PIE data for VVER fuels have been identified to facilitate burn-up credit for this fuel type or to validate the result of the sensitivity studies. Therefore, the topic of investigating the potential to obtain new PIE data for VVER fuel has become a more global interest.

Countries operating VVERs have no access to the mostly restricted proprietary data gathered in Russia. Several journal articles were published in Russia in the early eighties relative to experimental data for the VVER-440, but the sets of measured isotopes for BUC applications were incomplete and the data appear not to have been well documented. Only recently, supported by the US and reflecting the urgent need for such data, was a VVER-440 PIE focusing on BUC use performed in Dimitrovgrad as part of an ISTC project. This PIE project yielded data for the first eight samples of the VVER-440 spent fuel for actinides and major fission products and enables their use for the whole VVER criticality safety community without restrictions.

New and accurate experimental data will decrease the uncertainties in determining the multiplication factor, which will provide an appropriate degree of depletion code validation and therefore enable new technologies based on BUC to be safety implemented in spent fuel management systems.

In this context, the OECD/NEA Nuclear Science Committee organised this workshop to address the following topics:

1) expressing and justifying the need for fuel depletion calculation methodology validation;

2) evaluating the current status of validation work for different calculational methodologies and reactor types;

3) considering the current status of experimental validation work;

4) proposing experimental programmes to address these needs;

5) assessing the prospects for international co-operative programme(s).

The first topic is perhaps the most complex to address explicitly. Many of my comments herein are derived from the country reports given in the first session and are based on subsequent discussion during the final panel session. If one assumes that burn-up credit is to be used, then the need for having experimental data to validate the fuel depletion calculation is unquestionable for safe implementation. If one begins to examine the need for burn-up credit itself, there can be multiple reasons. The first is simply economics. Utilisation of burn-up credit has been shown to increase the amount of fuel that can be stored, transported or processed in a specific area/cask/equipment. The use of burn-up credit in spent fuel storage pools at reactors is only an intermediate solution to storage problems. The real need (and economic advantage) is downstream of pool storage at on-site storage, transportation, fuel reprocessing and/or geologic disposal. Reductions to the number of on-site storage casks, number of casks that must be transported, reduction of neutron poisons for reactivity control in reprocessing and the reduction in the number of waste packages and requisite space needed for geologic disposal are all strong economic incentives for pursuing burn-up credit as reported by many participants. The economic advantages are not the only reason to pursue burn-up credit. The reduction in the number of spent fuel shipments using burn-up credit provides a qualitative decrease in risk to the public, this is a good motivator particularly in areas of dense population. Reducing the size of the area required for a geologic repository could greatly increase the feasibility and reduce the complexity

and cost of characterising and licensing these sites. Last, but not least, the trend to increase the initial enrichment of nuclear fuel to increase cycle time, reduce outages (and therefore exposure to workers, etc.) is such that the facilities as designed cannot accommodate these fuels without some change to the regulatory basis.

The next two items are integral to one another and were generally addressed jointly. Presentations from a number of countries were made to address the process and data available for validation. Specific presentations on validation were made by French, Japanese, American, Ukrainian, German, Finnish, Hungarian and Slovakian colleagues. The extent to which measured data were emphasised in these analyses ranged from almost nothing (comparisons were made only between different calculational methods) to a high degree of rigor and multiple measurement types for each nuclide. These large differences were themselves revealing and strongly support the need for international co-operation in establishing standards and qualified benchmark databases. Presentations made to summarise the type and quality of benchmark data available, such as those in the NEA managed database, SFCOMPO further supported the need for establishing norms or requirements to qualify data as benchmarks. Several points related to the discussion of qualified data include the importance of detailed operating history data (this is a key parameter and should be established prior to measurements), lessons learned from radiochemical analyses about partially dissolved samples, the need for review of measurement plans and experimental procedures by independent and qualified contributors and also the enormous value of independent laboratory analysis of samples for cross checking. The nuclides to be included in the measurement plan should be greater than simply those of interest to burn-up credit for the purpose of near-term transportation and storage. Nuclides important to safety arguments for the disposal or reprocessing of spent nuclear fuel and management of the subsequent waste materials should be incorporated to realise greater benefit. Having measured data for parent and daughter nuclides provide additional data for establishing measurement quality. These data are of highest quality and use if all are measured at the same point in time to avoid potential errors in the decay calculations. Having the data available for measured nuclides increases the number of cross-checks that can be made to explain/understand when the measurements appear to be inconsistent with the calculations.

A discussion and review of a recent Russian proposal to obtain the necessary data was a focal point of the discussions. These include close co-operation between the research community and the nuclear power industry, reliable and detailed data about the operational history of the spent fuel, close proximity of the test fuel and hot cell facilities for extracting fuel samples, and qualified radioanalytical techniques to perform the dissolution and analysis measurements to adequate precision. An international co-operation that would involve countries like Japan, France, Switzerland and the United States that have performed these types of delicate experiments and analyses is important to capitalise on their experience and lessons learned and to assure the quality of the data with respect to the specific application to burn-up credit. Last but not least, the expectation is that these data would be used by both the utility and the regulator so it is important that the data be the result of collaboration among the constituent groups and not an NPP project only. The hope would be that experiments jointly funded by the regulatory body and industry (NPP and storage/transportation cask vendors) would have increased rigor in the review of both the experimental plan and results. Of course, care should be taken to ensure the independence of the regulator from the operator in the case of joint activities. A jointly-funded project could perhaps be more sophisticated and address a broader scope (e.g. range of fuel burn-up and enrichment) and larger numbers of samples than possible from the efforts of a single organisation. A co-operative experimental programme is important so that each VVER country will not have to independently bear the cost to obtain data or be forced to rely only on calculations for accepting burn-up credit. The tremendous economic and storage efficiency benefits of burn-up credit will ultimately result in high pressure to use this approach.

It was also reported at this meeting that analyses have been performed that indicate a programme to develop VVER data could be used to increase the experimental database for western-design PWRs as well. The western PWRs also suffer from the lack of data in the open literature in support of the inclusion of fission products in burn-up credit applications. Increasing the available benchmark data for the safe implementation of burn-up credit is of global benefit as demonstrated by the discussions and participation at this meeting.

In conclusion, I believe that there were two significant outcomes from the meeting. The first was the complete agreement that if burn-up credit is to be used for VVER fuel then experiments to validate the isotopic composition for irradiated VVER fuel are absolutely necessary and the experiments of the type proposed could produce the necessary data. Second, sensitivity studies show that the existing PWR fuel may be applicable to VVER fuel for the validation of burn-up credit applications and *vice versa*. However, there are no available VVER data to validate these analyses and there is no systematic evaluation process for assuring the quality of any existing PWR measurements for use as benchmarks. Therefore, there must be two recommendations from the meeting: (1) pursue VVER measurements as a precursor to the application of burn-up credit to VVER fuel (this means the measurements are needed quickly due to spent fuel management issues in countries like Ukraine and Slovakia) and (2) pursue establishing a systematic evaluation and database of spent fuel isotopic data for all light water reactors (including VVERs) in the format of SFCOMPO.

OPENING/INTRODUCTION

NEA ACTIVITIES IN NUCLEAR CRITICALITY SAFETY/BURN-UP CREDIT

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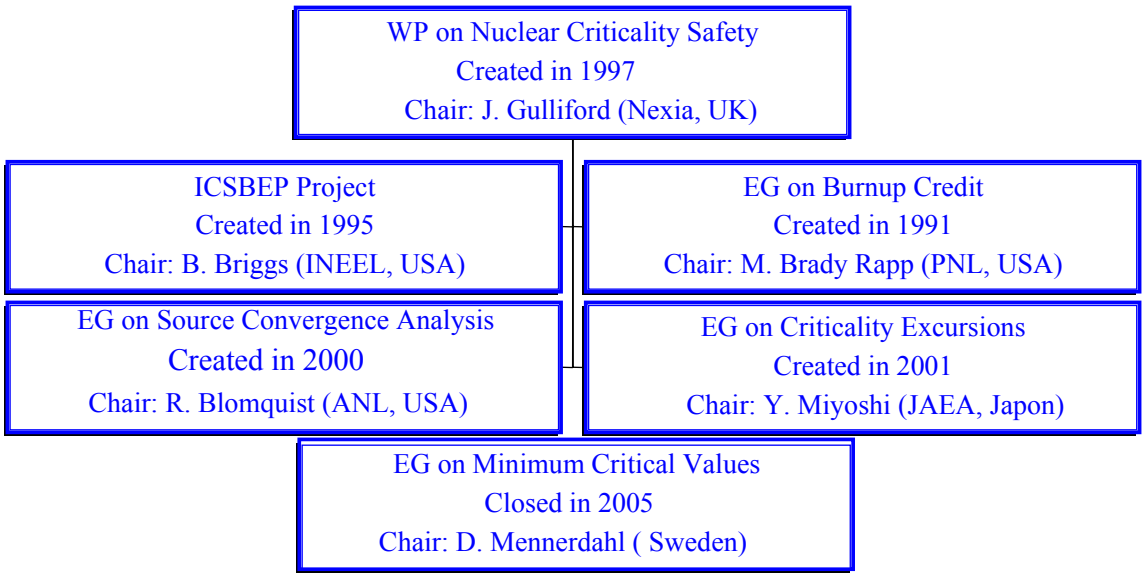
Abstract

The OECD Nuclear Energy Agency (NEA) started working on criticality-safety related subjects in the seventies. In the mid-nineties, several activities related to criticality-safety were grouped together into the Working Party on Nuclear Criticality Safety. This working party has since been operating and reporting to the Nuclear Science Committee. Four expert groups co-ordinate various activities ranging from Burn-up Credit issues to code and data inter-comparisons for the study of static and transient criticality behaviours. This paper describes current activities performed in this framework and the achievements of the various expert groups.

Introduction

The Nuclear Science Committee (NSC) is one of the six technical committees of the OECD Nuclear Energy Agency. Its mission is to help the NEA member countries identify, collate, develop and disseminate scientific and technical knowledge used for nuclear energy applications. Several Working Parties and Task Forces co-ordinate the work in the following areas: reactor physics, fuel cycle physics and chemistry, radiation shielding and criticality-safety. The Working Party on Nuclear Criticality Safety (WPNCS) was set-up in 1997 to co-ordinate the various activities in the area of criticality accident prevention in nuclear fuel cycle facilities. In the following paragraphs, the achievements of the different expert groups will be presented. References to the Expert Groups web site will be used in these paragraphs. The URL address of a specific Expert Group web site is constructed by adding an extra level to the Working Party's web site. For convenience, the following variable will be used throughout the paper: http://www.nea.fr/html/science/wpncs



Burn-up credit

The reactivity of nuclear fuel decreases with irradiation (or burn-up) due to the transformation of heavy nuclides and the formation of fission products. Burn-up credit studies focus on accounting for fuel irradiation in criticality studies of the nuclear fuel cycle (e.g. transport and storage of spent fuel).

The Expert Group on Burn-up Credit was established in 1991 to address scientific and technical issues connected with the use of burn-up credit in nuclear fuel cycle operations. Benchmark exercises were defined in order to compare the prediction capabilities of widely used computer codes and cross-section libraries. Basically, two kinds of benchmarks were considered: criticality calculations (where the fuel composition is given as input, the exercise then consists of comparing the calculated multiplication factors and fission densities) and depletion calculations (where the aim of the benchmark is to compare the calculated composition of spent fuel).

The Expert Group has attempted to cover a range of applications and to be as close as possible to actual configurations (e.g. transport flasks). Low-enriched uranium oxide fuels irradiated in pressurised water reactors were considered in four phases (I-A, I-B, II-A, II-B, II-C and II-D) and a fifth phase involving this fuel is still on-going (II-E). Phases III-A and III-B considered uranium oxide

fuels irradiated in boiling water reactors whereas Phases IV-A and IV-B investigated burn-up credit issues associated with MOX fuels irradiated in pressurised water reactors. Of the aforementioned exercises, five studied the effect of axial burn-up profile with an increasing level of complexity. Phase II-A and Phase II-B, have a simple axial burn-up profile. Phase II-C is devoted to the study of various axial profiles and to their effect on criticality calculations. Phase II-D has been recently completed and discusses the influence of control rods in the axial profile. Finally, Phase II-E merges Phase II-C and Phase II-D objectives and studies the end effect influence on the axial profile with a heterogeneous description of the system including the control rods.

The benchmark specifications and the analysis reports of the completed benchmarks are available for free download on the Expert Group's web site at: http://www.nea.fr/html/science/wpncs/buc. It should be noted that since the beginning of these activities, the availability of experimental data was considered as a key element for code validation. The composition of the spent fuel is a pre-requisite to any criticality calculation involving spent fuel and the accuracy of this input data is crucial for the subsequent calculations. Consequently, burn-up credit specialists stressed the need of Post Irradiation Examinations (PIE) for the purpose of measuring the isotopic composition of Spent Nuclear Fuel (SNF) at various occasions.

JAEA has initiated an effort for the collection of experimental data on spent fuel composition. A relational database called SFCOMPO was created in order to collect and disseminate these data. The database was transferred to the NEA web site (http://www.nea.fr/html/science/wpncs/sfcompo) in 2002 and is available for free consultation. The composition of the spent fuel is a pre-requisite to any criticality calculation involving spent fuel and the accuracy of this input data is crucial to the subsequent calculations. Consequently, burn-up credit specialists stressed the need of post-irradiation examinations (PIE) for the purpose of measuring the isotopic composition of spent nuclear fuel (SNF) on various occasions. More efforts are needed to complement the database with additional parameters and with other PIE composition data.

Two other groups at NEA have activities related with burn-up:

- Working Party in Reactor Systems (WPRS) – Depletion Calculation Benchmark devoted to Fuel Cycle Issues:
  - UOx Phase I
  - MOX Phase II
- Working Group on Operating Experience/Fuel Cycle Safety (WGOE/FCS) – Last edition of the report "The Safety of Nuclear Fuel Cycle"

Experimental benchmark evaluation

The Criticality Safety Benchmark Evaluation Programme was created in 1992 on the initiative of the US/DOE. The programme became an activity of the Nuclear Science Committee in 1995 when international experts joined the effort by providing experimental data and participating in the evaluation and review process. The aim of the programme is to identify, evaluate, verify, and formally document a comprehensive and internationally peer-reviewed set of criticality safety benchmark data that may be used to validate neutronics codes and nuclear cross-section data. The data are published in the International Handbook of Evaluated Criticality Safety Benchmark Experiments. The Handbook is updated every year to include new experiments and to revise previously published ones when new experimental data are found.

The 2006 edition includes 442 evaluations (over 38,000 pages) containing benchmark specifications for 3957 critical or subcritical configurations, 23 criticality-alarm-placement / shielding configurations with multiple dose points for each, and 20 configurations that have been categorised as fundamental physics measurements that are relevant to criticality safety applications in the 2006 Edition of the ICSBEP Handbook.

The Handbook contains benchmark data for seven fuel categories: uranium (high enriched uranium (HEU), low enriched uranium (LEU), intermediate and mixed enriched uranium (IEU)) plutonium (PU), mixed uranium and plutonium (MIX), ^{233}U, and special isotope (SPEC) fuels [^{244}Cm, ^{238}Pu, ^{237}Np, and ^{242}Pu]. The ICSBEP experiment classification includes two other levels which characterise the fuel form (SOLution, METal, COMPound and a mixture (MISC) of these forms) and the neutron spectra in the fuel (THERMal, FAST, INTERmediate and MIXED).

The ICSBEP Handbook contains not only benchmark data but also a set of calculated data (e.g. spectral indices) that provides some insights on the neutronics of each configuration. More detailed information about the neutron spectra was added in the new versions of the DVD-ROM. Neutron flux and reaction rates in the fuel region are now available in a 299-group energy structure. A detailed neutron balance in different regions of the configuration is also available. Finally, sensitivity coefficients, which characterise the relative change of k-eff due to a 1% change of multi-group cross-sections, are included for HEU solution configurations.

With the important growth of the Handbook and the addition of many numerical data, it was decided to develop an efficient and user-friendly means for accessing specific information contained in the Handbook. A relational database was designed and selected data was extracted from the Handbook. The main characteristics of a configuration (e.g. description of the geometry, fuel composition, moderating and reflecting materials…) are thus entered in the database together with all the spectra information described above. A users' interface was then developed to query the database and to extract the information needed by the user. Plotting capabilities were also included to compare the flux and reaction rates in different configurations. The database and the corresponding interface, called DICE, are part of the ICSBEP DVD-ROM.

Since ICNC 2003, a reactor physics counterpart to the ICSBEP, the International Reactor Physics Experiment Evaluation Project (IRPhEP) was initiated. The IRPhEP is patterned after its predecessor, the ICSBEP, but focuses on other integral measurements such as buckling, spectral characteristics, reactivity effects, reactivity coefficients, kinetics measurements, reaction-rate and power distributions, nuclide compositions and other miscellaneous types of measurements in addition to the critical configuration.

The purpose of the IRPhEP is to provide an extensively peer reviewed set of reactor physics related integral benchmark data that can be used by reactor designers and safety analysts to validate the analytical tools used to design next generation reactors and establish the safety basis for operation of these reactors. While co-ordination and administration of the IRPhEP takes place at an international level, each participating country is responsible for the administration, technical direction, and priorities of the project within their respective countries.

Minimum critical values

The Expert Group on Minimum Critical Values was established in 1999 with the aim of compiling and comparing minimum critical data used in different countries. Some of the values are from published handbooks, guides and other literature while other values were calculated mainly for the purpose of this study. With proper validation, an accurate estimation of the reference value based

on all contributions should be expected. The group has focused on data for homogeneous aqueous solutions of uranium oxides, plutonium oxides, uranium nitrate and plutonium nitrate. Efforts are being pursued to identify the origin of discrepancies between the compiled data.

The outcome of the group's findings is being published in a report containing a compilation and an evaluation of reference values from various participants. The accuracy of a reference value influences safety and economy of operations.

Source convergence

The Expert Group on Source Convergence Analyses revise the outstanding difficulty of convergence associated with the calculation of systems composed by loosely coupled fissile units. Four sets of benchmarks were defined and calculated by various Monte-Carlo and deterministic codes. The Expert Group's web site (http://www.nea.fr/html/science/wpncs/convergence) contains the detailed specifications of the benchmarks under study.

The most sensitive parameters of the convergence properties of k-eff series and generation-wise fission source distributions are: the initial spatial distribution of the starters, the number of skipped generations before tallying the results, the number of neutrons per generations and the overall number of generations. These parameters were considered in all the benchmarks.

The Phase I report, under publication, describes the work performed by the OECD/NEA Expert Group on Source Convergence in Criticality Safety Analysis during 1999-2004. Phase I consists mostly of specification and analysis of computational test problems of interest to criticality safety analysts that exhibit source convergence difficulties. It is aimed at improving the computational basis of criticality safety analyses.

Criticality excursions

The Expert Group on Criticality Excursion Analyses was established in 2001. Three main areas have been identified and included in the scope of the Expert Group:

- The evaluation of criticality excursion experiments. A draft format was designed by extending and adapting the format of the ICSBEP evaluation to include the sequence of excess reactivity introduction and to provide the time-dependent measured data (temperature, energy release, …).

- Inter-code comparison exercises. Two sets of benchmarks were submitted. These benchmarks are based on experimental programmes performed in the SILENE reactor (CEA, Valduc, France) and the TRACY reactor (JAERI, NUCEF, Japan). The specifications of the benchmarks are available on the Expert Group's web site: (http://www.nea.fr/html/science/wpncs/excursions).

- The development of Web-based information resources on criticality excursions including a description of experimental programmes and generated data, a short synopsis on existing modelling capabilities, and references to criticality accidents. This information is available at the Expert Group's website.

The Phase I report is being completed by the different participants and its publication is expected by the end of 2006.

Future plans

It is recognised that the WPNCS is a valuable forum for international co-ordination in the field of criticality-safety including:

- The exchange of information on on-going and projected national programmes.

- The organisation of the ICNC series of International Conferences on Nuclear Criticality Safety (selection of host country, set-up of the international advisory and programme committee).

- The co-ordination of international activities in different areas relevant to the analysis of reactivity of systems in the nuclear fuel cycle and associated properties (evaluation of experiments, code and data inter-comparison exercises…). To support other international initiatives (other committees within the OECD/NEA, IAEA, …).

IAEA ACTIVITIES RELATED TO CRITICALITY SAFETY

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Introduction

The IAEA's work on criticality safety is being carried out in the following three departments:

- nuclear sciences and applications (NA);
- nuclear energy (NE);
- nuclear safety and security (NS).

The three departments have their own work programmes as outlined in the IAEA Programme and budget. A good co-ordination is established between the NS an NE departments.

Activities of the Department of Nuclear Applications

In the NA department, the Division of Physical and Chemical Section (NAPC) is dealing, amongst others, with criticality safety. The Nuclear Data Section is active, e.g. in the development and dissemination of data, the application of data and technology transfer. It established and maintains a Nuclear Data Centre that makes databases, computer codes, electronic documents, etc. available on the web (www-nds.iaea.org). Co-ordinated Research Programmes (CRP) are also being carried out in order to improve the respective knowledge and expertise in the countries involved.

Activities of the Department of Nuclear Energy

The NE Department has a focal point on technological issues. Its Nuclear Fuel Cycle and Materials Section accomplishes a programme on spent fuel management that includes the application of burn-up credit as high priority issue. Some of the more prominent activities are related to technical meetings that led to the publication of three IAEA Technical Documents (TECDOCs) [1,2,3] with information of three earlier meetings and a CD that includes the essential information of the last technical meeting held in 2005 [4]. A CRP for China on burn-up credit technology was finished in 2005. An overview paper on IAEA spent fuel management activities [5] is included in Ref. [4].

Activities of the Department of Nuclear Safety and Security

The NS Department has a focal point on the development of safety standards and providing for their applications. Although criticality safety is addressed as an item in several (draft) safety standards, a comprehensive programme related to criticality safety and burn-up credit has not been established. Initial steps have been taken to correct this situation and prepare a safety standard on criticality safety in the near future. The Document Preparation Profile for such a safety standard has already been approved by all four Safety Standards Committees. Criticality safety has also been the subject of the IAEA conference on the management of spent fuel from nuclear power plants that was held in Vienna on 19-22 June 2006 (see http://www.iaea.org/NewsCenter/News/2006/spentfuel.html).

A workshop on criticality safety and burn-up credit is planned to be held for Eastern European countries in November 2006.

Summary

The IAEA has a lot of experience and expertise related to criticality safety. Three departments are involved in the work on criticality safety. Information can be retrieved from the IAEA web pages and also specific assistance can be provided upon request, including the assistance though Technical Co-operation projects. The IAEA will assist Eastern European countries by holding a workshop on criticality safety/burn-up credit, but does not plan to carry out specific activities on post irradiation experiments for VVER (or RBMK) fuel.

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PIE DATA NEEDS: OVERVIEW AND PRIORITIES

Chairs: E. Warnecke, Y. Rugama

CURRENT STATUS AND FUTURE NEEDS OF BURN-UP CREDIT: IMPLEMENTATION IN BULGARIA

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Abstract

This paper outlines the status of burn-up credit implementation in Bulgaria. WWER spent fuel inventory, spent fuel transportation and storage capacity at Kozloduy NPP site, as well as the country specific needs are shown. The regulatory status concerning burn-up credit is given. The burn-up credit methodology is presented, and the importance of depletion calculation validation against experimental data is emphasised.

Introduction

The most common assumption used in criticality safety analysis of spent nuclear fuel facilities is the “fresh fuel” assumption, resulting in significant conservatism in the calculated value of the system reactivity. The burn-up credit implementation allows accounting for the reactivity reduction associated with fuel burn-up, hence reducing the analysis conservatism and maintaining an adequate criticality safety margin.

Burn-up credit is already applied to transport systems, wet and dry storage facilities, and components of reprocessing plants in many countries. The implementation of burn-up credit in spent fuel management systems is an accepted possibility to achieve a reduction in fuel cycle costs. Besides the economical efficiency, the burn-up credit allows for safety enhancement in spent nuclear fuel management and reduction of environmental effect.

Implementation of burn-up credit in criticality safety analysis requires validation of the calculational depletion methodology against experimental data using fuel characteristics, operation history, axial distribution of burn-up, etc.

Bulgarian approach for spent nuclear fuel management

The safety of spent nuclear fuel and radioactive waste management is regulated by the Act on safe use of nuclear energy and is directed to fulfilment of the obligations on the Joint Convention on the Safety of Spent Fuel Managements and on the Safety of Radioactive Waste (1998) [1]. A National Strategy for management of spent fuel and radioactive waste is approved in 2004 [2].

The Republic of Bulgaria has two nuclear facilities, which during operation generate spent fuel and radioactive waste: Kozloduy NPP and Research Nuclear Reactor of the Institute for Nuclear Research and Nuclear Energy, Sofia. There are no local plants for conversion, enrichment and production of nuclear fuel as well as for reprocessing of spent fuel. Unit 1 and 2 of Kozloduy NPP are disconnected from the national grid. The research reactor is in process of reconstruction.

Status of the wet spent nuclear fuel storage at Kozloduy NPP site

The spent fuel of Kozloduy NPP is stored in both: pool type storage facility at each NPP unit and common wet storage facility on the site of the NPP [1, 3]. The Spent Fuel Storage Facility (SFSF) is designed for storage of spent fuel from WWER-440 and WWER-1000 reactors after at least 3 or 5 years, respectively, of initial storage in at-reactor storage pools. After that it is transferred for temporary storage in the spent fuel storage facility by the internal transport container. In SFSF the spent fuel assemblies are stored under water, placed in transport baskets. The time period foreseen for temporary wet storage is 30 years.

Strategy for management of spent fuel

As reported in Reference [1], the stored spent fuel at Kozloduy NPP site, collected in at-reactor spent fuel pools and wet spent fuel storage facility, at 01.01.2003 consists of 996.7 tHM. This amount is distributed into 5146 spent fuel assemblies from WWER-440 and 890 spent fuel assemblies from WWER-1000, or totally 6036 spent fuel assemblies.

The WWER spent nuclear fuel transportation to Russia is planned and provided upon contract basis. It is foreseen about 240 WWER-440 and 96 WWER-1000 spent fuel assemblies to be transported per year [2]. The strategy involves an increasing the capacity of the existing spent fuel wet storage facility on the site, as well as a construction of a new spent fuel dry storage facility. The capacity of the new storage facility considers total amount of spent fuel to be generated until the end of life of all units on site [1]. The first stage of the storage (for WWER-440 spent fuel assemblies) is expected to be commissioned in 2009. In the design of the first stage is included also the conceptual project of the second stage for storage of the WWER-440 and WWER-1000 fuel assemblies.

Regulatory status

The regulation for providing the safety of spent nuclear fuel management (August, 2004) [4] defines the matters related to the basic criteria and rules for providing nuclear safety and radiation protection in the management of spent nuclear fuel according to the provisions of the Safe use of nuclear energy act. According to this regulation to provide nuclear safety for both normal operation and design basis accidents the multiplication factor K_{eff} must be below 0.95 (K_{eff}<0.95).The spent fuel burn-up can be taken into account for criticality safety assessment if the burn-up control is assured through the technical tools [4].

Burn-up credit methodology. Validation against experimental data and benchmark problems

Our methodology for criticality safety analysis with burn-up credit implementation [5] is based on two worldwide used code systems for depletion and criticality calculations: SCALE-4.4 [6] for depletion and criticality calculations, and NESSEL-NUKO [7,8] for depletion calculations. The methodology is in process of validation for WWER applications.

The criticality safety control module CSAS6 (SCALE4.4) has been validated using both experimental data for rod power distribution in critical experiments (performed on the LR-0 experimental reactor in Nuclear Research Institute Rez, Czech Republic), and calculated results, obtained by MCNP code system [9].

The depletion code systems NESSEL-NUKO and SCALE4.4 (control module SAS2H) have been validated on the basis of comparison with the calculated results obtained by other depletion codes for the CB2 Calculational Burn-up Credit Benchmark [10].

As a first step of the methodology validation against experimental data, some preliminary calculations have been carried out for WWER-1000 spent fuel assembly. The data used for calculations and comparisons are taken from Ref. [11]. The considered spent fuel assembly (E-1591) is with initial enrichment 4.4% wt of ^{235}U. The assembly was operated for three fuel cycles in Unit 3 of Balakovo NPP. Two pellet samples (23-912 and 23-581) were taken at different distances of the fuel rod, located at the periphery of the fuel assembly. The burn-up level reached was 45.6 MWd/kg and 47.318 MWd/kg, respectively. The ratio of calculated to experimental (C/E) values of nuclide concentrations, for results obtained by SCALE, HELIOS [11] and NESSEL-NUKO [12], is shown in Figures 1-4.

The preliminary results presented show that the calculated concentrations of the most nuclides are in good agreement with the experimental ones. For further validation process more precise documented power history and some other input data are needed.

Fig.1. C/E values of nuclide concentrations, sample 23-912.
Calculations by SCALE4.4

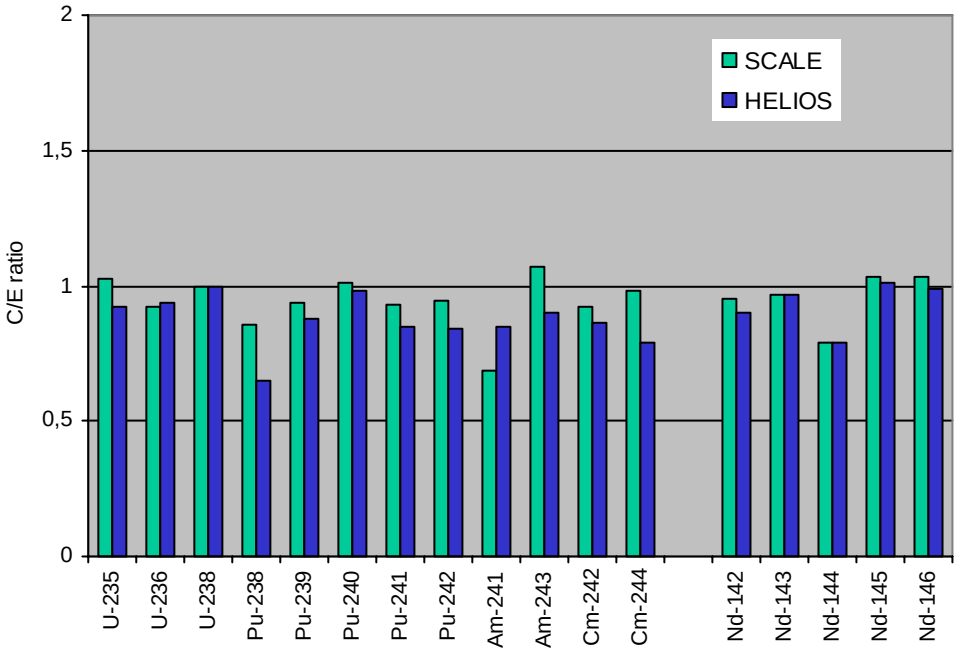


Fig.2. C/E values of nuclide concentrations, sample 23-581.
Calculations by SCALE4.4

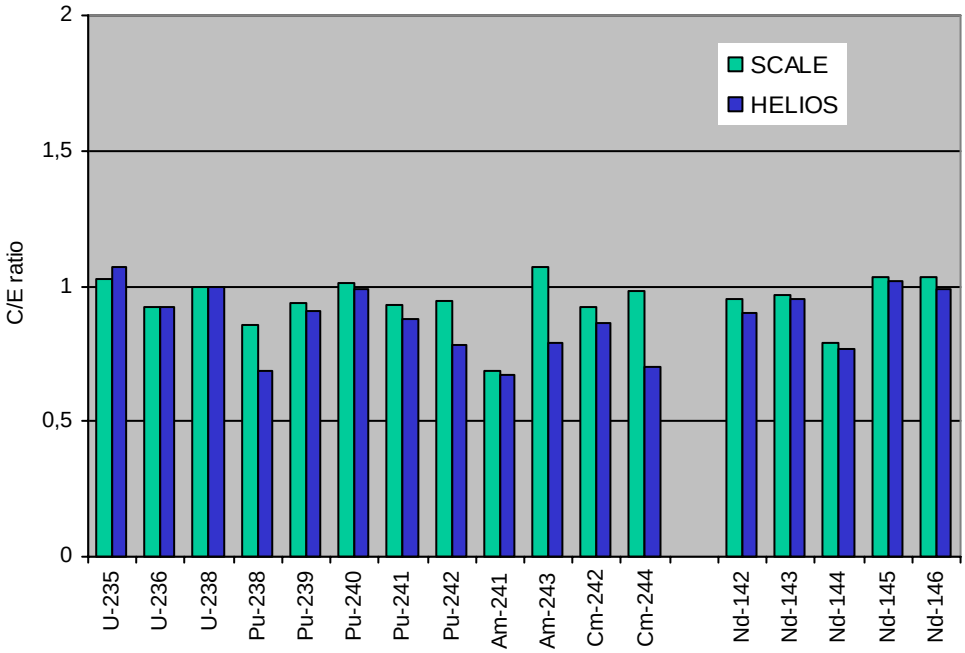


Fig.3. C/E values of nuclide concentrations, sample 23-912.
Calculations by NESSEL-NUKO

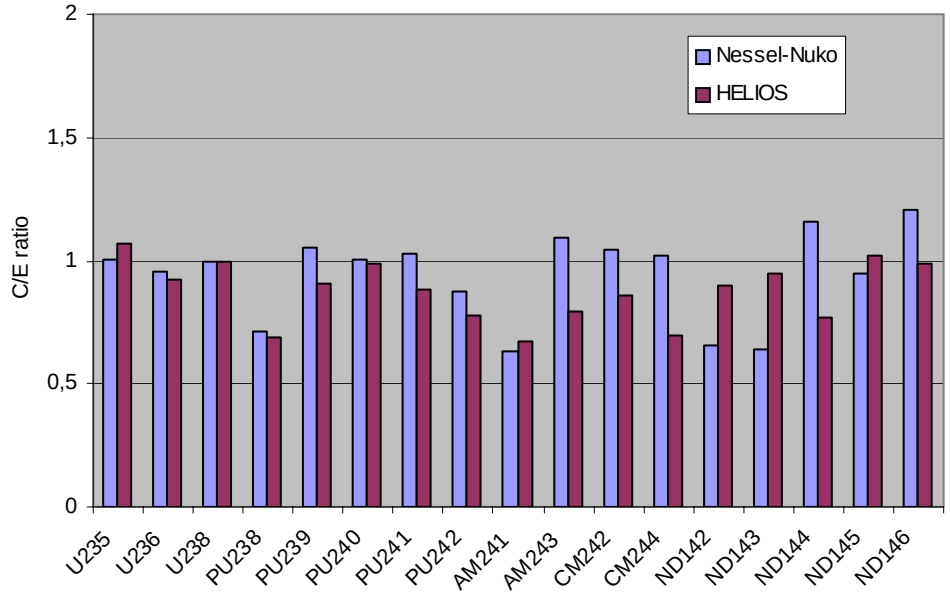
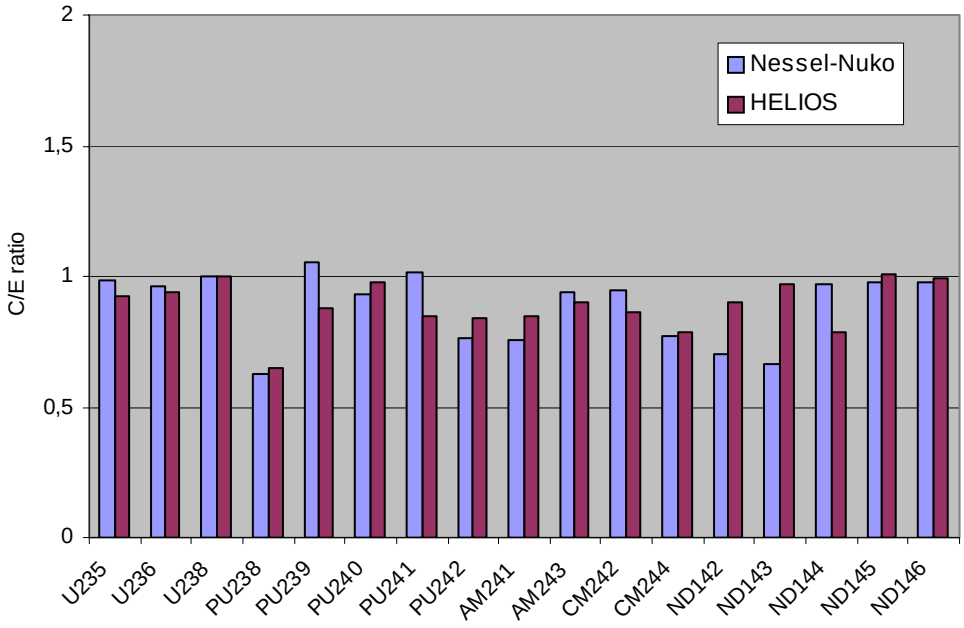


Fig.4. C/E values of nuclide concentrations, sample 23-581.
Calculations by NESSEL-NUKO



Conclusions

On the basis of the presented information the following conclusions about country specific needs could be drawn:

- Experimental data (nuclide concentrations) for selected WWER-440 and WWER-1000 spent fuel samples with well defined irradiation history and initial isotopic state are needed for depletion code validation;
- Support for an additional validation of the currently used codes and libraries for WWER environment is requested. This activity could be made on the basis of international collaboration;
- This would provide a suitable and reliable tool for burn-up credit analysis of WWER spent fuel management systems.

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NUCLEAR COUNTRY PROFILE AND SPENT FUEL MANAGEMENT: CZECH REPUBLIC

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SÚJB

The Czech Republic is medium size European country, both in population (about ten million people) and in size (about 80,000 square kilometres). It consists mostly of the three historic regions of Bohemia, Silesia and Moravia. The Czech Republic is bordered by Poland to the northeast, Slovakia to the southeast, Austria to the south, and Germany to the west and northwest.

Czech Republic is quite limited in hydro resources, and has practically no oil or gas. Poor quality light coal is the dominant energy source.

The uranium is supplied domestically; conversion, enrichment, and fabrication are carried out in a variety of other countries.

From the viewpoint of environmental protection and energy sources diversification, the nuclear energy plays significant role in country's energy supply structure.

Nuclear power was first considered as an option in former Czechoslovakia about 40 years ago.

There is highly developed nuclear infrastructure existing in the Czech Republic. The industry is capable to produce almost all of the main components of a Nuclear power plant (VVER design), including RPV's, primary piping, steam generators, pumps, balance of plant systems, etc.

Considerable R&D sector supports both the industry and regulatory authorities.

In late 1970s and beginning of 1980s there were four units of the VVER-440/213 design being built at the Dukovany site. The units were commissioned successively 1st unit – 1985, 2nd unit – 1986, 3rd unit – 1987, 4th unit – 1987. The four reactor units are owned by Czech power utility – ČEZ, a.s NPP Dukovany

These units represent about 21% of the total country electricity production.

Two additional NPP units of the VVER-1000 design, at the Temelín site essentially doubles the nuclear electric production capacity. The first fuel was loaded into the Unit No.1 in July 2000, into the Unit No.2 on 4 March 2002.

In the Czech Republic there are two experimental reactors located in vicinity of Prague at Nuclear Research Institute in Řež (LVR – 15 and LR – 0), training reactor VR - 1P operated by Faculty of Nuclear Science and Physical Engineering in Prague.

In connection with the nuclear installations there are the following spent fuel (SF) management facilities:

- for the NPP Dukovany – spent fuel pools and Interim Spent Fuel Storage Facility (ISFSF) Dukovany;

- for the NPP Temelín – spent fuel pools;

- for the NRI,plc– wet accumulator tank for spent fuel, Spent Fuel Storage Facility (SFSF) and HLW storage facility.

Spent fuel (SF) produced by operation of the research reactor LVR–15 in NRI, plc is stored in the HLW storage facility, which is in agreement with the Czech law classified as an independent nuclear installation. The other research reactors in NRI, plc (LR–0) and FJFI Prague (VR–1) do not produce any SF due to their small thermal output („zero power reactors“) and limited time of operation.

Nuclear power plant Dukovany – SF pools

To assure safe storage of SF removed from reactors, SF pool is provided next to each reactor unit, its volume being 335 m3, and the SF is stored in it for a period of time required to reduce the residual heat output. After that period the thermal output and radiation of SF assemblies drops to a level permitting their transport in CASTOR-440/84 design-approved casks for transport and storage in Interim Spent Fuel Storage Facility (ISFSF) Dukovany.

In the pools the SF is stored in a compact grid with the capacity of 682 positions. SF Pools also contains 17 positions for hermetic cases to store damaged SF. Depending on the number of removed fuel assembly (FA) in the annual reactor campaign, the pools enable to store SF for a period of at least 7 years. Only in case of emergency removal of fuel from the core or during inspection of the reactor pressure vessel, a reserve grid is inserted into SF pool.

As at 31 December 2004 all four pools contained 2 270 FAs with the total weight 488 050 kg, with the weight of heavy metals (HM) about 271 000 kg.

Interim Spent Fuel Storage Facility (ISFSF) Dukovany

ISFSF Dukovany is situated inside the NPP Dukovany site. The basic element of ISFSF Dukovany is CASTOR-440/84 cask. It is used for transport and storage of 84 hexagonal SF assemblies from a VVER 440 reactor. In the cask, SF assemblies are stored dry in the environment filled with inert gas (He). A CASTOR-440/84 cask consists of a thick-walled cylindrical body with a bottom, provided with a double head closing system in the upper part plus a built-in structure to store a fuel assembly. The radial ribs on the outside of the cask envelope are to extend the heat transfer surface.

For the operation of ISFSF Dukovany, the cask is primarily used for storage, the transport function is only used to carry the cask to/from ISFSF Dukovany. In the Czech Republic, this cask has a design approval for transport and storage of SF.

A central building of ISFSF Dukovany is a ground-level hall with a combined structural system, with fixed poles from reinforced concrete and steel roof structure with a 6-meter module. The poles bear a crane runway and roof steel open-web girders supporting the roof structure. The building shell is mounted from panels made of reinforced concrete 100 mm thick. The storage part of the building is surrounded with a shielding concrete wall 5 m high and 500 mm thick. The floor is a slab of reinforced concrete with dust-free consolidating surface finish.

The total capacity of ISFSF Dukovany is 60 casks, while on 31 December 2005 ISFSF Dukovany contained 58 CASTOR-440/84 casks (4 872 FA). Annually4-5 containers are moved to ISFSF Dukovany. The annual generation of SF per reactor unit is approximately 10 t HM. The SF is stored in the SF pools for five years at least to be consequently loaded into the type approved transport and storage cask CASTOR-440/84.

In connection with fulfilment of storage capacity of ISFSF was built new storage facility (SFSF), similar construction as ISFSF with the total capacity of 133 CASTOR-440/84M casks. Commissioning of the SFSF starts currently.

Nuclear power plant Temelín

Similarly as in NPP Dukovany, the main production building in NPP Temelín has a storage pool for SF removed from the reactor with the volume 1440 m3, immediately next to the reactor well. The removed SF is stored here for a period of 12 years (in the course of NPP operation) or for at least 5 years (after NPP operation is closed) in a storage pool. SF pool is divided into 3 parts: two bigger parts have two grid sections each and the third has only one storage grid section.

The entire SF pool enables to store 678 FAs, 25 FAs in hermetic cases and 2 cluster cases (one position is used now). To the 31 December 2005 Pool No 1 contained 126 FA and Pool No 2 contained 84 FA, in total 210 FA.

In a standard storage regime, however, at least 163 positions shall remain free for potential emergency removal of assemblies from the entire reactor core.

Taking into account the total capacity of the storage pools, the construction of the SFSF (dry type) is planed. The siting permission was issued early 2006, construction permission is assumed in 2010 and commissioning in 2014.

NRI, plc – SF pool in the reactor hall

The SF pool is designed to store SF removed from the reactor core of LVR-15. It is an aluminium vessel in the floor of the reactor hall, protected with concrete on all sides and a steel-plated case. The vessel is covered with three cast iron plates 500 mm thick. The plates have two handling openings with lids. The upper edge of the reactor vessel is connected to the tank with a slopány pipe ending at the tank bottom. In 1996 the fuel was taken out from the wet accumulator tank and its condition inspected. The level and physical and chemical parameters of water in the tank are continually monitored.

As at 31 December 2004 the tank contained 30 FAs IRT-2M with the initial enrichment 36% wt. ^{235}U.

Building 211/7 – SF storage facility

The object includes two pools - A and B. The internal dimensions of pool A are 230 x 120 cm, depth 6 m and of pool B 440 x 120 cm, depth 6 m. The pools are built of heavy concrete cast between the inner and outer jacket of the stainless steel vessel. The pool walls and bottom are made of stainless

steel inner jacket, 50 cm of heavy concrete and outer stainless steel jacket. Racks made of aluminium alloy are placed on the pool bottom to store SF.

The facility premises are used for temporary storage of activated probes, loops and other active material (pool B) and temporary storage of SF (pool A). Shielded casks are used to transport SF and activated parts of probes and loops from the reactor into the wet accumulator tank and storage facility and to transport SF from the storage facility to the HLW storage facility (Building 211/8).

As at 31 December 2004 the facility contained 51 FAs IRT-2M with the initial enrichment 36% wt. ^{235}U and 12 FAs IRT-2M with the initial enrichment 80% wt. ^{235}U (from which 2 were in hermetic cases).

Building 211/8 – HLW storage facility

The high-level waste storage facility is designed to store solid RAW and spent fuel produced in NRI,plc in the research reactor VVR–S or LVR–15, developed as a result of an extensive reconstruction of the original Soviet research reactor VVR–S, and at its research workplaces. The facility was built in 1981 – 1988. Subsequently, modifications were made to meet Czech Regulatory Body (SÚJB) requirements.

The building is a prefabricated hall, ca. 13 x 34 m and 15 m tall. The interior is made up of eight concrete boxes of square ground plan for dry storage of solid RAW and SF type EK-10 (used in the VVR–S reactor until 1975) and two cylindrical pools for wet storage of SF type IRT-M. The SF EK-10 is stored in dry conditions, in drums backfilled with concrete. The pools consist of inner stainless steel vessel placed in another vessel of carbon steel embedded in concrete. The pool diameter is 4.6 m and water level 5 m. The storage capacity in the pool A is 300 FAs and in the pool B 465 FAs.

In Pool A there is no FA, Pool B contains 256 FAs and in Box 5 there are190 FAs.

The storage facility for SF has to assure the following functions:

- subcriticality of the stored SF;
- removal of residual heat from PS;
- protection against radioactive radiation.

With the increasing number of spent fuel assembly an approach to criticality analysis is being reconsidered. Czech Regulatory Body is promoting projects to support development in burn-up credit possible application and to keep regulatory body an extremely conservative approach. The first project was “Technical Aspects and Technology of Burn-up Credit Implementation into Criticality Analyses of Spent Fuel Management Systems”. The solution enabled the nuclear safety regulatory body to review the topic and find technical knowledge related to burn-up credit implementation in spent fuel management systems which is at present a very advanced feature of spent fuel criticality analyses world-wide. The second project is “Burn-up Credit and Partial Boron Credit Implementation in Spent Fuel Pools at VVER Reactors”. The topic is burn-up credit (BUC) and partial boron credit (PBC) implementations in pools at reactors. So far the criticality analyses have supposed that the spent fuel is at its most reactive (fresh) state and no soluble absorber (boron as boric acid) is present in the pool water; both assumptions are considered now to be too conservative approach.

CURRENT STATUS AND POTENTIAL NEEDS OF BURN-UP CREDIT IN JAPAN

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Abstract

This paper describes the current status and future needs of burn-up credit (BUC) in Japan. The first application of BUC is the spent nuclear fuel (SNF) receiving pool and the dissolver at Rokkasho Reprocessing Plant. BUC in other fuel cycle facilities including SNF transport has not been considered in Japan. However, the concept of BUC has been attractive in Japan and the related techniques to BUC are continuously developed in the field of burn-up analysis of post irradiation examinations for validation of reactor physics methodologies. Further adoption of BUC in spent fuel management facilities is expected taking into account increase in burn-up value and initial ^{235}U enrichment of nuclear fuels.

Introduction

Nuclear power plants in Japan discharge approximately 900 to 1000 tonnes of spent nuclear fuels (SNF) a year. Because Rokkasho reprocessing plant (RRP) of Japan Nuclear Fuel Limited (JNFL) has the ability to reprocess SNF of 800 tonnes a year, the residual amount of SNF that is waiting for reprocessing will be increased. In this context, taking account of the decrease in reactivity of SNF according to burn-up into the criticality safety, which is well known as burn-up credit (BUC), is crucial to improve the efficiency of storage and transport of SNF.

Current status of burn-up credit in Japan

In Japan, BUC has already been introduced into the criticality safety evaluation of SNF receiving pools and dissolver at RRP. In the receiving pools which have been in operation since 1999, the burn-up value of SNF and remaining enrichment of uranium-235 in them have been measured by non-destructive methods. Then SNF is categorised by the remaining enrichment into (a) less than 2.0wt.% and (b) between 2.0wt.% to 3.5wt.%, in order to select a storage rack. For the dissolver, in order to decrease its neutron multiplication factor to satisfy the criticality safety requirement, gadolinium nitrate solution is added into the dissolver depending on the measured burn-up and initial enrichment values.

In other case, BUC has been introduced into spent fuel storage pools at BWR reactor sites at the level of integral burnable absorber (Gd credit).

Table 1. Status of BUC introduction in Japan

	Wet Storage at Reactor Site	Interim Storage [a]	Transport	Reprocessing	
				Spent fuel pool	Dissolver
PWR	N.A.[b]	-	N.A.	(U,Pu) [d]	(U,Pu)
BWR	Gd [c]	N.A.	N.A.	(U,Pu)	(U,Pu)

[a] Dry Storage system using metal casks. BWR fuel assemblies are considered.
[b] N.A: Not applicable; [c] Gd: Gd credit; [d] (U,Pu): U and Pu isotopes are considered as credit.

Table 1 summarises the status of BUC introduction into each fuel cycle facilities in Japan. It shows that BUC in other fuel cycle facilities including SNF transport is not yet considered in Japan. This is because the pressure of accumulation of SNF in Japan is released for a while since the operation of the spent fuel storage pools in RRP (3,000t) and proposal of construction of an interim storage facility (5,000t) using dry-type metal casks by Recyclable-Fuel Storage Company (RFS) in Aomori. The latter programme was accepted by the local government.

Future needs of BUC

However, potential demands for adoption of BUC and improvement of related techniques have been still high. First need is to decrease gadolinium usage into the dissolver of RRP by adopting BUC with fission products in order to reduce radioactive wastes generated by SNF reprocessing.

Other need is optimisation of managing SNF. For example, BUC is not yet introduced in the transport casks in Japan. However, SNF transport casks are one of the target facilities of BUC implementation in other countries. Considering that higher burn-up fuel with increased enrichment

will be introduced, BUC is a key technology in the design of transport cask. For this purpose, in Japan, safety research activities on fuel cycle involving uranium fuel beyond 5%-enrichment was initiated among several institutes in Japan from FY 2005 [1].

Status of PIE activities in Japan

We will focus on PIE activities in Japan Atomic Energy Agency (JAEA), former Japan Atomic Energy Research Institute (JAERI) since PIE conducted in private sectors have not been opened and will be used for limited purposes for their research and development.

There is no doubt that isotopic composition data, which are experimentally measured in post irradiation examination (PIE) are important to validate computer programs and data libraries to be used in BUC analysis. Japan was one of active countries in the field of PIE to obtain such data in 1980's and 1990's and JAERI published several reports [2,3]. Several PIE that were dedicated to evaluate mechanical properties and fuel behaviour of SNF have also been carried out at JAERI by collaborative works with other institutes.

After 2000, JAEA has no own PIE project because of restructuring of research organisation and resources. However, PIE for studies on fuel behaviour of SNF are still carried out in JAEA by collaborative works with other institutes, and PIE under contract with Japan Nuclear Safety Organisation (JNES) [4] was conducted. In the PIE, data were taken from the view point of validation of reactor physics methodologies for newly developed BWR fuel of 9×9 type.

Concerning international PIE programme, JAERI participated in the ARIANE programme [5]. But now JAEA does not participate in any international framework of PIE dedicated to obtain isotopic compositions.

Conclusion

BUC has already been introduced into the criticality safety evaluation of SNF receiving pools and dissolver at RRP. However, because the accumulation of SNF in Japan is increasing, BUC is still an attractive concept and further adoption of it in spent fuel management process is expected. This is because BUC is not yet applied to SNF transport cask in Japan and introduction of higher enriched fuel is under consideration. Also, BUC taking fission products into account will be interested for obtaining more benefit of application of BUC.

PIE in JAEA is conducted under contract with other institutes such as JNES in the field of burn-up analysis including PIE for reactor physics study. There is no PIE activities dedicated to obtain isotopic composition for BUC. However, the technical development in the field of reactor physics will be also applied to introduction of BUC.

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VALUABLE INFORMATION FOR BURN-UP CREDIT LICENSING REVIEW

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Abstract

Burn-up credit is an old and established criticality safety control. The traditional method using reactor design and simulation codes requires information about the reactor operation, including measurements and other observations, but not about the final individual nuclide composition of the fuel (though such information is always useful). Validation normally requires access to proprietary information. Some of the validation process is carried out as a routine procedure by the reactor operator. Recent (last 20 years) attempts of burn-up credit have often involved determination of individual fuel nuclide concentrations and validation of their properties. This is a very complicated method that involves a combination of a very large number of uncertainties. Independent burn-up measurements may be necessary to support this method. Post-irradiation experiments (PIEs) on fuel samples provide opportunities for discussions on importance of various specifications, on uncertainties and hopefully result in data that can be used in validation of burn-up credit methods. Physics differences between fuel types such as VVER and PWR may be quite small. Comparison of PIE results for different fuel types gives information about the flexibility and range of physics covered by the validated methods. This is valuable to licensing authorities even if that fuel type is not used in reactors in that country. Transport applications may lead to such unusual fuel being shipped on territory (including sea vessels) of that country.

Introduction

If the potential for criticality in a specific application may increase due to past or future reactor operation using the fuel, special action is required, This effect, not to be confused with the related effect of breeding (excludes influence of fission products), may apply to all the fuel constituents or to a selected sub range.

Light-water power reactors, through transmutation, produce the highly fissile nuclide ^{239}Pu from the neutron absorbing nuclide ^{238}U. Credit for other transmutation and decay processes is required to allow irradiated fuel to be stored and shipped under the same criticality safety controls as unirradiated fuel.

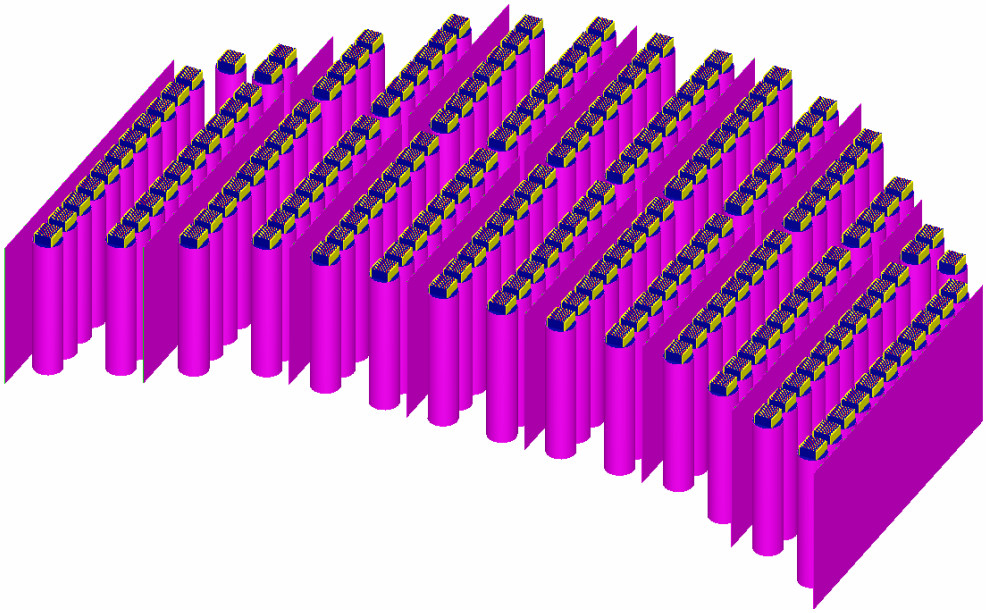
Addition of a burnable absorber to the fuel before reactor operation gives a similar effect by transmutation of burnable absorber nuclides into other less absorbing nuclides. If presence of burnable absorbers in the fuel can be assured, other controls may be relaxed or capacity can be increased. Such control is called burnable absorber credit. Reactor operation changes the composition of the absorber together with the remainder of the fuel and the net effect on criticality safety needs to be evaluated.

If the total effect of the transmutation and decay can be demonstrated to reduce the potential for a criticality accident and if a minimum degree of such transmutation and decay can be assured, other controls may be relaxed or capacity can be increased. Such control is called burn-up credit.

Whatever the result of reactor operation is, the criticality safety effects from the fuel properties in selected applications must be evaluated. The ambition level of such an evaluation should depend on the circumstances. Critical experiments together with other measurements and observations can be used to prepare benchmarks for different applications.

A post-irradiation experiment (PIE) on a selected irradiated fuel sample is a way to learn about the effects of reactor operation. There are many types of measurements and tests that can be performed on such a sample.

Figure 1. MTR fuel element storage, upper level with tubes and steel-clad Cd plates



Accounting for irradiation and radioactive decay

Storage and transport of irradiated fuel are operations that have been carried out since the first reactors were started. What type of evaluation is required to demonstrate that the criticality safety will be maintained after the fuel has been in an operating reactor?

The simplest type of accounting for irradiation and decay effects is to assure that the ^{239}Pu production and the ^{238}U loss are at least balanced by a net absorption effect from other nuclides. This is not burn-up credit but the irradiation effect must be evaluated. This is normally achieved by reference to "common knowledge". That knowledge is based on many years of studies and experience with light-water reactors and of methods for breeding of fissile nuclides. Formal evaluations for specific applications are normally not required. However, the regulator may request evidence in specific cases. The international transport regulations support such requirements. Information on individual nuclide concentrations is not required.

If it is found that the neutron multiplication factor in any application, including accident scenarios and consideration of uncertainty effects, is increased for irradiated fuel, a safety evaluation is required. It is not optional, not a credit.

Burnable absorbers are used to reduce the initial reactivity of fuel assemblies in the reactor. The reactivity of such fuel, in particular for Boiling Water Reactors (BWRs) is known to increase and reach an optimum with irradiation in the reactor. Similar (but not identical) behaviour is expected in applications outside the reactor. The irradiation effect must be accounted for if irradiation of the fuel is a realistic possibility. In the past, such accounting has been made without detailed studies of influences of individual nuclides. The integral effect has been requested and applied. This type of evaluation is required, not optional. If credit is taken for the presence of burnable absorbers to reduce the neutron multiplication factor, it is not burn-up credit but burnable absorber credit.

If it is found that the neutron multiplication factor in an application, including accident scenarios and consideration of uncertainty effects, is reduced for irradiated fuel, credit for the irradiation and decay effect may be obtained. It is an option, not a requirement.

The above application types refer to criticality safety design and operation. The primary responsibility and incentive for such evaluations normally belong to the nuclear industry, not to the regulator. However, there are regulatory activities that benefit from better understanding and information on irradiated fuel. These include emergency preparedness and response for which accurate information on the real margin to criticality can be taken advantage of to optimise emergency operations. Radiation protection, security and non-proliferation issues are not considered here.

Whatever consideration of the irradiation and decay effects that are requested, the value of measurements and other observations that give information on irradiated fuel properties is obvious. The value depends on the quality on the specifications of the fuel irradiation and decay history, on the similarity between these specifications and those of the application as well as on the quality of the evaluations. Even when traditional methods that are not based on individual nuclide properties and concentrations are used, having such information simplifies validation and control. PIE evaluation is one of the methods to obtain additional or to confirm existing information.

Requested information – scientific research or safety engineering?

It is important to recognise that the need for information is related to the application. Irradiated fuel from light-water reactors have been stored and transported for a long time and evaluation of the irradiation and decay influence on criticality safety is not new. During the last twenty years, it appears as if a more scientific approach, as opposed to a safety engineering approach, has become dominant. The result of this scientific approach often resembles inefficient bureaucracy rather than progress.

Burn-up credit and burnable absorber credit are additional criticality safety controls that increase the safety margin of an application. Advantage of this can be taken by reducing other controls or by increasing storage or transport capacity as long as the overall criticality and total (integral) safety is acceptable.

In the past, it has been noticed that unofficial burn-up credit has often been taken. The priority of administrative and technical controls has been reduced under the prejudiced conclusion that the reactivity decrease due to irradiation and decay is the same in the application as in the reactor. Even though some of the unofficial burn-up credit has had substance, it has been demonstrated that the margins may have been much smaller than expected without careful evaluation. Recognising false feelings of safety is the responsibility of any criticality safety specialist, not only of a regulator. This may be particularly serious in emergency situations.

The key property is the neutron multiplication factor for the system being evaluated. If it is maximised and subcritical, accounting for all credible scenarios as well as taking known and unknown uncertainties into account, the application is safe.

Scientifically, the solution is to model and verify the reactor and its operation in every detail, to determine the concentration of each nuclide accurately as a function of space and time and finally to determine and validate the cross-sections for those nuclides.

Using a safety engineering approach, advantage is taken of the reactor operator's method for designing the core and for following the fuel changes continuously during the reactor operation. The resulting information given for burn-up credit application contains specifications for the fuel properties. These properties may be individual nuclide concentrations, "lumped" nuclide concentrations or combinations of such specifications.

However, the properties may also be given as a spectrum-weighted, energy-group-condensed, fuel/water-homogenised and/or macroscopic cross-section set that covers all fuel zones and cooling times to be evaluated. This method would be validated by the normal operation of the reactor and the need for measurement-based adjustments to the simulation. It may be possible today to model a reactor and its operation in detail using Monte Carlo and continuous energy cross-sections, but it is not necessary for burn-up credit in general. The transfer from reactor application to burn-up credit application can be carried out if the reactor operator's method can handle the scenarios required for criticality safety evaluation or if the cross-sections can be transferred to the method used in criticality safety evaluations.

In Sweden, the ABB Atom company (later Westinghouse) about 1975 developed software to routinely transfer cross-sections from the PHOENIX core design and management system to a format recognised by the KENO Monte Carlo code. This has been taken advantage of in applications of burnable absorber credit and in studies of burn-up credit.

Independent verification of nominal fuel properties

Normal operation of a reactor gives many opportunities for direct verification of the properties of the reactor core and for indirect verification of individual fuel assembly properties. In addition, any measured or otherwise verified properties of each fuel assembly after reactor operation give valuable information on the history of the fuel. The histories of various assemblies should be consistent with each other and with the integral properties of the reactor (power generation, residual heat, radiation, activation, etc.).

The most valuable method to support evaluation of irradiation effects on criticality safety is one that can simulate the reactor operation well. Such methods require proprietary information from the reactor operator for verification and validation. The detailed reactor operating data can be simplified to various degrees in order to support independent generation of detailed fuel properties. Such simplification should be carried out by the reactor operator. The simplification process shall be made in a way that it can be verified by comparing with detailed records if the licensing authority so requests.

A realistic model of the radioactive decay process after reactor operation (cooling) is important.

The traditional method

The traditional method, available for burn-up credit applications since the 1960s, is to generate the cross-sections corresponding to the final fuel properties in a single process. This process may involve many iterations and updates but the result is a cross-section set that can be used to calculate the neutron multiplication factor for an application with requested accuracy. The geometry of the final application may not be possible to model with the core design or management methods but traditional criticality safety methods can be used to determine the differences in k_{eff} due a core design method model compared with the final application model.

The traditional method may be able to produce concentrations for many important nuclides. This is useful but not necessary. What is important is that they can reproduce the behaviour of the reactor and thus generate input for other methods such that the nuclide concentrations can be determined when needed. This will not be needed for all burn-up credit applications.

Validation of this method is carried out daily at reactors. Validation for particular criticality safety applications need to be carried out for each new application type. One parameter that often needs tightening in criticality safety applications is the mesh size in regions between the fuel assemblies. More regions between the fuel assemblies may require modification of the method. Treatment of voids between fuel positions needs consideration. Reflector and construction materials not usually found in reactors may also need consideration. A special complication may be that flux distributions with sharp peaks that could not be realistic in a reactor are possible in storage or transport applications. The method must be able to handle such peaks adequately to produce reliable cross-sections.

The traditional method has been used for many years in BWR fixed burnable absorber credit applications, in PWR burn-up credit based on minimum burn-up in any axial 50 cm region, in burn-up credit for MTR fuel storage and international transport, etc.

The new method

Since burn-up credit was rediscovered in the 1980s, a more complicated method has been dominating the studies related to future applications.

The process is divided into two distinct parts. The first sub-process is to generate nuclide concentrations and the second sub-process is to generate cross-sections for those nuclides. The properties of the irradiated and cooled fuel depend on the combination of all nuclide concentrations and all cross-sections. However, the validation requirements are often related to individual concentrations and cross-sections for each nuclide. This is new and such ideas are not applied to fresh fuel and to other common mixtures such as concrete, steel, boron, iron, etc. It is primarily the integral effect that is validated. Even increasing the uranium enrichment of ^{235}U a single percent requires additional validation. The individual cross-sections are not changed and the concentrations are not involved in this requirement.

There is no doubt that knowing all the details about every nuclide and every detail about every part of a fuel assembly can be useful in burn-up credit. The question is why do we require more knowledge than we need for a specific application? This question has not been answered very clearly in modern burn-up credit studies and guides.

Identification of credible variations and potential incidents

When the real properties of the fuel are adequately known, they need to be combined with other properties of the system being evaluated. Potential variations and incidents (during reactor operation as well as later) need to be identified and evaluated. Can the variations or incidents pass unnoticed or will they be acted on before the criticality safety control is reduced?

Will any fission products be lost due to misloading of a fresh fuel assembly into a burn-up credit controlled storage rack or will such a loss occur before the misloading is discovered and corrected? High burn-up fuel is more likely to fail during tests required by transport regulations but the required burn-up may be higher than accounted for in the burn-up credit based design and this can be taken into account in the criticality safety assessment. Criticality due to simultaneous accidental water flooding of the internals of a dry cask containing irradiated fuel during transport or storage and extreme damage to the same fuel in the same cask may have an acceptably low probability.

Visualising relations between k_{eff}, the neutron flux and specifications for the evaluated system

The neutron multiplication factor k_{eff} is a system property. It is determined by the individual specifications x_i for the system and on specifications on how these individual specifications are configured into a single system. The geometry configuration is required to define the system. The geometry configuration may determine the temperature distribution in the system (residual heat, cooling, etc.) and together they will determine the neutron flux, Φ. This flux is shown here as a single scalar variable but is really a complicated function of space and energy. In a criticality safety calculation requesting the system property k_{eff}, there is no heat generation from fission.

$$k_{eff} = k(x_1, x_2 \ldots x_n, T(x_1, x_2 \ldots x_n), \Phi(T, x_1, x_2 \ldots x_n)) = k(f_1(x_1,T,\Phi), f_2(x_2,T,\Phi) \ldots, f_n(x_n,T,\Phi))$$

If T is treated as an independent specification for each geometry region, it can be included in the specifications x_i. A realistic temperature distribution will usually reduce k_{eff} since the temperature is

higher inside the fuel assemblies than between the fuel assemblies. Here, a constant T is assumed in the system and it is covered in x_i.

