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# ESARDA Bulletin

NUMBER 20
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# News about ESARDA

## 14th Annual ESARDA Meeting (Restricted Participation)

Salamanca, Spain, 5-8 May 1992

The fourteenth Annual Meeting will be held in the old University "Paraninfo", Salamanca, Spain.

Attendance will be limited to the ESARDA Steering Committee members, working group members and observers, coordinators and a few experts invited to the workshops (see presentation at page 3).

**In memorian of Jean-Marie Leblanc**

ESARDA has the sad duty to announce here the passing away on Sunday 1st September 1991, at the age of 59, of Mr. J.M. Leblanc, member of the ESARDA Steering Commitee.

Mr. Leblanc was one of the pioneers of BELGONUCLEAIRE in Belgium where he started as young civil engineer in 1958. He was Manager of the MOX fuel fabrication plant at Dessel and also Deputy Director General of BELGONUCLEAIRE.

We all knew him as a very dynamic man who gave all his energy, even during his illness, to "his" Plutonium Plant. He was also known in the nuclear world as one of the pioneers of MOX fuel.

The ESARDA members would like to present their sincere condolescence to his family and his collegues at BN.

Corrigendum: The editor of the Bulletin wishes to apologize for the fact that in the table of the ESARDA members published in issue No. 19 the name of Dr. Weh disappeared from the list of the ESARDA Steering Commitee. He cannot understand how this happened but hopes that Dr. Weh has forgiven him.

ESARDA wishes to thank Miss M. Neuilly. She has now retired but she kept operating up to the end very efficiently as French Coordinator in ESARDA. She also participated in several ESARDA Working Groups and always with great competence and efficiency. We shall miss her. As a last task she greatly contributed to the organization and the succes of the Avignon Symposium.

We do not mention one by one all the changes in the ESARDA membership for the Steering Committee and all the other groups. The reader is kindly invited to note that there are several changes in the table "Who's who in ESARDA?" published on the next page.

## 15th ESARDA Symposium

Italy, 11-13 May 1993

The fifteenth Annual Meeting will be a general ESARDA Symposium on Safeguards and Nuclear Material Management. It will be held in Italy, probably in Rome.

Attendance and contributions to the ESARDA Symposium will be open to people from all the world.

# Who's Who in ESARDA?

(as of 1st December 1991)

Chairman 1991 *B.H. Patrick*, AEA Technology, Harwell, U.K.
Appointed chairman 1992 *P. Frederiksen*, Risø, Denmark
Secretary *C. Foggi*, CEC, JRC-Ispra, Italy
Permanent Symposium
Scientific Secretary *L. Stanchi*, CEC, JRC-Ispra, Italy

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*F. Pozzi*, ENEA Saluggia, Italy
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*A. Velilla*, CIEMAT, Spain
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*R. Carchon*, CEN/SCK Mol, Belgium
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*R.J.S. Harry*, ECN Petten, Netherlands
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*H. Lefèvre*, CEA Fontenay-aux-Roses, France
*R. Schenkel*, CEC, Safeguards Directorate, Luxembourg

**Working Group Convenors**

Techniques and Standards for Non-Destructive Analysis (NDA)
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Techniques and Standards for Destructive Analysis (DA)
*P. De Bièvre*, CEC, JRC-Geel, Belgium
Reprocessing Input Verification (RIV)
*T.L. Jones*, AEA Technology, Dounreay, U.K.
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*P.P.A. Boermans*, FBFC, Belgium
MOX Fuel Fabrication Plants (MOX)
*G. Le Goff*, CEA Paris, France

**ESARDA Bulletin Editor**

*L. Stanchi*, CEC, JRC-Ispra, Italy

# 14th Annual ESARDA Meeting

## 5-8 May 1992, Salamanca, Spain

**B.H. Patrick**
AEA Technology
Harwell Laboratory, United Kingdom

**L. Stanchi**
Commission of the European Communities
JRC Ispra, Italy

It was previously planned that this meeting would be held in Lanzarote, Canary Islands, on 12-15 May 1992. This site was chosen by the CIEMAT administration because of the large choice of rooms which completely fulfilled the needs of the Working Groups and the planned sub-groups, in addition to a well equipped plenary room and also because it is very well served by air connections from all the European countries at prices which in May are particularly attractive.

However, because of perceived presentational difficulties and in order to give participants the opportunity to visit a nuclear facility, the Steering Committee invited the Spanish Authorities to reconsider the choice of venue.

We should now like to thank the CIEMAT administration. They had to make additional and very great efforts to find quickly a new place satisfying the stringent conditions of a complex meeting associated with all the Working Group meetings. In addition they were faced with the fact that in 1992 there are other important events at the national level which prevent us from using most of the hotel and congress facilities. We greatly acknowledge the persistence, goodwill and dedication of the CIEMAT administration in reaching a satisfactory alternative solution.

The venue is now fixed in Salamanca, but, because of other engagements, the date had to be brought forward a week and the meeting will be held from 5 - 8 May 1992.

Salamanca is a cultural and historical town, capital of the homonymous province, and is situated on the right bank of the river Tormes.

Salamanca is a university town which has maintained its ancient character, its history being evident from its illustrious monuments. It is proud of the memory of its doctors who emanated from its famous university, the oldest in Spain.

Salamanca was firstly a Roman fortified town, which fell at the end of the Roman empire under the invasion of the Visigoths. All the traces were then eliminated by the Arab domination. Salamanca was finally taken over in the XIth century by Raimundo de Borgoña on behalf of King Alfonso VI. He operated an efficient policy of repopulating the region which had become nearly abandoned.

Christian peoples of different origins settled in the area. Jerónimo de Perigord received the care of the church of Salamanca from Raimundo de Borgoña. He became the first bishop of Salamanca and helped greatly to gather those peoples into a community. They in turn supported the construction of the church, the university and the setting up of a defence Army. Salamanca rapidly became a cross-roads for all the sciences and arts.

The town possesses two cathedrals, built side by side. The older of them was started in the XIIth century in pure Romanesque style. Both churches have a great number of artefacts which are worth a visit. The old cathedral contains the image of the "Virgen de la Vega", the patron saint of Salamanca, one of the most famous treasures of Spain. The cathedral was modified and extended in the following centuries. The new cathedral had a lengthy period of construction. Initiated in 1513, it was finished in 1733 and can be considered as one of the last Gothic buildings.

