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INTERNATIONAL WORKING GROUP
ON WATER REACTOR FUEL PERFORMANCE AND TECHNOLOGY

BURNUP DETERMINATION OF WATER REACTOR FUEL

PROCEEDINGS OF A TECHNICAL COMMITTEE MEETING
ORGANIZED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
AND HELD IN KARLSRUHE, 13–16 JUNE 1988

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FOREWORD

The present meeting was scheduled by the International Atomic Energy Agency in consultation with the Members of the International Working Group on Water Reactor Fuel Performance and Technology.

The meeting was hosted by the Commission of the European Communities, at the Transuranium Research Laboratory, Joint Research Centre Karlsruhe, in the Federal Republic of Germany.

This subject was dealt with for the first time by the IAEA. It was found to correspond adequately to this type of Specialist Meeting and to be suitable in a moment when the extension of burnup constitutes a major technical and economical issue in fuel technology.

Twenty-seven participants from eleven countries plus two international organizations attended the Meeting. Twelve papers were given during three technical sessions, followed by a panel discussion which allowed to formulate the conclusions of the meeting and recommendations to the Agency. In addition, participants were invited to give an outline of their national programmes, related to Burnup Determination of Water Reactor Fuel.

At the occasion of this meeting, it was stressed that analysis of highly burnt fuels, mixed oxides and burnable absorber bearing fuels required extension of the experimental data base, to comply with the increasing demand for an improved fuel management, including better qualification of reactor physics codes.

The local organizing committee deserves special acknowledgement for the excellent meeting arrangement and hospitality in spite of a short notice. The session chairmen and all the contributors are also responsible for the success of this meeting.

EDITORIAL NOTE

In preparing this material for the press, staff of the International Atomic Energy Agency have mounted and paginated the original manuscripts as submitted by the authors and given some attention to the presentation.

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GENERAL CONSIDERATION ON BURNUP DETERMINATION

J. FUGER
Commission of the European Communities

Mr. Sametband (Argentina)

2 reactors are in operation totalling 900 MW, a third one of 700 MW is being built. All these are heavy water reactors, natural uranium, zircalloy clad. All analyses carried out sofar are non-destructive and done in reactor pools. They involve visual examination and metrological determinations. In one of the reactors γ-spectrometry has been carried out on the 5.2 m long (active length) fuel. The latter measurements are useful for good axial distribution but not for burn up. Non-destructive burn up measurements have been carried out on the Al-UO_2 (90%) fuel at the spent fuel pond of the research reactor at Bariloche.

A proposal to IAEA has been presented for carrying out determinations related to safeguards in the spent fuel pond of the power plants. Adequate measurements without movement of the fuel is planned using CdTe detectors for γ-activity, and solid state detectors for passive neutron counting. This work will be done in cooperation with the Stefan Institute Ljubljana, Yougoslavia, and the Institute of Isotopes, Budapest, Hungary.

P. DeRegge (Belgium)

The CEN-SCK, Mol (B) has been involved for more than 20 years in prototype fuels irradiated in the BR III reactor (PWR Westinghouse) at burn up ranging between < 5000 MWd/t and > 75000 MWd/t and using natural urani ., enriched uranium, MOX and, more recently, fuels with burnable poisons. Stu es involved effects of thermal cycling, of power transients and very long irradiation histories. Deep involvement exists also in the study of the properties of new fuel concepts though irradiations in the BR II material testing reactor. The concepts tested included BW and PW capsules, Na and NaK cooled loops for MOX and carbide fast breeder fuels, gas cooled breeder fuels, high temperature coated particle fuels, and also BR II reactor fuel itself. This leads to an impressive data bank on burn up and post irradiation studies. Alsc were carried out determinations of proprietary nature in various reactors, on request of commercial nuclear companies.

Developments in burn up determinations and analyses have been oriented for more than 15 years towards mixed oxide fuels as the recycling of Pu in thermal reactors has been long advocated by Belgonucléaire which also carried out design of fast breeder fuels. Recent developments involve detailed fuel examination, axial burn up, radial burn up (burnable poisons by radial drilling). γ-scanning has been used for long as measurements of axial F.P. and burn up. An important activity has been the accumulation of selected pieces of fuel with particularly well known history and well characterized by full analysis of adjacent pieces. These selected pieces (Burnothèque) are used to calibrate non-destructive measurements on LWR fuels. Due to migration effects and contributions from the blankets, FBR fuels need a calibration with a complete rod from a well analyzed assembly.

Work was also carried out on the analysis of passive neutron measurements carried out on PWR spent fuel with the ION-1 fork detector developed by the IAEA, JRC and LOS ALAMOS laboratories. Using a point model depletion code,

in order to take account for the irradiation history, a fairly good correlation is obtained between the improved neutron measurements and the burn ups from the fuel management codes, if only one initial enrichment is regarded. Some more work remains still to be carried out to take the initial enrichment into account.

Mr. Alonso (Spain)

In Spain there are eight LWR in operation. Fuel for nearly all these reactors is supplied by ENRESA that also provides the core engineering for the fuel management of the plants. Benchmarked commercial codes are used for burn up determination.

The current national strategy for fuel disposal is direct storage instead of reprocessing and it is being managed by ENRESA.

Both ENUSA and ENRESA are now in a joint program to characterize the spent fuel coming from the Spanish plants. Real fuel burnup values of spent fuel are an important part of this characterization so burnup determination techniques are becoming very necessary at the present moment.

Mr. Liao (People's Republic of China)

For the past 10 years the activities in China have involved both destructive and non-destructive analysis.

Use has been made of isotopic dilution mass spectrometry and γ-spectrometry. In destructive methods U and Pu were separated using ion exchange, U being determined by IDMS and Pu by coulometry.

Mr. Simon, Mr. Wiese (Federal Republic of Germany)

The program on non-destructive assay is in continuation using neutron measurement and γ-spectrometry. For neutron measuring the work at KfK, meanwhile, concentrates on implementation of improved technical designs into prototypical devices fulfilling stringent accuracy limits and being operational on routine basis under actual plant conditions.

Active and passive neutron detection is carried out in storage ponds; U and MOX fuels are involved. The method applied can be used in reprocessing plants for the determination of burn up, for quality control, for Pu input balance estimation.

Combination of active, passive neutron and cross gamma counting fulfills all requirements for fuel assemblies in water. Without any indication on fuel a positive identification is possible, as well as consistency of shipment data. It was also demonstrated to what extent the measurements are influenced by specific reactor parameters.

Simplification of the spent fuel monitor are developed, to facilitate the application.

The theoretical investigations related to burn up need effective nuclear cross sections, fission product yields, decay constants etc. One of the main efforts at KfK in this field is the reliable determination of effective cross sections by using well established basic data from the KfK-file KEDAK and, recently, from the Joint Evaluated File, JEF, and proven codes as WIMS for spectral calculations. Further work was devoted to include modern yield data. These data are used for

depletion calculations with the code system KARBUS or the stand-alone program KORIGEN. Both codes were checked intensively against experiments. Recent investigations aimed at the determination of high-burn up (> 40 GWd/t) LWR fuel behaviour.

Mr. Girieud, Mr. Robin (France)

Burn up determination is approached in three different ways. First it is used for code qualification and fuel characterisation. Many fuels have been investigated. The other two methods are carried out in connection with reprocessing plants: the gravimetric input balance is obtained by isotopic correlation technique and non-destructive analysis using gamma spectrometry on the cesium activity ratios.

For all needs, the neutron physicists are the source for codes and data information and the good connection with neutron physicists is essential.

Mr. Watanabe (Japan)

Having stayed in Germany, Mr. Watanabe gave some information only about the activity in the field of fuel reprocessing. In Japan, a new reprocessing plant (800 t/y) is being designed. In this plant the burn up monitor will be adapted to check the residual activity in spent fuels from criticality safety point of view. Last year, the burn up measurement device was installed in the pool of the TOKAI reprocessing plant, and there the measurement was done by using fuels from power reactors.

Mrs. Matausek (Yougoslavia)

The three research reactors (1 TRIGA, 2 HW) and one power plant (PWR, Westinghouse, 600 MW) are the material for the studies on burn up in Yougoslavia.

Calculation methods for burn up being presented in a full paper. the present introduction is restricted to the presentation of non-destructive methods for fuel inventory. There are two lines of investigation. The first one is γ-spectrometry using a high resolution instrument allowing the distribution profile of ^{137}Cs and leading to burn up after correction for cesium diffusion. The second project deals with the detection of defective fuel by activation of the primary coolant, during operation. $^{137}Cs/^{134}Cs$ ratio is measured and used to localize the defective element, with the help of other parameters. These two projects are undergoing research.

Mr. Lysell (Sweden)

In Sweden there are twelve power reactors and one research reactor. Since the national policy is not to reprocess but to store the fuel, the perspective is different.

In Studsvik, the burn up is determined in hot cells using the neodymium method and γ-scanning is made using a standard rod with known burn up.

The Swedish Nuclear Inspectorate also requires a number of measurements in pools. For γ-spectrometry ^{137}Cs is used as monitor along the fuel bundle with collimators in the pool walls. There is a similar facility at the intermediate storage location. At present measurements are also carried out on MOX fuel received as "exchange" fuel from Germany.

Mr. Zwicky (Switzerland)

In Switzerland the companies which operate the 5 reactor power plants make calculations of fuel burn up for core management.

Destructive techniques are carried out only at PSI. For the burn up determination of oxide and carbide fuels, the Nd method is used. Experience has been accumulated during the past 20 years in the framework of many fuel development programs.

The SIMS method is used for radial Gd isotopic distribution determination [IAEA-TC-657/2.3]. The application of this technique to U and Pu radial isotope distribution is being developed.

Mr. Prosyolkov (USSR)

There are 11 power stations with 1000 MW power and more than 30 with 440 MW. Burn up up to 42 000 MWd/t have been achieved on the VVER-1000 reactors with 3-years cycle.

Burn up determination serves 3 purposes: safety in reactor operation; fuel transportation and refabrication; improvement of computer codes.

Both ND and D methods are used, for example

- passive neutron method for burn up estimation of whole fuel assemblies
- γ-scanning for individual fuel rods
- mass spectroscopy for exact measurements

Mr. Bayliss (UK)

The first PWR should be build in UK in the next few years. There will therefore be a change for BNFL from a fixed customer to a more competitive world. The information on new fuels likely to be developed and what this means towards reprocessing is therefore very important. At the next conference on modelling organized by IAEA in Preston, BNFL will present fundamental information on the latest fuel development code including radial power distribution. In connection with the above, the data dealt with in this meeting are needed for testing and validating purposes. Also is very important the knowledge of the effects of burnable poisons.

Mr. Koch (JRC Karlsruhe)

The burnup analysis by the ^{148}Nd method has been carried out routinely in the European Institute for Transuranium Elements since 1966. At the beginning, spent fuel samples of various LWR's were analysed for the purpose of reactor code verification. The LWR were: VA Kahl, Lingen, Gundremmingen, KWO, Sena, Doodeward, Garigliano, Trino Vercellese. Later the method was extended to fuel characterisation as needed in the development of MOX-fuel.

At present the Institute possesses chemical hot cells and α-laboratories equipped with automatic mass-spectrometers and robots which are used for the "isotope analysis" of spent fuel for different purposes such as fissile material control and post-irradiation examinations.

DESTRUCTIVE ANALYSIS: METHODOLOGY AND EXPERIENCE

G.G. SIMON
Federal Republic of Germany

M. ROBIN
France

1. Summary

Three papers were presented in this session.

The first paper by Mr. de Regge (Belgium) addressed the present experience in burnup determination of MOX fuel by mass-spectrometric and radiochemical measurements. Neodymium is determined by mass-spectrometry. Analytical data are compared with calculated data for check of consistency.

The present technique has been proven to be sufficiently reliable and accurate for the needs of the fuel supplier and related parties. Extension of the method to Gd-analysis will require some upgrading of the procedure. Effort to further automatization of the method is anticipated.

The second paper by Mr. Zu-min (P.R. China) was related to burnup determination of high enriched fuel from MTR's on basis of heavy isotope correlation. A calculation model for burnup determination using measured U-234, U-235, U-236 and U-238 concentrations before and after irradiation was presented.

When applying this technique a precision of approximately 3% in burnup could be achieved. Burnup of approximately 50% was determined.

The third paper by Mr. Zwicky (Switzerland) described the radial gadolinium burnup determination by secondary ion mass-spectrometry (SIMS).

The SIMS-technique was applied within the GAP-program. In comparison to the micro-drilling technique (micro-probe) SIMS is faster and provides greater lateral resolution. Samples of fuel with an initial U-235 enrichment of ~3.5% and 3 to 7 wt% Gd_2O_3 have been analysed in the burnup range 2 to 6 GWd/t. Based on several surface scans on one sample the residual isotopic abundance could be determined with an accuracy of $<1\%$; the corresponding relative standard deviation was $<10\%$. Model calculations with WIMS code confirmed the SIMS measurements.

2. State of the Art

Destructive methods for burnup determination

Three methods can be distinguished for the destructive burnup analysis.

2.1. Method Based on Neodymium Isotopes

The method based on the measurement of Nd-148 has been known for about 20 years as ASTM 321-69 and is widely used.

Advantages

The advantages of the method lies on the choice of the fission monitor which is insensitive to the source of the fissions. By the analysis of the other neodymium

isotopes which are available from the same procedure without additional work, it is possible to identify the fission source distribution and quantify the contributions of the different fissile isotopes. The method has been demonstrated within but is not limited to a burnup range between 0.15 and 10% FIMA. The accuracy of the method is 1.5%.

Disadvantages and limitations

The method necessitates elaborate and skillful laboratory measurements with calibrated instruments to achieve the stated accuracy.

The concentration Nd-148 needs to be corrected for Nd-147 neutron capture during irradiation in high neutron fluxes ($>5 \cdot 10^{13} ncm^{-2} s^{-1}$). The independence of the source of fission is least when advanced fuels are analysed incorporating burnable neutron poisons or mixed oxides due to the higher contributions of U-238 and Pu-241 to the fissions.

Sensitivity

The method is applicable to very small samples of about 100 µg of fuel and up to the scale of dissolved assemblies at the reprocessing stage.

2.2 Radiochemical Measurements based on gamma-emitting isotopes

The absolute measurement of gamma-emitting fission products in well-calibrated conditions provides a means for burnup determinations when the actinide concentrations in the liquid are also available.

Advantages

The advantage of the method is its application to dissolved fuel solutions without chemical separations or elaborate preparations being needed.

The method provides the simultaneous acquirement of data for all gamma-emitting isotopes which can be used for fuel characterisation and isotopic correlations. The accuracy is 2 to 3% under the best conditions.

Disadvantages and limitations

The in-pile decay corrections necessitate the accurate knowledge of the fuel power history and the accuracy is dependent on the chemical stability of the isotopes of interest in the fuel during irradiation when small samples are considered. For assemblies or entire fuel pins the migration of Cs isotopes does not influence the accuracy of the results.

Isotope ratio measurements may be less sensitive to the power history.

Sensitivity and range

The reliability of the corrections for decay and possible migration limits the range to within 0 - 4% FIMA. As for other destructive analytical methods the sensitivity is high and very small fuel samples of the order of a few µg up to whole assemblies can be analysed.

2.3 Heavy Isotopes Method

This method known as ASTM E 244-80 or the actinide mass-spectrometric method uses the differences induced in the isotopic ratios during irradiation as the basis for burnup determination.

Advantages

Only isotopic ratios for actinide elements are used in the calculations which eliminates the need for absolute concentration measurements and simplifies the calibration procedure. No fission-product measurements are necessary. An accuracy of 3% can be reached.

Disadvantages and limitations

The method although being described for all fuels, is in practice only applicable to moderately to high enriched uranium fuels such as MTR fuel elements or targets. For other fuels and for current fuel with a burnup exceeding approximate 20%, neutron spectrum averaged cross-sections are necessary to apply a number of corrections to the measured data. Those cross-sections are not always readily available.

Furthermore as the method relies on changes in isotopic ratios, the initial ratios in the fuel, also for the minor isotopes should be well-known.

Sensitivity and range

Because of the isotopic ratio differences involved the method will reach the stated accuracy when the burnup exceeds approximately 5%.

The method is applicable to small sample sizes, of a few µg, as well as to entire fuel elements.

2.4 Isotope Correlation Techniques

Isotopic data either obtained from nondestructive measurements or from destructive fuel analysis after dissolution provide a measurement of fuel burnup.

Advantages

The advantages of isotopic correlation techniques, particularly using ratios between the isotopes of a single element is their application to small or large samples without the need for quantitative measurements or absolute calibration. The correlations rely on the physical transmutation laws of the isotopes involved and can be selected to provide the optimal sensitivity for any particular case.

Disadvantages and limitations

The reliability of isotopic correlation techniques increases with the amount of data available and a minimal number of data obtained by other methods is necessary to establish the correlations. The correlations are perturbed to different extents by non-standard power histories or other factors affecting the isotopic pattern of the fuel, such as the vicinity of neutron reflectors, water gaps and neutron absorbing rods. Part of the perturbation can be corrected by calculations which take into account the sample history.

Sensitivity and range

The sensitivity of the method extends from very small samples to whole fuel assemblies but the reliability of the data is dependent on the type of correlation. If historical data for similar fuels and measurement accuracy are used, burnups can be determined to within 3 to 5% in the range of 1 to 5% FIMA.

NON-DESTRUCTIVE ANALYSIS: METHODOLOGY AND EXPERIENCE

P. DE REGGE
Belgium

G. LYSELL
Sweden

1. Summary

Seven papers were presented in this session.

The first paper by P. Blanpain (Belgium) et.al. addressed the experimental investigation of gadolinium at low burnup.

The "Burn" method uses the Cs-137 activity and standard burnup samples for burnup determination.The method yields accuracies of better than 3% (2σ). It was demonstrated in the GAP program and calibrated against destructively obtained data by SIMS.

The second contribution by Mr. Aleksandrov (USSR) consisted of two papers. Both addressed the burnup determination of VVER-440 Fuel.

The first report related to a mass- and gamma-spectrometric method based on Cs-137 buildup and comparison with standard fuel samples. Precision of measurements was approximate 8% when referred to mass-spectrometry data.

The second paper addressed a method based on passive neutron verification. The method is based upon calculation of the specific neutron emission rate as a function of burnup . It provides the basis for the construction of a burnup-meter.

The third contribution by Mr. G. Simon (Germany) described a measurement technique for burnup determination at the storage pond in the reprocessing plant.

It has been demonstrated that fuel identification and burnup determination is possible when using a combination of active and passive neutron measurements. The methods yields accuracies for PWR UO_2 fuel of 1.5 GWd/t for minimum cooling periods of 1.5 year.

The method is also suitable for determining the input balance of a reprocessing plant if the isotope correlation technique is applied. A simplified spent fuel monitor is under development.

The fourth contribution by Mr. Prosyolkov (USSR) was related to calculation methodologies. Experimental investigations for the determination of n, γ (U-238) and n, f (U-235) reaction rates in an elementary cell near the core were performed at BOL. On the basis of this information the Pu-239 buildup and U-235 fission rate were calculated and used as input in 2 DT calculations. These resulted in a 200°C central temperature decrease (at about 448 W/cm MLHR).

The sixth paper by Mr. Koch (CEC) addressed the isotope correlation technique (ICT). The performance of the method for burnup determination was demonstrated on a number of various isotopes (e.g. Pu-240, Xe-131/Xe-132, Xe-134/Xe-137, Cs-134/Cs-137).

The method is largely insensitive to fuel and reactor type. The method yielded high precision data within the burnup range 1 to 5% FIMA, and has been applied to fuel reprocessing and for fuel characterisation.

The seventh contribution by Mr. Smith (CEC) was related to the burnup determination method employed by EURATOM Safeguards.

Results of using the FORK device, a passive neutron and gamma measuring device, showed satisfying results for fuel exposure verification. For UO_2 fuel a 5% precision in burnup determinations was reached. For MOX fuel about 15% accuracy was shown.

2. State of the Art

2.1 Methods based on gamma-spectrometry

- The gamma-spectrometry technique for burnup determinations is applied in all countries as a standard method.
- In Argentine, France and Sweden underwater installations are operational.
- Two different standard methods are employed: Absolute method based on Cs-137 activity and the comparative method based on ratio of Cs-134/Cs-137.
- The gamma-spectrometry methods are applicable to burnup determinations of fuel assemblies, fuel rods, fuel pellet and samples including fuel containing solutions.
- They are often performed before destructive analysis is carried out.

Advantages

- Gamma-scanning can be performed on fuel assemblies, single fuel rods, fuel pellets or samples.
- The method can provide detailed information on axial and radial isotope distributions.
- If used comparatively, no absolute calibration is required (e.g. if based on "Burnotheque")
- The Cs-137 build-up is linearly related to burnup
- No burnup limitation in practice.

Disadvantages and limitations

- Long measuring times.
- For the method based on Cs-137 calibration of data is required.
- For the Cs-134/Cs-137 method, information regarding the formation of Cs-134 by capture in Cs-133 is needed.
- Migration of Cs-isotopes in fuel can influence the precision of the burnup determination.
- Complex data evaluation procedures.
- Not suited for defective fuel rods.

Sensitivity

- The accuracy of burnup determinations is about 3% when using the comparative method (e.g. with "Burnotheque").

2.2 Methods based on passive neutron and gross-gamma activity measurements

- These methods use a combination of detectors:
  - Fission chambers
  - Gamma-detectors (GM-tube)
- Neutron emission from Cm (Cm-244 being the main emitter) and the total gamma activity are measured.

- The measured data is converted into burnup data using a calibrated standard.
- the measuring device can be easily handled in storage ponds. It is mobile and usable in various locations.
- Its main application is within the EURATOM Safeguards activities, e.g. as a check of fuel exposure data and integrity of fuel assembly.
- The method is also suited for the determination of cooling times.

Advantages

- Simple measuring technique, applicable at reactor fuel storage pond.
- Mobile system, transferable between different locations.
- Easy to handle.
- Short measuring times (e.g. 2 minutes/fuel assembly and one point)
- Simple data evaluation (outline)

Disadvantages and limitations

- No information about isotopic composition
- Only gross burnup data measureable
- Fuel assemblies need to be lifted from storage position for measurement.
- Interference with refuelling operations
- Can only be applied to burnup levels higher than approximately 5 GWd/t.

Sensitivity

- For UO_2 standard WR fuel, burnup determination of an accuracy of approx. 3 GWd/t is feasible
- For MOX WR fuel, precision of burnup determination is approx. 10 to 15%.

2.3 Methods based on passive and active neutron measurement and total-gamma scanning

- This method employes a combination of detectors and a neutron source:
  - Several fission chambers (e.g. 2 to 4)
  - A gamma-detector (GM-tube)
  - A displaceable neutron source (Cf-152)
- The neutron emission rate from Cm-isotopes and the total-gamma-activity are measured simultaneously during the "passive" measuring phase.

  For cooling time > 1.5 years no gross γ-measurements are required.

  In combination with the neutron source the neutron flux multiplication originating from the fissile material is determined.
- Both data sets are analysed for:
  - Burnup
  - Criticality of the spent fuel
  - Fuel assembly identification for an independent check of supplier's data.

- Combined with isotope correlation data also the balance of fissile material input into reprocessing plants and the ratio of Cm to Pu can be obtained.
- Much experience with the method has been obtained in Germany. The method will be introduced in France for fissile material determination and criticality checks.

Advantages

- The method delivers several data required for spent fuel transport, storage and reprocessing:
  - Burnup
  - Criticality
  - Fuel identification
  - Ratio of Cm to Pu
  - Inventory of fissile material
- Nearly insensitive to irradiation history.
- Calibration can be performed on mock-up devices.
- High accuracy of measurement and burnup determination.
- Measuring equipment commercially available

Disadvantages and limitations

- Precise positioning of detectors and neutron source required.
- Dedicated measuring bench. Environment around measuring site must be free from disturbing activity.
- For determination of Cm/Pu ratio and inventory of fissile material complex data evaluation is required.
- MOX fuel can not fully be identified.
- No experience has been made for cooling times <0.5 years. Application is probably inhibited by large gamma-background and scatter in the decay functions.
- For cooling times <1.5 years additional gamma-activity data have to be measured:
  - Gross gamma-activity or
  - Ce-144 gamma-activity

Sensitivity

- For fuel assemblies with a cooling time >1.5 year the burnup can be determined with an accuracy of 1.5 GWd/t.
- Neutron emission rate (passive method) can be measured with an absolute accuracy of <10%.
- The neutron flux multiplication is determined to within 2%.

CALCULATION MODELS AND THEIR QUALIFICATIONS

K.A. ALEKSANDROV
Union of Soviet Socialist Republics

H.U. ZWICKY
Switzerland

1. Summary

The first paper by Mrs. Matausek (Yugoslavia) described results of calculations performed for prolonged fuel burnup in LWRs. A computer code package has been used comprising a neutronic code for calculating fuel pin and fuel assembly parameters versus burnup and a 2D diffusion theory code for calculating overall power and burnup distribution in the core. Repeating the global core calculations with a specified burnup step for a typical equilibrium cycle, the entire range of burnup values is covered.

The second paper of Mr. Crijns (IAEA) described work started under an IAEA coordinated research program. It concerned the creation of complete computer packages, consisting of several individual computer programs, for performing in-core fuel management in developing countries. In the meantime new developments have been made and will be reported during a TC meeting in July 1988 in Madrid. In addition a benchmark experience is being considered to verify and test the in-core fuel management program packages.

2. Field of Application

The requirements for burnup calculations depend on specific applications (see table).

Applications	Requirements
Fuel testing	- establishing reference point - feedback fuel development/design - isotopic composition vs burnup
Reprocessing	- checking utility specifications - criticality reasons - inventory
In-core fuel management	- verification of codes - reload patterns - economic considerations - safety considerations
Safeguards	- identification of fuel

3. State of the Art

- A number of institutes have developed codes for predicting burnup and have performed measurements for code validation. However, these codes are usually not commonly available for commercial reasons.

- Codes readily available can often not be used for certain fuel types (e.g. fuel containing burnup absorbers) or are not reliable.
- No openly available code was discussed during the meeting, however, reference was made to available codes such as:

  ORIGEN

  KORIGEN

  WIMS

  HAMMER
- Some codes would be inproved if they could be validated against real data and if newly available data were used (e.g. on n, α cross-sections).
- Data for benchmarking of codes are available in literature.

The group identified the below listed needs and recommends future R&D in the following fields:

1. The conversion from "atompercent burnup" to energy produced (MWd/t) can introduce systematic discrepancies (This observation has been made for different reactor types in different countries). This may be due to the different ways the energy is released with burnup.
2. The potential to determine the radial burnup by other techniques such as SIMS should be assessed.
3. There is a need for experimental data on spent fuel (gadolinium-absorber and MOX) in order to verify reactor physics codes and fuel performances. It is recommended that earlier benchmark tests and comparisons should be looked at. If information is lacking on the fuel history it should be investigated if these data could be made available.
4. In order to assess the accuracy of burnup determinations an interlaboratory test is proposed. For this purpose it should be investigated if "REIMEP" (Quality control of U, Pu analysis in spent fuel organized by BCMN Geel) could be extended for this purpose. The IAEA should investigate the possibility of enlarging the program outside the EC.
5. The burnup analysis of highly burnt water reactor fuel (~5%) and of MOX fuel will require the determination of the fission source. Therefore all information on the isotopic abundance of actinides and selected fission products should be made available for the evaluation of the burnup by different methods. The fission yields used in the determinations should be published with the results.
6. Results of NDA burnup determinations should be checked against calculations and where possible compared to results of destructive analyses. The findings should be published to assess the reliability of the NDA.
7. The accuracy of nuclear data related to neutron emission of spent fuel should be reviewed and reported.
8. The group recommends a follow up meeting to review the implementations of the proposed recommendations within 3 years.

DESTRUCTIVE ANALYSIS: METHODOLOGY AND EXPERIENCE

ACTUAL EXPERIENCE IN BURNUP DETERMINATION OF MIXED OXIDE FUEL

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Abstract

The current radiochemical techniques used at SCK/CEN for the destructive burnup determination of mixed oxide fuels are reviewed. The procedure for fuel sampling and dissolution, for the chemical separations and the mass-spectrometric and radiochemical measurements are briefly described. The concentrations of uranium and plutonium in the fuel are obtained by mass-spectrometric isotopic dilution using ^{233}U- and ^{242}Pu-spikes. Neodymium is determined by mass spectrometric isotopic dilution using a ^{146}Nd spike and ^{147}Nd as a tracer in the chemical separations. The transplutonium elements and the gamma-ray emitting fission products are determined by alpha- and gamma-ray spectrometry, respectively the calculation method to obtain the weighted fission yields averaged over the irradiation period is commented. The procedure contains several cross-checking points where the consistency of the data is evaluated and thereby provides virtually a number of independent burnup measurements, increasing both the reliability and the accuracy of the results. Typical results obtained for MOX fuels in thermal recycling and fast breeder reactor fuels are presented as well as data on the radial distribution of burnup, fissionable nuclides and fission products.

1. INTRODUCTION

The most accurate determination of the amount of energy produced by nuclear fuels still relies on destructive radiochemical procedures, aiming at the measurements of the concentrations of selected fission products after dissolution of the fuel. The procedure using ^{148}Nd as a fission indicator is successful and widely accepted as the reference method for burnup measurement, since it has been recognised as ASTM E321-69. Although the principles are always maintained, many laboratories use chemical separation techniques differing from the ASTM method and more extensively exploit the detailed isotopic composition of the separated neodymium to obtain information on the neutron spectrum and irradiation characteristics. For several years, burnup determinations were carried out at SCK/CEN on a great variety of commercial and prototype nuclear fuels using a procedure which is derived from the ASTM E321-69 method. Its features have been continuously optimised and adapted for the specific characteristics of the fuel to be analysed. Because of the analytical efforts and costs involved in a burnup determination, the SCK/CEN method also provides sufficient redundancy for the major parameters to obtain a very high level of confidence in the results obtained on a single fuel sample. Recently adaptions had to be made to analyse radially drilled samples from MOX or Gadolinia-doped fuel rods. The method is briefly described below and representative data obtained for different kinds of fuel are presented.

2. SAMPLING AND DISSOLUTION

Three kinds of samples are currently submitted for burnup determination :

- pieces of fuel rods with a length of 1 to 5 cm and containing 5 to 20 g ;
- aliquots taken from dissolved fuel solutions ;
- slurries containing fuel chips, lubricant and epoxy remnants obtained from radially drilled samples.

TABLE I : Plutonium Isotopic Composition in the Different Phases (Atompercent)

Experiment Code	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
DOD - P354 - 1					
Solution	1.26	52.73	31.48	9.86	4.67
Residue	1.71	37.79	38.43	14.48	7.59
DOD - V1670 - 1					
Solution	1.39	42.24	38.06	11.02	7.29
Residue	1.48	41.42	38.29	11.24	7.57
SENA - D1					
Solution	1.37	64.26	22.46	8.81	3.10
Residue	1.67	55.62	27.37	11.24	4.10
SENA - D2					
Solution	1.86	54.36	27.31	11.41	5.06
Residue	2.58	39.65	34.14	15.93	7.70
SPC III-ZOB 100					
Solution	1.19	50.18	33.84	9.98	4.81
Residue	0.24	76.71	19.39	2.66	1.00
SPC IV - G 55					
Solution	1.64	49.41	35.60	8.46	4.89
Residue	1.38	52.56	33.70	8.34	4.01

Completeness of dissolution of the sample is an essential feature for accurate burnup determination since undissolved residues have been shown to be enriched in refractory actinide and rare earth oxides. Particularly in MOX fuels, the manufacturing process or the radiation induced segregation of plutonium enriched zones can result in residues difficult to dissolve and requiring the use of hydrofluoric acid to complete dissolution. As an example, data reported earlier [1] for Pu solubilities are shown in Table I. From the data it is obvious that the undissolved plutonium in the residue corresponds essentially to recycled material whereas the radiation-induced plutonium is easily dissolved. The slurries obtained from radially drilled samples are evaporated while nitric acid is added to dissolve the fuel, eventually followed by a nitric-hydrofluoric acid mixture. For this kind of samples, the initial mass of the fuel cannot be obtained which prevents cross-checking of the results on the basis of material balance calculations. Also the evaporation is time-consuming and preliminary filtration might be an alternative ; selective leaching of rare earth fission products is not expected but more soluble fission products such as Cs will yield unreliable results.

Fuels with a burnup exceeding 4 atom percent FIMA will show undissolved residues even after treatment with hydrofluoric acid. They consist mainly of noble metal fission products and contain negligeable amounts of actinides. Typical compositions have been reported earlier [2].