The uncertainty in k_{eff} can be visualised with the following simplified equation. The correlation terms should be viewed as if a change δx_i in x_i leads to a change δx_j in x_j. The sums are over all non-identical indices i and j. The dependencies between two specifications are not necessarily reciprocal. An example is a vertical stack of assemblies relying on gravity for packing density. A change in the vertical geometry of an assembly may change the positions of assemblies above. The reciprocal relation is not there.

$$\delta k^2 = \sum((\partial k/\partial x_i)^2 * \delta x_i^2) + \sum\sum ((\partial k/\partial x_i) * (\partial k/x_j) * \delta x_i \delta x_j) + (\partial k/\partial \Phi)^2 * (\sum((\partial \Phi/\partial x_i)^2 * \delta x_i^2) + \sum\sum ((\partial \Phi/\partial x_i) * (\partial \Phi/x_j) * \delta x_i \delta x_j)) + \text{more correlation terms}$$

For simplicity, assume that there are no correlations between any of the x_i specifications (not credible in a general equation but useful for limited studies). The equation becomes:

$$\delta k^2 = \sum((\partial k/\partial x_i)^2 * \delta x_i^2) + (\partial k/\partial \Phi)^2 * (\sum((\partial \Phi/\partial x_i)^2 * \delta x_i^2)$$

This equation correctly shows that the effects of all flux changes are combined in one uncertainty term. The more flux changes, the more the likelihood for non-linearity becomes. The sensitivity to reactivity changes may be local; the overall change of the neutron flux will often determine the total reactivity effect.

Combination of variations or uncertainties using simplified model of k_{eff} and neutron flux

The traditional method of applying burn-up credit is not very complicated. It can be complicated to collect all essential parameters related to reactor operation. However, the simplified specifications resulting from the associated reactor simulation should be easy to use in applications of other methods.

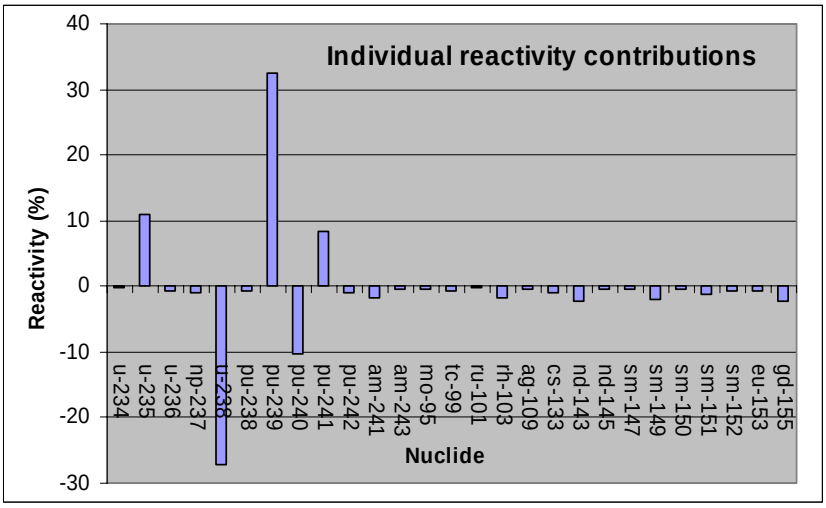
The new method of identifying each nuclide and determining its concentration is very complicated and resource-requiring. Correlations between variations, incidents, approximations, uncertainties, etc. can be estimated using the simplified relationship between k_{eff}, the neutron flux and the system specifications x_i given in the previous section.

An irradiated fuel assembly region from the OECD/NEA burn-up credit study Phase IIC 0 was selected for a preliminary study of correlations between reactivity influences of various nuclides.

Using SCALE 5 0, an infinite array resulted in $k_\infty = 0.904$. Figure 2 shows the effect of removing an individual nuclide from the fuel. Removing multiple nuclides will not result in the same combined reactivity as the sum of individual reactivities.

Further evaluations will support development of an engineering judgement for determining when correlations between parameters are important and not. It is not sufficient to study correlations between nuclides but also geometry variations need to be considered.

Figure 2. Calculated reactivities of individual nuclides removed from irradiated fuel



A burn-up credit reference library

During 2005, a compilation of references was made to support licensing review of burn-up credit and burnable absorber credit applications in Sweden 0. More than one thousand documents in the form of published reports, conference presentations, licensing requirements and guides, journal articles, working papers from various studies, design information from licensing applications, etc. were included.

The major part of the list of documents will be offered to OECD/NEA, for publication on its burn-up credit web page. Some of the references are proprietary or not intended for wide release and will not be included. Many valuable references, known to other specialists, are probably missing from the list. Further, the subject of burn-up credit touches on so many technical areas that some subjective selection criteria are needed. The development, testing and use of reactor physics theory, codes and data belong to a category that needs screening.

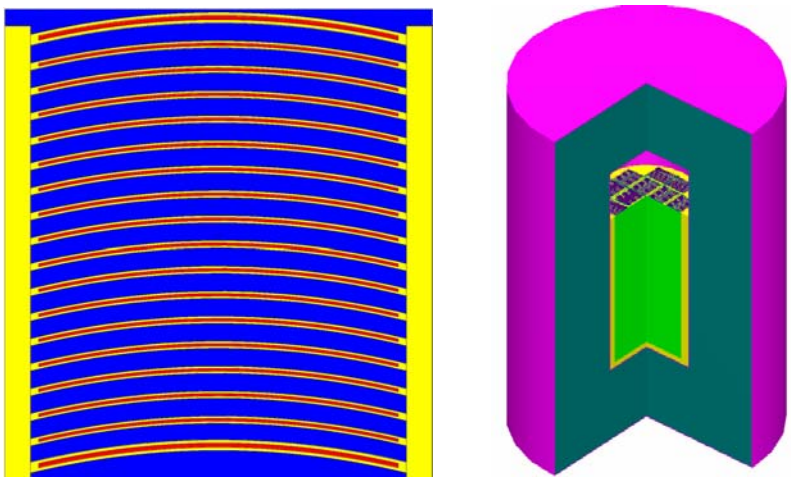
A recent burn-up credit application in Sweden

The Material Testing Reactors (MTRs) R2 (50 MWth) and R2-0 (<1 MWth) in Studsvik, Sweden were permanently shut-down in 2005. The conversion to low enrichment (from 93 wt-% to 20 wt-%) ^{235}U had been completed. There were no high-enriched fuel elements remaining at the site. However, some of the fuel elements were only slightly irradiated.

To take advantage of existing storage capacity in a separate storage facility, all fuel was planned to be moved from the reactor building to that facility. This facility had already been approved for storage of MTR fuel elements using burn-up credit to cover serious incidents. Since some of the fuel elements had lower burn-up than previously required, a more careful burn-up credit study was made to support storage of all existing elements 0.

The internal Studsvik area transfer was planned to be made using an existing transport cask, designed and previously licensed for international dry transport of the high-enriched fuel element type using burn-up credit. It had not been licensed for the low-enriched fuel element type which would

Figure 3. MTR fuel element (left) and R2 fuel element transport cask (right)



have required considerably more burn-up credit and other restrictions (much higher ^{235}U mass per element). The proposed solution was to block some of the central cask fuel element positions for the least irradiated elements. The final solution was to keep those positions blocked for all transfers. The internal transfer was planned with water in the cask.

No credit was requested for the fission products. The applicant used SCALE 5 0 with SAS2 0 to obtain the irradiated fuel composition for later calculations using KENOV.a 0. Comparisons with previous calculations using the reactor operator's core design and simulation method as well as with other methods were made. SAS2 seemed to be adequate. During the licensing review, SCALE 5 with TRITON/NEWT 0 was used to model fuel irradiation. Accurate descriptions of the curved-plated MTR fuel elements could be modelled. This was not necessary but provided a test of the TRITON/NEWT sequence. KENO-VI 0 and MCNP5 0 were subsequently used for criticality safety review of the proposed transport and storage systems. Figures 1 and 3-4 show KENO-VI geometry models of licensing review models of storage and transport.

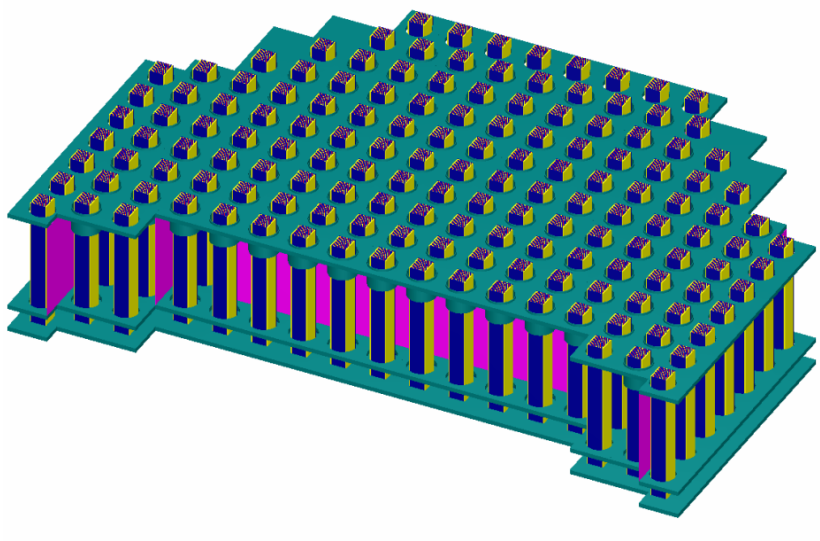
A problem with the LATTICECELL option for slabs was pointed out by the developers of SCALE during the review. A suggestion to use the MULTIREGION option was made. However, it turned out that the TRITON/NEWT sequence failed when the MULTIREGION option was selected. The fission products were dropped from the NEWT calculations, even after being particularly specified in the input. This probably resulted in a softer energy spectrum and less reactive fuel. This type of error for rarely used options should be expected in new software. TRITON/NEWT appears very valuable for future applications of burn-up credit. The next version of SCALE 5 will contain substantial improvements in speed and stability.

Value of post-irradiation experiment evaluation in licensing reviews

Any reliable information that can be used to test any of the complicated relations involved in fuel irradiation and decay is valuable. PIEs provide such information. Even when the results have higher uncertainties than requested or expected, the discussions and evaluations of the results are valuable in themselves.

For a designer, it is most useful to have data that are as similar as possible to the proposed design or operation. For a licensing authority, it is also valuable to have data that go outside the expected specifications of the proposed design or operation. Verification of a method should cover parameter

Figure 4. MTR fuel element storage, lower level with grids and steel-clad Cd plates



ranges that go beyond those that are expected in order to test the algorithms and approximations involved in the method. For this reason the difference between VVER and PWR fuel and reactor operation specifications may be considered small.

A licensing authority may be involved in transport approvals for fuels that have not been used in reactors in that country. Use of a validated method by the package designer and availability of benchmarks to validate methods used in the safety review of the design is important for such licensing evaluations.

Conclusions

Post-irradiation experiments on VVER fuel samples and the associated preparation and evaluation, including discussions, would be beneficial not only to the industry but also to the licensing authorities involved in burn-up credit and burnable absorber credit. The cost, shared between many parts, appears to be very reasonable. Care should be taken to specify the fuel history as accurately as possible and to reduce uncertainties in the experiments.

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SAFETY ASSESSMENT FOR THE TRANSPORTATION AND STORAGE OF SPENT NUCLEAR FUEL IN UKRAINE

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Abstract

The following main systems for management of VVER spent nuclear fuel acting at that moment in Ukraine:

- VVER-1000 reactor pools;
- VVER-440 reactor pools;
- TK-13 transport casks for VVER-1000 spent fuel assemblies;
- TK-6 transport casks for VVER-440 spent fuel assemblies;
- the Sierra Nuclear dry storage casks for VVER-1000 spent fuel of the Zaporizhya NPP.

The criticality analysis has been carried out according to the general requirements of documents in force, i.e. without the burn-up credit, without the credit of absorbers dissolved in water and removable absorbers.

The analysis has demonstrated that not all spent fuel management systems meet regulatory requirements in force for modern nuclear fuel. First of all, this concerns the reactor pools and transport casks for VVER-440 fuel and dry storage casks for VVER-1000 at the Zaporizhya NPP.

But the inconsistencies between these systems and regulatory requirements should not be considered as drawbacks of these systems. What is more, they should be considered the consequences of excessive conservatism of Ukrainian regulatory requirements in force.

It was demonstrated that the excessive conservatism laid previously into the requirements on nuclear safety in Ukraine due to insufficient development of means for modelling processes in nuclear fuel can be considerably decreased through using more real modelling fuel systems. If such modelling is performed at the state-of-the-art level, based on more complete understanding the processes in fuel systems, then removal of the excessive conservatism will not reduce the safety of nuclear dangerous systems.

Introduction

The following main systems for management of VVER spent nuclear fuel acting at that moment in Ukraine:

- VVER-1000 reactor pools;
- VVER-440 reactor pools;
- TK-13 transport casks for VVER-1000 spent fuel assemblies;
- TK-6 transport casks for VVER-440 spent fuel assemblies;
- the Sierra Nuclear dry storage casks for VVER-1000 spent fuel of the Zaporizhya NPP.

Implementing new types of fuel, new devices and mechanisms, including those developed and manufactured abroad, at enterprises of the Ukrainian power industry makes it necessary to license them in advance by the Ukrainian Regulatory Authority. From time to time, situations occur when these systems or their close analogues have been already used in some countries and have successively passed licensing by the relevant regulatory authorities, however, they do not meet the regulatory requirements in force in Ukraine.

It is possible to refer, as an example, to the spent nuclear fuel storage facility commissioned presently at Zaporizhya NPP site. This storage facility uses casks whose close analogues have already passed licensing and have been manufactured and used in the USA for more than 10 years. In spite of this, there were quite serious problems related to nuclear safety appeared in Ukraine in licensing these casks. They were caused by differences in the nuclear legislation in force in both countries, at first sight, not very significant. This example obviously demonstrates urgency and importance of a preliminary comparative analysis to be performed concerning nuclear legislation in force in manufacturing country and consuming one.

Preliminary analysis of the regulations in force in Ukraine concerning nuclear safety of spent nuclear fuel management systems shows that some regulatory requirements in force are too conservative in view of current international practice.

Requirements on nuclear safety in force in Ukraine

The main Ukrainian regulatory document containing requirements on nuclear safety as regards systems for spent nuclear fuel storage is the “Safety Rules for Storage and Transportation of Nuclear Fuel at Nuclear Power Facilities” (PNAE-G-14-029). The Rules cover both systems for management of fresh and spent nuclear fuel of NPPs and independent storage facilities for spent nuclear fuel, i.e. storage facility located beyond the reactor building.

The rules contain the following general principle on safety assurance: effective neutron multiplication factor should not exceed 0.95 in normal operation conditions and in case of design-basis accidents.

The following principle on safety assurance covers all systems for nuclear fuel management: the design should provide for technical and organisational measures aimed at the prevention of design-basis accidents and the limitation of their consequences, and the safety assurance for each of design

initiating events with one independent concurrent failure of any of the following elements of safety systems: active or passive element which has movable mechanical parts, or one human error independent on the initiating event. In addition to one failure independent of initiating event of one of the above-listed elements, it is required to consider the undetected failures of elements unmonitored during operation, which impact on the progress of an accident and cause the violation of safe operation limits.

The design of equipment for the nuclear fuel storage and management system should provide for nuclear safety mainly owing to the location of fuel assemblies with certain pitch.

Nuclear safety analysis should be performed taking into account the conditions when the nuclear fuel storage and management system has maximum K_{eff} in compliance with the following requirements:

- To consider that all fuel has maximum enrichment when nuclear fuel with different level of enrichment are located in the storage facility.

- It is necessary to consider such composition of spent nuclear fuel that causes maximum neutron multiplication factor if reactivity of spent nuclear fuel can increase in the process of burn-up.

- It is necessary to consider the maximum design capacity of the storage facilities.

- It is necessary to consider the inaccuracy of calculation methods, concentration and isotope composition of absorbers, tolerances in the production.

- It is necessary to neglect the presence of absorbent elements in fuel assemblies or structures of racks if they are not fixed or if their effectiveness decreases as a result of initiating events.

- It is necessary to consider such an amount, distribution and density of a moderator (in particular, water) in the system as a result of initiating events that causes maximum effective neutron multiplication factor.

- It is necessary to tolerate the presence of the reflector.

- It is necessary to consider the state which cause maximum K_{eff} in case of temperature change in normal operation conditions and due to initiating events.

- It is necessary to consider the possibility of increasing K_{eff} due to nuclear fuel burn-up because of the change of nuclide composition during burn-up related to the accumulation of nuclear-hazardous fissile nuclides. Spent nuclear fuel should be considered as fresh if K_{eff} decreases in the process of burn-up excluding cases when burn-up credit principle is applied and the burn-up is monitored by means of special devices.

- To consider that burnable absorbers are absent in fuel assemblies containing burnable absorbers.

- To consider that homogeneous absorbers are absent for the storage facilities with homogeneous absorbers (for example, borated water).

Results of conservatism in Ukrainian regulatory requirements on nuclear safety

Criticality analysis spent fuel management systems has been carried out according to the requirements of documents in force without the burn-up credit, without the credit of absorbers dissolved in water and removable absorbers. In addition, the analysis has assumed the optimal neutron moderation. Mathematically this assumption means that the system is filled with water-air mixture having the optimal density which corresponds to the maximum neutron multiplication factor in full compliance with regulatory documents in force as well as the existing international practice. The following results have been obtained under these conditions:

- the VVER-1000 reactor pool: $K_{eff\,max} = 0.9289 \pm 0.0008$;
- the VVER-440 reactor pool: $K_{eff\,max} = 1.3394 \pm 0.0007$;

  Consequences: Measurements under development.

- TK-13 transport cask for VVER-1000 fuel assemblies: $K_{eff\,max} = 0.8873 \pm 0.0010$;
- TK-6 transport cask for VVER-440 fuel assemblies: $K_{eff\,max} = 1.1235 \pm 0.0009$;

  Consequences: Partial loading of cask.

- the Sierra Nuclear dry storage cask for VVER-1000 spent fuel of the Zaporizhya NPP: $K_{eff\,max} = 1.1930 \pm 0.0008$.

  Consequences: Implementation control rod credit and burn-up credit, see below.

Hence, the analysis has demonstrated that not all spent fuel management systems meet general regulatory requirements in force for modern nuclear fuel. First of all, this concerns the reactor pools and transport casks for VVER-440 fuel and dry storage casks for VVER-1000 at the Zaporizhya NPP.

This situation has resulted from commissioning new fuel types and fuel cycles since the VVER-440 systems were originally designed for fuel with 3.6% enrichment, but now this value constitutes 4.4%. The hole in the fuel pellet center has decreased and the amount of uranium in each assembly has increased as a result. As far as VVER-1000 is concerned, these changes are not so fundamental in terms of nuclear safety. Therefore both the reactor pool and transport cask for VVER-1000 practically meet the regulatory requirements.

The situation with dry storage casks for VVER-1000 spent fuel at Zaporizhya NPP also requires attention. American analogues VSC-24 of these casks were designed for fuel assemblies with less amount of uranium in the assembly (PWR 17x17 assembly) and multiplication properties corresponding to fresh fuel with 1.35 weight percent enrichment by ^{235}U.

It should be also emphasised that American regulatory requirements are more flexible and less conservative than Ukrainian ones.

The most reliable solution in case of violating regulations in force is partial loading of the spent nuclear fuel management system with the following expert safety assessment.

But the inconsistencies between these systems and regulatory requirements should not be considered as drawbacks of these systems. What is more, they should be considered the consequences of excessive conservatism of Ukrainian regulatory requirements in force.

Some aspects of nuclear safety analysis for ZNPP spent nuclear fuel dry storage facility

Two types of credit were analysed:

1) control rod credit;

2) burn-up credit;

The neutron multiplication factor in case of cask loading with fuel assembly together with control rod clusters reaches the following value:

$$K_{eff} = 0.9851 \pm 0.0009$$

The credit of fuel burn-up results in reducing the neutron multiplication factor in the cask from 1.1936±0.0008 (fresh fuel without control rods, optimal moderation) to

$$K_{eff} = 0.9416 \pm 0.0007 \text{ (burn-up – 50 MW·d/kgU).}$$

The results show that spent fuel assembly with initial enrichment 4.4% may not be installed without additional absorbers, such as control rods etc., even with burn-up credit

Two Branch Regulatory Documents have been prepared: “Storage of Spent Fuel in VVER-1000 Ventilated Cask of Spent Fuel Dry Storage Facility. Authorization Procedure, Requirements on Documentation and Calculations of Neutron Physical Characteristics of VSB Loading at ZNPP Spent Fuel Dry Storage Facility.”

The document establishes requirements on nuclear safety justification for loadings of cask with sent nuclear fuel regarding:

- procedure for obtaining authorisations for loading of ventilated casks of the dry storage facility for VVER-1000 spent fuel;
- content of documents that justify the safety of cask operation;
- scope of nuclear safety calculations for cask loading.

The second document is entitled “Methodology of Spent Nuclear Fuel Burn-up Credit as Nuclear Safety Parameter for VSC Fuel Loadings at Spent Fuel Dry Storage Facility”.

The methodology establishes the work procedure and requirements in justification of nuclear safety of cask spent fuel loading with taking into account burn-up credit:

- Procedure for spent fuel assembly burn-up calculation. Procedure for calculation of spent fuel assembly conservative burn-up profile.

- Selection and determination of concentration of radionuclides that are the main contributors to Keff.

- Nuclear safety justification of cask loading with credit of spent fuel isotopic composition.

- Requirements on mathematical model of cask. Procedure of Keff calculation for fuel loading of cask.

- Requirements on documentation and finalisation of calculation results.

- The work procedure and requirements in monitoring of spent fuel burn-up at the stage of cask loading.

As known, burn-up is not uniformly distributed along fuel assembly height. To implement a conservative approach to nuclear safety analysis, in particular calculation of multiplication factor for cask loadings, two methods are applied to consider non-uniform fuel assembly burn-up by height:

- burn-up is accepted unchanged by height for each spent fuel assembly and equal to average burn-up between lower and upper (least burn-up) assembly parts;

- for spent fuel assembly of the same enrichment, conservative burn-up profile is formed so that burn-up of each part is equal to the least burn-up of associated assembly parts.

In the framework of this methodology, changes in concentration of only fuel isotopes:

$$^{235}U,\ ^{238}U,\ ^{239}Pu,\ ^{240}Pu,\ ^{241}Pu$$

are taken into account in VVER-1000 spent fuel depending on its burn-up, Fig.1.

In burn-up of 50 MW day/kgU, difference in the multiplication factor for FA infinite lattice with initial enrichment 4.4% calculated with account of only fuel isotopes and all isotopes (but without Xe) constitutes 14%. This value actually determines the additional safety margin which we incorporate in our justification in connection with possible errors in determining the concentration of U or Pu isotopes.

At the final stage of the nuclear safety assessment with the burn-up credit the “loading curve” has been calculated. This curve demonstrates the maximum admissible fuel enrichment and its minimum burn-up to satisfy the condition $K_{eff}<0.95$ under the optimum moderation, Fig.2. The area above the curve is nuclear safety and below the curve – nuclear dangerous.

Based on research results, the following main conclusion can be made: implementation (through supplementing or updating standards and rules in force) of approaches actively applied by many advanced countries in nuclear safety analysis of PWR spent fuel management systems into this analysis of VVER spent fuel management systems will allow elimination of most contradictions between regulatory requirements and actual state of affairs.

Fig.1. Multiplication purposes of irradiation VVER-1000 fuel

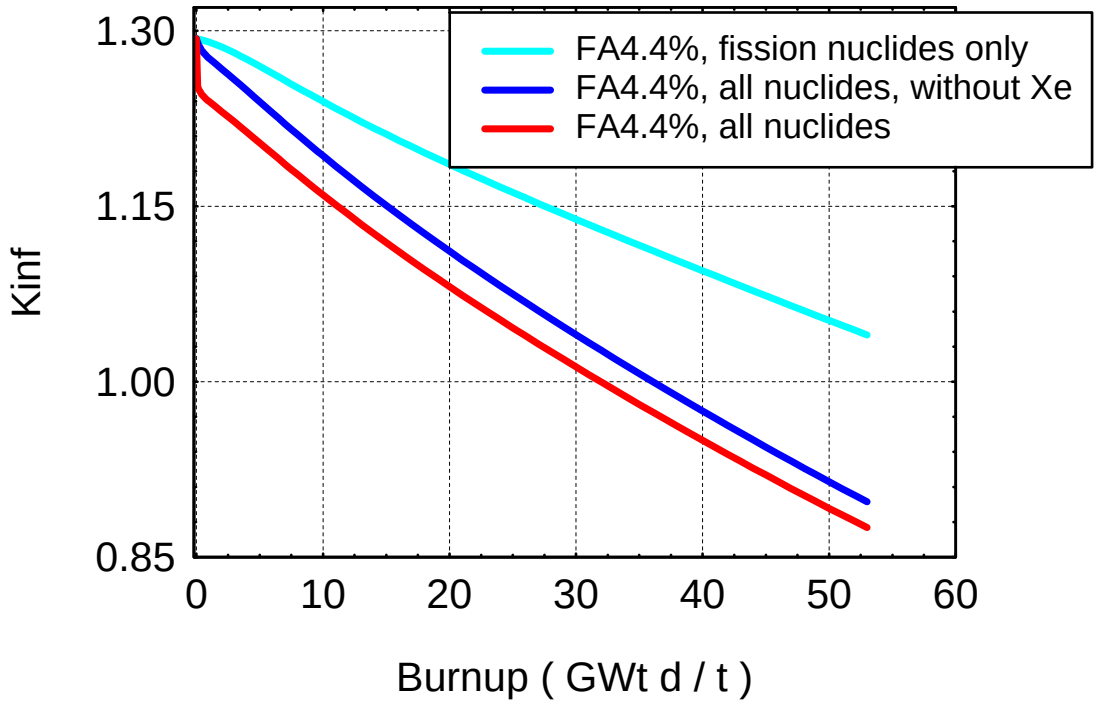
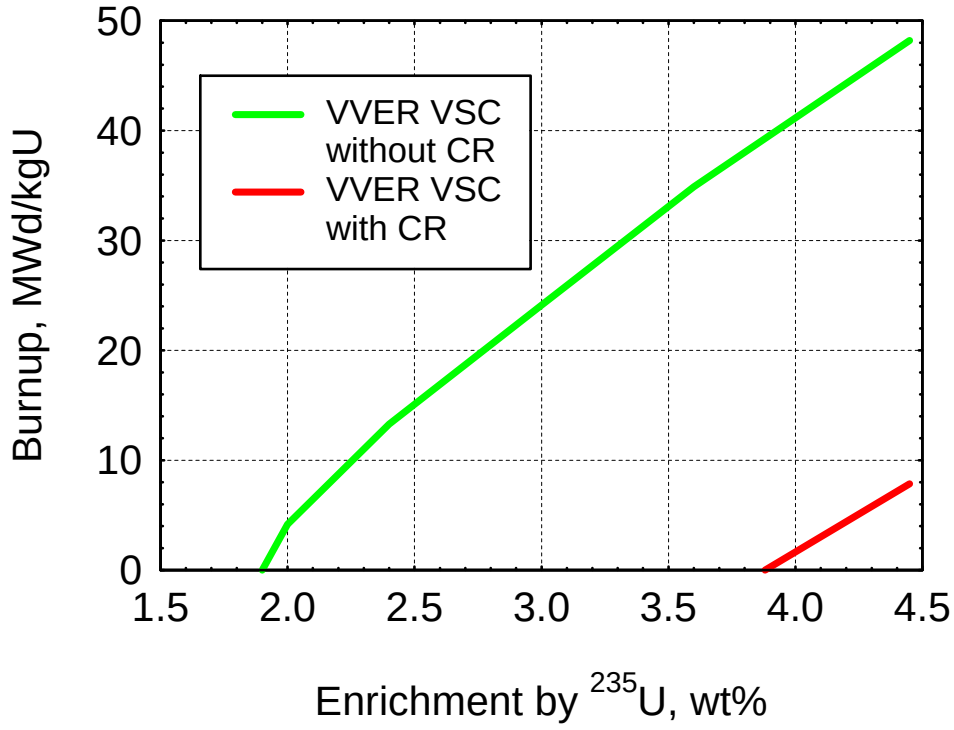


Fig.2. Loading Curve



Conclusion

This result demonstrated that the excessive conservatism laid previously into the requirements on nuclear safety in Ukraine due to insufficient development of means for modelling processes in nuclear fuel can be considerably decreased through using more real modelling fuel systems.

If such modelling is performed at the state-of-the-art level, based on more complete understanding the processes in fuel systems, then removal of the excessive conservatism will not reduce the safety of nuclear dangerous systems.

STATUS OF BURN-UP CREDIT IN THE UNITED STATES

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Abstract

Oak Ridge National Laboratory is leading an effort to expand burn-up credit for spent fuel storage and transportation casks to enable more cost-effective and higher density storage and transport while maintaining an adequate safety margin. A key objective of this programme is to develop the technical basis to credit the negative reactivity effects of both the actinide and major fission product compositions in irradiated fuel. To support this activity, new experimental data are being acquired and evaluated to validate the cross sections and the prediction of the isotopic compositions of spent nuclear fuel. This paper presents the status of burn-up credit activities in the United States and summarises these current research activities.

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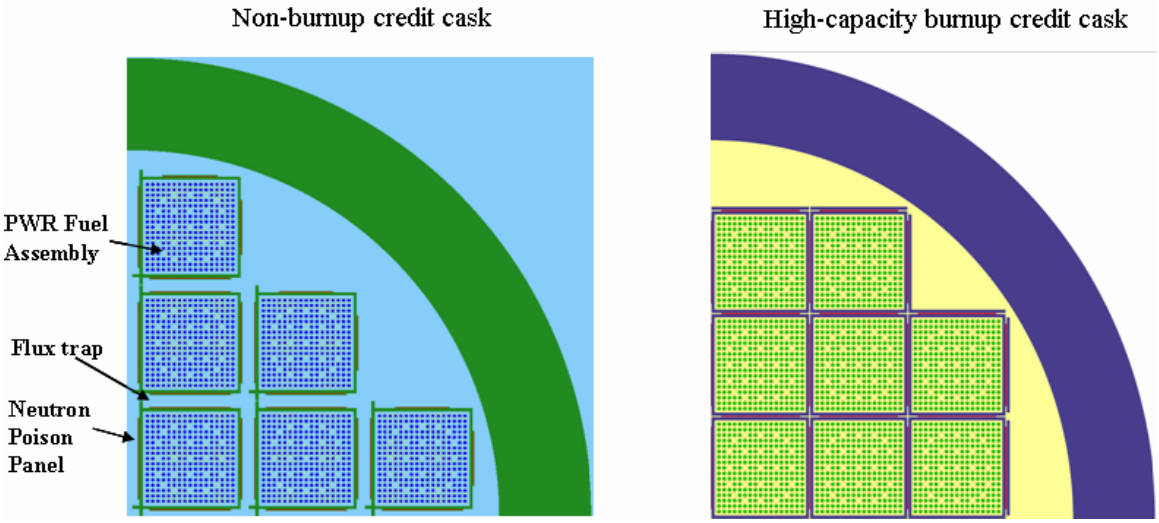
Introduction

The Nuclear Regulatory Commission (NRC) in the United States approved the principle of burn-up credit in a licensing application with the issuance of Interim Staff Guidance 8 (ISG-8) in 1999 containing recommendations on the use of actinide-only burn-up credit in storage and transportation of pressurised-water reactor (PWR) spent fuel. In September 2002, the NRC issued ISG-8 Revision 2 providing expanded guidance on the application of burn-up credit in criticality safety analyses [1]. An important requirement addressed by the guidance is the need to establish an experimental database to support burn-up credit and the general approach to take for establishing the bias and uncertainty in the analysis codes. The revised ISG-8 guidance recommends an actinide-only burn-up credit approach because of insufficient experiment data available to validate fission products.

A goal of burn-up credit in the United States is to enable more cost-effective, higher density storage and transport of spent nuclear fuel (Fig. 1) while maintaining an adequate safety margin. Several studies have demonstrated that actinide-only burn-up credit will allow loading and transportation of only a small fraction of the existing spent fuel inventory in high-capacity transportation casks in the United States [2]. Given the appropriate data for validation recommended in ISG-8, the most significant opportunity identified for increasing accuracy and expanding burn-up credit is to include negative reactivity credit for fission products. Oak Ridge National Laboratory (ORNL) is presently leading an effort with the Department of Energy, NRC, and the Electric Power Research Institute (EPRI) to acquire and evaluate the experimental data (mainly isotopic assay measurements and critical experiments) necessary to extend burn-up credit to include fission products.

This paper describes previous activities leading up to the release of ISG-8 and discusses the status of current burn-up credit activities for transportation and storage that focus on acquiring experimental data for the fission products.

Fig. 1. Illustration showing fuel assembly configurations for a traditional transportation cask and high-capacity cask with burn-up credit



Regulations

There are separate and distinct NRC licensing regulations that govern spent fuel storage, transportation and disposal. These include:

- pool storage at reactors (10 CFR 50);
- dry cask transport and storage (10 CFR 71 &72),;
- disposal (10 CFR 63).

Although the technical issues can be very much the same, the regulatory approach and concerns are often different. The areas of responsibility also fall under different groups organisationally with the NRC.

Regulatory guidance on burn-up credit

NRC Interim Staff Guidance 8, Rev. 2 (ISG-8), was issued in September 2002. The efforts leading up to the issuance of ISG-8 focused on establishing a solid technical basis of understanding of the physics and burn-up credit phenomenon for PWR fuels, and to establish burn-up credit to the maximum extent possible given the experimental data available. It is recognised, domestically and internationally, that the availability of experimental data for code validation is a limiting factor for implementation and expanded use of burn-up credit [2,3].

The revised ISG-8 guidance includes the following main components:

- criteria to determine whether spent nuclear fuel is eligible for burn-up credit determination;
- significant relaxation of the limits and restrictions on eligible assemblies from Rev. 1, but still recommends actinide-only;
- guidance on code validation: bias and uncertainty for subcritical margin established using measured data for spent fuel compositions and cross sections;
- modeling assumptions to consider in safety assessment: data may be needed for best-estimate model development;
- guidance for loading operations;
- request for estimate of additional (but unvalidated) reactivity margins.

An independent review of the guidance by the Electric Power Research Institute (EPRI) concluded that *"ISG-8, Revision 2 can be viewed as providing as much burn-up credit flexibility as can be currently expected (UO_2 fuel irradiated in PWRs only, with no credit for fission products) based on the extent and range of the available data" (Source: EPRI 1002879 [3])*.

The technical basis for ISG-8 Rev. 2 was developed by ORNL during several years of research and technical studies. The supporting activities and reports completed prior to the issuance of the revised ISG guidance included:

- a baseline report to review and prioritising technical issues (NUREG/CR-6665 [4]);
- facilitating an "expert group" PIRT meetings on burn-up credit (NUREG/CR-6764 [5]);
- developed and analysed a computational benchmark for estimating margins due to fission products (NUREG/CR-6747 [6]);
- evaluated separate effects on burn-up credit analysis due to:
  - control rods (NUREG/CR-6759 [7]);
  - integral burnable absorbers (NUREG/CR-6760 [8]);
  - burnable poison rods (NUREG/CR-6761 [9]);
  - cooling time (NUREG/CR-6781 [10]);
- performed assessment of axial burn-up effect and data (NUREG/CR-6801 [11]);
- performed assessment of validation data and uncertainty approaches (NUREG/CR-6811 [12]);
- developed automated software tools for burn-up credit analysis (NUREG-CR-6748 [13]);
- performed assessment of reactivity margins and loading curves (NUREG/CR-6800 [14]);
- comparison of US and French approaches to burn-up credit (NUREG/CR-6702 [15]).

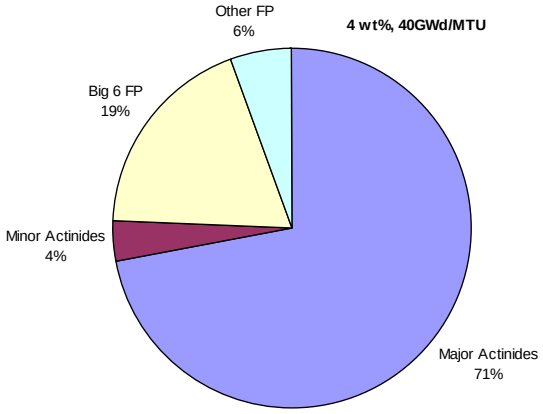
Limitations in burn-up credit

Several studies performed recently have demonstrated that actinide-only burn-up credit will allow loading and transportation of only a small fraction of the existing spent fuel inventory in high-capacity transportation casks in the United States [2]. Additional negative reactivity (beyond only the actinides) is required to accommodate the majority of US spent fuel in transportation casks in a cost-effective manner. Given the appropriate data for validation recommended in ISG-8, the most significant opportunity identified for increasing accuracy and expanding burn-up credit is to include the negative reactivity credit for fission products.

The primary impediment to actinide and fission product (full) burn-up credit is the lack of experimental data for demonstrating the available subcritical margin. The data needs extend to validation of the reactivity effects associated with fission product absorption (i.e., microscopic cross section validation) and the ability to accurately predict the fission product concentrations in spent fuel.

Figure 2 illustrates the relative contribution of the different nuclide groups to burn-up credit in a high-capacity transportation cask. It can be seen that six major fission products, ^{143}Nd, ^{149}Sm, ^{103}Rh, ^{151}Sm, ^{133}Cs, and ^{155}Gd, account for roughly 19% of the total reactivity effect. All other fission products combined account for an additional 6% of the total effect. The potential benefit from crediting fission product absorption in a burn-up credit analysis is illustrated in Fig. 3. The loading curve shows which assemblies have an enrichment and burn-up that are acceptable for cask loading (assemblies above the curve are acceptable). The loading curve is dramatically shifted when fission products are included. The change in the number of assemblies acceptable for loading depends on the specific assembly design. Based on the inventory of spent fuel in the U.S. (discharged through 1998), the percentage of assemblies acceptable for loading increases from only about 30% with actinides only (ISG-8) to roughly 90% when fission products are included.

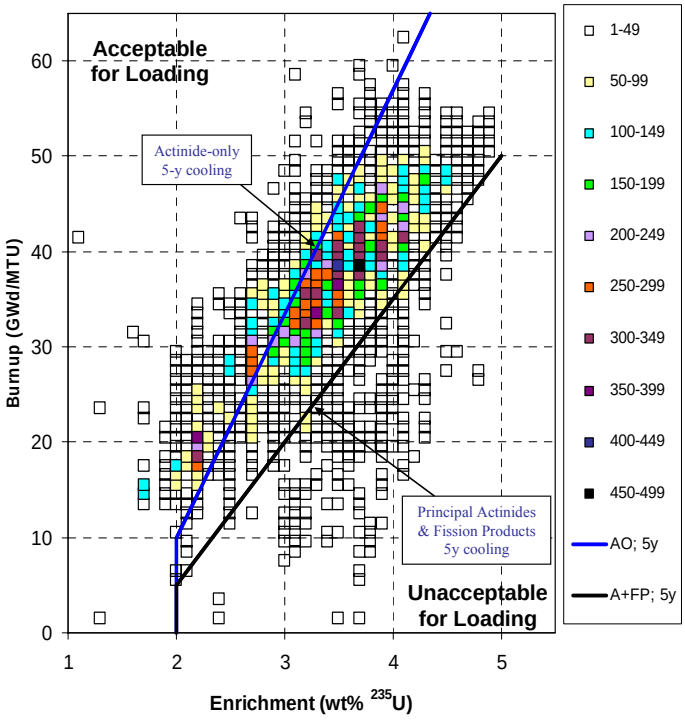
Fig. 2. Relative isotopic contributions to burn-up credit



Planned work programme

ORNL is leading an effort with the Department of Energy Office of Civilian Radioactive Waste Management (DOE/RW), NRC, and EPRI to expand the experimental database for validation by acquiring and evaluating new measurement data necessary to extend burn-up credit to include fission products. The goal is to solidify cask vendor ability to provide a technical basis for predicted subcritical margin and uncertainty using burn-up credit. Improved and expanded experimental data are needed specifically related to spent fuel isotopic assay measurements, more relevant critical experiments, and improved fission product cross sections.

Fig. 3. Illustrative loading curves for generic 32 assembly cask for actual US PWR discharged fuel through 1998



The approach being developed to acquire the necessary data is through international collaborations, data acquisition, and domestic programmes guided by improved tools for data assessment. The experiments currently being evaluated include:

- critical experiments with key fission products to be designed and performed at Sandia National Laboratory (^{143}Nd, ^{149}Sm, ^{103}Rh, ^{151}Sm, ^{133}Cs, ^{155}Gd);
- assessment and enhancement of high-quality isotopic assay data:
  - integrate with other NRC and DOE programmes assay data needs;
  - pursue evaluated existing sources of data;
- assessment and enhancement (as needed) of fission product cross-sections:
  - goal is to ensure rigor of key fission product cross sections is comparable to those of the actinides;
- procurement of relevant French experimental data (COGEMA):
  - enhance technical basis of actinide-only burn-up credit and add additional data to support fission product credit;
- identify and develop data to support BWR burn-up credit.

Summary

It has been demonstrated that the actinide-only approach to burn-up credit as described in the NRC ISG-8 guidance will not permit loading of the majority of US spent fuel in high-capacity casks. The experimental data and supporting technical basis for existing burn-up credit to include fission product presently is not publicly available. The goal of the current co-ordinated research programme involving ORNL, DOE/RW, NRC, and EPRI is to develop/obtain the experimental data needed to support extending burn-up credit to include fission products. It has been demonstrated that the potential savings in terms of reduced risk and reduced transportation cost associated with using extended burn-up credit will far outweigh the costs of the research programme [3].

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VALIDATION AND SESITIVITIES FOR BURN-UP CREDIT DEPLETION ANALYSES

Chairs: K. Suyama, M. Manolova

VALIDATION OF LATTICE ANALYSIS METHOD THROUGH PIE DATA OF HIGH BURN-UP BWR UO_2 FUEL

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Introduction

Post-irradiation experiments (PIE) of nuclear fuel provides us with valuable data to be used for validation of burn-up calculation methodologies for reactor core design and application of burn-up credit in spent fuel management. The calculational analysis has been conducted with a public deterministic code to reproduce the isotopic compositions of the burnt fuel pellets and the fission products distributions among the fuel rods in the BWR 8×8 UO_2 assemblies for the verification of the lattice analysis methods [1].

As a verification programme of fuel integrity, post-irradiation experiments has been progressing for the high burn-up BWR 9×9 UO_2 lead use fuel assemblies which have been burnt to almost licensed burn-up limit, 55 GWd/t (assembly average burn-up) [2]. The PIE includes radiochemical analysis of isotopic compositions of U, Pu, Am, Cm and Nd isotopes and burn-up evaluations based on those isotopic data for several samples. Those measurement data have been compared with the results of burn-up calculations with modelling of whole BWR fuel assemblies using a deterministic analysis code and a Monte Carlo analysis code.

Present paper shows the outline of the experimental data and the preliminary analysis results about these BWR 9×9 UO_2 fuel assemblies.

PIE data of BWR high burn-up UO_2 fuel

2.1 BWR 9 × 9 UO_2 lead use fuel assembly

Two different type BWR 9×9 UO_2 lead use fuel assemblies were inserted to the unit 1 of Fukushima power station 2 (2F1) as a part of reload fuel assemblies in December 1996. After three cycle irradiation, two assemblies (2F1Z3 and 2F1ZN2) were discharged in May 2000 for PIE, and after five cycle irradiation, two other assemblies (2F1Z2 and 2F1ZN3) were discharged in Jan. 2005 for PIE. Isotopic inventory measurements with radiochemical analysis were applied to the samples taken from 2F1ZN2, 2F1Z2 and 2F1ZN3. Table 1 shows the some core parameters of 2F1 and Table 2 the major specifications of the fuel assemblies, 2F1ZN2 and 2F1ZN3, for which the preliminary

burn-up calculations have been applied and the calculated results are presented in this paper. Fig. 1 shows the enrichment splitting in the assembly and the positions of the fuel rods for that isotopic inventory were measured.

Table 1 Core parameters of Unit 1 of Fukushima Power Station 2 (2F1)

Core Thermal Power	3 293 MW
Core Flow	48.3×10^3 t/h*
Core Pressure	6.93 MPa [gage] (70.7 kg/cm^2 g) *
Coolant Inlet Temperature	286 ^{o}C*
Core:	
Number of Fuel Assemblies	764
Core Effective Height	3.71 m*
Core Equivalent Radius	4.75 m*

*: Round number

Table 2 Major specifications of fuel assemblies (2F1ZN2 and 2F1ZN3)

Fuel Assembly	
Lattice	9 × 9
Fuel Rod pitch	1.45 cm*
Fuel Rod Gap	0.35 cm*
Number of Fuel rods	72
Assembly Av. Enrichment	3.4 wt.%*
Fuel Rod	
Outer Diameter	11.0 mm*
Cladding Thickness	0.70 mm*
Cladding Material	Zry-2
Pellet Diameter	0.94 cm*
Pellet-Cladding Gap	0.20 mm*
Pellet Density	97%TD*
Pellet Material	UO_2, UO_2-Gd_2O_3
Pellet Enrichment	2.1 to 4.9 wt.%*
Gd_2O_3 Density	5.0 wt.%*
Water Channel	Square
Dimension	38.5 mm*
Number	1

*: Round number

2.2 Isotopic inventory measurement

Radiochemical analysis was applied to the seven samples shown in Table 3. The isotopes of the analysis are $^{234\text{-}236,238}$U, $^{238\text{-}242}$Pu, $^{142\text{-}146,148,150}$Nd and ^{241}Am and ^{244}Cm. The typical measurement errors are less than 0.3% (one sigma) except ^{241}Am and ^{244}Cm which may have a larger error because they were measured by alpha spectrometry. The burn-up of the sampled were also evaluated by the ^{148}Nd method [3] with using these isotopic data. A part of the results of the isotopic inventory measurements are shown in Figs. 2(a), 3(a) and 5(a).

Table 3 Samples of isotopic inventory measurements

Assembly	Irrad. Cycles	Fuel Type	Rod Position	Axial Position*	^{148}Nd Burn-up (GWd/t)	Sample ID
2F1ZN2	3	UO2	c3	6/24	47.4	Cy3 UO2 c3 6/24
		Gd2O3-UO2	c2	5/24	35.6	Cy3 Gd2O3 c2 5/24
2F1ZN3	5	UO2	c3	6/24	68.0	Cy5 UO2 c3 6/24
				19/24	58.7	Cy5 Gd2O3 c2 19/24
		Gd2O3-UO2	c2	6/24	54.4	Cy5 UO2 c3 6/24
				19/24	46.1	Cy5 Gd2O3 c2 19/24
		UO2	a9	6/24	61.2	Cy5 UO2 a9 6/24
				19/24	56.1	Cy5 UO2 a9 19/24

*From bottom in total 24 divisions of the effective fuel height

Burn-up analysis of isotopic inventory

3.1 Analysis methods

3.1.1 Deterministic method

Burn-up calculations with a deterministic method has been performed using the Pij collision probability module coupled with burn up calculation in the SRAC code system. [4,5] The cross-section library is the 107 energy group library installed in SRAC that was created from the nuclear data library, JENDL-3.3. [6] The resonance library was produced by the PEACO module where hyperfine energy group calculation is done to treat resonance shielding in detail. The geometrical configuration is X-Y two-dimensional model for the relevant axial node of the 9×9 assembly and the one-fourth region of the section of the assembly considering the symmetry of the nuclear design with reflective boundary condition. A water channel was simulated by nine water rods with adjusting the number densities of the water and the material. One material region was assigned to each UO_2 pellet and ten material regions with a same volume to each Gd_2O_3-UO_2 fuel pellet. The burn up step is 0.25 GWd/t for 0 to 15 GWd/t and 1 GWd/t for larger than 15 GWd/t during the irradiation and one cooling step between the reactor shut down and the measurement date.

3.1.2 Monte Carlo burn-up method

Burn-up calculation with the Monte Carlo method has been conducted using MVP-BURN [7,8] as a reference detail analysis. The geometrical configuration is same as the SRAC code except applying an one-eighth region of the section. The burn-up step is 1 GWd/t for 0 to 15 GWd/t and 5 GWd/t for larger than 15 GWd/t during irradiation with 400 000 neutron histories for each burn-up step, and one cooling step. The predictor corrector method was applied to the calculation.

3.1.3 Irradiation history

Both burn-up calculations were carried out tracking almost exactly the irradiation history such as nodal powers and in-channel void fractions that the relevant nodes of the two assemblies had experienced in the reactor core. Those history was provided by the utility operating 2F1.

3.2 Analysis results

3.2.1 Burn-up

The calculated burn-up values are shown in Table 3 compared with the evaluated values by using the ^{148}Nd method [3]. In Table 3, the sample ID is corresponding to the one in Table 2. The calculated burn up for the three cycle irradiation samples in the lower axial position show underestimation of 2 to 4%. The underestimation for the lower position extends up to 8% for the five cycle irradiation samples. On the other hand, the calculation overestimates by 2 to 7% for the higher position for the five cycle irradiation samples. This discrepancy would be mainly caused by the uncertainty in the average burn up of the relevant node given by the reactor process computing system that was used as the input in the tracking calculation in SRAC and MVP-BURN. The calculated burn up values of SRAC and MVP-BURN agree each other almost within 1%.

Table 3 Calculated burn-up with SRAC and MVP-BURN

Sample ID	Burn up (GWd/t)		
	^{148}Nd Method (a)	SRAC(b) ((b)/(a))	MVP-BURN(c) ((c)/(a))
Cy3 UO2 c3 6/24	47.4	46.5(0.980)	46.1(0.972)
Cy3 Gd2O3 c2 5/24	35.6	34.3(0.963)	34.4(0.969)
Cy5 UO2 c3 6/24	68.0	66.8(0.982)	66.2(0.973)
Cy5 Gd2O3 c2 19/24	58.7	62.5(1.064)	62.1(1.057)
Cy5 UO2 c3 6/24	54.4	52.3(0.960)	52.6(0.966)
Cy5 Gd2O3 c2 19/24	46.1	49.0(1.062)	49.4(1.071)
Cy5 UO2 a9 6/24	61.2	56.2(0.918)	57.2(0.936)
Cy5 UO2 a9 19/24	56.1	57.2(1.019)	57.6(1.027)

3.2.1 Isotopic inventory

The measured isotopic inventory of the Cy3 UO_2 c3 6/24 sample is shown in Figs. 2(a) and 3(a) comparing with the calculated results of SRAC and MVP-BURN. Figs. 2(b) and 3(b) show C/E for those results. These figures show that the calculated inventory is larger than the measured by 6 to 7% for ^{235}U and smaller by 1 to 1.5% for the total plutonium inventory. This is mainly because the under estimation of the exposures in the burn up calculations. Fig. 4(a) shows the trend of C/E values of uranium isotopes depending on the calculated burn up of SRAC in Cy3 UO_2 c3 6/24 sample. Fig. 4(b) also shows that for the plutonium isotopes.

As an example of the five cycle irradiation samples, Fig. 5(a) and Fig 5(b) show the measured isotopic inventory of the upper position, Cy5 Gd_2O_3 c2 19/24 sample, comparing with calculated results of MVP-BURN and C/E for those results. These figures show that the calculation underestimate the inventory of ^{235}U by 7% and overestimate the inventory of plutonium by 2%, which is mainly because the over estimation of the exposure in the burn up calculation. This trend is almost same for the calculation of SRAC.

Discussion

It would be needless to say that the burn up of the sample is key value in analysing the isotopic inventory. In this work, the burn up of the sample is one of the calculated results through the tracking

analysis following the nodal data of the plant process computer. As the result, the calculation gives a small discrepancy in the burn up values from the measurement so that a significant deviation between the calculation and the measurements in C/E was seen in ^{235}U and a small deviation in plutonium isotopes.

As a future work, the burn up calculation will be adjusted to make the calculated burn up equal to the one evaluated by the ^{148}Nd method. In applying the ^{148}Nd method, the effect of neutron capture of ^{148}Nd and ^{147}Nd to the inventory of ^{148}Nd [9,10] will be evaluated as a correction to the current burn up evaluation in this study.

The burn-up calculation method with Monte Carlo transport calculation such as MVP, MCNP, etc. would give a reference result of the isotopic inventory that could be used to validate the model and evaluate the uncertainty of the deterministic method in place of the experimental data. For that use, the precise assessment of the uncertainty in the Monte Carlo burn-up calculation method would be necessary. For such purpose, the PIE data should be extended to cover the major parameters such as the neuron energy spectrum of reactor cores and the burn up; however, the set of the experiments would be saved compared with the necessary ones for the direct comparison with the deterministic codes.

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[10] Idem, "Correction to 148Nd method of evaluation of burn-up for the PIE samples from Mihama-3 and Genkai-1 reactors", Annals of Nuclear Energy, 33[4], 335 (2006).

Fig.1 Enrichment splitting and rod position of isotopic inventory measurement

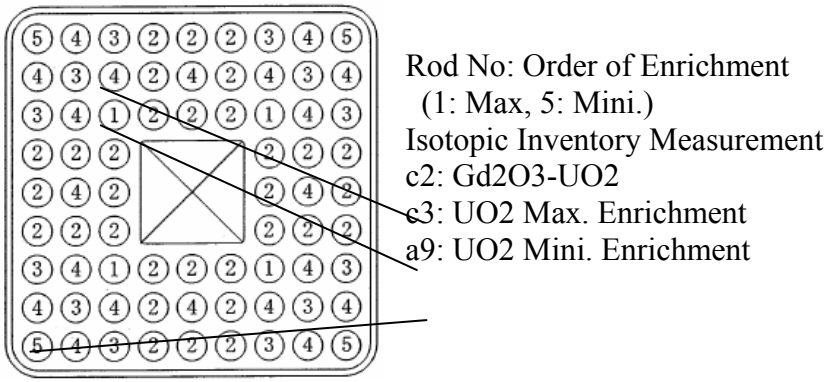


Fig. 2(a) Measured isotopic inventory compared with the calculated with SRAC for Cy3 UO_2 c3 6/24 sample

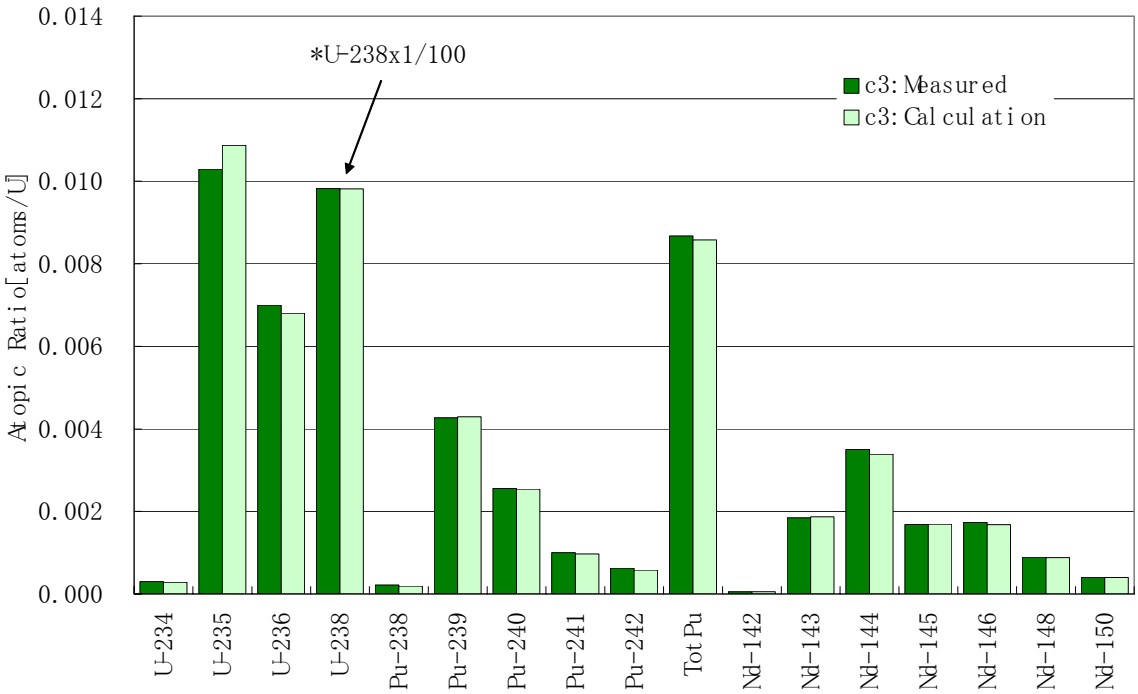


Fig. 2(b) Ratio of calculated results (C) with SRAC to the measured (E) for Cy3 UO_2 c3 6/24 sample

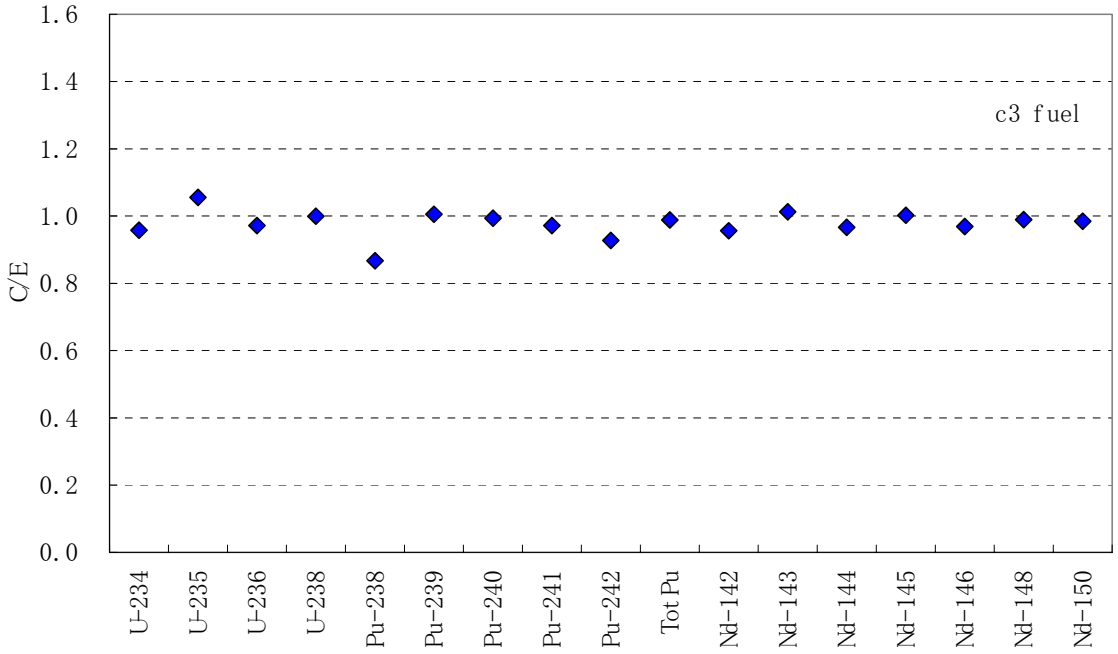


Fig. 3(a) Measured isotopic inventory compared with the calculated with MVP-BURN for Cy3 UO_2 c3 6/24 sample

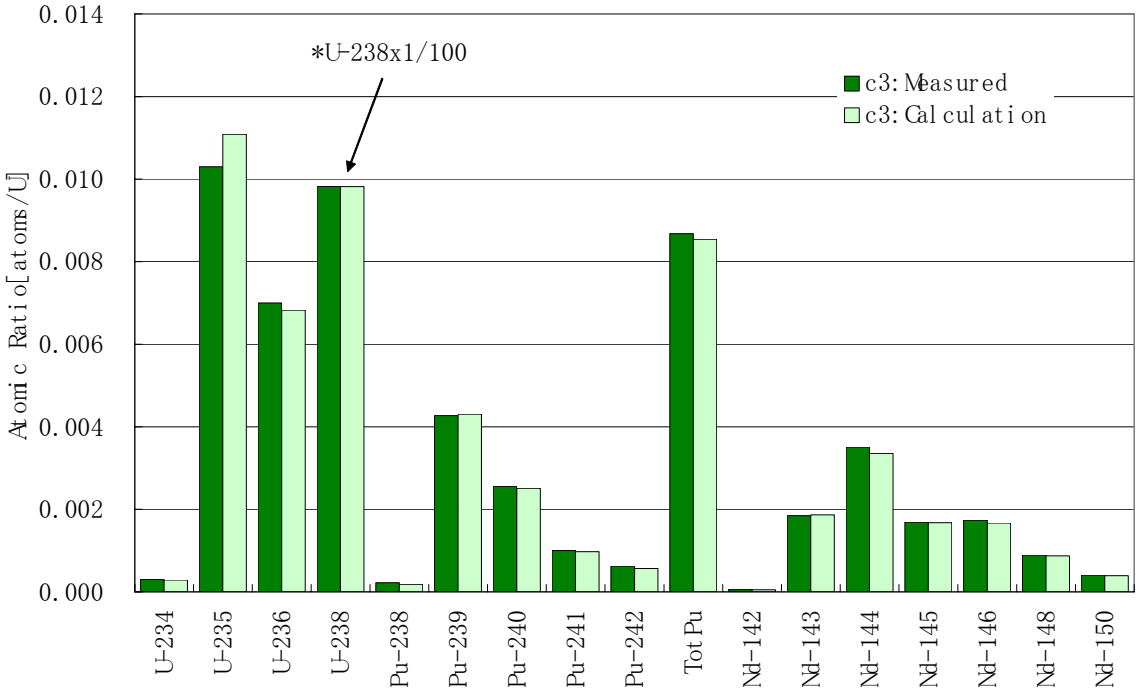


Fig. 3(b) Ratio of calculated results (C) with MVP-BURN to the measured (E) for Cy3 UO_2 c3 6/24 sample

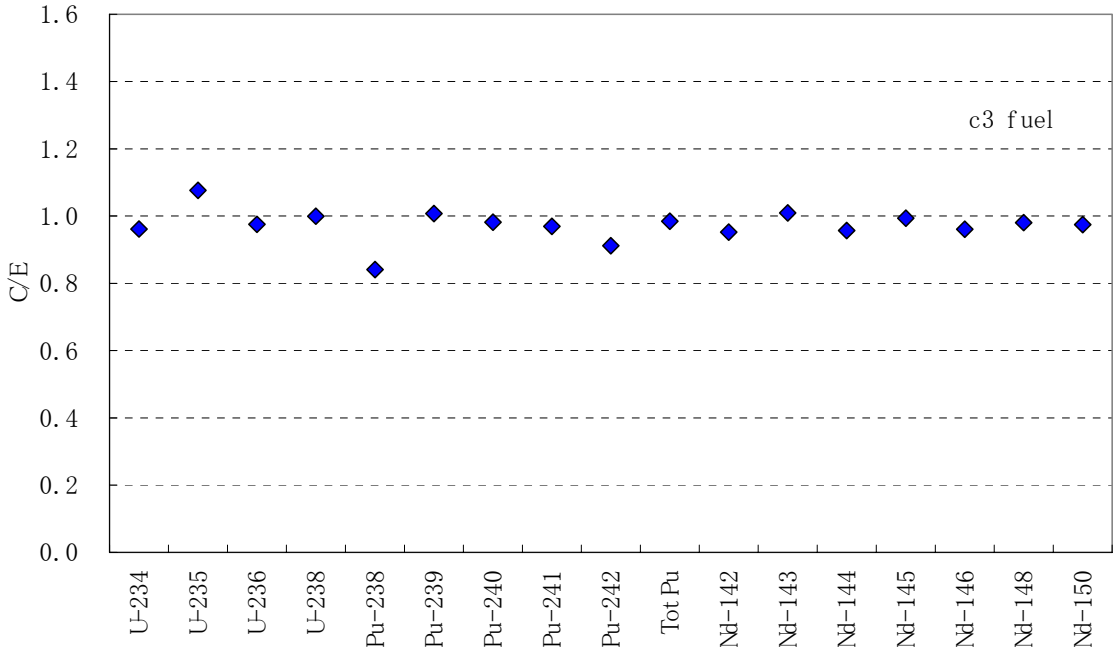


Fig. 4(a) C/E of uranium isotopes vs. calculated exposure of SRAC for Cy3 UO_2 c3 6/24 sample

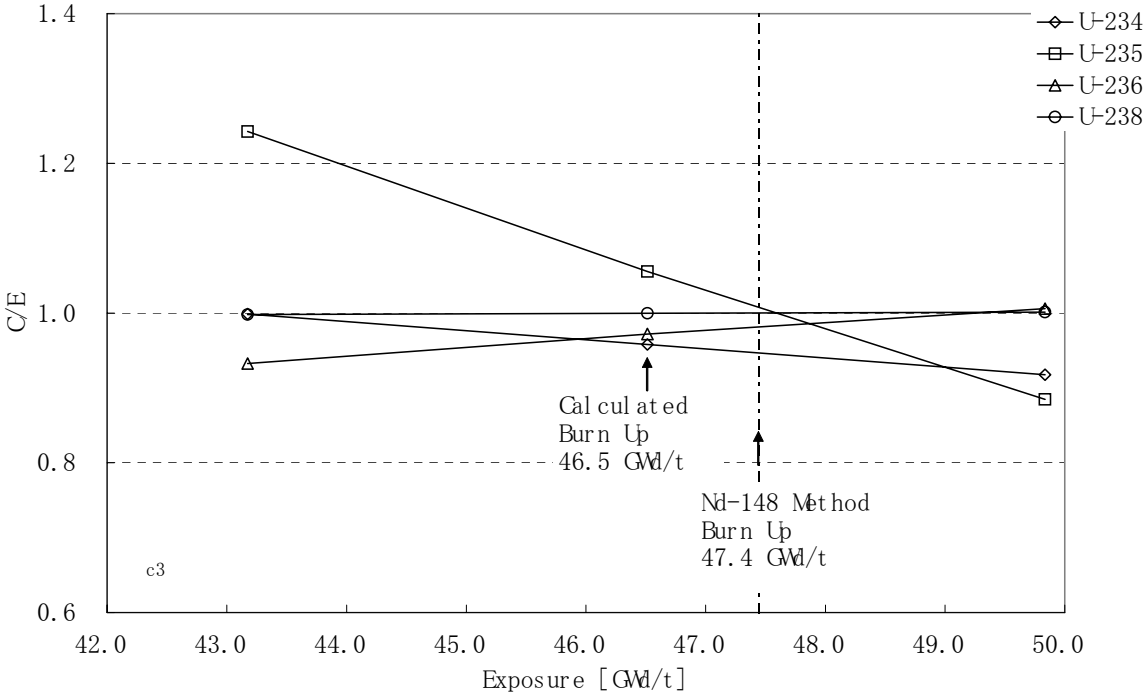


Fig. 4(b) C/E of plutonium isotopes vs. calculated exposure of SRAC for Cy3 UO_2 c3 6/24 sample

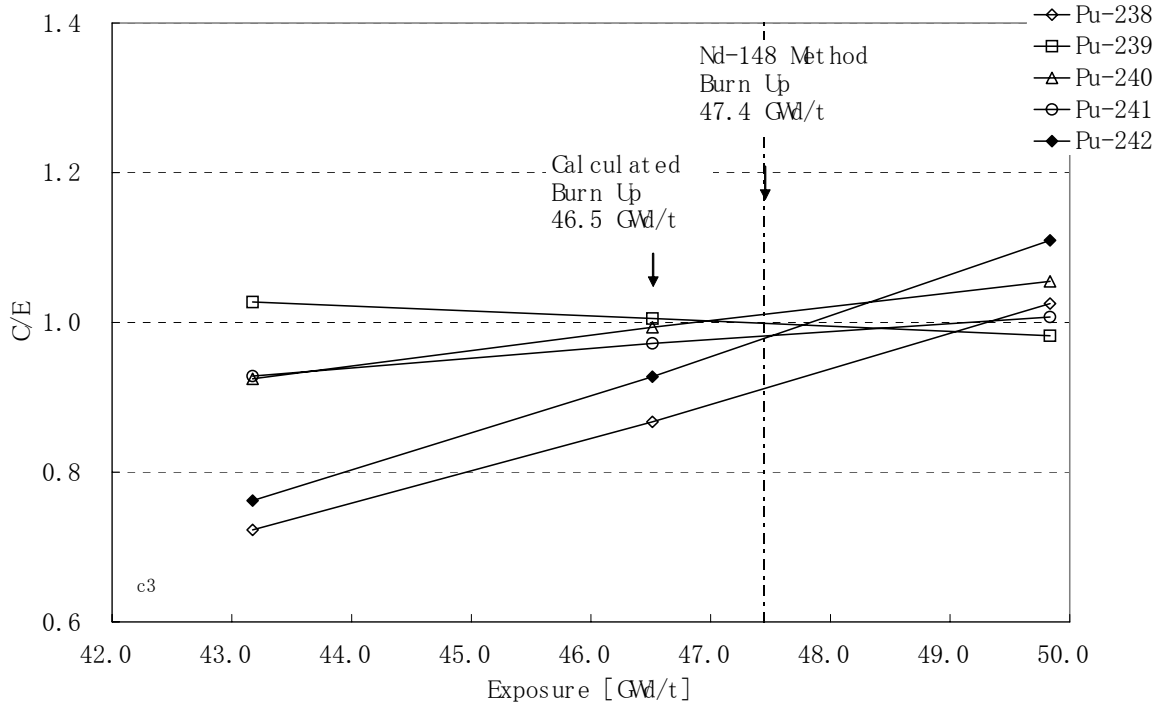


Fig. 5(a) Measured isotopic inventory compared with the calculated with MVP-BURN for Cy5 UO_2 c3 19/24 sample

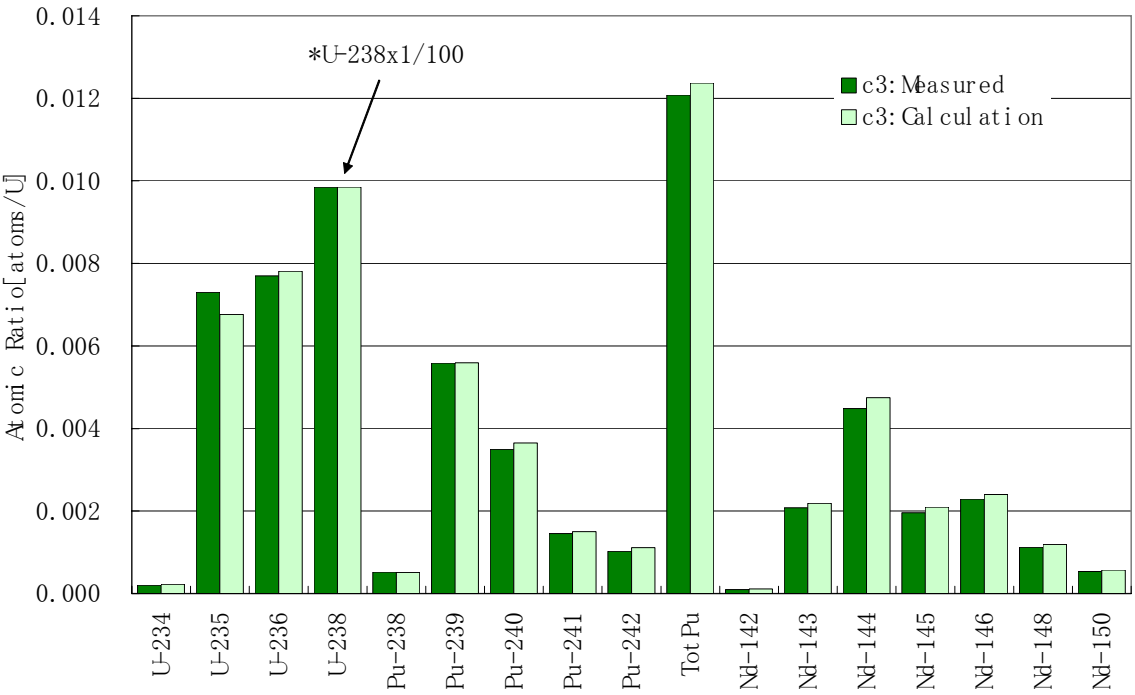
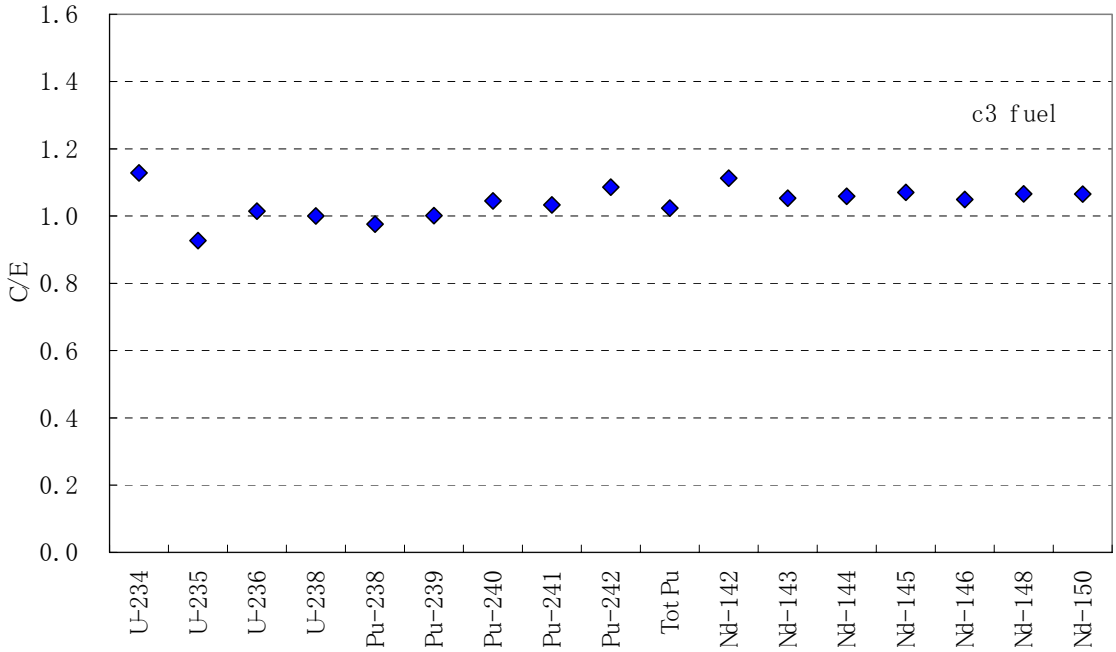


Fig. 5(b) Ratio of calculated results (C) with MVP-BURN to the measured (E) for Cy5 UO_2 c3 19/24 sample



VALIDATION OF THE 2-D/3-D DEPLETION AND REACTIVITY ANSLYSIS CODE SYSTEM KENOREST

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Abstract

For several years the 2-D/3-D code system KENOREST for reactivity and nuclide inventory calculations has been developed and validated successively at GRS. The present paper gives at the beginning a short overview on the development history of the coupled code system KENOREST, which is based on the well-known codes HAMMER for lattice calculations, the depletion code ORIGEN and the Monte-Carlo code KENO. The basic structure and main features are discussed briefly. The major part of the paper is dealing with validation and benchmark calculations using KENOREST. As an example some results from recalculation of PIE data from the Japanese Takahama-3 spent fuel samples are presented. KENOREST also offers a sequence, which includes the code KENO-VI for appropriate treatment of hexagonal lattice systems. Results from benchmark calculations for VVER-1000 fuel assemblies are presented. Similarities of VVER and western type PWR fuel characteristics are discussed by comparing neutron physics data.