**Fig. 1:** Salamanca from the river Tormes.

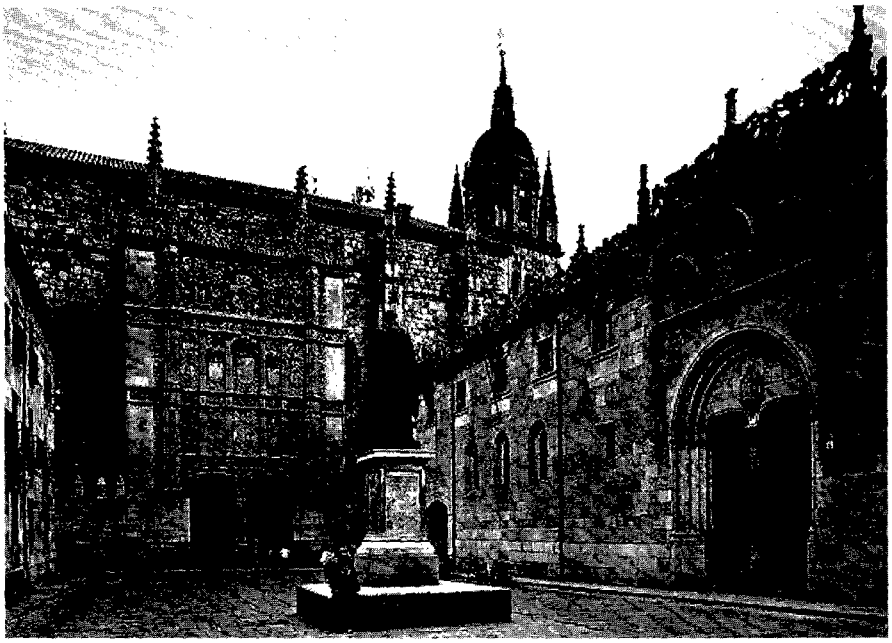

**Fig. 2:** The entrance of the old university.

Salamanca has three universities. The most ancient, called Paraninfo, is dated 1254 and is not only the oldest in Spain but also one of the oldest in the world.

The 14th Annual ESARDA Meeting will be held in this university, which is no longer used for studies but only as a conference and cultural centre. This meeting will be based on two workshops organized by the C/S and NDA Working Groups respectively. The titles of the two workshops are:

- C/S Safeguards Techniques Applicable to the Intermediate and Long Term Storage of Irradiated Fuels
- Non-Destructive Assay Techniques Applicable to Safeguarding Nuclear Materials in Wastes.

The workshops will run from Tuesday to Thursday (5-7 May 1992) and will include plenary and sub-group sessions. In parallel the WGs not directly involved in the workshops will meet in the same place as follows:

- from Tuesday to Thursday (5-7 May): DA
- from Wednesday to Thursday (6-7 May): LEU, MOX, RIV.

A normal meeting of the Steering Committee will also be organized on Thursday.

The conclusions of the two workshops will be presented to the Steering Committee in an extended plenary session on the morning of Friday 8 May. This extended plenary session will involve all the workshop participants (C/S WG and NDA WG members together with the invited experts), the ESARDA Coordinators and also all the other WG members.

The official language of the workshop will be English. No simultaneous translations are planned.

A technical visit to a nuclear factory will be organized for all interested participants on Friday afternoon.

The 14th ESARDA Meeting is the fourth in the series of restricted meetings which in recent years have alternated with the full ESARDA Symposia on Safeguards and Nuclear Material Management with open participation. The preceding restricted meetings were held in Copenhagen in 1986, in Karlsruhe in 1988, in Como in 1990. In 1993 a general ESARDA Symposium will be organized with the assistance of ENEA in Italy, probably in Rome.

Included with this article are some photographs to illustrate this presentation of the Salamanca meeting. We wish to draw attention to the school room of Fray Luis de León, a philosopher of the sixteenth century, who was imprisoned by the Inquisition for his ideas. Recognized as innocent after five years of jail he returned to his class-room and started the lesson with the famous "heri dicebamus", "yesterday we were saying"...

**Fig. 3:** The class-room of Fray Luis de Léon.

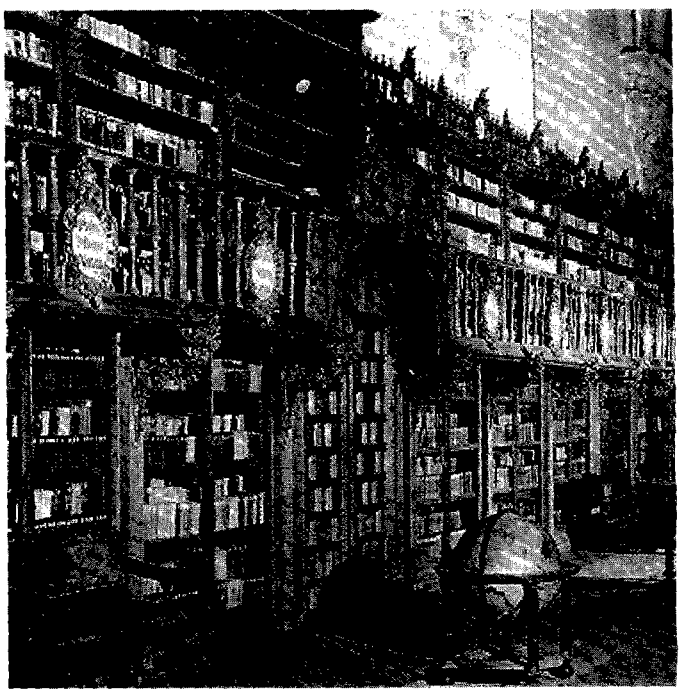

**Fig. 4:** The library of the old university.

# The 13th ESARDA Symposium

## Avignon, France, 14-16 May 1991

**B.H. Patrick**
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**L. Stanchi**
Commission of the European Communities
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Eight years after the Versailles Symposium, ESARDA returned to France to hold the 13th ESARDA Symposium. The former was held in a modern, purpose-built congress palace, while the latter was hosted in the magnificent premises of the Palace of the Popes, in surroundings reminiscent of the middle ages. If it was less suitable for hosting a symposium, the enchantment of the centuries of history escorting the participants compensated for all the small problems that such a palace, not created for symposia, could cause.

Moreover, the magnificent architecture of the Palace and its annexes, the old town and the surroundings were so beautiful that we think all participants left this part of Provence with marvelous memories and the desire to return to pay a longer visit.

The symposium had about 220 participants from many regions of the world. The general title of the meeting, "Safeguards and Nuclear Material Management", enabled the complete spectrum of safeguards topics to be covered, ranging from the political and regulation aspects, to all the technical areas which contribute to the subject.

The plenary sessions were held in the Room of the Conclave, as said above, which provided a magnificent setting for the discussions. The presentations were divided into 11 oral sessions and 6 poster sessions, with 60 presentations in the former and 66 in the latter.

The large number of papers in total and more specifically in the field of measurements shows that there is great interest in this field again, both in Nondestructive assay and in Destructive Analysis.

The considerable participation of Eastern European countries is a remarkable fact. There were several papers and 6 participants from the USSR, with appreciated contributions. We also had papers and participants from Czechoslovakia, Hungary and Poland, countries which are candidates to join an important European initiative, i.e. COST (Scientific and Technical Co-operation) which has the aim of promoting research in Europe on a transnational basis.

In the initiatives which are beginning to be brought forward on energy policy in the European Community, nuclear power is expected to continue to play an important role in which safeguards are an essential ingredient. They will also be of great importance in the increase in nuclear trade which could result when the open market comes into being at the beginning of 1993.

An important landmark was reached recently when fresh MOX fuel was verified for the first time under water by the Euratom Inspectorate. The anticipated increase in the flow and use of MOX in the future makes it essential to devise and demonstrate successful safeguards measures which can give the necessary assurance. The ability to verify under water is a key part of these measures.