3. ANALYSIS SCHEME

To preserve accuracy in the remote manipulations, all aliquotations and dilutions are based on weighed quantities. To provide internal verifications in the procedure those aliquotations are caried out with known volumes, which allows the density of every aliquot to be compared with the density of the mother liquor. This procedure allows tracing of operational errors and cross-checking without lengthy duplicate analyses. The analysis scheme for the burnup measurement is shown in figure 1 and is designed to provide duplicate measurements of the most important parameters, preferentially using different measurement methods. The basic analytical techniques are mass-spectrometric isotopic dilution and radiometric analysis. Both techniques are sufficiently sensitive to perform the measurements on very small quantities with low radiation exposure for the laboratory operators. The limiting aliquot for the chemical separations depends on the fuel composition and irradiation characteristics ; for MOX fuels, usually the neodymium quantity is the limiting factor and quantities of 1 to 3 µg are taken. At lower quantities, neodymium and samarium contaminat. from reagents and laboratory glassware, as indicated e.g. by the presence of ^{142}Nd, interferes with the mass spectrometric analysis.

For the isotopic dilution analysis, weighed spike aliquots ^{242}Pu, ^{233}U and ^{146}Nd are added to an aliquot of the solution. Mixed spikes are used for larger series of similar composition and irradiation characteristics, but normally each spike is added individually to obtain an approximate 1/1 ratio versus the major isotope in the sample. The ^{146}Nd spike has been irradiated in a high flux reactor to produce ^{147}Nd which traces Nd through the chemical separations.

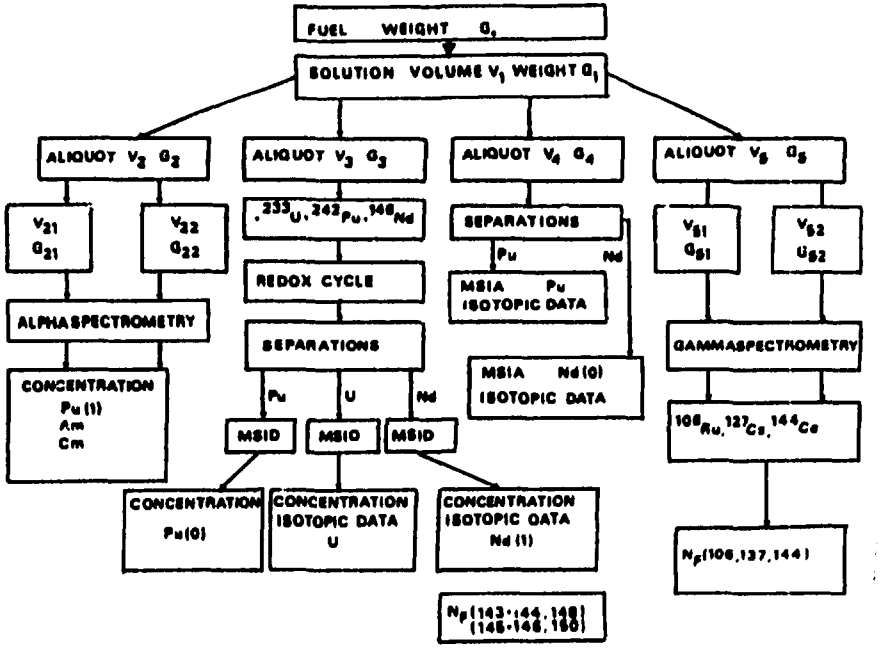

FIG. 1 PROCEDURE FOR BURNUP DETERMINATION

4. CHEMICAL SEPARATIONS

4.1 Separation of Uranium and Plutonium

Prior to any chemical separation, the complete isotope exchange with the ^{242}Pu spike is achieved using a $FeCl_2$-NH_2OH-$NaNO_2$ redox cycle. U and Pu are separated from the bulk of the fission products in 9 M HCl on a Dowex 1-X4 anion-exchange column. Uranium is eluted with 8 M HNO_3. Plutonium is recovered with 0.35 M HNO_3. This separation yields U- and Pu-fraction of sufficient purity for mass spectrometric isotopic analysis except for the measurement of ^{230}Pu, which is usually done by alpha-ray spectrometry. As this procedure also separates ^{241}Am from the Pu fraction, no additionnal separations are necessary as long as isotopic analysis is carried out within one week after the separation.

4.2 Separation of Neodymium

The effluents containing the fission products and trivalent actinides is evaporated and treated with hydrogen peroxide to destroy organic matter.

The residue is redissolved in 8 M HNO_3 and passed on a mixed anion exchanger-PbO_2 column. This eliminates Ce from the rare earth fraction, which not only isotopically interferes with ^{142}Nd and ^{144}Nd but also forms the main radiation source in short-cooled fuel. The effluent is again evaporated and converted to 0.1 M HCl. The solution is transferred to a column of kieselguhr supporting di-2-ethylhexyl phosphoric acid, which retains the trivalent rare earth and actinides separating them from other fission products. Actinides are washed out using their di-ethyl-triamino-penta-acetic acid complexes in a lactic acid buffer at pH 2. The rare earths are fractionated by gradually increasing the HCl eluent molarity. Nd is recoverd with 0.16 M HCl and is easily identified by the ^{147}Nd tracer.

4.3 Separation of Gadolinium

The use of gadolinium as a burnable neutron poison in advanced uranium fuels aiming at higher burnup frequently provokes requests for its isotopic composition and concentration in conjunction with the burnup measurements. For this purpose, enriched ^{152}Gd is added as a spike to the primary aliquot simultaneously with the other spikes. Preliminary irradiation of the spike in a high flux reactor produces the gamma-ray emitter isotope ^{153}Gd which can be used as a tracer for gadolinium throughout the chemical separations and particularly during the reversed-phase extraction chromatography for the rare earth fractionation. In gadolinia-doped fuels, it is also of particular importance to obtain information on the concentration levels of other rare earths in the unirradiated gadolinium from the fuel manufacturer, as their presence might seriously interfere with the measurements made for burnup determination. Figure 2 shows typical gadolinium isotopic analysis data for a series of radially drilled samples.

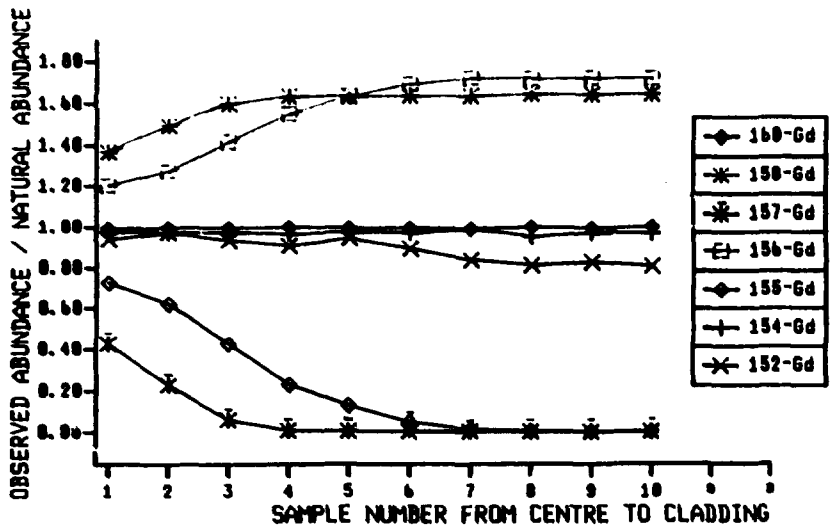

FIG. 2 GADOLINIUM DATA IN RADIALLY DRILLED SAMPLES
RATIO OF OBSERVED TO NATURAL ABUNDANCE

5. MASS-SPECTROMETRIC MEASUREMENTS

The separated U-, Pu-, Nd- and possibly Gd-fractions are evaporated to dryness and redissolved in 1 M HNO_3 to achieve a concentration around 1 g l^{-1} for U and Pu and 50 mg to 1 g l^{-1} for the rare earts. The solutions are loaded on the side filaments of triple rhenium filament beads for mass spectrometric analysis. The measurement procedures have been reported earlier [3]. The isotopic dilution measurements provide the concentrations of U and Pu in the fuel and the fission product concentrations for the masses 143, 144, 145, 146, 148 and 150. The high enrichment of the ^{146}Nd spike allows the isotopic abundances for the neodymium isotopes to be measured in the spiked as well as in the unspiked aliquot, the agreement being usually within 0.2 % for the major isotopes to about 1 % (relative) for ^{150}Nd. Significant differences provide an indication for faulty measurements or excessive contamination by natural neodymium or other rare earth elements. As will be further indicated the neodymium isotopic ratios are important parameters for accurate burnup calculations.

6. RADIOCHEMICAL MEASUREMENTS

Radiochemical measurements are carried out on weighed dilutions of the dissolved fuel solution, usually without chemical separations. Total alpha-ray counting combined with alpha-ray spectrometry provides the alpha desintegration rates for $^{239+240}Pu$, ^{238}Pu + ^{241}Am, $^{243+244}Cm$ and ^{242}Cm [4]. As ^{243}Cm has a negligeable contribution and the plutonium isotopic ratios are known from the mass-spectrometric analysis, the concentrations for Pu, Am and Cm are readily obtained. The agreement between the Pu concentration obtained from mass-spectrometric and from alpha-ray spectrometric measurements is normally within 2 % except when the $^{239+240}Pu$ fraction in the alpha-ray spectrum is very small due to large activities of ^{241}Am and Cm isotopes ; this can occur in high burnup recycled plutonium fuel.

Gamma-ray spectrometric assay of the solutions with a calibrated semiconductor spectrometer yields the concentrations of the gamma-ray emitting fission products, particularly ^{144}Ce-Pm, ^{137}Cs and ^{106}Ru-Rh. In order to use the data from radiochemical measurements for burnup calculations, they have to be corrected for out-of-pile and in-pile decay. Particularly long and complex irradiation histories at different power levels can compromise the usefulness and reliability of data obtained for radionuclides with shorter halflives. In principle, fission-product concentrations can be obtained for two additionnal mass numbers 106, 137 and a duplicate measurement for mass 144. However, in practice, at higher irradiation temperatures ^{137}Cs migrates radially and axially in the fuel, and ^{106}Ru might accumulate in insoluble residues or partially volatilise during chemical treatments. In spite of those drawbacks, good agreement is frequently observed between the different fission indicators.

7. BURNUP CALCULATIONS

The burnup value is expressed as follows as the number of fissions per initial heavy metal atom (FIMA) in the nuclear fuel

$$FIMA = \frac{N_F \quad \text{(number of fissions)}}{No_{HM} \text{ (initial number of heavy metal atoms)}}$$

The number of fissions is derived from the concentration of a given fission product in the irradiated fuel and from its fission yield [5].

$$N_F = \frac{C_{F_i} \text{ (concentration of fission product i)}}{Y_{F_i} \text{ (fission yield for mass i)}}$$

The number of initial heavy metal atoms, No_{HM}, is obtained by adding the number of residual heavy metal atoms (U + Pu + Cm + ...) to the number of fissions N_F.

Except for the fission yields, all parameters can be obtained in a straight forward way from the described analytical procedure. As the fission yields are dependent on the fissionable isotope and on the neutron energy

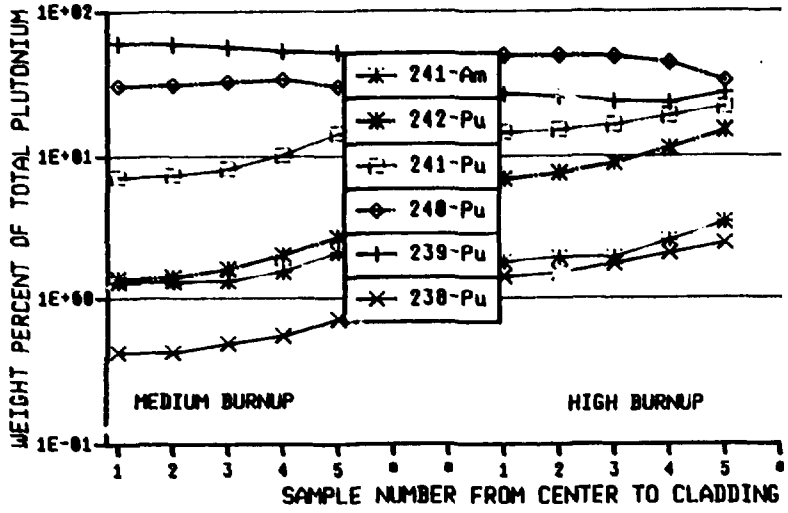

FIG. 3 PLUTONIUM RECYCLING FUEL
ACTINIDE DISTRIBUTION IN RADIALLY DRILLED SAMPLES

TABLE II : Pu recycling mixed oxide Fuel
Neodymium burnup data on radially drilled samples

MEDIUM BURNUP - Atompercent FIMA					
SAMPLE	I	II	III	IV	V
Indicator					
$^{143+144}Nd$	2.852	3.022	3.196	3.716	4.310
$^{145\ 146}Nd$	2.899	3.060	3.229	3.759	4.353
^{148}Nd	2.895	3.071	3.236	3.755	4.342
^{150}Nd	2.897	3.071	3.225	3.773	4.359
MEAN VALUE	2.886 ± 0.023	3.056 ± 0.023	3.222 ± 0.018	3.751 ± 0.024	4.341 ± 0.022
HIGH BURNUP - Atompercent FIMA					
SAMPLE	I	II	III	IV	V
Indicator					
$^{143+144}Nd$	7.444	7.611	7.819	8.570	10.024
$^{145\ 146}Nd$	7.529	7.706	7.059	8.642	10.116
^{148}Nd	7.491	7.693	7.999	8.579	10.065
^{150}Nd	7.416	7.617	7.891	8.444	9.939
MEAN VALUE	7.470 ± 0.050	7.657 ± 0.050	7.942 ± 0.107	8.559 ± 0.083	10.036 ± 0.075

spectrum, they are not necessarily constant over the irradiation period. Therefore a weighted average fission yield can be defined as

$$\bar{Y}_F = \frac{1}{1+P}\left[\left[\frac{\sum_j \phi_j N_{A_i} \sigma_{ij} Y_{ij}}{\sum_j \phi_j N_{A_i} \sigma_{ij}}\right] + P_{BOL}\left[\frac{\sum_j \phi_j N_{A_i} \sigma_{ij} Y_{ij}}{\sum_j \phi_j N_{A_i} \sigma_{ij}}\right]_{EOL}\right],$$

whereby the summations are carried out for different neutron-energy groups with a flux ϕ_j and a corresponding fission cross section σ_{ij} for fissionable isotope A_i. The formula assumes a linear interpolation between the situations with indices BOL and EOL referring to the beginning and at the end of the irradiation whereby the relative fission density between the two phases is given by P. This formula is very useful for MOX fuels at moderate burnup levels (2 to 4 % FIMA) in Pu recycling fuel and for fast breeder reactor fuel at higher burnup. In practice, the number of neutron energy groups is limited by availability of fission-yield data for each group and only epithermal and the thermal neutrons are considered. For advanced uranium fuel or MOX fuels with high burnup, the formula is unreliable mainly because a linear interpolation between BOL and EOL is not representative for the irradiation history. The weighted average fission yields are then calculated from the observed ratios of neodymium isotopes. In priciple, sufficient ratios are available to calculate the contributions of ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu but the redundancy of the measurements is lost as more parameters have to be calculated. Instead, the ratio of the fission contributions of ^{239}Pu and ^{241}Pu is calculated according to

$$\frac{N_{F241}}{N_{F239}} = \frac{1}{1+P}\left(\frac{\sigma 241}{\sigma 239}\right)\left[\left(\frac{N_{241}}{N_{239}}\right)_{BOL} + P\left(\frac{N_{241}}{N_{239}}\right)_{EOL}\right],$$

and the ratio of $^{148}Nd/^{150}Nd$ is then used to calculate the ratio of fission contributions from Pu isotopes to ^{235}U. The fast fissions originating in ^{238}U are thereby neglected. The weighted average fission yields are then calculated using the fractional fission contribution for each fissionable isotope as the weighting factors. The same weighting factors are used for the caluclations of the conversion factor FIMA to energy produced expressed in units of MWd kg^{-1}. Two aspects still deserve to be emphasized. First, the value obtained for ^{144}Nd has to be corrected for the quantity of ^{144}Ce still present at the Ce/Nd-chemical-separation time. Second, the appreciable conversion of ^{143}Nd to ^{144}Nd and ^{145}Nd to ^{146}Nd in a thermal neutron flux precludes the independent use of those fission indicators. Their respective sum is used in conjunction with the sum of the corresponding fission yields.

8. EXPERIENCE WITH BURNUP DETERMINATIONS IN DIFFERENT FUELS

A few examples can be given to illustrate the results obtained by the described procedure for burnup determination. Figure 3 shows the actinide isotopic composition for a series of radially drilled samples at medium and high burnup from plutonium recycling mixed oxide fuel. Table II shows the burnup results obtained from the neodymium isotopes in the same fuel

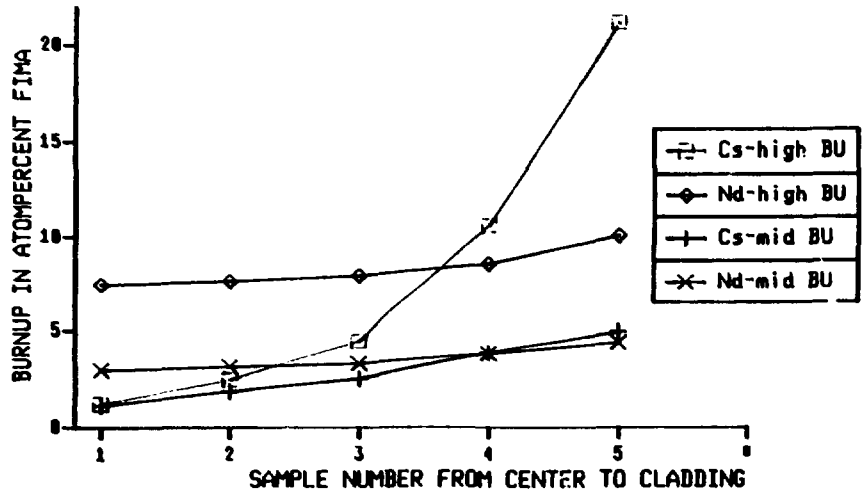

FIG. 4 BURNUP RESULTS FOR RADIALLY DRILLED SAMPLES

TABLE III : Burnup Measurements on Fast Breeder Reactor Fuel with enriched Uranium

Uranium weight %		Plutonium weight %		Burnup FIMA Atompercent	
U	71.2	Pu	28.1		
^{234}U	0.288	^{238}Pu	0.824	^{143}Nd	8.59
^{235}U	31.670	^{239}Pu	75.177	^{144}Nd	8.78
^{236}U	1.252	^{240}Pu	17.091	^{145}Nd	8.79
^{238}U	66.789	^{241}Pu	5.262	^{146}Nd	8.99
		^{242}Pu	1.647	^{148}Nd	8.83
				^{150}Nd	8.72
				MEAN	8.78 ± 0.13
Relative contribution to fissions $^{235}U/^{239}Pu/^{241}Pu$ = 27.8/67.2/5.1					

TABLE IV : Burnup Measurements for Fuel Cladding Interaction Studies

Uranium weight %	Plutonium weight %	Burnup FIMA Atompercent
U 74.4	Pu 25.1	
^{234}U 0.456	^{239}Pu 0.485	^{143}Nd 0.277
^{235}U 61.868	^{239}Pu 73.291	^{144}Nd 0.283
^{236}U 0.338	^{240}Pu 20.486	^{145}Nd 0.276
^{238}U 37.338	^{241}Pu 3.958	^{146}Nd 0.279
	^{242}Pu 1.780	^{148}Nd 0.278
		^{150}Nd 0.278
		MEAN 0.2785 ± 0.0024
		^{144}Ce 0.299
		^{137}Cs 0.241
Relative contribution to fissions $^{235}U/^{239}Pu/^{241}Pu$ = 74.6/24.1/1.3		

samples. The fission contribution of each fissionable isotope remains relatively constant for each sample series but the burnup value itself varies over 50 % in the medium burnup sample to 34 % in the high burnup sample. In figure 4, the burnup results from neodymium and ^{137}Cs measurements are shown for the same samples. Radial migration of ^{137}Cs is obvious and ranges from a depletion of about 60 % at the fuel central part to an enrichment of 18 % in the outer fuel rim. It has also been observed that the total ^{137}Cs quantity from radial drillings is lower than the value expected from burnup due to fact that part of the ^{137}Cs migrates out of the fuel and sticks to the cladding, which has not been taken into account.

The results obtained for a fast breeder reactor mixed oxide incorporating enriched uranium are displayed in Table III. In this case, the weighted average fission yields have been calculated using the actinide contributions at BOL and EOL. Good agreement is obtained for the different neodymium fission indicators and it can be observed that conversion of ^{143}Nd and ^{145}Nd into respectively ^{144}Nd and ^{145}Nd is quite small as expected in a fast neutron flux.

Particular fuel-cladding interaction tests necessitated the measurements of very low burnup in mixed oxide fuel irradiated under cadmium cover. Typical results are shown in Table IV.

9. CONCLUSION

The results presented illustrate the application of the procedure for burnup measurements to a variety of mixed oxide fuels. As the data in general are sufficiently reliable and accurate with respect to the needs of the fuel manufacturer or parties interested in the measurements, no real incentive has been felt to improve the procedure except for the implementation of optional features such as gadolinium analysis, and treatment of radially drilled samples. Further developments are mainly directed to reduce the analytical effort and personnel radiation exposure by increasing the sensitivity to analyse smaller quantities and by automation of the procedures.

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HEAVY ISOTOPE METHOD FOR BURNUP DETERMINATION OF HFETR FUEL ELEMENTS

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Abstract

In this paper, a set of formulae for burnup determination of fuel element of HFETR is deduced and found. The highest burnup level of U-235 is 60% and the uncertaintly of burnup values got by the set of formulae is 3%.

1. INTRODUCTION

Burnup Determination of nuclear fuel is very important to reactor evaluation, fabrication study of fuel elements and its economy etc. Our work in this area has been for ten years and some good results have been obtained [1,2].

Mass spectrometry is the most important method in destructive burnup determination of nuclear fuel because of no requirring chemical recovery and having good precision. It includes two ways: fission product method (burnup monitor method) and heavy isotope method. In the first one, the ^{148}Nd as monitor is generally accepted and used [3,4]. In the latter one, the change of isotope abundances of uranium and plutonium and the ratio of uranium and plutonium component are determined, as soon as the burnup value is calculated by formulae and nuclear constants. The both are standard methods of ASTM[5]. The first one has the best precision for various types of fuels, if the burnup monitors are suitably selected; the second one has measurement terms less than first one, but its usefulness is limited by reactor conditions, fuel types and measurement precision.

The HFETR is a High Flux Engineering Test Reactor in our Center, its fuel element is made of uranium and aluminium alloy, the weight of uranium in the element is 25.4%, the abundance of ^{235}U is 90%, the length of the active core 1000mm, thermal neutron flux 3.6-4.5 10^{14} n/cm^2.s, the fast neutron flux 1.2-1.5 10^{15} n/cm^2.s, the highest burnup level of ^{235}U 60%. Because of this conditions the formulae recommended in ASTM E244 couldn't be used here, so we have deduced and found a set of formulae for burnup determination of fuel element of HFETR, and the uncertainty of the burnup value is 3%.

2. PRINCIPLE OF HEAVY ISOTOPE METHOD AND DEDUCED FORMULAE

Let N_4 and N_4^0, N_5 and N_5^0, N_6 and N_6^0, N_8 and N_8^0 respectively stand for the number of ^{234}U, ^{235}U, ^{236}U, ^{238}U atom before and after irradiation; $R_{4/5}$ and $R_{4/5}^0$, $R_{6/5}$ and $R_{6/5}^0$, $R_{5/8}$ and $R_{5/8}^0$ respectively stand for the ratio of ^{234}U-to-^{235}U, ^{236}U-to-^{235}U, ^{235}U-to-^{238}U before and after irradiation; ϕ_t and ϕ_f respectively stand for the flux of thermal neutron and fast neutron; σ_{4t} and σ_{4f}, σ_{5t} and σ_{5f}, σ_{8t} and σ_{8f} respectively stand for the cross-section of thermal and fast neutron of ^{234}U, ^{235}U and ^{238}U; C_8^0 stands for the atom percentage of ^{238}U in pre-irradiation fuel.

2.1 Relativeship of Nuclei in Before and After Irradiation Fuel

$$dN_4 = -(\sigma_{4t} N_4 \phi_t + \sigma_{4f} N_4 \phi_f)\, dt$$
$$= -(\sigma_{4t}\phi_t + \sigma_{4f}\phi_f)\, N_4 dt \qquad (2.1.1)$$

Let $P = \phi_f/\phi_t$ (assume constant for fast to thermal neutron flux). The equation (2.1.1) is reformed as follows

$$dN_4 = -(\sigma_{4t} + P\sigma_{4f}) N_4 \phi_t\, dt \qquad (2.1.2)$$

Similarly, dN_5 is obtained as follows

$$dN_5 = -(\sigma_{5t} + P\sigma_{5f})\,(N_5 + dN_4)\phi_t dt \qquad (2.1.3)$$

for calculating the burnup value, the contribution of dN_4 is negligeble then

$$dN_5 = -(\sigma_{5c} + \gamma\sigma_{5f}) N_5 \phi_t dt \qquad (2.1.4)$$

Similarly

$$dN_8 = -(\sigma_{8c} + \gamma\sigma_{8f}) N_8 \phi_t \, dt \qquad (2.1.5)$$

Let

$$\sigma_4 = \sigma_{4c} + \gamma\sigma_{4f} \qquad (2.1.6)$$

$$\sigma_5 = \sigma_{5c} + \gamma\sigma_{5f} \qquad (2.1.7)$$

$$\sigma_8 = \sigma_{8c} + \gamma\sigma_{8f} \qquad (2.1.8)$$

divide the equation (2.1.2)by (2.1.4).We can obtain

$$\frac{dN_4}{dN_5} = \frac{\sigma_4}{\sigma_5} \frac{N_4}{N_5} \qquad (2.1.9)$$

its solution

$$\frac{N_4}{N_4^0} = \left(\frac{R^0_{4/5}}{R_{4/5}}\right)^{\sigma_4/(\sigma_5-\sigma_4)} \qquad (2.1.10)$$

The number of ^{234}U atoms reduced

$$N^0_4 - N_4 = N^0_4 [1 - (R^0_{4/5}/R_{4/5})^{\sigma_4/(\sigma_5-\sigma_4)}] \qquad (2.1.11)$$

divide the equation (2.1.4)by (2.1.5), we can obtain

$$\frac{dN_8}{dN_5} = \frac{\sigma_8}{\sigma_5} \frac{N_8}{N_5} \qquad (2.1.12)$$

its solution

$$\frac{N_8}{N_8^0} = \left(\frac{R_{5/8}}{R^0_{5/8}}\right)^{\sigma_8/(\sigma_5-\sigma_8)} \qquad (2.1.13)$$

2.2 Deduce Formulae of Burnup Value

The burnup value is the number of fission atoms of uranium. Let F5 stands for the number of ^{235}U fission

$$F_5 = N_5^0 - (N_5 + N_6) + (N_4^0 - N_4) \qquad (2.2.1)$$

Where the $(N_4^0 - N_4)$ is neglected because its constribution to burnup value is less than 10^{-4} at the highest burnup level of fuel element.

The fractional furnup of ^{235}U is

$$\frac{F_5}{N^0} = (N_5^0 - N_5 - N_6)/N^0 \qquad (2.2.2)$$

let $Bu = F_5/N^0$, then

$$B_u = C_8^0 \left(R^0_{5/8} - R_{5/8} \cdot \frac{N_8}{N_8^0} - R_{6/8} \cdot \frac{N_8}{N_8^0}\right) \qquad (2.2.3)$$

If the equation(2.1.3) is substituted into the equation(2.2.3) on N_8/N_8^0 , we can obtain

$$B_u = \left\{ R^0_{5/8} - \left(R_{5/8} + R_{6/8} \right) \left(\frac{R_{5/8}}{R^0_{5/8}}\right)^k \right\} C_8^0 \qquad (2.2.4)$$

Where $k = \sigma_8/(\sigma_5 - \sigma_8)$ (2.2.5)

and $k = 0.461 - 0.40Bu$ (2.2.6)

Where k depends on fractional burnup(see fig.1), and it has been got from fractional burnup determined by Nd method.

If desired, Calculate gigawatt days per metric ton of initial U from

$$Q=Bu\ (9.6+0.03)\ 100 \qquad (2.2.7)$$

2.4 Calculate the Burnup Value

The burnup Value shown in Table 1 is Calculated by the set of equations (2.2.4)and (2.2.6).Its distribution along axial direction of a element is shown in Fig.2.

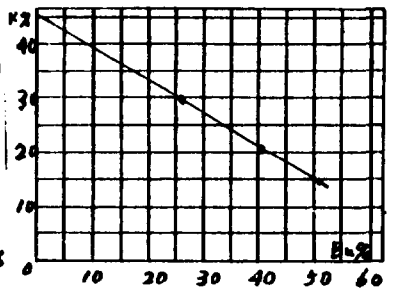

FIG.1. Relationship of k to burnup.

TABLE 1. BURNUP VALUES

NO	L.MM	H.I.%	ND %	CS-137%
1	150	25.95	25.92	24.24
2	350	42.90	—	41.31
3	500	48.54	—	—
4	630	50.78	50.20	48.27
5	720	48.27	—	45.02
6	800	44.44	45.46	44.56
7	900	33.84	—	—

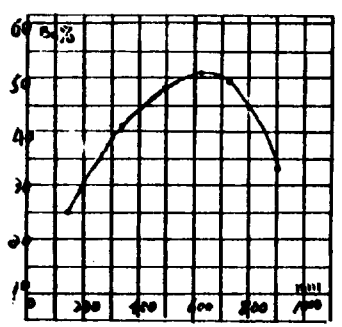

FIG.2. Burnup distribution along axial direction of an element.

3. CONCLUSION AND DISCUSSION

3.1 Measurement Precision

The isotope abundance of uranium is measured by surface source of Mass Spectrometer VARIAN MAT CH5, its values is shown in Table 2. The relative standard deviation of ^{235}U-to-^{238}U is 0.1%, and R.S.D. of burnup value is 0.5%(see Table 3).

TABLE 2. RATIO OF URANIUM

NO	R4/5	R6/5	R5/8
0	.01015	----	10.267
1	.01404	.0659	7.656
2	.01703	.1397	5.689
3	.01779	.1873	5.009
4	.01910	.2146	4.687
5	.02010	.2015	4.875
6	.01642	.1592	5.424
7	.01527	.0945	6.745

THE 0# IS UNIRRADIATED

TABLE 3. ACCURACY OF MS MEASUREMENT

NO	R4/5	R6/5	R8/5	BURNUP %
1	.02178	.2231	.2231	51.35
2	.02189	.2234	.2187	50.59
3	.01995	.2250	.2205	50.95
4	.01974	.2243	.2207	50.99
5	.01910	.2269	.2219	51.22
6	.01931	.2247	.2203	51.26
MEAN	.02033	.2246	.2207	51.26
RSD	5.8%	.6%	.1%	.5%

* THIS IS DATA OF 3# SAMPLE

3.2 Evaluation of the formulae

The formulae have the advantages: one, the uncertainty of burnup value is 3%, this is better than that of recommended formulae in ASTME 244. second the burnup value only depends on the change of uranium isotopes, and measuremehts of the isotope abundance of plutonium and the ratio of U to Pu is needless. but the burnup value have included its effect.The equation $k=\sigma_8/(\sigma_5-\sigma_8)$ is not real cross-section ,and it is only a coefficient. When k value is from 0.24 to 0.36, the change of burnup value is 3%, so the relativeship in Fig.1 is correct sufficiently. Third, because the burnup value only depends on the change of uranium isotope abundances, measurement process and chemical treatment become simple, so that the cost of the experiments is reduced.

So we concluded the equations(2.2.4) and (2.2.6) are a good set of formulae for burnup determination.

I am grateful to Dr Mao-Lian Li and Yon- xi pu for valuable suggestions, and to De-Yang Huan, Li-hua Ye, Jin-Yi Fen and Yu-liang shao for burnup experiments.