Introduction

Reliable prediction of the nuclide inventory of spent fuel from light water reactors is important for different tasks of reactor operation as well as safety issues related to transportation and storage of spent fuel. In burn-up credit (BUC) application the relevant nuclide inventory has to be determined as input for the criticality analysis in a licensing procedure. Therefore, the requirements on accuracy and reliability of the calculation method are high, and validation based on measured data is indispensable.

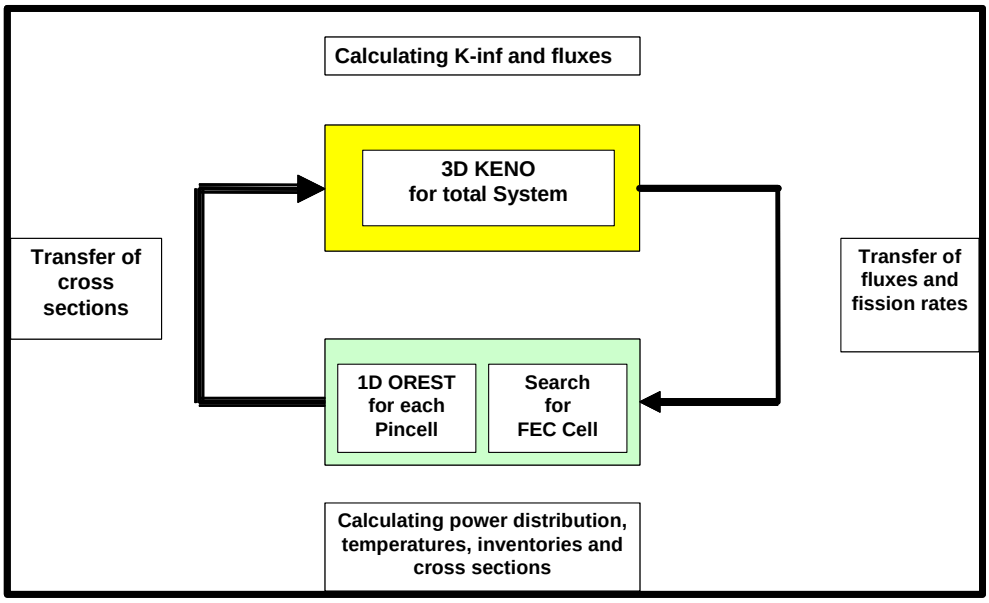
For a detailed depletion and inventory calculation the accurate modelling and representation of the operation conditions during reactor operation for the fuel assembly of interest is very important. Under this point of view the development of a depletion and burn-up analysis code system is being pursued in GRS since the 1980s. In the first stage the 1-D burn-up code system OREST [1] has been developed by coupling the infinite lattice code HAMMER (Savannah River Laboratory 1967) and the depletion code ORIGEN (Oak Ridge National Laboratory 1973). The OREST code system is preferably applicable to problems, which can be represented by a one dimensional pin cell model. However many problems in burn-up and depletion calculation are more complex and cannot be treated properly by one dimensional calculation, as there are for example fuel assemblies with burnable absorber rods, water channels, or different enrichment degrees. Therefore a 2-D/3-D burn-up code system KENOREST has been developed by coupling the OREST system with the 3d Monte Carlo code KENO-V.a (Oak Ridge National Laboratory 1995). The first version was available in 1998 [2], further improvements have been realised during the following years.

Main features of the KENOREST system

For developing the 2-D/3-D depletion and burn-up code system KENOREST the Monte Carlo code KENO-V.a has been directly linked to the OREST system in a time-step approximation. KENO is used for overall calculations of reactivity and power distribution, OREST is used for rod by rod inventory calculation. The cross section libraries of KENO and of OREST, based on JEF-2.2 coincide in the ‘infinite dilution’ starting point and in the energy group structure to avoid possible inconsistencies between different libraries. Resonance shielding and cross section calculations are treated by the Flux Equivalent Cell method (FEC) at each local position of the fuel rods. The pin cell of the one-dimensional calculation is adjusted to the local flux provided by the three-dimensional calculation. The method comprises the standard Nordheim resonance treatment and a search for effective one-dimensional cell moderator conditions. This step is performed fully automatically in a separate interface module. From a neutron-physical point of view the coupling of KENO and OREST is obtained by transferring the three-dimensional KENO results (neutron fluxes and fission densities) for each fuel rod position to the one-dimensional HAMMER code (fuel lattice simulation) and to the zero-dimensional ORIGEN depletion code by the basic relation of reaction rate balance. After a short OREST burn-up step, the cross sections and inventories for up to 1000 local positions are reloaded to KENO. The coupling is done using well defined data files during the calculation loops. A simplified diagram of program loops and of data transfers is shown in Figure 1. A detailed description of the system is given in references [3] and [4].

The actual version KENOREST 2004 includes updated versions of the lattice code HAMMER and the depletion code ORIGEN-X (GRS), and cross-section libraries based on JEF-2.2, ENDF/B-VI, JENDL3.2, and EAF97. A full update of the ORIGEN code for handling more reaction types and new cross-section libraries has been completed recently. For treating hexagonal rod lattices (e. g. VVER reactor types) the KENO-VI code has been included into the system. More information on the current state and capabilities is given in Ref. [5].

Figure 1: Simplified program flow in KENOREST



Validation of the code system

Along with improvement of the code system comprehensive testing and validation work was preformed and is still underway. GRS has access only to free available experimental data for validation, for example the SFCOMPO data base providing PIE data from Obrigheim PWR spent fuel up to 37 GWd/tHM. The latest experimental data used for validation are from the Japanese Takahama-3 (PWR) and Fukushima Daini-2 (BWR) providing maximum burn-ups of 47 and 44 GWd/tHM respectively. The full release of data from the ARIANE programme is still expected. So far chemical assay data from the ARIANE programme are available for public but without information on reactor operation during burn-up of the fuel samples, which is essential for validation calculations of burn-up codes. For VVER fuel up to now benchmark calculations but no validation based on measured inventory data have been performed. More details are given in the following.

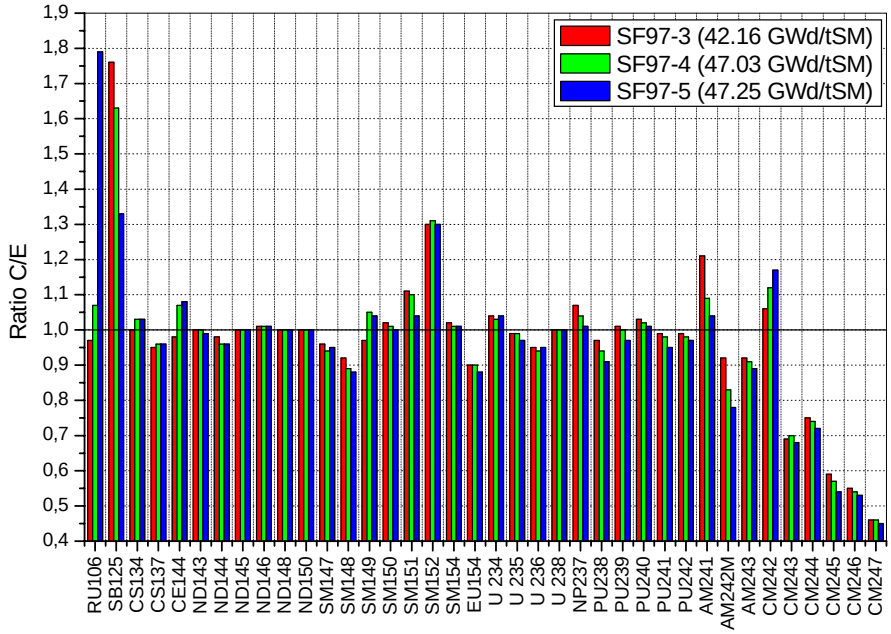
Validation with PIE data from Takahama-3 reactor

As an example for validation, some results of calculation of the PIE data from Takahama-3 reactor will be presented [6]. In 2000 the Japanese Atomic Energy Research Institute (JAERI) published extensive radiochemical analyses of spent fuel samples taken from two fuel assemblies of the pressurised water reactor Takahama-3 [7]. The fuel elements are of 17 × 17 design, with an initial enrichment of 4.11 wt.% ^{235}U and 14 fuel rods containing initially 6.0 wt.% gadolinium oxide Gd_2O_3 as burnable absorber. The burnable poison rods had a reduced initial enrichment of 2.63 wt.% ^{235}U. Furthermore the fuel assemblies include 25 water filled guide tubes. The samples under investigation received exposures up to 47 GWd/tHM, which were measured by applying the ^{148}Nd method. For every sample a detailed history of boron concentration and local power are given. The experimentally determined amounts of nuclides were calculated back to the date of reactor discharge, except the samarium nuclides which were given for 3.96 years after discharge. A detailed description of the assayed samples and the experimental setup can be found in Ref. [7] and at the NEA nuclear

database [8], respectively. These radiochemical analysis data provide in connection with the comparatively high initial enrichment and the high burn-up of the samples, these data a valuable basis for code validation.

From this work as an example the nuclide inventory calculations performed with KENOREST for the samples SF97-3, SF97-4 and SF97-5 are presented. These samples originate from a fuel assembly, which was irradiated in the reactor core for three operating cycles. The burn-up values reached by the samples were determined to 42.16, 47.03 and 47.25 GWd/tHM respectively. Figure 2 shows the ratios C/E of calculated and measured nuclide concentrations for the SF97 samples series.

Figure 2: Ratio of calculated (C) and measured (E) nuclide concentrations for the SF97 sample series.



As can be seen from this figure, the major differences between calculation and measurement are the distinct overestimation of the measured concentrations of ^{106}Ru for SF97-5, ^{125}Sb and ^{152}Sm for each sample, as well as ^{241}Am and ^{242}Cm in some cases. On the other side, ^{148}Sm, ^{154}Eu, ^{236}U, ^{242m}Am and again the curium isotopes beyond ^{242}Cm were underestimated. The main fissile nuclides as well as ^{148}Nd are met very well. As found above, most of the nuclide concentrations of this samples were met within an accuracy of ±10% or less.

It should be mentioned that sample SF97-4 has been proposed by the OECD/NEA Working Party on Scientific Issues of Reactor Systems (WPRS) as Depletion Calculation Benchmark Devoted to Fuel Cycle Issues Phase I [8], assuming average conditions concerning power operation history and boron concentration history, which are essentially different to the explicit modelling used in this work.

International benchmark calculations

As the number of experimental data on nuclide inventories after irradiation of fuel in a reactor core is very limited, inter-code comparisons of benchmark calculation results are useful as additional tool in validation a depletion calculation code. However, they cannot substitute experimental PIE data

completely. There are a number of OECD/NEA Working Parties and Expert Groups which conduct such benchmarks regularly.

In the frame of validating KENOREST, GRS participated in several international depletion calculation benchmarks. The most recent activities are participation in the OECD/NEA Working Party on Scientific Issues of Reactor Systems (WPRS) Depletion Calculation Benchmark Devoted to Fuel Cycle Issues Phase I [8] (based on Takahama-3 sample SF97-4 as mentioned above) and Phase II, a MOX fuel assembly surrounded by UO_2 assemblies. While the latter phase is still ongoing, for Phase I the distribution of a draft final report is expected soon. Both phases require the calculation of masses for more than 100 nuclides which include actinides, fission products and activation products, as well as decay heat (alpha, beta and gamma) and neutron emission, all those data for five different cooling times up to 300 years after discharge.

In Phase I, the comparison of benchmark results of KENOREST (at that time using version 03t01, a predecessor of the recent version 2004) and the other participants shows KENOREST performing very well in calculating nuclear inventories during burn-up. As an example table 1 shows the ratios C/MV of calculated mass fractions (C) to the mean values of calculated mass fractions for all participants respectively (MV), for a couple of actinides at reactor discharge.

Table 1: Comparison of results C/MV of the participants of the WPRS Benchmark Phase I

	CEA-DARW.	CEA-EAF01	BNFL	JAERI (J32)	JAERI (J33)	KAERI	KURCH.	VTT	GRS
U 232	0,93	0,89	0,68	1,31	1,31	0,69	1,06	1,09	1,06
U 233	0,55	0,78	0,32	1,62	1,80	0,44	0,72	1,71	1,06
U 234	0,99	0,99	0,99	0,99	0,99	0,99	0,99	1,04	0,99
U 235	0,99	1,01	1,05	1,05	1,05	1,00	0,86	1,00	1,00
U 236	1,00	1,00	1,00	0,98	1,02	1,04	1,02	0,98	0,98
U 238	1,00	1,00	1,00	1,00	1,00	1,00	1,00	1,00	1,00
NP236	1,79	0,50	0,94	0,63	0,72	0,61	0,58	2,49	0,75
NP237	0,99	0,97	1,02	1,02	1,05	0,90	0,97	0,97	1,10
PU236	1,07	1,02	0,59	1,29	1,34	0,36	1,23	0,86	1,23
PU238	1,00	0,96	1,08	1,00	1,04	0,88	0,92	0,96	1,13
PU239	0,97	0,97	0,99	1,06	1,06	0,91	1,06	0,96	1,02
PU240	1,03	1,03	0,99	1,03	1,03	0,95	0,95	1,03	0,99
PU241	0,99	0,99	0,99	1,11	1,11	0,92	0,86	0,99	1,05
PU242	1,01	1,00	1,00	1,02	1,04	0,94	0,91	1,04	1,05
PU243	1,05	0,99	0,94	1,05	1,05	0,89	0,89	1,10	1,05
PU244	1,11	0,53	1,09	1,24	1,22	0,69	0,50	1,58	1,03
AM241	0,99	0,95	1,05	1,16	1,09	0,97	0,81	0,91	1,07
AM242M	0,87	0,58	0,86	0,88	0,92	1,45	0,76	1,57	1,11
AM243	1,04	1,04	1,04	1,04	1,11	0,91	0,91	0,85	1,04
CM242	1,01	0,96	1,06	1,06	1,10	0,87	0,92	1,01	1,01
CM243	1,10	0,97	1,10	1,01	1,04	0,79	0,92	1,02	1,06
CM244	1,06	0,99	0,94	1,04	1,02	0,50	0,79	1,68	0,99
CM245	1,02	0,99	0,86	1,15	1,13	0,43	0,62	1,98	0,83
CM246	1,07	1,00	0,86	1,07	1,12	0,43	0,76	1,88	0,81
CM247	1,01	0,94	0,63	1,19	1,21	0,40	0,71	2,11	0,80
CM248	0,96	0,88	0,61	1,24	1,35	0,36	0,96	2,01	0,63

The numbers coloured in green assign an agreement of C/MW within 10% to unity, while yellow assigns values between 11 and 25%, and red of differences greater than 25%. It can be seen that for most nuclides KENOREST results are in agreement better than 5% to the mean value of calculation results, especially for the reactivity determining fissile and absorbing nuclides as ^{235}U, ^{239}Pu, ^{240}Pu and ^{241}Pu. Also good agreement was found for most fission products, and to a lesser extent for a number of

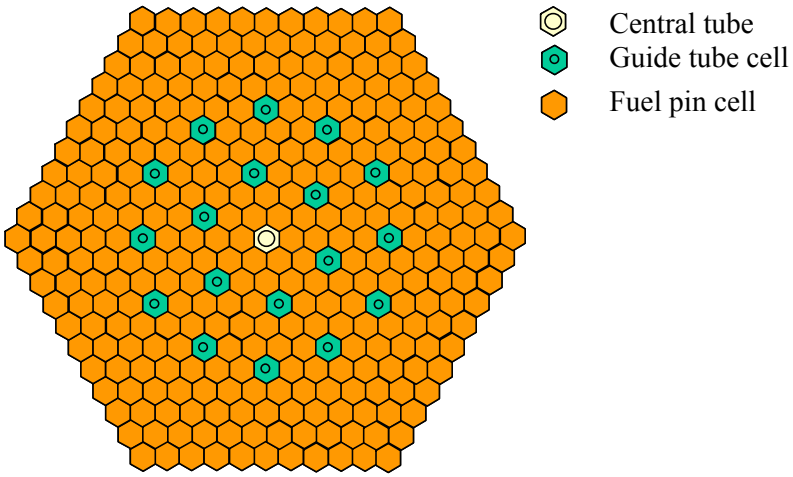
activation products. As KENOREST especially calculates the amounts of nuclides being essential in the field of burn-up credit with good accuracy, it has proven to be a useful tool in this kind of application. Regarding Phase II, no preliminary calculation results of other participants are available to GRS, so there is no comparison possible up to now.

Depletion calculations on VVER fuel

In this section, results of a depletion calculation with the hexagonal KENOREST version for a VVER-1000 fuel assembly are presented. To demonstrate the appropriateness of KENOREST-HEX for this problem, calculated results are compared with results of calculations with other codes obtained in a benchmark activity within the trilateral French-German-Russian co-operation AIDA-MOX. Then, results for the VVER-1000 fuel assembly are compared with results for a square-lattice PWR fuel assembly with similar nuclear parameters.

The depletion calculations were performed for an Uranium fuel assembly with an initial ^{235}U enrichment of 3.7%, at full power, with a boron concentration of 600 ppm, constant during the whole cycle. The layout of the fuel assembly is sketched in Fig. 3. It contains 312 fuel pins, 18 guide tubes, and a central tube. The moderator/fuel ration is 1.85 (taking into account the inter-assembly water gaps and the tube cells). The assumed fuel temperature is 1027 K, and the power is 39.2 kW/kg of heavy metal. A detailed description of the benchmark is found in [9].

Figure 3: Layout of the VVER-1000 uranium fuel assembly



In the frame of the AIDA-MOX fuel assembly benchmark, the Russian side performed calculations with four different computer codes:

- MCU-REA is a continuous energy Monte-Carlo code, which can also be applied for burn-up calculations.

- The CONKEMO code package consists of the CONSYST code for group cross-section preparation, the multi-group Monte-Carlo code KENO-VI for neutron flux calculations, and ORIGEN-S for isotope inventory calculations.

- TVS-M is a spectral code for VVER lattice and assembly burn-up calculations, using a broad-group nuclear library with 48 energy groups. Fuel assemblies are calculated with the diffusion fine-mesh method.

- WIMS-ABBN is based on the WIMS-D4 code, with a number of improvements, like an extended resonance self-shielding.

Moreover, calculations with the CASMO-4 code were contributed by SIEMENS, and calculations with APOLLO2 by CEA; both are two-dimensional cell and burn-up transport codes.

In Fig. 4, the multiplication factors as a function of the burn-up are displayed. It can be seen that the curves are in reasonable agreement, with a slight tendency of KENOREST to underestimate the results of the other codes for intermediate burn-up values. In addition, the pin power distribution calculated with KENOREST for the fresh state is compared with the corresponding distribution from a MCNP reference calculation with nuclear point data based on JEF-2.2. The relative differences for 1/6 of the fuel assembly are given in Fig. 5, showing excellent agreement between the KENOREST and MCNP results, with maximum discrepancies of ~ 2%.

Figure 4: Multiplication factor vs. burn-up for the VVER-1000 uranium fuel assembly. The KENOREST result is compared with the results of an international benchmark.

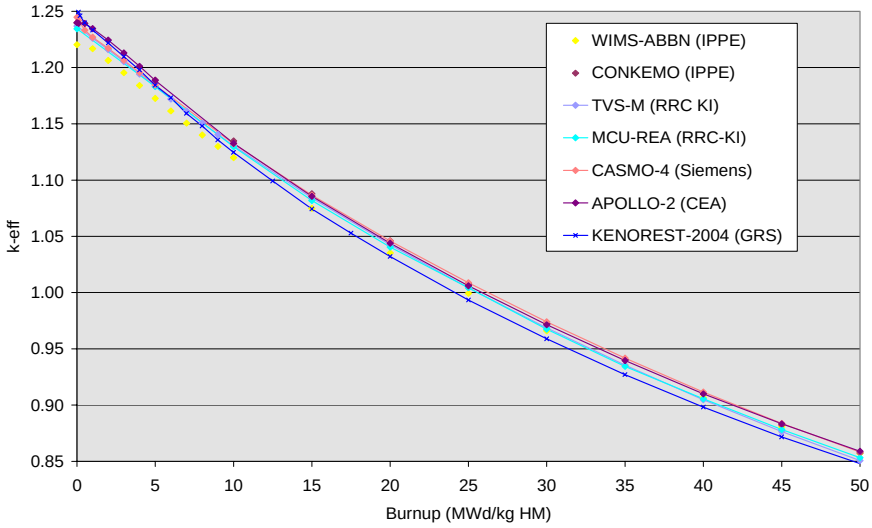
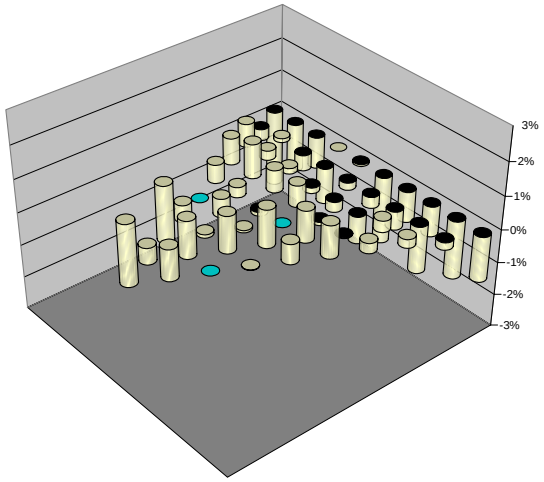


Figure 5: Pin power distribution for 1/6 of the VVER-1000 uranium fuel assembly in the fresh state. Relative differences between KENOREST and MCNP are shown (KENOREST/MCNP – 1). The centre of the fuel assembly is on the left.



To investigate the differences between a VVER-1000 fuel assembly and a corresponding square-lattice PWR fuel assembly, additional depletion calculations with KENOREST for such a PWR fuel assembly have been performed. The nuclear parameters were chosen similar to those of the VVER fuel assembly (same ^{235}U enrichment and boron concentration). The main differences are in the fuel temperatures (773 K for PWR vs. 1027 K for VVER) and the moderator/fuel ratios due to the different lattice types (1.99 for PWR vs. 1.85 for VVER).

In Figs. 6 and 7, results of the KENOREST calculations for both fuel assemblies are compared. Figure 4 shows the multiplication factors as a function of burn-up. At BOC, the PWR fuel assembly possesses a slightly higher reactivity, whereas during burn-up, its reactivity decreases somewhat faster than that of the VVER fuel assembly. The neutron spectra for BOC and at a burn-up value of 50 MWd/kg of heavy metal are displayed in Fig. 7, exhibiting a slightly harder spectrum for the VVER than for the PWR fuel assembly. These deviations in the neutronics behaviour of both fuel assembly types are consistent with the different moderation ratios and fuel temperatures.

Figure 6: Multiplication factor vs. burn-up for a VVER-1000 and a square-lattice PWR fuel assembly calculated with KENOREST.

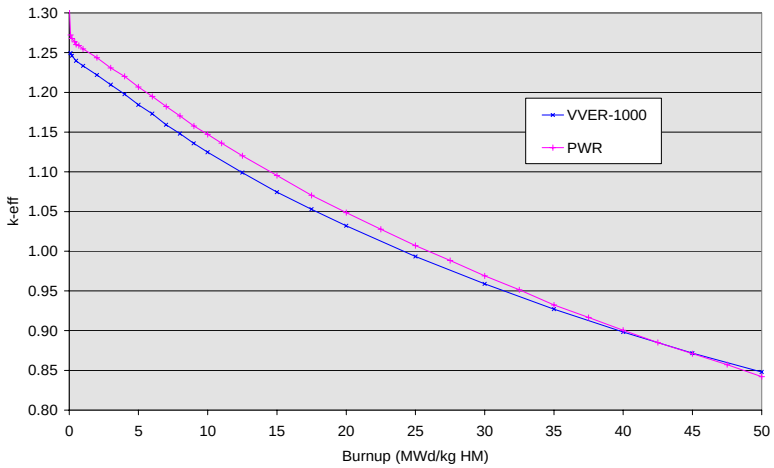
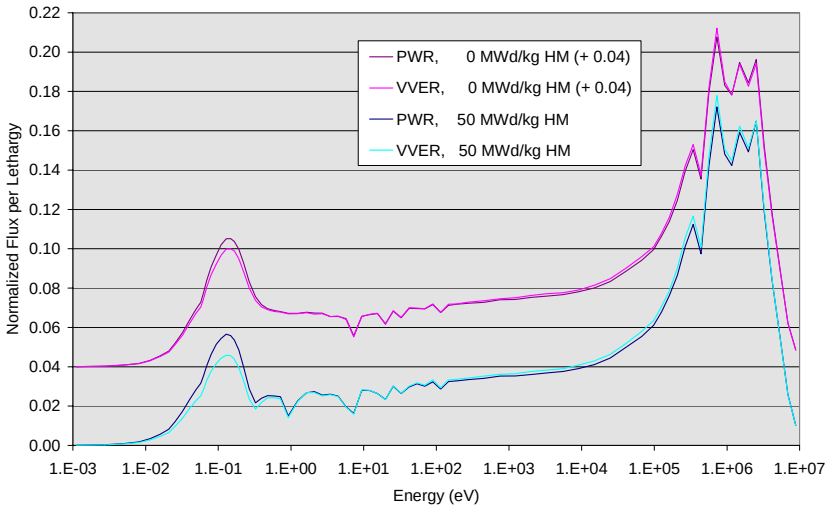


Figure 7: Neutron spectra for a VVER-1000 and a square-lattice PWR fuel assembly calculated with KENOREST. The values for the fresh state and for a depleted state are displayed. For a better separation of the curves, the values for the fresh state are shifted by 0.04.



The results show a good comparability in the neutronics properties of hexagonal VVER and square-lattice PWR fuel assemblies with only minor differences. Therefore, we conclude that the validation of depletion codes by experimental data from post irradiation experiments supports the application of these codes for both types of reactors.

Need for PIE data from VVER spent fuel samples

There are several reasons, why from our point of view there is currently a need for PIE data of VVER fuel. As shown above, VVER fuel is very similar to PWR fuel of western type with respect to neutron physics. So the VVER data can be used in a general validation data base of PWR fuel and are not limited to specific codes for VVER application.

For codes, which are used in safety analyses for licensing procedures, e.g. for BUC application, validation based on experimental data is absolutely necessary. The data base, which is freely available to the interested expert community is still limited. New PIE data from VVER fuel samples could be a contribution to broadening the free validation data base. GRS has only access to freely available data for code validation.

Additionally there are some reasons to justify a special interest in VVER PIE data. At first there is a need for testing the hexagonal lattice calculation sequence of the KENOREST system, which was developed primarily for VVER application. Secondly, an important part of GRS activities are co-operations with VVER-operating countries in the frame of bilateral and multilateral projects on reactor safety and fuel cycle issues. The computer codes used in these projects should be validated properly. Finally there are more than 5 000 spent fuel assemblies of VVER 440 type in the interim storage facility ZLN Lubmin located in the North-east of Germany. The fuel stems from the 4 reactors of the NPP Greifswald, which were finally shut down in 1990. This fuel must be disposed in a final repository in the future, where due to current plans BUC will be applied.

Summary

The KENOREST system provides a versatile depletion and inventory calculation tool, applicable to light water reactor fuel assemblies of square lattice structure as well as hexagonal lattice structure like VVER fuel.

By coupling the proven one-dimensional burn-up code system OREST with a three-dimensional Monte-Carlo code to calculate fluxes and power distributions at specified time steps, the system is capable to meet high demands in geometric modelling and simulating reactor operation history of the fuel assembly of interest. Also an update of cross section libraries and extension of reaction channels for burn-up calculations has been realised.

Validation calculations by means of available experimental data show satisfying results. Nevertheless a need for more freely available PIE data for code validation still exists. No validation based on measured data for VVER spent fuel has been done so far, but is planned for the future. Such data would also be an opportune contribution to a free access spent fuel data base like SFCOMPO. As the neutronic characteristics of western PWR and VVER fuel assemblies are very similar, VVER PIE data can be used also for code validation in general, independent of specific application to VVER. Beside this there is a special interest of GRS in VVER spent fuel data due to the residual spent fuel of the German NPP Greifswald. This fuel is currently stored for final disposal in the future.

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VALIDATION OF DEPLETION CODES AGAINST VVER-440 SPENT FUEL DATA

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Abstract

A recently published report (ISTC 2670) describes the initial composition and the radiochemical measurement results of eight samples taken from a VVER-440 assembly irradiated in Novovoronezh 4 during the years 1987-1991. The initial U-235 enrichment of the fuel rods was 3.6% and the average burn-up of the samples was about 37.5 GWd/tU. The scope of the isotopes measured was comprehensive. This paper reports the comparison of the predictions of CASMO-4E and ABURN against the Novovoronezh 4 isotopic measurements. For most of the considered isotopes the average C/E-values were close to unity but the variation between the samples was large. In some cases, very large biases and uncertainties were observed. The results of CASMO-4E and ABURN proved to be quite similar.

KEYWORDS: VVER-440, isotopic composition, CASMO-4E, ABURN

1. Introduction

The validation of depletion codes is important from the point of view of reactor safety and economics. It is also necessary if burn-up credit is to be applied in a criticality safety assessment. The amount of publicly available VVER-440 spent fuel data has been inadequate for validation purposes and limited to few reported measurements [1]. The AER community currently pursues new radiochemical measurements of spent VVER-440 assemblies.

Recently, an ISTC report "Radiochemical Assays of Irradiated VVER-440 Fuel for Use in Spent Fuel Burn-up Credit Activities" was published and it extends the database potentially useful for the validation and burn-up credit studies. The report describes the initial composition, irradiation history, and the spent fuel isotopic composition (8 samples) of a VVER-440 assembly irradiated in Novovoronezh 4 during the years 1987-1991 [2]. The average burn-up of the samples was about 37.5 GWd/tU and the measurements included a broad range of actinides and fission products.

This paper reports the comparison of the predictions of CASMO-4E and ABURN with the Novovoronezh 4 data. Cross-section libraries based on ENDF/B-VI and JEF-2.2 were applied to the calculations.

2. Specification of the assembly

Table 1 lists the dimensions of the assembly. The ISTC report did not include the specification of the pin pitch, so the value shown in Table 1 corresponds to a typical pitch used in VVER-440 assemblies. Figure 1 shows the initial enrichments and the positions of the sampled rods. The material compositions of the assembly applied in the calculations are shown in Table 2. All fuel rods had the same initial enrichment.

Table 1. Fuel assembly dimensions (p. 7 and 10, Reference [2])

Pellet hole diameter	1.2
Pellet diameter	7.6
Cladding inner diameter	7.72
Cladding outer diameter	9.1
Central tube inner diameter	8.8
Central tube outer diameter	10.3
Pin pitch	12.2
Inner distance between box walls	140.2
Thickness of the box wall	2.0
Thickness of the half water gap	1.15

Note: All lengths given in mm.

Figure 1. Fuel rod enrichments and the numbering of the sampled rods (Note: Fuel rod numbering differs from that presented in Reference [2])

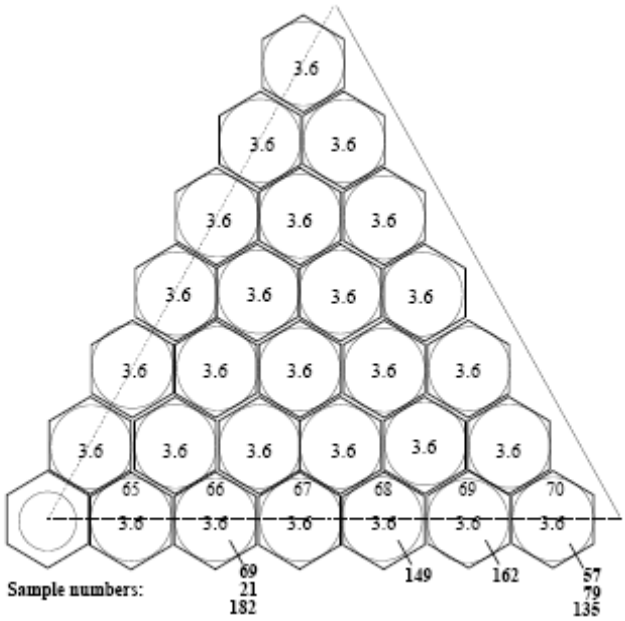

Table 2. Fuel assembly material compositions (p. 5, 8, and 10, Reference [2])

Component	Composition (weight percent)	Density (g/cm^3)
Fuel	3.6 % (U-235/U)	10.0[1]
Cladding	Zr-1%Nb alloy	6.55
Central tube	Zr-1%Nb alloy	6.55
Assembly box	Zr-1%Nb alloy	6.55
Spacer	Zr-1%Nb alloy	6.55

1) Calculated average density of fuel rod.

3. Specification of the irradiation history

The assembly was irradiated in the reactor during four cycles in 1987-1991. According to the assembly movement diagram described in the ISTC report, the assembly was not located next to control group assemblies during any of the irradiation cycles. Depletion calculations were made with the cycle-average operating history based on the data in the ISTC report. Table 3 lists the irradiation cycle data [2].

Table 3. Irradiation cycle data for assembly 135

Cycle	15(1)	16(2)	17(3)	18(4)
Effective days	296	389.8	98.2	325.1
Average burnup EOC	11.0	25.5	29.0	38.5
Power (W/gU)	37	37	36	29
Boron[1] (ppm)	394	522	150	487

1) Calculated with the data in Reference [2].

4. Radiochemical assay data

The radiochemical assay data covered measurements from 8 specimens. The specimen names and the corresponding fuel rod location numbers, sample heights, and burn-ups are listed in Table 4.

Table 4. Sample numbering and burn-ups

Fuel rod	Specimen	Height (mm)	BU (kg FP/tU)	BU (GWd/tU)[1]
66	182	100	22.86	22.77
	21	1000	41.50	42.61
	69	2150	31.32	31.95
68	149	1000	41.90	43.41
69	162	1000	44.20	44.93
70	135	100	29.90	30.37
	79	1000	46.30	47.36
	57	2150	36.20	36.11

1) Estimated by matching Nd-148 concentrations.

5. Codes and data libraries

CASMO-4/4E is a two-dimensional transport theory code for burn-up calculations of PWR and BWR assemblies. CASMO-4E is an extended version of the code for which JEF-2.2 and ENDF/B-VI based libraries and the hexagonal module for the calculation of VVER bundles are available. The CASMO-4E version applied was v2.10.12 with its libraries based on ENDF/B-VI (e60201), and JEF-2.2 (j20200).

ABURN is a Monte Carlo burn-up script developed at VTT, which combines the MCNP4C Monte Carlo code and the ORIGEN2 isotopic depletion code. The reaction rates are calculated with MCNP4C for each depleted zone and the predictor-corrector method is applied in the depletion calculation performed with ORIGEN2. The number of isotopes for which the reaction rates are estimated with MCNP4C equals 88. The cross-sections for other isotopes are taken from the ORIGEN2 cross section tables. The cross-section library applied in the calculations was based on the ENDF/B-VI.8 data.

6. Results and discussion

The results of the comparison of the predicted isotopic compositions with the measured ones is collected in Tables 5-10. On average, the calculated and measured actinide concentrations agree quite well. An exception is Np-237 for which a large underprediction is seen. The variation in the C/E values with different samples is large leading to significant standard deviations. Many of the important fission products are well predicted. However the predicted concentrations of Ag-109, Cs-134, Sm-147, Sm-151, and Eu-153 are far from the measured values. The standard deviation of Sm-149, which is one of the most important fission products from the BUC point of view, is large.

The difference in the predictions of CASMO-4E and ABURN can be seen in Tables 5-8. Tables 5 and 7 show that there was no large difference in the prediction of the actinide concentrations. Only in the case of Am-243 and Cm isotopes, the differences were significant. The predictions of the FP concentrations were similar with the exception of some Sm isotopes (Tables 6 and 8). The calculations

with the JEF-2.2 library (Tables 9 and 10) revealed only less than few percent differences compared to ENDF/B-VI. Only the Am and Cm concentrations were clearly sensitive to the cross-section library used.

Figure 2 shows the comparison of the calculated (CASMO-4E, ENDF/B-VI) and the experimental actinide concentrations for sample 79. A very large overprediction of U-235 concentration by nearly 40% is observed, which indicates a possible error in the measurements or the reported concentrations. This sample was excluded from the calculation of the bias and the uncertainty.

Figure 3 illustrates the difference of the predicted U-235 concentration between CASMO-4E and ABURN. The differences are shown for the rods in the assembly diagonal. The difference is seen to be largest for the corner rod.

Tables 11 and 12 show the results of the sensitivity calculations for the fuel density and the fuel rod pitch, respectively. The density of fuel has a significant impact on the predicted actinide concentrations and should therefore be selected carefully. The effect of the variation in the pin pitch is seen to be a minor one.

The study presented in this paper aimed at testing the usability of the VVER-440 spent fuel data described in the ISTC report for burn-up credit purposes and the validation of CASMO-4E and ABURN against experimental data. The results indicate that the data can be useful for code validation but at the same time some very large deviations between calculated and experimental data were observed that are difficult assign to the code or the data library applied:

- The variation in the C/E of the U-235 concentrations was large. Both codes CASMO-4E and ABURN gave similar results.
- Np-237, Ag-109, Cs-134, Sm-147, Sm-151, and Eu isotopes had unusually large biases.
- The large difference between the calculated and the measured U-234 concentration may be due to uncertainties in the initial composition.

Finally, the uncertainties estimated from the comparison of the calculated and experimental data were observed to be larger than previously seen for PWR and BWR lattices. Comparison against other codes and experimental data are needed in order to validate the calculational methods reliably.

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Table 5. C/E-values of the actinide concentrations (CASMO-4E with library ENDF/B-VI)

	Sample numbers							Statistics	
	69	21	182	149	162	57	135	ave	std
U-234	1.00	0.75	1.51	0.74	0.73	1.56	0.76	1.01	0.373
U-235	1.05	1.04	1.05	1.07	1.10	1.01	0.92	1.03	0.058
U-236	0.91	0.93	0.86	0.91	0.91	0.93	0.94	0.91	0.026
U-238	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	0.001
Np-237	0.40	0.40	0.35	0.39	0.39	0.40	0.44	0.40	0.026
Pu-238	0.93	1.09	0.73	1.09	0.92	0.93	1.08	0.97	0.131
Pu-239	1.00	1.04	0.97	1.00	0.99	0.97	1.02	1.00	0.026
Pu-240	0.92	0.97	0.90	0.91	0.89	0.93	0.99	0.93	0.038
Pu-241	1.04	1.08	0.99	1.01	0.99	1.01	1.12	1.04	0.048
Pu-242	0.91	0.96	0.85	0.88	0.85	0.93	1.09	0.92	0.084
Am-241	1.11	0.93	0.81	0.87	0.61	0.97	1.20	0.93	0.194
Am-243	1.08	1.23	0.90	1.29	1.25	1.03	1.38	1.16	0.169
Am242m	0.73	0.68	0.48	0.85	1.73	0.76	0.73	0.85	0.404
Cm-244	1.01	1.06	0.58	0.91	0.72	0.69	1.01	0.85	0.187
Cm-245	0.81	0.99	0.53	1.27	0.72	1.17	0.99	0.93	0.259
Cm-246	0.54	1.12	0.27	1.14	1.06	-	0.53	0.78	0.375

Table 6. C/E-values of the fission product concentrations (CASMO-4E with library ENDF/B-VI)

	Sample numbers							Statistics	
	69	21	182	149	162	57	135	ave	std
Mo-95	0.91	0.91	0.91	0.91	0.90	0.95	0.93	0.92	0.016
Tc-99	1.00	0.99	1.01	1.01	1.00	1.03	1.02	1.01	0.012
Ru-101	1.02	1.01	1.00	1.01	0.99	1.03	1.02	1.01	0.013
Ag-109	1.68	1.89	1.36	1.92	1.96	1.77	1.64	1.74	0.210
Cs-133	0.97	1.03	0.95	0.98	0.97	0.97	0.98	0.98	0.026
Cs-134	0.63	1.64	0.49	1.26	1.60	0.97	0.63	1.03	0.478
Cs-135	1.00	1.04	0.99	0.99	0.98	0.97	0.96	0.99	0.026
Cs-137	0.98	1.04	0.96	0.99	0.97	0.99	1.01	0.99	0.026
Sm-147	0.29	0.29	0.27	0.27	0.29	0.31	0.32	0.29	0.019
Sm-148	0.94	0.92	0.78	0.85	0.84	0.88	0.89	0.87	0.055
Sm-149	1.00	1.01	0.85	0.91	0.86	0.85	0.78	0.90	0.084
Sm-150	1.08	1.12	0.90	1.05	1.06	1.08	1.07	1.05	0.072
Sm-151	1.48	1.66	1.15	1.49	1.50	1.33	1.15	1.39	0.194
Sm-152	1.20	1.27	1.02	1.20	1.23	1.25	1.25	1.20	0.085
Sm-154	1.08	1.12	0.89	1.05	1.05	1.06	1.05	1.04	0.074
Nd-143	1.03	1.00	1.03	1.00	0.99	1.01	1.00	1.01	0.015
Nd-145	0.95	0.92	0.97	0.92	0.92	0.95	0.97	0.94	0.022
Nd-148	1.00	1.00	0.99	1.00	1.00	1.00	1.01	1.00	0.005
Eu-153	4.94	7.16	3.38	7.58	8.21	5.91	4.50	5.95	1.778

Table 7. C/E-values of the actinide concentrations (ABURN with library ENDF/B-VI.8)

	Sample numbers							Statistics	
	69	21	182	149	162	57	135	ave	std
U-234	0.99	0.73	1.50	0.72	0.71	1.54	0.75	0.99	0.373
U-235	1.04	1.04	1.04	1.07	1.10	1.02	0.93	1.03	0.053
U-236	0.90	0.91	0.85	0.90	0.90	0.92	0.93	0.90	0.024
U-238	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	0.001
Np-237	0.40	0.40	0.35	0.39	0.39	0.40	0.44	0.40	0.024
Pu-238	0.94	1.09	0.78	1.10	0.93	0.94	1.11	0.99	0.122
Pu-239	1.02	1.06	0.96	1.02	1.02	0.99	1.02	1.01	0.032
Pu-240	0.98	1.04	0.96	0.97	0.98	0.99	1.05	1.00	0.035
Pu-241	1.03	1.08	1.00	1.03	1.01	1.03	1.13	1.05	0.046
Pu-242	0.93	0.97	0.92	0.90	0.86	0.95	1.13	0.95	0.087
Am-241	1.09	0.93	0.79	0.88	0.61	0.94	1.18	0.92	0.188
Am-243	0.92	0.95	0.88	1.00	0.99	0.88	1.27	0.98	0.135
Am242m	0.72	0.67	0.47	0.87	1.75	0.75	0.74	0.85	0.414
Cm-244	1.73	1.68	1.14	1.42	1.14	1.16	1.77	1.43	0.289
Cm-245	1.48	1.67	1.16	2.13	1.25	2.29	2.08	1.72	0.450
Cm-246	0.98	1.86	0.58	1.90	1.82	-	1.03	1.36	0.567

Table 8. C/E-values of the fission product concentrations (ABURN with library ENDF/B-VI)

	Sample numbers							Statistics	
	69	21	182	149	162	57	135	ave	std
Mo-95	0.91	0.91	0.91	0.92	0.91	0.94	0.92	0.92	0.013
Tc-99	1.05	1.06	1.04	1.08	1.07	1.07	1.05	1.06	0.014
Ru-101	1.01	1.00	1.00	1.01	0.99	1.02	1.01	1.01	0.009
Ag-109	1.77	1.98	1.48	2.00	2.04	1.83	1.72	1.83	0.196
Cs-133	0.97	1.02	0.96	0.97	0.96	0.96	0.98	0.97	0.022
Cs-134	0.67	1.68	0.54	1.29	1.66	1.04	0.69	1.08	0.473
Cs-135	0.88	0.95	0.95	0.96	1.01	1.02	1.10	0.98	0.068
Cs-137	0.97	1.03	0.96	0.98	0.97	0.98	1.00	0.99	0.023
Sm-147	0.25	0.25	0.24	0.24	0.25	0.27	0.28	0.25	0.016
Sm-148	1.15	1.09	1.01	1.03	1.03	1.13	1.18	1.09	0.066
Sm-149	1.03	1.03	0.86	0.93	0.89	0.86	0.77	0.91	0.093
Sm-150	1.22	1.29	0.98	1.21	1.21	1.19	1.14	1.18	0.098
Sm-151	1.33	1.47	1.09	1.33	1.35	1.22	1.08	1.27	0.144
Sm-152	1.07	1.11	0.96	1.05	1.09	1.12	1.15	1.08	0.061
Sm-154	1.15	1.20	0.95	1.12	1.13	1.13	1.11	1.11	0.076
Nd-143	1.02	1.00	1.04	1.00	0.99	1.01	1.00	1.01	0.016
Nd-145	0.94	0.91	0.97	0.91	0.91	0.93	0.95	0.93	0.024
Nd-148	1.01	1.01	1.01	1.01	1.01	1.01	1.01	1.01	0.001

Table 9. C/E-values of the actinide concentrations (CASMO-4E with library JEF-2.2)

	Sample numbers							Statistics	
	69	21	182	149	162	57	135	ave	std
U-234	1.01	0.76	1.52	0.75	0.74	1.57	0.77	1.01	0.373
U-235	1.06	1.07	1.06	1.10	1.13	1.03	0.93	1.05	0.064
U-236	0.87	0.89	0.83	0.88	0.87	0.89	0.91	0.88	0.025
U-238	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	0.001
Np-237	0.40	0.40	0.34	0.39	0.39	0.40	0.43	0.39	0.027
Pu-238	0.90	1.07	0.71	1.07	0.90	0.91	1.05	0.95	0.132
Pu-239	1.03	1.07	0.99	1.03	1.02	1.00	1.05	1.03	0.028
Pu-240	0.94	1.00	0.92	0.94	0.91	0.95	1.01	0.95	0.038
Pu-241	1.04	1.08	0.98	1.02	0.99	1.01	1.11	1.04	0.048
Pu-242	0.91	0.96	0.85	0.88	0.85	0.93	1.09	0.92	0.083
Am-241	1.11	0.94	0.81	0.88	0.61	0.97	1.19	0.93	0.192
Am-243	0.95	1.09	0.78	1.15	1.12	0.91	1.21	1.03	0.154
Am242m	0.71	0.66	0.46	0.83	1.69	0.74	0.70	0.83	0.395
Cm-244	0.87	0.92	0.50	0.79	0.63	0.59	0.87	0.74	0.162
Cm-245	0.74	0.92	0.48	1.17	0.67	1.08	0.91	0.85	0.239
Cm-246	0.49	1.00	0.24	1.02	0.95	-	0.48	0.70	0.336

Table 10. C/E-values of the fission product concentrations (CASMO-4E with library JEF-2.2)

	Sample numbers							Statistics	
	69	21	182	149	162	57	135	ave	std
Mo-95	0.92	0.92	0.92	0.93	0.92	0.96	0.94	0.93	0.017
Tc-99	1.01	1.01	1.02	1.03	1.01	1.04	1.03	1.02	0.012
Ru-101	1.03	1.02	1.00	1.02	1.00	1.04	1.03	1.02	0.012
Ag-109	1.63	1.83	1.32	1.86	1.90	1.73	1.61	1.70	0.200
Cs-133	0.95	1.00	0.93	0.95	0.94	0.94	0.96	0.95	0.023
Cs-134	0.68	1.75	0.53	1.35	1.72	1.04	0.68	1.11	0.509
Cs-135	1.00	1.04	0.99	0.99	0.98	0.97	0.96	0.99	0.026
Cs-137	0.98	1.04	0.96	0.98	0.97	0.99	1.01	0.99	0.025
Sm-147	0.29	0.29	0.27	0.28	0.29	0.31	0.32	0.29	0.019
Sm-148	0.96	0.93	0.79	0.87	0.86	0.90	0.90	0.89	0.055
Sm-149	1.01	1.02	0.85	0.92	0.88	0.85	0.78	0.90	0.088
Sm-150	1.08	1.13	0.89	1.06	1.07	1.08	1.07	1.05	0.075
Sm-151	1.51	1.70	1.16	1.53	1.53	1.35	1.16	1.42	0.202
Sm-152	1.21	1.28	1.02	1.21	1.25	1.27	1.26	1.21	0.088
Sm-154	1.13	1.18	0.92	1.11	1.11	1.11	1.09	1.09	0.080
Nd-143	1.03	1.01	1.04	1.01	1.00	1.01	1.01	1.02	0.014
Nd-145	0.95	0.93	0.98	0.93	0.93	0.95	0.97	0.95	0.022
Nd-148	1.01	1.02	1.01	1.02	1.01	1.02	1.02	1.01	0.005
Eu-153	5.04	7.38	3.41	7.81	8.47	6.06	4.59	6.11	1.863

Figure 2. C/E-values of the actinide and the FP concentrations, sample 79

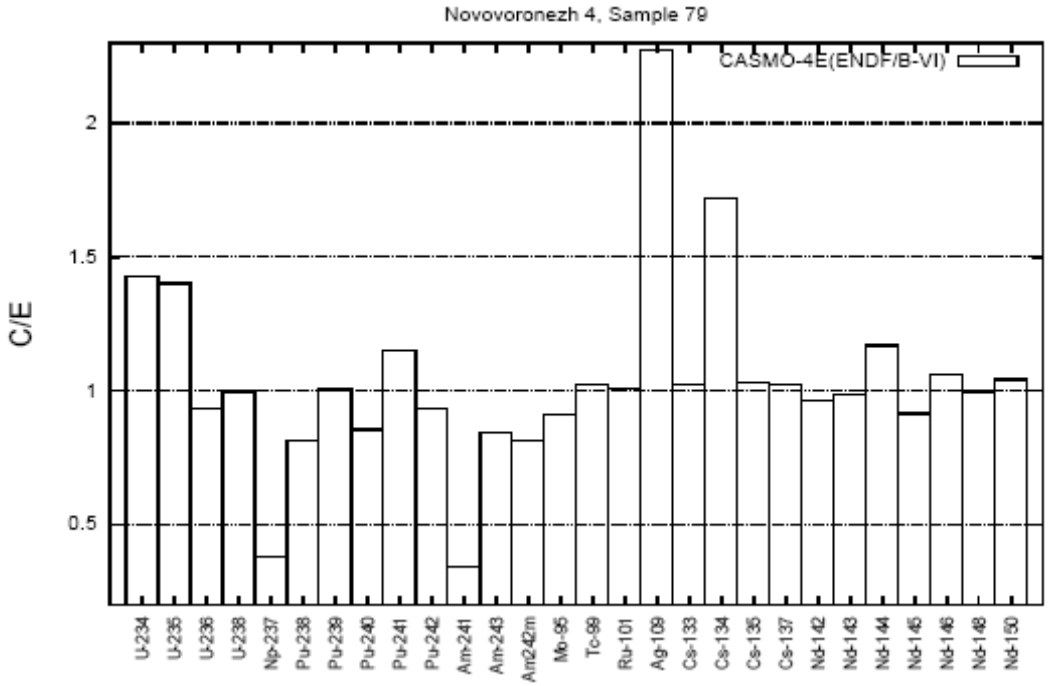
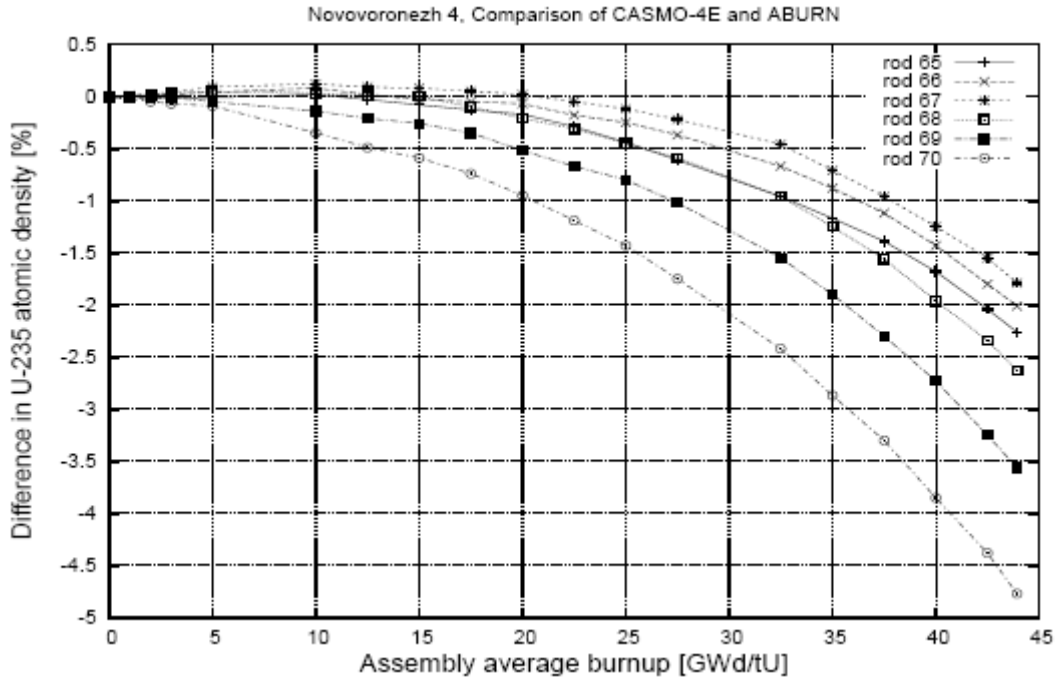


Figure 3. Fractional difference in the U-235 concentration as a function of burn-up (CASMO-4E -ABURN)



A Sensitivity of isotopic concentrations to fuel density

Table 11 lists the results of the comparison of the calculated and the experimental nuclide concentrations in the case where fuel density of 10.7 g/cm^3 was used instead of 10.0 g/cm^3 applied in the base cases. The choice of the initial density of the fuel clearly affects the calculated actinide concentrations of the irradiated fuel.

Table 11. N(Sensitivity case)/N(Base case). The ratio of the concentrations of actinides and some FPs (CASMO-4E, ENDF/B-VI).

	Sample numbers						
	69	21	182	149	162	57	135
Cs-133	1.00	0.99	1.00	0.99	1.00	0.99	0.99
Cs-134	1.03	1.02	1.04	1.02	1.02	1.03	1.03
Cs-135	1.03	1.04	1.03	1.04	1.03	1.03	1.02
Cs-137	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Sm-147	0.97	0.97	1.00	1.00	0.97	1.00	1.00
Sm-148	1.02	1.01	1.04	1.01	1.01	1.01	1.02
Sm-149	1.07	1.08	1.06	1.08	1.08	1.06	1.05
Sm-150	1.01	1.00	1.01	1.00	1.00	1.00	1.01
Sm-151	1.07	1.08	1.05	1.07	1.07	1.05	1.04
Sm-152	0.98	0.99	0.99	0.99	0.98	0.98	0.99
Sm-154	1.01	1.01	1.01	1.01	1.01	1.01	1.01
U-234	0.99	0.99	0.99	0.99	1.00	0.99	0.99
U-235	1.02	1.03	1.01	1.04	1.04	1.02	1.01
U-236	1.00	1.00	1.00	1.00	1.00	1.00	1.00
U-238	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Np-237	1.05	1.05	1.05	1.02	1.02	1.02	1.04
Pu-238	1.05	1.06	1.07	1.05	1.05	1.05	1.05
Pu-239	1.05	1.06	1.04	1.06	1.05	1.03	1.03
Pu-240	1.01	1.02	1.00	1.01	1.02	1.01	1.01
Pu-241	1.03	1.04	1.03	1.04	1.04	1.02	1.03
Pu-242	0.98	0.98	1.00	0.98	0.99	0.99	1.00
Am-241	1.04	1.04	1.03	1.05	1.05	1.03	1.03
Am-243	1.03	1.02	1.05	1.01	1.02	1.02	1.03
Am242m	1.07	1.09	1.06	1.08	1.08	1.06	1.05
Cm-244	1.07	1.04	1.10	1.05	1.05	1.05	1.07
Cm-245	1.13	1.12	1.16	1.12	1.11	1.11	1.13
Cm-246	1.10	1.06	1.14	1.07	1.06	-	1.11

B Sensitivity of isotopic concentrations to fuel rod pitch

Table 12 shows the results for the sensitivity calculation where the fuel rod pitch was increased from 1.22 cm to 1.23 cm while other dimensions remained similar with the base cases. The effect of the fuel rod pitch on the isotopic concentrations is seen to be small.

Table 12. N(Sensitivity case)/N(Base case). The ratio of the concentrations of actinides and some FPs (CASMO-4E, ENDF/B-VI).

	Sample numbers						
	69	21	182	149	162	57	135
Cs-133	1.00	1.00	1.00	1.00	1.00	0.99	0.99
Cs-134	0.98	0.99	1.00	0.99	1.00	1.01	1.00
Cs-135	0.99	0.99	1.00	1.00	1.00	1.02	1.01
Cs-137	1.00	1.00	1.00	1.00	1.00	1.00	0.99
Sm-147	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Sm-148	1.00	0.99	1.00	1.00	1.00	1.00	1.01
Sm-149	0.98	0.98	0.99	1.00	1.02	1.02	1.04
Sm-150	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Sm-151	0.99	0.99	0.99	1.00	1.01	1.02	1.02
Sm-152	1.00	1.01	1.01	1.00	0.99	0.99	1.00
Sm-154	1.00	1.00	1.00	1.00	1.01	1.01	1.00
U-234	1.00	1.00	1.00	1.00	1.00	0.99	1.00
U-235	0.99	0.99	0.99	1.00	1.01	1.01	1.01
U-236	1.00	1.00	1.00	0.99	0.99	1.00	1.00
U-238	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Np-237	1.00	1.00	0.97	1.00	1.02	1.00	1.02
Pu-238	0.98	0.99	0.99	1.00	1.01	1.02	1.02
Pu-239	0.99	0.99	0.99	1.01	1.01	1.01	1.01
Pu-240	1.00	0.99	0.99	0.99	1.00	1.00	1.00
Pu-241	0.99	0.99	1.00	1.01	1.01	1.01	1.02
Pu-242	1.00	1.00	1.00	1.00	1.01	0.99	1.00
Am-241	0.99	0.99	0.99	1.00	1.02	1.02	1.02
Am-243	1.00	1.00	1.00	1.00	1.01	1.01	1.01
Am242m	0.99	0.99	1.00	1.01	1.03	1.04	1.04
Cm-244	0.99	0.99	0.99	1.00	1.01	1.01	1.03
Cm-245	0.98	0.97	0.97	1.00	1.01	1.04	1.04
Cm-246	0.98	0.98	1.00	0.99	1.01	-	1.03

SCALE 5 ISOTOPIC PREDICTION FOR VVER-440 SPENT FUEL

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Abstract

In this article are compared theoretical results by new version of the SCALE 5 code with experiments or other theoretical calculation for:

- nuclide compositions:
  - measurement in Kurchatov institute for 3.6%;
  - measurement in Dimitrovgrad for 3.6% (project ISTC 2670);
  - measurement in JAERI (PWR 17 × 17);
  - numerical benchmark No. 2 (CB2).