A number of organisations are working on schemes for randomisation of inspections or zone approaches. Mr. Gmelin remarked that while the elegance of the techniques is unquestionable, the practice is not so encouraging. We suspect this is the first shot in a discussion that will last for some time and we look forward to seeing the outcome of the argument.

The advances in NDA techniques in recent years have been remarkable. In some areas, they are beginning to approach capabilities so far achieved only by DA techniques and this can only be good for safeguards. We would not be surprised if this infringement of the prime position held by the DA community stirs them into even greater efforts, resulting in further improvements in their armoury. This rivalry is a great stimulating influence which will inevitably lead to improved accuracy and therefore improved safeguards.

Recent developments in C/S techniques seem to have eliminated the Cinderella image which it

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has had. The application of modern, sophisticated techniques has brought about a revolution and raised the status of C/S very significantly. Data are now of higher quality which leads to greater assurance. This is a much-needed development because, with the limitation on inspection resources making it difficult to keep pace with the increasing number and complexity of nuclear installations, greater reliance on C/S is required. C/S seems to have risen to the challenge and those responsible for the advance are to be congratulated.

The participation of Eastern European countries in this Symposium is a very welcome addition. Both sides have much to offer and can learn from each other. I hope their participation is a trend which will continue to the benefit of all.

If we had to select only one highlight, it would have to be the trend towards integration. Integration has been with us for a long time but we lost count of the number of times the words "integrated system" were used by presenters. More and more, complete systems are called for, requiring contributions from several technical areas. Besides producing better, more reliable, safeguards systems, integration brings together personnel involved in different disciplines and this must be an additional benefit. Using the word "integration" in a looser sense, we have been struck by the number of papers involving authors from different organisations, not only R&D ones but also the inspection agencies. Safeguards is an international business and collaboration adds to this dimension and is therefore to be encouraged.

During the Symposium, a dinner was hosted by EdF, COGEMA and CEA in La Salle de la Grande Audience, a superb Gothic building. Both the food and the spectacular entertainment will long be remembered by those who were present. The delegates were also invited by the Deputy Mayor of Avignon to a reception in the Salle des Fêtes de la Mairie where some of the region's wines were experienced in delightful surroundings.

# Calculation of Passive Neutron Emission from Spent Fuel of WWER-440 and WWER-1000 Reactors

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## 1. Introduction

Spent fuel elements from nuclear power plants form a considerable portion of the nuclear material, whose peaceful use in the frame of international safeguards has to be reliably verified by national and international institutions. Counting of fuel elements, application of C/S-measures and, if occasion arises, the application of Cerenkov Viewing Devices comprise necessary conditions for the attainment of inspection goals. In the case of particular circumstances, as for instance long cooling times, or a poor visibility in the spent fuel ponds, or a reverification of fuel in case of conclusive negative C/S, or the verification of spent fuel prior to its becoming difficult-to-access, specific verification methods should be available to even then attain the admitted verification goals.

In this connection, the measurement of passive neutron emission is a method which can be used at comparatively low technical expenditure to carry out even partial defect measurements. However, a well-founded theoretical basis is necessary for the interpretation of the measurement results because the measured neutron signal in a complicated manner is connected with the fuel parameters of interest.

The present article summarizes the results of respective theoretical investigations carried out for spent fuel from Soviet power reactors of the types WWER-440 and WWER-1000 /1,2,3/.

## 2. Structure of WWER-440 and WWER-1000 Fuel

Pressurized water reactors of the WWER type (light-water-moderated, light-water-cooled energy reactor) utilize fuel elements having a hexagonal cross-section. The WWER-440 core is composed of 349 fuel elements, of which 37 are vertically movable to make possible the control of reactor power by means of absorber elements connected with them. Owing to the use of such compact control elements comparatively steep neutron flux gradients can arise at such positions.

In contrast to this, the WWER-1000 uses cluster rods for power control. WWER-1000 fuel elements, of which 163 are contained in the core, furthermore differ from WWER-440 elements by their larger dimensions and by the missing fuel element jacket. Some of the types of WWER-1000 fuel elements for purposes of neutron flux flattening have lower enriched fuel in the peripheral row of rods.

The following tables give a survey of some parameters of WWER-440 and WWER-1000 fuel including the thermohydraulic state variables used in the computations (Table I). Tables II and III summarize the different types of fuel elements existing for the WWER-440 and the serial WWER-1000.

Fuel element types K16 and R16 are exclusively used in the initial loading of WWER-440. Types K24, R36 and, as a function of the planned refuelling cycle, some elements of type R24 are exploited in the equilibrium core.

The lower enriched fuel element types 1 and 2 are used in the initial loading of WWER-1000 reactors. Types 7 and 8 or 9 and 10 are used in the intended equilibrium regime of the two-year and three-year cycle, respectively. The other fuel element types are used during the reactor cycles until the equilibrium state is attained.

**Table I**

| | WWER-440 | WWER-1000 |
|---|---|---|
| lattice pitch (mm) | 147 | 236 |
| core compositions | 349 | 163 |
| fuel element: | | |
| width over flats (mm) | 144 | 232 |
| lattice positions | 127 | 331 |
| fuel rods | 126 | 312 |
| central channel | water filled | water filled |
| cluster channels | – | 18 |
| thermohydraulic parameters: | | |
| fuel temperature (K) | 970 | 1000 |
| moderator temperature (K) | 558 | 578 |
| moderator density ($g/cm^{-3}$) | 0.754 | 0.7144 |
| linear fuel rod power (W/cm) | 125 | 166 |
| boric acid concentration in moderator ($gH_3BO_3/kgH_2O$) | 4 | 3 |

**Table II - Fuel element types of WWER-440**

| Type* | initial enrichment (wt.-%) | maximum burnup (GWd/tU) |
|---|---|---|
| K16 | 1.6 | 20 |
| K24 | 2.4 | 30 |
| R16 | 1.6 | 20 |
| R24 | 2.4 | 30 |
| R36 | 3.6 | 40 |

* K - fuel part of control element (wt. of uranium 114.5 kg)
R - normal fuel element (wt. of uranium 119.5 kg)

## 3. Method of Calculating the Rate of Passive Neutron Emission

The neutron radiation of spent and unloaded fuel is caused by the decay of actinides. This decay proceeds on the one hand through spontaneous fission, as a result of which primary fast neutrons are emitted. The half-lives for spontaneous fission vary largely from nuclide to nuclide but drop systematically with increasing mass numbers. On the other hand, fast neutrons are

**Table III - Fuel element types of WWER-1000**

| Type | initial enrichment (wt.-%) | maximum burnup (GWd/tU) | type of cluster heterogeneity |
|---|---|---|---|
| 1 | 2.0 | 30 | $H_2O$ |
| 2 | 2.0 | 30 | $B_4C$ |
| 3 | 3.0 | 45 | $H_2O$ |
| 4 | 3.0 | 45 | $B_4C$ |
| 5 | 3.3 | 45 | $H_2O$ |
| 6 | 3.3 | 45 | $B_4C$ |
| 7 | 3.3 (3.0) | 45 | $H_2O$ |
| 8 | 3.3 (3.0) | 45 | $B_4C$ |
| 9 | 4.4 (3.6) | 60 | $H_2O$ |
| 10 | 4.4 (3.6) | 60 | $B_4C$ |

emitted as a secondary process of the reaction of $\alpha$-particles with the oxygen isotope $O^{18}$ of uranium dioxide [($\alpha$,n)-process)]. The half-lives of $\alpha$-emitting actinides lie, of course, considerably below those of spontaneous fission, but the effective neutron yields in the ($\alpha$,n)-processes are by about 8 orders of magnitude smaller than those of spontaneous fission.