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32 DETERMINATION OF RADIAL GADOLINIUM BURNUP IN LWR FUEL BY SECONDARY ION MASS SPECTROMETRY (SIMS)

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Abstract

The "International Experimental Programme on Gadolinia Fuel Evolution in PWR's (GAP)" is devoted to the acquisition of experimental results in support of studies concerning the use of Gd_2O_3 as a burnable absorber in pressurized water reactors. One of the main tasks of the programme was to evaluate radial isotopic distributions of Gd in irradiated fuel samples. Mass spectrometry following radiochemical separations on samples taken by microdrilling is a laborious technique. Moreover, the lateral resolution is limited by the diamter of the drill. An alternative technique for the measurement of local isotopic compositions is Secondary Ion Mass Spectrometry (SIMS). This method is faster and provides greater lateral resolution. The major disadvantage of the SIMS technique is its limitation concerning quantitative analyses. It is therefore necessary to use average isotopic abundance values evaluated by dissolution and mass spectrometry for the normalization of the SIMS data. Alternatively, isotopic calibration standards with a chemical composition comparable to the fuel samples may be used. Samples of fuel with an initial ^{235}U enrichment of ~3.5% and Gd_2O_3 concentrations of ~3 and ~7 wt% have been analyzed. The burnup values lay between 2 and 6 GWd/tM. Several surface scans of the same sample showed a standard deviation of less than 10% (relative), provided that the residual isotopic abundances were higher than ~1%. Results of radial Gd burnup calculations with the aid of the LWRWIMS code were in good agreement with SIMS evaluated isotopic distributions.

1. INTRODUCTION

Utilisation of burnable absorbers in nuclear light water reactors (LWR's) is a method for implementing near term improvements such as 18 month fuel cycles and maximized average fuel burnup. Among the burnable absorbers, Gd_2O_3 is a leading candidate. It has been used in boiling water reactors (BWR's) for some years. When the two isotopes ^{155}Gd and ^{157}Gd with thermal neutron capture cross sections of 61000 and 254000 barn [1] are burnt out, the residual poisoning is negligible. As Gd_2O_3 can readily be mixed with UO_2, it can be implemented at the most appropriate locations in the fuel assemblies without negative effects on heat transfer or water/fuel ratio for example. Additionally, it does not adversely affect spent fuel storage and reprocessing.

Because uncertainties in the burnup calculation of Gd rods are still too large and since experience on the effect of Gd presence on high burnup behaviour is limited, it has not been possible to fully exploit its possibilities. The GAP programme[1] is devoted to the acquisition of experimental results required to complement the available data base for pressurized water reactors (PWR's).

Determination of burnup values on fuel samples by dissolution, chemical separation and mass spectrometric isotopic dilution analysis is a classical method used in destructive post-irradiation examination. It provides basic values for fuel rod performance and behaviour. To evaluate the radial isotopic distributions of Gd, the same method could be applied to samples produced by microdrilling. This technique is very time consuming and the lateral resolution is limited by the diameter of the drill. An alternative technique for the measurement of local isotopic compositions is Secondary Ion Mass Spectrometry (SIMS). This method is faster and provides greater lateral resolution. The major disadvantage of the SIMS technique is its limitation concerning quantitative analyses. It is therefore necessary to use average isotopic abundance values evaluated by dissolution and mass spectrometry for the normalization of the SIMS data.

Alternatively, isotopic calibration standards with a chemical composition comparable to the fuel samples may be used.

2. EXPERIMENTAL

In the hotlaboratory of the Paul-Scherrer-Institute (PSI) a modified A-DIDA-SIMS[2] apparatus has been installed for the analysis of highly radioactive samples [2]. The instrument is equipped with a quadrupole mass analyzer. An HP 9836 desk computer together with an HP 6942 multiprogrammer controls the spectrometer settings and is used for data processing.

For the analysis of polished cross sections of irradiated fuel, an O_2^+ primary ion beam of 12 keV energy was applied. The beam current used was about 50 nA with a spot size of around 10 µm. Surface scans were performed by moving the specimen across the primary beam in steps of 0.25-0.50 mm in the x- and y-directions, resulting in about 200-800 point areas analyzed per fuel sample. In order to smooth out inhomogeneities in isotopic composition caused by self-shielding in the gadolinia grains, the primary beam was scanned over an area of about 100x100 μm^2.

Evaluation of radial isotopic Gd distributions was effected using GdO^+ peaks, because the count rates for the oxide ions were about 2-3 times higher than for the metallic ions. The interference risk of fission product ions is also thus reduced.

Absolute secondary ion count rates can vary considerably, and some of the influencing factors are:

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[1] "International Experimental Programme on GAdolinia Fuel Evolution in PWR's (GAP)", programme conducted by BELGONUCLEAIRE (BN), Brussels, Belgium, and CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE/STUDIECENTRUM VOOR KERNENERGIE (CEN/SCK), Brussels, Belgium

[2] ATOMIKA Technische Physik GmbH, Postfach 450135, D-8000 Muenchen 45, FRG

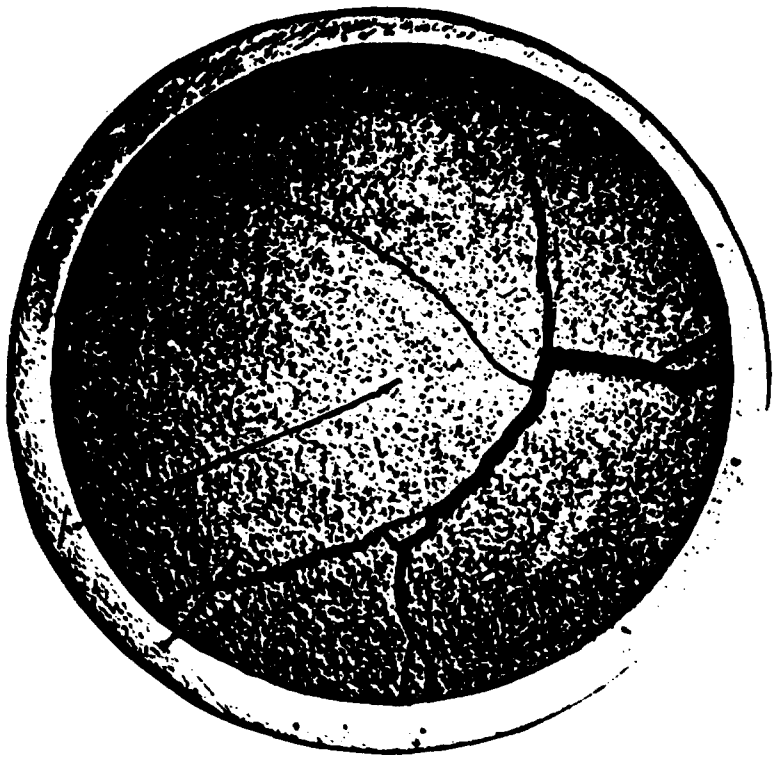

Figure 1 Photomacrograph of a typical cut and polished fuel cross section (sample A as indicated in Figure 3).

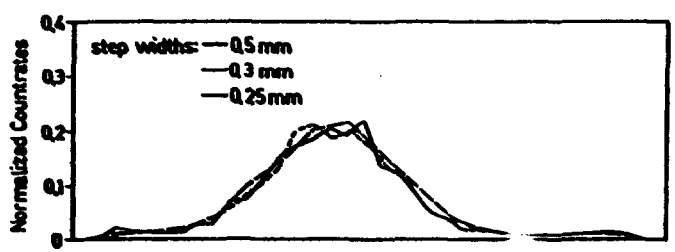

Figure 2 Comparison of three different scans for radial ^{155}Gd distribution. (Countrates normalized to ^{160}Gd countrates.)

- extraction field variations due to sample movement
- primary beam current variations
- variation of the primary beam incident angle and secondary ion take off angle due to sample cracks and pores
- true concentration variations within and between fuel and gadolinia grains
- and variation of secondary ion yield due to varying properties of the sample matrix

As a first step in data evaluation, all the raw gadolinium count rates were divided point for point by the count rate for ^{160}Gd. With a thermal neutron capture cross section of 0.77 barn, it can be assumed that the original ^{160}Gd content is not significantly changed during fuel irradiation at low burnup. This step eliminates most geometrical and chemical effects.

These normalized count rates were then transformed into local isotopic compositions by comparing the average SIMS values with radiochemically determined isotopic compositions of adjacent samples.

Fuel samples with an initial ^{235}U enrichment of about 3.5 % and Gd_2O_3 concentrations of about 3 and 7 wt% have been analyzed. The burnup values lay between 2 and 6 GWd/tM. A photomacrograph of a typical sample is shown in Figure 1.

Figure 2 shows ^{155}Gd count rates normalized to ^{160}Gd from three measurements of the sample shown in Figure 1. Datapoints along the same diameter were extracted from surface scans performed with different step widths. Shape and maximum value of all three measurements are similar. The difference between the lowest and highest value in the peak region of the curves, corresponding to values at the pin centre, is about 0.025 or less than 15 % of the approximate mean value of 0.2. Average values calculated for the whole specimen surface from the same three measurements show ranges of less than 10 % for the Gd isotopes 154, 155, 156, 158 and 160. ^{157}Gd is burnt out to about 0.1 % of the original content. This illustrates that SIMS measurements are reproducible, even if the sample surface is partly cracked and porous.

3. RESULTS AND DISCUSSION

In Figure 3 three-dimensional presentations of the radial ^{155}Gd distribution are shown together with the relative axial ^{137}Cs distribution evaluated by gamma-spectrometry [3]. The ^{137}Cs distribution is a measure of the local fuel burnup. The three-dimensional pictures show qualitatively, how the width and height of the ^{155}Gd distribution vary in function of the local fuel burnup.

For quantitative informations, this type of data presentation is not suitable. Quantitative information from the whole sample cross section can be displayed in contour plots. In Figures 4 and 5 the same data as in Figure 3 are used to show the ^{155}Gd distribution in the two samples A and D with the highest and lowest burnup values. This type of data presentation gives information about asymmetrical burnup and the width of single nuclide distribution. If informations on more than one nuclide are to be compared, it is easier to extract a part of the data, e.g. along a diameter, and to plot them in line graphs as in Figures 6 and 7. These figures show the distribution of the Gd isotopes 155-158 in samples A and D along the diameters indicated in Figures 4 and 5. The plots show how the two isotopes ^{155}Gd and ^{157}Gd are

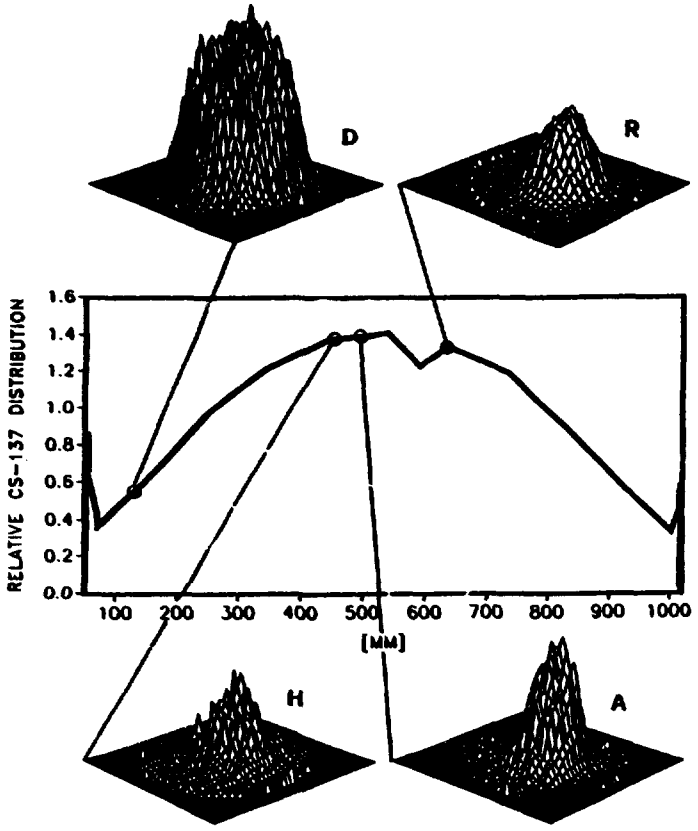

Figure 3 Axial ^{137}Cs distribution in a fuel pin [3] and radial ^{155}Gd distribution at different burnup levels.

converted into ^{156}Gd and ^{158}Gd respectively. The conversion is caused by thermal neutron capture and starts at the periphery of the pin. The lower isotopic contents in the central part of sample D compared to the original natural isotopic abundances of *14.80* % (^{155}Gd) and *15.65* % (^{157}Gd) are due to resonance reactions with fast neutrons.

4. COMPARISON WITH ISOTOPIC GADOLINIUM DISTRIBUTION CALCULATED BY LWRWIMS

In order to obtain values for isotopic composition, for comparison with theoretically calculated values, the SIMS data were further processed as follows:

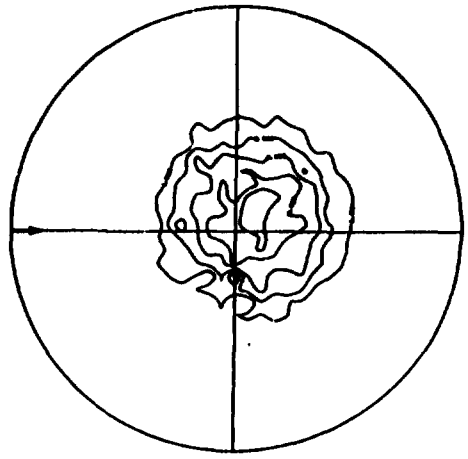

Figure 4 Contour plot of ^{155}Gd distribution in sample A. One contour interval corresponds to a difference in isotopic abundance of 1 %. The arrow indicates the direction of the radial plot in Figure 6.

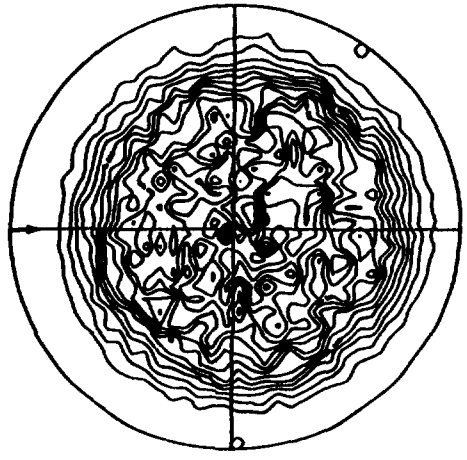

Figure 5 Contour plot of ^{155}Gd distribution in sample D. One contour interval corresponds to a difference in isotopic abundance of 1 %. The arrow indicates the direction of the radial plot in Figure 7.

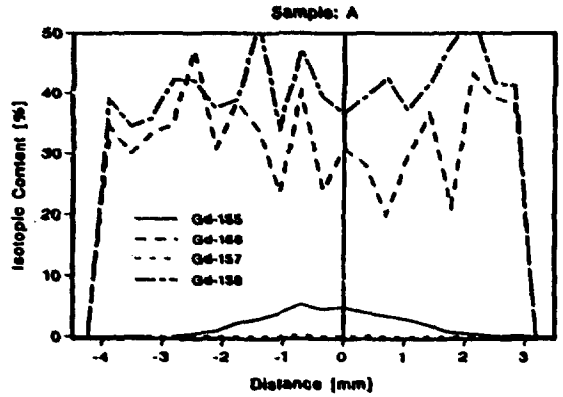

Figure 6 Radial distribution of Gd isotopes 155-158 in sample A along the diameter indicated in Figure 4.

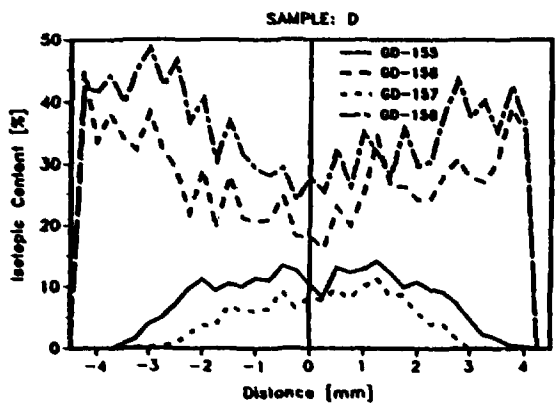

Figure 7 Radial distribution of Gd isotopes 155-158 in sample D along the diameter indicated in Figure 5.

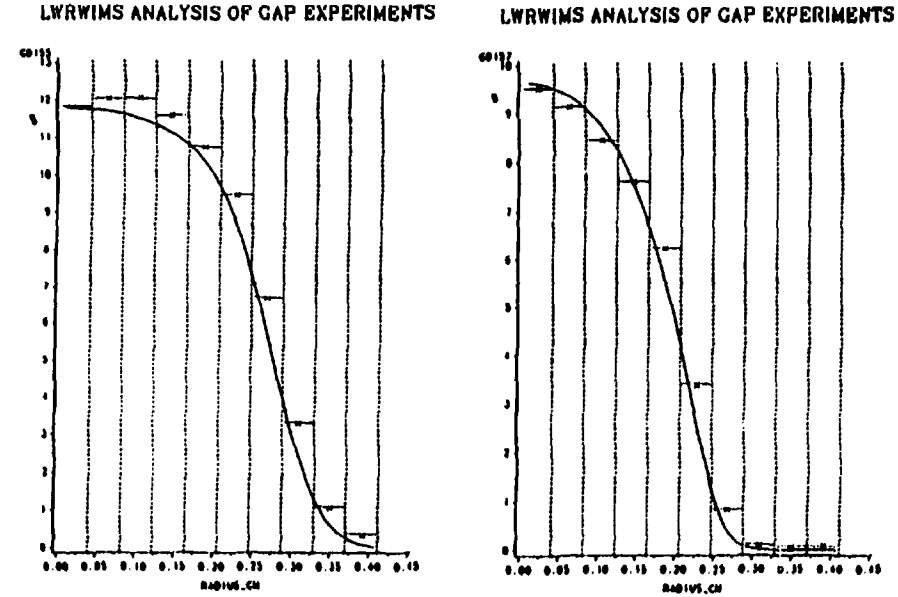

Figure 8 ^{155}Gd (left) and ^{157}Gd (right) variation with radius in sample D. Comparison of measured and calculated isotopic abundance. Local pin burnup: 3.03 GWd/tM.

- Based on the contour plot of the ^{155}Gd distribution the centre of the distribution was fixed. Usually this was not the geometrical pin centre.
- The original pellet cross section area was divided into 10 annuli. These annuli were superimposed on the isotpic distributions, the centre being the centre of the distribution.
- Average values of the ^{155}Gd and ^{157}Gd content were calculated for each annulus, taking into account all data points lying within the area concerned.

LWRWIMS [4] is a comprehensive scheme of computation for studying the reactor physics aspects and burnup behaviour of typical lattices occurring in modern Light Water Reactor designs.

The basic feature of the scheme is a coupled multi-cell spectrum calculation, which enables reduction of the energy group structure and the smearing of pin-cells containing fuel rods or poison pins. Diffusion theory and a variety of transport theory modules in Cartesian or explicit pin-type geometry are available for determining overall flux distributions in the reduced group structure. Special techniques are used to handle the positional effects of burnup and the buildup of fission product poisons. Approximate methods for dealing with highly absorbing regions in the context of diffusion theory, and for homogenising the interior of the BWR heterogeneous control cruciforms are available.

LWRWIMS ANALYSIS OF GAP EXPERIMENTS

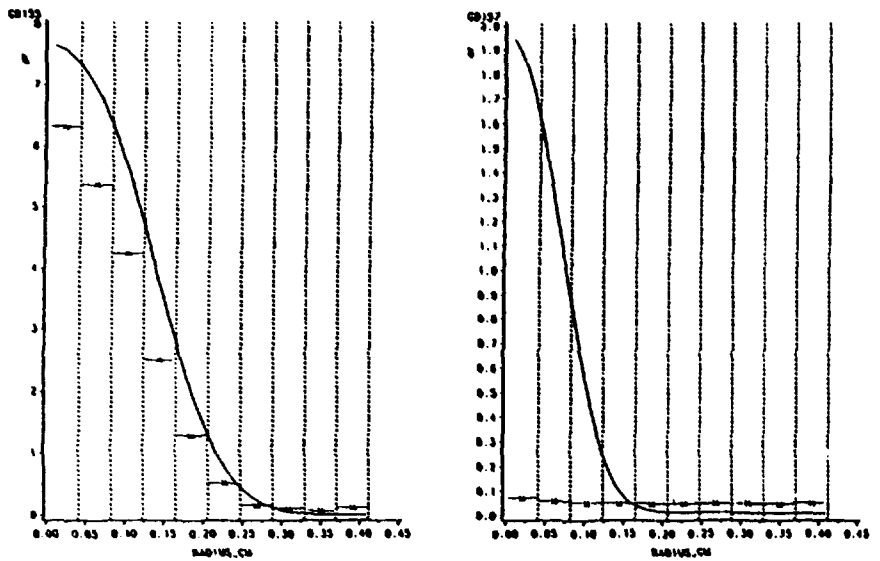

Figure 9 ^{155}Gd (left) and ^{157}Gd (right) variation with radius in sample H. Comparison of measured and calculated isotopic abundance. Local pin burnup: 5.78 GWd/tM.

This LWRWIMS code has been used to study the spatial variations of ^{155}Gd and ^{157}Gd for various samples examined under the GAP project and some results are given in Figures 8 and 9 for samples D and H as indicated in Figure 3. The LWRWIMS representation of the macrocell was as a 3x3 array with the gadolinium pin at the centre, other positions being filled by the fuel with a 3.2% ^{235}U enrichment. Reflective boundary conditions were applied on all four sides of the supercell. There is therefore no representation of th remaining sections of the reactor core in this model.

For the results discussed here the LWRWIMS burnup calculations for the Gadolinium pin were chosen to be equal to the radiochemically measured values. In this way the remaining fuel in the supercell serves only to give a sensible spectrum of neutrons into the gadolinium pin and we have discarded any information given in te GAP delivered neutronics package of the burnup of the surrounding fuel.

The results in general show reasonable agreement between the SIMS measurements and the LWRWIMS calculations for the ^{155}Gd results; however for the case of the ^{157}Gd distributions only for the low burnup sample (D) is there reasonable agreement.

5. CONCLUSIONS

SIMS is a suitable technique to evaluate relative radial isotopic Gd distribution in fuel pellets. Measurements on samples with Gd_2O_3 contents of 3 and 7 % and burnup values between 2 and 6 GWd/tM have been successfully carried out. This has lead to experimental data which can be compared with the results of theoretical calculations.

The SIMS technique is faster and provides greater lateral resolution than the microdrilling technique.

The major disadvantage of the SIMS technique is in limitations concerning quantitative analysis. In this work, SIMS data have been normalized by radiochemical and mass spectrometric analysis of adjacent samples.

If the pin burnup for LWRWIMS calculations is adjusted to the measured burnup, the calculated isotopic abundances agree with the SIMS determined values.

ACKNOWLEDGEMENTS

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Sample preparation was performed by the PSI hotcell group, in particular ALBERT ERNE.

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NON-DESTRUCTIVE ANALYSIS: METHODOLOGY AND EXPERIENCE

NON-DESTRUCTIVE BURNUP DETERMINATION: EXPERIMENTAL INVESTIGATION ON LOW BURNUP GADOLINIA FUEL

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Abstract

Neutronic codes benchmarking based on irradiated fuel rods needs a scale factor in order to compare calculated data with experimental results. A rod-to-rod non destructive, low cost burnup measurement with accuracies comparable with the accuracy of the ^{148}Nd radiochemical method provides the wanted check.

The "Burnothèque" method developed by the SCK/CEN physics group, consists in comparing the ^{137}Cs activity of the examined fuel rod with that of standard burnup samples. That comparison takes into account the decay correction during and after irradiation, the mass of investigated fuel material and shielding effects due to the eventual fuel density or cladding material differences.

The reliability and the limitation of the method will be discussed as well as recent data obtained in the framework of the "International Experimental programme on Gadolinia Fuel Evolution in PWR's (GAP)".

Seventeen absolute burnup results are compared to radiochemical and calculated values emphasizing the particular problems related to the low burnup gadolinia fuel rods.

---

1. INTRODUCTION

The examination of irradiated nuclear fuel by gamma-spectrometry is the most powerful and practical method for non destructive burnup determination. It is a comparative method and, therefore, it is subjected to major discrepancies if absolute burnups are wanted. Indeed, errors are accumulated when the experimental devices have to be calibrated in terms of absolute activities (standard source, self-shielding in source, crystal efficiency, statistics and self-shielding in fuel), and most of the measured fission products require irradiation history calculations, in order to obtain burnup values. Even fission product ratios, like $^{134}Cs/^{137}Cs$ do not completely escape the problems.

SCK/CEN and BELGONUCLEAIRE have been concerned for a long time with the qualification of design and follow-up calculation codes. In this field, critical experiments and irradiated fuel analyses are considered as powerful, and complementary, means to obtain accurate reference data.

As the presently available LWR calculation codes generally reach ~ 5 % precision in evaluating the burnup in standard irradiation conditions and ~ 10 % in more difficult situations, a high precision and low cost burnup determination is necessary to make calculation improvements possible.

Such accuracy comparable with the accuracy of the ^{148}Nd method is reached by the so-called "Burnothèque" method [1] using several reference standards, calibrated by different laboratories, by eliminating any calibration work of the gamma-spectrometry device (direct comparison of a source photopeak in both standard and investigated rod), and by taking account as precisely as possible of the irradiation history. For these reasons, burnup standard samples have been collected from destructive analyses of well followed rods or assemblies.

The present paper illustrates the reliability of this method on the basis of seventeen absolute burnup measurements carried out in the framework of the "International Experimental Programme on Gadolinia Fuel Evolution in PWR's (GAP)". The results are compared to radiochemical and calculated values emphasizing the particular problems related to the low burnup gadolinia fuel rods.

2. METHOD AND ACCURACY EVALUATION

The relationship typically used for the destructive analysis which gives the burnup using a fission product as burnup monitor is :

$$BU\ (MWd/tM) = 1.8563\ 10^{-24}\ \frac{E}{Y}\ .\ \frac{N}{P} \qquad (1)$$

N is the amount of atoms of the burnup monitor ;

P is the amount of fuel considered, in ton-metal ;

Y is the fission yield for the monitor ;

$\frac{N}{YP}$ is the amount of fissions per ton-metal ;

E is the energy emitted per fission.

To reduce the cost as well as the measurement delay, and also to allow intermediate examination before re-irradiation, a non destructive analysis using a radioactive fission product as burnup monitor is more suitable.

In that case, the equation (1) becomes :

$$BU = 1.8563\ 10^{-24}\ \frac{E}{\bar{Y}} \cdot \frac{C_\gamma \cdot f(\tau)}{P \cdot \epsilon_\gamma} \cdot f(E_\gamma, \text{geom.}) \cdot \frac{e^{\lambda t_c}}{k_\gamma \lambda} \cdot f(\text{power}, \lambda + \Phi \sigma_a, t_i) \qquad (2)$$

C_γ is the recorded counts for the gamma emitted by the fission product ;

$f(\tau)$ is the correction factor for counts lost by electronic deadtime and pile-up ;

ϵ_γ is the global efficiency of the measurement device for the chosen gamma-ray energy ;

$f(E_\gamma, \text{geom.})$ is the shielding and self-shielding correction for the examined fuel and its support ;

k_γ is the branching ratio ;

λ is the desintegration probability ;

$e^{\lambda t_c}$ is the correction for decay between the end of irradiation and the measurement ;

$f(\text{power}, \lambda + \Phi \sigma_a, t_i)$ is the correction for decay and possible absorption during irradiation.

As evident, the way to obtain the burnup absolute result from the spectrometry measurement through this procedure is quite long and each coefficient contributes directly to the global error (typically 5 to 6 %).

The "Burnothèque" method is based on a comparative measurement between the rod to be analysed and a set of reference samples. Equation (2) becomes :

$$\frac{BU^R}{BU^S} = \frac{\bar{E}^R}{\bar{E}^S} \cdot \frac{\bar{Y}^S}{\bar{Y}^R} \cdot \frac{C_\gamma^R}{C_\gamma^S} \cdot \frac{f^R(\tau)}{f^S(\tau)} \cdot \frac{P^S}{P^R} \cdot \frac{f^R(E_\gamma, \text{geom.})}{f^S(E_\gamma, \text{geom.})} \cdot \frac{e^{\lambda t_c^R}}{e^{\lambda t_c^S}} \cdot \frac{f^R(\text{power}, \lambda + \Phi \sigma_a, t_i)}{f^S(\text{power}, \lambda + \Phi \sigma_a, t_i)} \qquad (3)$$

Some sources of uncertainties disappeared (ϵ_γ) and most of the ratios include cancelling effects :

- if both rod and sample fuels have similar compositions :

$$\frac{\bar{E}^R}{\bar{E}^S} \sim 1 \quad \text{and} \quad \frac{\bar{Y}^S}{\bar{Y}^S} \sim 1$$

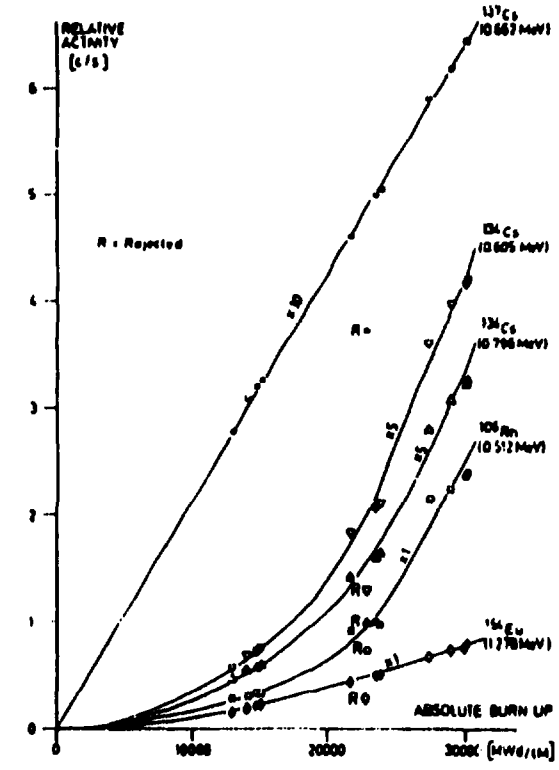


FIGURE 1 - MEASUREMENT OF THE REFERENCE SAMPLES

- if both rod and sample fuels have close geometries :

$$\frac{f^R(E_\gamma, \text{geom.})}{f^S(E_\gamma, \text{geom.})} \sim 1$$

- if the ^{137}Cs isotope is used ;

$f^R(\text{power}, \lambda + \Phi \sigma_a, t_i)$ and $f^S(\text{power}, \lambda + \Phi \sigma_a, t_i)$ are close to one

For a given fuel rod, the ^{137}Cs activity presents a linear relationship with burnup (Fig. 1), as a result of the characteristics of this isotope : similar fission yield for all the fuel isotope, large half-life (30 years) and low neutron absorption (0.1 b) and, however, its yield is nearly independent of the fissile isotope and the neutron spectrum.

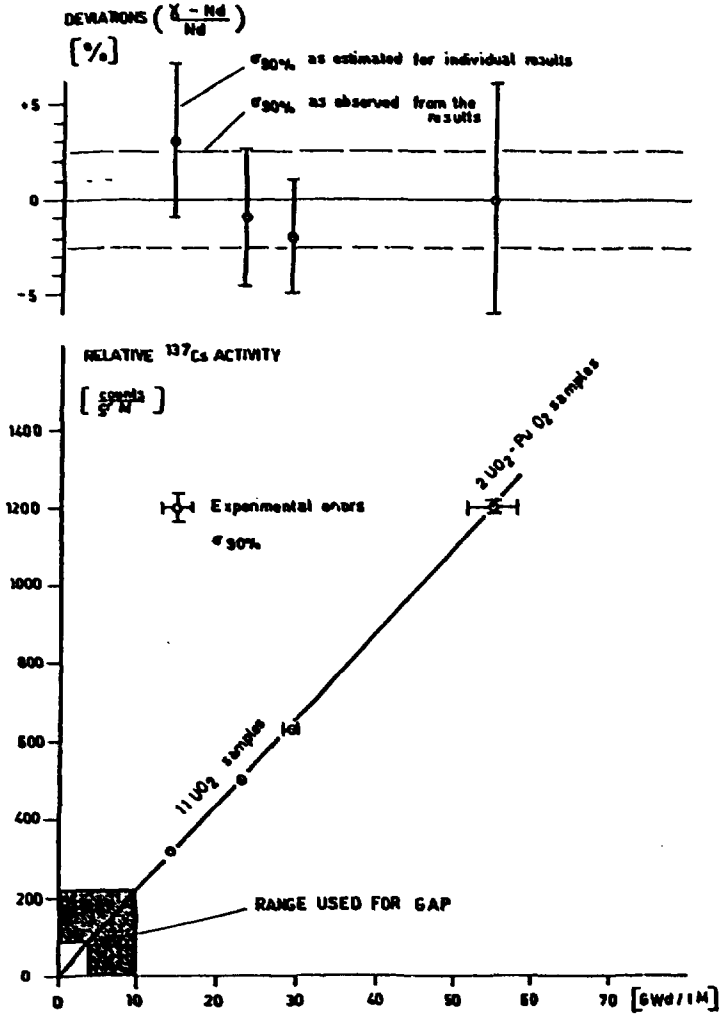


FIGURE 2 - BURNOTHEQUE CALIBRATION

The measurement procedure consists of measuring successively the ^{137}Cs activity of the examined fuel rods and of a series of standard samples of well know burnup.