The focus is on modules TRITON and ORIGEN-S.

Introduction

In Russian reactor VVER-440 are used hexagonal assemblies with triangular lattice pitch. In western PWR are used assemblies with square geometry. The change from square geometry to triangular is sometimes big problem. It is necessary to verify code used by calculations of spent fuel (VVER-440) storage. The best way is to compare theoretical results with experiments. If experiments do not exist with other calculations. In this paper are results of the SCALE 5 code (distributed in 2004) [1] for calculations of nuclide compositions. The computing system SCALE 5 includes several modules: KENO V.a, KENO VI, SAS2, SAS4, ORIGEN-S, TRITON etc. The 44GROUPNDF5 library was used.

Nuclide compositions

In ORIGEN-S calculations were used libraries prepared by the module SAS2. Basic library was used library 44GROUPNDF5 and 1 library per cycle. Time step by irradiation was maximal 100 days. The new 2-D modules (NEWT and TRITON) for inventory calculations were tested. By TRITON calculation was used 1 step per cycle.

The nuclide composition measurement (only some actinides) of spent fuel assembly VVER-440 was made in Kurchatov Institute in Moscow [4-7]. 12 samples (11 actinides) are from one assembly 3.6% from NPP Novovoronezh Unit 4 (In text Novovoronezh 1). In Fig.1 are measured pins shown. New nuclide composition measurement for burn-up credit application for VVER-440 reactor was made in RIAR in Dimitrovgrad (ISTC project) [8]. 8 samples (16 actinides and 32 fission products) are from one assembly 3.6% from NPP Novovoronezh Unit 4 (In text Novovoronezh 2). In Fig.2 are measured pins shown. We used module ORIGEN-S and library prepared for fuel VVER-440 (by module SAS2) and the new 2D module TRITON (April 2005 release). The results are in Tab.1 and Tab.2 and in Fig.3 and Fig.4 (maximal, minimal and average deviation) shown. For U and Pu agreement is good, with deviations of less then 15% (except Pu238), for Am and Cm may be large differences – up to 60%. The deviations according TRITON are smaller (except U235 and Am243) then deviation according ORIGEN-S. In the second experiment are results for actinides similar to the first experiment. Very high deviation is in one sample for U235 and in one sample for Am242m. The initial concentration of U234 is not known. For fission products are small deviation (less than 10%) for Nd143, Nd146, Nd148, Nd150, Cs133, Cs137, Ce140, Ce142, Sm150 and Sm154. Very high deviation (more than 50%) are for Sm147, Sm151, Eu151, Ag109 and Gd155. Extreme high deviation (more than 150%) are for Eu153, Eu154 and Eu155.

In Attachment 1 are dependence of actinides concentration (measured and calculated) on burn-up for both experiments (Novovoronezh 1 and 2) shown. Abbreviation in figures:

E = experimental
O = ORIGEN-S calculation
T = TRITON calculation
1 = fuel pin N^{o}. 25 (NV1) or N^{o}. 65 (NV2)
2 = fuel pin N^{o}. 63 (NV1) or N^{o}. 67 and 68 (NV2)
3 = fuel pin N^{o}. 64 (NV1) or N^{o}. 69 (NV2)
4 = fuel pin N^{o}. 107 (NV1)

Because some nuclides have very high deviation, the comparison was done for one PWR assembly. The measured data are from SFCOMPO data bank operated by OECD/NEA [9]. It was used 5 samples (SF97-2 up to 6, 19 actinides and 20 fission products) from TAKAHAMA-3 (PWR,

17 × 17). In ORIGEN-S calculation was used ARP library "17 × 17" and operational history with small time step. Additional data are in [10]. The results are in Tab.3 and Fig.5 shown. For U and Pu is agreement good – deviations are less then 10% (except Pu238), for Am and Cm may be large differences – up to 50 %. Generally results from TRITON are not better then from ORIGEN-S. For Cm246 and Cm247 (very low concentration) are very high difference between ORIGEN-S and TRITON. For all isotopes of Nd are deviation less than 5%, for other fission products are deviation up to 20%, except Sb125 (up to 80%), and Sm151 and Sm152 (up to 35%).

Calculational Burn-up Credit Benchmark No.2 was defined and evaluated in [11,3] (only 11 actinides and 15 fission products important for burn-up credit calculations). The library was prepared by the module SAS2, nuclide composition was calculated by the module ORIGEN-S. The results are in Tab.4 and in Fig.6 shown. Deviations are less then 10% (except Am243, Sm151 and Gd155). Big differences between concentration according SAS2 and TRITON are for U235 and isotopes of Pu.

For nuclides compared in all cases (except fission products, not exist in measurement Novovoronezh 1, 10 actinides and 7 fission products) the maximal deviation is usually in measurement Novovoronezh 2 and minimal deviation is usually for numerical benchmark CB2. Usually deviation for measurement Takahama is lower then for Novovoronezh 1 and 2.

Pins in assembly VVER-440 have different ratio uranium/water (by central tube, at corner, by side, surrounding by fuel pins) and therefore the concentration depends on pin position. In [12] is detailed analyse of nuclide concentration in pins. The maximal deviation is usually in corner's pin and for the most of nuclides is less 3%, maximal is 6%.

Conclusion

The nuclide composition calculation of spent fuel assembly has different deviation for different nuclides. 17 nuclides were compared in 3 (2) measurements and one numerical benchmark. Very good agreement (less 5%) is for U236, U238, Nd143, Nd145. Good agreement (less 15%) is for U235, Pu and some Sm isotopes. For Am243, Cm244 and some Sm isotopes is deviation higher (more than 25%). Some nuclides (Eu isotopes) were measured only in Voronezh 2 experiment and have extreme high deviation (more than 150%). The high deviation is usually for nuclides with very low concentration.

The system SCALE 5 is very good tool for calculation of spent fuel VVER-440. By nuclide composition the accuracy depends on nuclide, it is necessary to continue in investigation.

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Fig.1 Measured pins in experiment Novovoronezh 1 (CT = central tube)

Pin N^{o} 25 – 5 samples
Pin N^{o} 63 – 3 samples
Pin N^{o} 64 – 3 samples
Pin N^{o} 107 – 1 sample
Total – 12 samples

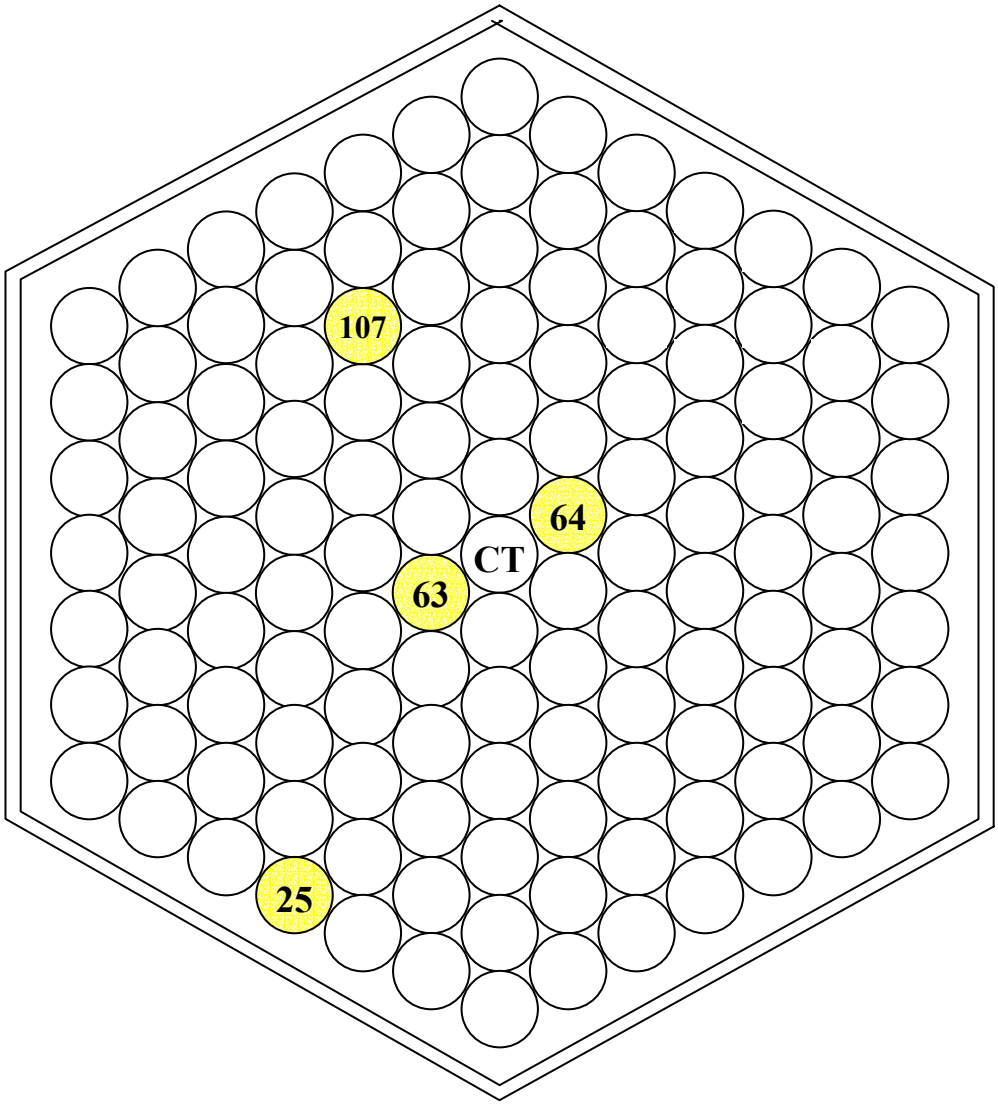
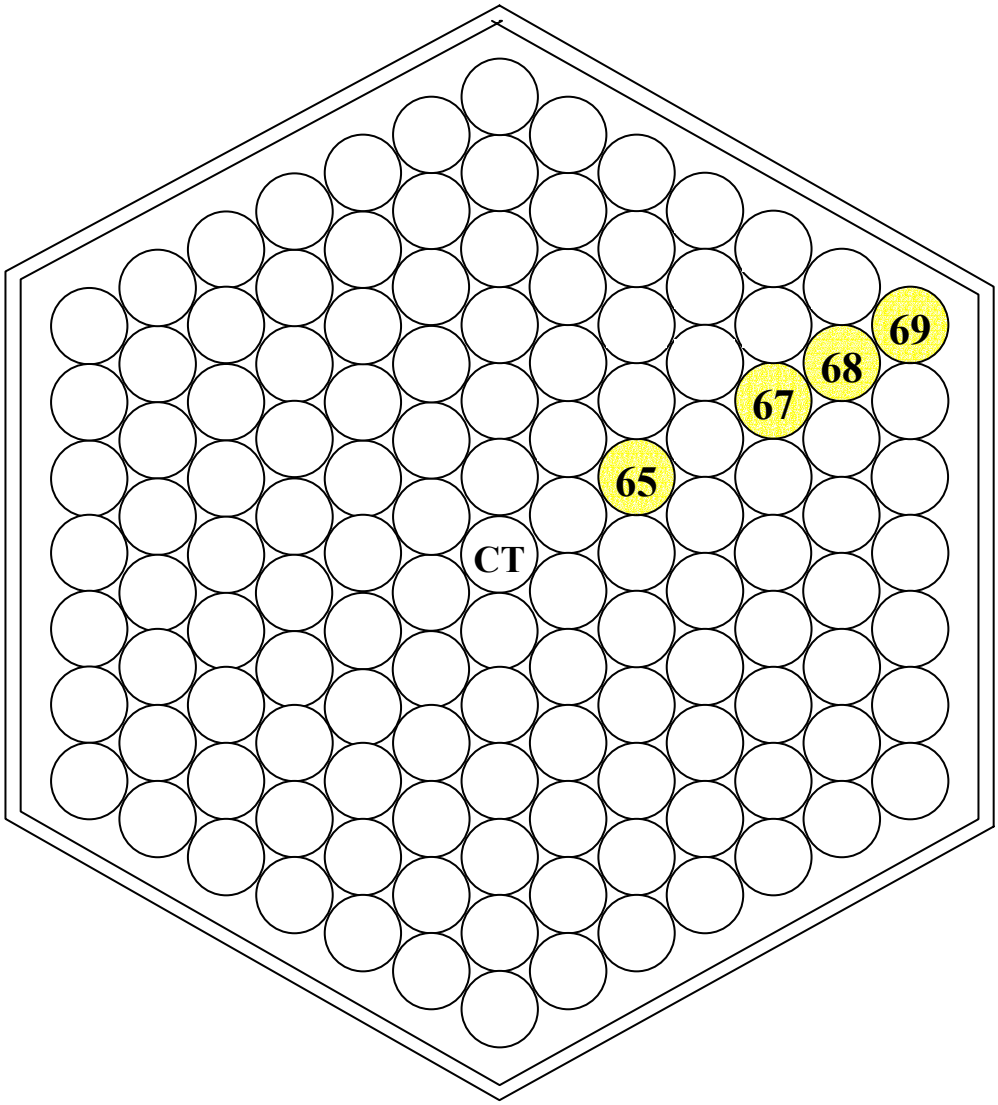


Fig.2 Measured pins in experiment Novovoronezh 2 (CT = central tube)

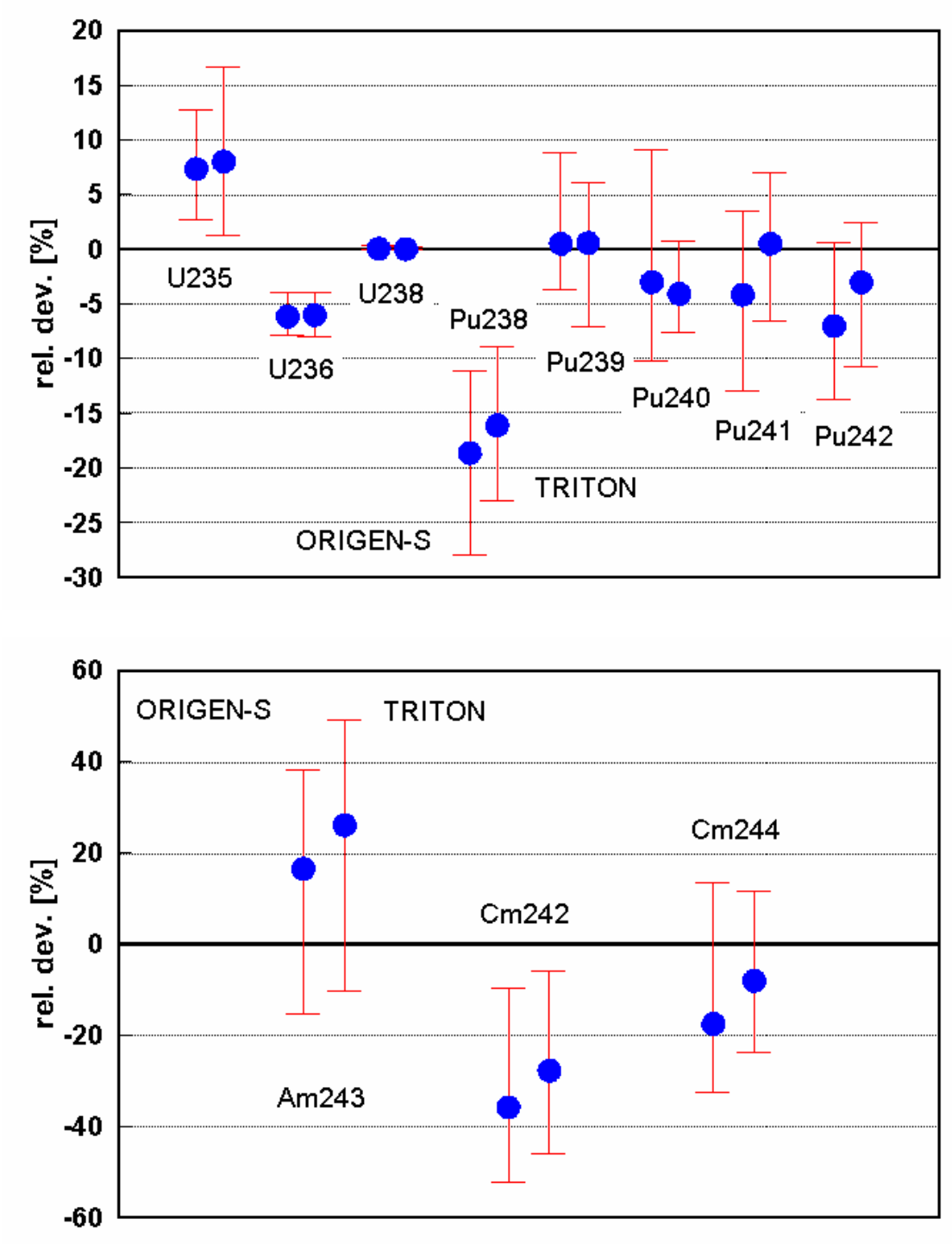
Pin N^{o} 65 – 3 samples
Pin N^{o} 67 – 1 sample
Pin N^{o} 68 – 1 sample
Pin N^{o} 69 – 3 samples
Total – 8 samples



Tab.1.Deviation (%) of nuclide concentration, experiment Novovoronezh 1 (12 samples)

	ORIGEN-S				TRITON			
nuclide	min	average	max	σ	min	average	max	σ
U235	2.75	7.33	12.69	7.96	1.24	8.00	16.63	9.24
U236	-7.94	-6.17	-4.00	6.25	-8.05	-6.03	-3.97	6.15
U238	-0.11	0.05	0.35	0.13	-0.11	0.01	0.22	0.08
Pu238	-28.01	-18.63	-11.19	19.30	-23.03	-16.11	-8.97	16.75
Pu239	-3.72	0.50	8.85	3.59	-7.10	0.57	6.07	3.87
Pu240	-10.21	-3.01	9.02	7.42	-7.58	-4.08	0.78	4.83
Pu241	-12.95	-4.16	3.52	6.03	-6.52	0.51	7.04	3.14
Pu242	-13.73	-7.03	0.60	8.29	-10.79	-3.03	2.42	5.14
Am243	-15.40	16.49	38.28	23.42	-10.29	26.08	49.26	31.17
Cm242	-52.17	-35.84	-9.80	38.14	-45.96	-27.78	-5.87	29.90
Cm244	-32.70	-17.55	13.32	22.38	-23.85	-8.07	11.59	12.99

Fig.3 Deviation of nuclide concentration, experiment Novovoronezh 1 (12 samples)



Tab.2. Deviation (%) of nuclide concentration, experiment Novovoronezh 2 (8 samples)

	ORIGEN-S				TRITON			
nuclide	min	average	max	σ	min	average	max	σ
u234	-99.28	-98.44	-96.72	98.44	-99.3	-98.34	-96.7	98.34
u235	-0.39	13.79	60.61	23.15	-4.04	13.05	46.92	19.68
u236	-13.1	-9.81	-7.11	9.99	-4.25	-2.25	1.56	3.05
u238	-0.13	0.06	0.25	0.12	-0.1	-0.01	0.14	0.08
pu238	-24.05	-4.88	20.59	13.8	-10.43	7.94	30.88	14.31
pu239	-3.73	4.28	15.18	7.14	-3.37	5.1	11.76	6.54
pu240	-12.93	-3.93	10.67	8.7	-10.35	-5.27	2.5	6.55
pu241	-15.23	-2.27	15.01	9.36	-7.79	0.13	7.15	4.7
pu242	-18.5	-10.65	1.56	12.25	-16.29	-8.58	5.54	10.7
np237	-64.28	-59.76	-52.74	59.85	-58.36	-55.1	-48.71	55.16
am241	-58.63	-5.54	36.95	27.62	-61.28	-1.3	34.84	28.49
am242m	-31.34	38.43	200.6	76.03	-13.98	40.32	194.4	72.45
am243	-18.4	12.08	35.53	23	-13.38	16.07	40.46	23.03
cm244	-52.89	-30.76	-7.12	33.84	-51.21	-26.88	-5.19	30.57
cm245	-64.03	-36.02	0.37	41.85	-55.8	-32.29	-6.3	36.71
cm246	-81.62	-46.43	-11.72	51.95	-74.18	-42.42	-12.25	47.48
nd142	-14.28	-6.48	12.6	10.69	-14.37	-5.92	12.9	10.32
nd143	-3.61	-1.28	2.69	2.29	-3.34	-1.35	2.86	2.28
nd144	12.38	21.81	29.27	22.31	15.73	21.93	27.65	22.37
nd145	-12.46	-9.2	-3.42	9.64	-12.48	-9.43	-4.05	9.84
nd146	1.68	3.88	5.88	4.08	1.85	3.87	5.42	4.03
nd148	-6.93	-4.83	-2.22	5.06	-6.99	-4.95	-2.26	5.15
nd150	-5	-2.5	0.43	3.08	-4.83	-2.6	-0.7	3
cs133	-4.92	-2.9	1.43	3.67	-5.42	-3.46	0.6	4.11
cs134	-58.17	-9.39	43.07	38.34	-55.99	-9.7	40.16	37.34
cs135	-6.89	-0.58	8.83	4.33	-5.14	-0.35	4.37	2.97
cs137	-7.14	-4.45	-1.54	4.83	-7.27	-4.56	-1.68	4.93
ce140	-4.22	-1.68	1.79	2.65	-2.43	-0.27	3.19	2.05
ce142	-5.8	-2.45	0.93	3.41	-5.43	-2.54	0.93	3.44
ce144	-50.17	-47.45	-44.77	47.48	-51.46	-48.03	-45	48.07
sm147	-69.85	-66.84	-65.62	66.86	-70.52	-66.88	-64.9	66.9
sm148	-22.39	-15.07	-10.38	15.62	-19.78	-15.05	-9.59	15.49
sm149	-21.49	-12.04	-4.76	12.82	-23.71	-15.4	-3.77	16.7
sm150	-11.06	2.84	7.71	6.39	-10.96	2.69	7.89	6.29
sm151	23.71	53.51	79.23	55.89	31.08	54	79.62	56.27
sm152	4.18	22.77	29.69	24.03	2.11	21.47	30.09	22.96
sm154	-12.92	-0.6	3.54	5.15	-11.24	-0.53	4.78	4.59
eu151	-92.52	-90.52	-84.75	90.55	-93.36	-90.48	-86.54	90.5
eu153	221.16	524.94	1008.55	573.91	226.73	523.73	1009.21	572.14
eu154	278.2	623.02	1147.13	672.43	303.46	626.64	1092.36	670.52
eu155	117.28	413.81	878.71	472.03	116.4	413.61	879.88	472.57
mo95	-12.64	-11.05	-7.4	11.19	-12.68	-10.88	-6.63	11.04
tc99	-2.7	-0.91	1.7	1.74	-3.14	-1.17	1.41	1.89
ru101	-3.51	-1.94	1.15	2.49	-3.83	-2.08	0.88	2.6
pd105	-11.72	-6.71	-1.93	7.29	-9.73	-6.64	-2.86	7.1
pd108	8.2	21.19	31.93	22.27	12.31	21.66	30.9	22.37
ag109	32.94	71.7	115.29	75.27	36.92	71.95	115.29	75.21
gd155	-94.55	-93.72	-91.5	93.72	-94.42	-93.52	-92.15	93.53

Fig.4a Deviation of nuclide concentration, experiment Novovoronezh 2 (8 samples)

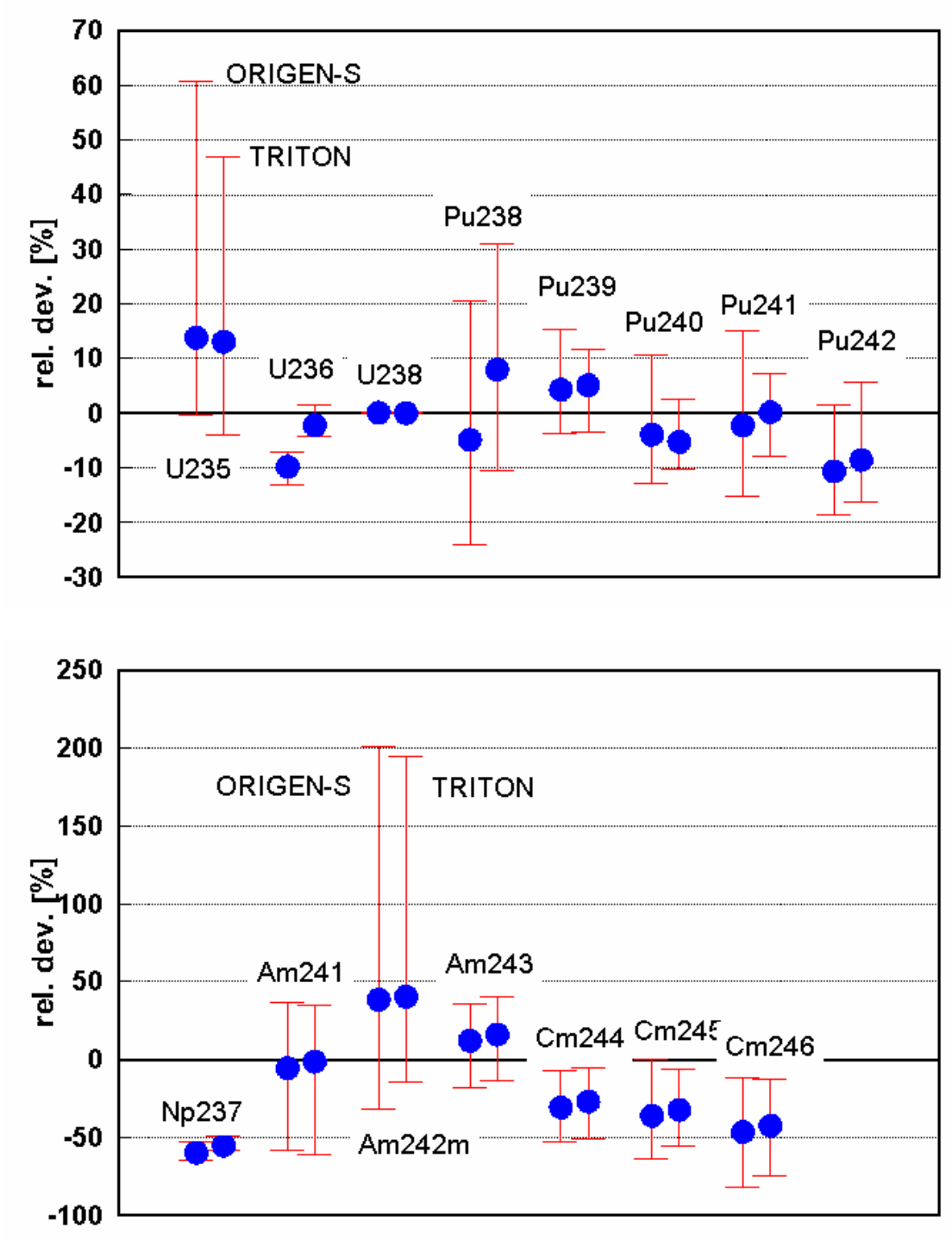


Fig.4b Deviation of nuclide concentration, experiment Novovoronezh 2 (8 samples)

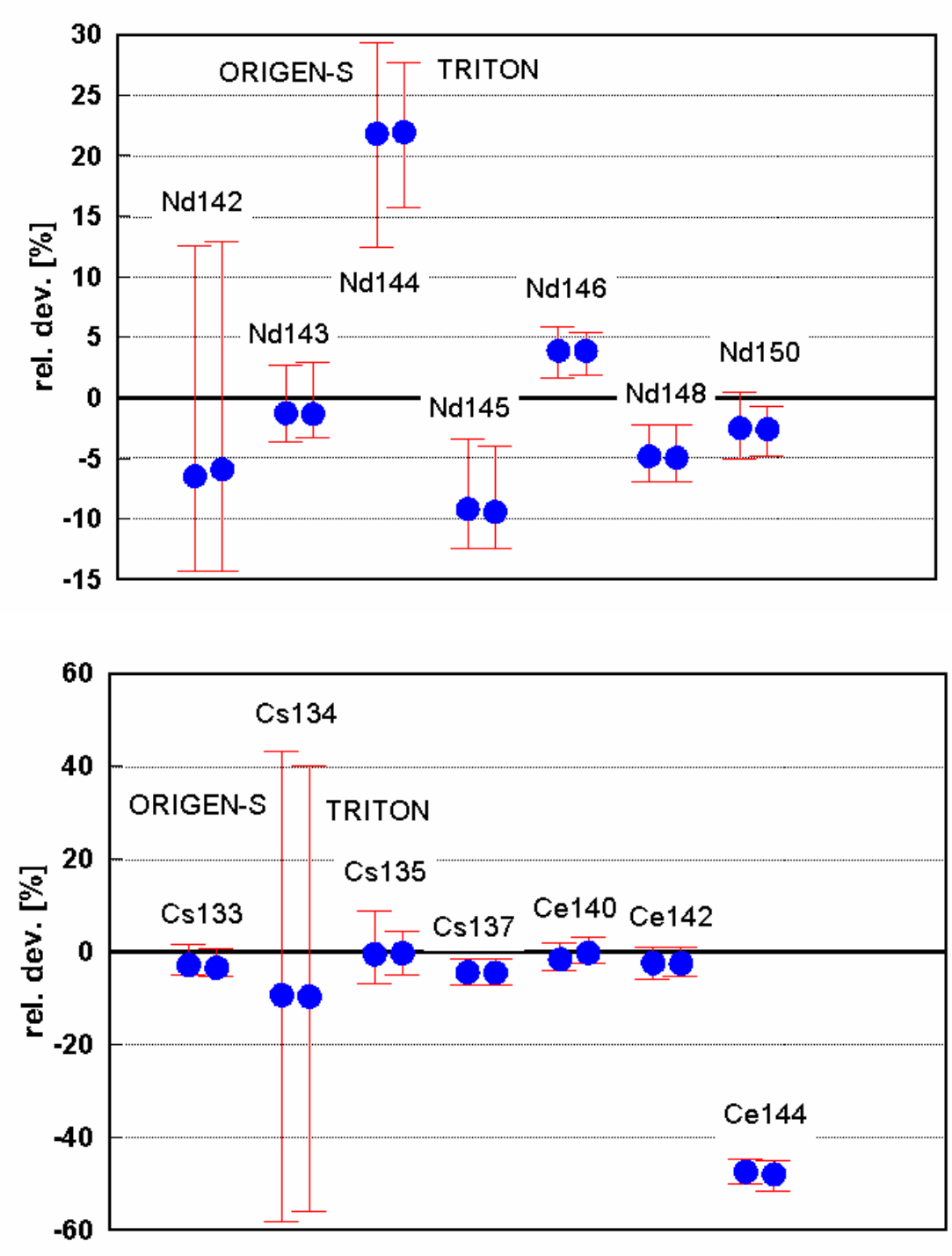
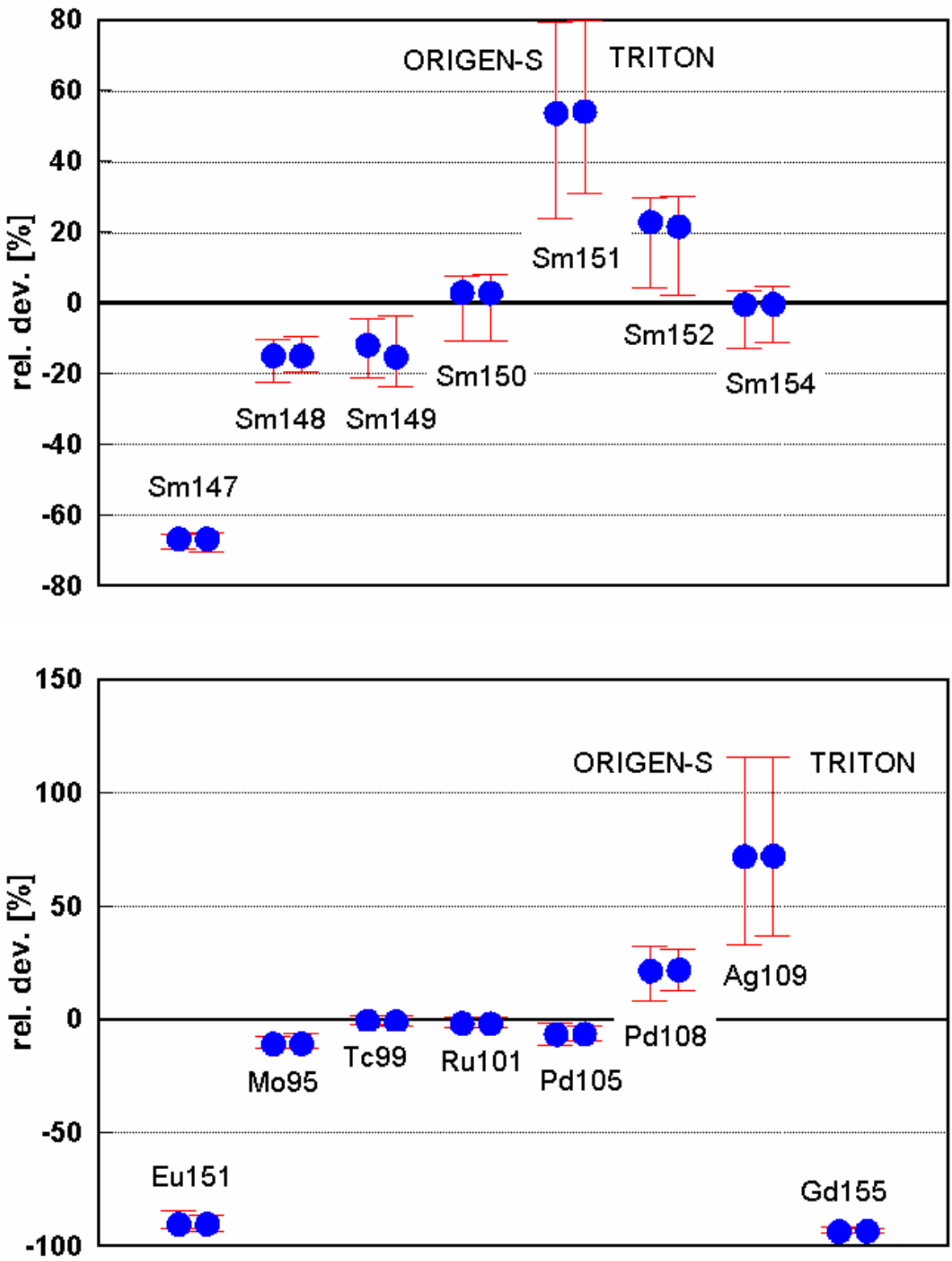


Fig.4c Deviation of nuclide concentration, experiment Novovoronezh 2 (8 samples)



Tab.3 Deviation (%) of nuclide concentration, experiment Takahama-3 (5 samples)

	ORIGEN-S				TRITON			
nuclide	min	average	max	σ	min	average	max	σ
u234	5.47	7.00	10.26	7.22	7.48	8.83	11.75	8.97
u235	-5.09	-1.91	1.77	3.00	-6.00	-2.91	0.79	3.68
u236	-0.49	-0.15	0.13	0.27	-0.34	-0.04	0.17	0.18
u238	-0.11	0.00	0.08	0.06	0.31	0.42	0.50	0.43
np237	-2.01	0.71	2.14	1.61	-4.39	-2.04	-0.54	2.42
pu238	-11.93	-9.16	-2.80	9.75	-15.61	-12.89	-6.62	13.29
pu239	-2.67	0.75	6.39	3.31	-6.10	-2.74	2.73	4.08
pu240	3.10	4.70	7.27	4.92	-2.47	-1.04	1.59	1.79
pu241	-7.37	-3.97	1.87	5.24	-9.06	-5.79	0.20	6.71
pu242	-1.79	-0.94	0.54	1.25	-1.43	-0.33	1.39	1.00
am241	8.77	16.91	31.67	18.86	13.03	21.55	37.12	23.27
am242m	2.84	10.75	26.08	13.67	1.78	9.71	25.15	12.89
am243	7.81	11.84	19.40	12.49	3.82	10.13	17.69	11.10
cm242	-8.44	3.83	15.71	8.98	-11.74	1.13	13.08	8.41
cm243	-25.24	-19.15	-12.64	19.65	-30.11	-22.88	-16.52	23.37
cm244	-11.63	-5.28	5.69	7.99	-18.14	-9.98	0.59	11.79
cm245	-45.73	-40.03	-27.38	40.57	-45.79	-37.95	-24.93	38.65
cm246	-19.74	-16.37	-5.16	17.31	-46.28	-41.33	-33.45	41.55
cm247	13.73	22.58	39.76	24.44	-52.04	-44.17	-36.57	44.51
nd142	-4.04	1.08	3.15	2.84	-4.86	0.64	2.92	2.90
nd143	-1.59	-0.60	0.23	0.84	-1.59	-0.60	0.33	0.88
nd144	-4.57	-1.72	2.52	3.03	-2.25	0.60	4.76	2.54
nd145	0.23	0.88	1.60	1.00	1.20	1.80	2.48	1.85
nd146	0.49	0.65	0.98	0.68	0.89	1.15	1.55	1.18
nd148	-0.27	0.03	0.59	0.31	0.21	0.52	1.06	0.62
nd150	0.49	0.87	1.64	0.97	0.94	1.35	2.21	1.43
cs134	-20.35	-15.29	-12.50	15.59	-23.36	-18.16	-15.35	18.43
cs137	-1.01	-0.63	-0.11	0.72	-2.47	-2.12	-1.66	2.14
eu154	-5.27	2.10	7.97	5.16	-10.29	-3.23	2.45	5.55
ce144	-9.44	-0.35	7.28	6.49	-12.32	-3.70	3.55	7.17
ru106	-14.34	-3.81	8.11	8.21	-17.10	-6.99	4.60	9.92
sb125	14.93	54.05	83.57	59.94	19.27	58.59	89.22	64.29
sm147	-12.51	-10.44	-6.88	10.63	-2.18	-0.10	3.37	2.01
sm148	-17.18	-14.48	-11.22	14.71	-13.07	-10.22	-6.87	10.58
sm149	-4.88	8.89	20.51	12.83	-17.43	-6.27	3.47	9.92
sm150	7.24	8.31	9.68	8.35	4.57	5.13	6.63	5.19
sm151	23.32	33.68	45.44	34.58	15.17	23.83	34.85	24.84
sm152	24.66	28.76	31.82	28.91	24.87	28.70	31.59	28.83
sm154	0.20	1.75	3.66	2.08	-2.08	-1.00	0.97	1.45

Fig.5a Deviation of nuclide concentration, experiment Takahama (5 samples)

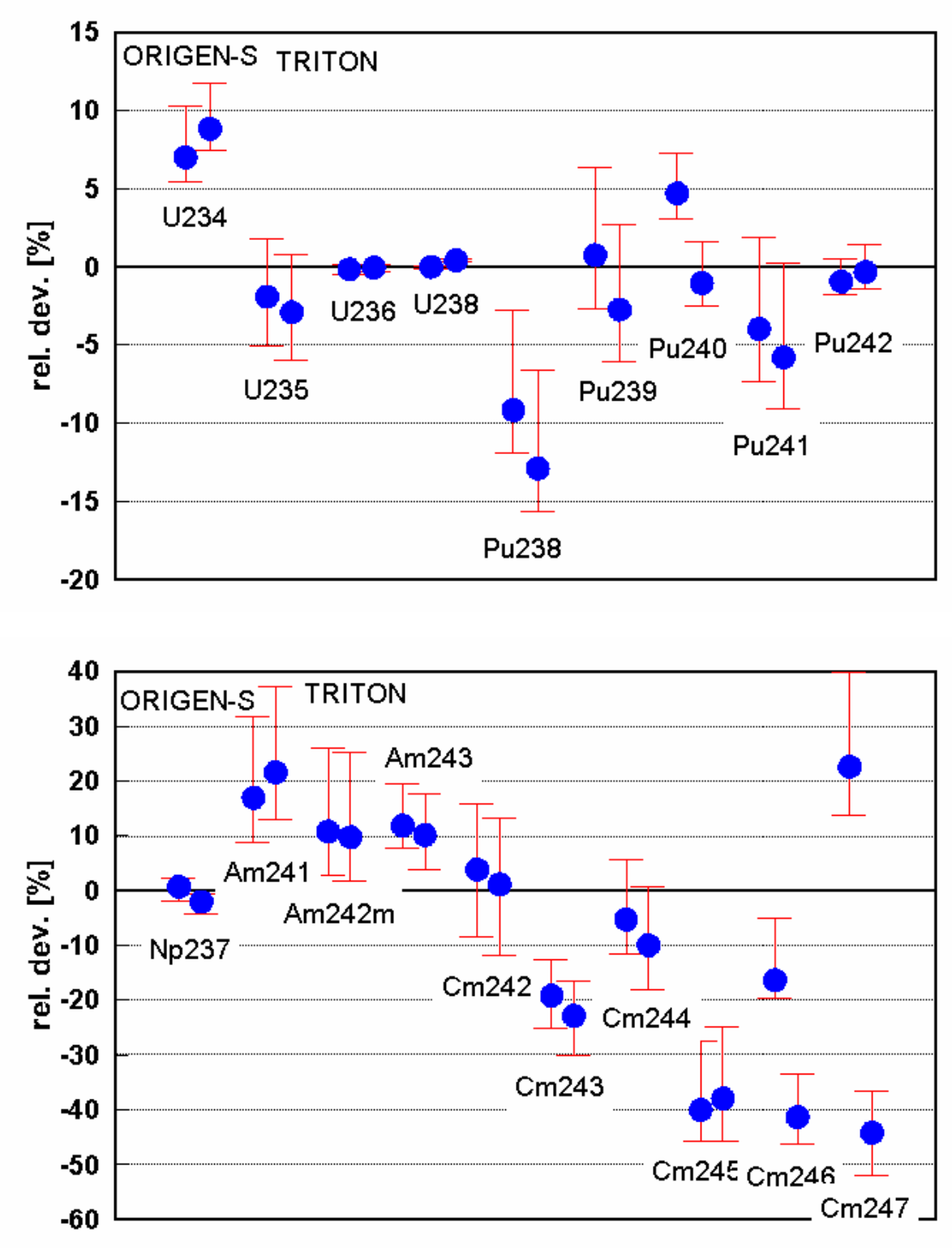
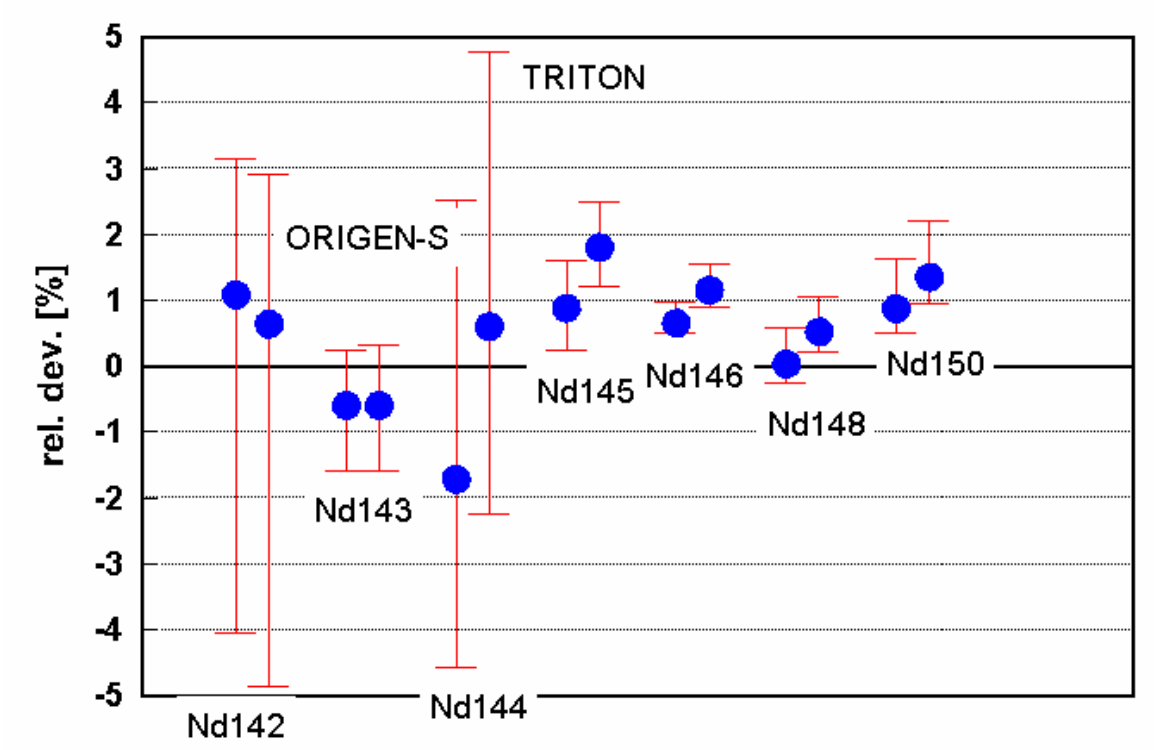


Fig.5b Deviation of nuclide concentration, experiment Takahama (5 samples)



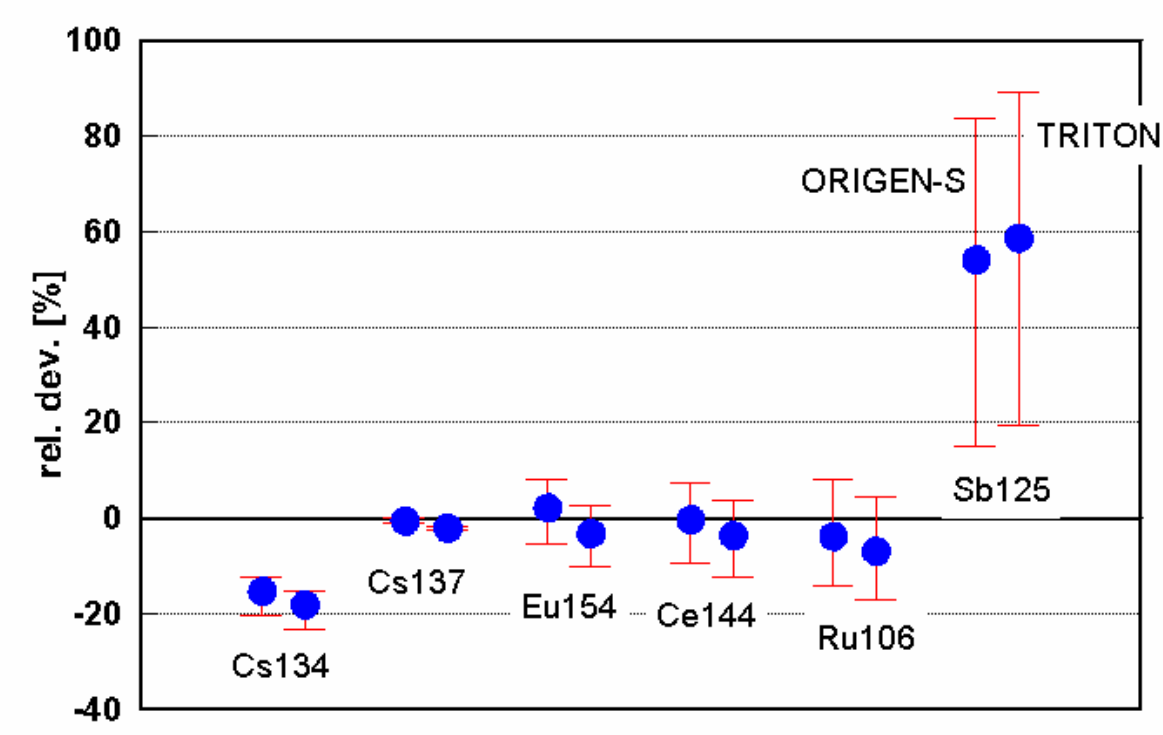
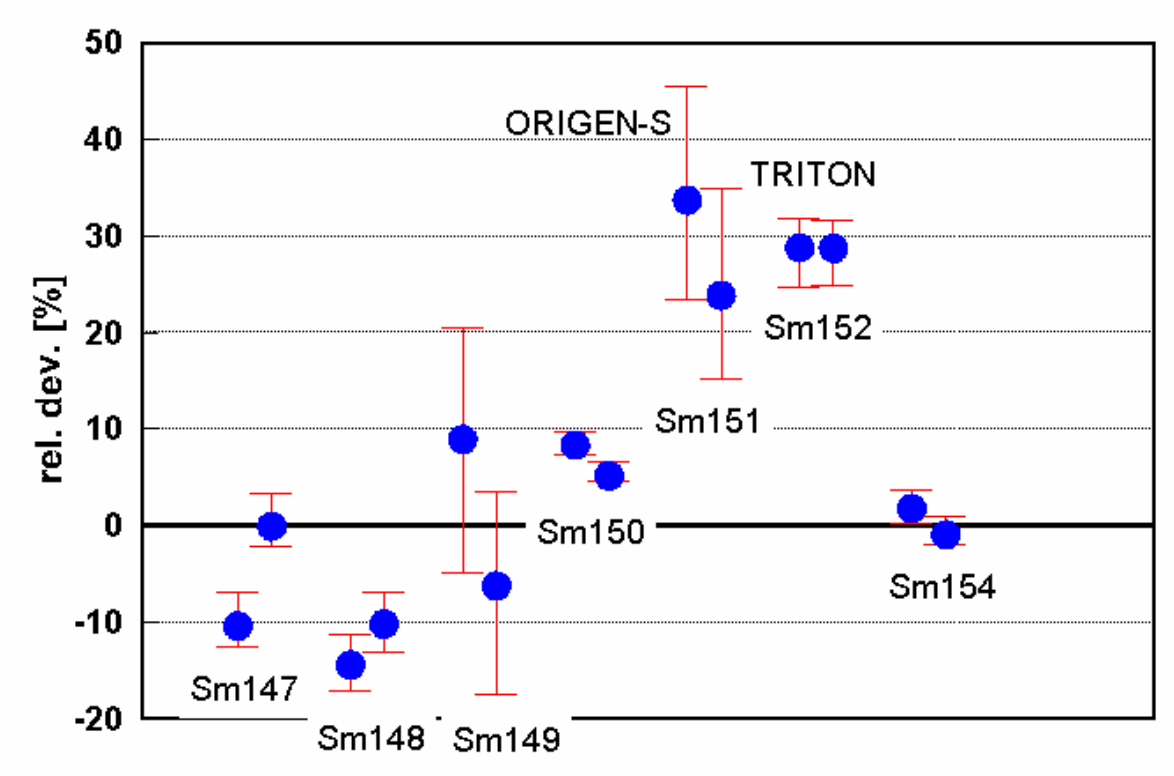


Fig.5c Deviation of nuclide concentration, experiment Takahama (5 samples)



Tab.4 Deviation (%) of nuclide concentration, numerical benchmark CB2 (6 samples)

	SAS2				TRITON			
nuclide	min	average	max	sigma	min	average	max	sigma
u235	-1.29	-0.79	-0.51	0.83	-5.82	-4.53	-3.46	4.64
u236	0.69	1.03	1.26	1.05	1.36	1.56	1.94	1.57
u238	-0.13	0.10	0.33	0.25	-0.14	0.09	0.33	0.25
np237	0.26	0.63	0.98	0.68	-0.89	-0.50	-0.08	0.57
pu238	-0.87	-0.02	1.36	0.86	-3.17	-2.40	-1.16	2.50
pu239	-1.64	-0.49	0.50	0.97	-5.23	-3.90	-2.48	4.04
pu240	-1.09	0.21	1.67	1.12	-1.31	0.10	1.88	1.29
pu241	1.50	1.98	2.78	2.03	-0.66	0.34	1.63	0.96
pu242	3.74	4.19	5.26	4.22	7.31	7.75	8.83	7.77
am241	-0.37	1.63	2.78	2.00	-2.48	0.02	1.90	1.43
am243	10.72	12.04	13.97	12.11	13.22	14.45	16.29	14.50
mo 95	-1.75	-0.60	0.09	0.90	-0.17	0.69	1.40	0.89
tc 99	-0.24	-0.03	0.08	0.12	0.83	1.03	1.18	1.04
ru101	0.90	1.10	1.20	1.11	2.09	2.22	2.39	2.23
rh103	2.04	3.74	4.77	3.85	3.14	4.52	5.33	4.59
ag109	-5.28	-3.44	-2.09	3.60	-4.91	-3.59	-2.76	3.67
cs133	1.44	1.92	2.38	1.95	2.33	2.63	2.95	2.64
nd143	-0.17	0.01	0.33	0.17	-0.47	0.08	0.73	0.46
nd145	0.27	0.46	0.96	0.52	1.29	1.58	1.99	1.59
sm147	-0.37	0.36	0.90	0.61	0.69	1.69	2.60	1.85
sm149	1.15	2.78	4.11	2.95	-9.02	-6.28	-4.11	6.46
sm150	1.64	2.58	3.39	2.67	1.49	2.38	3.15	2.48
sm151	8.50	10.31	11.70	10.39	3.38	5.31	7.16	5.48
sm152	3.55	4.83	5.91	4.94	5.00	6.24	7.27	6.32
eu153	-3.12	-2.37	-0.89	2.50	-2.30	-1.49	-0.08	1.69
gd155	-42.93	-32.11	-6.46	34.59	-42.60	-32.81	-6.22	35.16

Fig.6a Deviation of nuclide concentration, numerical benchmark CB2 (6 samples)

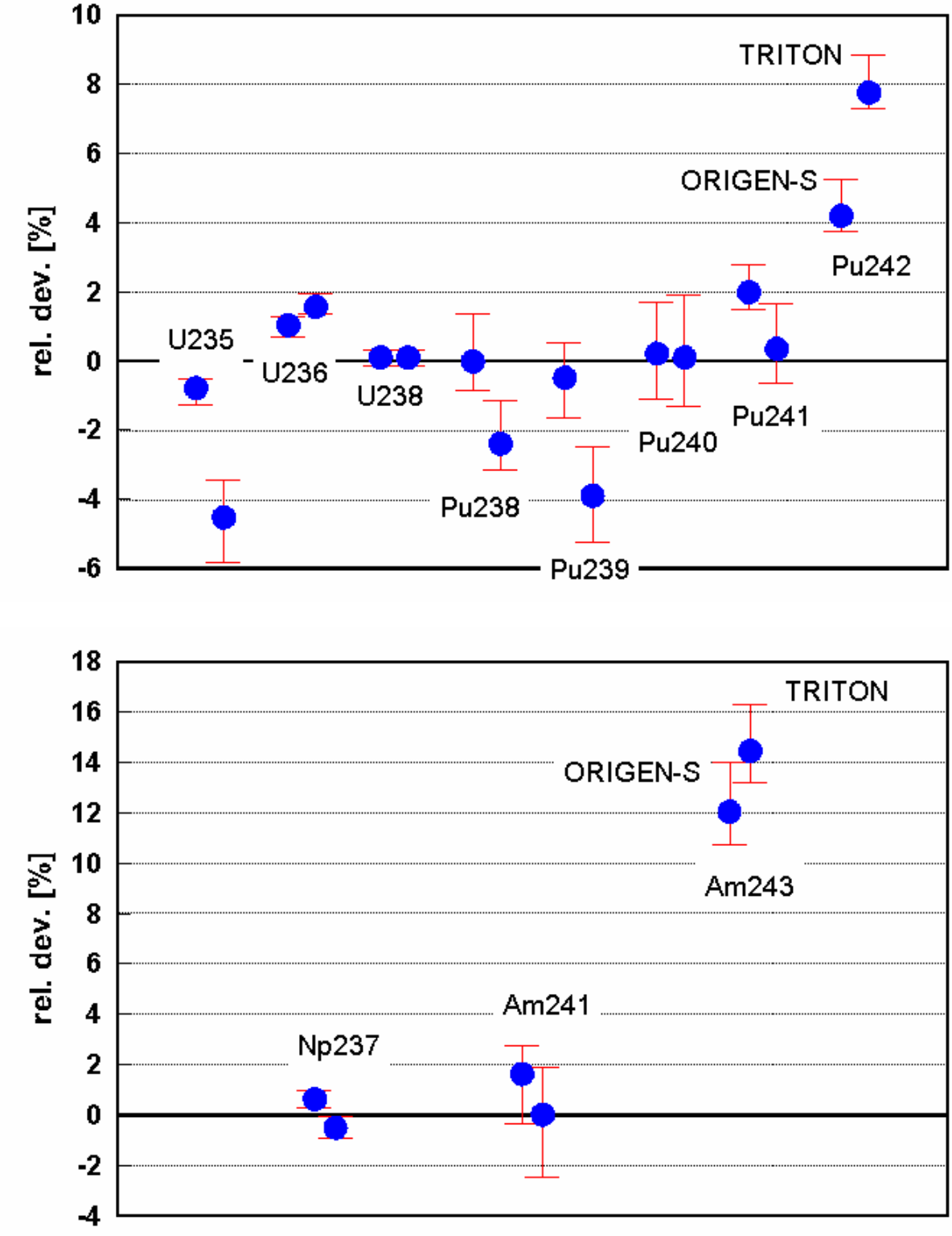
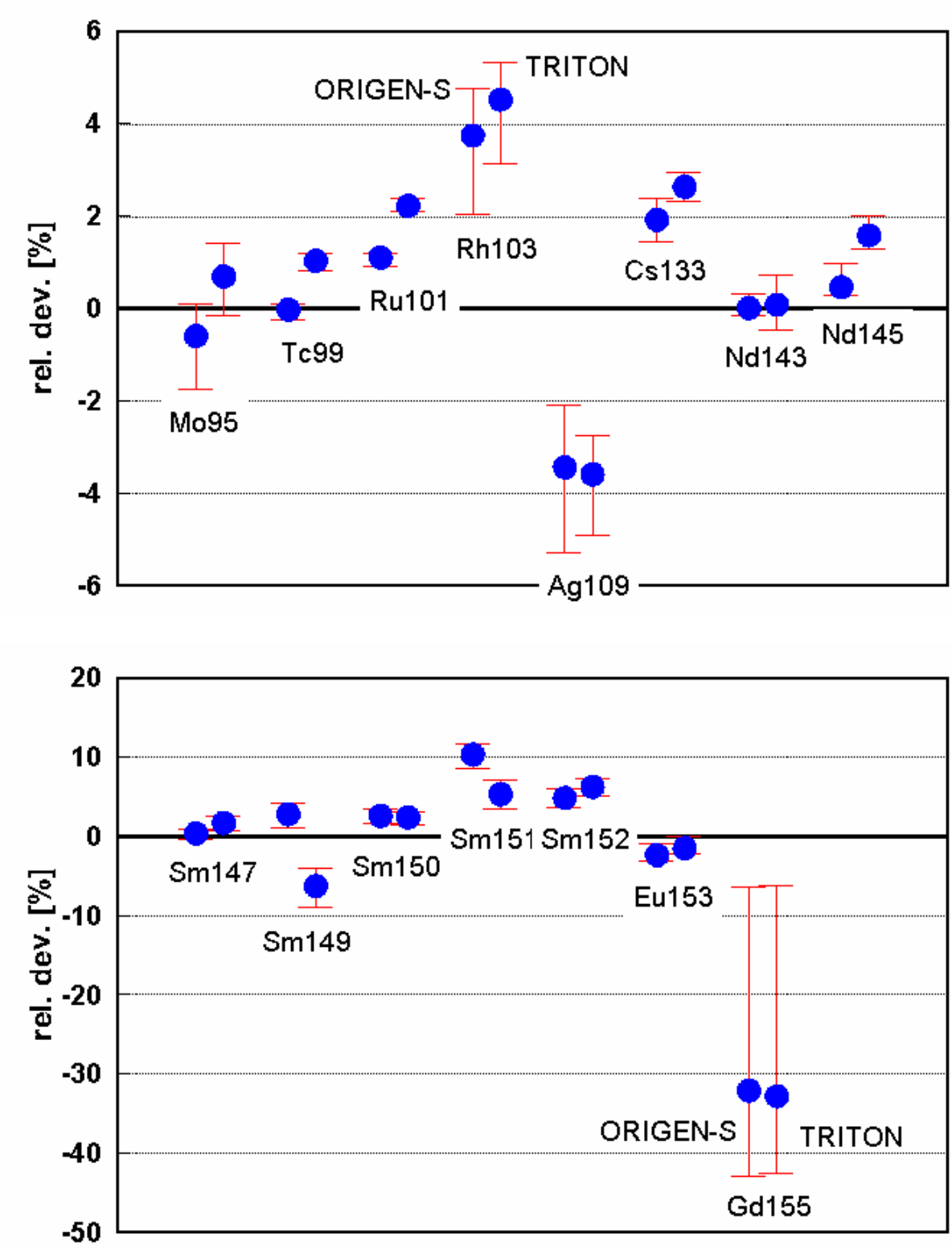


Fig.6b Deviation of nuclide concentration, numerical benchmark CB2 (6 samples)