In neutron production, the ($\alpha$,n)-process plays a decisive role only at low burnup. The contributions of individual actinide isotopes to the neutron emission rate $Q_n$ are thus highly different owing to the differing half-lives and neutron yields of actinides, with their content in spent fuel being a complicated function of burnup (BU), irradiation history, initial fuel parameters and decay time (cooling time $t_c$). Therefore, a definite and simple functional relation between $Q_n$ and BU in the form of $Q_n = f(BU)$ cannot be expected to exist.

According to this, the calculation of the passive neutron emission rate has to be carried out in two steps:

1) Calculation of the actinide content of the nuclear fuel as a function of burnup, allowing for various operating conditions;
2) Calculation of the neutron emission rate in connection with a given fuel composition.

### Actinide Content

Actinide inventory as a function of specific burnup is generally calculated in two separate steps:

1) Calculating those isotopes which mainly determine the neutron-physical properties of the fuel lattice, in particular the macroscopic cross-sections and the neutron spectrum;
2) Calculating those isotopes which are present in concentrations too low to have any obvious influence on the macroscopic cross-sections and the neutron spectrum.

In the case of uranium fuel the first group includes U-235, U-236, U-238, Pu-239, Pu-240, Pu-241, and Pu-242. With the exception of Pu-241, they can be considered stable for the purpose of deriving practical numerical results owing to their long half-lives. From this it follows that their concentrations explicitly only depend on the specific burnup but not on the time in which a given burnup is attained. For calculations including these isotopes the spectrum- and burnup code NESSEL-4 /6/ was used.

The second group mainly includes all transplutonium elements (americium, curium, californium), but in addition also Pu-238, U-237 and Np-237 whose concentrations are very low, too. In the calculation of neutron emission, however, this group has to be given particular attention. Since some of these isotopes have shorter half-lives, their concentrations depend not only on burnup but also more or less on the irradiation history. Hence, in computations with these nuclides, their time dependence has to be explicitly taken into account.

In the corresponding differential equations, the above-mentioned group (1) of nuclides appears as a source term. These equations are:

$$\frac{dC_i}{dt} = -\left(\lambda_i + \hat{\sigma}_i^a \hat{\Phi}\right) \cdot C_i + \sum_{j=1}^{z} \left(\lambda_j \cdot g_{ij} + \right.$$

$$\left. + \hat{\sigma}_j^c \cdot f_{ij} \cdot \hat{\Phi}\right) \cdot C_j + \hat{S}_i \qquad (1)$$

where

$$\hat{S}_i = \sum_{j=1}^{z_0} P_{ij} \cdot \hat{\sigma}_j^{o,x} \cdot \hat{\Phi} \cdot C_j^o$$

is the source term of the inhomogeneous equation system.

In detail, the following nomenclature is used:

- $C_i$ nuclear concentration of (second group) nuclide i
- $z$ number of nuclides to be treated simultaneously (i = 1, ..., Z)
- $t$ time
- $C_j^o$ nuclear concentrations of group 1 isotopes (j = U-235, U-236, ..., Pu-242)
- $\lambda_i$ decay constant of nuclide i
- $\hat{\sigma}_i^c$ effective one group capture cross-section of nuclide i
  $$\hat{\sigma}_i^c = \int \sigma_i^c(E) \cdot dE / \hat{\Phi} \qquad (2a)$$
- $\hat{\sigma}_i^a$ effective one group absorption cross-section of nuclide i
  $$\hat{\sigma}_i^a = \int \sigma_i^a(E) \cdot \Phi(E)\, dE / \hat{\Phi} \qquad (2b)$$
- $\hat{\sigma}_j^{o,x}$ effective one group cross-section for group 1 nuclides [x = c or x = (n, 2n)].
- $\hat{\Phi}$ total flux in fuel
  $$\hat{\Phi} = \int_o^\infty \Phi(E)\, dE \qquad (2c)$$
- $g_{ij}$, $f_{i,j}$ transition probabilities for transmutation of nuclide j to nuclide i by decay or by capture
- $p_{ij}$ 1 if $C_i$ arises from $C_j^o$
  0 otherwise.

The neutron spectrum is calculated using the computer code NESSEL-4 in a 34-group model (10 thermal and 24 epithermal energy groups). Accordingly, the integrals (2a), (2b) and (2c) are represented as

$$\hat{\sigma}^{c,a} = \sum_{g=1}^{34} \sigma_g^{c,a} \cdot \Phi_g / \hat{\Phi}$$

$$\hat{\Phi} = \sum_{g=1}^{34} \Phi_g$$

c = capture; a = absorption

The 34-group cross-sections $\sigma_g^{c,a}$ are considered burnup-independent data sets, while the 34-group neutron spectrum depends on burnup. Thus, all cross-sections occurring in the equation system (1) depend on burnup. The solution of equations (1) which can be written in the compact form

$$dC/dt = M \cdot C + Q$$

(M: Z x Z - coefficient matrix) (2d)

is obtained by a matrix exponential development of

$$C(t) = \exp[(t-t_o) \cdot M]\,(C(t_o) + (\exp[(t-t_o) \cdot M] - E) \cdot M^{-1} Q$$

(E = identity matrix).

The time-dependent nuclide concentrations and the parameters derived from them are calculated by means of the computer code NUKO /4/.

This basic approach can similarly be applied to both the WWER-440 fuel and to the WWER-1000 fuel. But, for calculating the nuclide inventory of a concrete fuel element type it is necessary to define the integration area for the above differential equation. As regards the WWER-440 fuel, all fuel rods can be described by a uniform fuel rod cell being composed of the fuel pellet, the zirconium cladding tube and an adjacent, effective water region. Besides this fuel area, in the NESSEL-4 calculations the central channel of the fuel element (water-filled zirconium tube) and the peripheral jacket of the fuel element (zirconium jacket with adjacent interior and exterior water region) are treated as specific areas.

Due to the existence of the cluster channels and the deviating enrichment of some outer fuel rods, for the WWER-1000 it is no longer possible to define one uniform fuel rod cell. Rather the fuel area has to be subdivided into three different zones for which the relationship between nuclide concentration and burnup has to be individually calculated.

In this subdivision, zone 1 comprises all fuel rods in neighbourhood and thus in close physical interaction with cluster channels. Zone 3 comprises the fuel rods of the marginal belt and some other corner rods; these are subjected to the influence of spectral disturbance of the external medium between fuel elements. Finally, zone 2 covers all remaining fuel elements in the intermediary, more or less undisturbed area of the fuel element.