The second step in obtaining the burnup is the transformation of the ^{137}Cs gamma activity into a quantity independent of rod and irradiation particularities expressed in counts/unit mass of fuel. This transformation takes account of the decay correction during and after irradiation, of the mass of the investigated fuel material and of the shielding effects due to the possible fuel density or cladding material differences. These corrections and modifications to the gamma activities finally provide ^{137}Cs activities that are related to the burnup by a linear function, by comparing them to the burnup values of the "Burnothèque" standards.

Fig. 2 shows eleven UO_2 standard samples together with $(U,Pu)O_2$ with the same fuel geometry ; note that the destructive burnup measurements were carried out by two different laboratories. They were used as reference for the GAP burnup measurements. An accuracy better than 3 % (2σ) is reached, even without irradiation history corrections.

The method is limited by possible cesium migration. The latter can, however, be detected, even by gross gamma-scanning, and generally occurs in extreme irradiation conditions. Nevertheless, good fuel rod average burnups may be measured by accumulating counts during both axial scanning and rotation.

3. THE GAP PROGRAMME

Utilization of "burnable poison" in light water reactors (LWR) is a requirement for implementing near term improvements such as 18 month cycles, increased discharge burnups and alternative in-core fuel reshuffling schemes [2]. Among the "burnable poisons", UO_2 - Gd_2O_3 is a favourite candidate. It has been used in boiling water reactors (BWR) since many years. It shows a negligible residual poisoning after the first cycle. As it can easily be mixed with UO_2 fuel, it can be implemented at the most appropriate locations in the assemblies without negative effects on heat transfer or water/fuel ratio for example. Finally, it does not effect spent fuel storage and reprocessing. As uncertainties in the burnup calculation of Gd rods are still too large, and since experience on the effect of Gd presence on high burnup behaviour is limited, it has not been possible to fully exploit the possibilities. The "International Experimental Programme on Gadolinia Fuel Evolution in PWR's (GAP)" is devoted to the acquisition of experimental results required to complement the available data base. Experimental data are obtained from UO_2 - Gd_2O_3 rods and the surrounding UO_2 rods irradiated during one cycle in the Belgian BR3 reactor. The first objective of this programme is to provide a data base useful for benchmarking neutronic codes.

These rods were located in four macrocells comprising 3 x 3 rod lattices introduced in standard assemblies positioned in four symmetrical positions of BR3/4C core. All rods have the outer diameter, fuel pellets as well as the cladding characteristic of a 17 x 17 PWR.

The four macrocells contained one fresh gadolinium fuel rod (^{235}U enriched) in the centre, surrounded by seven or eight fresh UO_2 fuel rods :

- macrocell 4C/1 : 3 w/o gadolinium rod surrounded by similar uranium rods;
- macrocell 4C/2 : as 4C/1, but with an empty corner uranium rod position, simulating a PWR guide-thimble ;
- macrocell 4C/3 : as 4C/1, but the central gadolinium rod contains 7 w/o Gd_2O_3 ;
- macrocell 4C/4 : as 4C/3, but with a water hole.

The gadolinia rods reached a peak pellet burnup between 5.5 MWd/tM (7 w/o Gd_2O_3) and 9.1 MWd/tM (3 w/o Gd_2O_3), whereas the UO_2 rods reached peak pellet burnup around 13 MWd/tM.

Seventeen fuel rods (4 gadolinia and 13 urania rods) were involved in a non destructive examination campaign performed at SCK/CEN (Mol). It included mainly gamma-spectrometry work to provide relative an absolute data on burnup and thermal neutron flux rate. On eleven of the UO_2 rods, burnup data were obtained for the rod average and the peak location. More data was obtained, at fifteen different axial levels, on six rods selected for destructive examinations. These destructive examinations were performed at the Paul-Sherrer-Institute (ex-EIR). It included rod punctures and fission gas analyses, twelve burnup determinations as well as isotopic analysis of U, Pu and Gd, and seven determinations of the radial isotopic Gd distributions by Secondary Ion Mass Spectrometry (SIMS) [3].

In addition to the postirradiation examination data, results of neutronic calculations of these rods (including mainly power, flux and burnup axial distributions as a function of irradiation time) were provided in such a way that the set of data can be directly used for calibration of the depletion codes on Gd fuel rods of the Participants to the Programme.

This Programme managed by BELGONUCLEAIRE was launched in 1983 and will be completed in 1989 (including an extension to the above mentioned work).

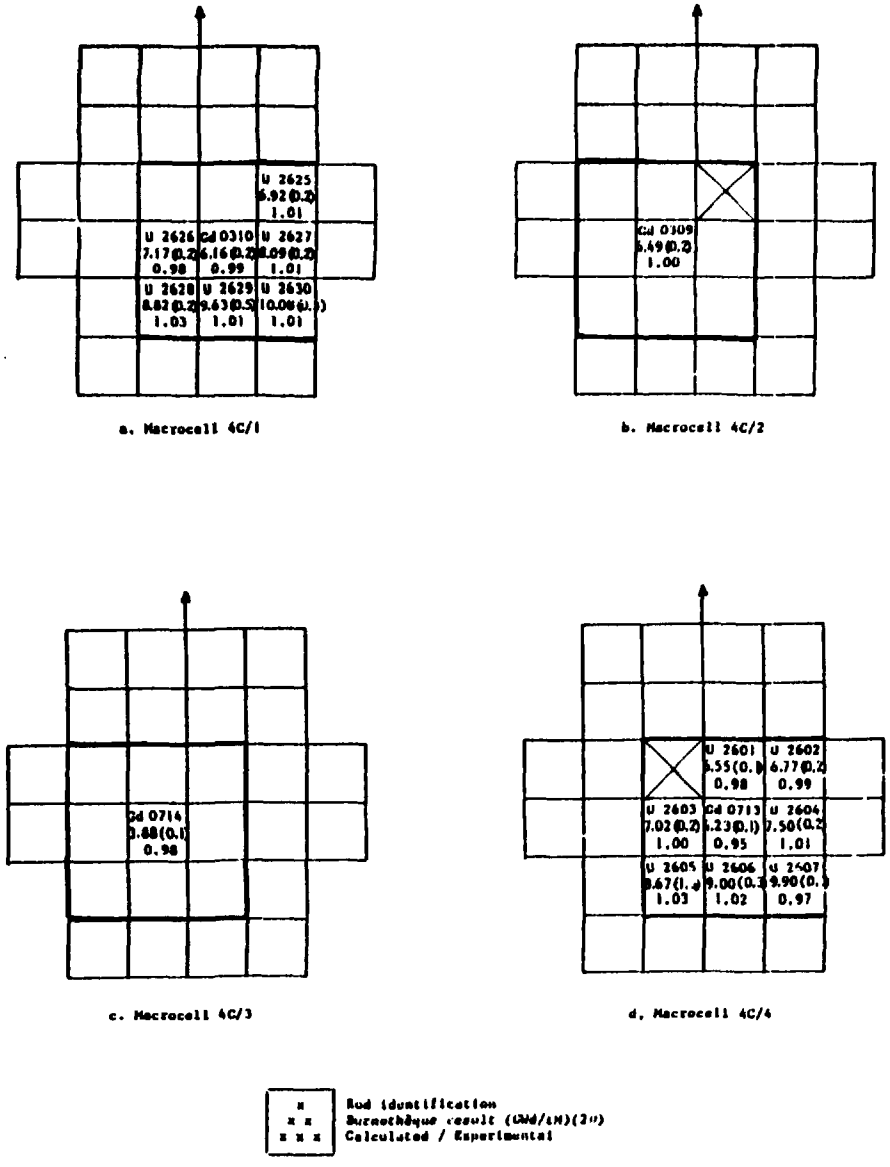
4. RESULTS

The rod average burnup has been determined for the four gadolinia rods and the thirteen urania rods by the "Burnothèque" method.

Two ^{137}Cs gamma-scanning at least were carried out over the whole rod length, whereas five measurements were made for each "Burnothèque" standard sample. With regard to the usual corrections to be applied, problems related to gadolinium rods were considered :

- Due to the lateral consumption of gadolinium, the distribution of the ^{137}Cs accumulated across the pellet is different from a standard fuel. Use was made of the ^{137}Cs radial distribution obtained by destructive analysis (SIMS and radial microgamma-scanning) to evaluate the self-shielding correction for the 7 w/o gadolinia rods. This correction $f^R(E_\gamma, \text{geom.})$ was computed as follows :

flat distribution	1.4279
SIMS measured distribution	1.4140
gamma-scanning distribution	1.4169



a. Macrocell 4C/1

b. Macrocell 4C/2

c. Macrocell 4C/3

d. Macrocell 4C/4

x — Rod identification
x x — Burnothèque result (MWd/tM)(2σ)
x x x — Calculated / Experimental

FIGURE 3 - THE FOUR GAP MACROCELLS - COMPARISON BETWEEN EXPERIMENTAL AND CALCULATED ROD AVERAGE BURNUPS

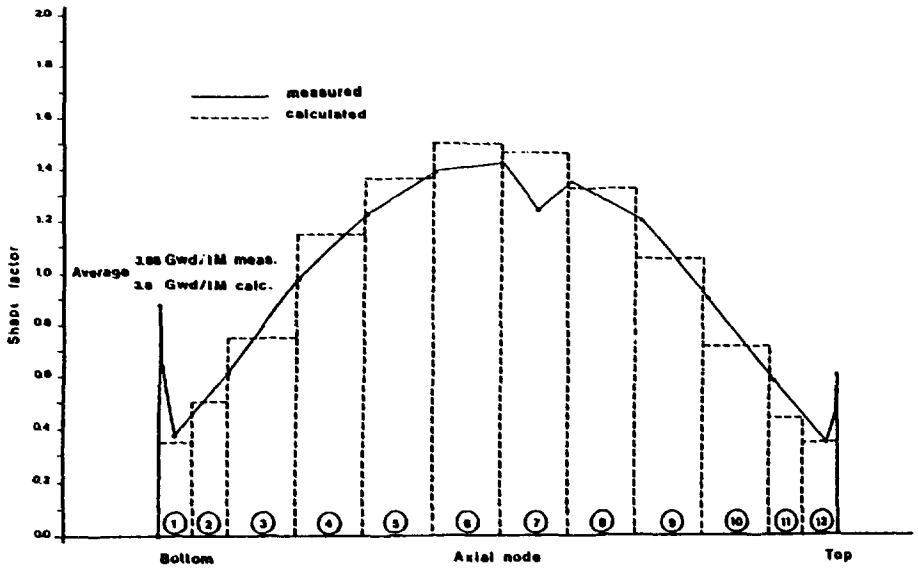


FIGURE 4 - COMPARAISON BETWEEN CALCULATED (12 AXIAL NODES) AND MEASURED (Cs137 SPECTROMETRY) AXIAL BURNUP DISTRIBUTIONS OF ROD GD0714

The maximum effect is - 1 % relatively to the commonly used flat distribution.

- Due to the high absorption of thermal neutrons, the fissile isotopes contribute differently to the fission rate (high fission contributions of ^{238}U and ^{239}Pu leading to a somewhat different ^{137}Cs yield). The correction was evaluated at maximum $\pm$ (1 $\pm$ 1) % ; it was not applied, but an extra $\pm$ 1 % error has been considered.

- Due to the progressive consumption of gadolinium, the pin power rate variation versus time is very different from the reactor power. Nevertheless, the maximum effect of the influence on the irradiation history correction was estimated less than 0.1 %.

The results (rod average burnups) are gathered in Fig. 3 ; they are compared to the values given by the neutronic calculations. For six rods, an experimental peak burnup value as well as the burnup axial distribution were obtained from the ^{137}Cs spectrometry data. Fig. 4 compares the calculated and the measured burnup axial profiles for one 7 w/o Gd_2O_3 rod. The measured burnup values were given with an accuracy of $\pm$ 3 % (2σ), except one result at $\pm$ 5 % (2σ).

The radiochemical ^{148}Nd burnup determination on twelve selected fuel samples cutted cut the four gadolinia rods has been performed by dissolution, chemical separation and mass spectrometric isotopic determination. The results are given in Table I. They are compared to local non destructive burnup values inferred from the "Burnothèque" rod average value and the axial distribution of the ^{137}Cs.

A relative precision of 1 % on the radiochemical burnup values could be considered as realistic based on an interlaboratory comparison, and on the comparison of the results obtained for three adjacent samples G714K, G714N and G714M.

Nevertheless, all the radiochemically determined values, with one exception, are higher than the values given by the ^{137}Cs spectrometry. The differences in most cases are higher than the given precision of the two methods. A first reason could be an assumed fractional fission yield for ^{148}Nd too low for the calculations. The yield depends upon the distribution of fission between the fissile isotopes present during irradiation ; it is approximately equal for fission in ^{235}U and ^{239}Pu, but increase by 29 % for fast fission of ^{238}U. The burnup evaluation of this report was made using thermal yield for ^{235}U. However, a contribution of about 12 - 13 % to the total burnup from fast neutron fission in ^{238}U is calculated for the low burnup 7 w/o gadolinia rods. This error which leads to a 2 - 3 % overestimation of the radiochemical burnup, must be added to the 1 %

TABLE I

Comparison of radiochemical burnup results with values based on axial ^{137}Cs activity profile and "Burnothèque"

Rod	Sample	Burnup (GWd/tM)		Difference (%)
		^{148}Nd	Spectrometry	
Gd0309	E	5.02	4.70	- 6
	K	9.00	8.80	- 2
Gd0310	D	4.83	4.43	- 8
Gd0713	E	2.75	2.60	- 5
	L	5.88	6.02	+ 2
	O	5.59	5.45	- 3
Gd0714	E	3.03	2.50	- 17
	K	5.78	5.51	- 5
	N	5.88	5.51	- 6
	M	5.78	5.51	- 5
	S	5.40	5.13	- 5
	U	2.37	2.20	- 7

uncertainty already mentioned. Moreover, one observes that the difference between the destructive and non-destructive burnups is higher for the low burnup samples. That could be explained by a lower relative accuracy of the ^{148}Nd method for very low burnup samples due to the higher relative effect of a possible neodynium contamination.

Another potential error source is the precision of the spectrometry measurements which give the local burnup values from the rod average value given by the "Burnothèque" : ± 1.5 % should be added to the uncertainty of the non destructive burnup determination. The uncertainty on the local burnup evaluation could have been avoided by a local evaluation by the "Burnothèque" technique.

Accuracy of the non-destructive burnup determination allows to discriminate small effects such as the influence of the water holes on the burnup of the gadolinia rods Gd0309 and Gd0713. This is not the case for the ^{148}Nd burnup determination which was performed mainly to determine the average gadolinium isotopic abundance used for the normalization of the SIMS data.

5. CONCLUSION

The use of a comparative and non destructive method such the "Burnothèque" is an easy, low cost and accurate way for checking the burnup of a nuclear fuel. An accuracy better than 3 % (2σ) is reached. This technique was successfully used in the framework of the GAP International programme, for the analysis of urania and gadolinia rods unloaded after one irradiation cycle. The obtained accuracies were sufficient for the benchmarking of neutronic codes.

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TECHNIQUES AND EQUIPMENT FOR DETERMINATION OF FUEL BURNUP IN WATER COOLED REACTORS

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Abstract

This report involves methods and equipment to determine the burnup of fuel for water-cooled reactors. The mass- and gamma-spectrometric data for determining the burnup of fuel in the VK-50 BWR are given as an example. The burnup distribution and "hardness" parameter of neutron spectrum with height and section of fuel assembly (FA) have been measured by nondestructive test (NDT) of gamma-scanning.

I. INTRODUCTION

The estimation of burnup for BWR fuel is very important problem the solution of which is of scientific and practical interest for reactor physics, radiation material science and spent fuel reprocessing. Among the modern existed nuclear physics and radiochemical methods to assess the burnup the most developed and extended that is based on nondestructive test of radioactive fuel fission products contents. The extensive use of this method is responsible for its simple realization, relative high accuracy, reliability and reproducibility of resut tested. Its positive feature is expressive and informative ability. In addition to burnup this method makes possible to use the distribution of energy release and fast neutron fluence (for claddings) with height and section of FA, change of "hardness" parameter for neutron spectrum, fuel mass-transfer and fission products

migration. The more energy-stressed regions of FA are identified and individual features of fuel rod irradiation are estimated.

2. TECHNIQUE AND EQUIPMENT FOR BURNUP DETERMINATILN BY GAMMA-SPECTROMETRIC METHOD

The burnup of fuel is assayed by comparing ^{137}Cs gamma-ray fluxes with energy E_j = 0.662 MeV emitted from fuel rod region under analysis and standard fuel specimen the burnup of which has been measured predominantly by the mass-spectrometric method. The burnup of fuel under analysis in terms of the gamma-scanning results is obtained by following formula:

$$\beta^{FR} = \beta^{St} \frac{f_5^{St} \cdot S_{PLP}^{FR} \cdot e^{\lambda t_{St}}}{f_5^{FR} \cdot S_{PLP}^{St} \cdot e^{\lambda t_{St}}} \cdot K$$

where β^{St} = the burnup of fuel of "standard" specimen

f_5^{St}, f_5^{FR} = ^{235}U content per unit length of initial fuel both for "standard" and analysed fuel rod, respectively

$S_{PLP}^{St} \cdot S_{PLP}^{FR}$ - ^{137}Cs photopeak area when measuring both "standard" specimen and investigated fuel, respectively

$e^{\lambda t_{St}}, e^{\lambda t_{FR}}$ - correction is due to ^{137}Cs decay in storing time of "standard" specimen and fuel investigated, respectively

K = correction allowed for "dead" time of measuring equipment

The use is made of semi-automated facility for fuel gamma-scanning to measure burnup the scheme of mechanical part of that is shown in Fig.I.For automated fuel researches the facility is provided a rotated storage of fuel rods.The fuel tested traverse vertically and passes a collimator slit. After measuring the fuel rod returned to the cell of storage and the following fuel rod is set for operative position when turning the storage. There are all 9 cells in the storage. In most cases one of them contains a standard fuel rod intended to calibrate the facility. Ge(Li) detector is usually

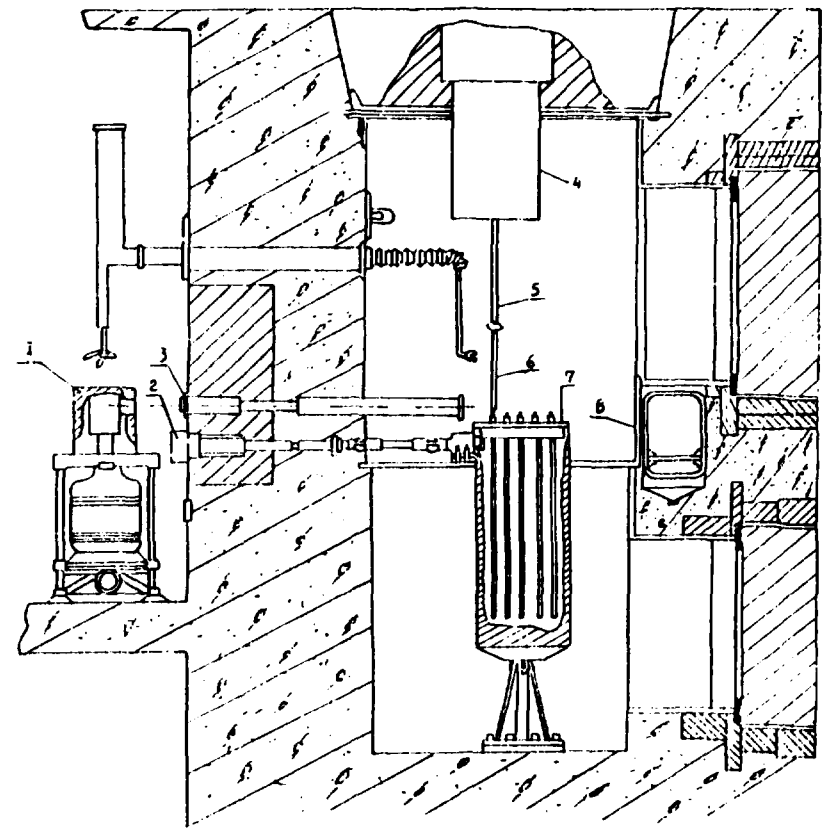

Fig.1. GAMMA-SCANNING FACILITY

I-detector, 2-drive of storage, 3-collimator, 4-drive, 5-mobile bar with capture, 6-fuel rod tested, 7-storage, 8-interchamber transporter

used. The control of the facility as well as data processing is automatically performed with using a microcomputer. A total error in determining the burnup by gamma-scanning method is achieved 10% within 95% probability confidence range.

3. TECHNIQUE OF MASS-SPECTROMETRIC MEASUREMENT

The fuel are sampled for test with mass-spectroscopy method and dissolved in 8N nitric acid in heating. The samples are prepared according to the technique /1/. Isotope content of heavy elements and fission products is determinated by using the mass-spectrometry of the above-described technique /2/. The relative content of uranium, plutonium and fission products is obtained by the isotope dilution method with using the three-complex marks /4/. The burnup is estimated by using both the heavy atoms (HAM) and fission products buildup methods /3/.Used by HAM the values of capture cross-section and fission ratios for ^{239}Pu and ^{241}Pu are computed with the POP program /6/. In estimating the burnup in terms of fission products the use is made of their yields values presented in /5, 6/. The mass-spectrometric uncertainty makes the main contribution to the error of the burnup determination. The uncertainty of d=Ga/Gf parameters for ^{239}Pu and ^{241}Pu is also important. The total uncertainty of the burnup measurement for fuel in terms of fission product buildup does not exceeded about 3 % for 95 % probability confidence range.

As an example both techniques application is considered when testing the VK-50 boiling reactor FA. In this case the common burnup affected by heavy isotopes fission is tested, sometimes ^{235}U and ^{238}U burnup ratio is measured individually.

4. Characteristics of tested VK-50 FA

The VK-50 FA tested is similar to the VVER-440 SA. It contains 162 fuel rods and 6 absorbing rods with 9.1 mm outer cladding diameter. Fuel rods are contained in triangular lattice with 13 mm pitch. The fuel are clad with zirconium alloy. The core reactor height is 200 mm. The UO_2 pellets of 10.2 g/cm^3 density are used as a fuel. The initial fuel enrichment with ^{235}U is about 3 %. The fuel pellet

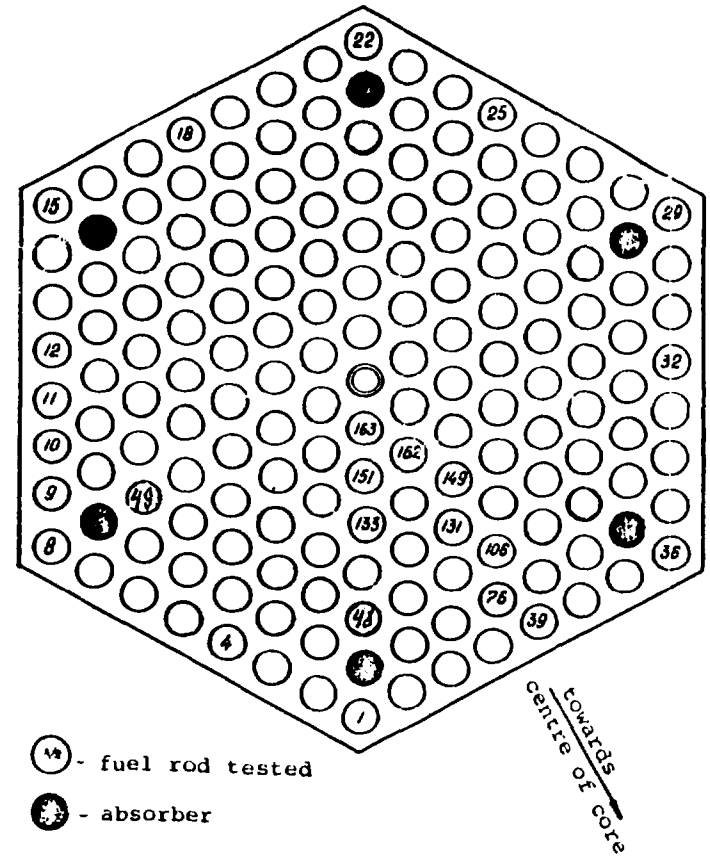

Fig. 2 Fuel arrangement in FA tested

diameter is 7.58 mm. There is a hole of 1.4 mm diameter in the centre of pellet.

FA is in reactor throughout 1820 days from which the reactor operated with nominal power during 1260 days. The tested FA was in operation with the following parameters:

- pressure = 6.9 MPa;
- coolant temperature = 284^{o}C;

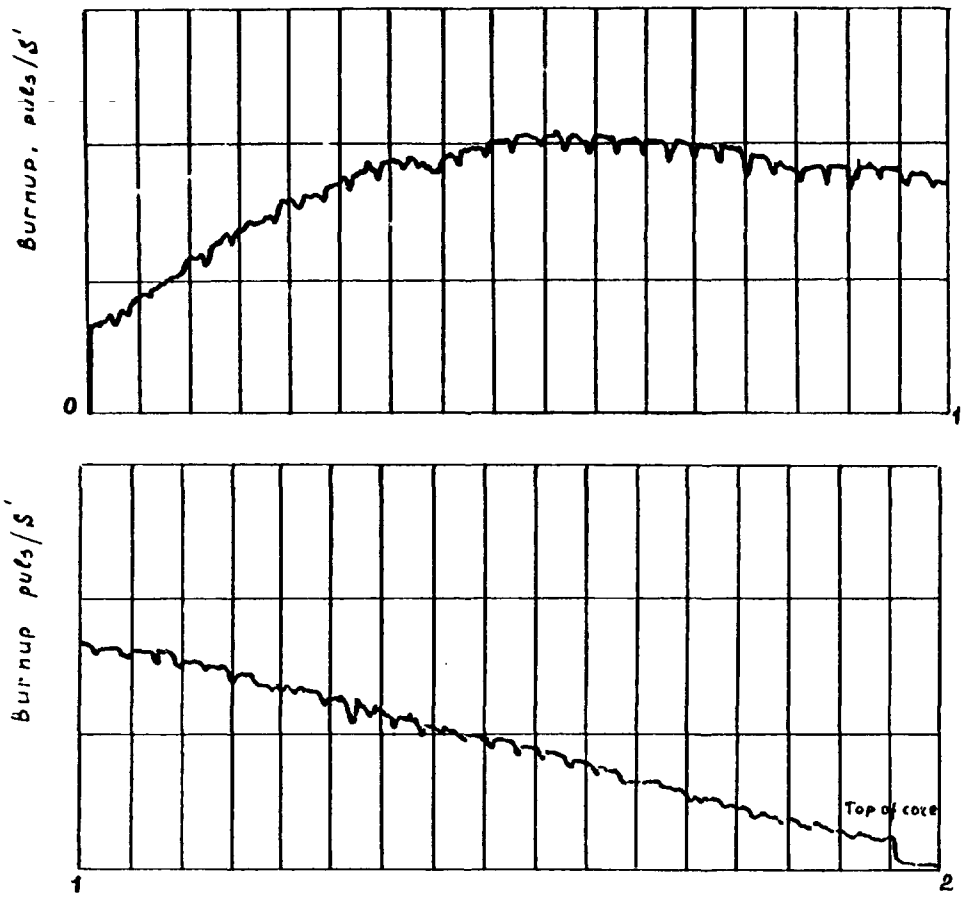

Fig. 3. Distribution of the integral gamma activity lengthwise rod 161; Number of measuring position

- inlet coolant rate = (0.9 ± 0.1) m/sec;
- subcooling to saturated state at entry to FA = 5°C;
- power = (1.85 ± 0.20) MW;
- non-uniformity energy release ratio with height = 1.45;
- volume coolant steam content at entry to FA = (0.60±0.05)

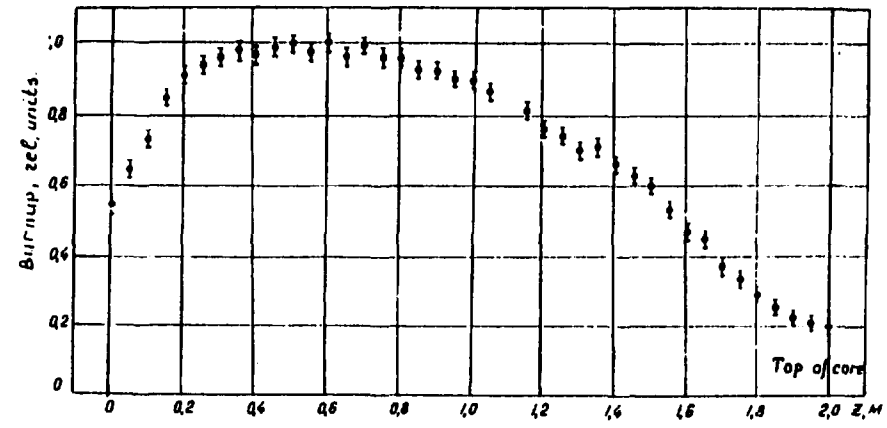

Fig. 4. Distribution of fuel burnup lengthwise rod 8

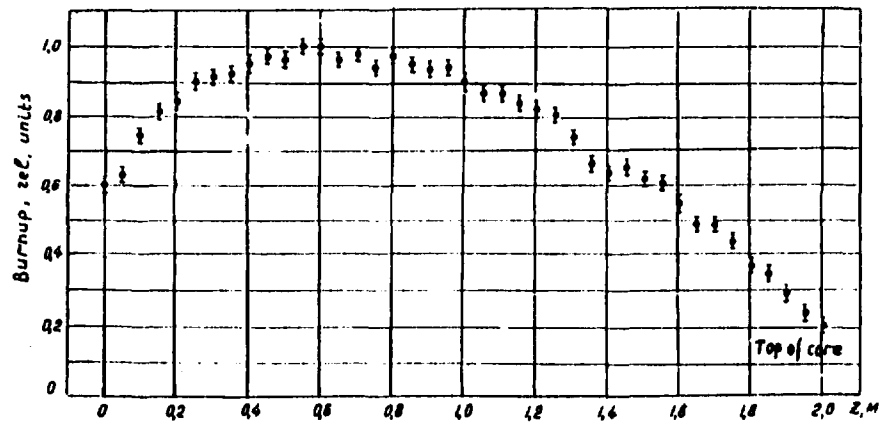

Fig. 5. Distribution of fuel burnup lengthwise rod 106

TABLE I. MAXIMUM BURNUP AND HEIGHT RATIO FOR NON-UNIFORMITY OF FUEL BURNUP

Rod number	Maximum burnup, kg ml/t U	Height ratio for non-uniformity of fuel burnup
I	36.0 ± 2.5	I.33
22	3I.I ± 2.2	I.35
49	27.8 ± I.9	I.33
76	32.8 ± 2.3	I.35
I06	28.2 ± 2.0	I.33
I63	25.6 ± I.8	I.36

The average specific volume power of the present FA was 30 KW+l. In operation FA there was at the same place of core. The calculated value of the average burnup of fuel was 20.5 kg/tU. The FA arrangement with indication of orientation within the core is shown in Fig.2.The fuel rods to have been subjected to gamma-spectrometric tests are marked by the digits.

5. RESULTS OF GAMMA-SPECTROMETRIC TESTS

To analyse the distribution of burnup for fuel with FA cross-section and height the fission product activity of fuel was measured. The distribution function of the total fission products gamma-activity was obtained at first wise-length of every fuel rod within the uninterrupted movement regime. As an example the plotted function to be obtained the above mentioned method is given in Fig.3 for fuel rod I63. Small activity dips in curve corresponds to the boundaries of pellet.

^{106}Ru, ^{134}Cs and ^{137}Cs fission products distribution with fuel height was measured using Ge(Li)-detector. ^{137}Cs distribution with height of fuel rod 8 and I06 is shown in Figs.4,5, the specimens of which were cut out subsequently for mass-spectrometric analysis. ^{137}Cs distribution with height is also produced in details for fuel rods I,22,49,76,I63 placed on different points of FA cross-section. The maximum values of burnup obtained by the gamma-spectromrtric method and the height ratios of non-uniformity for burnup of the above mentioned fuel rods are given in the Table I.

As the observed results indicate the height ratios of burn-up non-uniformity for fuel from different cores (circumferential, central and positioned near fuel rod) are in good agreement. The average value of the height ratio for non-uniformity of burnup is K_z= I.35.