Attachment 1

DEPENDENCE OF ACTINIDES CONCENTRATION ON BURN-UP

Fig.1a Dependence of U235 on burn-up, experiment Novovoronezh 1

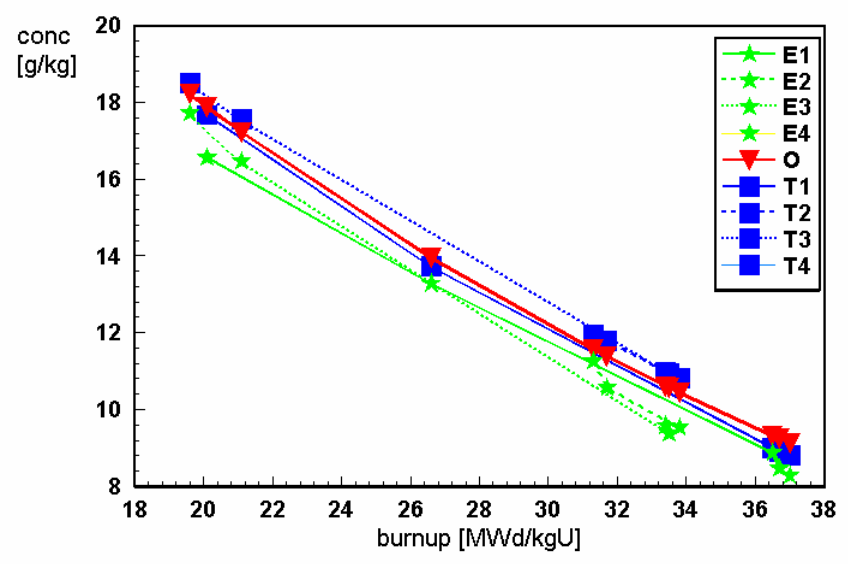


Fig.1b Dependence of U235 on burn-up, experiment Novovoronezh 2

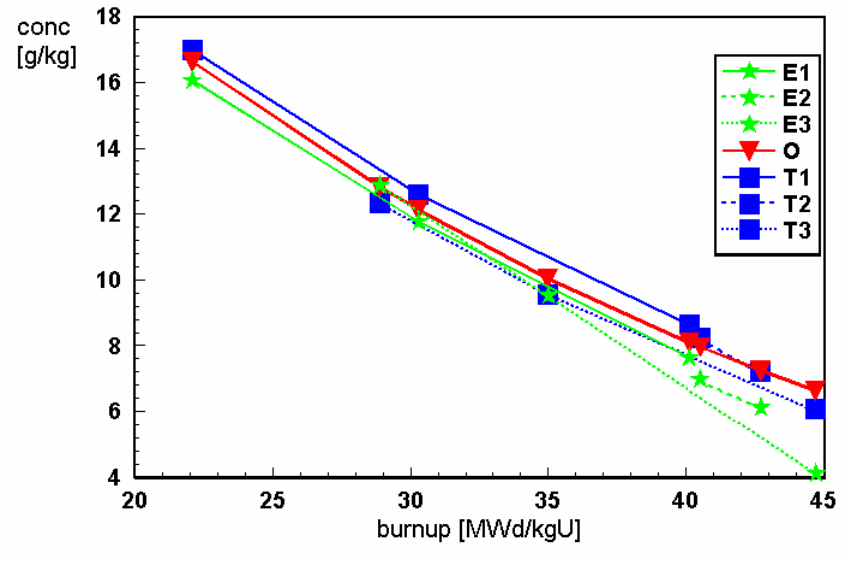


Fig.2a Dependence of U236 on burn-up, experiment Novovoronezh 1

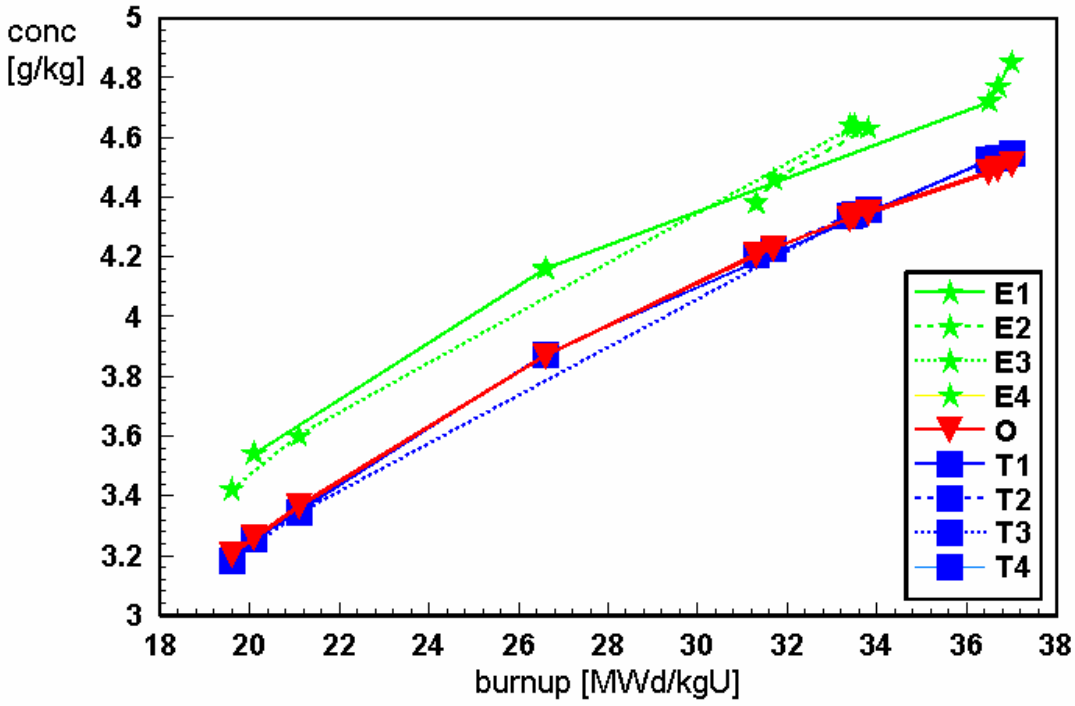


Fig.2b Dependence of U236 on burn-up, experiment Novovoronezh 2

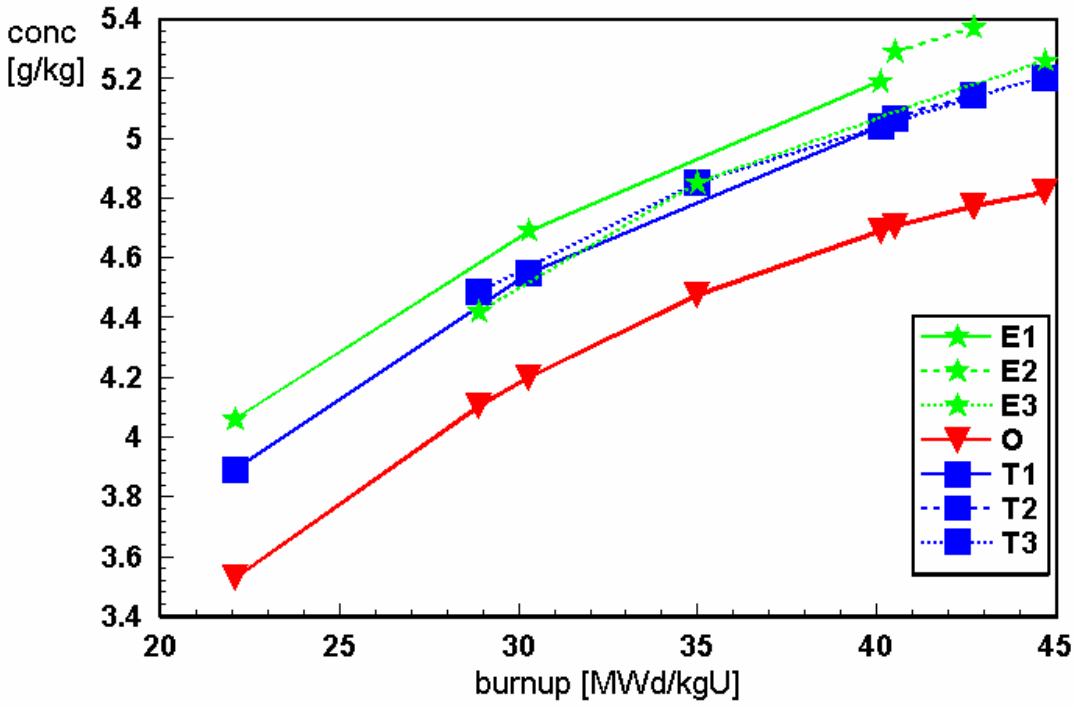


Fig.3a Dependence of U238 on burn-up, experiment Novovoronezh 1

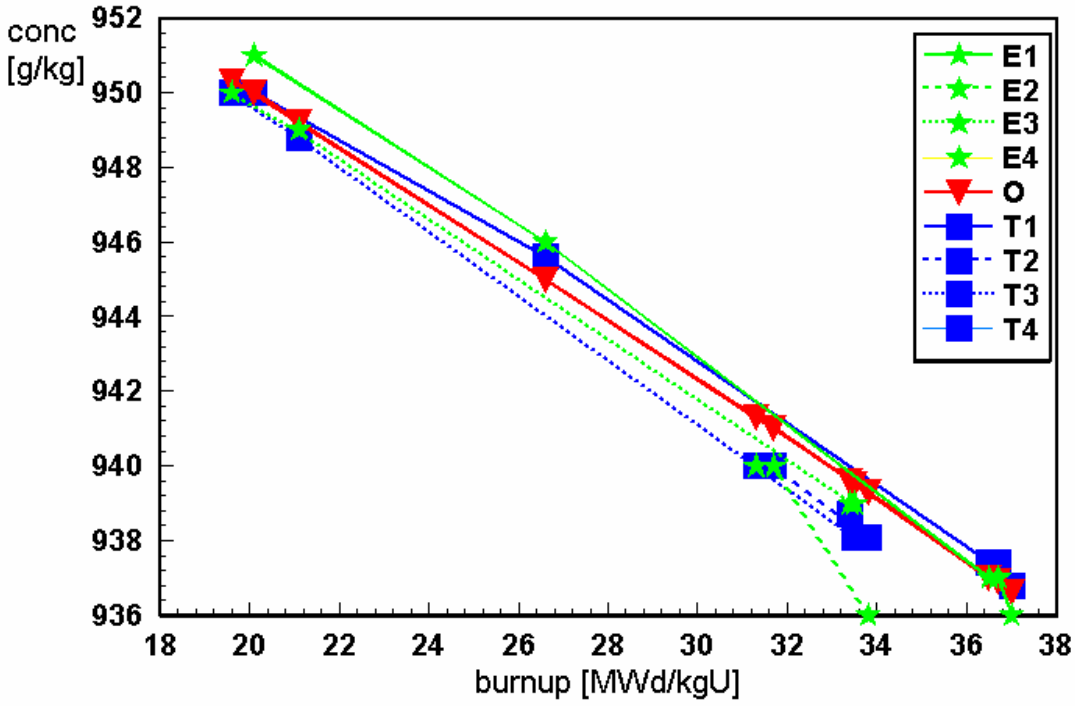


Fig.3b Dependence of U238 on burn-up, experiment Novovoronezh 2

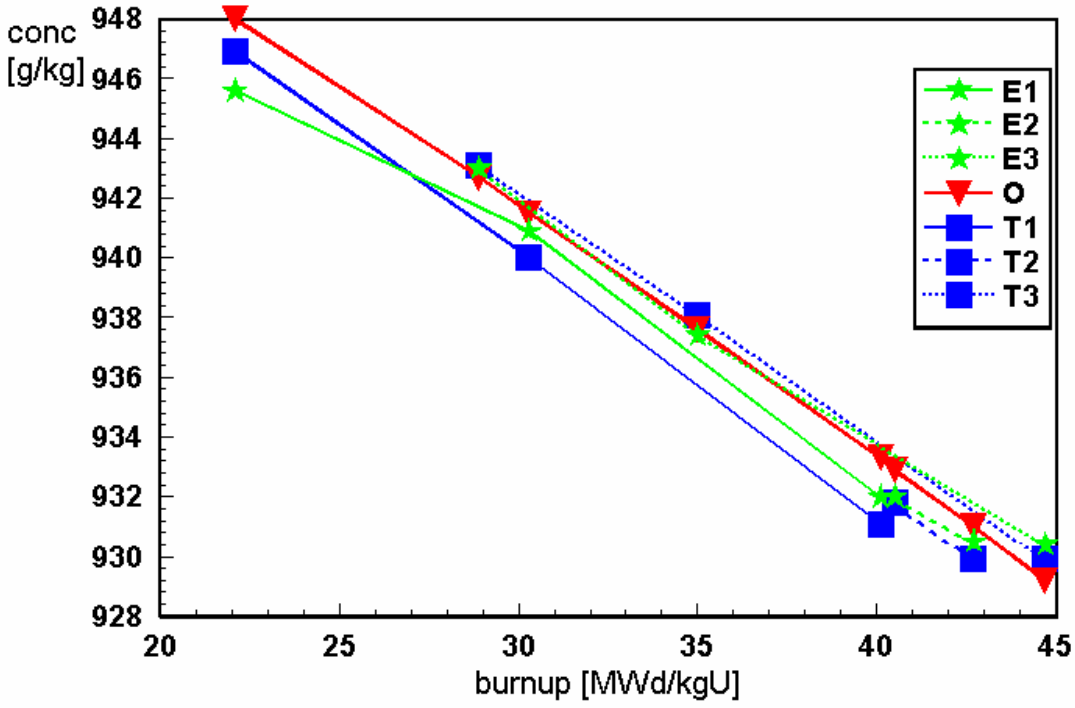


Fig.4a Dependence of Pu238 on burn-up, experiment Novovoronezh 1

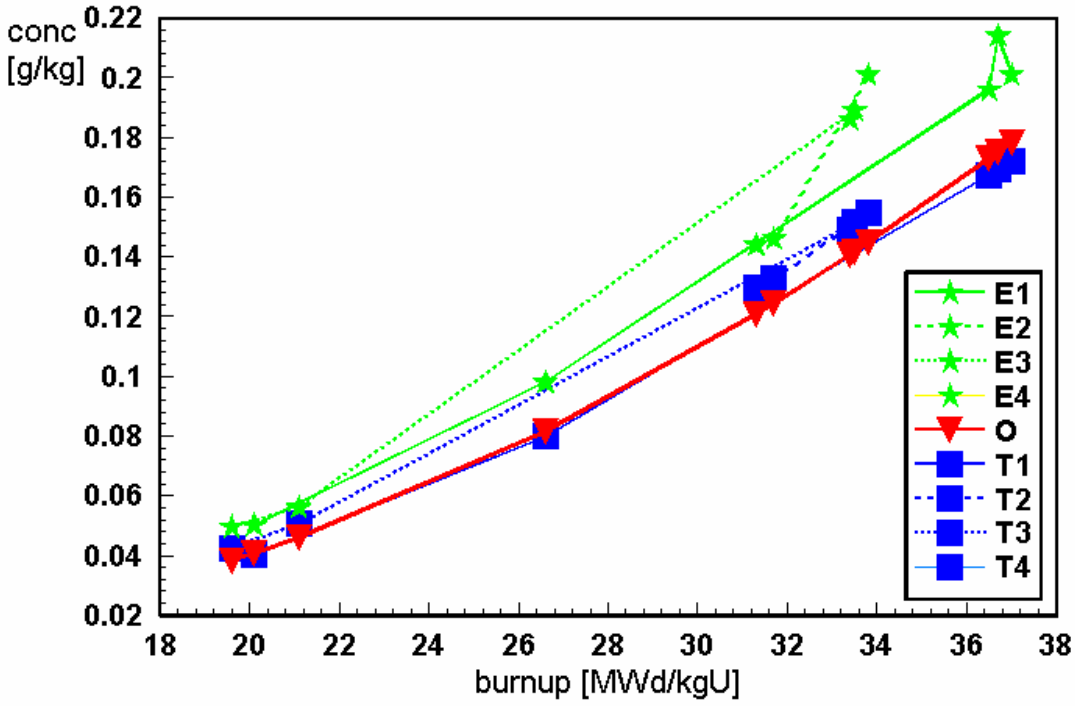


Fig.4b Dependence of Pu238 on burn-up, experiment Novovoronezh 2

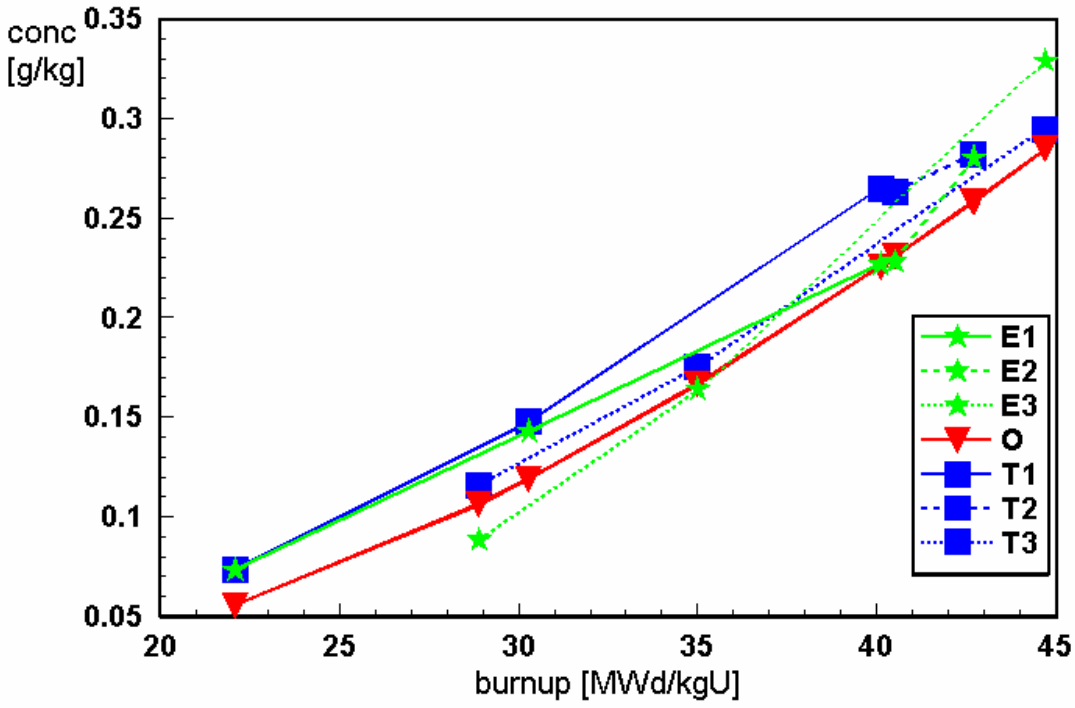


Fig.5a Dependence of Pu239 on burn-up, experiment Novovoronezh 1

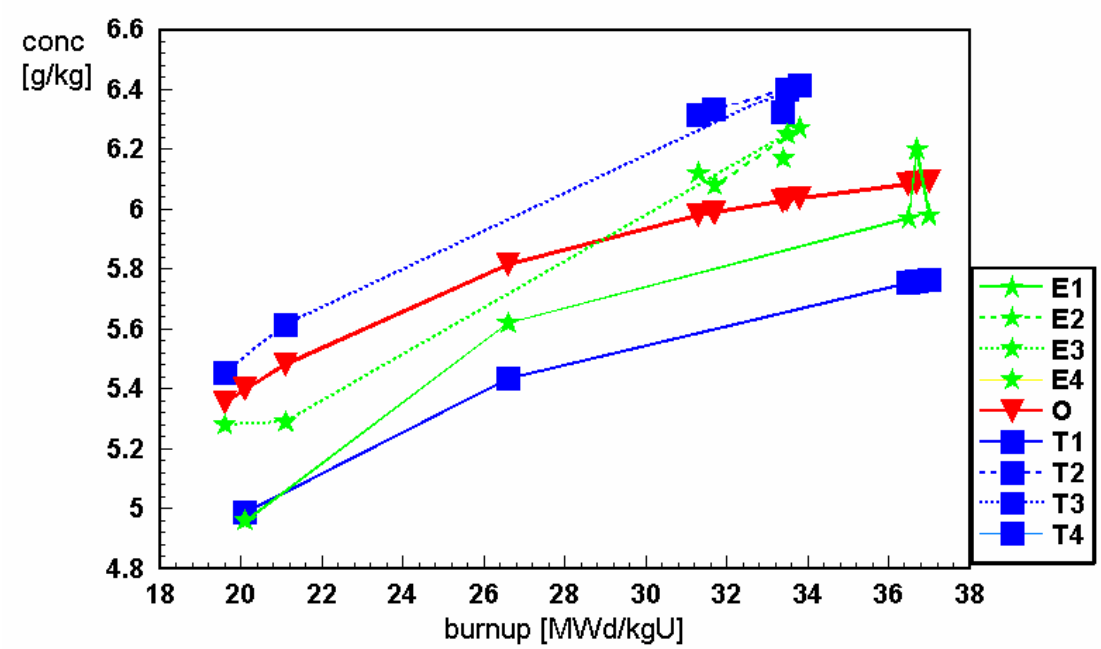


Fig.5b Dependence of Pu239 on burn-up, experiment Novovoronezh 2

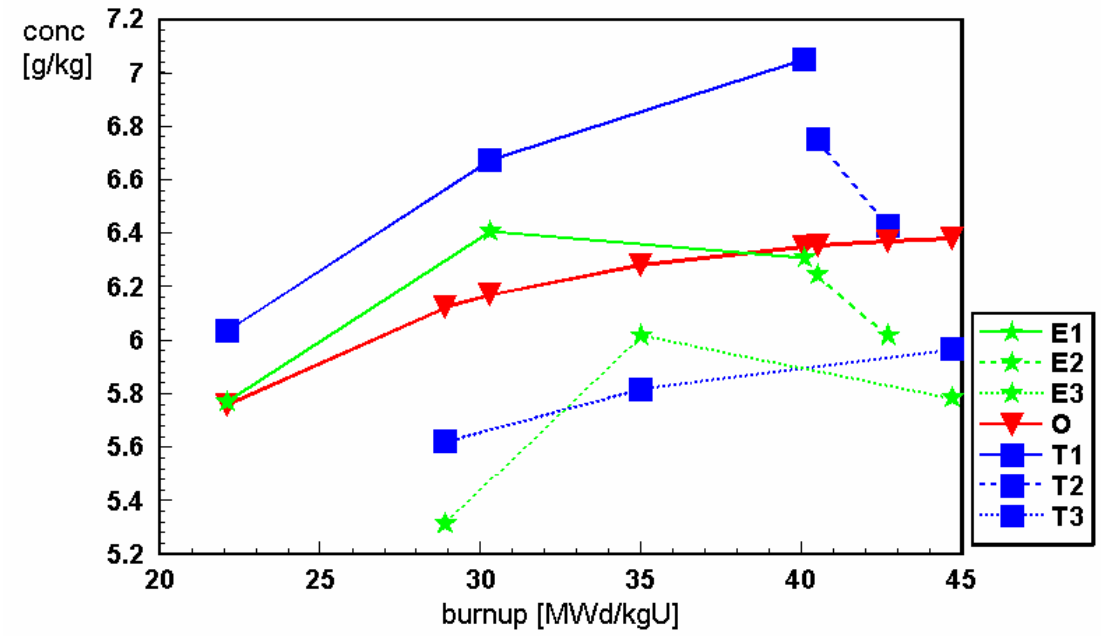


Fig.6a Dependence of Pu240 on burn-up, experiment Novovoronezh 1

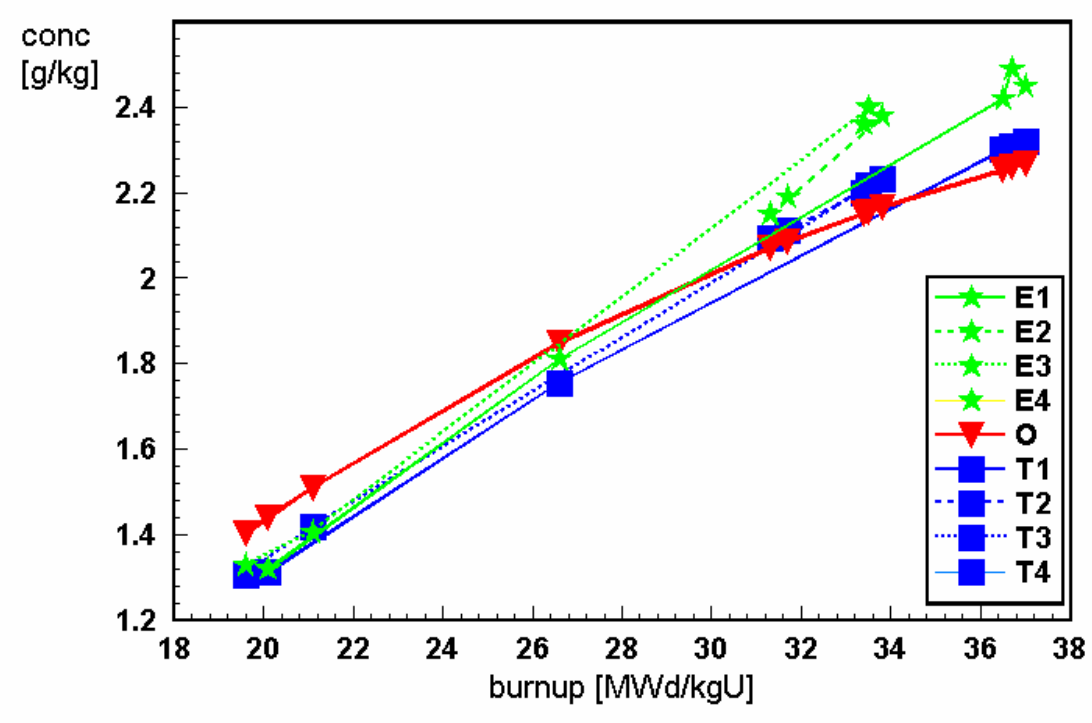


Fig.6b Dependence of Pu240 on burn-up, experiment Novovoronezh 2

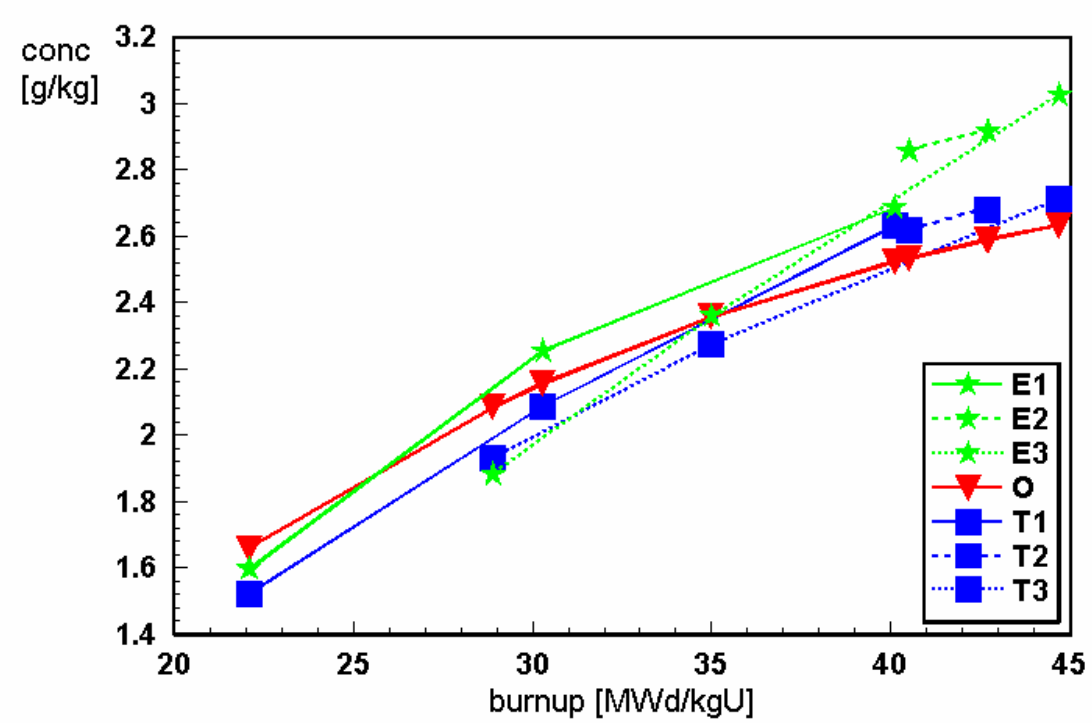


Fig.7a Dependence of Pu241 on burn-up, experiment Novovoronezh 1

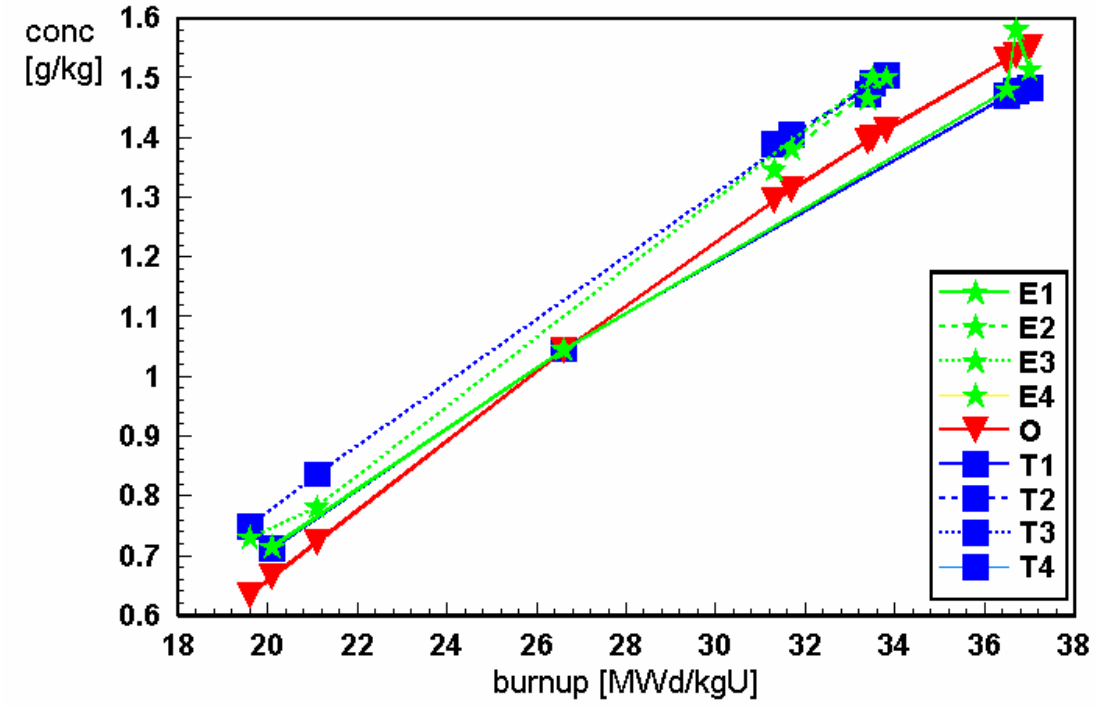


Fig.7b Dependence of Pu241 on burn-up, experiment Novovoronezh 2

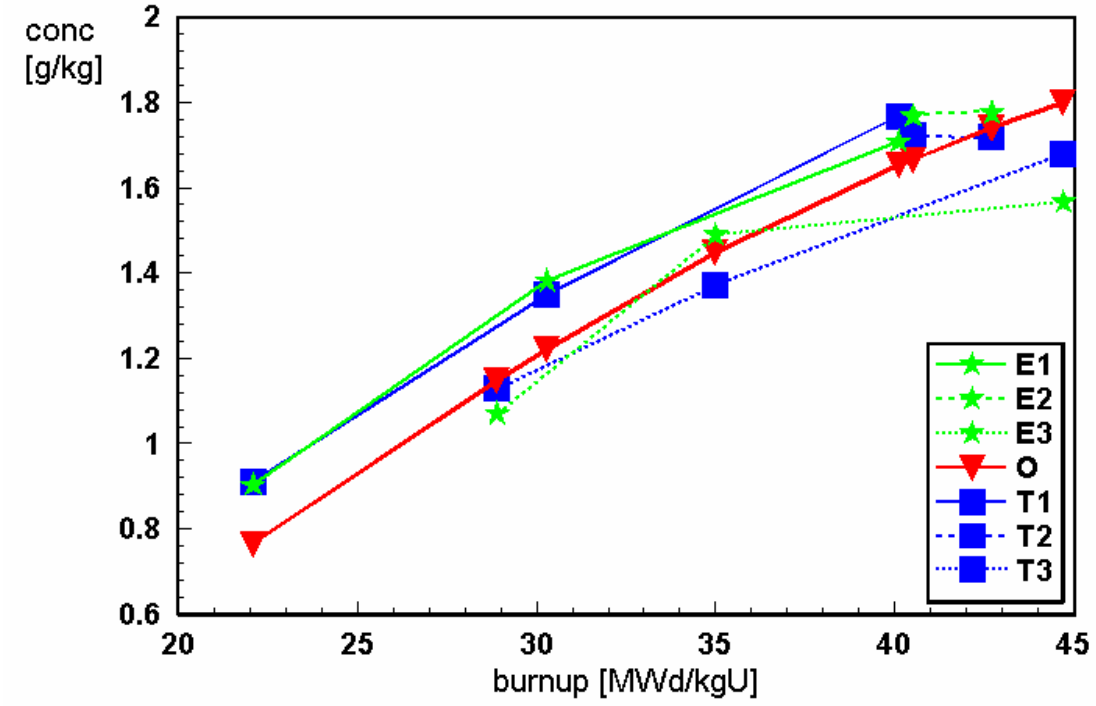


Fig.8a Dependence of Pu242 on burn-up, experiment Novovoronezh 1

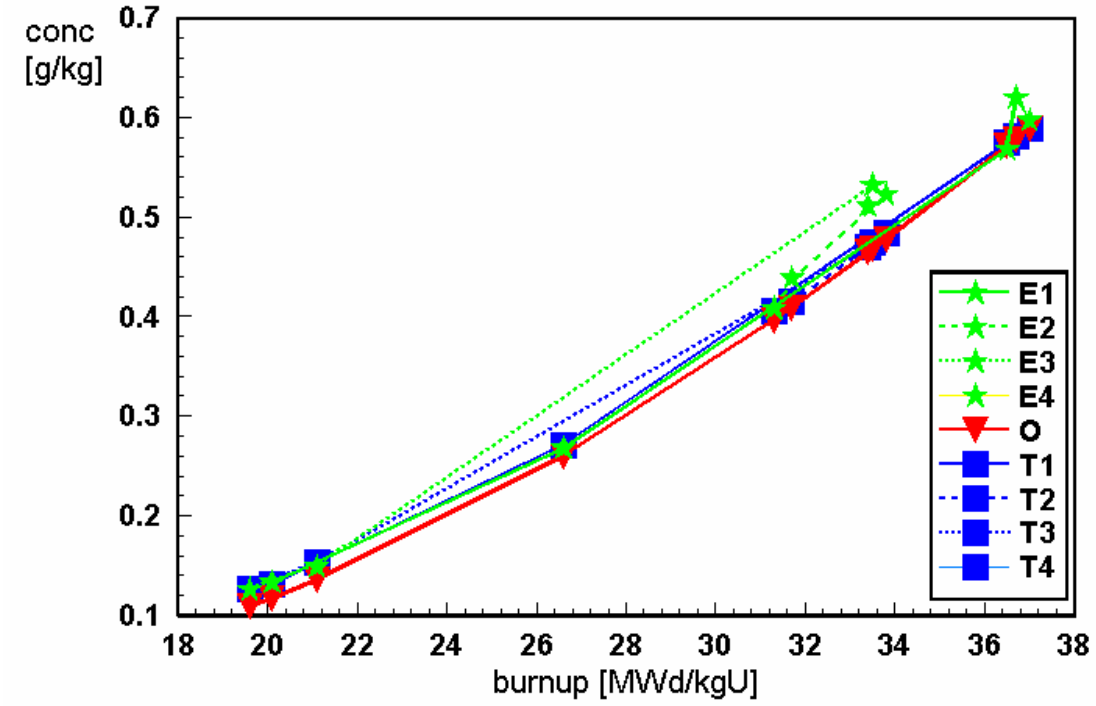


Fig.8b Dependence of Pu242 on burn-up, experiment Novovoronezh 2

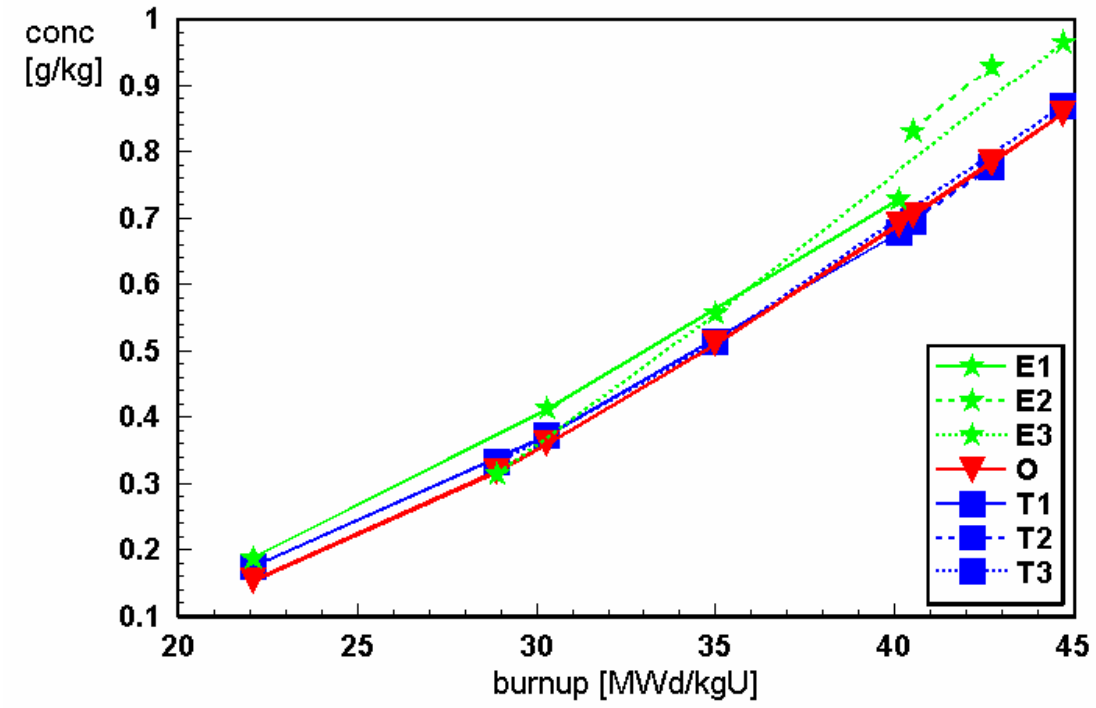


Fig.9b Dependence of Np237 on burn-up, experiment Novovoronezh 2

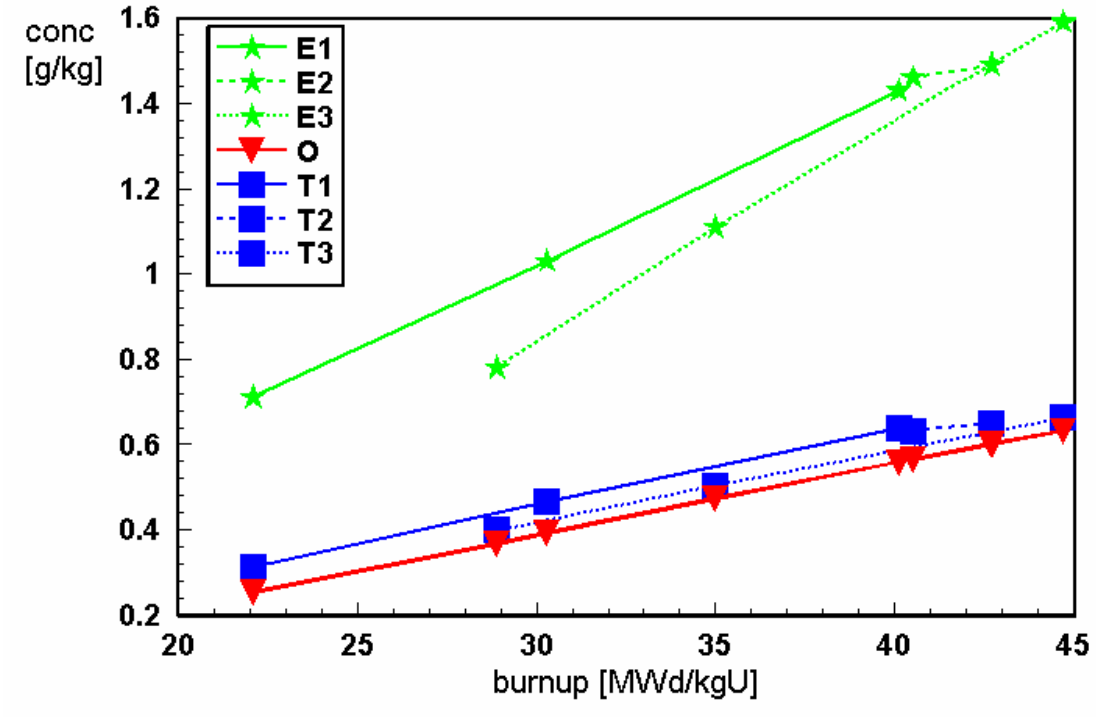


Fig.10b Dependence of Am241 on burn-up, experiment Novovoronezh 2

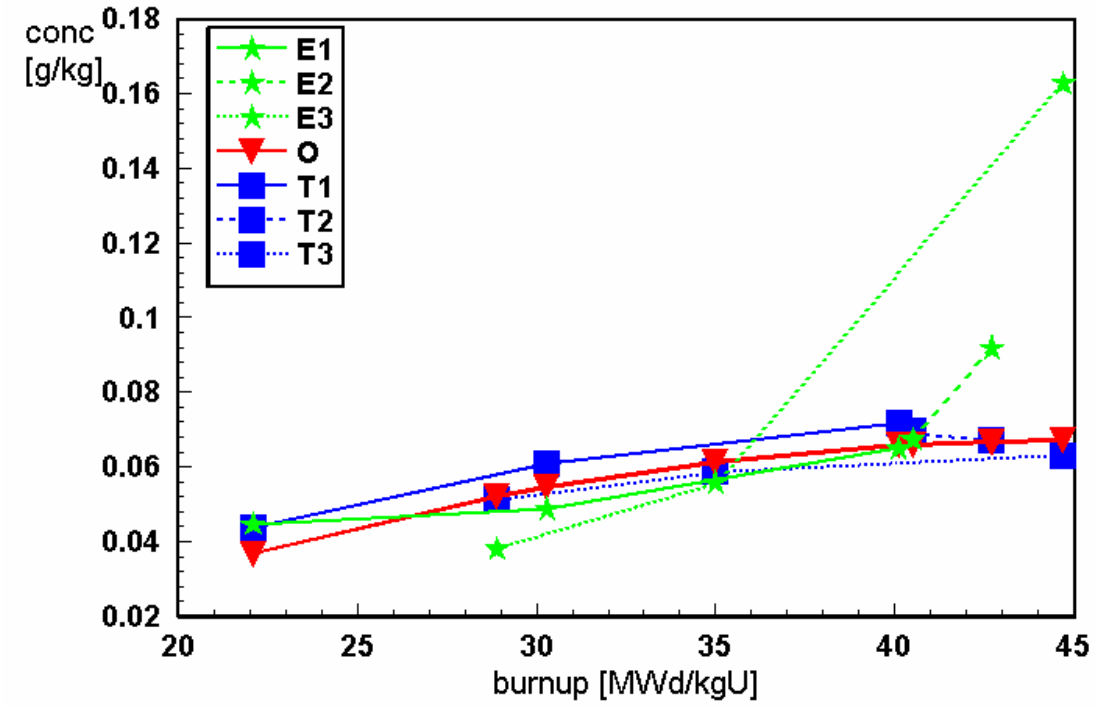


Fig.11b Dependence of Am242m on burn-up, experiment Novovoronezh 2

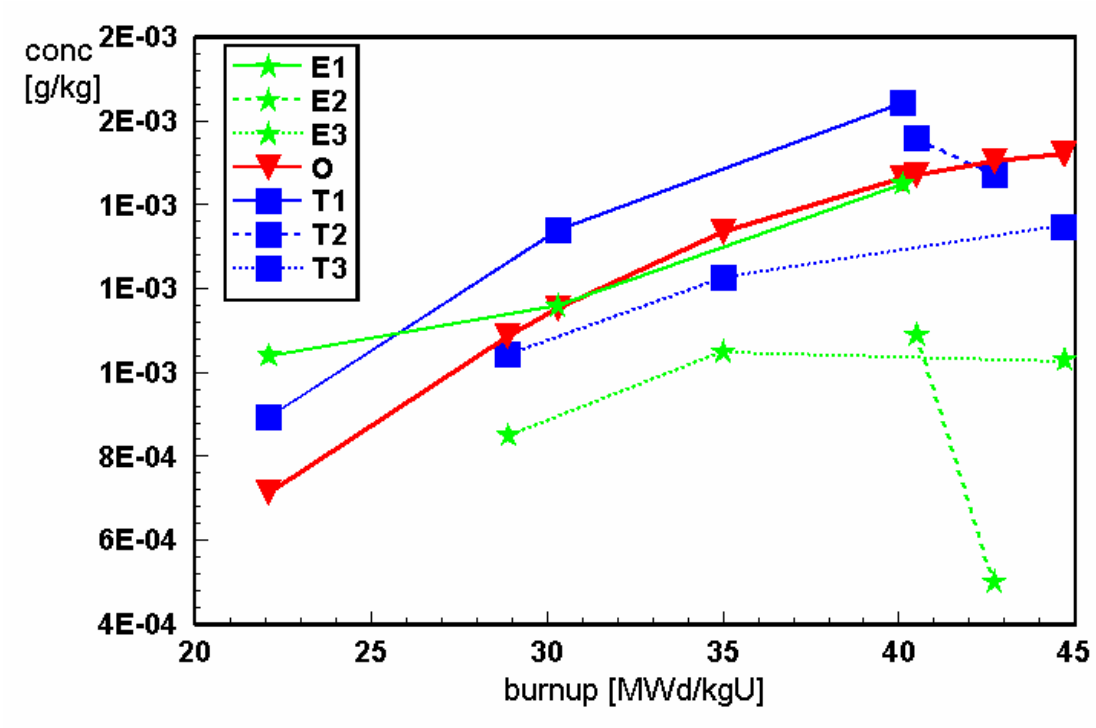


Fig.13a Dependence of Cm242 on burn-up, experiment Novovoronezh 1

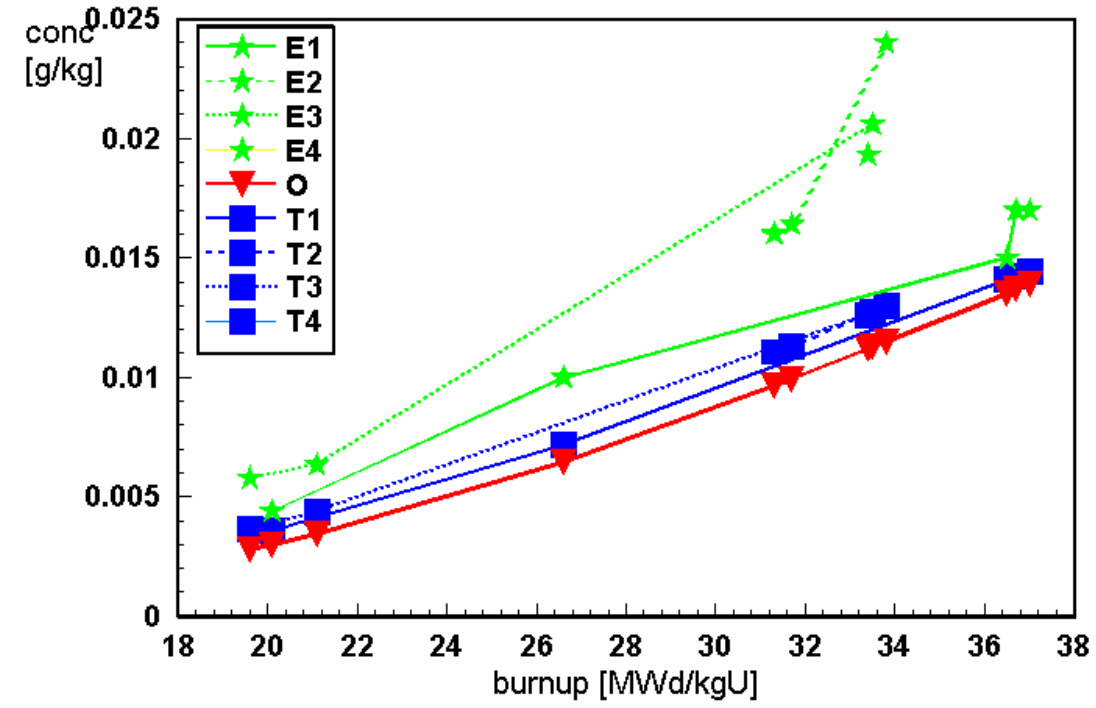


Fig.12a Dependence of Am243 on burn-up, experiment Novovoronezh 1

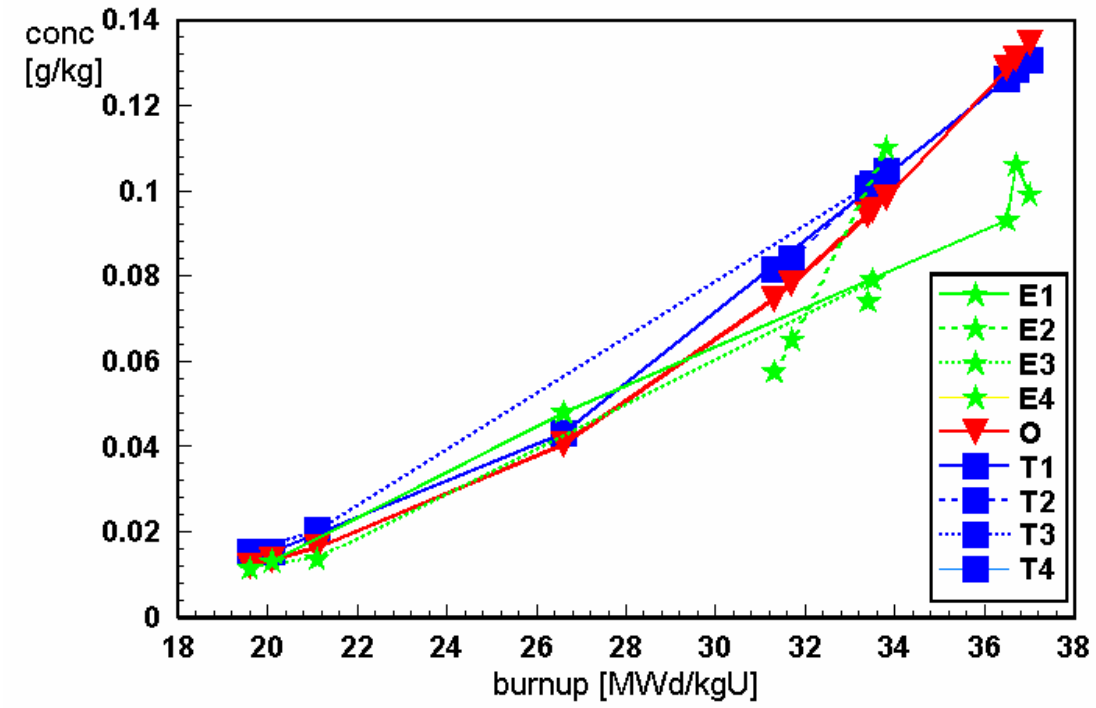


Fig.12b Dependence of Am243 on burn-up, experiment Novovoronezh 2

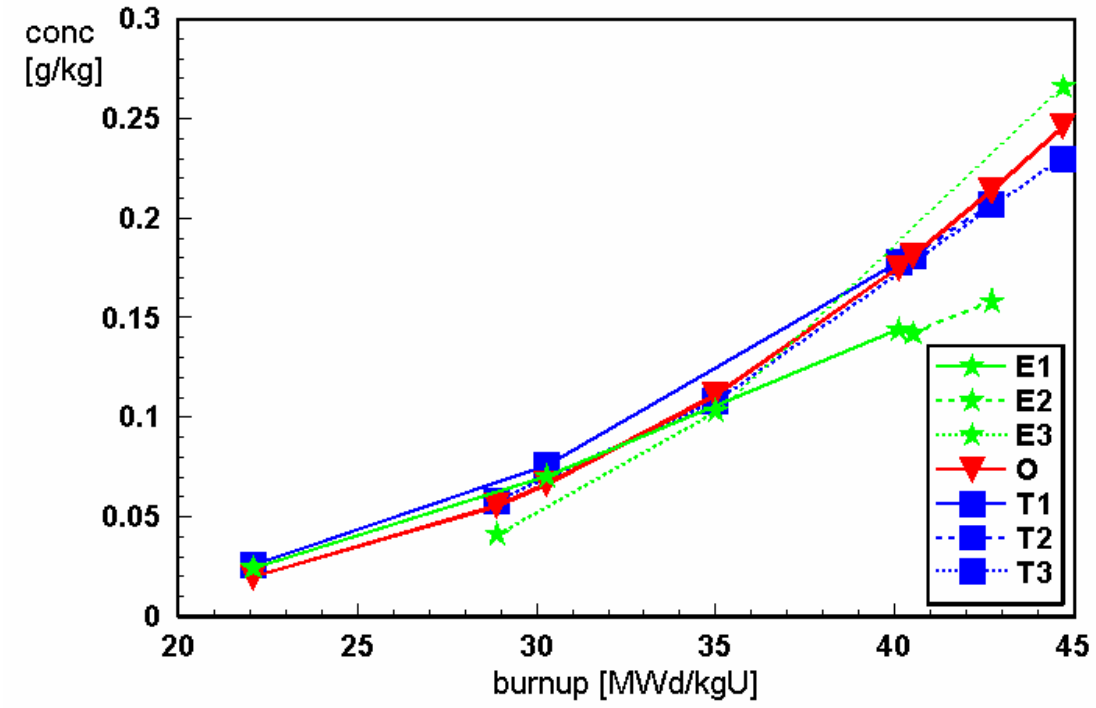


Fig.14a Dependence of Cm244 on burn-up, experiment Novovoronezh 1

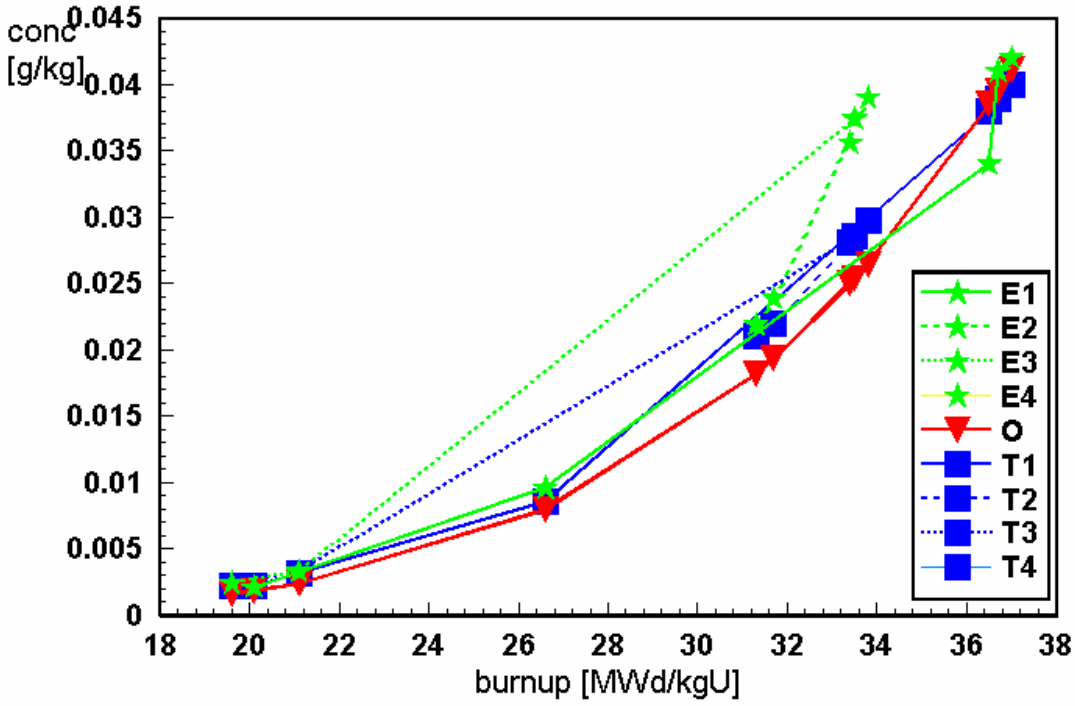


Fig.14b Dependence of Cm244 on burn-up, experiment Novovoronezh 2

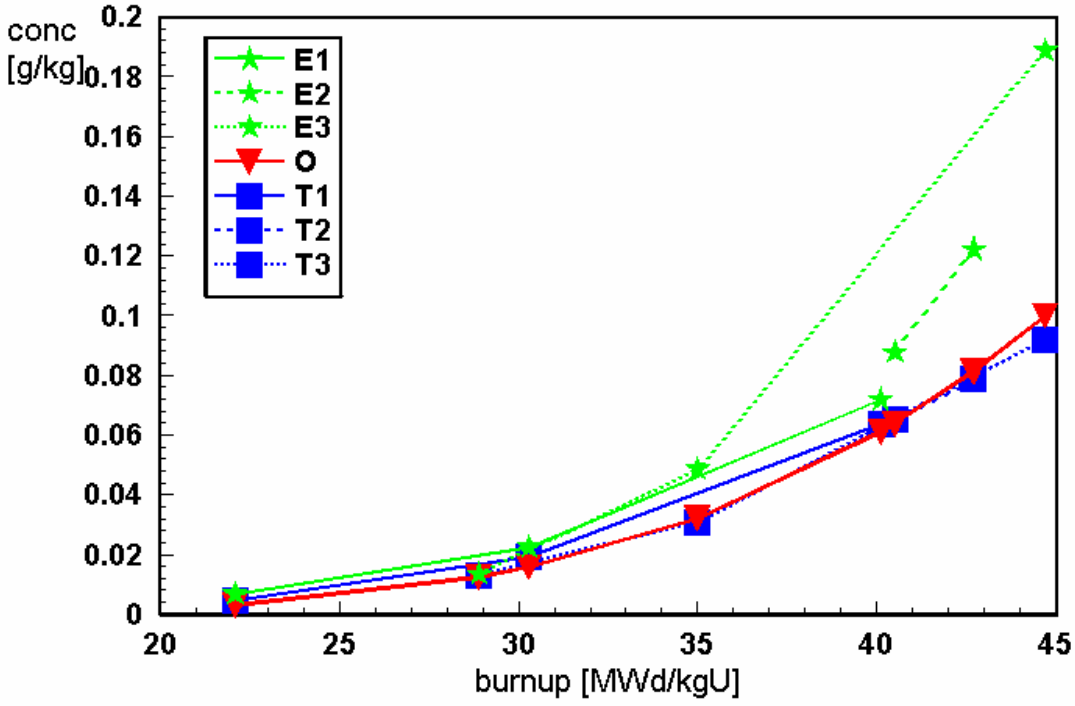


Fig.15b Dependence of Cm245 on burn-up, experiment Novovoronezh 2

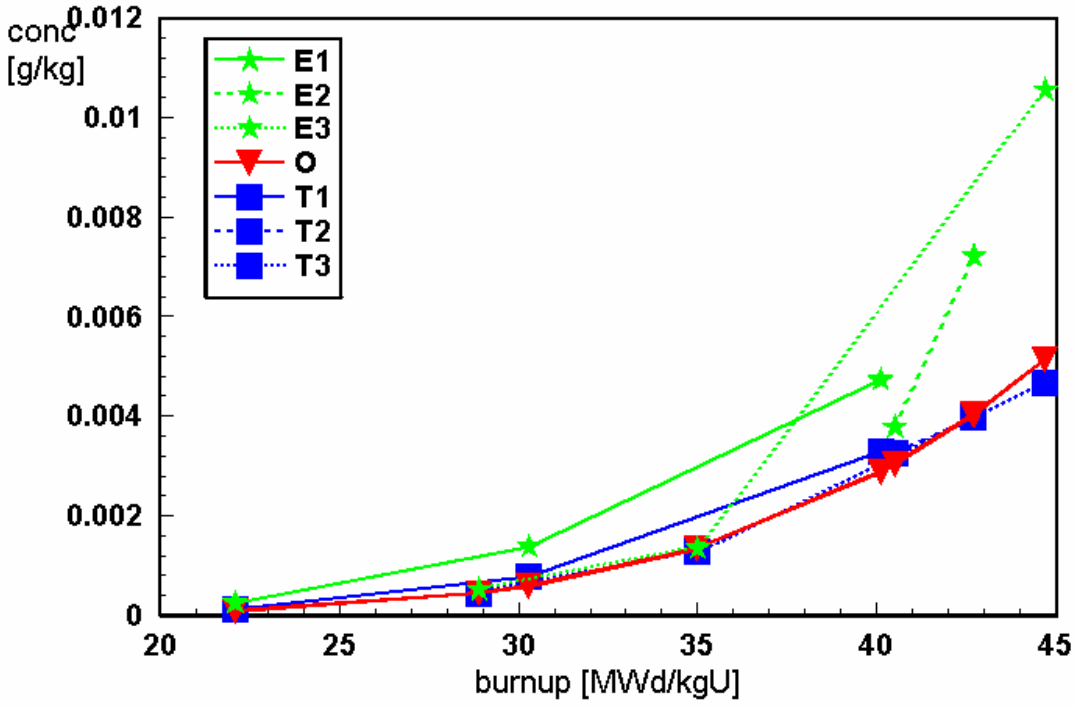
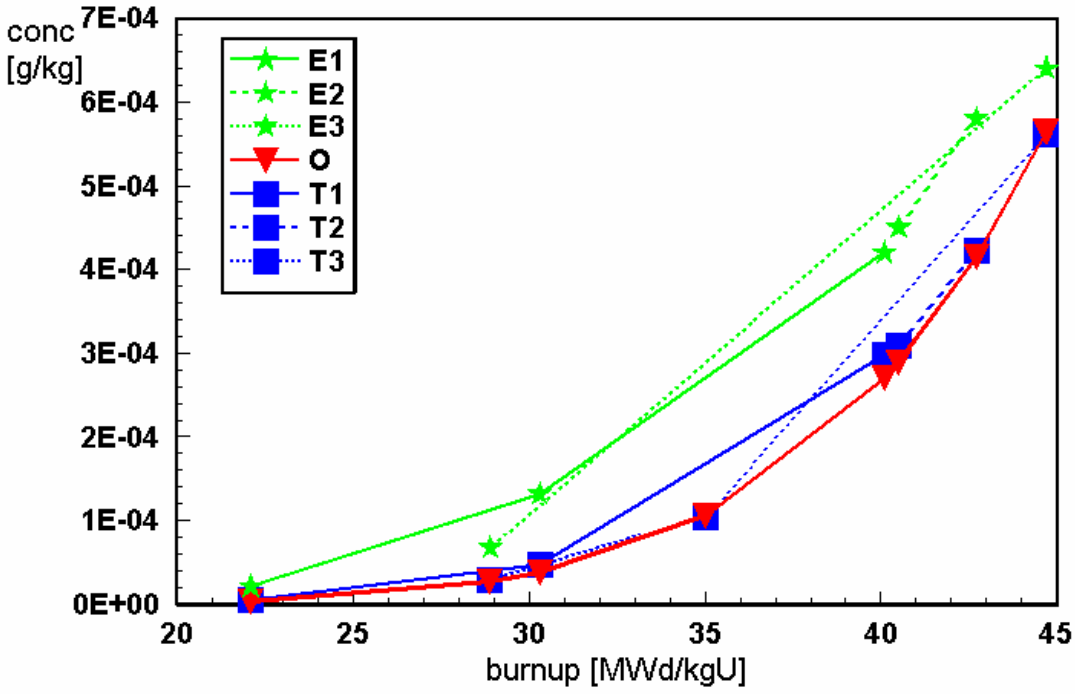


Fig.16b Dependence of Cm246 on burn-up, experiment Novovoronezh 2



BENCHMARKING OF SCALE 5 NUCLIDE PREDICTIONS FOR VVER-440 SPENT FUEL SAMPLES FROM THE NOVOVORONEZH REACTOR*

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Abstract

We report on comparisons between calculated results and nuclide assay measurements that involve two-dimensional simulation studies with the SCALE 5 code system. These studies consider VVER-440 spent-fuel measurements compared with predictions using the two-dimensional depletion sequence in the SCALE 5 TRITON module. The study addressed a range of actinides and fission products important to burn-up credit. Eight samples were studied whose burn-ups were between 22.6 and 47.5 GWd/t. This allowed us to examine the agreements between simulation and measurement as a function of burn-up over a meaningful range.

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Introduction

As part of an ongoing effort to benchmark the SCALE 5 system (1) comparisons were made with radiochemical analysis results from spent-fuel samples obtained from the Novovoronezh Unit 4 VVER-440 reactor (2). These samples achieved burn-ups ranging from about 22.6 to 47.5 GWd/t. The radiochemical measurements on the spent-fuel samples were performed at the Research Institute for Atomic Reactors (RIAR). A report covering the irradiation and the post-irradiation analysis was prepared by the Lawrence Livermore National Laboratory (LLNL) (2). At Oak Ridge National Laboratory (ORNL) burn-up simulations were carried out for these samples using the two-dimensional depletion analysis sequence TRITON in the SCALE 5 code system.

Experimental details

Four fuel rods in a VVER-440 assembly were used to obtain samples for analysis. These are identified as Rods 65, 67, 68, and 69 in Fig. 1. Eight spent-fuel samples were examined. Three samples were from Rod 65 and three were from Rod 69. Rods 67 and 68 each contributed one sample. All samples were of about one cm in length. All fuel rods had an enrichment of 3.6 %. Table 1 lists the samples and gives the rod number, the RIAR sample number, and the sample burn-up used in the simulations. These burn-ups were determined from the ORNL calculations using the measured ^{148}Nd in each sample.

The fuel rods were irradiated over the course of four reactor cycles. The irradiation history of the assembly for the four cycles was quoted in terms of the year and month for the beginning and end of each cycle. However, for each cycle, the effective number of days of irradiation was reported although there is uncertainty as to the downtime between cycles. We used the quoted effective-day values for the cycle lengths and assumed a downtime of 30 days between cycles. Power levels were assumed to be uniform throughout the four reactor cycles as detailed power variations during the cycles were not available. Power levels were adjusted so as to match total burn-up on the basis of the ^{148}Nd concentration. Measured concentrations had been adjusted to correspond with the end-of-irradiation date.

The design parameters for the VVER-440 fuel assembly were typical for that type of reactor. As explained, the burn-up chosen for each sample was the calculated value that matched the experimentally measured concentration for ^{148}Nd. Burn-up values in terms of kg fissioned per 10^3 kg of initial uranium were quoted in the LLNL report and are based on combinations of ^{148}Nd, $^{145+146}$Nd, and ^{137}Cs measurements. These were found to correspond with burn-up values significantly lower than the values estimated by us in matching ^{148}Nd concentration values.

Burn-up simulation calculations were carried out using the TRITON and NEWT codes that are part of the SCALE 5 system. The calculations employed the 44-group ENDF/B-V cross-section library and the NITAWL code was employed in the pin-cell calculations to account for self-shielding effects.

Fig. 1. The VVER-440 fuel assembly showing the location of the four rods (65, 67, 68, and 69) from which the experimental samples were obtained.

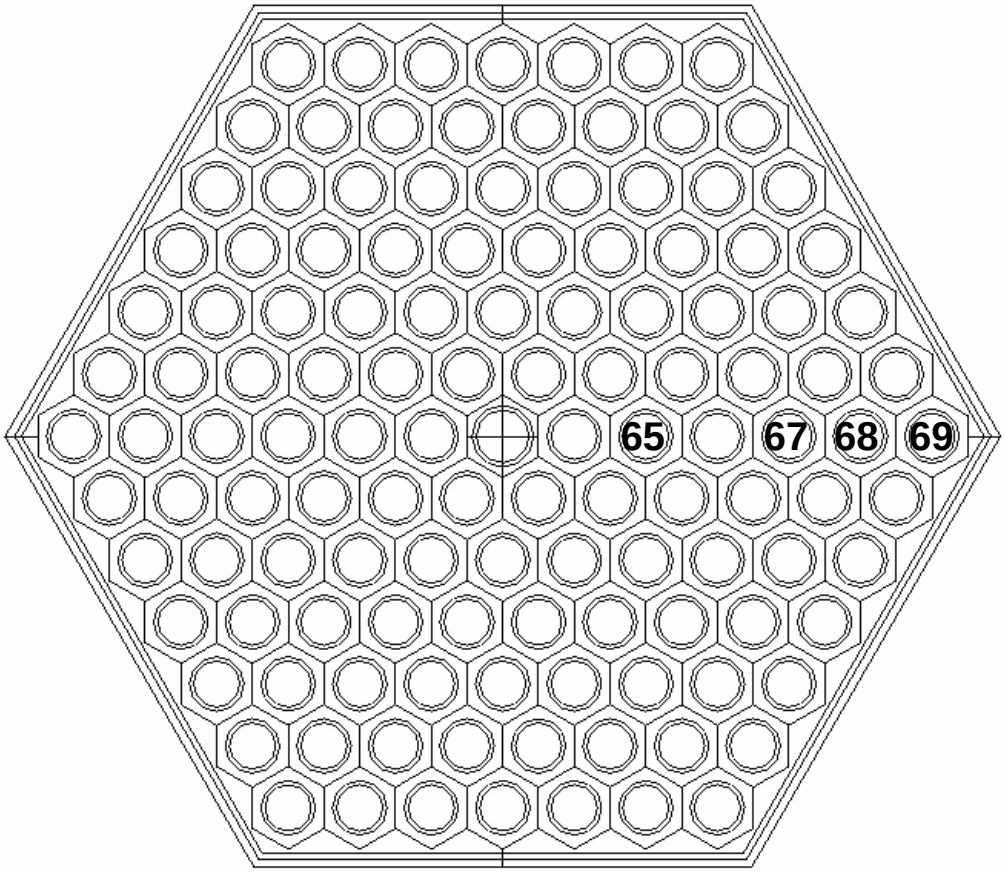


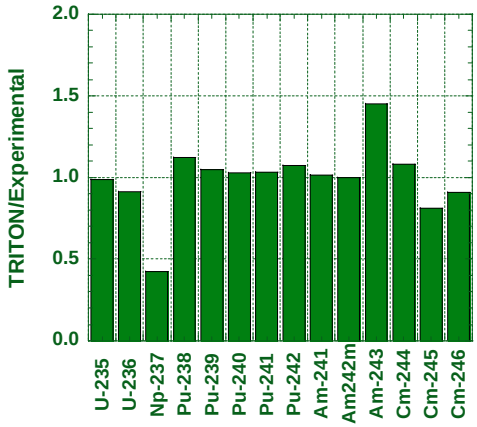
Table 1. Listing of irradiated VVER-440 samples

Rod number	Sample number	Burn-up (GWd/t)
65	21	42,650
65	69	31,789
65	182	22,599
67	149	43,457
68	162	44,983
69	79	47,485
69	57	35,972
69	135	30,185

Comparison results for actinide species

Figure 2 is a typical example of Calculated-to-Experiment (C/E) ratios for the actinides in the eight samples. The burn-up for this sample (no. 21) was 42.65 GWd/t. In general, there is reasonable agreement for the actinide species. For some individual cases agreement is poorer. Results are not shown for ^{234}U; agreements are poor because fresh-fuel concentrations were not reported for ^{234}U. For ^{237}Np, concentrations are consistently underestimated. Concentrations for the curium isotopes were found to be underestimated for high burn-ups (45 GWd/t and above).

Fig. 2. Calculated-to-Experiment (C/E) ratios for actinides in sample 21 from Rod 65. Sample burn-up was 42.65 GWd/t.



With eight samples that span a wide range of burn-ups, a much better sense of the trends can be obtained by comparing measurements and calculations as functions of burn-up. In Fig. 3, measured and calculated concentrations for ^{235}U and ^{239}Pu are plotted. Measured values are identified as RIAR and calculated values are identified that are associated with the four separate rods. Rods 67 and 68 have only one sample each (43.5 and 45.0 GWd/t) and they show very good agreement between measured and calculated values. Calculated points for Rods 65 and 69 have been fit with separate curves. The other actinide plots have a similar layout to that of Fig. 3; the measured values are identified as are the calculated values resulting from the TRITON burn-up simulations. Figure 4 shows comparisons for ^{240}Pu and ^{241}Pu. Figure 5 shows comparisons for ^{244}Cm. In the case of ^{244}Cm, the results are more or less independent of rod position and only one trend curve is shown.

Fig. 3. Measured values (RIAR) and calculated values for ^{235}U (left panel) and for ^{239}Pu (right panel) for 8 samples from 4 rods plotted as a function of sample burn-up.