For practical use, of course, particularly the mean values of all parameters computed over the entire fuel element cross-section are of interest, as a function of the means of burnup. These means have to be established then from the values of the individual zones by weighting with the corresponding numbers of fuel rods.

### Passive Neutron Emission

Having calculated the concentrations of actinide isotopes, passive neutron emission caused by these isotopes can be calculated by a simple summation formula:

$$q_i = C_i \left(\lambda_{sp}^i \cdot \nu_{sp}^i + \lambda_a^i \nu_a^i\right) \qquad (3)$$

where

- $C_i$ nuclear concentration of nuclide i [$cm^{-3}$]
- $\lambda_{sp}^i$ decay constant of spontaneous fission,
- $\gamma_{sp}^i$ neutron yield of spontaneous fission,
- $\lambda_\alpha^i$ decay constant of $\alpha$-decay
- $\gamma_\alpha^i$ effective neutron yield per $\alpha$-decay (dependent on nuclear material).

The overall neutron emission rate in a fuel element follows by summation of $q_i$ over all actinide nuclides

$$Q_n = \sum q_i \; (cm^{-3} \cdot s^{-1})$$

The constants in equation (3) for calculating neutron emission are taken from literature /5/.

For a WWER-1000 fuel element, of course, this summation at first has to be carried out for each of the three zones. Then, these three values have to be summed up to get total passive neutron emission acccording to the following formula, where w(z) are weighting factors resulting from the shares of fuel rods in the three zones:

$$Q_n = \sum_{z=1}^{3} w(z) \cdot Q_n(z)$$

[w(1) = 120/312, w(2) = 114/312, w(3) = 78/312]

## 4. Calculation Results for the Passive Neutron Emission of WWER-440 and WWER-1000 Spent fuel

In order to make possible a comparison between different types of fuel and to investigate the influence of possible changes of parameters in the boundaries of normal operating conditions, for each of the reactor types a set of reference conditions was defined. They include the thermohydraulic parameters given in Table I and the time stability of these parameters until the final burnup of the fuel element is attained. This continuous irradiation period is followed by the decay period during spent fuel storage. These irradiation and decay conditions in the following are called the nominal state.

Since neutron emission is a direct result of the respective concentrations of the plutonium and transplutonium isotopes which depend on a large number of parameters, only very few general remarks are possible on the course of neutron emission:

- The number of primary passive neutrons emitted per unit of volume and unit of time is very small. Even at high fuel burnup a value of $2x10^4$ $cm^{-3} \times s^{-1}$ is not exceeded.
- Neutron emission is a strongly increasing function of burnup because the dominant emitters, the curium isotopes, are formed only after 5 - 6 conversion steps.
- After unloading spent fuel elements from the reactor core, during the following 1 - 2 years neutron emission comparatively strongly decreases because of the decay of Cm-242.

These effects are especially pronounced for fuel elements having an unloading burnup above 20 GWd/tU which will everytime be the case under normal operating conditions of the equilibrium core.

To illustrate the principal relationship between neutron emission and burnup, Fig. 1 shows this relationship for WWER-440 fuel elements ($t_c = 0$). From Figs. 2, 3 and 4 the contributions of individual actinide isotopes to total neutron emission during decay can be seen for different unloading burnups (WWER-440, fuel element type R36 as an example).

Since the calculation of neutron emission was carried out for the nominal state and the evaluation of measurement data will start from these results, the justification of the application of nominal state data was investigated by determining the influence of deviations from the nominal state on neutron emission.

With regard to changes of neutron emission as a result of variations of thermohydraulic parameters, the following conclusions can be drawn:

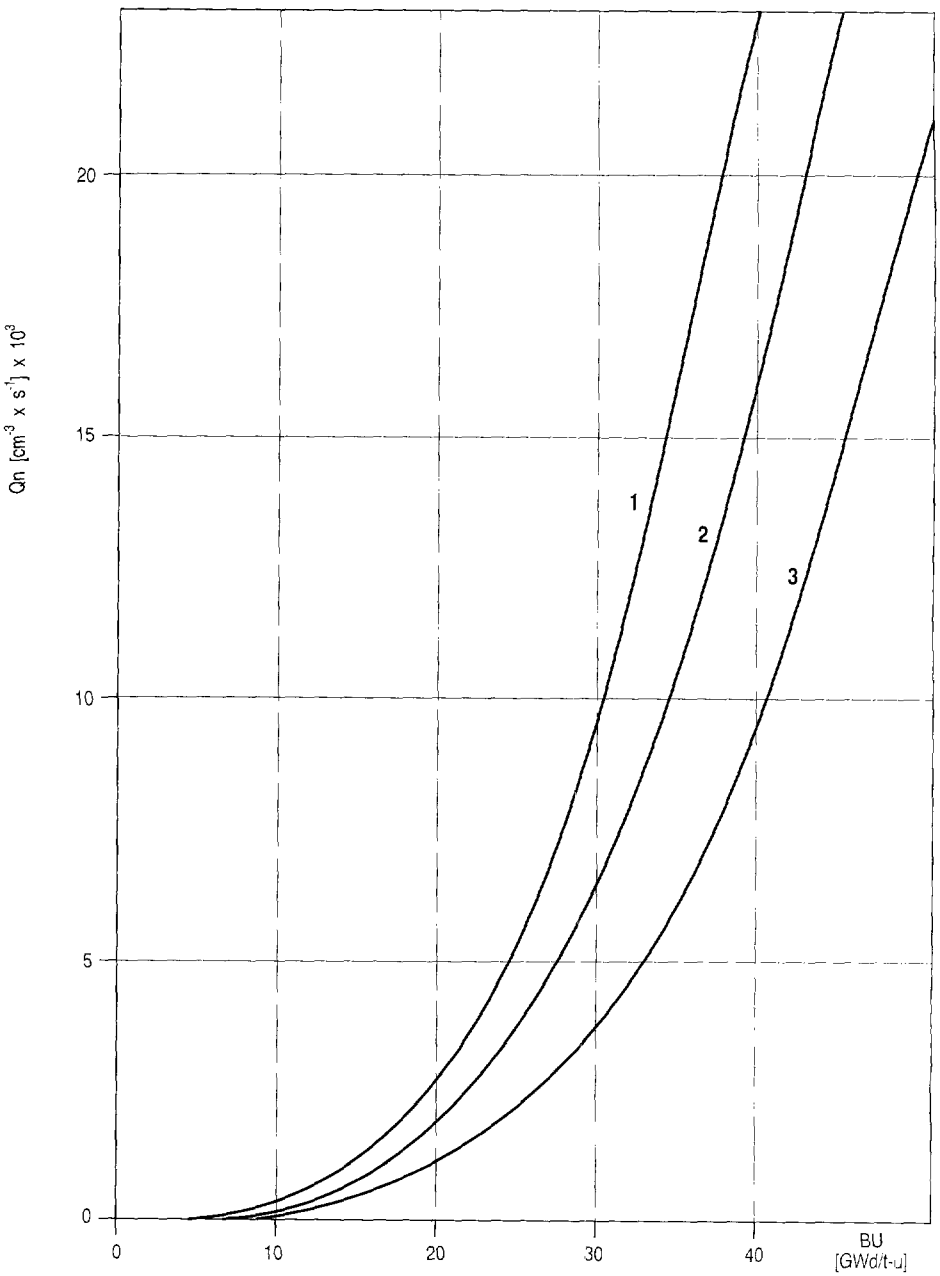


**Fig. 1:** Primary Neutron Source Strength as a Function of Burnup for WWER-440 Fuel Element Types
1) K16, R16 2) K24, R24 3) R36.