The fuel burnup distribution of fuel rods located along FA perimeter is observed in Fig.6. The burnup distribution

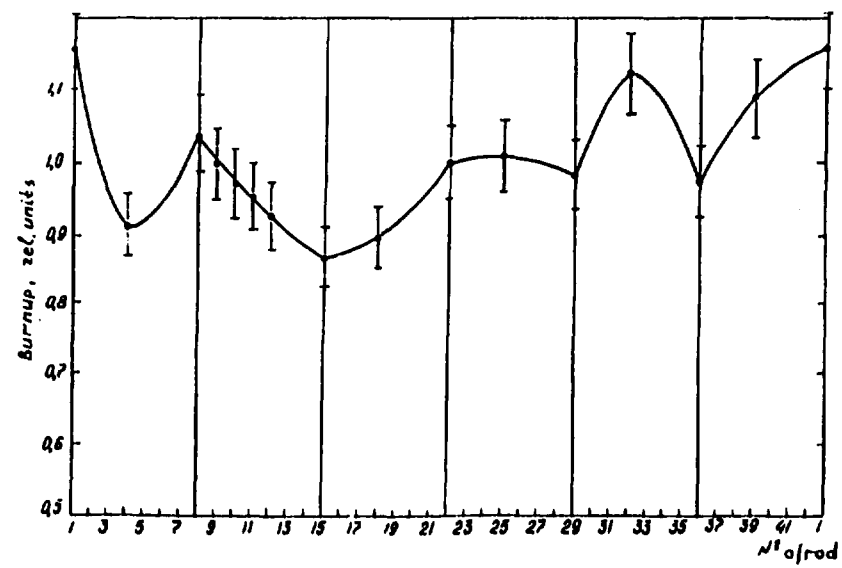

Fig. 6. Distribution of fuel burnup for the outer FA row

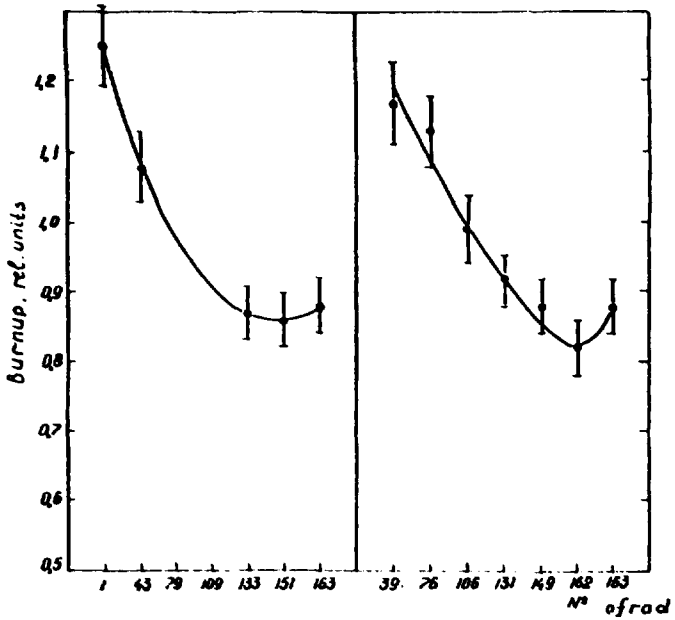

Fig. 7. Distribution of fuel burnup in cross-section of FA

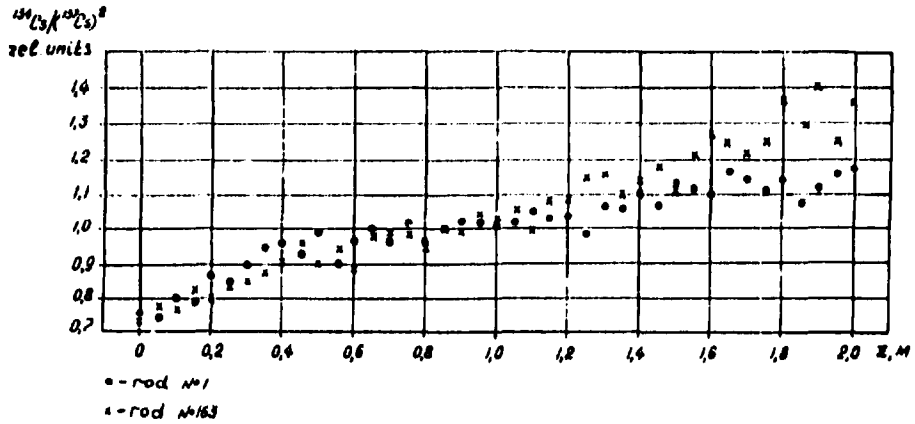

Fig. 8. Distribution of $^{134}Cs/(^{137}Cs)^2$ activity ratio with height of F

on FA cross-section over two directions is presented in the Fig.7. The specified distributions are produced for cross-section at 600 mm level from core base, i.e. the region of maximum activity. The radial ratio of non-uniformity for burnup on FA cross-section is K_r = I.25.

The distributions of ^{134}Cs and ^{137}Cs activity ratio with FA height and radius are shown in Figs. 8,9. These data indicate that the spectrum "hardness"goes up with increasing the vapour-content and removing from external boundary towards the centre of FA.

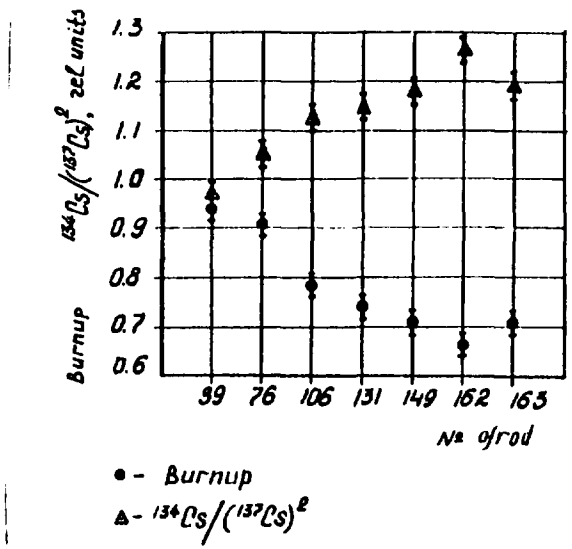

Fig. 9. Distribution of burnup and $^{134}Cs/(^{137}Cs)^2$ activity ratio in FA cross-section

TABLE 2. RESULTS OF MASS-SPECTROMETRIC BURNUP DETERMINATION IN VK-50 FUEL SPECIMENS (%)

Monitor	Number of specimen					
	6-1,0	I06-0,2	I06-0	I06-1,0	I06-1,4	I06-1,8
$^{133}Cs + ^{134}Cs$	27,5	23,0	25,2	23,3	I7,8	8,23
^{137}Cs	27,8	23,3	25,8	23,7	I7,8	8,34
^{140}Ce	27,4	22,2	25,3	-	I7,2	7,99
^{142}Ce	27,2	22,3	25,1	-	I7,2	7,97
$^{143}Nd + ^{144}Nd +$ $+ ^{144}Nd$	28,0	22,9	26,3	23,7	I7,9	8,24
$^{145}Nd + ^{146}Nd$	27,6	22,6	26,0	23,6	I7,6	8,04
Average values:						
-FP	27,6±0,3 27,1±1,2	22,7±0,5 22,4±0,9	25,6±0,6 26,7±1,1	23,6±0,4 23,8±1,1	I7,6±0,4 I7,9±0,8	8,I8±0,I7 8,72±0,88
-RIAR						
Content of FP						
^{235}U	I6,74±0,22	I4,78±0,23	I7,II±0,2I	I5,37±0,24	I2,28±0,23	7,0±0,8
^{238}U	I,8±0,5	I,5±0,4	I,7±0,4	I,5±0,4	I,I5±0,29	0,55±0,I4
$^{239}Pu + ^{24I}Pu$	8,60±I,0	6,I±0,8	7,8±I,0	6,9±I,0	4,5±0,7	I,I6±0,27

6. MASS-SPECTROMETRIC TEST RESULTS

The specimens of fuel rod I06 located in the centre of FA are sampled to measure. The specimens were cut out from the regions placed with height of fuel rod at a distance 200, 600, I000, I400, I800 mm from base of core. In addition to one specimen was cut out from the rod 8 at a distance I00 mm from the core base. The results of the mass-spectrometric burnup analysis of fuel specimens are summarized in Table 2.

The results of the mass-spectrometric burnup test differ from that of the gamma-scanning method by 8 % what does not exceed the above estimated errors.

7. SUMMARY

In RIAR at present to measure burnup of thewater cooled reactor fuel the techniques are used which base on NDT of accumulated fission products as well as those based on mass-spectrometric measure of fission products and trans-uranium elements concentration. The error of burnup measurement for first and second cases is about I0 % and 3 %, respectively.

The results of burnup measurement with gamma-spectrometric and mass-spectrometric methods agree within the probability confidence ranges. The correlative relationships between the ratio of burnup and ^{134}Cs and ^{137}Cs concentration relations are obtained.

At present the neutron methods of burnup determination are under development.

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PASSIVE NEUTRON VERIFICATION OF VVER-440 SPENT FUEL BURNUP

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Abstract

This paper presents a method based on passive neutron verification. The method is based upon calculation of the specific neutron emission rate as a function of burnup. It provides the basis for the construction of a burnup-meter.

To-day the most accurate data on the composition and burn-ups spent power water reactor fuel have been received using destructive methods of analysis. However, these methods have not an adequate rapidity that is required to solve some practical tasks. Non-destructive methods based on the gamma-radiation spectrometry and recording the passive neutron radiation of spent fuel assemblies (FA) make it possible to rapidly receive information on the characteristics of nuclear fuel under study. At the same time the require information can be also obtained from the declared data on the average specific burn-up of FA to be reprocessed, e.g., from the known different nuclide accumulation VS burn-up relations. The error in the Pu-239 and U-235 contents found from the known burn-up is several per cent / 1,2 /. However, some possible mistakes in the selection of FA to be reprocessed can result the discrepancy between the FA characteristics and the declared data. Therefore, the task arises to determine experimentally the average specific fuel burn-up to check the declared data on FA.

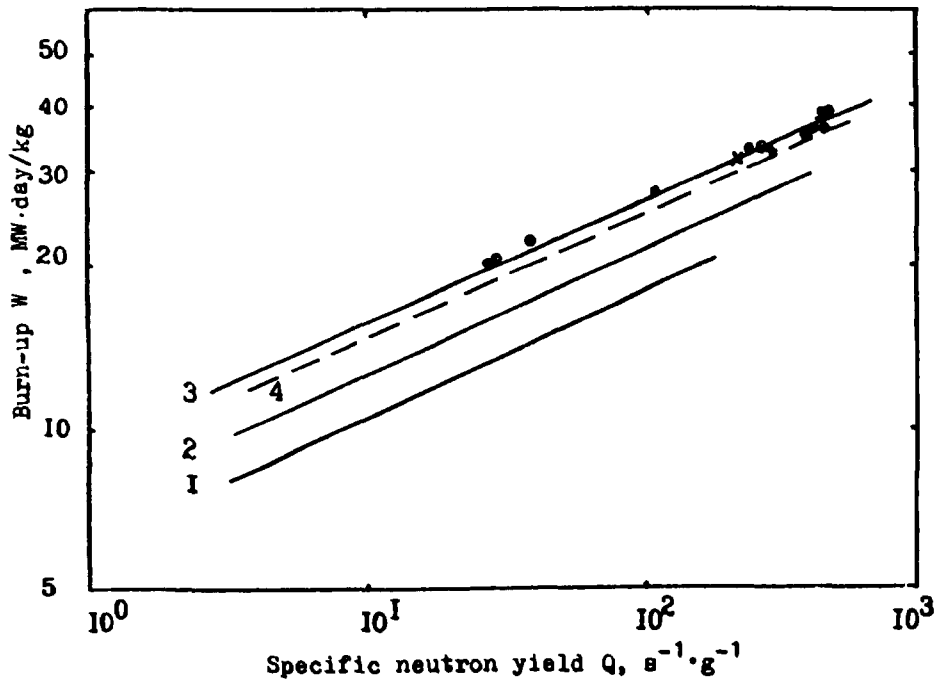

Fig. 1 VVER-440 fuel burn-up VS specific neutron yield
Initial ^{235}U enrichment of fuel:
1 - 1.6 % ; 2 - 2.4 % ; 3 - 3.6 % ; 4 - 3.6 %
1,2,3 - fuel pin specimens
4 - FA on the average

With this aim in view a method is considered based on the FA passive neutron measurements. In spent fuel neutrons are produced by spontaneous fission of transuranium nuclides and (α,n) reaction on the oxygen of uranium dioxide. In principle, the possibility of determining the spent fuel burn-up is thus related to the regularities in the accumulation of transuranium nuclides thus cause neutron radiation. These nuclides are Pu-238, Pu-239, Pu-240, Pu-242, Am-241, Cm-242, Cm-244. Fig.1 shows the calculated burn-up specific neutron yield relations. The relations were derive from the results of the calculations of the transuranium nuclides content of spent FA. For dependence 3 in fig.1 data are presented on the results of destructive methods of determining the transuranium nuclides content / 3,4,5 /. It can be seen from the comparison of the calculated and experimental data on 3.6% enriched fuel

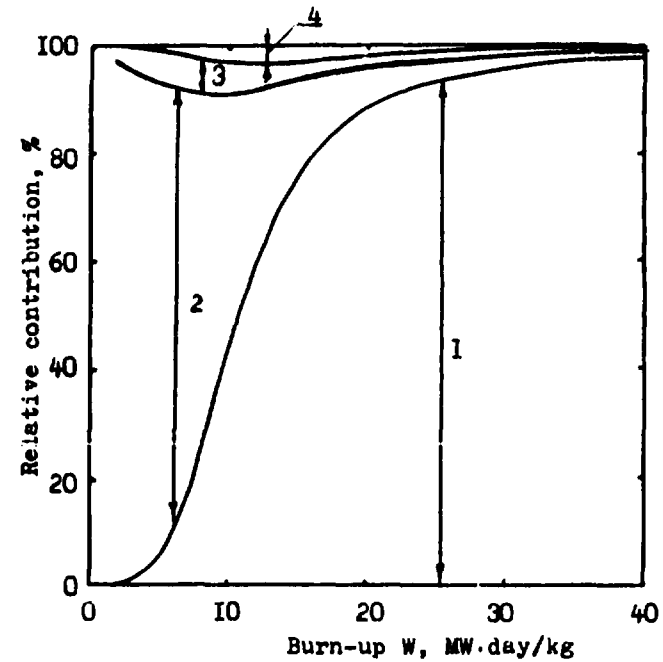

Fig. 2 Relative contribution to neutron radiation of FA VS fuel burn-up
1 - ^{244}Cm; 2 - Pu species sum ;
3 - ^{241}Am ; 4 - ^{242}Cm

Table 1

Formulae Relating specific Burn-up of VVER-440 FA of Specific Neutron Yield

Initial enrichment :	Formula	: Burn-up range
1,6	$W = 6,1\ Q^{0,23}$	$8 \leq W \leq 20$
2,4	$W = 7,3\ Q^{0,23}$	$10 \leq W \leq 40$
3,6	$W = 9,0\ Q^{0,23}$	$12 \leq W \leq 45$

that using the results of specific neutron yield measurements it is possible to determine the fuel burn-up with the error of ~10%. Fig.2 shows per cent contributions of nuclides responsible for neutron radiation to the neutron yield of spent three-year cooled VVER-440 3.6% U-235 enriched fuel VS burn-up. The results indicate that the major contribution to the specific neutron yield at the burn-up 20 MW·day/kg is made by Cm-244 as well as by plutonium species and Cm-244 at lower burn-ups. These conclusions apply to all the reactors of the VVER type and agree with the results of / 6,7 /. All the constants required to calculate neutron yields (half lives, neutron yields per fission, neutron yields in (α,n) reaction on UO_2 and oth.) were taken from / 8,9 /.

In the approximation of the dependencies the power law function was used:

$$W = aQ^b ,$$

where W - is burn-up (MW·day/kg),

Q - is specific neutron yield ($s^{-1}g^{-1}$),

a,b - are coefficients.

The coefficients a,b were fitted by the least square method. The fit assumed that W_i values of burn-up are known with the same relative error. Table 1 lists fitting power law functions that relate the burn-up to the specific neutron yield of spent VVER-440 fuel.

It should be noted that the derived relations and formulae are true for fuel pin samples or assemblies the burn-up non-uniformity

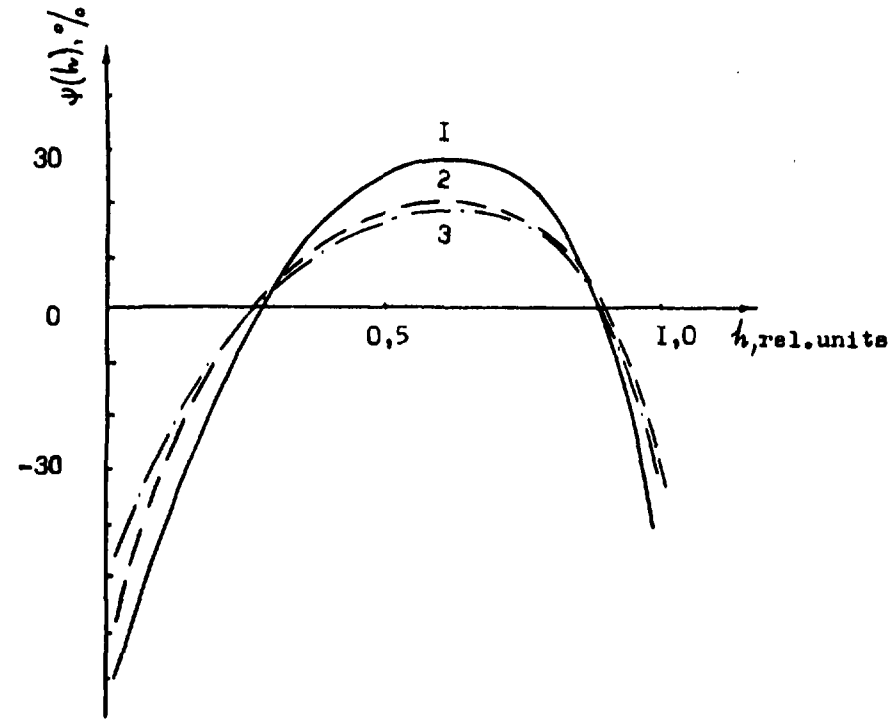

Fig. 3 Burn-up distribution over VVER-440 FA length.
Initial ^{235}U fuel enrichment :
1 - 1.6 % ; 2 - 2.4 % ; 3 - 3.6 %

coefficient of which is equal to unity. In this case different conditions of neutron radiation measurement can be realised, e.g., at one spot of FA or over the whole FA length. Consider the trends in the variation of the derived relations taking account of the real non-uniformity of burn-up in the determination of the average specific burn-up an assembly. Let's assume that the whole non-uniformity over an assembly is due the non-uniformity of burn-up over the length. Fig.3 shows the graphs at $\psi(h)$ - the relative deviation from the average specific burn-up VS the length of the VVER-440 FA. Having measured the rate of neutron counting which in the first approximation will be taken proportional to the specific neutron yield, at some height h_o, and thereby having determined $Q(h_o)$, find $W(Q(h_o))$ values from the relations of fig.1. Then, using the graph of the $\psi(h)$ function shown in fig.3, find how much $W(h_o)$ differs from the average burn-up of the fuel in the analysed FA and then define $\bar{W}$. It is possible to take account of the constant systematic shift between $\bar{W}$ and $W(h_o)$ due to the influence of the burn-up non-uniformity and determine directly the average specific burn-up of FA from the relation:

$$\bar{W}(Q(h_o)) = \frac{W(Q(h_o))}{1 + \psi(h_o)}$$

Then the non-uniformity type $\psi(h)$ must not vary from one assembly to another. To determine the average burn-up when the neutron radiation of the whole assembly is measured another relationship must be plotted that is different from $W(Q)$, as $\bar{W}(\bar{Q}) \neq W(\bar{Q})$, which can be seen from

$$\bar{W} = \int_0^1 \frac{W(h)}{1 + \psi(h)}\, dh \quad , \quad \bar{Q} = \int_0^1 Q(h)dh$$

The graph $\bar{W}(\bar{Q})$ for spent VVER-440 fuel initially enriched to 3.6% is shown in fig.1 (curve 4). The relationship $W(\bar{Q})$ used instead of $\bar{W}(\bar{Q})$ to determine the average specific burn-up of an assembly results in a systematic approximately 5% over estimation of the specific burn-up. For the assemblies of the initial enrichment of 1.6 and 2.4% the discrepancy will be more up to ~ 10%, since the non-uniformity of their burn-up is large. The burn-up distribution over the length was assumed constant for all the assemblies of the same enrichment. In reality the character of the non-uniformity can change from one assembly to another, which will introduce an additional random error into the average

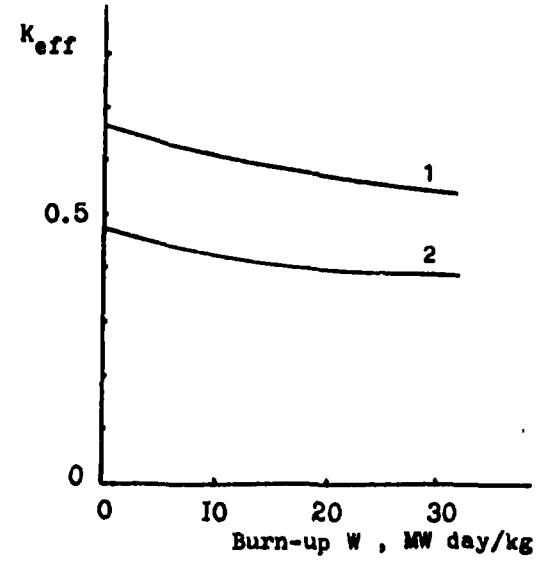

Fig. 4 K_{eff} VS burn-up (∞ reflector, H_2O)
1 - water containing FA , 2- FA without water

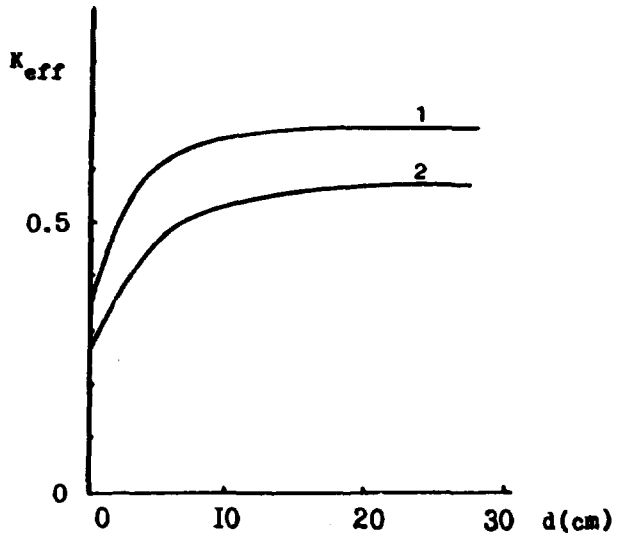

Fig. 5 K_{eff} VS reflector thickness d.
1 - fresh fuel, 2 - W= 30 MW·day/kg

specific burn-up. At the same time the burn-up is observed to be more uniform at the end of life-time / 10 /; when the fitting power law function is used to describe the average burn-up - average specific neutron yield relation the coefficient b is increased to approximately 0.25-0.27.

Another important point that needs discussion is the effect of neutron multiplication that is determined by the fuel burn-up and the conditions of neutron flux measurements (a water containing assembly or an assembly without water).The effective coefficients of neutron multiplication K_{eff} of spent VVER-440 fuel assemblies were calculated using the calculation code WIMS-D4 / 11 /. Actinide and fission product contents were found in preliminary calculations. The cooling time for FA was assumed equal to 3 years. Fig.4 shows the relationship between K_{eff} variations and the burn-up of the 3.6% enriched fuel. Curve 1 corresponds to the water containing assembly and curve 2 - to the assembly without water. The assembli were supposed to be surrounded with an infinite water reflector. It can be seen from fig.4 that K_{eff} is weakly dependent on the fuel burn-up and when the burn-up is above 20 MW·day/kg(the water containing assembly) and more than 10 MW·day/kg(the assembly without water) K_{eff} can be neglected. To choose the conditions for measuring neutron radiation the influence of the water reflector thickness on K_{eff} was calculated / 11 /. The derived dependences are shown in fig.5. It can be seen from the figure that K_{eff} almost doubles with an increase of the reflector thickness and at its thickness more than 15 cm it practically does not change. When assemblies in an air environment are measured the multiplication effect may be neglected.

The analysis of the data on the influence of the non- uniformity and multiplication effect on the fuel burn-up - specific neutron yield initial ratios that were derived by calculations or from the results of the investigation of individual specimens indicates that the relative character of these ratios is retained and offer having been refined a little they can be used to determine fuel burn-up. At the same time a 10-15% error in the determination of the average burn-up can be reached provided measurments for plotting dependence are conducted throughout the entire range of the controlled burn-ups. This stage should compris dissolution and subsequent analysis of composition of individual

FA to determine the average burn-up. The possible way of deriving the relationships between burn-ups and neutron counting rates is to correlate declared data on fuel burn-up and measured neutron counting rates, but the error in plotting this relationship will be higher through,e.g., accountancy mistakes.

Thus, the error in passive neutron measurments of spent FA can be ~ 10-15%, which is quite tolerable for the check up of the declared data on the burn-up of fuel to be reprocessed. In view of the rapidity of the method it can be employed prior to the process of the removal of the tails of fuel assemblies to be dissolved.

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NON-DESTRUCTIVE MEASUREMENTS OF WATER REACTOR FUEL AND EVALUATION OF MEASUREMENT TECHNIQUES FOR IMPROVED ASSAY ACCURACY

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Abstract

In the nuclear research center Karlsruhe, a continuing program for nondestructive assay of power reactor fuel elements is going on. The work, meanwhile, concentrates on implementation of improved technical designs into prototypical devices fulfilling stringent accuracy limits and being operational on a routine basis under actual plant conditions.

The assay is based on active and passive neutron detection. To demonstrate the method, several extensive series of measurements have been undertaken in storage ponds at reprocessing plants and power reactors. Uranium and MOX fuel elements have been assayed. It has been shown that the method applied can be used in reprocessing plants for determination of burn up and the type of fuel (Uranium or MOX). Due to the method's unique features it can also be used in reprocessing plants for criticality control and Pu input balance estimations.

A combination of active-passive neutron and gross gamma measurements fulfills all requirements for fuel assembles immersed in water. Without knowledge of any fuel assembly's data a complete identification of power reactor fuel is possible. The more simple passive neutron and gross γ-method allows a consistency check of the shipment data if and only if the fuel type is known.

Neutron measurements have the advantage of high transparency and easy detectability. The high neutron emission of spent fuel together with rather large amount of fissile material present yields a favorable signal-to-background ratios. The passive neutron measurement is made on the inherent neutron emission of spent fuel with Cm being the main neutron emitter. The active measurement accounts for the fissile material in spent fuel.

The demonstration program makes available calibration curves for criticality determination and representative burn up depend neutron emission data for Uranium and MOX fuel or PWR and BWR power reactor having a cooling times above 2 years. It was further demonstrated to what extent the measurement is influenced by specific reactor parameters such as irradiation history and steam void. Both quantities having only minor influence for the assay of PWR fuel assemblies but have some consequences for BWR fuel.

The knowledge acquired with the demonstration program was used to built a prototype incorporating all relevant features based on improved technical designs. Criteria for fulfilling stringent accuracy limits and for computerized control of these limits were established. After detailed laboratory tests, the prototype will undergo hot tests and routine measurement campaigns in storage ponds at reprocessing plants and power reactors.

1. INTRODUCTION

For several years the nuclear research center Karlsruhe has engaged in the development and implementation of NDA methods for identification spent LWR fuel assemblies /1/. Originally, a simple device was used in a measurement program to demonstrate the possibilities of the adopted NDA method and to establish a data base for NDA of spent fuel. This includes burn-up dependent neutron emission, neutron flux multiplication and the influence of irradiation history on this data.

Meanwhile, inclose cooperation with the nuclear industry the work concentrates on implementation of improved technical designs for a computerized prototype device fulfilling stringent accuracy limits and being operational on a routine basis.

Results of the demonstration program for identification of spent LWR fuel assemblies are presented. The prototype device is described together with results of the test program. Finally, the different nondestructive methods for identification of LWR fuel assemblies are discussed.

2. METHOD AND SIMPLE MEASUREMENT SYSTEM

Neutron measurements are used because of 1) high transparency of the fuel assembly 2) simple detectability, 3) high neutron emission of the spent fuel and 4) rather large amounts of fissile material present thus yielding favourable signal to background ratios.

Both active neutron interrogation and passive neutron measurements are done in order to obtain optimum neutronic characterization of spent fuel and to test achievable accuracy and reproducibility of the system.

The principle of the method is shown in Fig.1. The assay is done under water. The assembly is placed between a neutron source and neutron detectors. For the active assay only the detector opposite to the source is used. For the passive assay up to 4 detectors, one on each side of the assembly are used.

The passive neutron measurement is made on the inherent neutron emission of the spent fuel from spontaneous fission and (αn) reactions augmented by neutron multiplication due to the presence of fissile material. Active interrogation accounts for the fissile material in spent fuel. The neutrons of the external source induce fissions in the fissile isotopes. The neutron flux increases due to induced fissions, the neutron flux multiplication M_{th} is determined from the active and passive measurement.

The simple measurement system used in the demonstration program is shown in Fig.2. The neutron source and the detectors are mounted on a frame. The system is positioned on the fuel assembly's storage rack or is suspended from the wall of the storage pond as shown in Fig.2. The fuel assembly is introduced into the monitor by the storage pond handling device.

In total 92 fresh and spent Uranium and MOX fuel assemblies from different KWU built reactors were measured. Data of the fuel assemblies are listed in Table 1.

3. NEUTRON EMISSION AND BURNUP

The neutron emission of spent fuel mainly is due to the buildup of Curium isotopes in the course of irradiation. In spent fuel removed from the reactor, Cm-242 decays rapidly due to it's short halflife. After a cooling time of 1,5 years. Cm-244 remains the main neutron emitter in spent Uranium fuel.