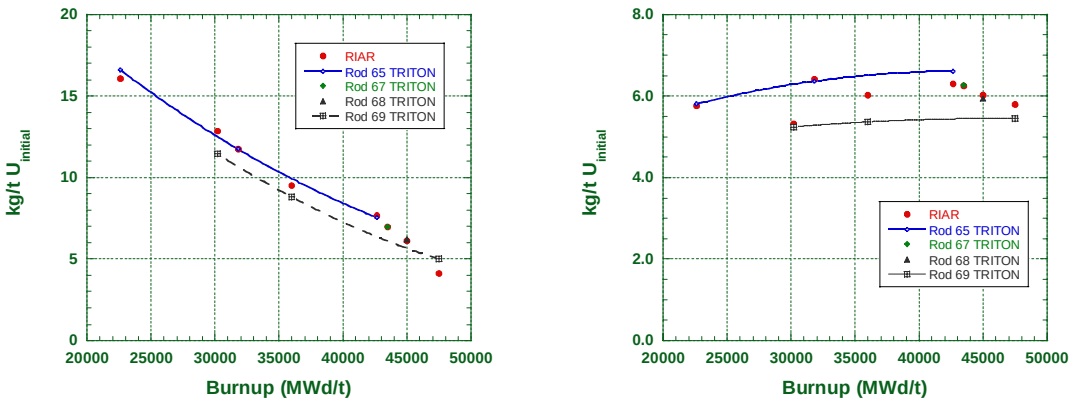


Fig. 4. Comparisons of measured and calculated values for ^{240}Pu (left panel) and ^{241}Pu (right panel).

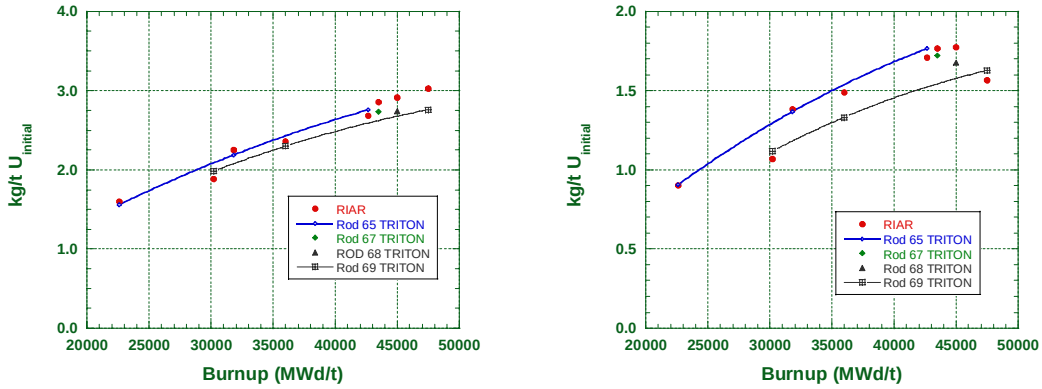
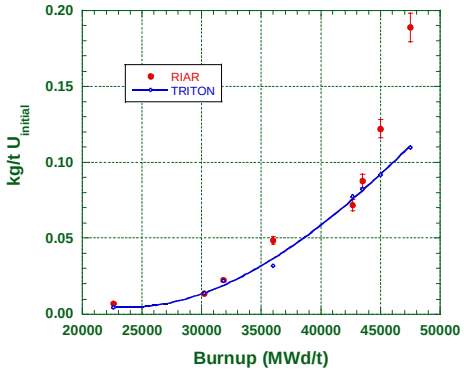


Fig. 5. Comparisons of measured and calculated values for ^{244}Cm. One trend line is shown for all the TRITON calculations because there are negligible differences between the fuel rods.



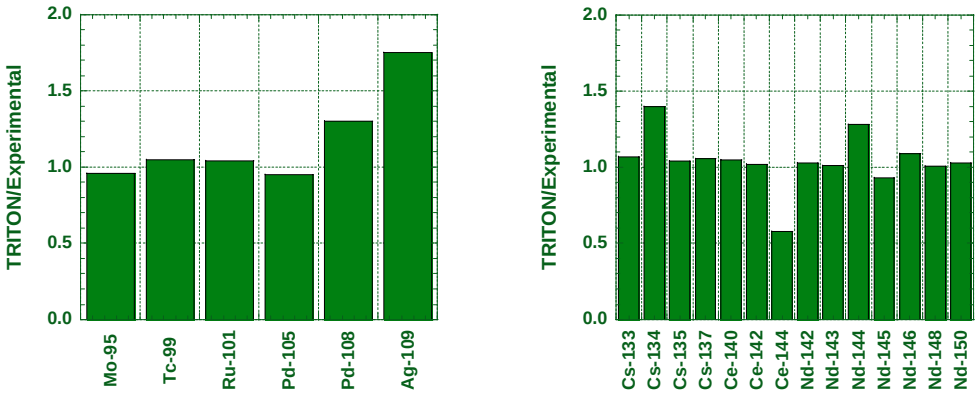
Comparison results for fission-product species

The fission products can be conveniently divided in to three groups: There are the so-called metallic species; ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{105}Pd, ^{108}Pd, ^{109}Ag; there are the caesium, cerium and neodymium species; and there are the samarium, europium and gadolinium species. The metallic species are treated separately because they are often difficult to dissolve and therefore challenging to measure accurately. Concentrations for the caesium, cerium, and neodymium isotopes are generally well predicted in burn-up simulation exercises. It is convenient to treat the third group of fission products (samarium, europium, and gadolinium) separately because many are strong neutron absorbers.

Figures 6 and 7 show C/E ratios for the three sets of fission products. One of the plots in Fig. 6 shows results for a sample from Rod 69 where the burn-up was 36.0 GWd/t; there is good agreement between measurement and calculation. Previous experience with international assay programmes demonstrates that ^{109}Ag is generally well predicted in LWR samples. The large deviation observed in this VVER-440 case suggests possible incomplete dissolution of Ag and/or failure to account fully for Ag in the residue. The other plot in Fig. 6 shows comparison results for the caesium, cerium, and neodymium isotopes for a sample (Rod 65) with a burn-up of 42.7 GWd/t. As expected, these species show reasonably good agreement. However, there are a few items worthy of note: C/E values for ^{144}Ce are low and for ^{144}Nd they are high. This is a general feature that was seen in all the samples and is inconsistent with past experience. Note that for this particular sample, the C/E value for ^{134}Cs is high

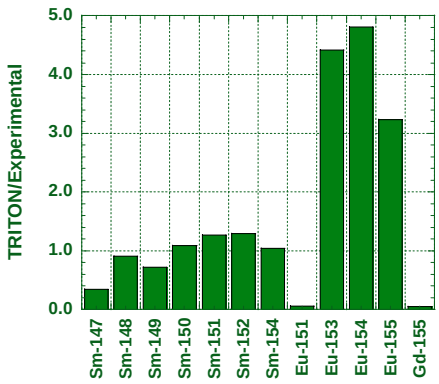
although, in general, other assay studies, for this species, show C/E values that tend to be on average 15% low (3, 4). But, among the eight samples in this study, the C/E values for ^{134}Cs show no consistency. The values are variable and have no trend. This strongly suggests a problem with the ^{134}Cs measurements.

Fig. 6. On the left are shown the C/E values for the metallic species in a sample (no. 57) burned to 36.0 GWd/t and on the right are shown the C/E values for Cs, Ce, and Nd isotopes in a sample (no. 21) burned to 42.7 GWd/t.



An example of the comparisons found for the samarium, europium, and gadolinium species is shown in Fig. 7. These comparisons are for a sample (Rod 69) with a burn-up of 30.2 GWd/t and they compare end-of-irradiation values from simulations and reported end-of-irradiation values determined from the measured values. The agreements for ^{153}Eu, ^{154}Eu, and ^{155}Eu are poor. We have generally found better agreement for these species in other comparison studies. C/E values for ^{147}Sm, ^{151}Eu, and ^{155}Gd are consistently under-predicted for all samples. The three species ^{147}Sm, ^{151}Eu, and ^{155}Gd are fed, respectively, by ^{147}Pm, ^{151}Sm, and ^{155}Eu. The half-lives for these decays are 2.6, 90, and 4.96 years so that there will be significant amounts of the parent species that will decay to the daughter species following the end of irradiation. The experimentally measured values will contain contributions from the decays of the parent species. However, the experimentally measured values were adjusted to the end-of-irradiation; but they will retain the contributions from their parent species whereas the values calculated for end of irradiation will not contain this contribution. Consequently, the calculated values will be low.

Fig. 7. C/E values for the samarium, europium, and gadolinium species. These results are for a sample (no. 135) subjected to a burn-up of 30.2 GWd/t. Subsequently, the ^{147}Sm, ^{151}Eu, and ^{155}Gd values were adjusted.



It was reported that the cooling time of the samples was 4518 days and predicted concentrations were therefore determined for ^{147}Sm, ^{151}Eu, and ^{155}Gd at that cooling time (however, it is doubtful that all concentration measurements were carried out on exactly the same day). These predicted concentrations were then compared to the measured values. This produced a C/E ratio of 0.98 for ^{147}Sm. However, both ^{151}Eu and ^{155}Gd are now considerably overestimated yielding C/E values on the order of the values for the other europium isotopes in Fig. 7. The improvement in the case of ^{147}Sm emphasises the importance of reporting the actual measured values and the date of the measurement rather than endeavouring to adjust for the end-of-irradiation date. The poor results for ^{151}Eu and ^{155}Gd indicate a measurement problem for these two nuclides (as in the case of ^{153}Eu, ^{154}Eu, and ^{155}Eu) in all probability.

Figures 6 and 7 show results that are, in general, typical for the samples. As in the case of the actinides, a better overall impression of the trends can be obtained from plots of C/E results as a function of burn-up. Some examples are shown in Figs. 8 and 9. Figure 8 contains plots of both ^{134}Cs and ^{137}Cs measured and calculated concentrations as a function of burn-up. As was mentioned, the ^{134}Cs results show no consistency. This possibly indicates a problem with the measurements. By contrast, in the case of the ^{137}Cs results, there is quite good agreement between measured and predicted values. In Fig. 9 we show results for ^{135}Cs and ^{144}Ce. The predictions are quite good for ^{135}Cs and one can see differences among the rods. However, the under-prediction that was noticeable for ^{144}Ce in Fig. 6 is seen here to be repeated for all the samples. But, in contrast to the ^{134}Cs case, the ^{144}Ce measured values show a consistent trend.

Fig. 8. Comparisons of measured and calculated values for ^{134}Cs (left) and ^{137}Cs (right).

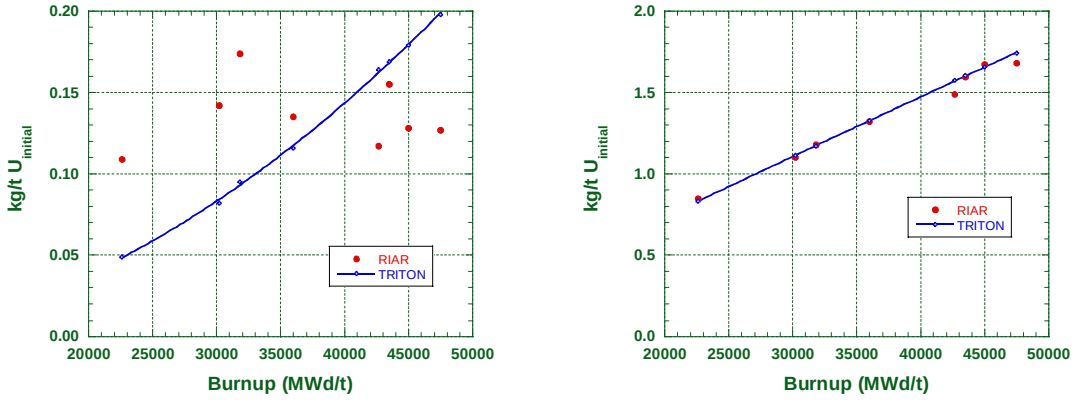
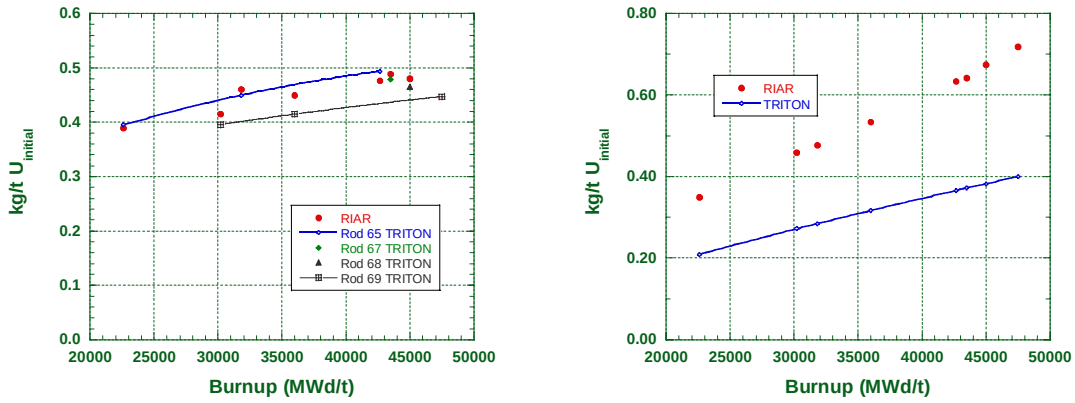


Fig. 9. Comparisons of measured and calculated values for ^{135}Cs (left) and ^{144}Ce (right).



Summary and conclusions

Using two-dimensional transport and depletion methods in the SCALE 5 system, burn-up simulations have been performed and results compared with measurements for a set of samples from a VVER assembly. Actinide concentrations are reasonably well predicted, but ^{237}Np predictions are noticeably poor. Some fission-product predictions show good agreement with measurements. However, isotopes of europium and gadolinium, in particular, show high C/E ratios. From our experience with other assay results (4) we expect much better agreement between calculations and measurements. The ^{134}Cs values are erratic. They show no particular trend and we suspect measurement problems.

We found that reporting measured values corrected to the end-of-irradiation date introduces much confusion and uncertainty to these kinds of studies. For the case of a species that is subject to simple decay this correction is acceptable but, for species that grow following discharge because of the decay of precursors, the approach may lead to the reporting of incorrect measured values. We would strongly suggest that all assay values should be reported as measured together with the measurement date for each particular nuclide. Also, beginning and end-of-irradiation dates (together with downtime between reactor cycles) should be reported so that simulations can be conducted effectively and true comparisons can be made.

Tables 3 and 4 give numerical summaries of the differences between Calculated and Experimental values for sets of actinides and fission products, respectively. The tables list the percentage difference between the Calculated and the Experimental value for each of the eight samples and they also give the average and standard deviation for that percentage difference.

Table 3. Percentage differences between calculated and experimental values for actinides in the Novovoronezh VVER-440 samples.

Calculated - Experiment (%) for Actinides												
Sample		182	135	69	57	21	149	162	79			
Burn-up (Gwd/t)		22.6	30.2	31.8	36.0	42.7	43.5	45.0	47.5			
Nuclide		Calculated - Experiment (%)									Av.	Std. Dev.
U-235		3.4	-10.9	-0.2	-7.4	-1.3	0.0	1.2	21.0		1	9
U-236		-14.1	-6.2	-9.9	-7.4	-8.7	-9.8	-10.3	-7.5		-9	2
Np-237		-61.5	-53.7	-58.3	-59.1	-57.6	-58.6	-59.0	-60.7		-59	2
Pu-239		0.6	-1.4	-0.6	-10.8	4.9	0.5	-1.4	-5.8		-2	5
Pu-240		-2.2	5.4	-3.0	-2.7	2.7	-4.2	-6.0	-8.8		-2	5
Pu-241		0.4	4.5	-1.1	-10.6	3.4	-2.7	-5.4	4.0		-1	5
Pu-242		-1.7	16.6	1.6	-0.9	7.3	-1.7	-5.9	2.1		2	7
Am-241		-8.3	20.0	18.1	-9.0	1.6	-7.1	-36.3	-67.8		-11	29

Table 4. Percentage differences between calculated and experimental values for fission products in the Novovoronezh VVER-440 samples.

Calculated - Experiment (%) for Fission Products												
Burn-up (GWd/t)		22.6	30.2	31.8	36.0	42.7	43.5	45.0	47.5			
Nuclide		Calculated - Experiment (%)									Av.	Std. Dev.
Ag-109		43	63	74	75	100	103	105	133		87	28
Cs-133		-2.7	-0.7	0.5	-1.0	7.2	2.1	0.9	5.4		1	3
Cs-134		-55	-42	-46	-14	40	9	40	56		-1	44
Cs-135		2	-5	-2	-8	4	-2	-3	0		-2	4
Cs-137		-2	1	0	0	6	0	-1	4		1	3
Ce-144		-40	-40	-41	-41	-42	-42	-43	-44		-42	2
Nd-143		5.0	0.0	3.4	-0.6	1.0	0.2	-1.2	-3.2		1	3
Sm-149		-18.2	-28.3	-13.4	-28.5	-10.4	-19.5	-22.9	-20.6		-20	6
Sm-151		33.1	27.0	54.6	34.1	76.7	59.3	60.9	58.9		51	17
Gd-155		221	286	318	333	398	384	394	435		346	70

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[1] *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluations*, ORNL/TM-2005/39, Version 5, Vols. I–III; Oak Ridge National Laboratory, Oak Ridge, TN; April 2005. Available from Radiation Safety Information Computational Center at Oak Ridge National Laboratory as CCC-725.

[2] *Radiochemical Assays of Irradiated VVER-440 Fuel for Use in Spent Fuel Burn-up Credit Activities*, L. J. Jardine, UCRL-TR-212202, Lawrence Livermore National Laboratory, April 2005.

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[4] *Strategies for Application of Isotopic Uncertainties in Burn-up Credit,* I. C. Gauld, NUREG/CR-6811, ORNL/TM-2001/257, June 2003.

COMPARATIVE ANALYSIS OF ORIGEN LIBRARIES FOR THE CALCULATION OF VVER SPENT FUEL POST IRRADIATION EXPERIMENTS

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Abstract

The ORIGEN-ARP computer code sequence and the SAS2H control module of the SCALE code package were validated in the framework of BOA No 409199-A-R4 (Task Order No. 1) for possible use in determining the isotopic composition of VVER and RBMK spent fuel. The validation was carried out by comparing the data received in the isotopic composition calculation with the results of experiments and calculations with other codes.

At the initial stages of this work, information was collected as required for testing the ORIGEN-ARP computer code and SAS2H control module of the SCALE package for determining the isotopic composition of VVER and RBMK spent fuel. These data are the results of experiments done to determine the composition of spent fuel and calculations performed using different codes. Recommendations were developed based on these sources.

After this information for testing was collected, the isotopic composition of VVER and RBMK spent fuel was calculated with the ORIGEN-ARP code sequence and the SAS2H control module of the SCALE package. The results were compared with experimental data and calculational benchmarks. General conclusions were made on the possibility of calculating the isotopic composition of VVER-440, VVER-1000, and RBMK-1000 spent fuel, and errors in calculating the concentrations of individual isotopes were indicated.

Acknowledgements

The authors would like to express their appreciation to the US Nuclear Regulatory Commission for its support of the project.

1 Introduction

The work reported here documents the testing of SCALE for calculating the isotopic composition of VVER spent fuel.

The SCALE [1] computer code, including program sequences ORIGEN-ARP[2], and SAS2H [3], is one of the most advanced tools with which isotopic content of spent nuclear fuel can be calculated. However, before using these codes to calculate isotopic content of spent fuel from VVER they must be tested and their applicability determined. Previously, the suitability of SCALE was successfully tested for calculation of nuclear safety (k_{eff}) of fuel systems for VVER and RBMK [4].

Experimental information was collected concerning of VVER-440, VVER-1000 spent fuel that can be used for testing the codes' ability to calculate the isotopic composition of spent fuel.

The experimental results considered the most valuable in terms of testing programmes. But most all of these are obsolete because they were obtained for fuel no longer used at NPPs. In addition, there are uncertainly errors because no data are available concerning the evaluation and validation of the methodologies and equipment used while measuring. In some cases, input data are insufficiently described and relate to the history of operating fuel.

At subsequent stages of testing the SCALE package, geometrical models of VVER spent fuels were developed for calculations with the SAS2H control module and ORIGEN-ARP using libraries of neutron-physical constants generated by SAS2H. The selection of geometrical models and fuel types for calculations was determined by the scope of information that was collected for testing. The geometric models incorporated the peculiarities of the SCALE package for calculating isotopic composition of spent fuel. The SAS2H control module uses several codes (ORIGEN-S is one of them) to model fuel in one-dimensional (1-D) approximation. A fuel assembly is described using two 1-D cylindrical models. The first model (Path A) simulates an infinite lattice of fuel pins; the second (Path B) approximates a fuel assembly by using homogenised fuel, clad, and water from the Path A model in one region plus other regions that simulate the central tube, water gap, and burnable absorber rods.

For VVER fuel, this lattices are hexagonal. However, several composition elements disturb their regularity.

For VVER-440 fuel, the components that disturb the regularity of the fuel pin hexagonal lattice are:

- central tube; water gap between assemblies;
- radial profiling with fuel pins of different enrichment; fuel assembly shroud.

For VVER-1000 fuel:

- central tube; water gap between assemblies; guiding channels;
- radial profiling with fuel pins of different enrichment; presence of burnable absorber in fuel pins.

Taking this into account, individual geometric models were developed for VVER fuel modelling; they describe the assembly as a whole (for preparing ORIGEN-ARP libraries) or as individual fuel pins (more reasonable for calculating experimental data).

Calculations were done with the 44groupndf5, 27burnuplib, 238groupndf5 and 27burnuplib (correction) SCALE code package standard libraries of neutron-physical constants.

2 Review and analysis of literature related to isotopic composition of spent nuclear fuel

2.1 A. Herman, W. Möller "Bestimmung der Abhängigkeit von Nuklidgehalten in VVER-440 Brennstoff nach rechnerischen und experimentellen Methoden", Kernenergie 27 (1984).

B. Bibichev, V. Mayorov, Yu. Protasenko, P. Fedotov. "Measuring fuel burn-up, and U and Pu content in FA for VVER-440 as regards activity of ^{134}Cs, and ^{137}Cs", Atomic Energy, Vol. 64, No. 2, p. 147 (1988) (in Russian).

- *Overall description*: Information presented is based on the results of measuring isotopic composition of spent nuclear fuel from VVER-440.
- *Error*: Error of fuel burn-up value and concentration of isotope - about 5-7%.
- *Comments*: Concentration of isotopes was measured quite long time ago, and for obsolete fuels. During experiment, only concentration of fuel isotopes, and actinides, was measured.
- *Possibility of simulation*: Possibility of simulation concerning conditions of fuel burn-up, and its subsequent cooling is limited. Fuel is of typical VVER design.

2.2 A. Stepanov, T. Makarova, B. Bibichev, et al. Determination of burn-up, and isotopic composition of SNF from VVER-440, Atomic Energy, Vol. 55, No. 3, Sept. 1983 (in Russian).

- *Overall description:* For measurement, four fuel assemblies were used from VVER-440 of Novovoronezh NPP. Initial enrichment of fuel was 3.3% and 3.6%. Selected assemblies were used during 1, 3, and 5 campaigns. Totally, isotopic content of 21 specimens with different burn-up levels was determined. Presents the data needed to calculate loading assembly during operation inside core: schemes of fuelling and refuelling, schedules of in-service loading, etc.
- *Error:* Error of fuel burn-up value and concentration of isotope – about 5÷7 %.
- *Comments:* Concentration of isotopes was measured quite long time ago, and for obsolete fuels. During experiment, only concentration of fuel isotopes, and actinides, was measured.
- *Possibility of simulation:* Possibility of simulation concerning conditions of fuel burn-up, and its subsequent cooling is limited. Fuel is of typical VVER design.

2.3 A. Tataurov, V. Kvator, Calculated-experimental studying nuclide composition of SNF from VVER-1000, and RBMK-1000, R&D Report, RNC "KI" (2002) (in Russian).

- *Overall description*: The results of isotopic content measurements of spent fuel in VVER-1000-type reactors are presented. For this measurement, one assembly was used from a VVER-1000 operating at Zaporizhya NPP. Its initial enrichment was 3.92%. This was operating during one campaign, and unloaded from the core in 1999. The isotopic composition of three specimens with different burn-up levels is presented. Information necessary to calculate loading assembly throughout its operation inside the core (fuel pattern and refuelling, reactor power during operation, etc.) is available.
- *Error:* Error for each value of burn-up and isotope concentration approximately 1–7%.

- *Comments:* Isotope concentrations were measured recently for a modern type of fuel. However, this assembly was operated for one campaign, and the achieved level of burn-up is insufficient. During the experiment, only fuel isotopes and actinide concentration were determined.

- *Possibility of simulation:* Conditions of fuel burn-up and subsequent cooling are described in detail. Fuel is of the known design.

2.4 Analysis of collected data

Collected information covers the following range of fuel burn-up (GW·Day/tU):

Reactor	***Experiment, fuel burn-up (GW·Day/tU)***
VVER-440	***8.7–54.8***
VVER-1000	***15.0–16.1***

Table 1. Sharing the information by isotope according to the source

Isotope	**Source (availability of information is presented as VVER-440/VVER-1000)**			
	Lit. 2.1.1	Lit. 2.1.2	Lit. 2.2	Lit. 2.3
^{234}U	-/-	-/-	X/-	-/X
^{235}U	X/-	X/-	X/-	-/X
^{236}U	X/-	-/-	X/-	-/X
^{238}U	X/-	-/-	X/-	-/X
^{238}Pu	-/-	-/-	X/-	-/X
^{239}Pu	X/-	X/-	X/-	-/X
^{240}Pu	X/-	-/-	X/-	-/X
^{241}Pu	X/-	X/-	X/-	-/X
^{242}Pu	X/-	-/-	X/-	-/X
^{241}Am	-/-	-/-	-/-	-/X
^{243}Am	X/-	-/-	X/-	-/X
^{242}Cm	X/-	-/-	X/-	-/-
^{244}Cm	X/-	-/-	X/-	-/X

3 Modelling isotopic composition of VVER-440 spent nuclear fuel

3.1 VVER-440 fuel assembly modelling

SAS2H employs two 1-D models, a fuel pin cell and a fuel assembly lattices. For VVER, this lattices are hexagonal. However, several composition elements disturb the regularity of fuel pin lattice: the central tube, the water gap between assemblies, and fuel assembly shroud (Figure 1) for VVER-440 fuel. These components have an impact on the neutron spectrum and fuel irradiation conditions, and their modeling should be accounted for in calculations.

The VVER-440 fuel assembly can be divided into three main zones:

1. fuel pins near the central tube (irradiation conditions are affected by additional water in the tube);

2. fuel pins in the assembly central part (their lattice is regular);

3. fuel pins on the assembly periphery (irradiation conditions are affected by additional water in the gap).

Experimental data on isotopic content were obtained for individual fuel pins. Their location in the fuel assembly lattice affects the isotopic composition. In this regard, modelling should account for the actual location of the fuel pin selected for analysis.

Table 2 lists the location of fuel pins sampled for further analysis. Figure 2 shows the numeration of fuel pins in the fuel assembly.

Table 2. Position of fuel pins sampled for further analysis in the fuel assembly (Figure 2)

No.	Burn-up, GW·d/tU	No. of fuel pin position	No.	Burn-up, GW·d/tU	No. of fuel pin position
1	8.7	97	11	32.6	63
2	11.7	97	12	33.1	63
3	13.3	58	13	34.8	107
4	13.9	63	14	34.9	64
5	14.3	97	15	35.2	63
6	16.9	63	16	38.1	25
7	20.4	64	17	38.3	25
8	21.0	25	18	38.6	25
9	22.0	64	19	54.7	63
10	27.7	25	20	54.8	63

3.2 Results of calculations

Table 3 show deviations of calculated isotopic concentration from experimental data given with use of different neutron-physical constant libraries. The best results are obtained when the “27burnuplib” library is used and the calculation model is corrected to incorporate the fuel pin location in the assembly.

Figure 1. Schematic of fuel pin lattice of VVER-440 fuel assembly
(1 – central tube; 2 – fuel pin; 3 – fuel assembly shroud)

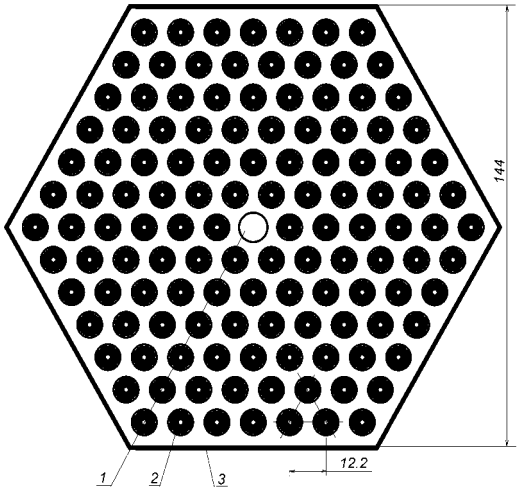
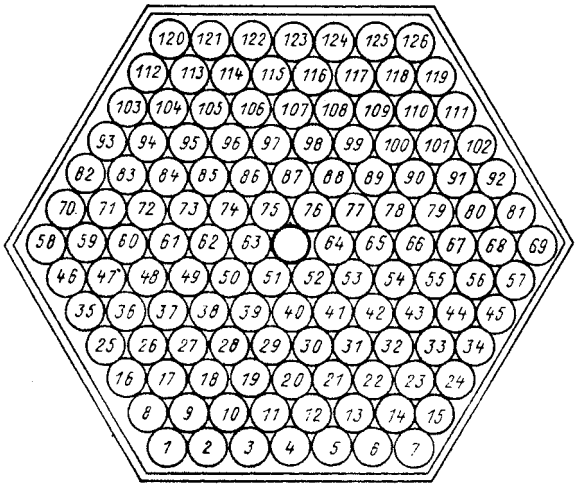


Figure 2. Numeration of fuel pins in VVER-440 fuel assembly



After the SAS2H control module was tested for calculating of individual isotopes content in VVER-440 spent nuclear fuel, the ORIGEN-ARP code was tested based on the same data. The ORIGEN-ARP employs the methodology for calculating isotopic content in SNF based on libraries of neutron-physical constants obtained with the SAS2H code. Calculations with the ORIGEN-ARP code were carried out after final correction of the fuel model for the SAS2H control module. Then, based on the results, libraries needed for the ORIGEN-ARP code were prepared and isotopic composition of SNF calculated for comparison with experimental and benchmark data. The “44groupndf5” library was used to calculate the ORIGEN-ARP libraries.

4 Modelling isotopic composition of VVER-1000 spent nuclear fuel

4.1 VVER-1000 fuel assembly modelling

The VVER-1000 fuel assembly (Figure 3) can be conditionally divided into seven main zones. The presence and geometry of these zones depend on a specific fuel design.

1. fuel pins near the central tube (their irradiation resulted from additional impact of water in the tube);
2. fuel pins near the guide tubes (their irradiation resulted from impact of extra water);
3. fuel pins with gadolinium;
4. fuel pins near those with gadolinium (their irradiation resulted from additional impact of absorber);
5. fuel pins in the FA central part (their lattice is regular);
6. fuel pins near the profiling zone (their irradiation resulted from additional impact of fuel pins of different enrichment from the fuel assembly periphery);
7. fuel pins on the FA periphery (their irradiation resulted from additional impact of water in the inter-assembly gap). In case of profiling, these assemblies have other enrichment.

Table 3. Average errors in calculating concentrations of individual isotopes in VVER-440 spent fuel

Isotope	Average error (calculation/bench.-1)*100%			
	SAS2H			ORIGEN-ARP
	27burnuplib (%)	27burnuplib (%) (correction)	44groupndf5	
^{234}U	-7.3	-5.9	-9.1	-10.2
^{235}U	5.1	-2.2	5.0	4.5
^{236}U	-2.1	-2.6	-2.1	-2.3
^{238}U	0.2	0.4	0.3	-0.2
^{238}Pu	-5.9	-9.8	-12.0	-12.7
^{239}Pu	10.1	-4.6	7.2	3.2
^{240}Pu	0.9	-5.6	4.9	-1.6
^{241}Pu	14.1	-5.8	7.3	18.9
^{242}Pu	-3.3	-0.1	7.1	17.5
^{243}Am	54.4	34.8	67.9	79.4
^{242}Cm	4.0	-1.2	-7.7	-2.6
^{244}Cm	82.2	78.2	121.4	133.2

To increase the accuracy of results, experiments should be modelled taking into account the actual location of the fuel pin selected for further analysis. The description of the experiment selected for testing the SCALE package indicates the position of individual fuel pins in the fuel assembly.

Table 4 lists the positions of fuel pins sampled for further analysis. Figure 3 shows the numeration of fuel pins in the assembly. For modelling experiments the fuel pin location listed in Table 4 were proposed:

Table 4. Position of fuel pins sampled for further analysis in fuel assembly

No.	Burn-up GW·d/tU	Fuel pin position in fuel assembly (Figure 4)
Experiment		
1	16.08	40
2	16.47	31
3	17.23	86

Figure 3. VVER-1000 fuel assembly lattice

(1 – fuel pin; 2 – guide tube for control rod or burnable absorber; 3 – central tube)

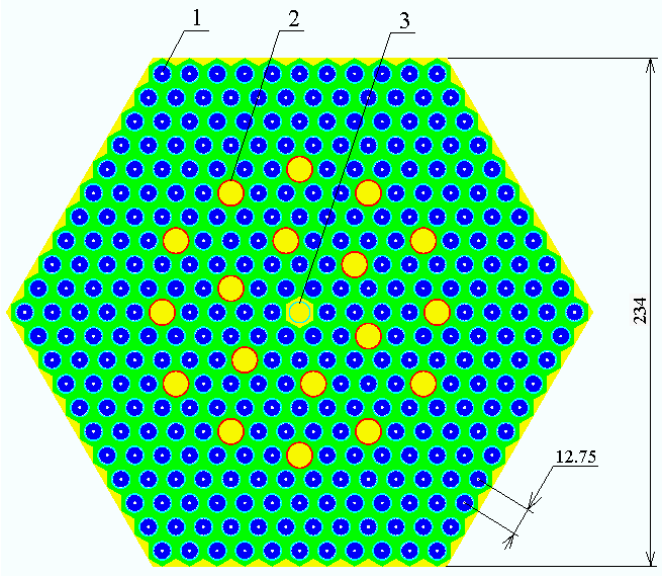
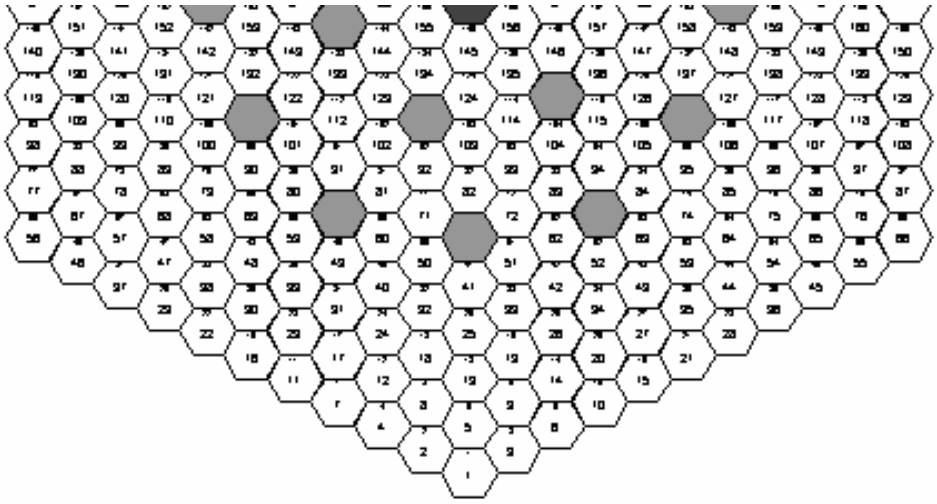


Figure 4. Numeration of fuel pins in VVER-1000 fuel assembly



4.2 Experimental calculations with SAS2H control module and ORIGEN-ARP

Table 5 show deviations of calculated isotopic concentration from experimental data given using different neutron-physical constant libraries.

The best results are obtained when the "27burnuplib" library is used and the calculation model is corrected to incorporate the fuel pin location in the assembly.

After the SAS2H control module was tested for calculating of individual isotopes content in VVER-1000 SNF, the ORIGEN-ARP code was tested based on the same data. ORIGEN-ARP employs the methodology for calculating isotopic content in SNF based on libraries of neutron-physical constants obtained with the SAS2H code. Calculations with the ORIGEN-ARP code were carried out after a final correction of the fuel model for the SAS2H control module. Then, based on the results obtained, libraries needed for the ORIGEN-ARP code were prepared and isotopic composition of SNF calculated for comparison with experimental and benchmark data. The "27burnuplib" library was used to calculate the ORIGEN-ARP libraries.

Average errors in calculating concentrations of individual isotopes obtained in experimental calculations are as follows:

Table 5. Average errors in calculating concentrations of individual isotopes in VVER-1000 spent fuel

Isotope	Average error (calculation/bench.-1)*100%			
	SAS2H			ORIGEN-ARP
	27burnuplib (%)	27burnuplib (%) (correction)	44groupndf5	
^{234}U	-16.0	-18.7	-17.1	-16.0%
^{235}U	-2.7	0.6	-2.4	-2.9%
^{236}U	-19.1	-18.5	-19.3	-19.3%
^{238}U	0.2	0.1	0.2	0.2%
^{238}Pu	-26.5	-29.4	-30.9	-27.4%
^{239}Pu	-7.9	-10.0	-10.8	-8.9%
^{240}Pu	-13.9	-18.9	-9.3	-17.2%
^{241}Pu	-16.1	-6.1	-20.1	-12.0%
^{242}Pu	-27.1	-20.5	-18.0	-24.7%
^{243}Am	-81.7	-80.0	-82.1	-81.2%
^{242}Cm	22.9	27.9	37.7	25.8%
^{244}Cm	-41.2	-41.1	-25.8	-40.6%

5 Conclusion

The isotopic composition of VVER-440, VVER-1000 spent fuel assemblies was calculated with the SCALE4.3(4.4a) code package SAS2H control module and ORIGEN-ARP code. Calculations were made with “44groupndf5”, “238groupndf5” and “27burnuplib” libraries. The obtained data were compared with results of experiments. Analysis of the results permits the following conclusions.

1. Both the SCALE SAS2H control module and ORIGEN-ARP code permit sufficiently accurate determination of isotopic composition of VVER-440 and VVER-1000 spent nuclear fuel. In general, calculations with the ORIGEN-ARP code demonstrate that the results deviate only slightly from calculations with the SAS2H control module.

2. Having compared the data, one can see that results obtained with the SAS2H code are the most accurate with regard to ORIGEN-ARP where location of experimental fuel pins and their operational conditions can be incorporated.

3. The greatest deviations in both VVER-440 and VVER-1000 are observed for ^{244}Cm.

4. Libraries “27burnuplib,” “44groupndf5,” or “238groupndf5” are recommended for calculating isotopic composition of VVER-440 and VVER-1000 SNF. But “27burnuplib” library does not contain data for calculating the concentration of gadolinium isotopes in spent nuclear fuel, so its application is reasonable only in case of old fuel types of VVER-1000.

5. New and accurate experimental data will decrease the uncertainties in calculating of isotopes concentration, which will provide an appropriate degree of depletion code validation.

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EXISTING ISOTOPIC INVENTORY DATA FROM PIE: AVAILABILITY, APPLICABILITY, QUANTITY AND QUALITY

Chairs: A. Santamarina, V. Chrapčiak

CURRENT STATUS OF SPENT FUEL ISOTOPIC COMPOSITION DATABASE SFCOMPO AND RELATED TECHNICAL DEVELOPMENT BY JAEA

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Abstract

In the field of criticality safety evaluation of spent nuclear fuels, Japan Atomic Energy Agency (JAEA), the former Japan Atomic Energy Research Institute (JAERI), has carried out continuous research and development.

In view of the fact that we had only a few opened post irradiation examination (PIE) data, JAERI had developed isotopic composition database SFCOMPO, which was transferred from JAERI to OECD/NEA in 2002, and it is now operated at the WWW site of OECD/NEA. This report summarises the PIE activities and related technical development by JAERI in past and current progress on PIE data evaluation by JAEA.

Introduction

In the field of criticality safety evaluation of spent nuclear fuels (SNF), Japan Atomic Energy Agency (JAEA), the successor institution of the former Japan Atomic Energy Research Institute (JAERI), has carried out continuous research and development. The effort includes (1) obtaining isotopic composition data of SNF irradiated in light water reactors by the post irradiation examinations (PIE) and (2) developing burn-up calculation code systems SWAT [1], SWAT2 [2] and new libraries for ORIGEN2 code [3] adopting the latest nuclear data libraries [4,5] at the times. The PIE data obtained by JAERI have been used worldwide for benchmark of calculation codes and libraries [6].

In view of the past experience that we had few opened PIE data, JAERI had developed isotopic composition database SFCOMPO. It was transferred from JAERI to OECD/NEA in 2002, and it is now operated at the WWW site of OECD/NEA. After the transfer of SFCOMPO to OECD/NEA, JAEA began to operate a closed version of SFCOMPO for internal usage and developed a function to edit and insert new PIE data into the database.

Because JAEA has no own PIE activities, we focused on more detailed analysis of PIE data taken in past. We studied on the value of burn-up because it is important to determine the amount of the exposure. By detail analyses of PIE data, the effect of neutron capture reactions of neodymium-147 and 148 on the burn-up was shown.[7] Moreover, the burn-up values of PIE samples from Mihama-3 and Genkai-1 PWR were revised by considering the effect of ^{147,148}Nd neutron capture reactions, effective fission yield of ^{148}Nd, and effective energy release a fission reaction.[8]

For burn-up credit introduction as well as reactor physics, PIE data are required to study and validate methods, computer codes and libraries. The effort to obtain new PIE data will be continued and international collaboration will be necessary.

Past PIE and related activities of JAERI

Activities of JAERI

JAERI has carried out several PIE since 1960s. Table 1 summarises PIE, which were conducted at JAERI and published. In these examinations, isotopic composition of actinides as well as fission products of UO_2 and UO_2-Gd_2O_3 fuel samples were measured.

Table 1. List of post-irradiation examinations performed at JAERI

Reactor	Type	Burn-up [GWd/t]	SFCOMPO
JPDR [9,10,11]	BWR	0.1-5	○[a]
Tsuruga-1 [12]	BWR	9-28	○
Mihama-3 [13]	PWR	7-31	○
Genkai-1 [13]	PWR	36	○
Not Opened [14][b]	PWR	22-39	not yet included
Takahama-3 [15]	PWR	14-47	○
Fukushima-Daini-2 [15]	BWR	4-44	○

a: Included in SFCOMPO
b: The name of reactor is not opened in the report [14].

Another activity of JAERI in the filed of PIE is development of isotopic composition database SFCOMPO. This activity was initiated in JAERI with agreement and support of taskforce on burn-up credit criticality safety of OECD/NEA in 1990's and two reports [16,17] were published. In order to distribute data, it was transferred to the environment of world wide web (WWW) server [18,19]. SFCOMPO was transferred from JAERI to NEA Databank to obtain and disseminate PIE data among NEA member countries in 2002, and it is now operated at the WWW site of OECD/NEA[20].

Collaborations with other institutes

Several PIE that were dedicated to evaluate mechanical properties and fuel behaviour of SNF have been carried out by collaborative works with other institutes in JAERI. Since many of them are controlled by the contact, isotopic composition data and related information are not opened. Concerning international PIE programme, JAERI participated in the ARIANE programme [21] to obtain PIE data of irradiated MOX fuel and high burn-up UO_2 samples.

Current PIE and related activities at JAEA

PIE

After the PIE of Takahama-3 and Fukushima-Daini-2, JAERI and JAEA have no their own PIE programme. Several contractual PIE were conducted for Japan Nuclear Safety Organization (JNES) by using facilities in JAEA [22]. In the PIE, data were taken from the view point of validation of reactor physics methodologies for newly developed BWR fuel of 9×9 type. PIE for studies on fuel behaviour of SNF are still carried out in JAEA by collaborative works with other institutes. However, the isotopic composition measurement is conducted only for major actinides, and such date will not be published.

PIE in JAEA such as done at JAERI in the past ten years seem to become difficult in future if we have no new PIE programme to obtain isotopic composition data of fission products and minor actinides as well as major actinides.

SFCOMPO

JAEA began to operate a closed version of SFCOMPO for internal usage and has developed a function to edit and insert new data into the database since FY2005. This function is based on common WWW system using perl script working in common gateway interface (CGI) of apache http server and SQL language to access relational database server in the network. Using the function, we have already archived data of ARIANE carried out from 1994 to 2000 into the "local" SCOMPO in JAEA. Negotiations among several institutes will be required when we transfer these new data to SFCOMPO in NEA for disseminate them to public.

Another technical development during 2000-2005

Since we finalised own PIE programme in JAERI after 2000, the current technical development of JAEA in the field of PIE and burn-up credit criticality safety evaluation consist from two subjects.

One is a study on the detailed chain analysis of neodymium isotope generation to be used in burn-up evaluation and re-evaluation of burn-up values of PIE samples taken at JAERI. This is because the value of burn-up is the most important indicator to determine the amount of exposure. The first report [7] shows that the effect of neutron capture reactions of neodymium-147 and -148 on the burn-up values. These effects are not negligible and these magnitude depends on the burn-up values and irradiation condition. The second report describes the re-evaluation of burn-up values of PIE samples from Mihama-3 and Genkai-1 PWR by considering the effect of ^{147,148}Nd neutron capture reactions, effective fission yield of ^{148}Nd, and effective energy release by a fission reaction.[8] These revised burn-up values should be adopted in SFCOPMO in NEA.

Another is evolution of SWAT2[2] code system to utilise continuous energy Monte Carlo Code MVP[23] in burn-up calculation and development of new criticality calculation code system [24] to integrate burn-up calculation code and following criticality evaluation codes. SWAT2 has been used to analyse UO_2 and MOX PIE samples obtained through the ARIANE programme to demonstrate its ability to treat assembly geometries. It will be the core part of the new criticality calculation code system, which has a controller to drive codes through WWW browsers and evaluation of isotopic composition of SNF is conducted by SWAT, SWAT2 and other codes such as ORIGEN2 with new cross section libraries based on JENDL-3.2[25] or JENDL-3.3[26].

Conclusion

As we described in the paper of review session [27], potential demands for adoption of BUC and improvement of related techniques has been still high in Japan. For this purpose, new PIE data, especially for fission products data, which is not sufficiently obtained in past activities, will be required. And PIE data are not only required by BUC implementation. For example, evaluation of reactor physics methodologies and validation of nuclear data also require PIE data. This concludes that continuous efforts to obtain PIE data are necessary for future nuclear energy development. International collaborative works would be important because to conduct PIE just by ourselves will become difficult and many countries share common interests and requirements in such data.

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SPENT FUEL ISOTOPIC VALIDATION ACTIVITIES IN THE UNITED STATES: STATUS AND LESSONS LEARNED

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Abstract

Oak Ridge National Laboratory (ORNL) is leading an effort to acquire and evaluate experimental isotopic assay data for the purpose of establishing the technical basis for the credit of fission products in criticality safety analyses for spent fuel storage and transportation facilities. An important component of this effort is acquisition of fission product assay data that can be used to validate computer code predictions of isotopic compositions of spent nuclear fuel. This paper presents a summary of the actinide and fission product isotopic assay data previously evaluated in the United States and current ORNL activities to obtain and evaluate data from public and proprietary programmes. Important lessons learned during evaluation of measurement data from domestic experimental programmes and from participation in several international programmes are presented.

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Introduction

In September 2002, the US Nuclear Regulatory Commission (NRC) issued Interim Staff Guidance 8, Revision 2, (ISG-8) providing guidance on the application of burn-up credit in criticality safety analyses involving pressurised-water reactor (PWR) spent fuel in transportation and storage casks [1]. Areas addressed by the guidance include experimental data needed to support burn-up credit and the general approach to take for establishing the bias and uncertainty in the analysis codes. This guidance requires the use of measured PWR isotopic data to validate fuel depletion calculations.

A review of available PWR isotopic measurements and uncertainties published in 2003 [2] identified 56 spent fuel samples with actinide measurements [2] suitable for use in code benchmark studies. The same review found that the majority of these experiments did not include measurements of fission products of importance to burn-up credit. Of the 16 most important fission products identified, 3 had no measurements at all, and 5 isotopes had 4 measurements or less. The paucity of fission product measurements makes a statistical evaluation of isotopic bias difficult and leads to large bias uncertainty. Based on the availability of isotopic assay data at the time of the ISG-8 revision in 2002, the guidance endorsed burn-up credit only for actinide compositions.

Several studies performed more recently (e.g. [3]) have demonstrated that actinide-only burn-up credit will allow loading and transportation of only a small fraction of the existing spent fuel inventory using high-capacity transportation casks in the United States. Given the availability of appropriate data for validation, the most significant opportunity for increasing the accuracy and expanding the range of burn-up credit is to include reactivity credit for both actinides and fission products in spent fuel. Oak Ridge National Laboratory (ORNL) is presently leading an effort with the Department of Energy Office of Civilian Radioactive Waste Management (DOE/RW), NRC, and the Electric Power Research Institute (EPRI) to acquire and evaluate new experimental data necessary to extend burn-up credit to include fission products.

This paper describes earlier efforts evaluating spent fuel isotopic assay data and discusses the status of current activities that focus primarily on acquiring isotopic assay data for the fission products. The ORNL experiences and lessons learned from evaluating more than 100 PWR spent fuel samples are summarised in this paper.

Experimental programmes

Isotopic assay measurements for 56 spent fuel samples from PWR fuel assemblies have been evaluated by ORNL [2]. The results have been used to validate computer code predictions and determine the code bias and uncertainties associated with the calculations. Further, the results have been applied to criticality calculations to estimate the effects of the predicted isotopic composition uncertainties on the neutron multiplication factor for a typical transport cask [2]. The fuel experiments evaluated in this earlier work included samples from the following PWR reactors:

- Calvert Cliffs Unit 1 (ATM-103, ATM-104, and ATM-106)
- H. B. Robinson Unit 2 (ATM-105)
- Turkey Point Unit 3
- Trino Versellese
- Yankee Rowe
- Obrigheim KWO
- Takahama Unit 3

Isotopic assay measurements on these samples provided a database with a sufficient number of samples and range to provide the technical basis for expansion of ISG-8 Revision 2 to allow burn-up credit for PWR fuels with up to 5 wt.% initial enrichment and burn-ups extending to 50 GWd/MTU.

The measurement data for these samples are publicly available and have also been compiled as part of the Isotopic Compositions Database System on Spent Fuels (SFCOMPO: http://www.nea.fr/html/science/wpncs/sfcompo/). Note that the database contains other samples that have not been evaluated by ORNL at this time due to concerns related to the quality and consistency of the data [4]. The measurements from the Approved Testing Material (ATM) programmes were performed in the 1980s and early 1990s to establish a high-quality database of irradiated fuel measurements. Because many of the measurements were performed in the 1980s, the enrichment and burn-up for the samples is considerably less than that of modern fuels. These measurements, and many others that are publicly available, lack comprehensive data for fission products. Of the experiments described, only the Calvert Cliffs ATM-104 and Takahama samples include measurements for any fission products of interest to burn-up credit, and the fission product measurements for these samples are relatively incomplete.

To develop a technical basis to expand burn-up credit to fission products, additional fission product assay data are being acquired and evaluated. The experimental programmes presently being evaluated that contain fission product measurements for PWR fuels are listed in Table 1. The data from these programmes are being evaluated for applicability to burn-up credit; a process that involves a careful review of the experimental procedures, measurement uncertainty, consistency of the measurement data, and comparisons with other independent measurements and calculations involving fuels of similar type.

Table 1. Experiments being evaluated for fission product isotopic validation data

Reactor (country)	Reactor type	Assembly design	Measurement laboratory (country)	Enrichment (wt.% ^{235}U)	No. of samples	Burn-up (GWd/MTU)
Takahama (Japan)	PWR	17 x 17	JAERI (Japan)	4.11	10	14.3 – 47.3
Calvert Cliffs (USA)	PWR	15 x 15	PNNL (USA), Khlopin Institute (Russia)	2.45, 2.72 2.45, 3.04	5 4	18.7 – 46.7 27.4 – 44.3
Gosgen (Switzerland)	PWR	15 x 15	SCK-CEN/PSI/ITU/CEA (Belgium)	4.3 3.5, 4.1	2 3	47.2, 68 29.1 – 59.7
Vandellós II (Spain)	PWR	17 x 17	Studsvik Nuclear (Sweden)	4.5	7	42.9 – 73.8
Neckar GKN II (Germany)	PWR	18 x 18	SCK-CEN (Belgium)	3.8	1	54
Novovoronezh (Russian Fed.)	VVER	Type 440	RIAR (Russia)	3.59	8	23 – 47
TMI-1 (USA)	PWR	15 x 15	ANL (USA)	4.013	11 (1 rod)	45.7 – 52.1
TMI-1 (USA)	PWR	15 x 15	GE-VNC (USA)	4.657	8 (3 rods)	25-3 – 31.4
Gravelines (France) EdF/ COGEMA/CEA	PWR	17 x 17	CEA (France)	4.5	5	39 - 61
H. B. Robinson (USA)	PWR	15 x 15	ANL (USA)	2.89	2	74 – 76

The Takahama and Calvert Cliffs measurements are both publicly available. The Three Mile Island Unit 1 (TMI-1) measurements are also publicly available [5]; however, some details of the fuel design are presently not published. Similarly, the H. B. Robinson measurements (different than those published previously) will be made public but may involve fuel design information considered proprietary.

Measurements of VVER 440 fuel samples from the Novovoronezh reactor are published with the related design and operating history data as part of the International Science and Technology Center (ISTC) project [6]. The VVER reactor and fuel design is distinct from western PWR designs, e.g., triangular pitch rods, smaller fuel diameter, tighter fuel pitch, etc. Therefore, establishing the similarity and applicability of the measured VVER isotopic data to the validation of computer codes for western PWR fuel designs is an essential task under the data evaluation programme. This task is being done using TSUMANI sensitivity/uncertainty analysis tools developed as part of the SCALE computer code system [7].

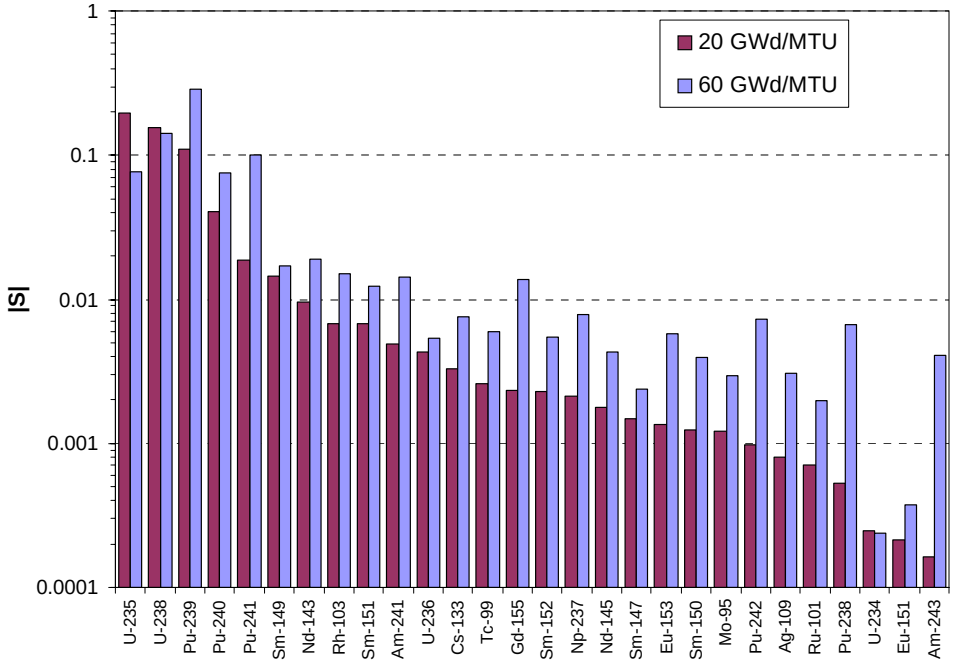
All other experimental programmes listed in Table 1 are proprietary and generally involve a data nondisclosure period of several years after completion of the programme. The Gosgen and Neckar GKN reactor samples have been measured as part of the international ARIANE, MALIBU, and REBUS proprietary experiment programmes co-ordinated by Belgonucleaire in Belgium. The Vandellós samples were measured as part of a proprietary high-burn-up fuel validation programme in which ORNL is participating with the Spanish organisations CSN, ENUSA, ENRESA, and other international partners. Finally, ORNL is negotiating to evaluate proprietary isotopic assay data measured in France involving fuel from the Gravelines reactor, data owned by COGEMA, CEA, and EdF.

The current effort to validate for fission products is focusing on six of the major fission product neutron absorbers in typical spent fuel: ^{143}Nd, ^{149}Sm, ^{103}Rh, ^{151}Sm, ^{133}Cs, and ^{155}Gd. These six isotopes represent approximately 75% of the negative reactivity associated with all fission product absorption (volatile isotopes being excluded). Isotopic data for other important fission products is also being evaluated including ^{152}Sm, ^{99}Tc, ^{145}Nd, ^{147}Sm, ^{153}Eu, ^{150}Sm, ^{99}Mo, and ^{109}Ag. The relative importance of all nuclides (actinides and fission products) in typical spent light-water reactor fuel is illustrated in Fig. 1.

Most of the experimental programmes listed in Table 1 (with the exception of the older Calvert Cliffs and Takahama measurements) include measurements for many of the fission products important to burn-up credit applications. Because most of the newer experimental programme involve modern fuel types, the samples have relatively high enrichments and burn-up values as compared to the samples evaluated earlier that were used to provide the technical basis for actinide-only burn-up credit. In fact, the availability of adequate fission product data for low-enrichment and low-burn-up fuels is a concern; particularly since the fission product measurements for the lower burn-up Calvert Cliffs samples include relatively few nuclides of importance to burn-up credit. Other considerations are that the samples represent the range of fuel designs, and the enrichment, burn-up, and operational parameter space of the application.

The following sections provide a brief summary of our experiences from participating in the international experimental programmes, evaluating the measurement data for the purposes of code validation, and some of the lessons learned.

Figure 1. Relative importance of nuclides in spent nuclear fuel to criticality safety (ranking in order of descending relative importance for a fuel burn-up of 20 GWd/MTU).



Experimental uncertainties and measurement error

Experimental uncertainty is an important consideration in evaluating the quality of isotopic assay data. The radiochemical analysis techniques required to measure specific isotopes, sometimes at very small concentrations, are extremely complex. There is no calibration *standard* available for spent nuclear fuel that can be used to quantify the accuracy of radiochemical assay procedures and measurement instruments. The reliable determination of measurement accuracy is not easily accomplished without some kind of independent analysis of duplicate samples.

Experimental error is sometimes expressed as the variance of repeated measurements (precision), i.e. repeatedly running the dissolved fuel sample through the instrument and measuring the variance of the results. However, this does not provide an accurate measure of error. Errors can be introduced in all phases of measurements: from cutting of the samples from the fuel rods, weighting the samples, techniques used to dissolve the fuel, chemical separation procedures, preparation of the samples for counting, isotopic dilution, mass spectrometry measurements, and measurements of the prepared solutions and the undissolved residues from the dissolution. There is also a "human factor" element that must also be considered in the handling and preparation of the samples and operation of the instruments. Therefore, any estimate of error made from repeat measurements alone is likely to underestimate the true uncertainty and will not account for measurement bias caused by instrument drift, radiochemical procedures, isotopic interference, etc. Even if duplicate samples are cut from the fuel rod and analysis, it is likely not possible to completely account for the uncertainty and bias unless independent analysis procedures and instruments are used.

The Belgonucleaire programmes have employed a system of independent laboratory cross-check measurements to ensure high confidence in the measurement results and measurement errors. Such cross checks must be considered an important component of a modern experimental isotopic assay

programme because of the complexity of the measurements and difficulty in confirming the reliability of measurements by other means. The importance of having these cross checks was reaffirmed in the ARIANE programme. The programme used three independent laboratories, and cross checking of duplicate samples was performed for a subset of samples. Comparison of the duplicate samples indicated that estimates of measurement uncertainty varied widely between different laboratories, even when similar techniques were used. In several cases cross-check measurements did not agree within the range of estimated error by the respective laboratories, and in other cases systematic biases in laboratory results were identified. The lesson from this experience is clear: all measurements and estimates of uncertainty must be carefully and critically reviewed. The use of high-accuracy instruments does not guarantee high-accuracy results. Any error in the measurements will be directly reflected as code uncertainty when the data are used for the purposes of computer code validation (i.e., a code can only be validated to an accuracy that is no better than the accuracy of the data).

The importance of interlaboratory cross checks, or qualification of laboratory methods against independent laboratories with an established track record and experience, can not be understated as a method to ensure a high degree of confidence in the results. In cases were cross checking is not possible or available, it is important to (1) thoroughly evaluate the measurement data for self-consistency using multiple samples and internal cross checking of data, (2) to compare data against other measurements for fuel samples having similar properties, and/or (3) to compare results against predictions for nuclides that have been established to be reliably calculated. Alternatively, duplicate measurements of a similar sample, cut from an adjacent location in the same rod, may provide an improved measure of overall accuracy, particularly if the measurements can be done at different times by different staff, using independent methods/instruments as available.

Analytical measurement methods

Measurement costs can vary considerably depending on the type of analysis performed. The costs can depend on whether or not chemical separation and/or isotopic dilution (ID) are required. The most commonly used methods for mass spectrometry (MS) are TIMS (Thermal Ionisation Mass Spectrometry) and ICPMS (Inductively Coupled Plasma Mass Spectrometry), with TIMS being more accurate and expensive. There are a number of variants in the ICPMS methods, e.g., single magnet, multiple collector (MCs), quadrapole (Q) etc., that can also influence the accuracy. The methods can also be used in combination with isotopic dilution (sample spiking) for improved accuracy, or a lower-accuracy external calibration can be used.

The measurement accuracy and precision will also depend on the analytical methods, equipment type, element being measured, and concentration of the isotopes. While it is not easy to generalise about the accuracy of different methods for all samples and isotopes, several of the more commonly used methods, listed in the order of decreasing accuracy (and costs), are listed below as a general guide.

- TIMS with Isotopic Dilution (ID): < 1%
- ICPMS with
  - Isotopic dilution method: < 3%
  - External calibration method: < 10%
- Gamma spectrometry: 5 – 10%
- Alpha Spectrometry: 10%

While there is always a desire to obtain the highest accuracy measurements possible for all isotopes to be measured, the practical consideration of programme cost is also an important constraint. At the same time, the use of relatively low-cost, low-precision methods can yield results of such high uncertainty as to make the data of very limited value for code validation purposes. The required accuracy will be guided to a large extent by the isotopes that are most important to the intended application, such as burn-up credit (actinides or fission products), decay heat, radiological sources, or others. What is important is to establish clearly defined accuracy targets with the measurement laboratory before the methods are selected.

For most applications, high-precision measurement methods (1 – 2%) are highly desirable for the major actinides (uranium and plutonium isotopes), neodymium, and other key fission products that are excellent indicators of fuel burn-up. Burn-up is generally determined using one or more of the fission product indicators, such as ^{148}Nd or ^{139}La. Any error in the estimated burn-up of the fuel sample will result in a bias for all predicted nuclides in the sample. Therefore, it is essential to establish this value to very high accuracy, using multiple measures of burn-up if possible.

Measurement data must ultimately be represented in the form of isotopic concentration per unit of initial heavy metal (or fuel) in the sample. The initial mass of the irradiated fuel sample (e.g., grams UO_2) can be weighed directly (corrected for the mass of zirconium cladding), or commonly the results are normalised to the mass of measured ^{238}U in the sample, and this value is then adjusted to correct for the reduction in ^{238}U caused by irradiation. A preferred method that is less reliant on calculations is to measure all of the major actinides (U-Np-Pu-Am-Cm) to determine experimentally the heavy metal in the irradiated sample. This value can then be corrected for the reduction in actinide mass due to fission to determine the initial heavy metal in the sample. The integral number of fissions is determined experimentally using the value for ^{148}Nd (or another suitable monitor) that has a similar fission yield for ^{235}U and ^{239}Pu. Ideally, the initial heavy metal derived from this procedure can be cross checked against the weighted fuel mass as a means of verifying mass balance of the radiochemical measurements.

Analysis of undissolved fission products

It is well known that Tc, Mo, Ru, Rh, Pd, Ag, and Sb segregate in irradiated fuels into a metallic submicron, particulate phase that is virtually insoluble in nitric acid. Any radiochemical analysis of these elements must therefore account for the insoluble residues present after the fuel is initially dissolved. Analysis of the residues can be made by further dissolution of the residues or counting techniques. Final reported concentrations are then determined by combining the inventory in the dissolver solution and the residues.

Based on previous experience with the ARIANE programme, the fraction of the inventory present in the residues can be substantial and vary significantly between different samples. There also does not appear to be a significant correlation between sample burn-up and the fraction of inventory in the residues. The variations observed for several fission products important in burn-up credit are listed in Table 2.

Burn-up estimation

Nd-148 is widely used to estimate the burn-up of the fuel samples. Although reactor burn-up records for may be available from the utility for the selected fuel rods and/or samples, the ability to accurately predict the burn-up for a single rod at a specific axial segment corresponding to the sample position

Table 2. Summary of undissolved metallic fission products in ARIANE programme fuel samples

Fission product nuclide	Percent undissolved (%)	
	Minimum	Maximum
Tc-99	3	50
Mo-95	4	40
Sb-125	3	35
Ru-101,-106	15	70
Rh-103	2	70
Ag-109	5	90

can be quite poor. Our experience has shown that sample burn-up estimates from reactor records may deviate from measured burn-up by more than 10% for heterogeneous fuel assembly designs. Any error in the estimated burn-up of the samples will bias all of the calculated results, and it is therefore critical that this value be determined as accurately as possible. In addition to high precision ^{148}Nd measurements, it is advisable to also use high-precision methods for several other independent fission products that are good measures of the number of fissions in the sample to improve the confidence in the sample burn-up derived from ^{148}Nd. These fission monitors may include ^{137}Cs, ^{139}La, or others.

As an example, recent analysis of a spent fuel sample using a burn-up value derived using reactor physics simulations that matched U and Pu isotopic compositions yielded fission product concentrations, including ^{148}Nd, that were significantly different than measurement. Calculations were then repeated, and the residuals between calculations and measurements were evaluated as a function of burn-up. The residuals the fission product isotopes ^{106}Ru, ^{133}Cs, ^{137}Cs, ^{145}Nd and ^{148}Nd were weighted by the measurement variance σ^2 to determine the burn-up corresponding to the minimum weighted residual. The selected isotopes are either known fission product monitors or are generally well predicted in previous isotopic benchmark studies. The burn-up corresponding to the minimum residual was found to be almost 10% larger, or about 4,000 MWd/t, than the value based on reactor physics simulations. This discrepancy could clearly have a large potential impact on code validation studies. Unfortunately, the uncertainty of the fission product measurements, with the exception of ^{148}Nd (2%), was relatively low (~10%), which does not allow independent confirmation of the burn-up value based on ^{148}Nd with high degree of confidence. As a result, new measurements on duplicate samples are being performed to help resolve the discrepancies, at significant added cost to the project.