### Fuel temperature $T_F$

Inaccuracies in the determination of $T_F$ have a small effect on neutron emission (inaccuracies of $T_F$ in the range of 20% produce a change of the emission rate in the order of 1%).

### Moderator temperature $T_M$

As for $T_F$-inaccuracies, the genuine moderator temperature effect, i.e. a change of $T_M$ without a corresponding change of moderator density, has also a small influence on $Q_n$ which can be neglected under normal operating conditions.

### Moderator density $d_M$

Changes in moderator density, even in the range caused by local deviations under normal operating conditions, can produce comparatively large changes in neutron emission (normal operating conditions: mean coolant heat-up 30 K, resulting difference in $d_M$ at $T_M$ = 558/578 K: $\Delta d_M$ = ~ 6/10%). This is a result of changing effective cross-sections caused by the changing neutron spectrum.

Since the thermal cross-section of the predominant fissile isotope U-235 decreases with a harder spectrum, larger flux times are required to obtain a certain specific burnup. So, the conversion of U-238 into Pu-239 and the build-up of the following isotopes into the transplutonium isotopes mainly responsible for neutron emission (Cm-242, Cm-244) will also increase. A competing effect is the reduction of the capture cross-section of Am-243 in a harder spectrum which results in a reduced production of Cm-244 in very hard spectra.

As a result, at a constant density difference, the resulting changes of $Q_n$ will decrease with increasing burnup because of the growing contribution of Cm-244 to total neutron emission. Generally speaking, in the range of normal unloading burnups a change in $d_M$ will effect a change in $Q_n$ which has the same magnitude, but a different sign.

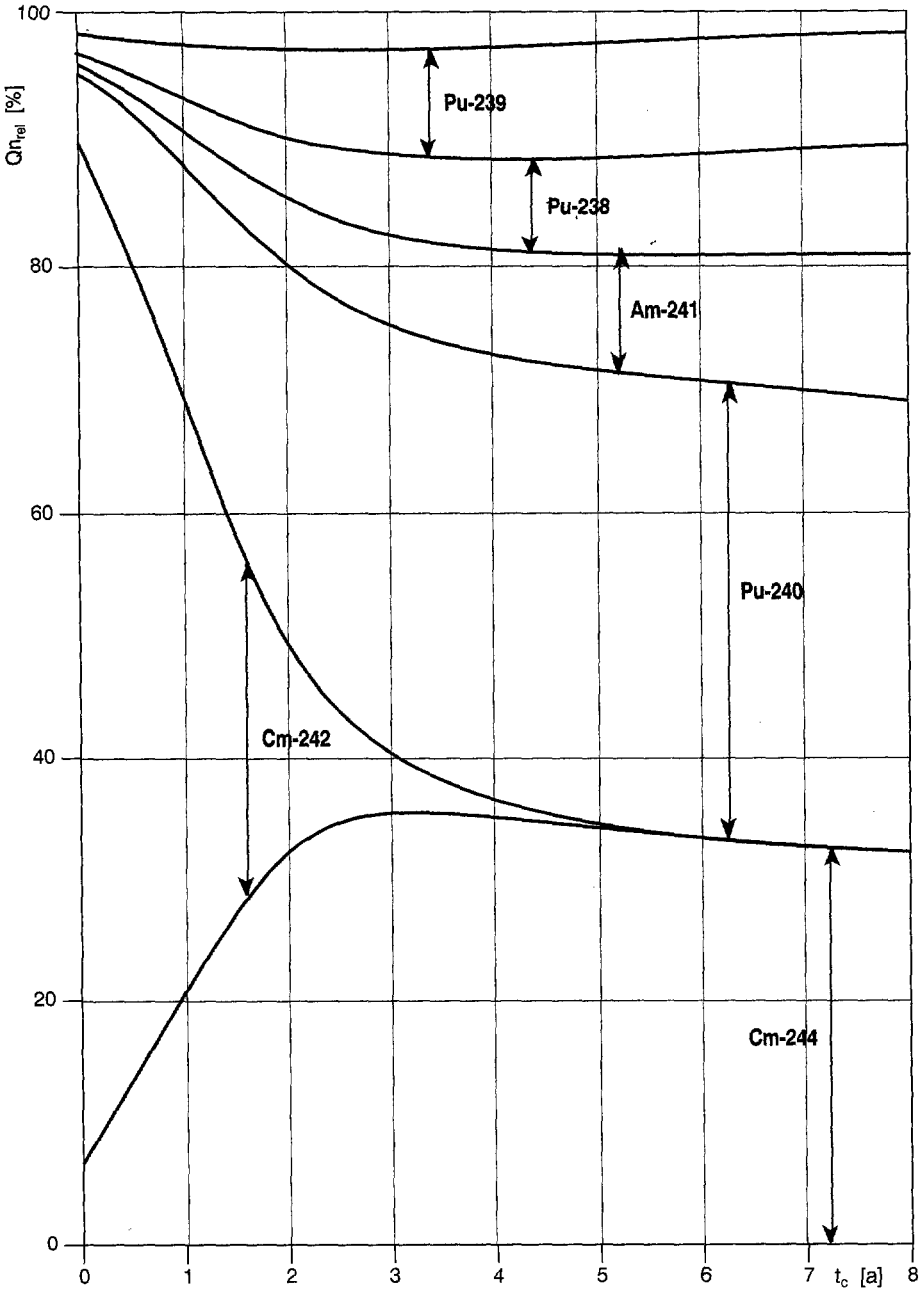


**Fig. 2:** Relative Contribution of Individual Isotopes to Total Neutron Emission as a Function of Cooling Time (WWER-440 Fuel Element Type R36, BU = 10 GWd/tU)

This shall be illustrated by the following examples (density increase $Wd_M$ due to a moderator temperature drop from $T_M^0$ to $T_M^1$, $t_c$ = 0), see Table IV :

**Table IV**

| Fuel Element Type | $T_M^0$ (K) | $T_M^1$ (K) | $\Delta d_M$ (%) | $\Delta Q_n$ (%) | BU (GWd/tU) |
|---|---|---|---|---|---|
| WWER-440 R36 | 558 | 538 | 4.4 | -3.7 | 45 |
| WWER-1000 | | | | | |
| 3 | 595 | 583 | 4.4 | -2.6 | 40 |
| 4 | 595 | 583 | 4.4 | -2.1 | 40 |
| 4 | 617 | 584 | 18 | -10.3 | 40 |

$$\Delta Q_n = \frac{Q_n^1 - Q_n^0}{Q_n^0} \qquad \Delta d_M = \frac{d_M^1 - d_M^0}{d_M^0}$$

### Boric acid concentration $C_{BA}$

Changes in boric acid concentration have effects similar to those of changes in moderator density because an increasing concentration has the same qualitative effect on the neutron spectrum as a decreasing moderator density.