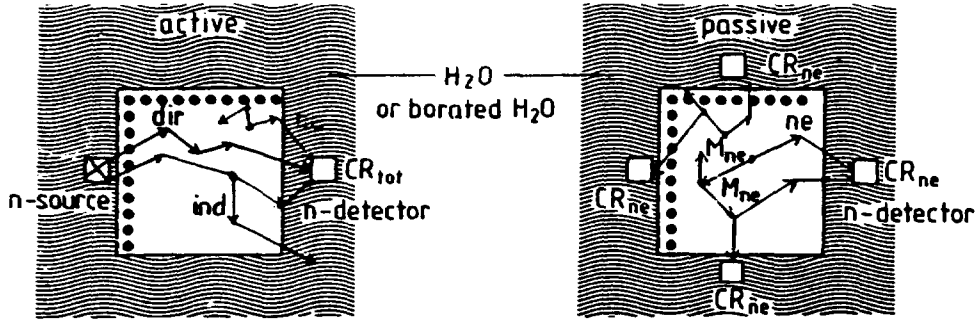

Fig. 1: Principle of NDA-method

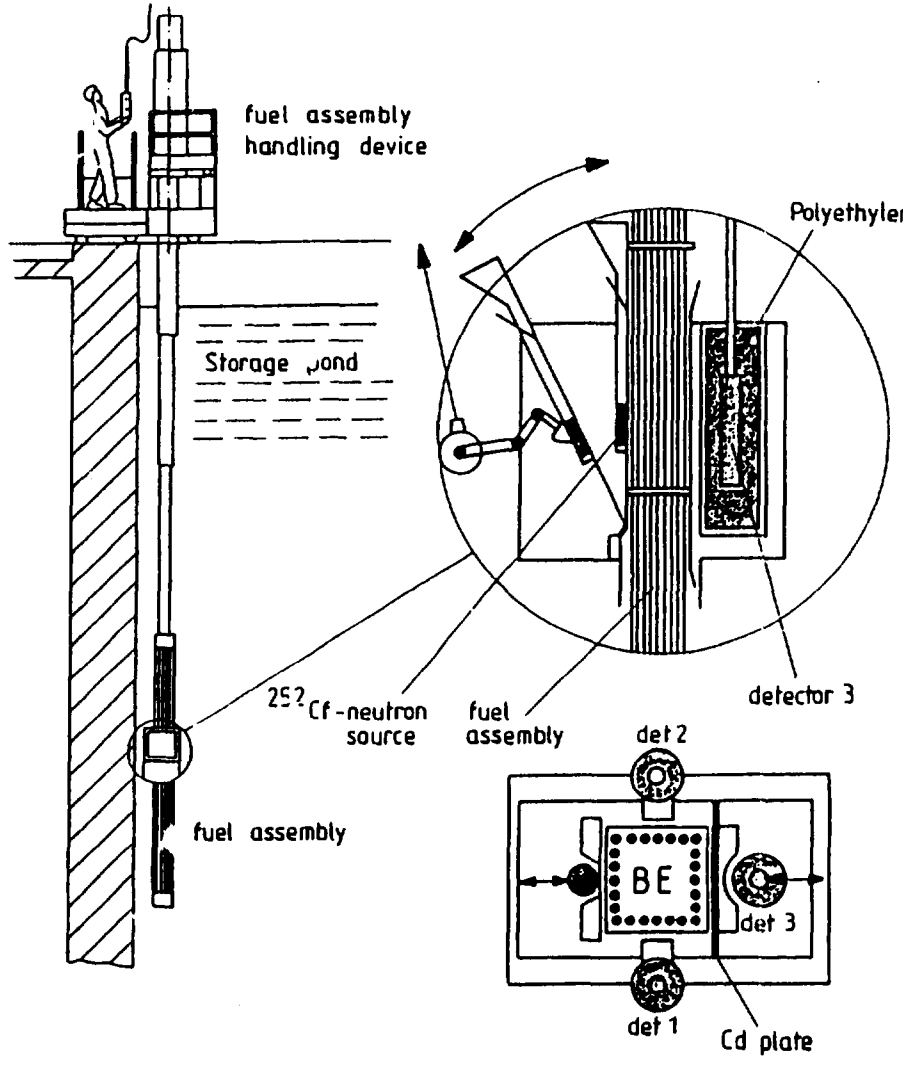

Fig. 2: Measurement system

measurement under water

detection of thermal neutrons

$$CR_{tot} = CR_{dir} + CR_{ne} + CR_{ind} \qquad CR_{ne} = \frac{1}{4} \sum_{i=1}^{4} CR_{ne}^{i}$$

thermal multiplication M_{th}:

$$M_{th} = \frac{CR_{tot} - CR_{ne}}{CR_{dir}} = 1 + \frac{CR_{ind}}{CR_{dir}} = f\,(k_{\infty}^{max})$$

total neutron emission of the spent fuel

$$CR_{ne} = A\, ne_{tot} \; ; \; ne_{tot} = f\,(IE, Bu, M_{ne})$$

$$M_{ne} = f\,(M_{th})$$

$$ne_p = \frac{ne_{tot}}{M_{ne}} \qquad \text{primary neutron emission}$$

Table 1: Data of the measured LWR fuel assemblies

reactor	measured fuel assemblies	range of burnup (GWd/tU)	initial enrichment (%)	lattice pitch (mm)
Obrigheim	25 Uranium	fresh-34,8	3,1	14x14
KWO	11 MOX	fresh-31,3	2,8	14,3
Biblis	3	13 - 17	2,16	16x16
	2	18	2,39	14,3
Stade	5	30,4-36,3	3,18	15x15
KKS	3	33,1-36,2	3,29	14,3
Neckarwestheim	3	21,9	1,9	15x15
GKN	5	33,3-34,3	3,2	14,3
Kahl	5	16,1-18,6	2,7	6x6
VAK				
Gundremmingen	15 Uranium	2,2-21,7	2,4	6x6
KRB	9 MOX	fresh-14,8	1,98	17,8
Würgassen	6	11,0-23,4	2,2	7x7
KWW				18.75

The neutron emission ne of spent fuel easily is detected by passive neutron measurement. For obtaining representative ne-values neutron detectors at least 2 off the 4 sides of the assembly have to be installed. γ-insensitive n-detectors are required. This is best accomplished using U-235 fission chambers.

The count rate CR_{ne} depends on the distance fuel assembly and detector as shown in Fig.3. There exists a count rate maximum at a distance of 2-3 cm. Positioning of n-detectors in this region results in minor changes in count rate caused by changes in fuel assembly position.

The neutron emission ne is determined from the detector countrate CR_{ne} according to

$$\overline{ne}_{tot} = A\, CR_{ne}$$

with A being a calibration constant and $\overline{ne}_{tot}$ the total neutron emission. $\overline{ne}_{tot}$ is given according to

$$\overline{ne}_{tot} = M_{hom}\, ne_p$$

with M_{hom} the multiplication of the primary nep due to the fissile material present in the fuel. nep is correlated with the burnup dependent buildup of the neutron emitting isotopes Cm-242, Pu-238, Pu-240 and Pu-242.

$\overline{ne}_{tot}$ has been determined for 92 LWR spent fuel assemblies. Due to axially varying power profiles in the reactor core the burnup and hence $\overline{ne}_{tot}$ are dependent on axial position. This is shown in Fig. 4a for PWR and in Fig.4b for BWR fuel assemblies. The axial profile for PWR approximately is symmetrical with respect to half height but is asymmetrical for BWR showing higher n-emission in the upper part (Fig. 4b). This is caused by spectral hardening due to increased steam fraction (void) of the coolant.

Fuel assembly averaged neutron emission values

$$\overline{ne}_{tot} = \frac{1}{21} \int_{\circ}^{1/2} ne_{tot}(z)\, dz$$

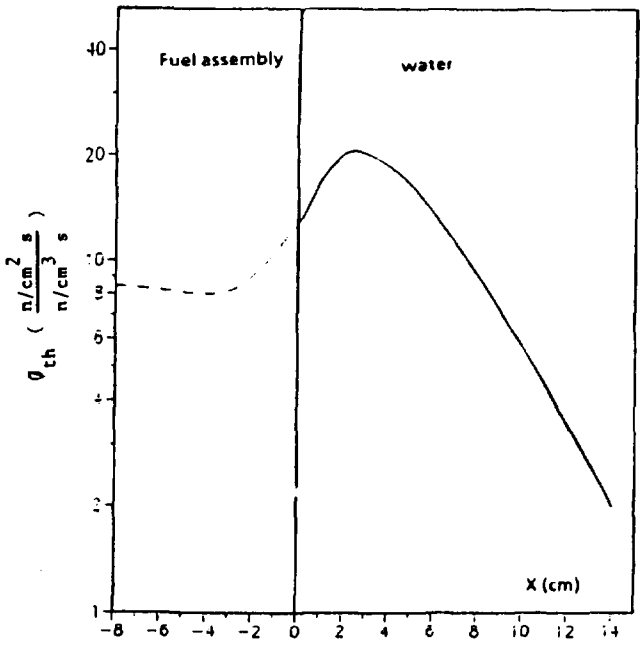

Fig. 3: Thermal neutron flux for neutron emission in fuel

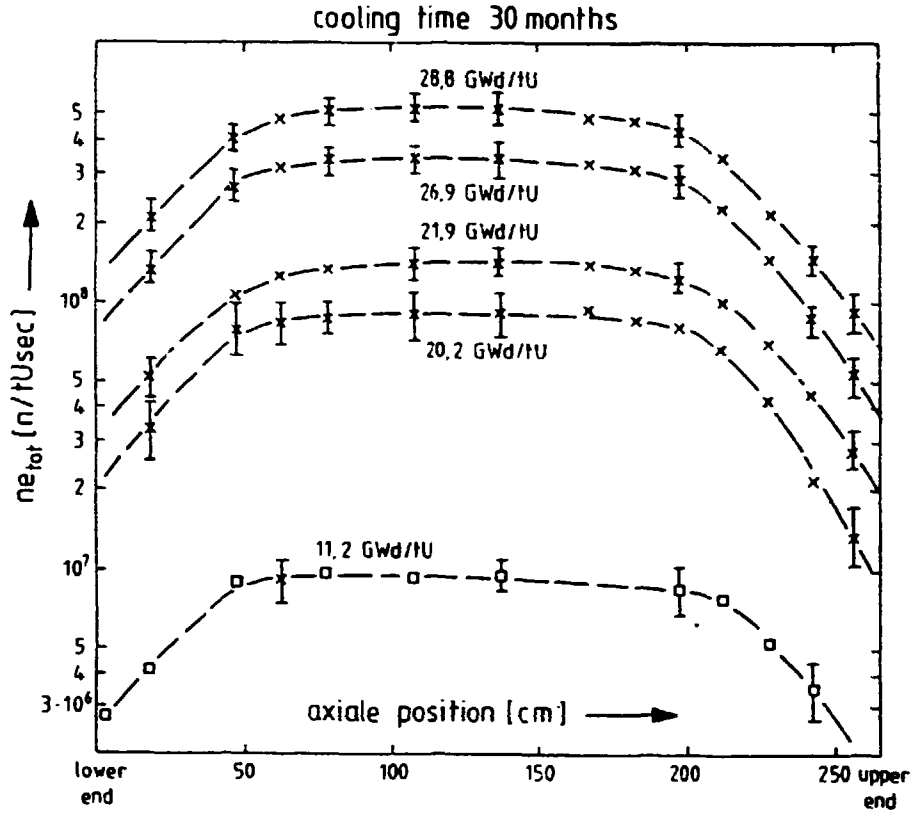

Fig. 4a: Total neutron emission ne_{tot} for PWR (KWO)
Uranium fuel assemblies
inital enrichment 3,19% U-235

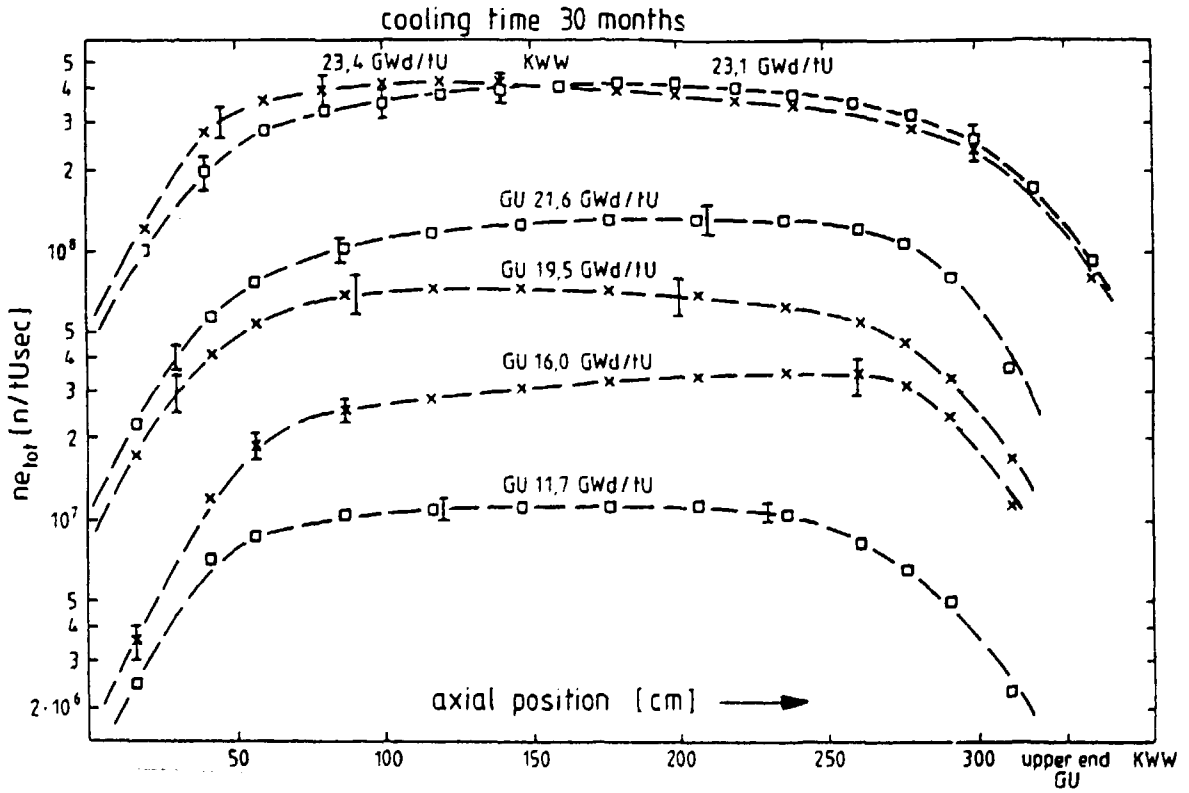

Fig. 4b: Total neutron emission ne_{tot} for BWR
Uranium fuel assemblies
GU: Grundmessungen, inital enrichment (IE) = 2,4% U-235, KWW:
Würgassen, IE = 2,2% U-235

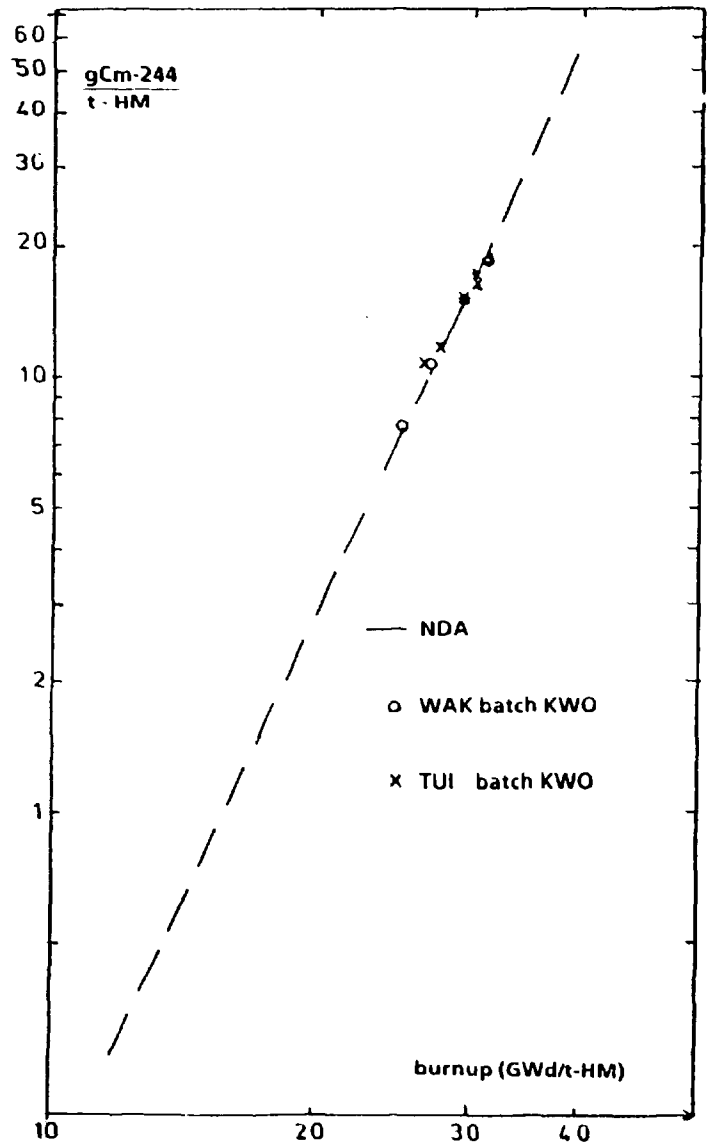

Fig. 5: Cm-244 production as a function of fuel burn up. Experiment NDA and DA results

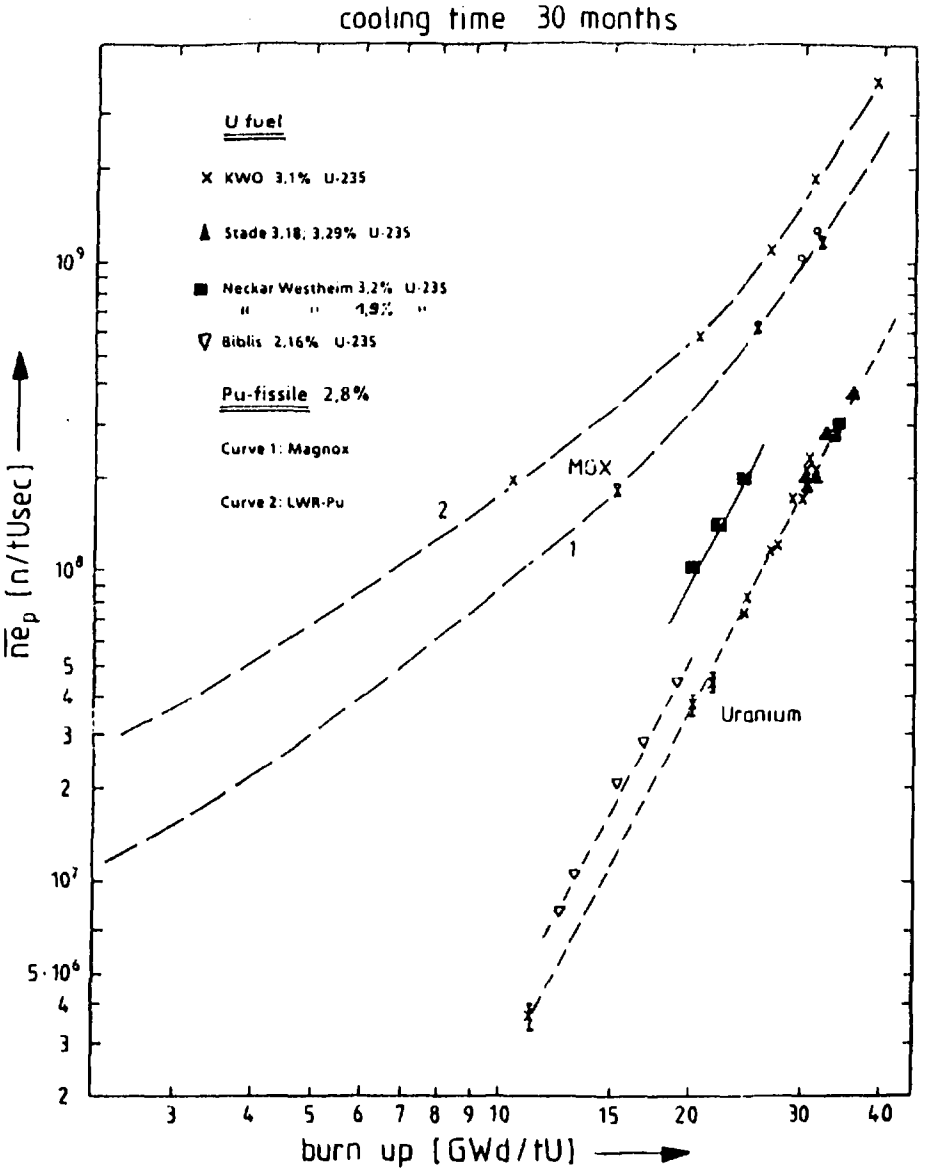

Fig. 6: Mean primary neutron emission ne_p for PWR fuel

have been determined. The ratio $R = ne_{tot}(z = l/2)/\overline{ne}_{tot}$ was evaluated for PWR fuel having different burnup and initial enrichment. R is independent of both parameters. Thus scanning is not required for PWR fuel assemblies but a measurement either in the region of max. ne_{tot} or at a fixed axial position is sufficient to obtained the assembly averaged value $\overline{ne}_{tot}$. After correction for M_{hom}, $\overline{ne}_p$ is obtained. From ne_p the amount of Cm-244 is calculated. Plotting this against the operator's declared mean burnup value results in burnup dependent Cm-244 build up as shown in Fig.5, in comparison with results of chemical analysis of feed solutions. In the limited burnup range where result of destructive analysis are available, the agreement with the nondestructive measurement data is very good.

4. EXPERIMENTAL RESULTS AND EXPERIENCE

The experimental program has made available absolute total and primary neutron emission data for LWR fuel assemblies from different reactors and for different initial enrichments. In Fig.6, fuel assembly averaged nep values for PWR fuel are shown plotted as function of the operator's declared burnup values. Conclusions from these curves are:

- the neutron emission strongly increases with burnup. For Uranium fuel with burnup above 15 GWd/tU the functional dependence is given according to

$$ne_p = a\,BU^b$$

with a = 170,3
b = 4,09

- neutron emission increases with decreasing initial enrichment
- the slope of the curves $ne_p = f(BU)$ is independent from initial enrichment
- the operator declared fuel averaged burnup values are consistent. Non outlayers larger than + 1,0 GWd/tU have been recognized
- the slope of the ne_{tot} curves for fuel assemblies in water is smaller due to the influence of neutron multiplication. Again, a power dependence is obtained, the value being b = 3,71. For borated water the slope only slightly is changed as compared with ne_p, i.e. b = 4,07
- the MOX fuel has a significantly higher neutron emission than the Uranium fuel
- the MOX neutron emission depends on the Pu vector of the recycled Plutonium. Curve 1 belongs to Magnox, curve 2 to LWR-Pu
- all PWR fuel assemblies having the same initial enrichment show the same burnup dependent primary neutron emission ne_p. They are only influenced by the moderating rate.

Measured primary neutron emission values for BWR fuel assembly are shown in Fig.7. The burnup dependence of the Uranium fuel is obtained as nep $(BU)^{4,06}$ in the burnup range above 11 GWd/tU. The same conclusions are valid as for PWR fuel.

During the experiments no problems occurred with passive neutron measurements. Due to the shielding properties of water in the storage ponds

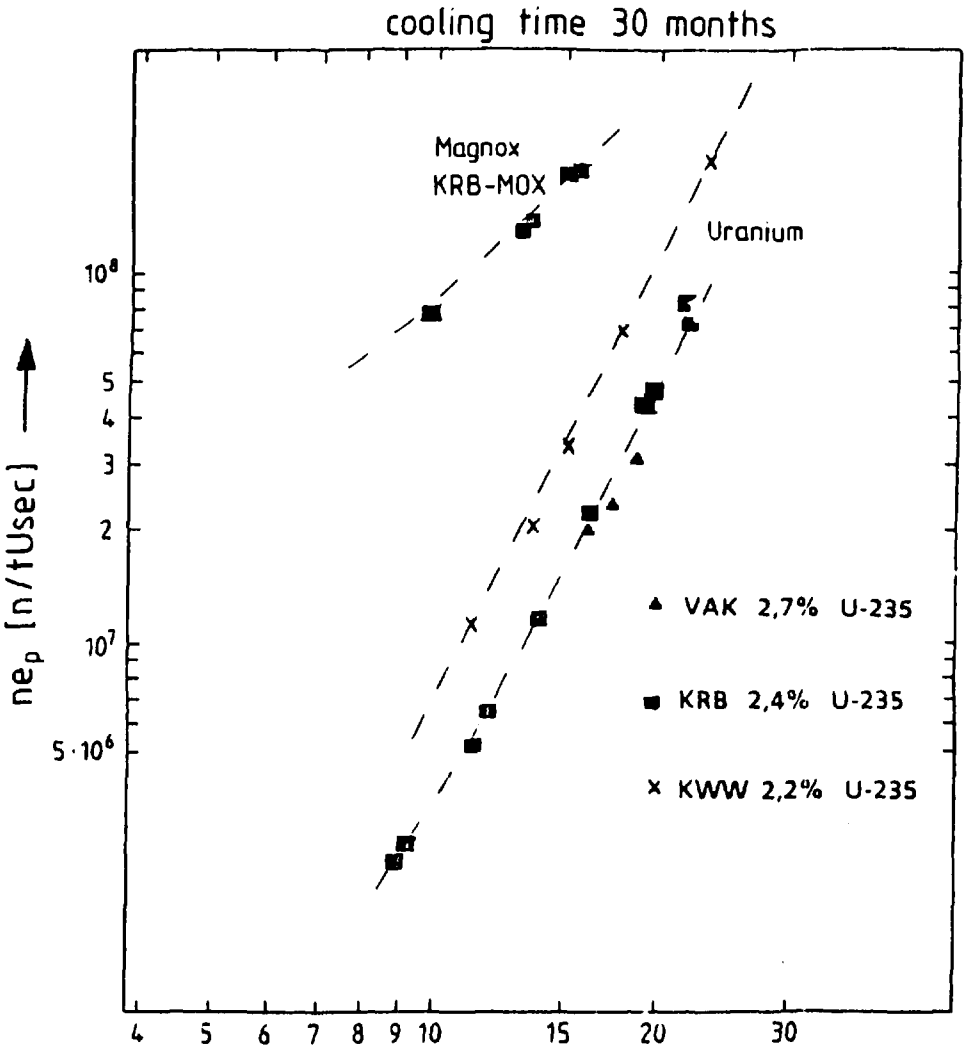

Fig. 7: Mean primary neutron emission ne_p for BWR fuel

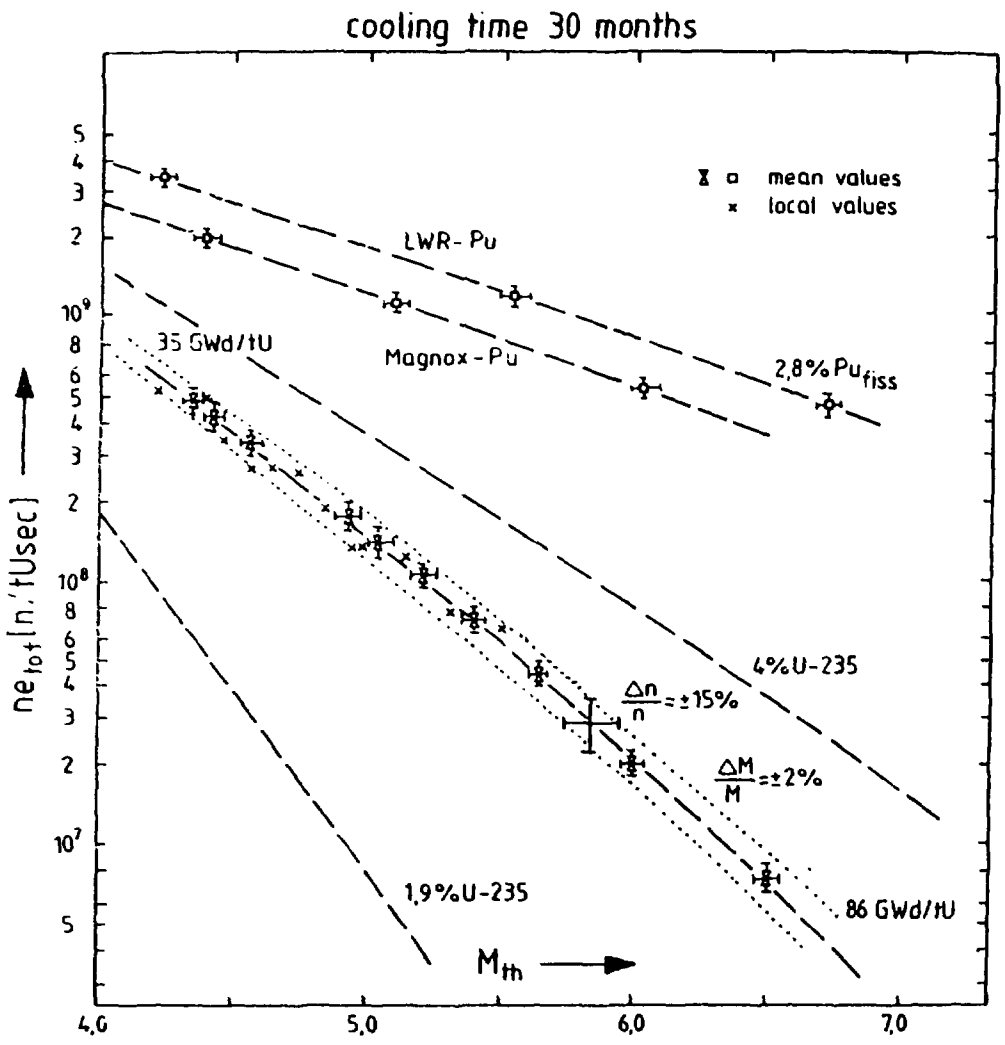

Fig. 8: Correlation of the measured quantities for KWO Uranium and MOX fuel assemblies

neutron emissions could be measured down to the burn up values of 3 GWd/tHM.

In several cases radial gradients in ne up to 30% have been observed. Examination of irradiation history revealed that these fuel assemblies had been located for most their irradiation time at the reactor core periphery. Normaly the fuel assembly position is changed from the core periphery to the center of the core with increasing burn up.

Typical measuring times are 5-15 min yielding count rates in the order of 1000 per second. Thus the statistical error can be neglected. The overall error in the count rate is less than 2%.

5. COMPLETE AND INDEPENDENT IDENTIFICATION

As a means for burnup determination, the passive neutron assay has to rely on the operators data initial enrichment and type of fuel (U or MOX). Performing an additional active measurement results in a complete and independent identification of the fuel assembly. This has been demonstrated experimentally as is shown in Fig.8. A simple plot of the experimental results of the passive assay ne_{tot} as function of the active assay M_{th} allow the determination of the initial enrichment and of the type of fuel. The results are taken from 36 fuel assemblies. The curves for U fuel of enrichment other than 3,2% U-235 are calculated.

The band width of the curves $ne_{tot} = f(M_{th})$ is due to the influence of the irradiation history. Nevertheless, the initial enrichment of PWR U-fuel assemblies can be determined for those curves with accuracies better than ± 5% as has been demonstrated for BIBLIS fuel of initial enrichment of 2,16% and 2,39% U-235 /4/.

The situation is not so favourable for MOX fuel assemblies. Neutron emission is influenced by burnup and the Pu vector of the recycled Plutonium. The quantity M_{th} depends on burnup and initial enrichment (% Pu fissile). A combination of both cannot result in a determination of 3 parameters.

6. DETERMINATION OF TOTAL PLUTONIUM

For immediate and in-field determination of the total Pu content of Uranium fuel a simple correlation between the mean primary neutron emission ne_p of the spent fuel and the total Pu content has been suggested /2,7/. The correlation is shown in Fig.9. It is based on measured primary neutron emission values ne_p and as a result of chemical analysis of reprocessing solutions. No chemical analysis data for PWR fuel assemblies were available having an initial enrichments other than 3,1% U-235.

As is indicated in Fig.9 the correlation for PWR fuel depends on the initial enrichment. Burnup dependent Pu_{tot} values for 1,9% U-235 were calculated using KORIGEN /8/ and correlated with measured ne_p values. From the overall accuracy achievable with the NDA measurement for ne_p, M_{th}, initial enrichment and from the reprocessors data on burnup dependent Pu build-up it is concluded that the total Plutonium content of spent PWR U-fuel assemblies can be determined with uncertainties of ± 0,3 kg/tHM.

7. PROTOTYPICAL DEVICE: DESCRIPTION AND TESTING

The prototypical system is shown in Fig.10. The neutron detectors are arranged symmetrically around the assembly. Each detector can be moved independently from one another with an accuracy of ± 0,1 mm. The movement is computer controlled. The actual detector positions are permanently monitored and documented. The high accuracy in detector positioning is necessary because

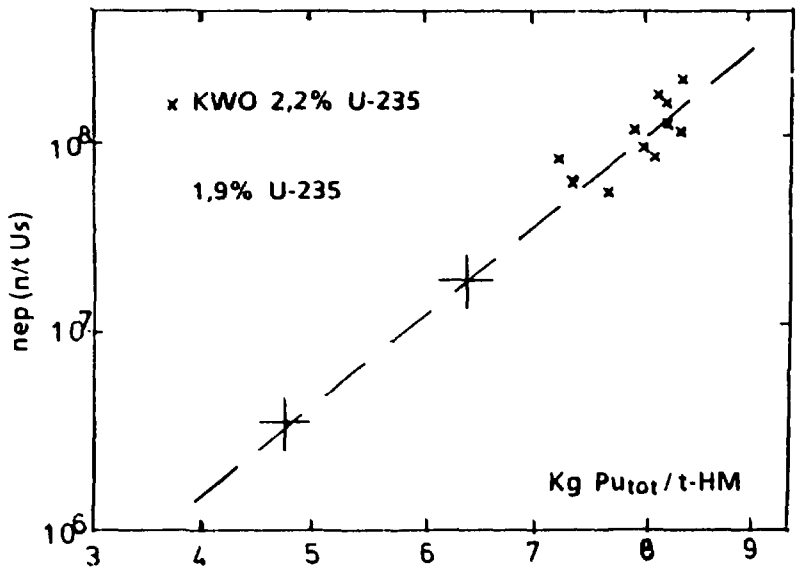

Fig. 9: Primary neutron emission as a function of total Plutonium in spent PWR fuel assemblies

of the rapidly changing count rate in relation to the distance between the external neutron source and the detector (2%/mm).

In the laboratory, the count rate is determined by moving the detectors repeatedly over a given distance to the neutron source. Fig. 11 shows that the fluctuation of the points are <0,2% what corresponds to ± 0,1 mm.

The system is equipped with a special device allowing for self centering of the monitor around the fuel assembly with an accuracy of 0,5 mm. Thus, correct measurements are also possible on banana shaped fuel assemblies. Moving the detectors independently permits measuring the dimension of the assembly that facilitates identification of fuel assemblies.

The neutron source for the active measurement is positioned in front of detector 2. For the passive measurement the neutron source is withdrawn over a distance of more than one meter from the detectors and the assembly so as not to influence the passive measurement and not to cause activation in the assembly.