It is also recommended that laboratory estimates of burn-up be treated as just that: estimates. The methods routinely used to estimate burn-up from ^{148}Nd are only approximate and may be less accurate than estimates made using codes, particularly for high-burn-up fuels where the approximations can break down. Again, any differences may materialise incorrectly as code bias. The procedure generally used by ORNL is to perform code simulations to obtain a ^{148}Nd value within the uncertainty of the experimental value. This procedure effectively normalises the calculated integral number of fissions in the sample to be equal to the measured fissions determined by ^{148}Nd.

Presentation of experimental results

Measurement results are sometimes reported for a single cooling time, often at discharge, to simplify use of the data in code benchmark studies. This is done by adjusting the actual measured values according to the decay between the time of measurement and the reference time (often taken as discharge). For some nuclides this correction is related simply by the half-life of the nuclide. For other nuclides, particularly those with decay precursors, the correction is complex and requires explicit

measurement of the precursor concentrations to adjust the measured values correctly. To avoid introducing errors due to back calculating spent fuel inventories to a reference date, it is essential for laboratories to always report the results at the date of the measurements. If adjusted data are also reported, details of the methods, assumptions, and nuclear data should be provided.

Fuel design and reactor operating data

A large amount of detailed design and operational data is required to accurately simulate the fuel isotopic inventory evolution during irradiation and decay after discharge from the reactor. For code benchmarking applications, it is highly desirable to acquire the physics data as early as possible and ensure that the data are of sufficient detail and accuracy. Rigorous lattice calculations may require information that includes:

- fuel and assembly geometry specifications and design dimensions,
- fuel enrichments, zoning patterns (if applicable),
- guide tube configuration; burnable poison rod compositions and exposure period,
- integral burnable poison rod configuration, compositions and poison content,
- control rods exposure and percent axial insertion (if applicable),
- fuel and moderator temperatures during operation (at sample position),
- soluble boron variation during cycle irradiation,
- moderator density at the axial position of the sample,
- neighbouring assembly data (enrichment, burn-up, by cycle) particularly for fuel rods obtained from the outer edge of the assembly,
- cycle start and end dates, and date for all measurements,
- cycle power variations and burn-up or average power by cycle.

For fuel samples obtained from reconstituted, or rebuilt assemblies (rods taken from one irradiated assembly and inserted into a second host assembly), detailed design and operational data are required for all assemblies and fuel rods of the primary and host assembly.

Another important data consideration is that some detailed design information, particularly for the complex modern assembly designs, may be protected or commercially sensitive. This information may be needed to perform rigorous inventory simulations. If it is desired to make the measurements and design data publicly available as a code benchmark it is essential that public disclosure of the data or distribution conditions be approved by the fuel manufacturer, preferably before the measurements are started.

Fuel sample selection

A key lesson based from our experiences in evaluating assay data is the importance of obtaining spent fuel measurements from samples that are representative of standard commercial fuels. Some of the fuel samples evaluated to date have included non-standard fuels that were not selected for the purposes of isotopic assay, but were made available from other programmes (e.g. cladding irradiation, fuel failures, etc.). Selection of these fuels can significantly reduce costs because the fuel is already at the hot cell, thereby avoiding additional transportation costs. Although well intentioned, these fuels in some cases experienced operational conditions that were not typical of commercial fuels, and were also poorly characterised for the purposes of fuel depletion calculations. Non-standard fuel can introduce additional analysis complexities and computational uncertainty, and ultimately these uncertainties will be reflected as potentially unrepresentative high margins of safety and conservatism.

Another source of assay data, particularly for high-burn-up fuel samples, has come from reconstituted or rebuilt assembly. These assemblies are typically rebuilt with a combination of previously irradiated and fresh fuel rods to achieve high burn-up in a reasonable irradiation time by effectively driving fissions in the high-burn-up rods at a higher rate than could be achieved otherwise. In some instances this has involved low initial enrichment fuel irradiated to burn-ups than could not be achieved ordinarily. However, since the burn-up is unrepresentative of typical discharge fuel of that enrichment it is not clear that any isotopic uncertainties obtained from using the sample as a benchmark are typical of standard commercial fuel. In the end, it is important to develop clear guidelines and criteria for selecting representative fuel samples a priori, rather than trying to rationalise large differences between calculations and experiments if the results are poor.

Another issue that must be considered is obtaining an adequate number of fuel samples for a reliable statistical evaluation of the data. If the sample size is too small, the tolerance factor that accounts for the uncertainty due to the finite number of measurements can be excessive. However obtaining a large number of measurements is no guarantee of reduced isotopic uncertainty if the measurements themselves are low quality/precision and exhibit inconsistent or high variability. Additionally, it is recommended that samples be acquired from different rods from several representative locations in the assembly. Because of spectral variations within the assembly (e.g., near the assembly periphery, water regions, poison regions, etc.) it is important to obtain as representative a sample population as possible.

Experience and recommendations

ORNL has previously evaluated experiments measuring the isotopic concentrations for 56 PWR spent fuel samples and is currently engaged in evaluating more than 40 new samples from other modern experimental programmes in support of burn-up credit and other code validation studies. Some of the primary recommendations based on our evaluations follow:

- Independent laboratory measurements are highly desirable to establish reliability of the radiochemical analysis methods by cross checking results.
- Use high-precision measurement methods and instruments as available.
- Undissolved residue must be analysed for the metallic fission products.
- Acquire the necessary design and detailed operating history data early in the programme.
- Use standard design fuel assemblies representative of spent fuel inventory.

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NEED FOR VVER PIE SPENT FUEL COMPOSITION DATA

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Abstract

This paper briefly describes the situation of non-Russian VVER countries with respect to their nuclear fuel facilities. The nuclear energy production rates, number/type/operation period of the VVER reactors, fuel designs/fabrication/supplies to the individual NPPs, hot cell laboratories in Eastern Europe and several published details on the RIAR laboratory are reviewed. Finally, an overview of the status of available data for depletion code validation in the VVER environment is given.

Introduction

Today there are 16 VVER-440 units operating in OECD/NEA member countries (the Czech Republic, Finland, Hungary and Slovakia); the full list of VVER 440 and 1000 units is provided below.

In the following discussions, several facts aimed at describing the spent fuel issue in VVER operating countries will be given based on information of IAEA, other public sources or referenced publications within the criticality safety community.

VVER country	Country nuclear share in electricity generation in 2005 [%]
Armenia	42.7
Bulgaria	44.1
Czech Republic	30.5
Finland	32.9
Hungary	37.2
India	*2.8*
Russia	15.8
Slovakia	56.1
The Ukraine	48.5

Source: IAEA 2005, http://www.iaea.org/dbpage/images/share.jpg

(India: 2 VVER-1000 units under construction, Libya: Considering obtaining 2 VVER 440 units)

Countries operating VVERs

country	number of VVER 440 units/ operation from/ (name)	note		number of VVER 1000 units/ operation from / (name)	note
Armenia	1/1980 (Armenia-2)	out of operation in 1989-1995		-	
Bulgaria	2/1980, 1982 (Kozloduj NPP)	EU insists on closing both		2/1988,1993 (Kozloduj NPP)	
Czech Republic	4/1985,1986,1986,1987 (Dukovany NPP)			2/2002,2003 (Temelin NPP)	design/instrumentation was changed by Westinghouse in the course of construction
Finland	2/1977,1981 (Loviisa NPP)			-	
Hungary	4/1983,1984,1986,1987 (Paks)			-	
India	-			(Kudankulam NPP)	2 units under construction from 2002
Russia	6/1972-84 (4 Kola NPP, 2 Novovoronezh NPP)			8/1981-2001 (4 Balakovo NPP,2 Kalinin NPP,1 Novovoronezh NPP, 1 Rostov NPP	
Slovakia	6/1978, 1980 a 1984, 1985, 1998, 2000 (4 Bohunice NPP, 2 Mochovce NPP)	EU insists on closing 1-2 first units		-	
The Ukraine	2/1981,1982 (Rovne)			11/1981-1989 (1 Khmelnitski NPP,1 Rovne NPP,3 South Ukraine,6 Zaporozhe)	
total	27 VVER-440 units (21 of them outside of Russia)			23 VVER-1000 units (15 of them outside of Russia)	2 under construction in India (?)

Source: IAEA, 2002, for more details see:
http://www-pub.iaea.org/MTCD/publications/PDF/cnpp2003/CNPP_Webpage/pages/countryprofiles.htm

Fuel supplies to the individual VVER 440 units from JSC 'MBP' (Joint Stock Company 'Machinary Building Plant' situated about 50 km east Moscow) [1]

unit \ year	1983	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99	00	01	02	03	04	05
Kola 3	3.6 %		4.4 % (4 cycles)					4.4 % (5 cycles)												4.25 %			
Kola 4	3.6 %															4.4 % (with Gd, 5 cycles)							4.25%
Rovne 1	3.6 %											4.4 % (4 cycles)					4.21 % (without Gd, 4 cycles)						
Rovne 2	3.6 %												4.4 % (5 cycles)			4.21 % (without Gd, 5 cycles)							
Dukovany	3.6 %															3.82 %					4.38 (Gd 1)		4.25 (Gd 2)
Bohunice 3,4 Mochovce, Paks	3.6 %																3.82 %						
Loviisa 1	3.6 %																	3.82		3.7% *			
Loviisa 2	3.6 %																	3.82		4.0%			

Fuel Designs for VVER-440 of V-213 reactor vessel type characterised by the average enrichment

unit \ year	1986	87	88	89	90	91	92	93	94	95	96	97	98	99	00	01	02	03	04	05
Novovoronezh 3	3.6 %																3.82 % **			
Novovoronezh 4	3.6 %									3.82 %										
Kola 1	3.6 %																		3.82 %	
Kola 2	3.6 %																3.82 %			
Bohunice 1	3.6 %																		3.82 %	
Bohunice 2	3.6 %																	3.82 %		

Fuel Designs for VVER-440 of V-179 and 230 reactor vessel type characterised by the average enrichment

* BNFL fuel, ** fuel made in 'Novosibirsk Chemical Concentrates Plant' Joint-Stock Company

Hot cell laboratories in the Eastern European countries

There are only a few laboratories in Eastern Europe with the capability to manipulate and cut spent fuel, one is in Romania and the others in Russia. According to IAEA 'Nuclear Fuel Cycle Information System' (**http://www-nfcis.iaea.org**, data of 2003-4), here is the list of main laboratories with hot cells available in Eastern Europe:

Spent fuel of VVER and fast reactors can be examined in:
RIAR (Russian Federation Research Institute of Atomic Reactors), Dimitrovgrad, Ulyanovsk Region, Russia

Spent fuel of RBMK can be examined in:
RI (Radium Institute), St. Petersburg, Russia

The other laboratories in Russia but not assigned for regular VVER spent fuel examination:
IRM (Institute of Reactor Materials, NIKIET) Zarechny, Sverdlovsk Region, Russia
IPPE (Institute for Physics and Power Engineering) in Obninsk, Russia
Kurchatov IAE, Moscow, Russia

Institute for Nuclear Research, Pitesti, Romania (NOTE: Romania operates only CANDU reactors, no VVERs)

Due to the current international law (based on proliferation concerns), it does not seem feasible to move any VVER spent fuel assemblies to Romania or outside of Eastern Europe in the near term. Therefore, only the Russian laboratories are considered in the ongoing discussion of future VVER PIE experiments.

Details on the RIAR laboratory (http://www-nfcis.iaea.org, http://www.niiar.ru/oit/en/oite.htm)

ACCEPTANCE	
Acceptance as either assemblies or rods	both
Acceptance in either wet or dry condition	both
Mode of transfer cask/cell Maximum weight of cask which can be handled	horizontal (rods) & vertical (assemblies) 50 t
Maximum length of rod which can be accepted	4.3 m
Maximum fissile content of both U or Pu (weight and/or enrichment)	125 kg
Acceptance of failed rods, with or without auxiliary cladding	Yes, without
CELL CHARACTERISTICS	
Main purpose of facility	PIE of fuel assemblies & rods
γ-Activity limits per type	10000 TBq concrete/300 TBq lead
Maximum length of rods which can be handled	4.3 m
Cell atmosphere	Air

Scheduled maintenance (Yes or No)		Yes
Largest Cell Width (m)		8
Largest Cell Length (m)		6
Largest Cell Height (m)		8
PIE techniques - nondestructive	**availability**	**comments**
Visual Examination (maximum magnification) a) Photographic or video storage b) Colour or black & white c) Stereo microscope	 x x	x30 both both
Length and diameter	x	
Gamma scan a) Quantitative	x x	
Eddy Current testing a) Encircling coil b) Point coil	x x x	
Oxide thickness	x	
X-Radiography a) Tomography	x x	
Neutron Radiography a) Tomography	x	
Clad-fuel gap	x	
Non-destructive gas release	x	thermal conductivity
Rod puncture a) Fission gas release b) Rod internal volume	x x x	
PIE techniques - destructive	**availability**	**comments**
Optical microscopy	x	F+C
SEM a) Elemental analysis (EDAX or WDAX) b) Polished surface c) Fracture surface	x x x x	
Image analysis	x	
EPMA	x	
Micro_γ-scan	x	F
TEM		
Micro-coring	x	
Auger spectroscopy	x	
Fission gas diffusivity	x	F
X-ray fluorescence	x	
α/β autoradiography	x	
X-ray diffraction	x	
Density	x	
Open porosity	x	F
Burn-up	x	F
O/M ratio	x	F
Thermal diffusivity	x	
Specific heat	x	
Melting point	x	
Hydrogen analysis	x	C

Clad chemical composition	x	C
Thermal conductivity of Zircaloy oxide	x	C
Tensile testing	x	C
Tube burst testing	x	C
Clad creep testing	x	C
Clad fatigue testing	x	C
Other mechanical testing		C
Other SIMS Impedance spectroscopy	x	
Refabrication and instrumentation Specify instrument(s)	x	
STORAGE and CONDITIONING	**availability**	**comments**
Intermediate storage capability	x	
Encapsulation for re-insertion into FAs		
Encapsulation for other purposes	x	for irradiation in research reactors
Connection to a reprocessing plant	x	

The VVER PIE data availability outside Russia

In the past, short pieces of information on the measured data of the isotopics of VVER fuel spent in 70' in Novovoronezh NPP for specified isotopes of major actinides (U, Pu, Am, Cm) were published in a Russian journal 'Atomnaya Energia' in early 80' (Tom 51, Vypusk 1, 1981, p. 53-54; Tom 55, Vypusk 3, 1983, p. 141-145). The publication of 1983 included a certain part of the measured data but the unpublished operational history in the course of depletion in details didn't allow any independent calculations. In the published article, some results of calculations with (at that time) currently used codes in Russia were mentioned. The calculations resulted in discrepancies less or up to 25 % for U and Pu isotopes and up to more than 40 % on Am and Cm. The kind and amount of the data published are neither sufficient for the depletion code validation nor usable for validation focused on the BUC implementation which takes actinides and major fission products into criticality analyses.

Only in 1998, the analysts of Kurchatov Institute (KI) presented the calculational exercise on fuel burn-up and inventory of VVER-440 spent fuel which included measured data for a comparison (Kravchenko Y.N., et al., 'Benchmark Calculation of Fuel Burn-up and Isotope Composition of VVER-440 Spent Fuel', paper at the 8th AER Symposium (Bystrice nad Pernstejnem, Czech Republic, Sept. 21-25, 1998). The exercise was specified as checking on PIE data from measurements of 11 samples of the same fuel as those above mentioned in publication of 1983 (VVER- 440, 3.6 % enriched, spent in 1973-1976 in Novovoronez NPP). Unfortunately, the exercise was not preprepared as a typical calculational 'benchmark' as several essential input parameters for isotopics prediction were not known and several codes including the operational one had to be employed to obtain them. Thus, the deviations between the calculation and experimental results could be hardly split into parts belonging to several different collaborating codes involved in the calculation and therefore the results are not able to contribute essentially to the individual code (calculational methodology) validation. The reason of discrepancies was very probably connected with the fact that the operational histories of fuel were old (not recorded digitally and incomplete), also not supported by any sophisticated core on/off line simulator. The results were presented by the several participants of the following AER Symposiums in 1999 held in Slovakia and 2000 held in Moscow. The calculations of the exercise authors of KI consisted of several steps using various codes and approaches, incl. e.g. fuel temperature

calculation. One of the conclusions made was that the isotopic differences are not large if FA surroundings are taken into account in the calculation. Several thoughts were expressed on the possibility of using the newly acquired PIE data in Russia which were unfortunately proprietary and not openly available.

Eastern European countries operating VVERs unfortunately have no specific funds to purchase these newer classified or proprietary Russian data. However, thanks to the recent OECD/NEA initiative for reactor physics benchmarks, Russian scientists have released a list of the measurements performed (shown in the Table 1 below).

As the reactor type installed in the VVER countries outside Russia in most cases was the VVER 440, these countries are mainly (the OECD VVER countries: only) interested in availability of the PIE data for VVER 440 spent fuel. Unfortunately, the VVER-440 spent fuel listed in the Table 1 is either the 3.6 wt.% enriched fuel of Novovoronezh coming from a control FA which is not too suitable for the validation calculations and spent fuel of 4.4 wt.% enrichment which is not used outside Russia except the Ukraine. But anyway, all these data were said proprietary. As for the VVER 1000 data, some of them seem to be released in the future.

Finally, the ISTC project No. 2670 on VVER-440 PIEs funded by US and led by L. Jardine of LLNL and A.Chetverikov of Dimitrovgrad was performed in RIAR Dimitrovgrad under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory. The project was completed in 2005; the final report was issued in LLNL [2] and released without any restrictions. The resulted well-documented VVER-440 PIE data for the 'BUC nuclides' (actinides and major fission products) from radiochemical measurements of 8 spent fuel samples thus became accessible to the whole VVER criticality safety community. The final report on the ISTC #2670 results listed another selected VVER-440 FAs (shown in the Table 2 below) that were subjected to post-irradiation examinations in the RIAR laboratory in 1988-2002. Unfortunately, except for the ISTC results belonging to the FA listed as No. 5 (D26135 FA of the Novovoronezh NPP), none of the PIE results were released outside of Russia.

Notes to the Table 2 items:

#7 E22198: the FA of KOLA NPP-3 is one of those FA mentioned also in the previous Table 1
#4 D19159: the FA is one of those FAs offered for the new VVER PIE
#5 D26135: the FA selected for the examination under the ISTC project (2003-5) was, as mentioned in the table, used for measurements in 1995. If the samples were the same but new measurements were performed within the ISTC project the results of the measurements made earlier would be very interesting if added (e.g. as an appendix) to the final report for the comparison (in spite of the fact that not all the 'BUC' isotopes are likely to have been measured in 1995).

As far as the final report on the ISTC #2670 project is concerned, some missing data/results in the final report were indicated as needed for independent recalculations or data comparison,e.g.:

- weight of FA spacers
- z-co-ordinate of each of 25 layers along z-axis taken for the Russian supporting calculations
- changes of pellet/pin dimensions could be mentioned, dishing of fuel pellets if observed after cutting
- full definition of the alloys (cladding, shroud, central tube)
- core and FA pattern positions are marked in a different way (sometimes non-consistently) by the experimentalists (RIAR) and analysts (KI) making the supporting calculations, legend of

marking is not clear, types of FA are not fully/clearly described (H,G,S,E - description is missing)
- the data of the core irradiation history before cycle No.15
- records on the fuel assembly orientations during the campaigns
- results of the temperature/mechanical predictive calculations
- tables of isotopics of Rh-103 resulting from the radiochemical analyses
- the references seems to be entirely unavailable from outside of Russia (most of them are not fully defined), the main subject of reference would be appreciated if at least shortly described in the report body

Efforts to acquire VVER PIE data should continue, the new results are needed to enlarge the applicability range in the measured data set and to increase the statistical data sample for evaluation and subsequent use for depletion code validation. Therefore, the new VVER-440 measurements are being negotiated with RIAR and a new project reflecting the recent RIAR offer [1] is being proposed to be funded by a consortium of countries interested in the data, which are mainly the VVER countries outside Russia.

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Anatoly Chetverikov: The VVER PIE offer, RIAR 2005, presented in discussion on PIE at 10th Meeting of AER Working Group E, Modra, Slovakia, April 2005.

L. J. Jardine, Lawrence Livermore National Laboratory 'Chemical Assays of Irradiated VVER Fuel for Use in Burn-up Credit' ISTC Project #2670 (between the International Science and Technology Center, the Department of Energy of the United States of America and the State Scientific Center "Research Institute of Atomic Reactors, RIAR, Project Manager Alexey V.Smirnov, RIAR), UCRL-TR-212202.

Table 1. Spent fuel samples measured in Russia recently

Sample origin	VVER type	number of samples	Fuel irradiated /measured	Actinides measured	FP measured
Balakovo, Unit 2 4.4 wt.%	1000	2	1990-93/ 1996,1997	^{232}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{236}Pu, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm	^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd, ^{151}Eu, ^{152}Eu, ^{153}Eu, ^{154}Eu, ^{155}Eu, ^{147}Sm, ^{148}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{154}Sm, ^{147}Pm, ^{106}Ru, ^{144}Ce, ^{99}Tc, ^{103}Rh
Balakovo, Unit 3 4.4 wt.%	1000	2	1990-93/1996	^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, ^{244}Cm	^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd
Balakovo, Unit 3 3.6 wt.% UO_2 + 8 wt.% Gd_2O_3 as integrated burnable absorber	1000	12	1994-99/ 2001,2002	^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm, ^{247}Cm, ^{248}Cm	^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd fuel burnable absorber: ^{154}Gd, ^{155}Gd, ^{156}Gd, ^{157}Gd, ^{158}Gd, ^{160}Gd
Kalinin, Unit 1 3.6; 4.4 wt.%	1000	3	1988-89/ 1996,1997,1999	^{232}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{236}Pu, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm	^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd, ^{151}Eu, ^{152}Eu, ^{153}Eu, ^{154}Eu, ^{155}Eu, ^{147}Sm, ^{148}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{154}Sm, ^{147}Pm
Zaporozhe, Unit 1 4.4 wt.%	1000	4	1988-1991/ 1994	^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm	^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd
Zaporozhe, Unit 1 4.4 wt.%	1000	9	1988-94/ 1997,1998	^{232}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{236}Pu, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, ^{244}Cm	^{133}Cs, ^{134}Cs, ^{135}Cs, ^{137}Cs, ^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd
Zaporozhe, Unit 6 3.6, 4.4 wt.%	1000	3	1988-89/2000	^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{244}Cm	^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd
Kola, Unit 3 4.4 wt.%	440	5	1986-90/1995	^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am	^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd
Novovoronezh, Unit 4 3.6 wt.%, control FA	440	5	1990-97/1999	^{232}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{236}Pu, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, ^{244}Cm	^{133}Cs, ^{134}Cs, ^{135}Cs, ^{137}Cs, ^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd

Table 2. The VVER-440 FAs subjected to post-irradiation examinations in the RIAR laboratory in 1988-2002 [2]

No	Fuel assembly No	Fabrication date	Date of loading	Unloading	Reactor, power unit	Beginning of examination	Completion of examination	U-235 enrichment, %	Number of fuel cycles	Operation period, effective days	Average specific generated power of FA, MWd/kgU
1	D11066	01.03.83	06.08.83	23.06.86	Rovno NPP-1	01.01.89	30.12.89	3.6	3	857.6	36.8
2	D11809	01.05.83	11.09.83	06.08.86	Novovoronezh NPP-4	01.01.90	22.10.90	3.6	3	994.8	32.8
3	D15687	01.06.84	25.09.84	06.08.86	Novovoronezh NPP-4	01.01.90	16.10.90	3.6	2	605.2	24.2
4	D19159	01.07.85	02.09.85	16.08.88	Novovoronezh NPP -4	01.01.91	01.01.92	3.6	3	989.1	34.1
5	D26135	01.06.87	13.10.87	04.07.91	Novovoronezh NPP -4	01.01.95	01.09.95	3.6	3	1109.7	38.5
6	D52380	01.02.96	10.06.96	18.06.99	Kola NPP-2	01.01.01	01.06.01	4.4	3	649.1	27.6
7	E22198	01.05.86	24.09.86	01.11.90	Kola NPP-3	01.01.94	30.12.94	4.4	4	1259.8	46.2
8	E22222	01.05.86	24.09.86	25.10.91	Kola NPP-3	01.01.94	30.12.94	4.4	5	1564.9	48.2

APPLICABILITY OF VVER-440 ISOTOPIC ASSAY MEASUREMENTS TO VALIDATE CODES FOR WESTERN PWR FUEL DESIGNS (AND *VICE VERSA*)

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Abstract

Recent isotopic assay measurements on VVER 440 fuel samples published as part of an International Science and Technology Center (ISTC) project are evaluated for applicability to validating depletion codes used for western-design pressurised-water reactor (PWR) fuels. The measurements are potentially of high benefit to burn-up credit in the United States because they include most of the fission products important to burn-up credit. However, the VVER reactor and fuel design is distinct from western PWR designs, e.g. triangular pitch rods, smaller fuel diameter, and tighter fuel pitch, etc. This paper quantitatively evaluates the similarity and applicability of isotopic measurements for VVER 440 fuel to western-design PWR fuels using modern TSUNAMI sensitivity/uncertainty analysis tools developed as part of the SCALE computer code system.

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Introduction

Isotopic assay measurements were recently performed at the Research Institute for Atomic Reactors (RIAR) in Dimitrovgrad, Russia, on eight VVER 440 fuel samples from the Novovoronezh reactor and published [1] with the related design and operating history data as part of an International Science and Technology Center (ISTC) project. The measurements are potentially of high value to burn-up credit in the United States and other countries operating western-design pressurised-water reactor (PWR) plants because the measurements include most of the fission product isotopes of importance to burn-up credit. Publicly available isotopic measurements on western PWR fuels of low to moderate enrichments and burn-up generally include few fission product isotopes of importance to burn-up credit, largely because these measurements were performed before the time that burn-up credit data needs had been clearly identified.

The VVER reactor and fuel design is distinct from western PWR designs, e.g., triangular pitch rods, smaller fuel diameter, and tighter fuel pitch, etc. Therefore, establishing the similarity and applicability of the measured VVER isotopic data to the validation of computer codes for western PWR fuel designs is an essential task before the measurement data can be applied. Establishing applicability of isotopic measurements from VVER 440 fuel to western PWR fuel would also imply that PWR fuel data can be similarly applied to validating depletion codes used for VVER fuels. This paper quantitatively evaluates the similarity and applicability of isotopic measurements for VVER 440 fuel to western-design PWR fuels using TSUNAMI sensitivity/uncertainty analysis tools [2] developed as part of the SCALE computer code system [3].

Sensitivity methods

This study compares an infinite array configuration of VVER-440 spent fuel assemblies with similar configurations of Westinghouse (W) 17 × 17 and Combustion Engineering (CE) 14 × 14 PWR spent fuel assemblies under typical reactor operating conditions. The comparisons were performed by generating detailed sensitivity and uncertainty information for each configuration using the TSUNAMI methodology. The sensitivity and uncertainty information for each configuration was integrated together into c_k and g integral indices of system cross section similarity. These indices range from 0 to 1, where a 0 value indicates no similarity from a neutronic point-of-view between the two systems and a c_k value near 1 indicates virtually identical systems, and a g value of 1 indicates complete coverage. The c_k values represent the systems' similarity on a global basis, while the g values are tabulated for each important nuclide and reaction type to indicate similarity on a differential level. A description of the development and applications of these indices is given separately [4,5].

Assembly models

Irradiated fuel isotopics were computed based on an initial fuel enrichment of 5 wt.% UO_2 and burn-up values of 10, 30, and 60 GWd/MTU and 5-years cooling. The spent fuel nuclides included in the assembly comparisons are given in Table 1. The isotopics were generated using the ORIGEN-ARP code [6] using the standard cross-section libraries for the respective fuel types distributed with the SCALE system. The cross-section libraries were generated based on typical assembly design and operating information given in Table 2 for the three assembly types considered in this study.

Table 1. Actinide and fission product nuclides used in similarity comparison

^{234}U	^{235}U	^{236}U	^{238}U	^{238}Pu	^{239}Pu
^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{99}Tc	^{133}Cs
^{153}Eu	^{143}Nd	^{145}Nd	^{147}Sm	^{149}Sm	^{150}Sm
^{151}Sm	^{152}Sm	^{155}Gd			

Table 2. Fuel assembly model specifications

Assembly parameter	CE 14 × 14	W 17 × 17	VVER 440
Fuel type	UO_2	UO_2	UO_2
Enrichments (wt.% ^{235}U)	1.5-5.0	1.5-5.0	1.6 - 4.4
Fuel temperature (K)	873	811	980
Effective fuel density (% TD)	91.7	94.5	84.2
Moderator density (g/cm^3)	0.7332	0.7295	0.73
Moderator temperature (K)	570	570	575
Clad material	Zirc2	Zirc2	Zr (1% Nb)
Clad temperature (K)	620	620	630
Fuel pellet ID (cm)	N/A	N/A	0.15
Fuel pellet OD (cm)	0.956	0.819	0.756
Gap OD (cm)	0.986	0.836	0.776
Fuel pin OD (cm)	1.117	0.950	0.910
Pin pitch (cm)	1.4732 (square)	1.2598 (square)	1.22 (triangular)
Number of central/instrument tubes	1	1	1
Guide tube ID (cm)	2.628	1.1430	0.880
Guide tube OD (cm)	2.832	1.2243	1.030
Guide tubes-water holes/assembly	4	24	0

The fuel rod and assembly lattice details given in Table 2 were used to construct three-dimensional KENO-V.a models for each fuel assembly type. The VVER 440 infinite assembly model is shown in Fig. 1 where the assembly is modeled with a reflective boundary condition to simulate an infinite array of assemblies. The CE 14 × 14 and W 17 × 17 models are shown in Fig. 2. Each of the three assembly models were input into the TSUNAMI-3D module for this study.

Results

A review of the design specifications in Table 2 indicates that the VVER 440 fuel rods are smaller in diameter than the typical range for PWR assemblies and that lattice pitch is tighter. A wider survey of PWR assembly designs was performed, and the PWR design and physics parameters were compared to those for the VVER 440 lattice in Table 3. The comparison results confirm that the VVER 440 design data are outside the range of typical PWR fuels and that the assembly moderation ratio (H/X) is also less than PWR assemblies. However, many design and operating parameters (Table 2) are very similar.

Figure 1. Fuel assembly configuration in VVER 440 model.

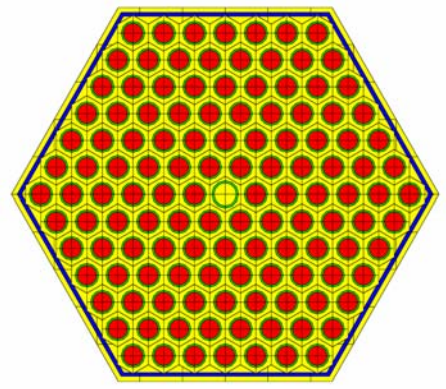


Figure 2. Fuel assembly configuration in W 17 × 17 and CE 14 × 14 models (shown approximately to scale in comparison with VVER model illustrated in Fig. 1).

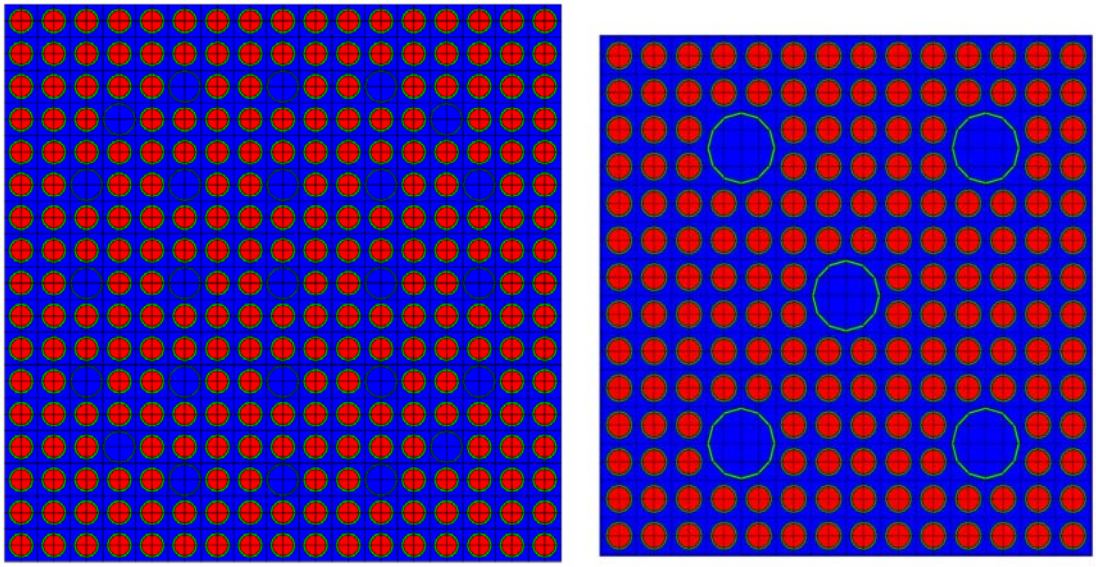
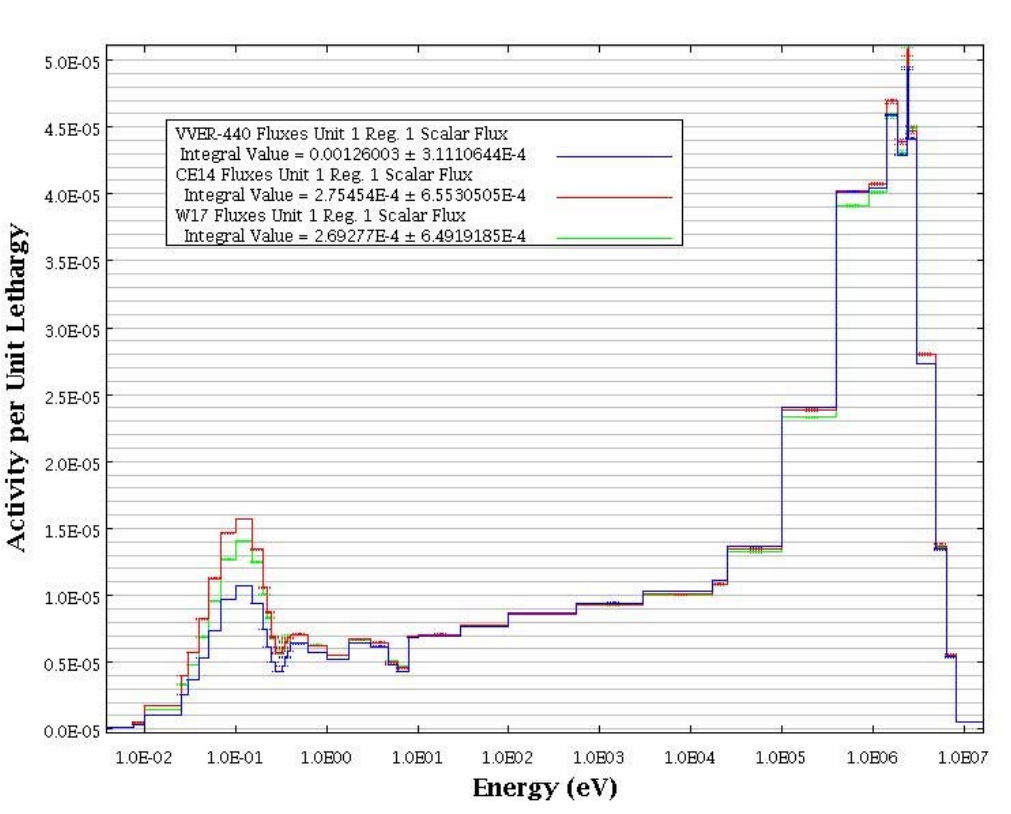


Table 3. Fuel assembly data and physics parameters

Fuel parameter	PWR	VVER 440
Fuel OD (cm)	0.95 - 1.12	0.91
Fuel rod pitch (cm)	1.26 - 1.47	1.22
Lattice type	Square	Hexagonal (triangular)
H/X ratio	1.23 - 1.27	0.98
Fission neutrons / thermal absorption	1.52	1.851
Resonance escape probability	0.597	0.608
Thermal utilisation	0.826	0.845
Reactivity control	Soluble boron / B_4C / Gd	Soluble boron / absorber assemblies
Soluble boron levels (avg ppm)	300 - 700	460

A comparison of the neutron flux spectra in the fuels is shown in Fig. 3 for normal operating (hot) conditions. The results show that the neutron energy spectra are very similar in the intermediate and resonance energy range, but that the spectrum for the VVER 440 fuel is considerably harder (lower thermal flux) than for the two PWR assemblies considered; an observation consistent with the assembly physics parameters.

Figure 3. Comparison of neutron flux spectra in VVER 440 and PWR fuel



Ultimately it is the similarity of the neutron spectrum in the fuel (and its influence on the cross-sections and reaction rates) that will determine the degree of similarity of the different fuel types.

Further quantitative analysis was performed using the TSUNAMI-IP module. The TSUNAMI comparison results are given in two forms, first the c_k values which represent the global similarity between two selected systems, followed by the g values which are tabulated similarities on a nuclide-by-nuclide and reaction basis between two selected systems. The c_k values given in Table 4 compare each of 3 burn-up values for the VVER 440 assembly model (three rows) to corresponding burn-up values for the two PWR assembly models (6 columns). The last two columns in Table 4 contain a comparison of the c_k values between the various VVER 440 burn-ups and the CE 14 × 14 and W 17 × 17 assemblies with the **same** isotopic compositions as the VVER 440 60-GWd/t case to isolate the effects caused by differences in fuel composition from those associated with the assembly design. The c_k values greater than 0.9 are highlighted in red for emphasis. In previous experience comparing different critical systems, a c_k value > 0.9 is an indicator of high system similarity.

The c_k elements with corresponding burn-ups (i.e., 10 GWd/t for VVER-440 vs. 10 GWd/t for both CE 14 × 14 and W 17 × 17 assemblies) are all 0.97 or higher, indicating very similar neutronic systems in terms of the similarity of the energy-dependent cross sections to the system neutron multiplication factor (k_{eff}). The practical importance of the similar c_k indices is that any bias in the energy-dependent cross sections will be manifest as a similar level of k_{eff} bias in the two systems.

The results for burn-up values of 30 and 60 GWd/t also indicate high similarities between these systems, because each of these (even with different geometry) have c_k values above 0.9. Comparing the results in the last two columns with the corresponding values for each PWR assembly type at 60 GWd/t burn-up indicates that the results are relatively insensitive to the different assembly geometries, and thus the geometries are considered similar among the three assembly types, with most of the differences seen due to the change in importances of the various isotopes with burn-up. Not explicitly shown in the table is a comparison of the c_k elements for the two different PWR assembly designs (CE 14 × 14 vs. W 17 × 17). The c_k value, for equivalent burn-up values, was found to be > 0.995, indicating effectively neutronically equal systems. Thus, although the VVER 440 results were found to be very similar to the PWR assemblies (e.g. > 0.97), the similarity index of is significantly lower than that between the different PWR assembly designs (> 0.99).

A more detailed method was used to supplement and extend the similarity determination to the individual actinides and fission products in the fuel. Nuclide indices, or g values [5], are useful in this regard and were used in this study to compare the important nuclide similarities. The g results are shown graphically in Fig. 4 for the VVER 440 vs W 17 × 17 and CE 14 × 14 assembly cases. All results shown in Fig. 4 correspond to a burn-up of 60 GWd/t. Clearly the most of the dominant nuclides have g values greater than 0.8, and many have values greater than 0.9. In general the g values for the actinides indicate a greater degree of similarity (nearer to unity) between the VVER 440 and W 17 × 17 assembly than the CE 14 × 14 assembly; again, a result that is consistent with degree of moderation in the respective assemblies. A notable exception is the result for ^{240}Pu capture. Both the CE 14 × 14 and W 17 × 17 contain more water regions (higher moderation) than the VVER 440 assembly, which may influence the ^{240}Pu cross-section similarity between the systems. Difference might also be attributed to the different number densities. Further investigation of these differences was beyond the scope of this preliminary study.

The results for the c_k values are observed to be similar to the results of the g values: although a general high degree of similarity between PWR spent fuel assemblies and those of VVER 440 assemblies is observed, the differences on a nuclide-by-nuclide basis are observed in many cases to be greater than the differences between the PWR assembly designs (e.g. ^{238}U capture). Similar trends are observed in the comparison of the g values for the major fission product nuclides in Fig. 5.

Figure 4. Comparison of actinide *g*-values between CE 14 × 14 vs. VVER 440 assembly cases and W 17 × 17 vs VVER 440 assembly cases for 60-GWd/t burn-up (f = fission, c = capture).

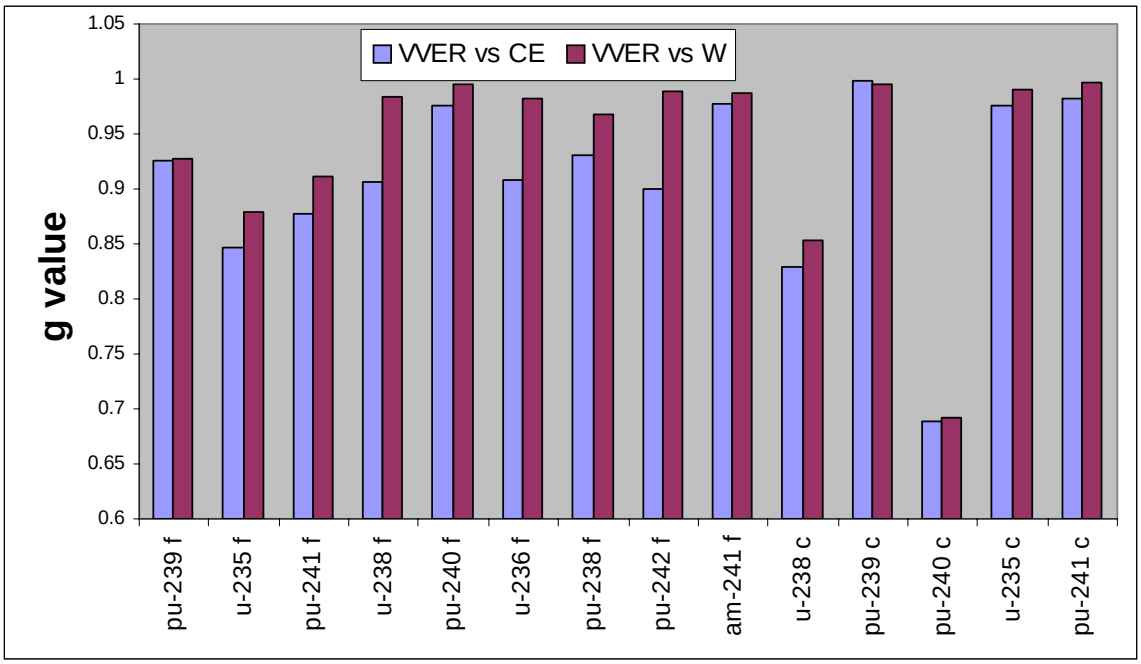


Figure 5. Comparison of fission product *g*-values between CE 14 × 14 vs. VVER 440 assembly cases and W 17 × 17 vs. VVER 440 assembly cases for 60-GWd/t burn-up.

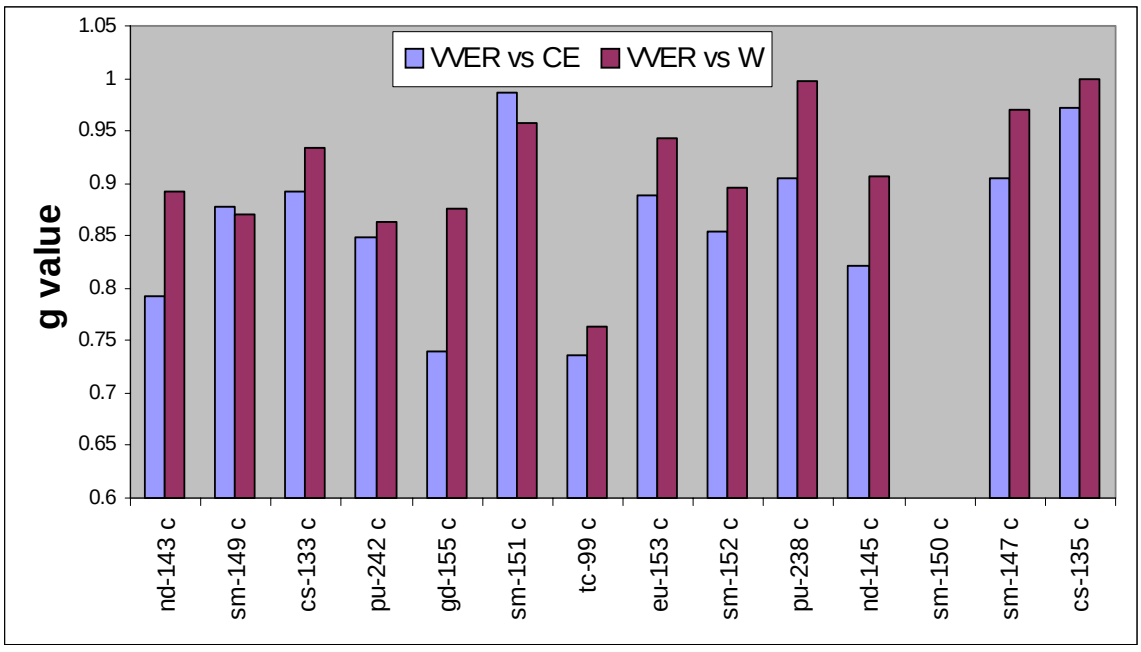


Table 4. c_k results for CE 14 × 14 and W 17 × 17 vs VVER 440*

System	CE 14 × 14 10 GWd/t	CE 14 × 14 30 GWd/t	CE 14 × 14 60 GWd/t	W 17 × 17 10 GWd/t	W 17 × 17 30 GWd/t	W 17 × 17 60 GWd/t	CE 14 × 14 60 GWd/t†	W 17 × 17 60 GWd/t†
VVER 440 10 GWd/t	0.9826	0.9272	0.7631	0.9829	0.9178	0.7654	0.7600	0.7632
VVER 440 30 GWd/t	0.8879	0.9756	0.9321	0.8984	0.9771	0.9349	0.9341	0.9356
VVER 440 60 GWd/t	0.7293	0.9151	0.9752	0.7458	0.9241	0.9765	0.9783	0.9781

*c_k values greater than 0.9 are shown in red.
† Isotopic compositions are taken from the VVER 440 60-GWd/t case

Summary

Using conventional measures to establish levels of similarity between different critical systems, the VVER 440 and PWR assembly designs evaluated are shown to be very similar, with global $\boldsymbol{c_k}$ indices > 0.97. However, the similarity is found to be less than that between different western PWR assembly designs studied ($\boldsymbol{c_k}$ > 0.995). The similarity, evaluated on a nuclide-by-nuclide and reaction type basis using the g values, indicates that many nuclides exhibit g values > 0.9, with some exceptions. The large g values indicate that the reaction rates will respond in a similar fashion to biases in the cross section evaluations for criticality applications. Some caution is needed in extrapolating the similarity to depletion applications (i.e., assumption that the cross sections will result in similar isotopic composition biases between the two systems). For example, ^{241}Am and ^{155}Gd in spent fuel are generated primarily by ^{241}Pu and ^{155}Eu decay after discharge from the reactor. Therefore any similarity for ^{241}Am and ^{155}Gd during irradiation is not necessarily of good measure after discharge. For a number of nuclides, the differences in the g values between the VVER 440 and PWR systems are observed to be much greater than that between the two PWR systems studied. This is likely caused by the harder neutron spectrum in the VVER 440 fuel relative to PWR fuels.

Although the systems are found to be highly similar for many nuclides of importance to burn-up credit, it is not possible, without further study, to determine the degree to which the VVER 440 isotopic assay measurements can be applied to validate depletion code predictions for western-design PWR assemblies and fuel. This conclusion is reached even though the conventional criteria for the global c_k index used for criticality has been met (c_k > 0.9). The additional requirement that each of the g values also be greater than 0.9 for the major actinides and fission products is deemed appropriate because the individual isotope values are to be used for validating the isotopic predictions on a nuclide-by-nuclide basis. Additional work is needed to evaluate the importance of the nuclide cross-section sensitivity differences on the prediction of isotopic compositions.

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EXPERIMENTAL TECHNIQUES FOR SPENT NUCLEAR FUEL

Chairs: B. Gmal, G. Hordosy

ISOTOPIC AND ELEMENTARY ANALYSIS TECHNIQUES APPLIED TO THE SPENT NUCLEAR FUEL CHARACTERISATION

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Abstract

The precise knowledge of the inventory of the radionuclides for various stages of the fuel cycle is mandatory for the comprehension of the management of the nuclear fuel and the neutronic calculation codes qualification. The quantitative determination of the radionuclides requires specific methods for sample preparation, chemical separation of the various elements and finally isotopic and elementary measurements.

The Laboratory of Nuclear, Isotopic and Elementary Analysis (LANIE) of the Chemical Physics Department (DPC) of the French Nuclear Research Center at Saclay (near Paris) operates as expert in the field of the isotopic analysis of high precision applied to the characterisation of irradiated nuclear fuels.

Mass spectrometry techniques which require specific and complex equipments permit the precise isotopic characteriSation of the radio elements after separation of the various elements to eliminate some possible isobaric interferences and matrix effects.

The best performances in accuracy and repeatability are obtained by Thermo Ionisation Mass Spectrometry (TIMS) and by Multi Collector Mass Spectrometry with Inductively Coupled Plasma (ICP-MS-MC). In the case of elementary analysis, accuracies of about 0.1% can be reached by using the isotope dilution technique after separation of the various elements of the sample. Atomic ratio of the different isotopes of the element relative to ^{238}U can be obtained. When only one isotope has to be determined or in the case of mono isotopic elements, quadrupole ICP-MS is used to determine the concentration of the isotope of interest without separation. 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 %.

These techniques, extremely significant, are developed for the characterisation of irradiated nuclear fuels in order to qualify the neutronic codes to gather data for a better management of fuels.

The different techniques used in the laboratory for irradiated fuel samples analysis will be presented: Q ICP-MS, MC ICP-MS and TIMS. The general principles of an analysis of a nuclear irradiated fuel will be presented including some recent developments and some new measurements performed in our laboratory.

Introduction

Laboratory of Nuclear, Elementary and Isotopic Analysis develops and implements high performance chemical specific separations and measurement techniques for trace level analyses. It is an expert laboratory for the isotopic analysis of spent fuel. Actinides and fission products in spent nuclear fuel samples are measured with a few per thousand to a few per cent accuracy and uncertainty [1-3]. Our laboratory is a support to EDF and AREVA for the qualification of neutronic codes, development of new fuels, and in the studies for increasing the burn-up rates.

In the nuclear domain, isotopic measurements are usually performed using the Thermo-Ionisation Mass Spectrometry (TIMS). Elementary measurements or analysis of mono isotopic fissions products can be easily performed by Inductively Coupled Plasma Mass Spectrometry (ICPMS). Multiple Collector ICP-MS (MC ICP-MS) can improve the determination of precise and accurate isotopic compositions of radio elements in spent nuclear fuel samples due to its high sensitivity and time saving compared to TIMS. This instrument is available in our laboratory and used for example for the determination of isotopic composition of rare earth elements and some other fissions products in spent nuclear fuel samples.

All of these techniques are combined with liquid chromatography if necessary and will be described. Analytical results will be presented.

Dissolution procedures

The analytical techniques used in our laboratory are limited to the measurement of liquid solutions. The CEA owns two dissolution facilities both located in the south of France. About 10 g of fuel are solubilised in one of the two and transported in solution to Saclay. Each team has its own dissolution procedure which gives to the CEA two complementary dissolution procedures at disposal.

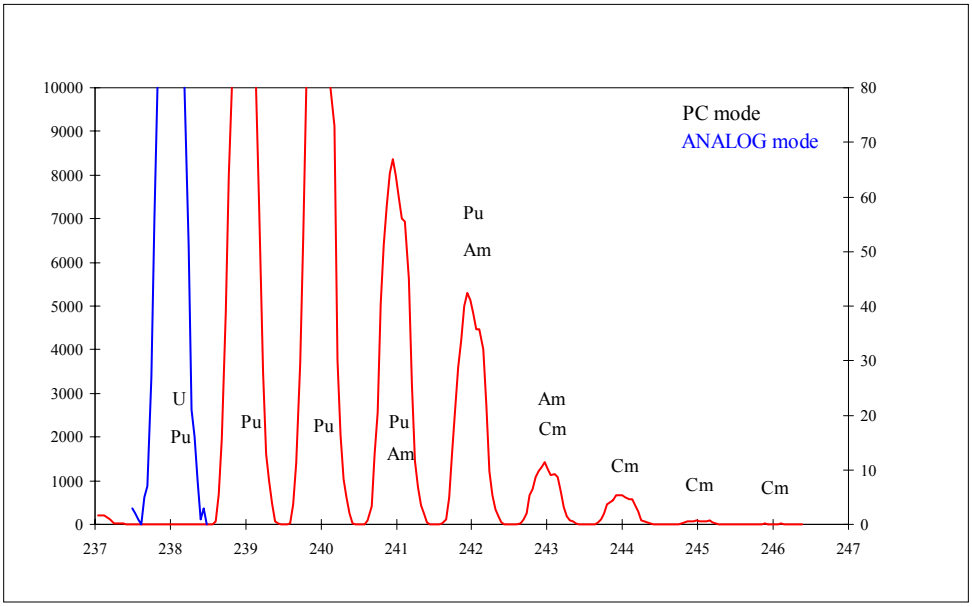
The facility of COMIR is located in Cadarache. The procedure consists of a single stage nitric attack under very corrosive conditions in regards to acidity and temperature. A small quantity of HF is added to the nitric acid. Nevertheless, it has be noted that this procedure can produce very small quantities of insoluble residues. With this procedure, one unique nitric solution is obtained and analysed.

The facility of Atalante is located further north in Marcoule where a total dissolution (which means a total absence of insoluble) of part of the rod is performed. The procedure is divided in four stages. The first stage consists of a nitric attack under quite soft conditions if compared to the procedure in COMIR. A first solution S1 is thus obtained. The residues are then collected, attacked by sintering with sodium peroxide and finally poured in water. After filtration of the remaining residues, a second solution is obtained and named S2. The remaining residues are then washed by nitric acid. The nitric solution is S3. At last a second sintering is performed with potassium persulfate solubilising the last remaining insoluble residues. As a conclusion, 4 solutions are obtained and transported to Saclay for characterisation.

Analytical techniques

As can be seen in the mass spectrum shown in Figure 1, there is an overlap between some isotopes of the actinides : U and Pu at mass 238, Pu and Am at mass 241 and 242 and Am and Cm at mass 243. It is not possible in 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 spectrometry techniques (TIMS or MC ICP-MS).

Figure 1. Mass spectrum of spent nuclear fuel sample, actinide zone, showing overlap between some isotopes



Chromatographic separations

At first, U/Pu/fission products + minor actinides separation is performed on the irradiated fuel sample solution. Fission products and minor actinides are eluted first in nitric acid 8M by gravitational chromatography on an opened column with an ion exchange resin. The procedure involves Dowex AG1X4 resin and has been previously described [1]. Uranium and plutonium are then isolated by elution with different nitric acid concentrations. The U and the Pu fractions are then ready to be analysed by TIMS, since the isobaric interference at mass 238 (U, Pu) has been resolved.

The fraction not retained by the resin (fission products and minor actinides fraction) is then used to separate all the fission products and Am and Cm by high performance liquid chromatography (Figure 2). The separation is performed under gradient elution by using a 2-hydroxymethylbutyric acid (HMB) as eluent.

Cs is detected with the scintillation detector. The lanthanides are detected by UV - visible detection after post column derivatisation and Am and Cm are detected simultaneously by the two detectors. An example of chromatogram obtained is shown in Figure 3. Each fraction obtained after separation is ready to be analysed by mass spectrometry.

Thermal Ionisation Mass Spectrometry (TIMS)

A scheme of the technique is presented Figure 4. An amount of the element of interest (about a few tens of ng to some µg) is deposited on a filament. The filament is heating in a vacuum source allowing the atomisation of the element deposited and then the ionisation of the atoms. The ions thus formed are separated according to their mass/charge ratio by a sector field. Currents of ions corresponding to each isotope present are then detected in a simultaneous way by the use of judiciously settled collectors.

Figure 2. Scheme of the separations method

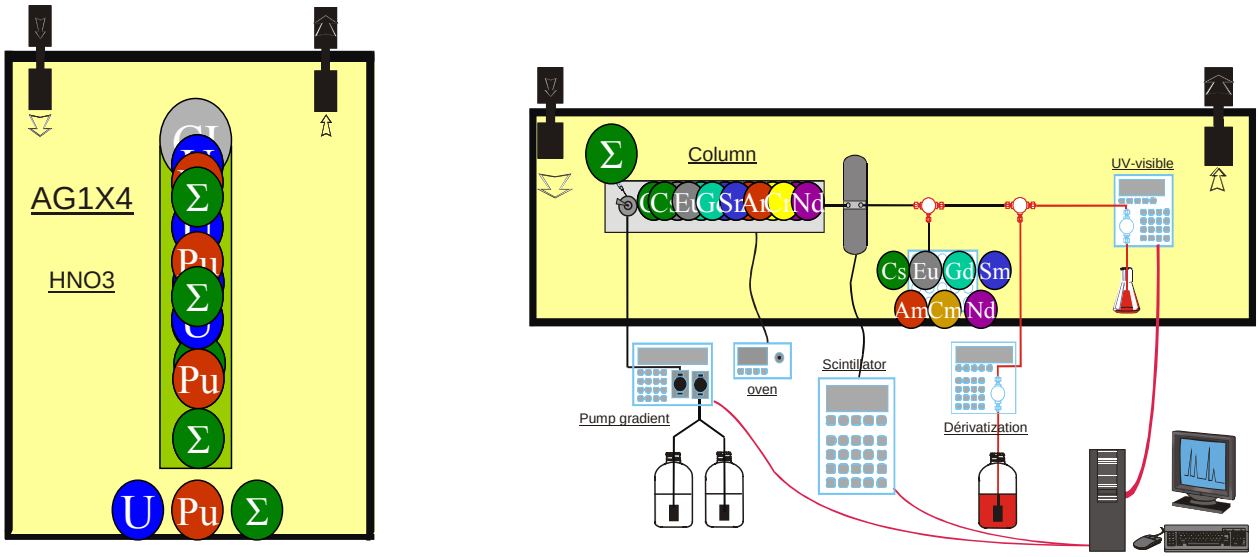


Figure 3. Chromatogram of the separation of lanthanides, Cs, Cm and Am (UV detector: top, scintillation detector: bottom)

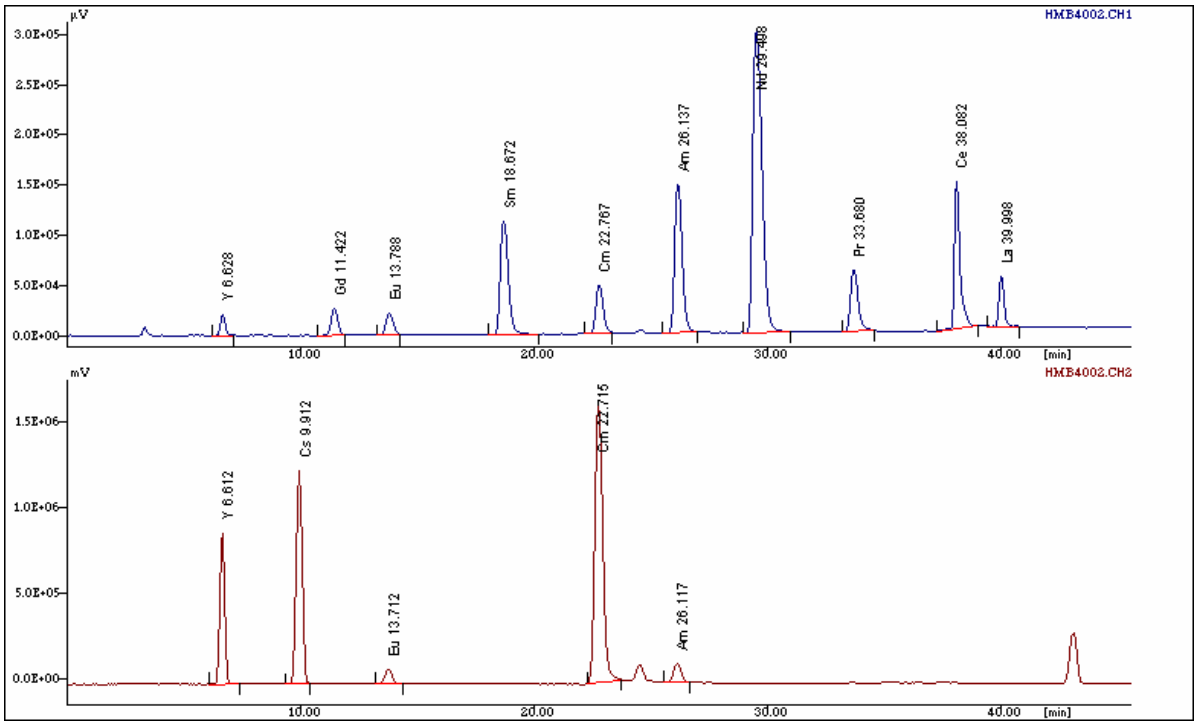
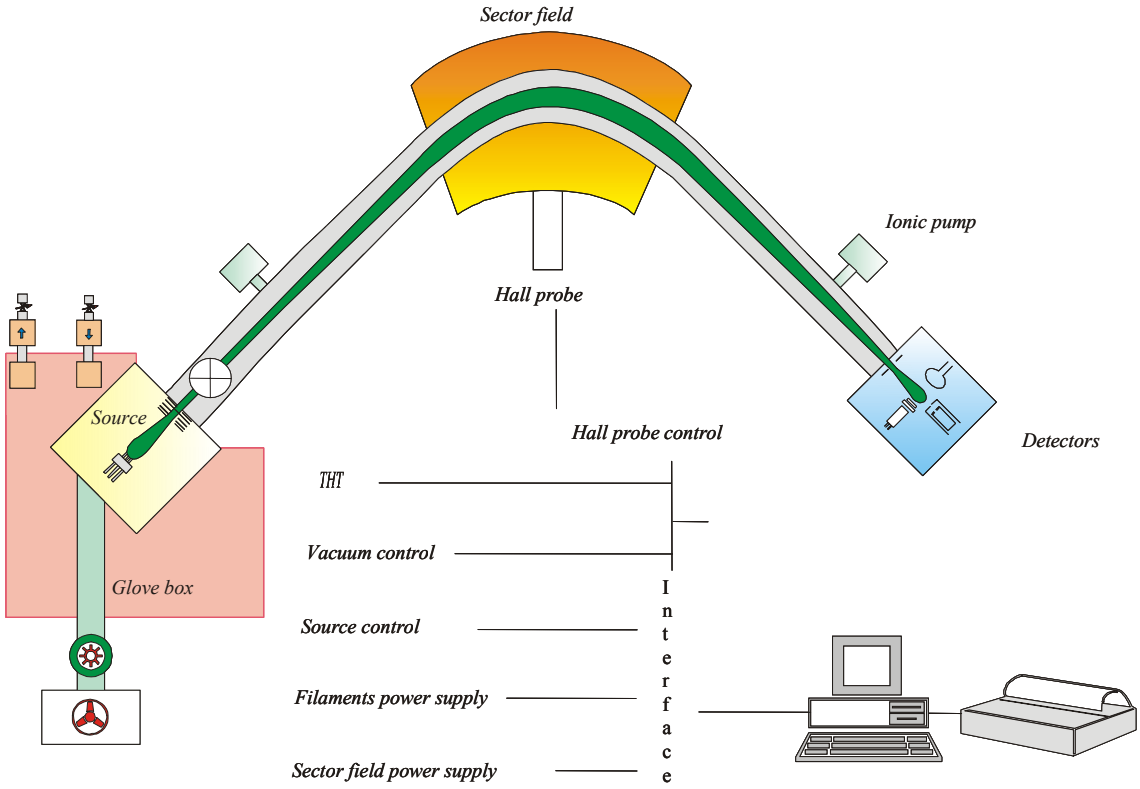


Figure 4. Thermal Ionisation Mass Spectrometer



The quality of the analysis is related to different phenomena which govern the accuracy of the results. Among the most important is the isotopic fractionation. Indeed, the thermal ionisation has for inconvenience to introduce a systematic bias of some one per thousand. Fractionation is not constant but decreases with time. It is independent from the isotopic content but depends on the range of mass. To take into account the isotopic fractionation, there are several analytical solutions. The first one is to use isotopic standards to adapt the analytical procedure to obtain highly precise and accurate ratios. The second one is the flash evaporation method. With this method, the sample is entirely volatilised while the signal from each isotope is integrated. Thus, the effect of isotope fractionation is eliminated. The quality of the 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.

It is necessary to note that the concentration of an element, if it possesses several isotopes, can be measured by TIMS or MC ICPMS in a very precise way with the isotope dilution technique [1,4]. This technique is only based on measurement of isotopic ratios. The principle is to add to the element to measure a known quantity of the same element but with a different isotopic composition, the spike. Knowing perfectly the isotopic composition of the spike and its concentration, the measurement of the element isotope composition in the sample and in the mix (sample + spike) and with the equations of the isotope dilution technique, it is possible to obtain the initial concentration of the element with an accuracy and uncertainty which depends on accuracy of the isotope ratios measurements. Typically, accuracy and uncertainty of few per thousand can be obtained by this way. Another advantage of this technique is that the yield of the chemical separation does not need to be known or constant because only isotopic ratios are measured.

The double spikes isotope dilution technique is usually applied in our laboratory to determine the concentrations of actinides and fission products (Cs, Sr, Nd, Am, Cm, Gd, Eu, Sm,…) with respect to ^{238}U by TIMS and MC ICP-MS. Some results of elemental isotopic composition and relative atomic ratios of one radio nuclide to ^{238}U are shown in Table 1. In all cases 2 repeats with independent sample preparation and analysis are performed.