However, whereas moderator density at a fixed reactor core position remains constant during steady-state operation, boric acid concentration is continuously decreased starting from a maximum value at the beginning of the reactor campaign. In the nominal state, the mean value of this transient was used.

A comparison of the emission rates occurring after a continuous irradiation at constant $C_{BA}$ = 0 with the rates at constant $C_{BA}$ = 7 g $H_3BO_3$/kg $H_2O$ gives the results of Table V:

**Table V**

| Fuel Element Type | $\Delta$Qn (%) | BU (GWd/tU) |
|---|---|---|
| WWER-440 R36 | 10 | 45 |
| WWRE-1000 | | |
| 3 | 10 | 40 |
| 4 | 7 | 40 |

$$\Delta Q_n = \frac{Q_n^7 - Q_n^0}{Q_n^0}$$

Here, a general remark shall be made referring to a principal difference between WWER-1000 fuel elements with and without cluster absorbers. Similar to an increased boric acid concentration, the insertion of cluster rods into the fuel element results in a harder local neutron spectrum. At a given mean burnup, this produces a higher Cm-concentration and higher neutron emission rate in fuel elements with inserted absorber rods as compared to absorber-free fuel elements (under otherwise equal conditions). With increasing burnup, this difference diminishes because of the decreasing boron content of the absorber.

Since under nominal state conditions the final burnup is attained by a continuous irradiation process (rod power q(t) = const.), the effects of the succession of full power operation and refuelling shutdown periods on neutron emission have also to be investigated.

Whereas the concetration of some isotopes only depends on the attained burnup, the concentrations of short-lived isotopes more or less sensitively follow the time course of irradiation. Here, the isotopes Pu-241, Am-241, Pu-238, Cm-242 and Cm-244 are of special interest. After reactor shutdown, the concentrations of Pu-241 and Cm monotonously decrease according to their half-lives. Am-241 increases its concentration as a result of Pu-241 decay. The concentration of Pu-238 has a small maximum after shutdown because of the decay of Cm-242 which has a much smaller half-life than Pu-238.

To investigate the effects of these interdependencies on neutron emission, a standard irradiation history was compared with two extremely deviating cases (standard case 1: I-R-I-R-I-S, case 2: I-R-I-C-I-S, case 3: I-R-I-C-C-C-I-S; with I - irradiation period of 1 year, R - reloading shutdown period of 40 days, C - interim cooling

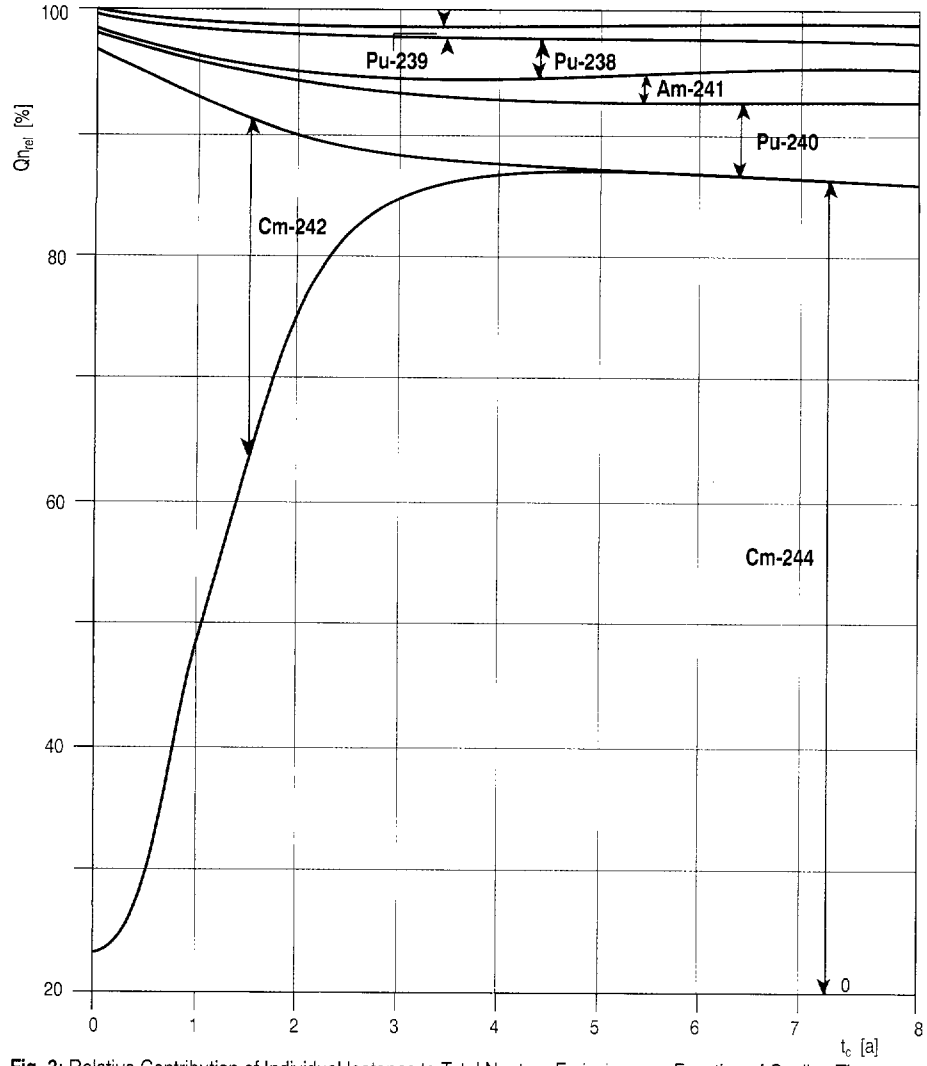


**Fig. 3:** Relative Contribution of Individual Isotopes to Total Neutron Emission as a Function of Cooling Time (WWER-440 Fuel Element Type R36, BU = 20 GWd/tU)