All moving parts with the detectors have the feature of identical structure allowing for easier maintenance.

All components were tested thoroughly before assembling the device. Then, operation tests were performed to establish the positioning accuracy of ± 0,1 mm. After calibrating the system, it was installed in the storage pond at the WAK reprocessing plant. Presently, final operational tests are being performed.

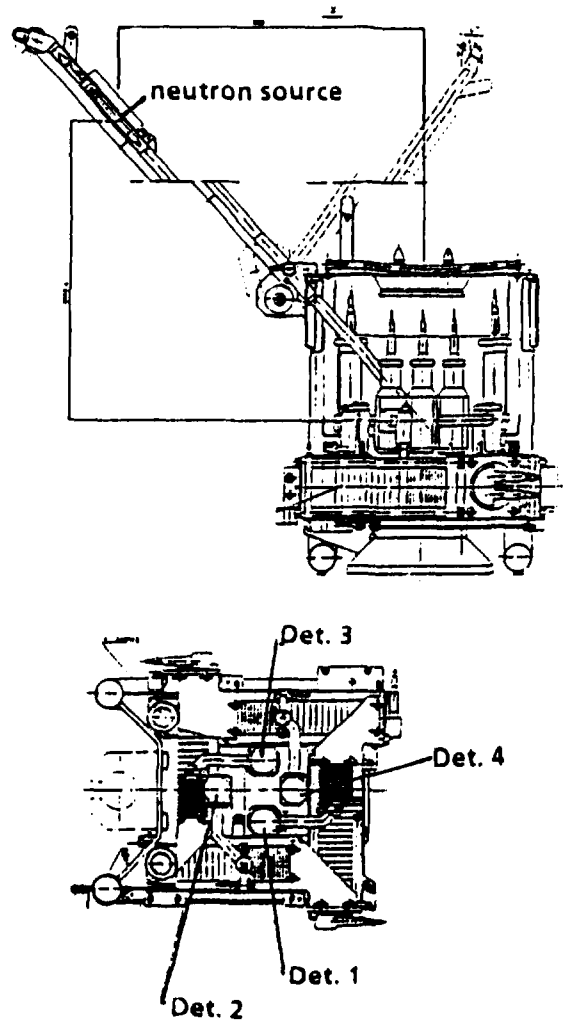

Fig. 10: Prototypical device for identification of spent LWR fuel assemblies

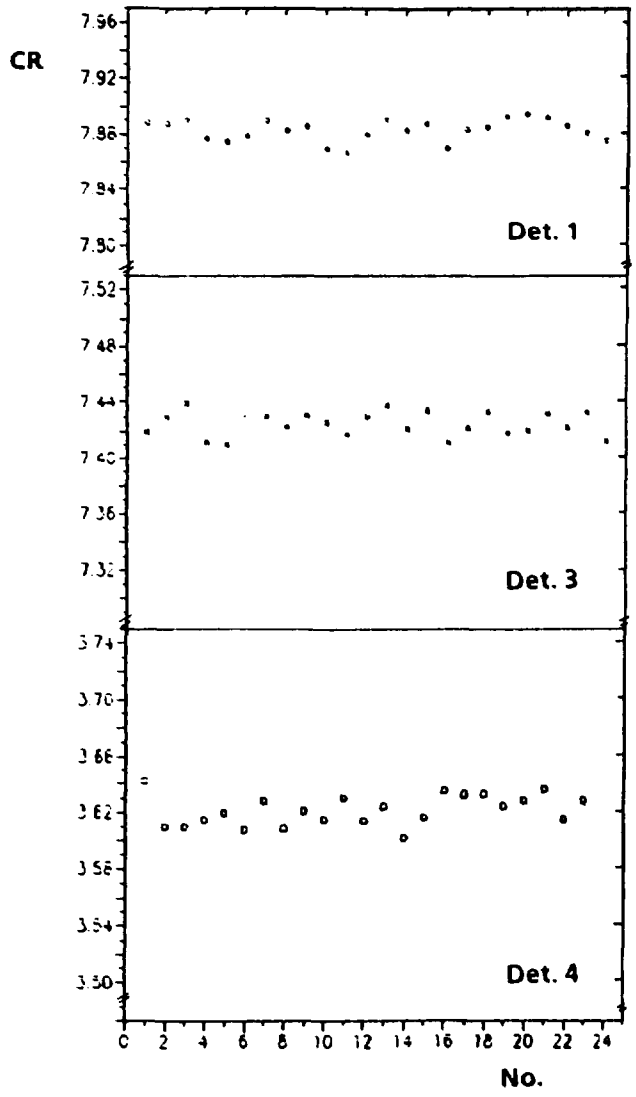

Fig. 11: Countrate determined by moving the detectors repeatedly over a given distance to the neutron source

8. FUEL IDENTIFICATION METHODS

The most sensitive parameter of a spent fuel assembly is the amount of Plutonium. When applying nondestructive methods, it is not possible to directly obtain this parameter. It has to be determined from measured quantities using established isotopic correlations verified by results of chemical analysis and burnup calculations.

The determination of Pu_{tot} requires a knowledge of the type of fuel (U or MOX), of initial enrichment, and of burnup. These quantities in principle are available from the reactor operator. However, the operator's values have to be verified. The degree of verification determines the nondestructive assay method to be applied.

8.1. NEUTRON MEASUREMENTS

As has been shown, the active and passive neutron measurement enablies the complete identification of LWR Uranium fuel assemblies. The prototypical device is a rather complicated instrument due to its movable parts which still needs to demonstrate reliability, reproducibility and durability. Therefore, a simple alternative has been developed.

The neutron source and the detectors are set at a fixed distance on the frame. This distance is a few centimeters (~5 cm) greater than the fuel assembly's dimensions. The fuel assembly will be lowered near the detector positioned oposite to the neutron source. In this position there are no affection to the countrate when the assembly is slightly moved (± 0.8 cm). For different type of fuel assembly the distance between the neutron source and the detector has to be properly selected. There is sufficient space to move the fuel assembly into the monitor without need further adjustments.

Presently, this monitor system is tested in the laboratory with 1.5% enriched fuel. Measurements with real LWR fuel assemblies are planned.

8.2. γ-SPECTROSCOPY AND PASSIVE N-MEASUREMENT

This method is used in France /9/. No movable parts are necessary. Precise positioning of the fuel assembly is not required, therefore, the device is rather simple, but voluminous and γ-spectroscopy compared to n measurements is rather complicated. γ-spectroscopy allows for the determination of cooling time and burnup. In combination with a passive n-measurement, the initial enrichment can be obtained. Thus, fuel identification is possible. The accuracy in burnup is somewhat inferior to that obtainable with n-measurement. This is due to two effects, because the γ-results are only representative for the fuel assembly's surface and because the line intensity ratio Cs-134/Cs-137 is only a weak function of burn up. On the other hand, the evaluated burn up values using neutron measuring increase of short cooling time (< 1,5 years). Thus the knowledge of cooling time is needed. This is achieved simply by doing a gross γ-measurement using a GM ionization detector.

8.3. SIMPLE SYSTEMS

The most simple method uses a passive n- and a gross γ-measurement /10/. A verification of burn up is possible (± 1,5 GWd/tHM) when the specification of the fresh assembly is known. Often, this condition is given at one reactor station.

The cross γ- measurement allows the determination of the cooling time especially for < 1,5 years. In the case, where the cooling time is greater than 1 year the measurement indicate a deviation of about 1,5 GWd/tHM.

Table 2: 2-examples for failed fuel identification in case of passive n-and gross γ-measurement: declared values are confirmed

Case 1	actually U-fuel	burnup 36 GWd/tU initial enrichment 3,2% U-235 cooling time 5 years
	declared U-fuel	burnup 23 GWd/tU initial enrichment 1,9% U5 Cooling time 5 month
Case 2	acutally MOX-fuel	burnup 20 GWd/tU initial enrichment 2,8% Pu_{fiss} Cooling time short
	declared U-fuel	burnup 35 GWd/tU initial enrichment 3,2% U5 Cooling time long

Without knowledge of the operators data, parameters such as initial enrichment and the type of fuel have to be taken into account. This method is unable to carry out a fuel assembly identification as is demonstrated in Table 2 in two different cases. Not only could a MOX assembly be declared as U-fuel, but also for U-fuel misleading results are possible. Without means for independent identification this simple method is only able to discriminate between fresh and spent fuel, a task which can be done more easily by the measurement of Cerenkow radiation /11/.

8.4. CONCLUSIONS

It has been demonstrated that fuel identification is possible using a combination of active and passive neutron measurement. The results of a demonstration program performed at 92 LWR fuel assemblies have been so encouraging, that a decision was made to built a prototypical NDA system. This system must still demonstrate that the accuracies hitherto obtained with a manually operated system, also are obtained in routine measurements using a fully automated system. A simplified version of this device is believed to be the NDA system which fulfills all requirements for fuel identification. And, due to its simple technical layout, guaranteed durability.

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EXPERIMENTAL INVESTIGATION OF THE RADIAL FISSION PRODUCT DISTRIBUTION IN VVER FUEL

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Abstract

This paper presents experimental investigations for the determination of n,γ (U-238) and n,f (U-235) reaction rates in an elementary cell near the core performed at BOL. On the basis of this information the Pu-239 buildup and U-235 fission rate were calculated and used as input in 2 DT calculations. These resulted in a 200°C central temperature decrease (at about 448 W/cm MLHR).

Increase of the VVER power density makes tougether requirements to the fuel rod reliability which depends on the temperature fields in the fuel rod and the moderator. The temperature field in the reactor is determined by power distribution depending both on the initially charged nuclear fuel and plutonium buildup during the core lifetime. Distribution of the ^{239}Pu fission rate is connected with the plutonium buildup rate which is not the same over the fuel rod radius, which is specially important for the fuel rods nearby the medium inhomogeneities in the core.

Experimental investigation of distribution of the $^{238}U(n,\gamma)$ - and $^{235}U(n,f)$ reaction rates in the elementary cell near the core inhomogeneity in the beginning of the core lifetime and subsequent use of the information obtained in the computer codes permit the average temperature of the fuel rods to be estimated.

The nuclear reaction rate distribution was studied on the subcritical facility simulating the VVER fuel assembly with the UO_2 with 6.5% enriched ^{235}U and a density of 10.4 g/cm^3. The fuel pellet diameter was 7.65 mm /1/. For the inhomogeneity modelling one fuel rod was removed, and thus a water rod was formed or replaced by a niobium-zirconium hollow tube or by the absorbing rod. The PE consisting of small B_4C pellets (7.65 mm in dia with a density 1.78 g/cm^3 and natural boron content 1.4 g/cm^3) has the same cladding as the VVER-440 fuel rods.

Distribution of the $^{235}U(n,f)$, $^{238}U(n,\gamma)$ reaction rates in the elementary uranium-water cell near the core inhomogeneity was measured after irradiation of physically thin foils of natural metallic uranium and 90% enriched uranium cladded with aluminium. For determination of the spatial distributions in the small-pitch array of VVER a scheme of setting detector foils for irradiation was developed.

The 12 mm-long foils sandwiched between the two parts of the fuel and absorber slugs out vertically over the diameters were inserted into dismountable rods with the similar slugs. In the moderator the foils were vertically sandwiched between the fuel rod, poisoning element or any other inhomogeneity and fastened with thin (1 mm thick) plastic clips.

The dismountable rods were irradiated in the center of the subcritical facility and after that the uranium foils were cut into pieces and the γ-ray intensities of the products of the ^{238}U radiation capture reaction, ^{239}Np($T_{1/2}$=2.346±0.004 day, E_γ = 277.63± ±0.01 keV), and of the ^{235}U fission, ^{143}Ce($T_{1/2}$=33.0 ±0.2 hr, E = 293.262 ± 0.021 keV, were measured. The measurements were carried out simultaneously on two semiconductor Ge-Li detectors with energy resolutions of 2.3 and 2.9 keV and E_γ = 1332 keV, connected

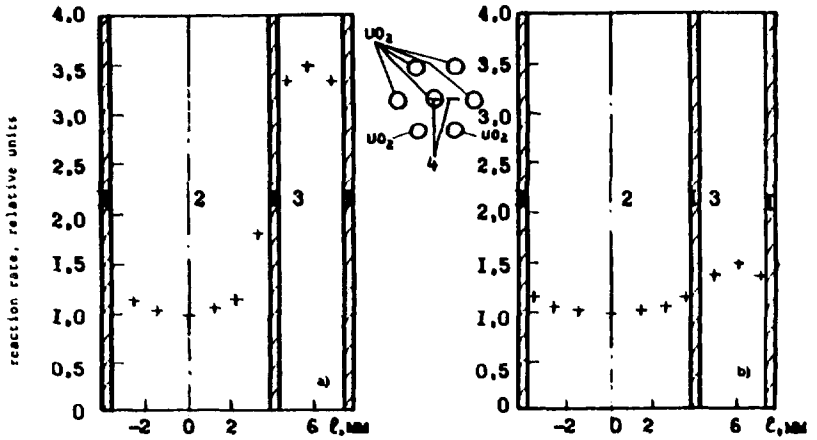

FIG.1. Distribution of the nuclear reaction rates in the elementary cell:
a) ^{238}U (n,γ) reaction, foil of natural metallic uranium.
b) ^{235}U (n,f) reaction, foil of 90% enriched ^{235}U.

Notes:
1 — cladding
2 — UO_2 fuel
3 — moderator with air spacing between the cladding and fuel of 0.125 mm
4 — foil.

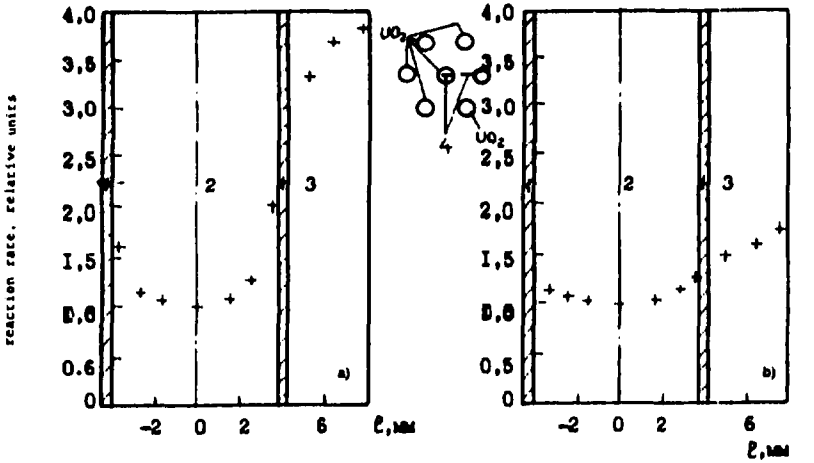

FIG.2. Distribution of the nuclear reaction rates in the elementary cell with the water rod. (Notes: see Fig.1).

to the input of the analyzer working online with the computer on which the γ-spectra obtained were processed.

The measurement results of relative distributions of the reaction rates in a homogeneous elementary cell, and in a cells with water, air and absorber rods are shown in Figs.1-5.

The errors in the distributions measured are both statistical and systematical. Statistical are the errors in determination of the intensities of the γ-lines measured, determination of the concentrations of the starting nuclides in the experimental foils and determination of temporary functionals.

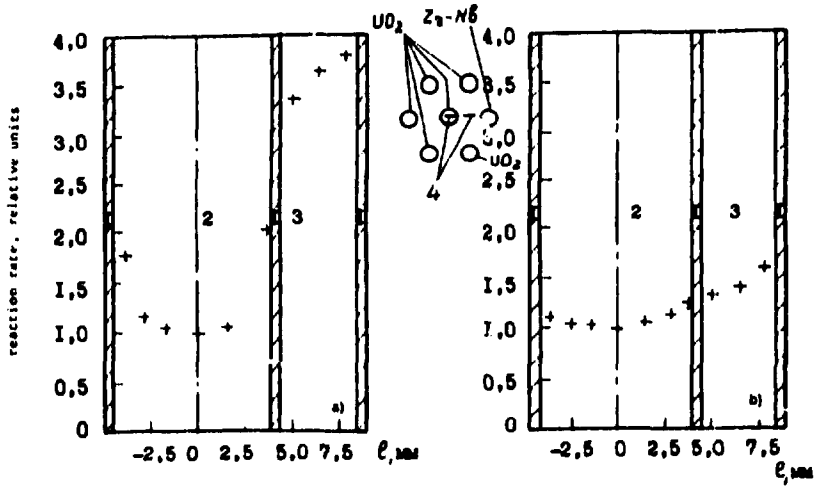

FIG.3. Distribution of the nuclear reaction rates in the elementary cell with the hollow Zr-Nb tube air rod. (Notes: see Fig.1).

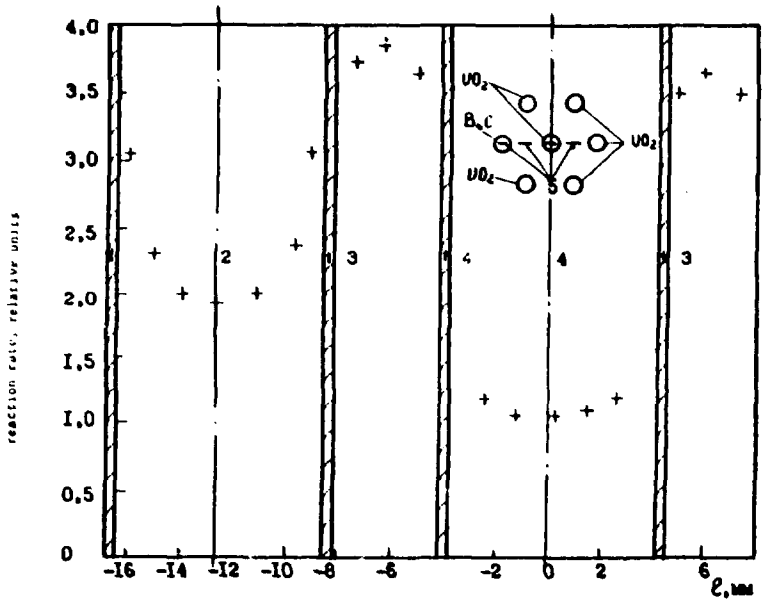

FIG.4. Distribution of the ^{238}U (n,γ) reaction rate, measured with a foil of material metallic uranium in the elementary cell with the B_4C rod.

Notes:
1 — cladding
2 — B_4C
3 — moderator
4 — UO_2 fuel; 0.125 mm spacing between cladding and fuel
5 — foils.

Systematical are the errors in foil setting in the fuel rod and the moderator in height of the fuel assembly and the errors arising from inaccuracies in determination of the radial coordinate of specimen location after cutting.

The analysis revealed that the errors of the measured rate distributions depend slightly on the inhomogeneity type and do not practically differ from the errors of distribution in the homogeneous cell. The main contribution to each error comes from the systematical error arising from the inaccuracy of the radial coordinate determination. To minimize it a special technique for cutting specimens on the coordinate electrospark machine has been developed.

The error in measurement of the spatial distribution of the $^{238}U(n,\gamma)$ reaction rates varies from 2% in the fuel rod center to 3.5% at its edges, and that of the $^{235}U(n,f)$ reaction rates from 2% to 2.5%, respectively. In the moderator the error in distributions of these reactions is 2.5%.

The studies have shown that the technique used is sensitive to the inhomogeneities giving shifts of not more than 3.5% in distribution

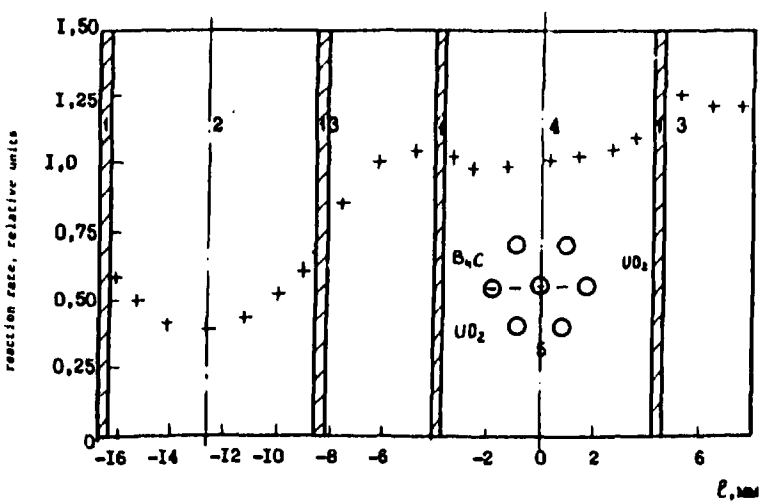

FIG.5. Distribution of the ^{235}U (n,f) reaction rate, measured with foil of uranium 90% enriched. ^{238}U in the elementary cell with the B_4C rod.
(Notes: see Fig.4).

of the $^{238}U(n,\gamma)$ reaction rate and not more than 2.5% in distribution of $^{235}U(n,f)$ reaction.

The greatest asymmetry in distribution of the plutonium build up rate in the fuel rod (up to 20-30%) was observed near the air and water channels. The greatest asymmetry in distribuyion of the ^{235}U fission rate in the fuel rod (up to 10%) was observed near the poisoning element. In the second row of the fuel rods this effect have not been found.

For the homogeneous array of fuel rods the average fuel rod temperature was estimated taking into account the measured radial distributions of the ^{239}Pu buildup and ^{235}U fission rates /2/. For the maximum linear power of the fuel element q_e=448 W/cm the maximum temperature in the fuel center is lower by 200°C than in the constant power distribution over the fuel rod cross section. This evidences the necessity of allowance for the azimuthat distributions in accurate calculations of the fuel rod temperature regimes.

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BURNUP DETERMINATION BY THE ISOTOPE CORRELATION TECHNIQUE

(Summary)

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1. GENERAL

The burnup of actinides and the consequent formation of fission products or higher actinides is inherently related. This can be expressed empirically by correlations, the basis for which has been proven theoretically[1]. The burnup of the spent fuel can be obtained from the difference between the heavy nuclide concentrations before and after irradiation or from the amount of fission products formed relative to the initial actinides. There exists a third possibility which is exploited by the isotope correlation technique based on the buildup of isotopes by neutron capture. The buildup is proportional to the integrated neutron flux. This can be seen from changes in the isotopic ratio of plutonium, e.g. $^{242}Pu/^{240}Pu$ which increases proportionally to the burnup or for those fission-product ratios containing a nuclide with an appreciable neutron capture cross-sections e.g. $^{131}Xe/^{132}Xe$. Even the recycling of Pu in thermal reactors is not prohibitive to the application of the isotope correlation technique[2]. In earlier publications[3,4,5,6] we have listed possible correlations (Tab. I) which could be used for this purpose.

Correlation F_T vs	Reactor Type	
	BWR	PWR
$^{242}/^{240}Pu$	.81	.97
$^{132}/^{131}Xe$	.92	.98
^{242}Cm	.86	.87
^{244}Cm	.89	.94
^{240}Pu	.91	.93

Tab.I Correlations between burnup (F_T) and isotope ratios or isotope concentrations expressed as correlation coefficients

2. APPLICATIONS

An application to burnup determination by the isotope correlation technique can be seen in the field of reprocessing where it is used to establish a material balance and in the storage of spent fuel where burnup is one of the parameters characterising the fuel.

2.1 Material balance

In order to establish a gravimetric balance at the head-end of the reprocessing plant the final (after irradiation) plus the proportion fissioned and initial (before irradiation) amounts of actinides are compared[7]. The amount fissioned, expressed as burnup, has previously bee: determined by fission product analysis e.g. ^{148}Nd. If heavy isotope correlations are used, as for instance $^{242}Pu/^{240}Pu$ vs burnup the analytical effort is considerably reduced because the plutonium and uranium isotopic abundances are obtained in routine analysis. From the analysis of 77 PWR fuel samples the following correlation was obtained: $(F_T) = 0.2429 + 14.36\ ^{242}Pu/^{240}Pu$ (Fig. 1). The accuracy of the isotope correlation technique is better than 5% which is sufficient for the purpose of establishing the material balance at the input. For a normal burnup of 3% the contribution to the to' ' error of the material balance would be less than 0.15%.

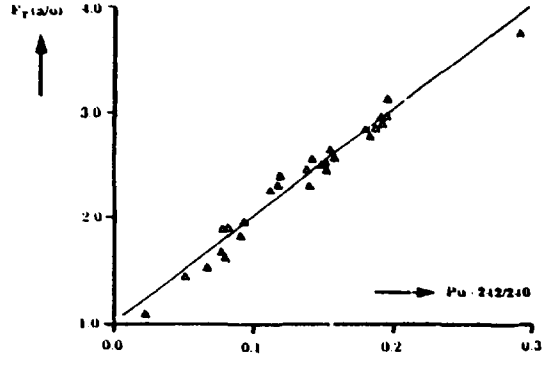

Fig. 1 Example of Isotope Correlation between (F_T) and $^{242}Pu/^{240}Pu$

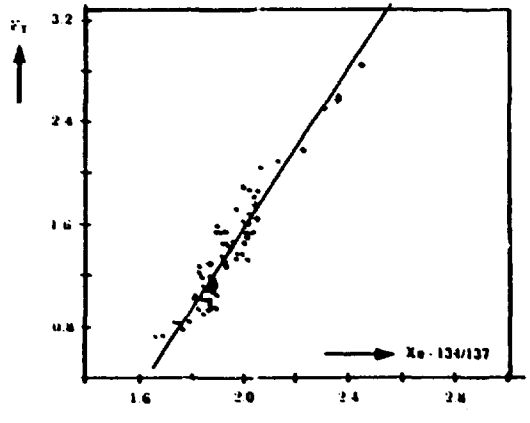

Fig. 2 Correlation fission gas ratio Xe-132/131 vs. fuel exposure (F_T) in atom percent
(+) PWR (KWO, SENA, TRINO)
(-) BWR (DODEWAARD, KAHL, GARIGLIANO).

For large reprocessing plants with a quasi-continuous dissolution of fuel the identity of the fuel assemblies is disturbed by batch mixing. In this case there is no direct relationship between the burnup determination from the dissolver solution and that of the spent fuel. However the fission gas released during dissolution of a fuel assembly is not effected by this batch mixing process. Hence correlations based on fission-gas isotope ratios such as $^{132}Xe/^{131}Xe$ can be used to determine the burnup[8]. This particular correlation seems to be rather insensitive to changing parameters during irradiation and fuel fabrication (Fig. 2). Here fuel samples of different cores of several pressurised and boiling water reactors were used. The correlation equation is obtained only for 29 PWR samples: $-4.11 + 2.844\ ^{132}Xe/^{131}Xe$.

2.2 Burnup of stored spent fuel

Among other parameters, the burnup of unreprocessed fuel has to be known before storage. There are in principal two possible ways to measure the burnup by nondestructive analysis, making use either of neutron or gamma interrogation.

As outlined above the neutron capture of certain fission products e.g. $^{133}Cs\ (n,\gamma)\ ^{134}Cs$ can be related by a correlation to the burnup[9, 10] (Fig. 3). Of course this correlation is sensitive to the cooling time of the spent fuel and to certain fuel management schemes which would influence the ^{134}Cs content due to in-pile decay. Where the irradiation history is not known it can be partially reconstructed from the presence of other radioactive gamma emitters.

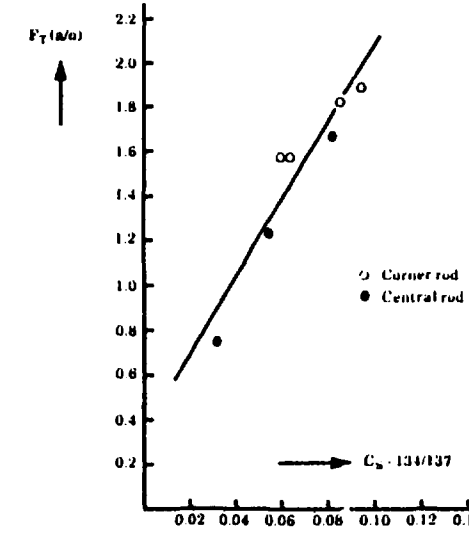

Fig. 3 Correlation of (F_T) vs. C_S-134/137

The formation of transuranium nuclides could also be used in this case for burnup determination. The neutron radiation from ^{242}Cm for freshly discharged assemblies or ^{244}Cm for longer cooled fuel can be directly related to the burnup[11] (Fig. 4). The dependence of the counting geometry can be partly eliminated if the neutron emission rate is measured relative to the gamma radiation of selected fission products. For very long fuel storage ^{240}Pu is an alternative for passive neutron interrogation.

3. OUTLOOK

The need to determine the burnup directly for the purpose of the material balance or to verify reactor physics calculations is diminishing. Two other requirements are gaining in importance:

- The authentication of spent fuel for nuclear material safeguards which could be established by verifying among other parameters the declared burnup.
- The risk assessment of the storage of spent fuels which requires verification of the declared transuranium and fission product content. Safety authorities would need nondestructive techniques which could directly measure burnup and the transuranium content of spent fuel.

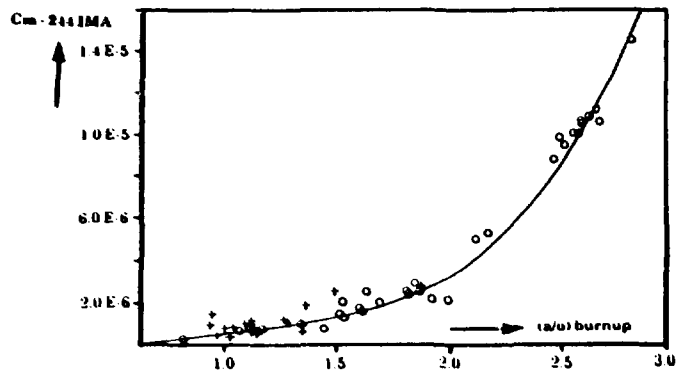

Fig. 4 Isotope correlation (Fy) vs. Cm-244 observed for various LWR: PWR Trino Vercellese, PWR Obrigheim, PWR Sena; BWR Gargliano, BWR Kahl, BWR Lingen, BWR Gundremmingen (IMA = per initial heavy metal atom)

For the isotope correlation technique to be accepted more experimental evidence is needed in order to assess the accuracy and the reliability of the nondestructive analyses. This can only be obtained from a comparison of results from nondestructive and destructive techniques.

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NON-DESTRUCTIVE BURNUP DETERMINATION OF SPENT FUEL IN THE FRAMEWORK OF NUCLEAR MATERIAL SAFEGUARDS
(Summary)

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1. INTRODUCTION

The Euratom Safeguards Directorate has spent considerable effort during the last few years in the test, acquisition, calibration and implementation of new equipment for nuclear materials verification.

One of the instruments introduced is a passive gross neutron and gross gamma counting instrument, which is primarily used to determine the burn-up and cooling time of irradiated Light Water Reactor Fuel assemblies.

This paper presents a short summary of results obtained. For more detailed information, reference is made to the literature.

2. DESCRIPTION OF EQUIPMENT

The measurement equipment consists essentially of four components: a fork shaped polyethylene det. co. body containing neutron (fission chambers) and gamma (ionisation chambers) detectors; a set of pipes which allows the detectors to be fixed from the bridge of the fuel handling machine and suspended over the top of the fuel assembly storage rack; a portable neutron and gamma-ray detector electronics and a small computer for data acquisition and evaluation.

A detailed description of the equipment is given in references [1, 2 and 3].

For the actual measurement, the fuel assembly is lifted about half way out of its storage position so that the fork shaped detector surrounds the element at about the middle of the assembly (area of maximum burn-up).

3. SURVEY OF RESULTS OBTAINED

From the first use of the equipment in 1983, Euratom has measured more than 500 assemblies in the various facilities within the European Community including PWR fuel, BWR fuel, various special fuel assemblies and irradiated MOX fuel assemblies.

The initial enrichment of the fuel varied from natural uranium to 3.5% U235, the declared burn-up varied between 1400-41000 MWd/tU and the cooling time was between 36 and 5000 days.

For standard PWR and BWR fuel assemblies, the consistency between declared and measured burn-up was about 5-6% for fuel with normal irradiation history and same initial enrichment. For the determination of the cooling time the observed standard deviation of the difference was about 15-20%.

Some typical measurement results were published recently [3,4,5].

4. CONCLUSIONS AND OUTLOOK

In order that the instrument is further developed and becomes a valuable burn-up meter for all types of fuel the following R&D activities are at present under study [6,7].