Table 1. Results obtained by TIMS for Cs and U isotope compositions and Am/U and Nd/U ratios by isotope dilution technique

Isotopic composition Cs				Isotopic composition U			
	133/135	134/135	137/135		234/238	235/238	236/238
fraction A	2,5004	0,07005	2,6070	fraction A	0,000174	0,003747	0,007289
fraction B	2,5015	0,07004	2,6051	fraction B	0,000173	0,003752	0,007304
Mean	2,5010	0,07005	2,6060	Mean	0,000174	0,003750	0,007297
Δ %/ Mean	0,04	0,01	0,05	Δ %/ Mean	0,57	0,09	0,15

Ratio Am/U (241Am/238U)	
Mix A	2,556E-03
Mix B	2,557E-03
Mean	2,556E-03
Δ %/ Mean	0,04

Ratio Nd/U (145Nd/238U)	
Mix A	1,658E-03
Mix B	1,656E-03
Mean	1,657E-03
Δ %/ Mean	0,07

Quadrupole Inductively Coupled Plasma Mass Spectrometry (ICPMS)

With this technique, a liquid solution is continuously introduced as an aerosol in an argon plasma produced by application of an RF field (the applied power is typically 1 200-1 400 W). The temperature within the plasma being above 5 000 K, the spray is vaporised, desolvated, atomised and atoms obtained are ionised. The ions are then separated according to their ratio mass/charge by a quadrupole. The separated ions are then detected with an electron multiplier. The sequential measurement of the various ions currents produced, allows then after calibration with standards to perform a quantitative analysis of the various isotopes. There are some limitations with this technique.

It can be quoted first of all the relatively weak mass resolution with Q ICPMS. Indeed, quadrupole system presents mass resolution in the order of one mass unit, limiting the field of application. This is particularly penalising in the case of fuels analysis where numerous isobaric interferences are present. As an example and within the framework of burn-up credit, is the case of the ^{109}Ag which is interfered by the molecular ion $^{93}Zr^{16}O$ at the level of few %. This interference must be taken into account by the measurement of natural zirconium and its oxide on standard solution to perform a correction on the ^{109}Ag measurement because the mass resolution is not sufficient to resolve this interference. It is necessary to add to this example, the recombination of molecular ions in the plasma. These ions are mainly hydrides, oxides or based on argon recombination.

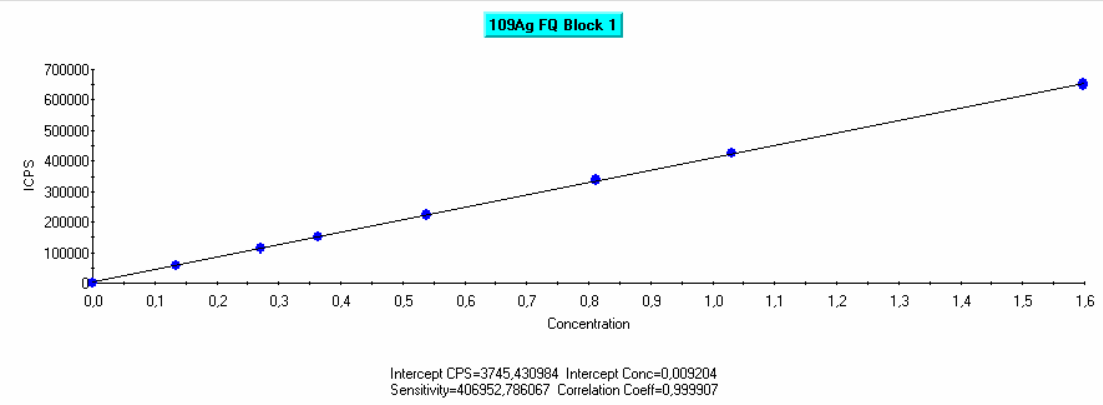
A collision cell is available on our Q ICPMS (Thermo X7). It can be declined under various forms according to the provider of ICPMS: quadrupole, hexapole or octopole. Besides to reduce the widespread energy, its potential function is to reduce the interferences as doubly charged ions, poly atomic ions by collisions with a gas.

Q ICPMS does not allow determining the isotopic compositions with a so good accuracy and uncertainty as TIMS does. Indeed, a difference between the isotopic ratios measured by Q ICPMS and the true isotopic ratios is always observed. Even if this mass bias is taken into account by analysing isotopic standards, the accuracy that can be obtained in isotopic measurements with a Q ICPMS is, in the favourable cases, of the order of 1%.

Q ICPMS is used here for the elemental concentration measurement for which only one isotope is of interest: ^{79}Se, ^{93}Zr, ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{107}Pd, ^{126}Sn and ^{109}Ag. The measurement is performed without preliminary chemical separation, with associated uncertainties of a few %.

Typically, a calibration curve is obtained with a minimum of 6 standards of different concentrations. For each standard, a minimum of 20 replicates is realised in order to obtain a good repeatability of the measurements. Internal standards are used (indium and bismuth for example) in order to correct a potential deviation of the instrument. An example of a calibration curve obtained in Q ICPMS is shown in Figure 5.

Figure 5. Calibration curve obtained in Q ICPMS for ^{109}Ag between 0.1 and 2 ng/ml



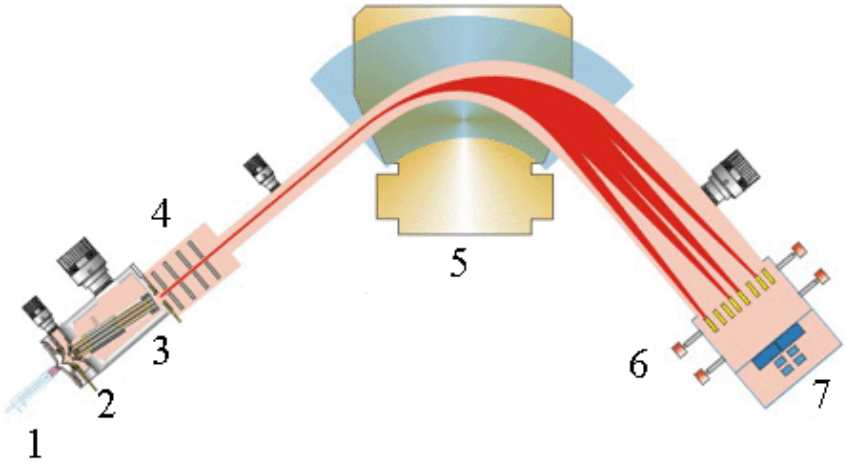
Multi Collection Inductively Coupled Plasma Mass Spectrometry (MC ICPMS)

The laboratory is also equipped with a MC ICPMS (Isoprobe, GV Instruments). This kind of ICPMS allow to determine the isotopic composition of an element with the same accuracies than the one obtained by TIMS. It is presented in Figure 6. These elements can be separated in 3 parts:

The first part of this instrument is constituted by the sample introduction system and by the plasma torch, the whole is noted 1 on Figure 6 (ICP). The second part consists of the interface, the collision cell (hexapole based) and the ionic optics, tracked down by the numbers 2-4. The last part is constituted by the magnetic analyser and by the multi collection detection system (5-7). The sample introduction system is based on the same principle as for quadrupole ICPMS.

The interface allows to extract the ions from the argon plasma while allowing at the same time the decrease of temperature and the passage from atmospheric pressure to the mass spectrometer vacuum. The interface between the torch and the collision cell is constituted by three nickel cones. The first one, the sampler serves for extracting the ions of the plasma. The skimmer takes the centre of the supersonic jet and finally, the last cone, the extraction cone, allows to extract the ions to bring them to the collision cell.

Figure 6. Schema of the Isoprobe (GV Instruments)



The ionic optics is a system constituted by electrostatic lenses which allow to focus and to accelerate the ions. They also allow to stop photons so that they do not affect the detectors. The sector field allows to separate the ions according to their mass to charge ratio (m/z).

The detection system is constituted by several collectors which allow measuring several masses simultaneously. The Isoprobe is equipped with 9 Faraday collectors and a Daly electrode. These detectors are used for concentrations in the ppt to ppm range. The multi-collection allows consequently quantifying at the same time several isotopes of the same element or several elements of more or which allow greatly improve the reproducibility of the measurements.

The analytical procedure applied is the standard bracketing technique. Isotopic analysis of the element is performed and before and after the sample, an isotopic standard is analysed in order to determine the mass bias that has to be applied to the element of the sample. An accuracy of 5 $^{0}/_{00}$ is easily achievable for the most abundant isotopes. As previously described for TIMS measurements, two repeats with independent sample preparation and analysis are systematically performed. Validation of the analysis is performed by the bias between the two measurements.

MC ICPMS is used for example with preliminary chemical separations for the determination of the isotopic composition of samarium, europium and gadolinium as well as for the determination of ratios Sm/^{238}U, Eu/^{238}U and Gd/^{238}U by double spikes isotope dilution technique. Some results have been published [3,5] and are shown in Table 2.

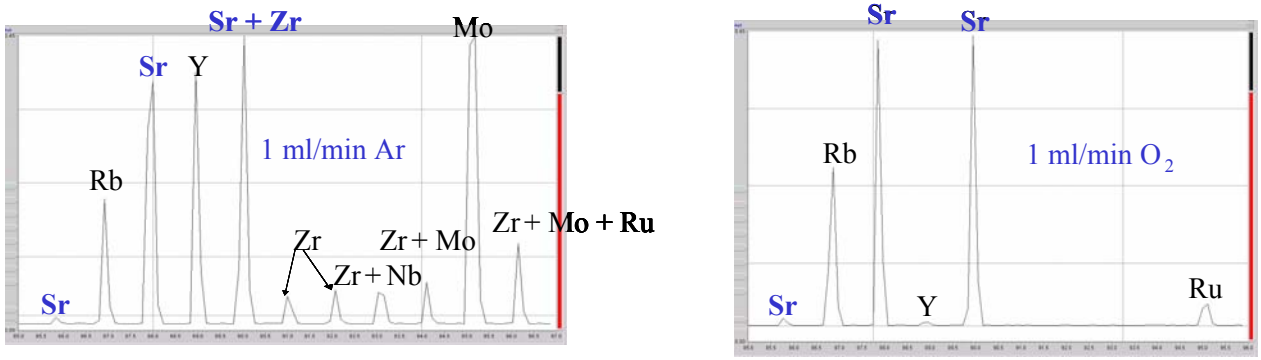
Developments in progress

Recently, appears on the ICPMS, a new tool to improve analysis performed by ICPMS, the collision-reaction cell. It is, as previously mentioned, a multipole set in a chamber under vacuum where a gas can be introduced in order to perform collisions to brake molecular ions and/or to perform ion molecule reactions to eliminate mass interferences. As an example, Figure 7 shows the mass spectra of spent nuclear fuel under 1 ml/min Ar (collision gas) and the same sample with 1 ml/min O_2 (reaction gas) with the isobaric interference ^{90}Sr/^{90}Zr. It can be seen a simplification of the spectrum in Figure 7b. There is no overlap between Sr and Zr since all the Zr reacts with O_2 to form the oxide ZrO^+. By the use of ^{84}Sr as spike with the isotope dilution technique, it is also possible to measure ^{90}Sr/^{238}U in the fuel solution without taking care of pollution with natural strontium [3].

Table 2. Results obtained by MC ICPMS for Gd isotope compositions and Gd/U atomic ratios by the double spikes isotope dilution technique

		+-
$^{154}Gd/^{156}Gd$	0.11685	0.00006
$^{155}Gd/^{156}Gd$	0.03305	0.00018
$^{157}Gd/^{156}Gd$	0.00096	0.00010
$^{158}Gd/^{156}Gd$	0.20542	0.00004
$^{160}Gd/^{156}Gd$	0.01565	0.00008
$(^{158}Gd/^{238}U)_A$	0.00010480	Δ(A–B)%
$(^{158}Gd/^{238}U)_B$	0.00010478	0.027
$(^{155}Gd/^{238}U)_A$	0.00001686	Δ(A–B)%
$(^{155}Gd/^{238}U)_B$	0.00001685	0.026
$(^{154}Gd/^{238}U)_A$	0.00005961	Δ(A–B)%
$(^{154}Gd/^{238}U)_B$	0.00005960	0.027

Figure 7. Mass Spectra of spent nuclear fuel solution obtained in MC ICPMS with 1 ml/min Ar in the collision cell (left) and 1 ml/min O_2 (right) in the range 85-97 uma



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SSC RF RIAR'S EXPERIMENTAL POSSIBILITIES CONCERNING INVESTIGATION OF IRRADIATED NUCLEAR FUEL FOR VERIFICATION OF BURN-UP CREDIT CALCULATIONS

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Abstract

Combination of data on radiochemical analysis and irradiated nuclear fuel (INF) reactivity measurements, obtained as a result of complex experiment, give us an unique possibility for validation of burn-up credit calculations. Such investigation of INF can be performed at FSUE "SSC RIAR" and is determined by availability of:

- spent VVER UO_2-fuel including with burning up absorbers over the wide range of burn-up at different initial enrichment;
- fresh VVER UO_2-fuel;
- hot cells for investigation and re-fabrication of fuel rods;
- radiochemical methods of analysis;
- critical stands and research reactors for reactivity measurement.

Complex investigation of INF includes:

1. Performance of radiochemical analysis in the volume of the work fulfilled under VVER-440 spent fuel investigation (A.P. Chetverikov, *et al*., Investigation of Burn-up and Nuclide Composition of Spent Nuclear Fuel for Use when Solving "Burn-up Credit" Tasks – RIAR Experience, IAEA, 29 August 2005-2 Sept. 2005, London, see proccedings at the following address: http://www.llnl/tid/pdf/documents/pdf/319467.pdf.

2. Fresh and burn-up fuel reactivity measurement.

In order to measure INF reactivity, different variants of model FA from fuel rods with non-irradiated and irradiated fuel of different burn-up will be investigated in the pool-type water-moderated reactor RBT-6. Error of reactivity effects measurement is not more than 5%.

The resulting data can be used for verification of the calculation codes, with the help of which system neutron multiplication factors on all stages of INF handling (storage, transport, reprocessing, etc.) are calculated.

Introduction

Development of nuclear industry inevitably leads to increase in loading of interim water and long-term dry storage facilities with spent nuclear fuel (SNF) and to necessity of clarifying basing of their safety due to rise in enrichment of using NPP fuel and tendency to tight placement of SNF. Ways of burn-up credit calculation are widely discussed and introduced for solving these questions at present. Burn-up credit is calculated by means of codes according to results of reactor power control in different periods of its operation. Accuracy of using calculation codes is verified basing on radiochemical analysis data and by means of experimental determination of SNF effect on multiplying characteristics (reactivity) of systems with fissile materials. The most reliable data for verification are obtained during complex application of these methods as e.g. it has been performed in international program "Rebus" intended for PWR reactor fuel [1]. There are no programmes of such investigations for VVER reactor fuel. The programme of complex investigation of spent VVER-440 UO_2-fuel is introduced in the present work. Aim of the programme is carrying out of experiments regarding verification of reactor physics codes for calculation of reactivity loss because of fuel burn-up at energetic reactor spent fuel rods. The following methods are used: radiochemical analysis and measurement of reactivity in reactor facility. The investigations can be performed at SSC RF RIAR since unique experimental possibilities of its scientific and technological base find an application for full-scale reactor investigations of VVER-440, VVER-1000, RBMK-1000 and BN-reactors' FAs including the problem regarding investigation of burn-up credit at all stages of SNF handling as well. There are irradiated fuels of different types, technological equipment for radiochemical analysis, fuel rod re-fabrication and investigation of reactivity at channels of critical stands and research reactors at RIAR. The complex investigation of SNF includes the following milestones:

- fuel selection;
- gamma-spectrometric and material science investigations;
- radiochemical analysis in volume of the work [2], performed under investigation of spent VVER-440 fuel;
- fabrication of experimental (model) fuel rods and FA from non-irradiated and irradiated fuel;
- reactivity measurement.

Fuel selection

In the beginning the fuel at 3.6% initial enrichment ^{235}U was used in VVER-440. Large volume of spent FAs with this fuel is accumulated for the years of operation. Mass introduction of the fuel at 4.4% enrichment has been commenced since 1999, and it will need to be utilised in the near future as well. So complex investigations should be carried out regarding fuel at both enrichments.

Table 1 shows data on availability of spent FAs VVER-440 at RIAR.

Table 1. Characteristics of spent FA VVER-440 storing at SSC RF RIAR

No.	FA No.	Date of unloading	Reactor (NPP), unit	U-235 enrich-ment, %	Operation time, eff. days	Average burn-up, MWt day/kg U
1	Д11066	23.06.86	Rovenskaya- 1	3.6	857.6	36.8
2	Д11809	06.08.86	N-Voronezhskaya- 4	3.6	994.8	32.8
3	Д15687	06.08.86	N-Voronezhskaya- 4	3.6	605.2	24.2
4	Д19159	16.08.88	N-Voronezhskaya- 4	3.6	989.1	34.1
5	Д26135	04.07.91	N-Voronezhskaya- 4	3.6	1 109.7	38.5
6	236-35228	15.02.97	N-Voronezhskaya- 4	3.6	1 514.0	50.5
7	13642198		Kolskaya -2	3.6	1 242.3	44.7
8	Д52380	18.06.99	Kolskaya- 2	4.4	649.1	27.6
9	E22198	01.11.90	Kolskaya- 3	4.4	1 259.8	46.2
10	E22222	25.10.91	Kolskaya- 3	4.4	1 564.9	48.2
11	144-46879	14.08.02	Kolskaya- 3	4.4	1 871.4	56.5

As the table shows, there are FAs with wide range of burn-up and at different fuel enrichments. Composition of non-irradiated fuel is described in detail in the documents during its fabrication.

Gamma-spectrometric and material science investigations

Gamma-spectrometric investigations are performed in order to obtain data on axial and radial distributions of fuel burn-up in FA. The base of measurements is method of replacement, in which burn-up depth in investigating fuel is determined relative to the known value of burn-up in a standard specimen. At the same time intensity of gamma radiation of ^{137}Cs fission product is measured. Burn-up distributions throughout the height of fuel rod No. 69 of FA No. 135 and the cross-section of this FA for fuel rods of diagonal 1-126 are demonstrated in Fig. 1 and 2 as an example. Fig. 3 shows schematic arrangement of fuel rods in the FA.

Fig. 1. Distribution of fission products throughout the height of fuel rod

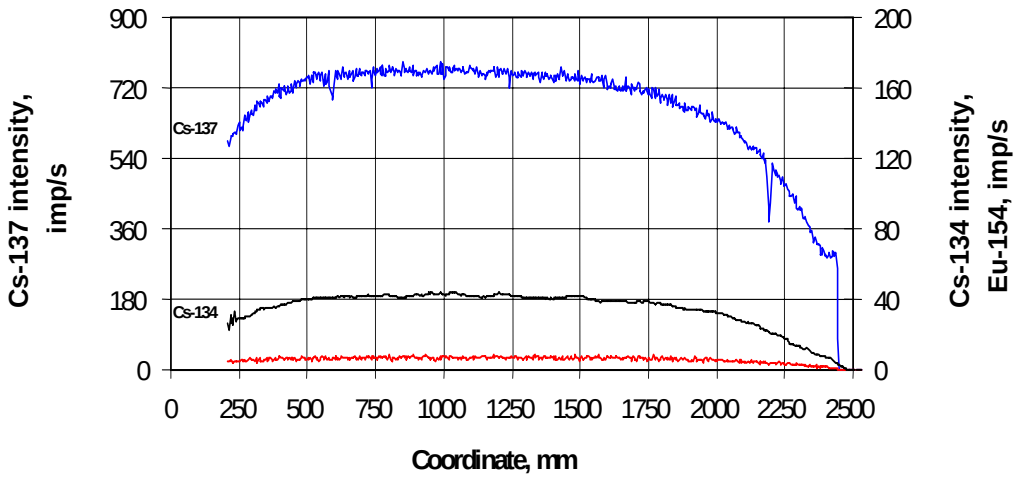


Fig. 2. Distribution of maximum burn-up at fuel rods in diagonal directions of the FA

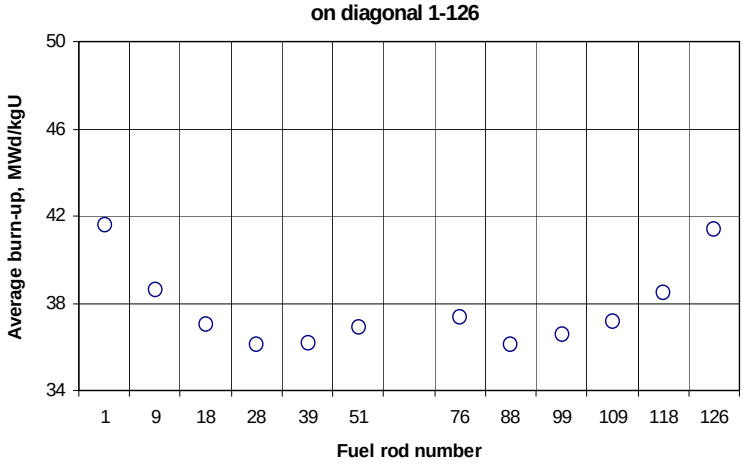
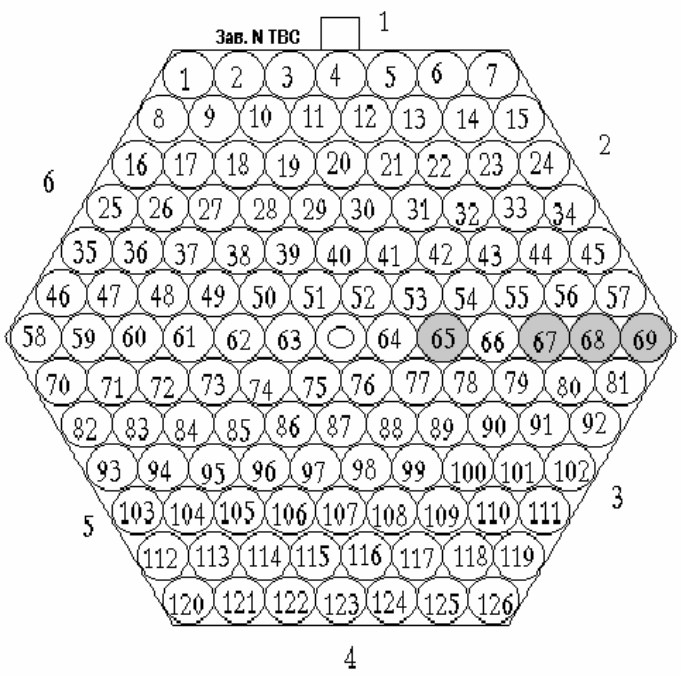


Fig. 3. Schematic arrangement of fuel rods in the FA



Gamma-scanning results are initial data on determination of co-ordinates of fuel specimen selection for radiochemical investigations, fabrication of model fuel rods for reactivity measurement experiments.

Material science investigations in their full volume are not necessary for burn-up credit calculations. They have independent importance for validation of operational reliability of fuel rods, FA and new kinds of fuel. But part of these investigations is necessary for determination of experimental fuel rods characteristics, which are required for further calculation modelling of reactivity measurement experiments. Material science investigations include determination of:

- fuel rod lengthening;
- fuel rod diameter, distribution of its values throughout the length;
- thickness of oxide film along cladding;
- quantity and composition of gas under fuel rod cladding and dissolved in solid phase of fuel core;
- fuel-cladding gap;
- structure and dimensions of fuel seed;
- distribution of nuclides and burn-up throughout fuel core radius including in "rim"-layer as well;
- carrying out of eddy-current detection of fuel rod cladding.

The aim of *radiochemical analysis* is determination of content of actinides, burn-up monitors ($^{145+146}$Nd, ^{148}Nd) and the most important fission products providing >80% of neutron absorption in irradiated fuel (Table 2).

Table 2. Nuclides, content of which is subject to determination

Actinides	^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, ^{244}Cm, ^{245}Cm
Burn-up monitors	$^{145+146}$Nd, ^{148}Nd
Fission products	^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd, ^{133}Cs, ^{134}Cs, ^{135}Cs, ^{137}Cs, ^{140}Ce, ^{142}Ce, ^{144}Ce, ^{147}Sm, ^{148}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{154}Sm, ^{151}Eu, ^{153}Eu, ^{154}Eu, ^{155}Eu, ^{95}Mo, ^{99}Tc, ^{105}Pd, ^{108}Pd, ^{109}Ag, ^{155}Gd

Radiochemical analysis consists of quantity chemical dissolution of irradiated fuel specimen, determination of isotope composition and mass of element by means of isotope dilution method with mass-spectrometric completion of the analysis. ^{242}Cm and ^{244}Cm content is determined by alpha-spectrometer method, ^{237}Np – by spectrophotometric one. Error of determination by mass-spectrometric and alpha-spectrometric analyses is in the range 2.5-6%, ^{237}Np determination error – 10% under confidence probability 0.95.

Burn-up depth is calculated on the basis of $^{145+146}$Nd content measurement results. The obtained data on nuclide composition and fuel burn-up is used for verification of burn-up credit calculations.

Quantity of fuel specimens for radiochemical analysis is determined during discussion regarding performance of the experiment.

Fabrication of experimental (model) fuel rods and FA for reactivity measurements

Measurement of 8 model FAs is anticipated under the present programme. These ones are as follows:

- one FA with non-irradiated fuel at 3.6% enrichment;
- three FAs with irradiated fuel with burn-up of 40, 50 and 60 MW·day/kgU and at 3.6% initial enrichment;
- one FA with non-irradiated fuel at 4.4% enrichment;
- three FAs with irradiated fuel with burn-up of 50, 60 and 70 MW·day/kgU and at 4.4% initial enrichment.

Each FA consists of 18 model fuel rods with length of fuel core 200 mm. This size is selected taking into account the height of active zone of the reactor where the experiments are planned to carry out.

Fuel rods of FAs Nos. D26135, 13642198 and 236-35228 (Table 1) can be used for fabrication of model FA with fuel at 3.6% initial enrichment. Fuel rods with burn-up of 40, 50 and 60 MW·day/kgU are selected according to the data on axial and radial burn-up distributions. Fragments with length ~ 1 500 mm (Fig. 1) and uniform burn-up distribution are cut out from the selected fuel rods. ~ 7 re-fabricated fuel rods with fuel core with length 200 mm are fabricated from each such fragment. Model FA cluster is assembled from 18 re-fabricated fuel rods with burn-up of 40 MW·day/kgU. Model FAs are similarly assembled from re-fabricated fuel rods with burn-up of 50 and 60 MW·day/kgU.

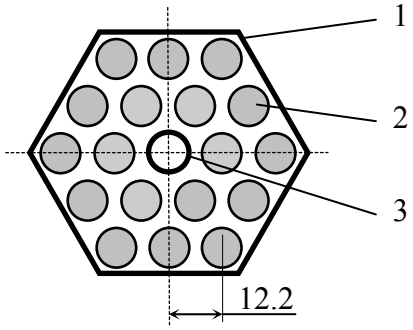
Fuel rods from FA Nos. E22198, E22222 and 144-46879 with burn-up of 50, 60 and 70 MW·day/kgU are selected for fuel at 4.4% initial enrichment. Three model FAs containing 18 re-fabricated fuel rods each are fabricated from fragments of these fuel rods with uniform burn-up distribution.

Experimental FAs with non-irradiated fuel are constructively made in the same way as the FAs with irradiated fuel, and contains 18 re-fabricated fuel rods each.

Fig. 4 shows geometry of experimental FA cross-section. All experimental fuel rods are passed through certification (documentation of nuclide composition of fuel composition and geometrical dimensions). Construction of experimental FA is worked out along with drawing up project of irradiating device and approving project documentation by state control authorities.

Fig. 4. Experimental FA cross-section

1 – hexagon casing (zirconium alloy; across flats dimension 53.4 mm, thickness 1 mm); 2 – short-cut fuel rod of VVER reactor (length of active part 200 mm; casing - ∅9.15x0.65 mm from zirconium alloy; fuel pellets - ∅7.5x3.05 mm of Uranium oxide with density 10.4 g/cm^3 and at U^{235} enrichment – 4.4%); 3 – central tube (zirconium alloy; ∅ 13x1 mm).



Reactivity measurement

Experiment scheme

Preliminary analysis lets us draw a conclusion that from the whole variety of reactor facilities and critical stands at RIAR it is expedient to use reactor RBT-6 for reactivity measurement experiment. This conclusion is caused by:

- availability of pool reactor;
- absence of principal restrictions regarding time of the experiment performance;
- possibility to carry out detailed investigations during use of the reactor under critical assembly operating regime, i.e. at minimally controlling level of power;
- possibility to implement "big" reactivity effects when loading the experimental FAs with irradiated and non-irradiated fuel (<1% Δk/k);
- availability of verified mathematical reactor models allowing calculation of reactivity effects from 0.5% Δk/k with an error not more than 5% relative to value of the measuring effect;
- availability of certified experimental methods and reactivity change controlling means when loading-unloading the experimental FAs;
- availability of large base of earlier obtained experimental data on reactivity effects and experience of reactor use for implementation of various research programmes including the ones under IAEA contracts [3];
- availability of technology of irradiated fuel handling (transport-technological equipment, shielded box, cooling pond and etc.);
- location of FA in the reactor along square lattice with water gap between them, i.e. in geometry, which is often used in irradiated fuel cooling ponds;
- availability of controlling means as cadmium plates located in gaps between FAs, i.e. in geometry of neutron plate absorbers, which are used at cooling ponds and transport containers in a number of cases.

Milestones of the experiment preparation and performance:

1. Implementation of preliminary neutron-physical, heat-hydraulic calculations and analysis of the experiment performance safety.

2. Carrying out of the experiments in the reactor with sequential loading of experimental assemblies (with fresh and irradiated fuel) into cell of experimental channel in active zone of the reactor and with determination of critical positions and reactivity effects from experimental FA loading.

3. Implementation of verification calculations using the experiment results following the possible variants:

   – calculation by RIAR's employees according to certified code MCU [4] by means of verified mathematical models of reactor RBT-6;
   – calculation by RIAR's employees according to the codes provided by customer (it takes some time to master the code and to develop calculation model);
   – passing the detailed experimental data to customer (detailed description of the reactor and collaborative work with customer on development of calculation models are required).

Brief description of reactor RBT-6

Pool water-moderated thermal reactor RBT-6 is intended for carrying out of experiments on change of material properties during long-term irradiation [5]. The reactor powers up to 6 MW. Active zone of the reactor is situated in the vessel filled with distilled water (Fig. 5). Height of the active zone is 350 mm. 56 FAs of square section are placed in the active zone along the square lattice (Fig. 6). The rest 8 cells are intended for placement into experimental channels. Reactor RBT-6 operates in a regime

of partial reloading of fuel, the spent FAs of reactor SM are used as which [5]. Water is as a moderator, a reflector and a coolant.

Six compensating controllers, each of which is two neutron absorber plates of sheet cadmium as half of disc canning with stainless steel, and automatic control rod are used as controllers (AZ-KO controllers). The plates of compensating controllers are moved in gaps between FAs. When turning (Fig. 5) they completely enter the active zone from two its side edges at a depth of three rows of FA. During withdrawal part of the plates move is used for emergency protection, the rest – for compensation of reactivity margin and assurance of reactor initial subcrit. Automatic control rod section is of T-shape with cadmium absorber canning with stainless steel. It is vertically moved.

Fig. 5. Vertical section of reactor RBT-6

1 – suction pipe; 2 – supporting structure; 3 – active zone; 4 – heat shield; 5 – vessel; 6 – drive AZ-KO; 7 – box of exhaust ventilation; 8 – control rod AZ-KO; absorber plate in position: 9 – “up KO”; 10 – “up AZ-down KO”; 11 – “down AZ”

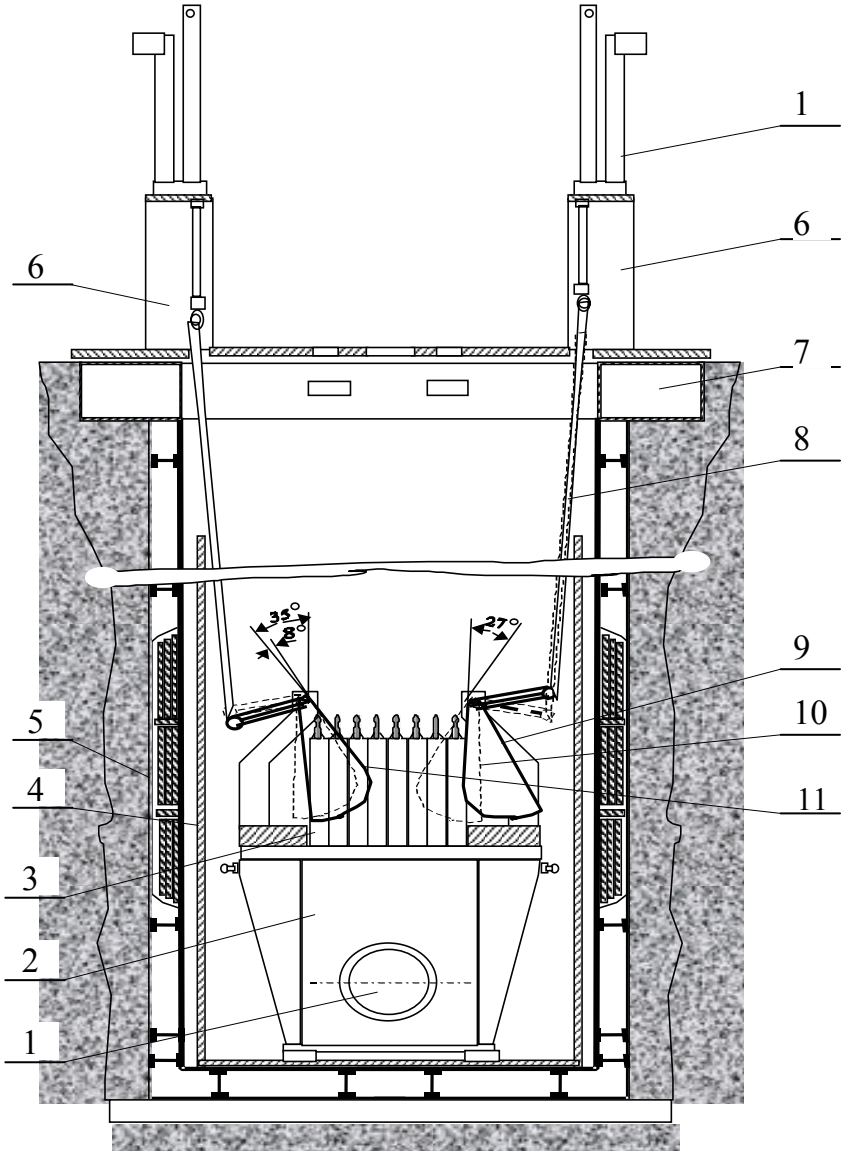
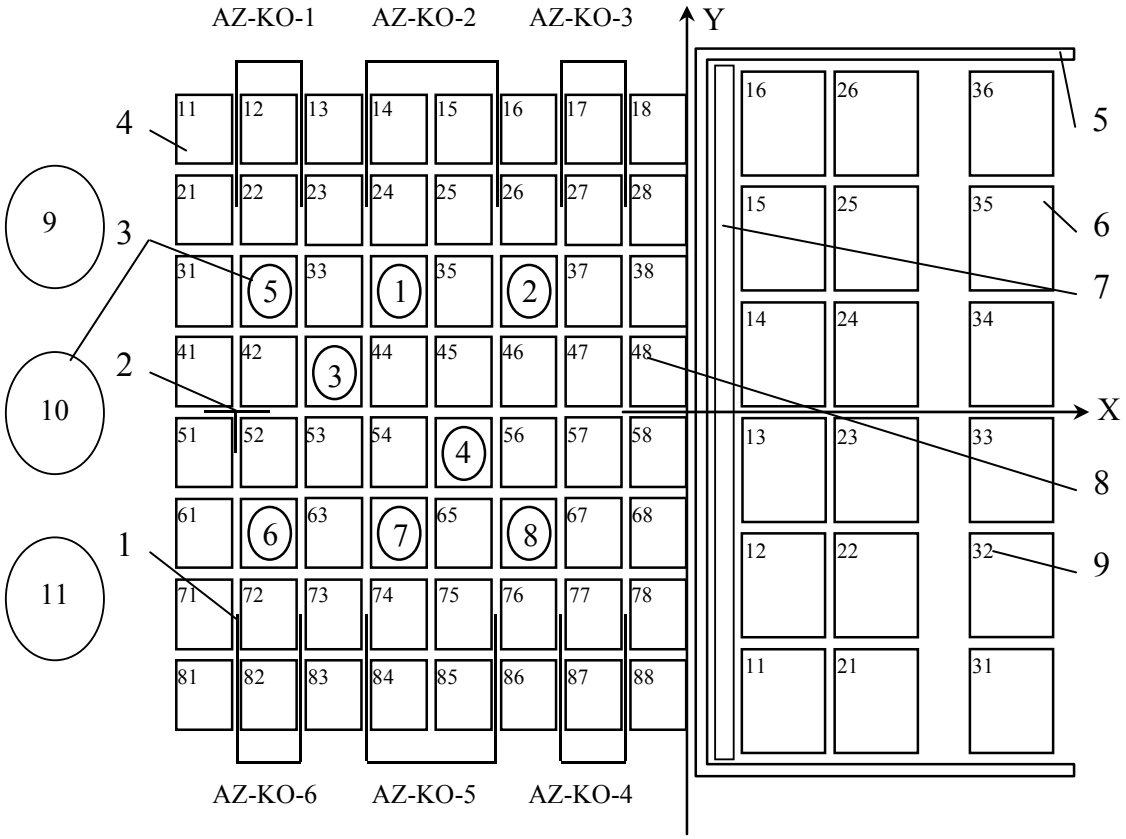


Fig.6. Cross-section of active zone of reactor RBT-6 and stand "KORPUS"

1 – movable operating element of emergency protection and reactivity compensation (AZ-KO);
2 – movable operating element of automatic controller; 3 – irradiation channel; 4 – FA;
5 – core buffle of stand; 6 – ampoule; 7 – lead shielding; 8 – number of active zone cell;
9 – number of stand cell; X, Y – co-ordinate axes (axe Z is perpendicular to surface of figure and up-directed).



Brief description of experimental methods of reactivity effects determination and method of the experiment carrying out

Methods of backward multiplication, asymptotic period, overcompensation with known reactivity and methods based on reversed solution of kinetics equation performed at digital and analogue reactimeters are used during reactivity effects determination. Methods of determination of SCS elements efficiency, reactivity effects, reactivity margin and reactor subcrit are certified according to national standards with error norms assignment. Every method includes description of: field of its application, using equipment characteristics, practical algorithms of using method implementation, sequence of operations when performing measurements, data handling and presentation requirements. The reactor is equipped with the analogue reactimeter of SCS "KARPATY" apparatus industrially produced (range of values of calculating reactivity is from –20 □eff up to +0.5 □eff at six measurement subranges; limits of main reactivity calculation error reduced to subrange value is not more than □ 6%) and with the digital multi-channel reactimeter of RIAR's fabrication. Computer using in the digital reactimeter for solution of reversed reactor kinetics equation depending on alternating current of neutron detector allows increasing accuracy and immediacy of results obtaining, carrying out various methods of statistical handling of big recorded data arrays, immediate presenting reactivity calculation results in digital, analogue and graphical form. In order to increase precision,

reliability, immediacy and representability of reactivity determination the digital multi-channel measuring system - hardware/software system “Reactimeter”, which is used for carrying out of experiments at RIAR’s research reactors, was developed. The system includes neutron flow detectors, object coupling device, personal computer, special software.

To increase reliability and confidence of obtained results during performance of experiments at the reactor, several methods are applied as rule. Usually combination of simple but reliable method and more precise one with measuring signal handling equipment is used. For example, efficiency of withdrawing part of compensating controller is determined by asymptotic period method and according to reactimeter indications. All applying methods of reactivity determination in measurement error limits show the same results under operable condition. Variety of applying methods and independence of measurement means increase confidence of obtained results.

During carrying out of experiment on determination of influence of VVER experimental FA with non-irradiated and irradiated fuel rods on reactor RBT-6 reactivity the following methodic sequence of operations is performed:

1. Selection of reactor loading schematic arrangement according to the results of preliminarily performed calculations.

2. Reloading of the reactor according to the selected schematic arrangement of active zone loading. Running the reactor to the minimally controlling level (MCL) of power without experimental FA in the channel, fixing one or several positions of SCS elements in critical condition, which are the most convenient for further calculation modelling. (For example, all the controlling elements except one are completely withdrawn or completely dipped). Determination of controllers efficiency, reactivity margin, active zone subcrit with dipped compensating controllers under this reactor condition. The reactor shutdown.

3. Loading experimental FA with fresh fuel rods in the experimental channel. Running the reactor to MCL of power, fixing one or several positions of SCS elements in critical condition, which are the most convenient for further calculation modelling with reactivity loading compensation when loading experimental FA by moving one or two controllers with respect to the critical conditions under Paragraph 2. Determination of efficiency of controllers parts which are required for reactivity compensation when loading with experimental FA. Efficiency of these parts corresponds to the reactivity effect from experimental FA loading. Determination of controllers efficiency, reactivity margin, active zone subcrit with dipped compensating controllers. The reactor shutdown.

4. Repeat of the procedures under Paragraph 3 when successively loading experimental FAs with irradiated fuel of various burn-up into the reactor channel.

5. Calculation of the obtained reactor conditions for verification of calculation methods and mathematical models. For preset adjustment of calculation program models, not using at RIAR, intended to take into account specialties of SM type fuel, it is possible to run the special verification experiments on the critical assembly – physical model of rector SM, in which non-irradiated fuel of SM type is used.

Brief description of calculation methods and preliminary results of calculation estimates

Calculations of values of efficiency rate of neutron multiplication when evaluating reactivity effect of experimental FA loading into channel of reactor RBT-6 are performed by means of numerical model developed on the basis of application package MCU (version MCU-RFFI/A [4]). Analogue method Monte Carlo of modelling of neutrons paths in three-dimensional geometry is realised in the program. Constant provision of the package is based on nuclear data library DLC/MCUDAT-1.0. The library includes several chapters, each of which contains cross-sections and other constants of interaction of neutrons and gamma-quanta in specified field of energy with nuclides being part of reactor material composition.

Cross-sections of neutrons interaction over the range of energies 1 eV-10.5 MeV are shown in 26-group format of constant system BNAB [6]. Data on resonance characteristics of cross-sections is presented as subgroup parameters.

Cross-sections in the heat field of neutron energies 0-1 eV are shown in 40-group partition with an even speed step (library TEPKON). Differential cross-sections of scattering are calculated under specified temperatures taking into account chemical links and crystal structure of materials.

The reactor active zone is modelled in three-dimensional geometry with the parameters as follows:

- all AZ-KO are equally dipped in active zone to the depth corresponding to a statistically average position;
- core AR is in midposition;
- aluminium displacers with aluminium plugs are placed into all the cells intended as experimental channels except cell 55.

Cell 55 is free under one variant of calculation, and it is occupied with experimental FA, cross-section of that is shown in Fig. 4, under another variant of calculation.

Difference of calculated systems reactivities (i.e. reactivity effect of experimental FA placement into cell 55) amounted to about 1.05% □k/k.

Conclusion

The investigations provided in the programme include performance of radiochemical analysis and reactivity measurements on the reactor facility. Their implementation will achieve the results for validation of calculations of burn-up credit of spent fuel at two enrichments (3.6% and 4.4%) using at VVER-440 reactors.

In case of limitation of funds for the proposing investigations, it is possible to limit ourselves to the investigation of 4 FAs (two model FAs are investigated: one – with non-irradiated fuel, another – with irradiated fuel for each fuel at enrichment 3.6% and 4.4%). In this case good results can be obtained as well.

If the second variant is also not funded, we can investigate two model FAs with fuel at one enrichment. But this variant will not provide completeness of data on burn-up credit.

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DRAFT PROPOSAL

DRAFT OF PROPOSAL ON VVER-440 PIE SUPPORTING BURN-UP CREDIT METHODOLOGY

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Abstract

This paper describes a draft project proposal to perform new radiochemical assays of 12 VVER-440 spent fuel samples in Dimitrovgrad, Russia. The need for new VVER PIE measurements was indicated due to a total lack of the well-documented VVER PIE data, particularly isotopic composition data, accessible in the open literature that is considered adequate for validation of VVER fuel depletion calculations as used in burn-up credit analyses. The draft proposal discussed in this paper has a similar structure as the ISTC #2670 project recently performed in Dimitrovgrad under the US support (completed in 2005). In the framework of the ISTC #2670 project, well-documented PIE data for 'BUC nuclides' (actinides and major fission products) from 6 VVER-440 spent fuel samples were acquired and released without any restrictions. These data are available to the entire criticality safety community. The main goal of the proposed VVER PIE project is to continue VVER spent fuel examinations in order to enlarge the measured data set applicability range and increase the statistical data sample for evaluation and use for depletion code validation. As requested by regulatory bodies, validation of the depletion codes is now the important condition for acceptance and licensing the future implementation of burn-up credit for VVER spent fuel management systems.

Introduction

The project was initiated in 2004 by a representative group of criticality safety specialists from nuclear research, design, manufacturing and nuclear safety surveillance institutions. These experts represent the European countries operating VVER-440 outside of Russia. As a result of a number of discussions the group intends establishing a consortium and negotiates the project with the State Scientific Center "Research Institute of Atomic Reactors" (RIAR, Dimitrovgrad, Russia) for their experience and capability in performing the radiochemical analyses needed to produce the required data. Discussions on the project preparation has been largely conducted in parallel with the activities of the OECD/NEA/NSC Working Party on Nuclear Criticality Safety (WPNCS) Expert Group on Burn-up Credit (EGBUC) and the Atomic Energy Research (AER) Group E, research activity of the Eastern and Central European Countries operating VVERs, which is focused on "Physical Problems on Spent Fuel, Radwaste and Decommissioning of Nuclear Power Plants".

Scope of the project

Background

In 2003-5, under the ISTC #2670 project [1] supported by the USA, eight VVER-440 UO2 fuel samples were subjected to radiochemical analyses in the RIAR Dimitrovgrad laboratory. It was the first VVER Post Irradiation Examinations (PIEs) where the isotopics of actinides and major fission products were measured for VVER spent fuel to support the burn-up credit methodology. The objective of these data was for use in the validation of the depletion codes in the VVER environment. The ISTC #2670 project data were released by US NRC as not restricted and are available for use by all analysts. Before these important data were available, the analysts' only option was to establish simple calculational exercises and perform code intercomparison benchmark exercises under the AER Group E. The CB2 [2] benchmark performed by this group addresses the issue of isotopic benchmarks but was not based on comparison with any PIE data since none were available at the time. This calculational approach to validation is generally not considered sufficient for criticality safety applications and the data comparison against measurement is urgently needed.

The ISTC #2670 data provide some information that can be used for validation. However, there are only eight measured samples of limited enrichment and burn-up ranges and is still not sufficient to solve the need of the VVER PIE data supporting the VVER spent fuel application area for broad use. The proposal discussed in this paper is aimed at acquiring more data points for each isotope and to increase the range of enrichment and burn-up conditions for which the validation is applicable. Additional PIE measurements, having statistically significant amounts of data and adequately addressing the range of fuel conditions for VVERs should be covered by additional VVER-specific measurements. Some of these data may also be useful for PWRs as it is also acknowledged that there are not a sufficient number of data points available for all BUC nuclides, using freely available PWR data. To address the issue, a review of all options available was performed and discussions within the analyst community resulted in the conclusion that clearly additional VVER PIE measurements for isotopic composition data are needed.

Facilities that have the capability to perform such analyses of spent fuel are very limited. The only known country with this capability and with accessibility and experience with VVER fuel is the Central all-Russia hot laboratory for examinations of the VVER-440 situated in RIAR, Dimitrovgrad. On this basis, the inquiry for the new VVER-440 PIE was sent to the RIAR laboratory early in 2005. After certain clarifications and requests/exchanges of information, the RIAR identified several VVER-440 fuel assemblies that could be made available for new PIE measurements.

The scope of the proposed project is similar to the ISTC #2670 project performed in 2003-2005 in RIAR [1] and is based on information provided by RIAR regarding experimental possibilities [3]. The specific needs of the VVER community for the BUC applications related to the validation of depletion calculation methodologies used for predicting specific isotopes needed for implementing BUC at the detailed 'actinides + major fission products' level have also been integrated into the proposed project scope. In the discussions below, the "contractor" refers to the consortium of non-Russian VVER-operating countries seeking to use the data for BUC implementation.

Project objective

The objective is to describe a specific VVER- 440 reactor spent fuel initial state before irradiation and its operational (irradiation) history and examine the resulting nuclide inventory and burn-up distributions in the fuel after the irradiation.

Work includes the following stages:

- The selection of a specific spent (irradiated) fuel assembly (FA) for examination taking into consideration the results of the "quick look" FA examination and contractor's special requests related to the selection.
- The description of the FA initial parameters.
- The description of the irradiation history enabling that the full set of input data for the criticality/depletion codes can be derived.
- Predictive thermal calculations.
- The non-destructive examination of the gross physical state of the FA fuel pins (first assessment of clad integrity based on photos/gamma scan, fuel pin dimensional inspection, gamma scan measurements of radionuclides/burn-up distribution along the height and FA cross-section, fission gas sampling).
- The selection of fuel pins of the selected FA and preparation of samples from the selected fuel pins for the destructive examinations.
- The destructive examination for the determination of the radionuclide compositions, isotope masses and burn-up in the fuel pin samples.
- Making analyses, uncertainty determination, documentation and processing the results.

According to the contractors' request the assays of spent fuel, which will be performed in the hot laboratory of RIAR, Dimitrovgrad, Russia, will use fuel samples of No. ..... fuel assembly of av. ..... MWd/kg_U burn-up irradiated in the ..... NPP unit ..... given the FA examination and preliminary PIE indicate good condition of the fuel pins and fuel behaviour is as expected relative to the FA operational history.

Based on agreed criteria and on the FA irradiation history and preliminary non-destructive measurements, the agreed number of pins of the fuel assembly will be selected and removed. Further, the agreed number of samples will be cut out of the pins for the destructive radiochemical analyses to be carried out. The results will be documented as a series of reports published within a period of about one and half years. The language of the reports is agreed to be in English. The measurement supporting calculations will be well-documented for detailed understanding and possible independent repetition, the reference documents will be available at request, electronic files for the submitting the reports and data are highly preferred. The other references in the reports will be either publicly or at request available or the subjects of the references will be described comprehensively enough in the report.

If any analyses of any spent fuel samples taken of the FA selected for this PIE have been performed earlier the results and exact times of the measurements will be mentioned in appendix of the final report for comparison if this is technically possible.

The project deliverable reports will be organised into 7 submittals relating to the individual phases of the project task fulfilment. These occurred in the logical sequence of performing the work over an approximately 1 1/2 year period. These reports will be contained in the final document as prepared by the Russian investigators with minimal editing.

Project phases and tasks

Seven phases of the project task fulfilment are:

Phase I.

Task No. 1. Selection methodology of fuel assemblies, fuel pins and fuel samples for measurements of isotopic composition, isotope masses and fuel burn-up.

Deliverable1: Interim Progress Report 1

The report will describe the selection methodology of VVER-440 fuel assemblies, fuel rods and fuel samples to perform their radiochemical analysis and mass spectrometry in order to obtain data on nuclide composition and fuel burn-up.

Selecting the proper FA for the PIE, the facts on VVER-440 FA availability in RIAR/Mayak for measurements, suitability for the the general validation purpose (working assembly rather than control FA (ARK), burned in positions not adjacent to the control FA, FA which is as new as possible,..), if FA design was/is in use in NPPs operated by the contractors and similar aspects must be assessed. This stage of the selection needs a collaboration of the contractors and the laboratory. After this non destructive examination of the FA technical shape as level of oxidation, elongation and shrinkage of cladding tubes, presence/absence of fuel-to-cladding gap, (non)existence of corrugation-traces due to fuel-cladding interaction and fuel pin tightness can be made and assessed by the laboratory. The assessment will be based on the results of the FA shape examination conducted preliminary PIE (the “quick look” documented by photos and accompanied by supporting thermal calculations for findings comparison). In addition of this, quality and comprehensiveness of the documentation on the operational history and (recorded/pre-calculated) cycle average burn-ups (to recognise the level of the FA exploitation: standard/extremely) play an essential role for the final FA selection.

Table 1 FAs currently available for radiochemical analyses in RIAR

FA no.	NPP unit	Operation [eff. days]	Av. burn-up [MWd/kg_U]	[wt. %] U-235 enrichment	Comment
D 19159	HBNPP-4	989.1	34.1	3.6	WA, has been measured in 1991-92
D 42198	KolNPP-2	1242.3	44.7	3.6	WA
E 46879	KolNPP-3	1872.0	56.6	4.4	WA

As for the selection of fuel pins from the selected FA, they must be proposed to meet the contractors' validation goals. It means that the measured data must be comparable with the data

resulted from depletion calculations simulating the documented FA irradiation history. The selection will be based on gamma spectrometry measurements aimed to finding the fuel pins of maximum/minimum/intermediate average (along z-axis) burn-ups and positions at which **12 samples** will be cut out for the quantitative analysis of the fuel isotopic composition.

Phase 2.

Task No. 2. Design of the selected VVER-440 working assembly.

Deliverable2: Interim Progress Report 2

The report will describe the design details of the selected VVER-440 working assembly. Geometry, nuclide and chemical composition of the fuel pellets as well as parameters of fuel pins and fuel assembly as a whole will be given there.

After the FA selection, all data related to the design characteristics will be checked and tabulated so the input data entering validation calculations (depletion and criticality) can be specified. The data related to the FA as fuel fabrication data, design type, design characteristics (geometry, initial enrichment and initial isotopic inventory of the fuel impurities, the geometry features and materials of the components as shroud, spacer grids etc.) before the FA first irradiation and cooling will be described in details. All the alloys and standard compositions will not be only named but also described in details (density, composition in wt. % of the individual nuclides), masses related to the initial state of fuel and FA spacer grids will be mentioned as well as uncertainties.

Phase 3.

Task No. 3. Irradiation history of the selected VVER-440 working assembly.

Deliverable 3: Interim Progress Report 3

In the report there will be described in necessary details the history of the core loadings before the selected FA was inserted in the core and operational history core loadings where the selected FA was working.

Similarly to the Phase 2 giving the full description of the FA and their fuel pins as for the geometry and material data, further data must document the FA irradiation history, which includes the set of the core load patterns mapping FA reshuffling in the core, FA orientation in the core in each cycle, number of the irradiation cycles, FA up- and down- times incl. unplanned shutdowns, specific power related values, temperatures, average boron cycle data as well as positions of operating control FA bank etc.

The history description will be exhaustive enough to enable the irradiation to be simulated by a depletion code calculation taking a case dependent flux into consideration in the course of the irradiation. Thus, the predicted fuel isotopics can be compared with the results of the radiochemical measurements as a piece of validation of the methodology used for the calculation.

To check the completeness of the data for such a calculation and provide some intermediate results supporting the measured data evaluation, the detailed calculation of reference made by Russian codes capable to describe the VVER-440 operation (e.g. BIPR, PERMAK, TVS-M) will be performed and documented.

Phase 4.

Task No. 4. Non-destructive post-irradiation examinations of the fuel pins of the selected VVER-440 working assembly.

Deliverable 4: Interim Progress Report 4

The results of non-destructive post irradiation examinations of the fuel pins of the selected VVER-440 fuel assembly will be described. Resulting from the fuel pin condition assessment and according to agreed criteria the agreed number of the fuel pins will be chosen for follow-up destructive examinations. Keeping to the same criteria, the positions for cutting samples for quantitative analysis (destructive examination) of fuel isotopic composition will be chosen as the next step. To predict fuel behaviour during irradiation, thermal/mechanical calculations will be performed, the results of them will be described as well as their comparison with the findings of the fuel condition (pellet-to-clad gap, pellet dishing, fuel pin outer diameter, gas pressure within the fuel pins, temperatures, etc.).

The following non-destructive examinations of the fuel pins will be performed:

- Visual inspection using the viewing windows of the hot cell and in-cell periscope.
- Measurements of length and outer diameter of the fuel pins.
- Eddy current testing for diagnostics of fuel pin clad conditions.
- Gamma spectrometry for determination of the axial peaking factor of ^{137}Cs long-lived fission product accumulation as well as maximum and average fuel burn-up values in fuel rods.
- Analysis of gas release from fuel.
- Measurement of pellet-to-clad gaps.
- X-ray radiography.
- Temperature measurement.

A correlation analysis of fuel pin parameters should prove the proper selection of fuel pins supposed for the analyses of the isotopic composition based on the selection methodology agreed for the Phase 1.

Phase 5.

Task No. 5. Selection and preparation of samples for the radiochemical analyses of VVER-440 fuel isotopic composition.

Deliverable 5: Interim Progress Report 5

The report will describe the fuel sampling technology and cutting procedure as well as will give the characteristics of the fuel samples.

The agreed fuel pins from the agreed positions in the FA will be removed from the dismantled FA and cut in accordance with the requirements described in the earlier Phases. **12 fuel samples** will be cut out and placed into the marked containers for the follow-up radiochemical analyses. After sampling all the fuel samples will undergo dissolution process and the radiochemical analyses.

Phase 6.

Task No. 6. Determination of the nuclide compositions, isotopic masses and fuel burn-ups related to the selected VVER-440 spent fuel samples; description of the techniques and analytical methods applied as well as equipments used.

Deliverable 6: Interim Progress Report 6

Results of nuclide composition examination and fuel burn-up determination for twelve samples of the selected VVER-440 fuel pins will be presented. Analyses will be focused on finding abundances of the following nuclides:

U-234, U-235, U-236, U-238, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Am-241, Am-243, Np-237, Mo-95, Tc-99, Ru-101, Rh-103, Ag-109, Cs-133, Sm-147, Sm-149, Sm-150, Sm-151, Sm-152, Nd-143, Nd-145, Eu-153, Gd-155.

As subsidiary but important from the shielding and heat points of view, contents of Cm-242, Cm-244, Cs-134, and Cs-137 will be evaluated. Further, in agreed or needed cases, parent and daughter nuclides will be measured to provide additional data for establishing measurement quality.

Fuel burn-ups will be calculated using the methods of heavy atoms and the fission product accumulation. The main techniques and analytical methods applied as well as equipments used will be shortly described.

The examination will include the following stages: the dissolution of the selected samples, radiometric measurements, the radiochemical extraction of the specified elements from the fuel solution and determination of isotopic compositions of the extracted elements using mass-spectrometer analysis, the measurement of mass fractions using isotopic dilution followed by mass-spectrometer analysis, data processing and determination of fuel burn-up. The report will present the analytical results of the isotopic concentrations for the nuclides which are important for the BUC criticality analyses according to the BUC implementation methodology:

U-234, U-235, U-236, U-238, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Am-241, Am-243, Np-237, Mo-95, Tc-99, Ru-101, Rh-103, Ag-109, Cs-133, Sm-147, Sm-150, Sm-151, Sm-152, Nd-143, Nd-145, Eu-153, Gd-155.

Method of isotope dilution with mass-spectrum ending of analysis will be used as the main method of determination of the nuclide content. Error of the method doesn't exceed 1.5-3.0% and is mainly determined by error of standard samples using as tracers. When determining content of U, Pu, Am, Nd, Ce the standard solutions of these elements prepared in Radium Institute (Saint Petersburg) are used as labels When determining content of Gd, Eu, Sm, Cs, Mo, Ru, Ag, Pd the standard solutions of these elements prepared from samples of natural isotope composition are used as labels. To determine Tc-99 content the standard solution prepared from Tc sample with Tc-98 content not less than 12% is used. Such a Tc sample is made according to (n,2n) reaction when irradiating Tc-99 in nuclear reactor. In spite of the good analytical possibilities of the method the Ru-101 data will have to be evaluated as dissolving fuel sample some Ru can be lost due to high volatility of this element. Because of complexity of the chemical composition and therefore difficulties of chemical separation of elements maybe the obtained Mo-95, Tc-99, Ag-109, Pd-105, -108 mass values will have to be considered as evaluating as well. It is

impossible to determine Rh-103 by this method since its share in nature is 100%. Therefore spectral method will be applied to determine content of these elements at the same time.

Burn-up of fuel will be determined by two methods:

- fission products accumulation method as the main method with error of 3-4%. The Nd-145+146 and Nd-148 are used as burn-up monitors
- heavy atom method, error of the method is 5-7%.

Isotopic concentrations will not be adjusted to discharge but will be reported to the date at which each measurement was made. The isotopics will be given in agreed units, preferably in [(b-cm)$^{-1}$] and burn-up in MWd/kgU; The uncertainties as well as date of the measurements will be mentioned.

Phase 7.

Task No. 7. Chemical assays of irradiated VVER fuel – summary.

Deliverable 7: Final Report

The Final Report will summarise the whole technical work made for the project tasks fulfillment. All the needed explanations related to the terminology, units and constants used will be mentioned there as well as the reference list.

The data resulting from the PIE measurements will be used for the depletion/operational code validation for criticality safety in spent fuel operations. Identifying the initial data of fuel as well as the operation history as taken from the reactor records and sample identification all need great detail and attention in the project documentation in order to assure correctness and the usefulness of the final data.

Cost of project

The project proposal described above is the 'Basic PIE' and includes the basic requested examinations for depletion code validation purposes. As a possible option, an additional Phase *) could be included to focus on more detailed examinations of the fuel pins/pellets for the purpose of collecting general cladding and fuel performance and clad testing data. Incorporating this additional Phase is referred to as the 'Extended PIE' option. Those two alternatives, the 'Basic PIE' and the 'Extended PIE' will differ also in the cost and term:

A 'Basic PIE' cost preliminary estimation is to be on the order of USD 250 000 which includes radiochemical examinations of 12 spent fuel samples, non-destructive examinations, selection of fuel pins, cutting out samples, initial fuel data and irradiation history data documentation as well as supporting calculations.

An 'Extended PIE' cost preliminary estimation is to be on the order of USD 340 000 which includes radiochemical examinations of 12 spent fuel samples, non-destructive examinations, selection of fuel pins, cutting out samples, material testing examinations, initial fuel data and irradiation history data documentation as well as supporting calculations.

Term

The basic PIE will be performed and documented in the course of 1½ years, the extended alternative will last 2 years.

***) Additional (optional) phase**

Phase

Task: Detailed examination of the fuel pins/pellets for the purpose of collecting general cladding and fuel performance data.

Deliverable: Interim Progress Report

Material testing and further more detailed analyses related to quantity and composition of gas under fuel cladding, quantity and composition of gas solute in solid phase of fuel core, fuel-cladding gap, structure and dimensions of fuel seed, thickness of the oxide layer on the outer surface of the selected fuel pins, the elongation of the fuel pins as well as vortex-current defectoscopy of fuel cladding will be described and results presented.

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[2] *L. Markova*: Calculational Burn-up Credit Benchmark No.2 (CB2) (7th AER Symposium on VVER Reactor Physics and Reactor Safety, Hornitz near Zittau, Germany, Sept.23-26, 1997; NRI 10979 R).

[3] A. Chetverikov: RIAR offer of the new VVER PIE, March 2005, additional items to extend the offer: Oct. 2005, personal communication.

Appendices

NEED FOR EXPERIMENTAL DATA ABOUT NUCLIDE INVENTORY AND REACTIVITY OF IRRADIATED NUCLEAR FUEL AS SEEN BY THE GERMAN AUTHORITY RESPONSIBLE FOR THE TRANSPORT AND DRY STORAGE OF SPENT FUEL

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Abstract

The German authority responsible for the transport and dry storage of spent nuclear fuel (BfS) can only access the publicly available PIE data. In the case of package design approvals including credit for high burn-up and/or the inclusion of fission products the applicant must provide all data necessary for the validation of the depletion and multiplication factor calculation software, followed by an independent review by BfS. This procedure could be much accelerated if the international community could actively participate. Therefore BfS would really appreciate help by OECD/NEA in enlarging the number of publicly available PIE data.

The application of burn-up credit in the criticality safety assessment of transport and storage of spent fuel is getting more and more important with increasing initial enrichment of the fuel. In Germany, the VVER fuel elements remaining from shut-down nuclear power stations can be stored and transported without burn-up credit due to their low enrichment. However, fuel burn-up credit has to be considered for PWR and BWR fuel elements.

There is a German standard for the application of burn-up credit. One main idea of this standard is that any method for the depletion and criticality analysis can be used provided that this method has been validated against experimental data.

The German authority responsible for the transport and dry storage of spent fuel BfS (Federal Office for Radiation Protection) can only access the experimental data that are publicly available, especially the data in the SFCOMPO database at OECD/NEA. The German competent authority does not carry out PIE measurements and is not involved in any international commercial PIE project.

This corresponds to the status quo of package design approvals with burn-up credit in Germany: So far the use of burn-up credit in German package design approvals has been limited to max. 10 GWd/tU and restricted to accounting for actinides only. For this application the publicly available information on PIE data and calculational research, e.g. by ORNL, is adequate.

Future applications for package design approvals including credit for higher burn-up and/or the inclusion of fission products will require additional assessment procedures to be carried out by BfS:

- review of the validation report for the depletion calculation method that was supplied by the applicant, based on PIE data provided by the applicant,
- validation of the depletion software used by BfS using PIE data to be provided by the applicant,
- review of the validation report for the applicants method for calculating k_{eff}, supplied by the applicant and based on experimental reactivity data provided by the applicant,
- validation of the software used by BfS for calculating k_{eff} using experimental reactivity data to be provided by the applicant

This extensive procedure will cause significant delay in package design approvals. The procedure could be much accelerated if the international community could actively participate. However, this would require more publicly available PIE and reactivity data. BfS would really appreciate help by OECD/NEA in enlarging the number of publicly available PIE data.

ON ESTABLISHING THE CONSORTIUM TO FUND THE NEW VVER PIE PROJECT

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Abstract

In this article is overview through participated or interested institutes for new chemical assay of VVER-440 spent fuel.

Participated or interested institutes

The Soviet type of reactor VVER-440 is/was in operation in Eastern and Central Europe and in Finland. Countries with this type of reactor don't have hot laboratories for chemical analyses of spent fuel, except Germany and Russia. In Germany were in operation 4 Units VVER-440, now are closed and are decommissioned. Germany colleagues think that measurement is possible, but the operation data about irradiation history are not complete to prepare "well documented" description. Transport of spent fuel through boundary is practically impossible (too expensive and too bureaucracy). There is only one possibility – spent fuel from Russian Unit and Russian hot laboratory.

Institutes from different countries have interest to participate in international project for chemical assay of VVER-440 fuel:

Czech Republic	NRI Rez (Lida Markova) SKODA JS (Pavel Mikolas)
Hungary	KFKI Budapest (Gabor Hordosy) NPP Paks (Imre Nemes)
Finland	VTT (Markku Anttila) FORTUM (Pertti Siltanen) Reactor and Safety Systems Radiation and Nuclear Safety Authority (Riku Mattila)
Slovakia	VUJE (Vladimir Chrapciak) GovCo (Vojtech Adamovský) Slovak electric company (Jozef Lukacovic) Nuclear Regulatory Authority (Juraj Vaclav)
Bulgaria	INRNE (Maria Manolova) NPP Kozloduy (Iskra Stoianova)
Ukraine	SSTC (Yuriy Kovbasenko)
Russia	Kurchatov Institute (Lev K. Shishkov)
Germany	DG Joint Research Centre, Institute for Transuranium Elements (Paul VanUffelen)
Armenia	Nuclear Regulatory Authority (Ruben Aydinyan)
United Kingdom	BNFL – Nexia Solutions (Jim Gulliford)
Int. org.	IAEA (William Danker) OECD/NEA (Yolanda Rugama)

Hot laboratory

Russia	RIAR Dimitrovgrad (Anatoly Chetverikov)