- calculation of influence of unusual irradiation histories, different initial enrichments, and difference in reactor and fuel type (neutron flux distribution) on neutron emission of spent fuel
- measurement of neutron emission with normal fission chambers and cadmium-shielded fission chambers to measure and correct for different boron concentrations in pond water

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CALCULATION MODELS AND THEIR QUALIFICATIONS

PROLONGED FUEL BURNUP IN ADVANCED LWRs

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Abstract

Nuclear power plants expected to be built in Yugoslavia in the future will probably be of an advanced type, with prolonged fuel cycles leading to discharge burnup values considerably higher than the ones achieved with the present day plants. In order to obtain elements needed in the procedure of bid evaluation, a comparative study of fuel utilization in different types of advanced PWR's, including the advanced type of VVER, has been performed. Some typical results are presented and discussed here. In treating the effects of prolonged fuel burnup, changes in the design of fuel assemblies, core configuration and reactivity control system, introduced in order to provide longer fuel irradiation, are to be considered. Here, the specific design characteristics, as increased initial enrichment, advanced type burnuble absorbers and low-leakage loading patterns, are taken as for the advanced 1000 MWe LWR power plants. Calculations are performed using the NET IBK computer code package. The calculational scheme comprises: neutronic calculations of fuel pin cell parameters versus burnup; neutronic calculations of few group parameters for fuel assemblies with inserted or withdrawn control rods and for specified values of fuel, coolant and moderator temperatures and specified fuel burnup; few group diffusion theory calculation of control rod position and neutron flux and power distributions in a reactor core, divided into a number of zones having different fuel burnup values. Repeating the global reactor core calculations with a specified burnup step for a typical equilibrium cycle, the entire range of fuel burnup values is covered. The feedback between neutronic and thermo-hydro-dynamic calculations can be provided by introducing new temperatures in neutronic calculations when necessary. The obtained results provide a good basis for estimating the economic and safety impacts of prolonged fuel burnup, as well as for judging the possibilities of fuel utilization improvements.

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1. INTRODUCTION

For a developing country buying its first nuclear power plants from a foreign supplier, disregarding the type and scope of the contract, there is still a number of activities which has to be performed by local stuff and domestic organizations. This particularly applies to the choice of the fuel cycle strategy and the choice of the type and the size of nuclear power plants, to the bid parameters specification and bid evaluation, the evaluation of safety analysis reports, as well as to the in-core fuel management activities.

At the Nuclear Engineering Department of the Boris Kidrič Institute of Nuclear Sciences (NET IBK) systematic work has been going on for quite a period of time with the aim to complete a computer code package for reactor core analysis and design /1/. Nuclear data available on the basis of international cooperation and exchange are being collected, while the already formed nuclear data libraries are being updated. Own methods, algorithms and codes are being developed. Foreign codes, supplied from different code centers, are being modified for application on the avilable computes and for the own needs. Developed computer programs are being transformed into well documented codes and modules. Computer codes are being designed for connecting the independent modules into complex modular schemes.

Fuel burnup analysis comprises: neutronic calculations of fuel pin cell parameters versus burnup; neutronic calculations of few group parameters for fuel assemblies with inserted or withodrown control rods, versus burnup and for specified values of fuel, coolant and moderator temperatures; few group diffusion theory calculation of control rod position and neutron flux and power distribution in a reactor core and determination of global fuel burnup parameters; calculation of related thermo-hydro-dynamic parameters. Repeating the global reactor core calculations with a specified burnup step for a typical equilibrium cycle, the entire range of fuel burnup values is covered. The feedback between neutronic and thermo-hydro-dynamic calculations can be provided by introducing new temperatures in neutronic calculations when necessary.

The standard NET IBK calculational scheme consists of the WIMS code, for pin cell and fuel assembly calculations, and of several 2 D (RZ or XY) or 3 D (XYZ) codes for overall reactor core calculations and criticality search. They are coupled and modified to compute neutron flux, power density distribution and burnup taking into account spatial variations of temperature and xenon poisoning, as well as the reactivity changes due to xenon transients during the start-up and shut-down. Presently, codes for overall reactor calculations are based on finite difference solution of group diffusion equations. Efforts are being made to improve reactor cell and fuel assembly parameters calculations, and to develop advanced methods for solving diffusion equations.

The NET IBK computer code package has been extensively used to solve in-core fuel management problems of the own research reactors, as well as for studying advanced fuel utilization schemes in different types of power reactors. Particular attention has been paid to experimental verification of the calculational procedures. This paper presents and discusses the current status of the NET IBK computer code package for burnup predictions and in-core fuel management, as well as the results obtained in the frame of several studies performed for the utility organizations /2,3/.

2. PIN CELL CALCULATIONS

In order to get elements for evaluating the overall fuel cycle strategy, first a comparative study of typical burnup parameters for different types of 1000 MWe nuclear power plant has been performed /3/. For this kind of study, it is appropriate to consider the results of pin cell calculations /10/. The basic input data concerning the standard fuel management schemes in present day nuclear power plants, provided by different suppliers /4-9/, are presented in Table I.

In Table II the calculated values of heavy isotope compositions of fuel discharged from different types of nuclear power plants are compared with the reference values. Satisfactory agreement can be noticed. Fission products inventory in fuel discharged from different types of nuclear power plants is given in Table III. The heavy isotope compositions and corresponding intensities of ionizing radiation, 3 and 15 years after the fuel has been discharged from the reactors, can also be calculated using the programs for solving the fuel depletion equations /11/. The results are presented in Table IV and Table V.

3. FUEL ASSEMBLY CALCULATIONS

Average discharge fuel burnup in present day nuclear power plants can be increased by improving the fuel assemblies design, by optimizing the fuel loading patterns and by adequate choice of operational regimes. Increasing the average power density in fuel (smaller pin radius) and spatial flattenning of power generation (convenient loading pattern schemes, use of burnuble apsorbers and axial blanket) can lead to considerable saving in fuel material requirements and in requirements for fuel cycle services. However, from both economical and operational point of view it is particularly important to prolong the stay of fuel in the reactor. In light water reactors this can be achieved by either increasing the length of particular cycles (18 month cycles instead of 12 month cycles), or by increasing the number of 12 month cycles per a fuel assembly (four instead of three).

In considering the effects of prolonged fuel burnup, two aspects of the problem should be noted: (1) changes in the design of fuel assemblies, core configuration and reactivity control system, introduced in order to provide longer fuel irradiation; (2) changes of safety and operational characteristics caused by prolonged irradiation. In the present paper the following design characteristics, belonging to the category (1) are considered: increased initial enrichment, advanced type burnable absorbers and low-leakage loading patterns. The basic design data are taker as for typical advanced 1000 MWe PWR's offered by potential suppliers /4,5,6,7/.

The fuel assembly is supposed to have square lattice with 17x17 positions. As presented in Fig.1., 264 positions are occupied by fuel pins, 24 positions are occupied by quide tubes for control absorber rods and one

Table I - Data used in pin cell fuel burnup calculations

type of 1000 MWe NPP	PWR	VVER	BWR	PHWR
first core initial enrichment (kg ^{235}U/MTU)	25	24	20	7.11
equilibrium core initial enrichment (kg ^{235}U/MTU)	33	44	7.11	25.6
average discharge fuel burnup (6WD/MTU)	33	40	28.5	7.5
average power density in fuel (kW/kgU)	38.3	45.5	24.2	19.1
fuel pellet radius (mm)	8.19	7.55	10.6	14.2
thickness of Zr cladding (mm)	0.57	0.65	0.66	0.38

Table II - Calculated and reference values of heavy isotope compositons in fuel discharged from different types of nuclear power plants (kg/MTU)

isotope	^{235}U	^{236}U	^{238}U	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
			PWR				
calculated	8.376	4.193	941.0	5.932	2.354	1.428	0.581
reference	9.000	3.800	940.7	5.407	2.379	1.299	0.473
			VVER				
calculated	12.575	5.188	930.3	5.497	2.379	1.644	0.654
reference	12.600	5.000	930.0	5.600	2.100	1.800	0.600
			BWR				
calculated	8.428	3.596	946.7	4.261	2.867	1.337	1.289
reference	8.400	3.500	947.0	4.576	2.056	0.928	0.360
			HWR				
calculated	2.112	0.743	985.6	2.665	0.878	0.211	0.588
reference	2.270	0.733	985.5	2.599	0.973	0.174	0.530

Table III - Fission products inventory in fuel discharged from different types of nuclear power plants

Isotope (kg/MTU)	PWR	VVER	BWR	HWR
^{83}Kr	0.039	0.049	0.035	0.009
^{95}Mo	0.791	0.994	0.975	0.177
^{99}To	0.832	1.029	1.041	0.192
^{101}Ru	0.796	0.970	1.065	0.176
^{103}Ru	0.047	0.053	0.053	0.029
^{103}Rh	0.396	0.458	0.465	0.094
^{105}Rh	0.001	0.001	0.001	0.001
^{105}Pd	0.383	0.419	0.594	0.078
^{108}Pd	0.163	0.174	0.281	0.040
^{109}Ag	0.081	0.086	0.127	0.022
^{113}Cd	0.0001	0.0002	0.0001	0.00004
^{115}In	0.001	0.002	0.001	0.0006
^{127}I	0.051	0.061	0.067	0.012
^{131}Xe	0.427	0.521	0.478	0.122
^{133}Ca	1.126	1.393	1.386	0.275
^{134}Ca	0.146	0.174	0.238	0.011
^{135}Xe	0.0002	0.0001	0.0001	0.00004
^{135}Ca	0.341	0.385	0.336	0.026
^{143}Nd	0.827	0.986	0.747	0.177
^{145}Nd	0.672	0.835	0.790	0.154
^{147}Pm	0.144	0.169	0.128	0.031
^{147}Sm	0.053	0.093	0.061	0.008
^{148}Pm	0.003	0.003	0.001	0.0085
^{149}Sm	0.003	0.003	0.001	0.0006
^{150}Sm	0.341	0.409	0.438	0.063
^{151}Sm	0.022	0.021	0.017	0.002
^{152}Sm	0.123	0.151	0.164	0.041
^{153}Eu	0.114	0.129	0.157	0.018
^{154}Eu	0.048	0.052	0.068	0.003
^{155}Eu	0.006	0.006	0.008	0.0006
^{157}Gd	0.00004	0.00003	0.00002	0.000

central position is reserved for the instrumentation tube. The "cluster" option of the WIMSD-4 code /10/ has been used to calculate group values of the efeective fuel assembly physical parameters versus burnup. The portion of the fuel assembly assigned to one control rod, i.e. its 1/24 part is simulated by a multizone cylindrically symmetrical lattice cell in the manner shown schematically in Fig.2. Two central zones are occupied by a

Table IV - Heavy isotope concentrations and corresponding intensities of ionizing radiation 3 years after the fuel has been discharged from the reactors

isotope (kg/MTU)	PWR	VVER	BWR	HWR
^{235}U	8,353	12,538	8,419	2,110
^{236}U	4,195	5,191	3,578	0,740
^{238}U	940,950	930,236	941,687	985,320
^{239}Pu	5,923	5,484	4,247	2,663
^{240}Pu	2,359	2,382	2,974	0,870
^{241}Pu	1,221	1,405	1,140	0,209
^{242}Pu	0,631	0,660	1,293	0,580
(Bq/kg ^{235}U)	$1,946 \cdot 10^{13}$	$1,990 \cdot 10^{13}$	$1,800 \cdot 10^{13}$	$1,045 \cdot 10^{13}$

Table V - Heavy isotope concentrations and corresponding intensities of ionizing radiation 15 years after the fuel has been discharged from the reactors

isotope (kg/MTU)	PWR	VVER	BWR	HWR
^{235}U	8,257	12,503	8,388	2,070
^{236}U	4,207	5,191	3,574	0,738
^{238}U	940,770	930,154	941,681	985,300
^{239}Pu	5,887	5,471	3,806	2,610
^{240}Pu	2,381	2,395	3,382	0,861
^{241}Pu	0,664	1,202	1,108	0,203
^{242}Pu	0,806	0,661	1,343	0,578
(Bq/kg ^{235}U)	$1,344 \cdot 10^{13}$	$1,357 \cdot 10^{13}$	$1,362 \cdot 10^{13}$	$9,341 \cdot 10^{12}$

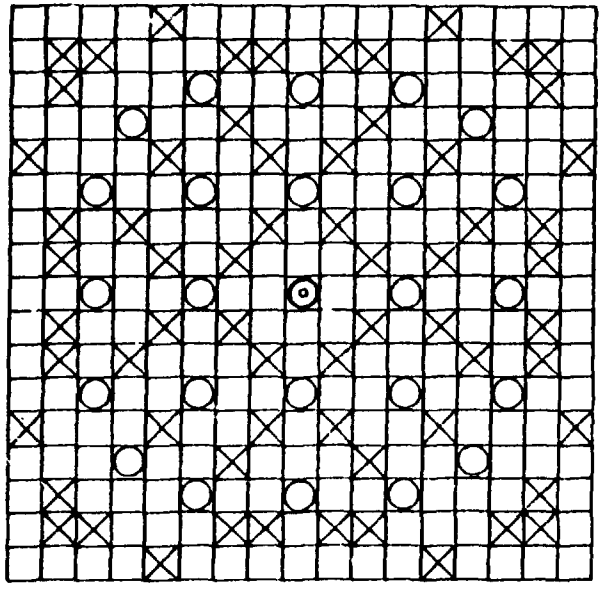

Figure 1. Fuel assembly - top view

control rod and its cladding, i.e. by moderator if control rods are not inserted. The next four annular zones correspond to moderator, control rod guide tube, structural materials of instrumentation channel and moderator. The seventh zone contains four fuel pins adjacent to the control rod. Moderator again occupies the zone number eight. Zone number ten contains 1/24 portion of fuel pins whose lattice cells have no contact with the control rod cell, while the zone number nine contains 1/24 portion of the remaining fuel pins. The outermost eleventh zone then contains the corresponding portion of the fuel assembly structural material.

Fuel assembly calculations have been performed for an equilibrium cycle with the fresh fuel initial enrichment being 3.35% or 3.55% of ^{235}U. The following cases have been treated: fuel assembly without control rods or burnable absorbers, fuel assembly with integral type burnable absorbers (a thin boride coating applied to the surface of the fuel pellets, i.e. combination of fuel and burnuble absorber material in a single rod) and fuel assembly with inserted control absorber rods. Infinite medium criticality parameters versus burnup for the first two cases are presented in Table VI.

4. BURN-UP PREDICTIONS FOR REACTOR CORE

The reactor core burnup analysis has been performed using the program VAMPIR /12/, which solves few group diffusion equations for a multizone

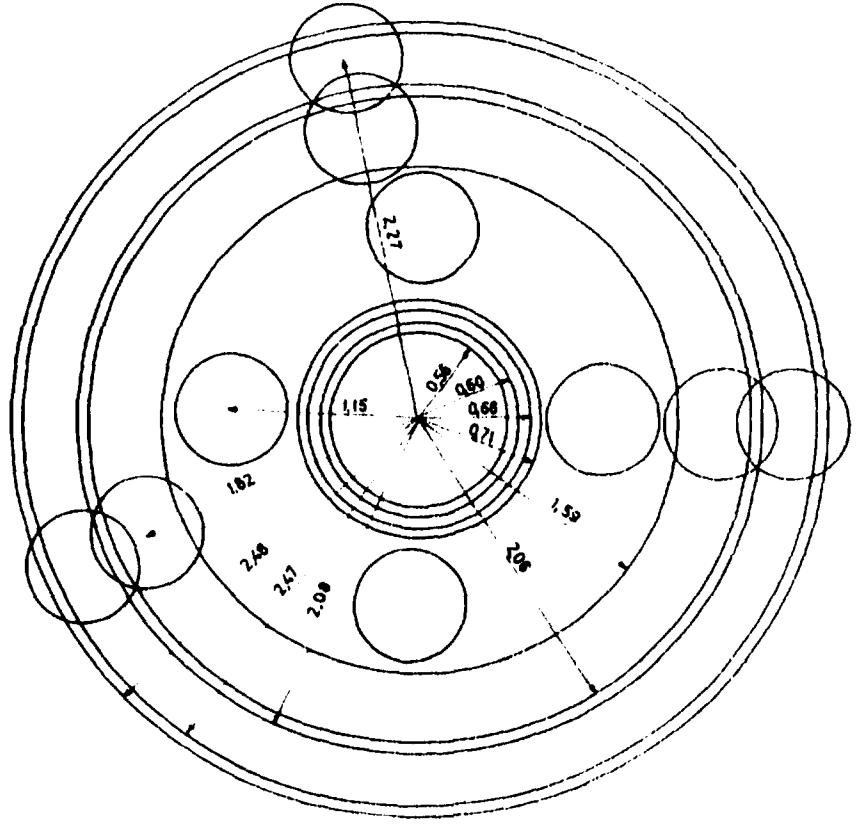

Figure 2. Schematic representation, in the "cluster" option of the WIMSD-4 code, of a portion of the fuel assembly assigned to one control rod

Table VI - Infinite medium criticality parameters versus burnup for a fuel assembly with and without integral type burnuble absorber

burnup (MWD/MTU)	with burnuble apsorber		no burnuble apsorber	
	k-inf	reactivity (pcm)	k-inf	reactivity (pcm)
0	.94765	-5523.75	1.36586	26786.05
150	.93415	-7019.61	1.31632	24030.60
4000	1.06837	6399.12	1.26539	20973.17
8000	1.12315	10965.01	1.21606	17766.88
12000	1.13187	11651.02	1.17163	14648.82
16000	1.11748	10513.18	1.13151	11622.21
22000	1.07833	7264.01	1.07721	7167.76
28000	1.03285	3180.33	1.02732	2659.35
34000	.98705	-1275.69	0.98079	-1259.06
40000	.94439	-5888.98	0. 3778	-6635.06

reactor in twodimensional R-Z or X-Y geometry by the finite difference method. As input data, the program uses precalculated tables of group parameters versus burnup, for different values of the reactor material temperatures and densities. The program calculates spatial distributions of neutron flux, power density and fuel burnup as functions of reactor operating time. Two options are available: (1) calculation of criticality parameters versus full power operating time or (2) calculation of control rod positions providing the given criticality conditions.

Effects of prolonged fuel burnup have been studied by considering fuel management scheme based on four 12 month cycles. At the begining of life (BOL) the equilibrium core thus contains 1/4, i.e. 48 fresh fuel elements, besides the once, twice and three times burned fuel. Assemblywise power and burnup distributions for hot and full power core with no control rods or burnuble absorbers, at BOL of an equilibrium cycle, have been calculated by the R-Z option of the program VAMPIR. The fresh fuel enrichment was 3.35% ^{235}U. The results given in Fig.3. show that in this case the maximum value of power form factor, i.e. assembly power/core averaged power, is very high (1.94).

One of the possible ways to perform power flattening is by insertion of control absorber rods. Here it is supposed that control rods are grouped in three banks moving independently. Using the option (2) of VAMPIR program the position of control banks is determined which considerably flattens power generation in the reactor core. The results are presented in Fig.4.,

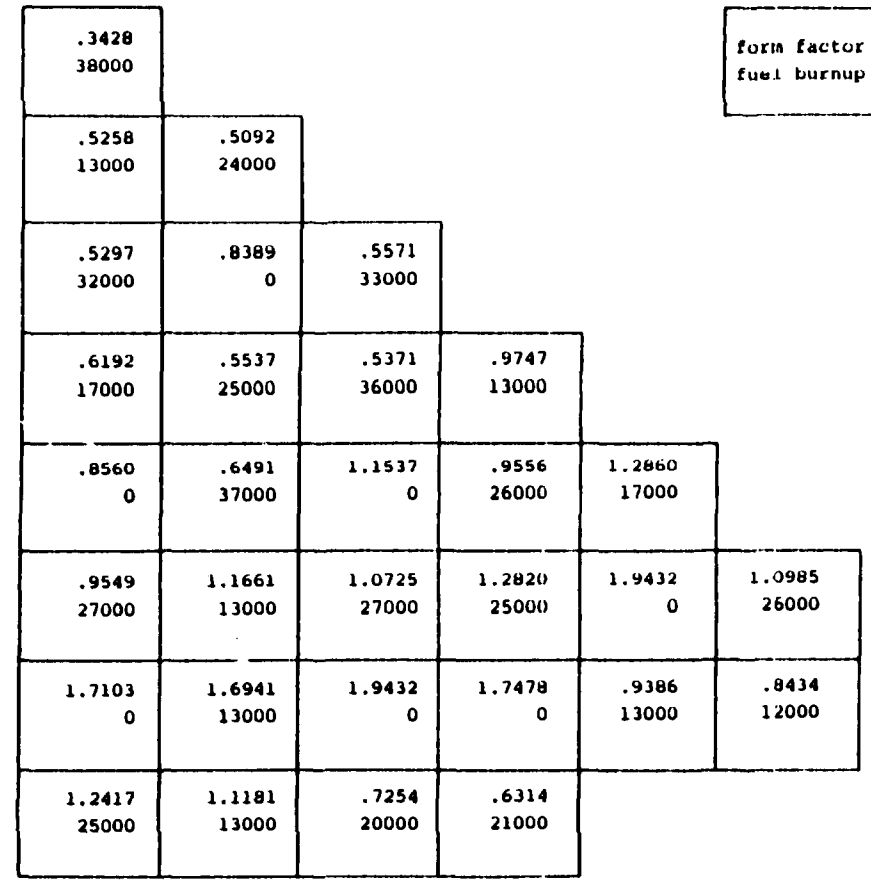

Figure 3. Assemblywise distribution of power form factors and burnup (MWD/MTU) for hot full power core, no control rods or burnable absorbers, BOL of an equilibrium cycle

.6763 38000 X1					form factor fuel burnup contr. bank
1.3433 13000	1.0125 24000				
.7480 32000	.6571 0	.9437 33000 X1			
.9648 17000	.9201 25000	.8058 36000	1.4411 13000		
1.0965 0 X2	.9261 37000	1.5013 0	1.1396 26000	.8524 17000 X2	
.8810 27000	.9691 13000	.6948 27000 X3	.6405 25000	1.4618 0	1.0337 26000 X1
.9449 0 X3	1.1681 13000 X3	1.2363 0	1.4435 0	.7269 13000 X1	.9508 12000
1.155 25000	.9978 13000	.8318 20000	.6754 21000		

Figure 4. Assemblywise distribution of power form factors and burnup (MWD/MTU) for hot full power core with inserted control rods, no burnable absorbers, BOL of an equilibrium cycle

.9759 38000 1					form factor fuel burnup zone number
1.4618 13000 1	1.3553 24000 1				
1.3239 32000 1	1.4734 0 3	1.1316 33000 1			
1.1442 17000 1	.8370 25000 1	.5709 36000 1	.6495 13000 1		
.6863 0 2	.4969 37000 1	.7222 0 2	.6723 26000 1	1.0327 17000 1	
.7613 27000 1	.9540 13000 1	.8790 27000 1	.9506 25000 1	1.2459 0 3	.9227 26000 1
1.2043 0 2	1.1961 13000 1	1.4414 0 2	1.3487 0 3	.8626 13000 1	.8423 12000 1
.9855 25000 1	.9833 13000 1	.6897 20000 1	.6176 21000 1		

Figure 5. Assemblywise distribution of power form factors and burnup (MWD/MTU) for hot full power core with integral type burnable absorbers, BOL of an equilibrium cycle

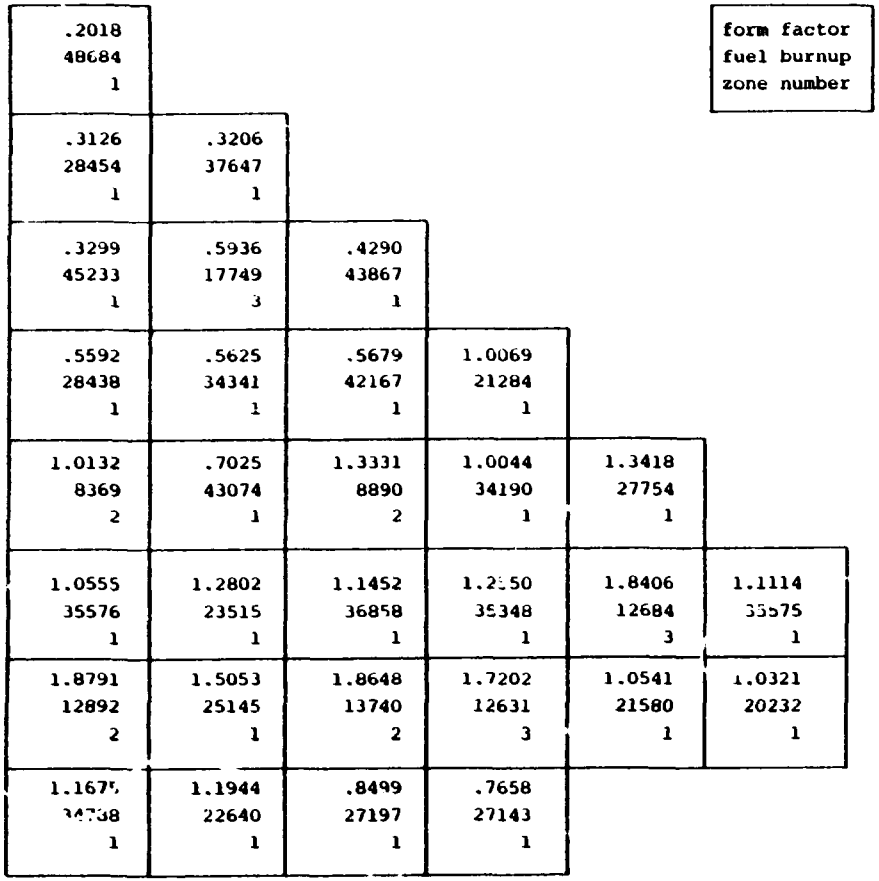

Figure 6. Assemblywise distribution of power form factors and burnup (MWD/MTU) for hot and full power core with integral type burnable absorbers, EOL of an equilibrium cycle

Table VII - Criticality parameters versus burnup for the equilibrium core with 1/4 (48) fresh fuel assemblies

enrichment 3.35% number of burnuble absorbers 0			enrichment 3.55% number of burnuble absorbers 24		
oper. time	reactivity (pcm)	burnup (MWD/MTU) average	oper. time (days)	reactivity (pcm)	burnup (MWD/MTU) average
0	9537.4	16608	0	6420.3	16607
100	4756.0	20060	100	2763.1	19988
200	1479.5	23512	200	1741.1	23367
280	-1152.8	26273	300	-822.3	26742

Table VIII - Redistribution of zone averaged power and zone averaged fuel burnup during the life of an equilibrium core containing burnuble absorbers (1/2 of the core)

	operating time of the reactor (days)				
zone	0	4.2	100	200	300
	zone averaged power (GW)				
1	.9555	.9699	.9542	.9281	.9261
2	.1205	.1208	.2151	.2457	.2553
3	.3363	.3216	.2430	.2385	.2309
total	1.4123	1.4123	1.4123	1.4123	1.4123
	zone averaged burnup (MWD/MTU)				
1	22105	22237	25259	28363	31383
2		94	2230	6200	10734
3		261	5895	10342	14706
average	16607	16751	19987	23366	26741

where X1, X2, and X3 denote control banks inserted up to 73.134 cm, 10.445cm and 20.895 cm from the bottom of the reactor vessel. Comparing to the previous case the range of the power form factors is considerably decreased (0.67 - 1.46 instead of 0.34 - 1.94).

In a PWR core control rods are primarily used to meet rapid transient reactivity requirements, as well as safety shutdown requirements. When the fresh fuel initial enrichment is increased in order to provide prolonged

fuel burnup. besides boron dissolved in the reactor coolant, burnable absorbers have to be used to control excess reactivity and avoid strong power peaking at the beginning of a cycle. In the present paper the so called integral type of burnable absorber, i.e. the combination of fuel and burnable absorber material in a single rod, is considered. About 80 percent of fuel pellets in the central part of a fuel rod are supposed to have thin (less than 2.5×10^{-3} cm) boride coating, while top and bottom portion of the fuel pellets are supposed to be uncoated in order to reduce axial peaking factors. In Fig. 5. calculated values of assemblywise distribution of power form factors and fuel burnup are presented for hot and full power core at BOL of an equilibrium cycle. The core configuration is supposed to consist of 145 fuel assemblies having initial enrichment 3.35% ^{235}U (zone 1), 24 fuel assemblies having initial enrichment 3.55% ^{235}U and integral type burnable absorber (zone 2) and 24 fuel assemblies having initial enrichment 3.55% ^{235}U and no burnable absorber (zone 3). In this case the range of power form factors is 0.61 - 1.46, i.e. approximately the same power flattening is achieved as in the case when groups of control rods have been used.

During the fuel irradiation boron is depleted, so that at the end of life its influence on the power distribution is practically eliminated. In spite of the fact that fuel depletion itself tends to flatten the flux and power distribution, power peaking can again become a problem at the cycle end. It can be seen in Fig. 6. that for the case treated here, the range of power form factors at end of life (EOL) is 0.2 - 1.86, i.e. almost as in the case presented in Fig. 3.

In Table VII criticality parameters versus burnup for the equilibrium core with lower fresh fuel enrichment and no burnable absorbers are compared with criticality parameters of the equilibrium core with higher fresh fuel enrichment and burnable absorbers. In the later case about 9% longer cycle, i.e. about 4.5% higher average discharge burnup, can be achieved.

Redistribution of zone averaged power and zone averaged fuel burnup during the life of an equilibrium core containing burnable absorbers is presented in Table VIII.

5. CONCLUSION

The purpose of the work presented here was to analyse the improved fuel designs and advanced in-core fuel management schemes in order to get elements for evaluating bids for future nuclear power plants, as well as to verify the available methodology and codes for burnup prediction. It can be concluded that different improvements in fuel design and fuel management, introduced in order to get prolonged fuel burnup and better fuel cycle economy, require more sophisticated calculational models and codes. Particularly desirable are faster codes which make possible more detailed and numerous calculations in the frame of different optimization procedures.

The results obtained also show that safety margins can be a limiting factor in achieving high burn-up values, even with the advanced fuel designs and fuel loading patterns. Methods and codes for coupled neutronic - thermodynamic calculations are thus important to perform detailed analysis of burnup related safety parameters.

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IAEA IN-CORE FUEL MANAGEMENT ACTIVITIES
(Summary)

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Background

In the early seventies, the Agency had organized lectures and seminars on reactor physics.

Because of interest shown by Member States in having comprehensive information concerning available computer programs applicable to the in-core fuel management of light and heavy water moderated power reactors, the Agency has carried out a survey during the late seventies. This resulted in the publication of the technical document "Computer Programs for the In-Core Fuel Management of Power Reactors", IAEA-TECDOC-250, in 1981.

In the framework of the IAEA/NENP activities on In-Core Fuel Management and related Core Physics Matters, this survey has been followed in the early eighties by a Co-ordinated Research Programme (CRP), with the objective to establish in those developing nations, seeking to use nuclear energy, a capability for in-core fuel management. This was done through the creation of co-ordinated sets of computer codes (i.e. complete computer packages) for performing extensive neutronic analysis. These computer codes included the determination of burnup and power distributions for complex operating histories and geometries for PWRs, BWRs and HWRs. These code packages have been reported in the IAEA TECDOC-314 entitled "In-Core Fuel Management Programs for Nuclear Power Reactors" and published in 1984.

After 1984, new developments have been made by some institutes in the program packages created under the previously mentioned CRP and new packages have been developed by other institutes.

Current Programme

The Advanced Nuclear Power Technology Section in the Division of Nuclear Power will convene a Technical Committee meeting and Workshop on Improvements of In-Core Fuel Management Codes at the Polytechnical University of Madrid in Spain, from 12 to 15 July 1988.

The purpose of the meeting is to provide an opportunity to review and discuss the current status of and recent progress made in in-core fuel management programs of PWR, BWR and PHWR; to update and complement those reported in the Agency's TECDOCs 250 and 314; to exchange experience with the code packages; to discuss their application; to explore their availability and to identify specific needs of developing countries.

Parallel to this meeting the ANPT Section is considering to initiate a Co-ordinated Research Programme (CRP) in order to set up appropriate test cases for checking fuel management program packages of PWR and BWR. In a second phase, it is considered to set up similar cases for PHWR. The programme of the CRP could consist of the following:

- specification of a set of appropriate and accurately measured parameters;
- compilation and provision of such a set of parameters; and
- computation of parameters and comparison with measured ones.

With respect to the above programme, the ANPT Section could make programs and results available to the IWGFPT. However, it should be noted that the programs do not deal with (detailed) fuel design but with general reactor core design. Of course, average and maximum pin peak powers can be calculated with the developed code packages, but they do not calculate a very fine power distribution within the fuel pins as requested by the IWGFPT.

Core design studies are also a long standing activity in the IWGFR (more than 20 years) and in the IWGGCR (eight years). More recently the IWGATWR has been set up, which covers design studies of water-cooled reactors.

The ANPT Section would hope to arrive at receiving advise/input for the proposed CRP on Benchmarks from the TC on Burnup Determination of Water Reactor Fuel and from the IWGFPT.

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