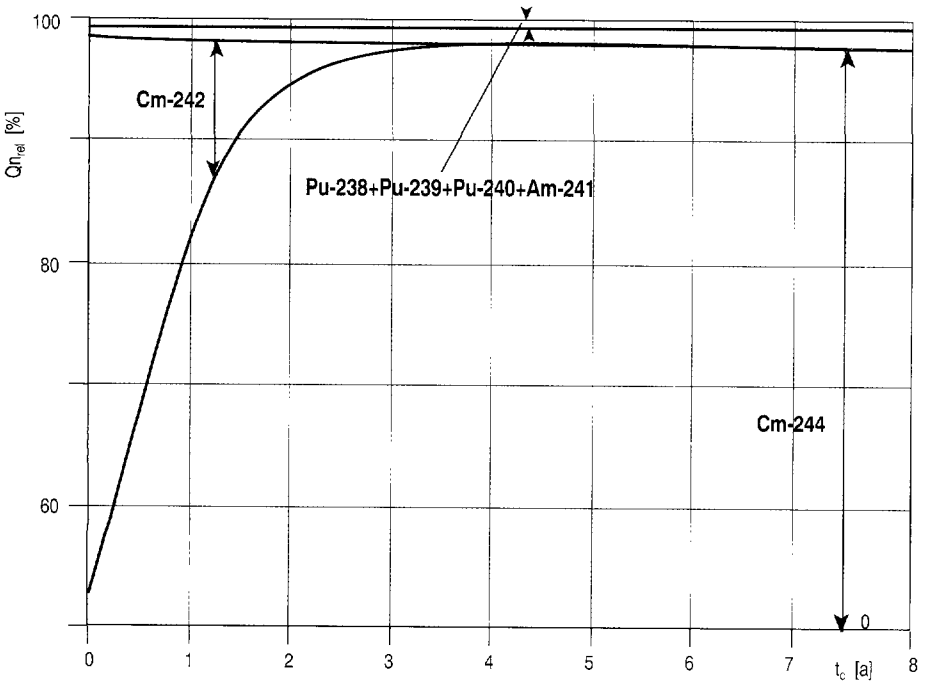


**Fig. 4:** Relative Contribution of Individual Isotopes to Total Neutron Emission as a Function of Cooling Time (WWER-440 Fuel Element Type R36, BU = 40 GWd/tU)

period of 1 year, S - storage following final unloading). After final unloading, for both reactor types the following qualitative course of the deviation from the standard case results (BU = 45 GWd/tU, fuel elements types R36 (WWER-440) and 4 (WWER-1000)), see Table VI:

**Table VI**

| | $t_c$ (a) | $\Delta Q_n$ (%) |
|---|---|---|
| case 2 | 0<br>0.5<br>2<br>5 | 15<br>9<br>1<br>-0.3 |
| case 3 | 0<br>0.5<br>2<br>5 | 50<br>30<br>3<br>-1 |

$$\Delta Q_n = \frac{Q_n^i - Q_n^1}{Q_n^1} \quad i = 2,3$$

It can be concluded that after 2 years of cooling even in case 3 the deviation from the standard case is below 5%. After about 5 years, a significant difference in the emission rates of fuel elements with the same unloading burn-ups as a result of different irradiation histories cannot be detected. From that point on neutron emission is governed by the concentration of isotopes which are insensitive to the course of irradiation (Pu, Cm-244). This conclusion is of special importance for the verification of spent fuel with long cooling times which should constitute the majority of passive neutron measurements.

Another factor affecting the course of burnup is the linear fuel rod power. The dependence of neutron emission on rod power is mainly determined by the change of the relative contributions of Cm-242 and Cm-244 to total neutron emission. Whereas the concentration of Cm-242 decreases with increasing rod power the concentration of Cm-244 slightly increases. Therefore, immediately after unloading the emission rate of fuel elements with the same burnup decreases with increasing rod power. The magnitude of this effect is reduced with increasing burnup and decreasing initial enrichment. WWER-1000 fuel elements therefore have a lower emission rate than WWER-440 fuel elements of the same burnup and initial enrichment.

With increasing cooling time the differences become smaller. After cooling times above 5 years they cannot be detected any longer.

## 5. Subcritical Multiplication during Under-Water Storage

The emission rate calculated for the nominal state only gives the number of fast neutrons emitted per second and per $cm^3$ of fuel. As measurements are usually carried out under the conditions of under-water storage, multiplication effects have to be taken into account. These effects result in an increase of the primary neutron source strength by a factor q:

$$q = (k_{eff} - 1) / k_{eff}$$

Its magnitude depends on the material properties of the fuel and of the surrounding medium together with the geometry of the whole arrangement.

In the calculational model used the fuel element was mapped onto coaxial cylinders. The calculation of $k_{eff}$ and of the neutron flux distribution was carried out one-dimensionally. Neighbouring fuel elements were not considered.

The curves $Q_n$ (BU) measured under storage conditions are in every case flatter than the calculated primary source strength curves because the multiplication factor $k_{eff}$ decreases with increasing burnup. The dependence of $k_{eff}$ with respect to cooling time is only slightly and can be neglected for purposes of evaluating measurements. Therefore, $k_{eff}$ was only considered to be a function of burnup and of the boric acid concentration in the storage pond.

The calculations, whose numerical results are given in chapter 7, show that the rise of neutron source strength is in the range of a factor of 1.5 - 2 for WWER-440 fuel and in the range of 2 - 3 for WWER-1000 fuel.

## 6. Fuel Element Verification by Neutron Spectrum

The neutron spectrum, i.e. the ratio of epithermal and thermal neutron flux, on the one hand, very sensitively depends on the physical parameters of the fuel lattice and, on the other hand, can be measured without significant expense. Therefore it suggests itself to try to conclude from measured spectra to the properties of spent fuel elements.

Figures 5, 6 and 7 show the radial behaviour of fast (epithermal) and thermal flux inside a fuel element and in the surrounding water zone for fuel element types 16 and 36 of WWER-440 and 9 of WWER-1000. Whereas the fast flux behaves like a Bessel function modified by interior heterogeneities of the fuel element, the thermal flux has a characteristic bulge at the edge of the fuel element. This bulge is reduced with increasing boric acid concentration on the pond water.

The behaviour of the spectrum, i.e. of the ratio between the flux values, shows that spectral differences occur only in the interior of the fuel element whereas already at distances of a few centimeters outside the element edge the type-independent (asymptotic) water spectrum appears. According to this, if at all, a spectrum measurement can only give evidence in the interior of a fuel element. Hence, a comparison of the spectra has to be carried out for an internal detector position.

For WWER-440 fuel, a comparison of the burnup-dependent spectra of the individual fuel element types for a measurement position inside the central (water-filled) dosimetry channel shows that such measurements can differentiate the enrichment types with reasonable certainty. Gross spectrum values are in the range between 5 and 7 for a boric acid concentration in the spent fuel pond of 7 g/kg /2/.

In contrast with this, an identification of the fuel element types for the WWER-1000 is possible only with some restrictions /3/. Assembly types 1, 2, 9 and 10 are unequivocally identifiable, i.e. in particular without having detailed knowledge on burnup. A separation of fuel element types 5 and 7, or 6 and 8 respectively, is basically impossible because they distinguish only in their enrichment profiles (type "3.3./3.3" contrary to type "3.3./3.0"). In theory, an experimental isolation of a 3.0% fuel element (types 4 and 3) would be conceivable provided that, in addition, the burnup is known. However, the actual measurement accuracies might not be sufficient.

## 7. Parametrization of the Results

The numerical results obtained by using the respective computer codes can only be presented in extensive tables which are not easy to manage. They are of restricted use if statements are to be given in a quick manner and at low expenditure. For purposes of a simplified data management a representation of the results as compact as possible had to be found which also can be applied with simple computer technology. This was made by means of a parametrization of the numerical results.

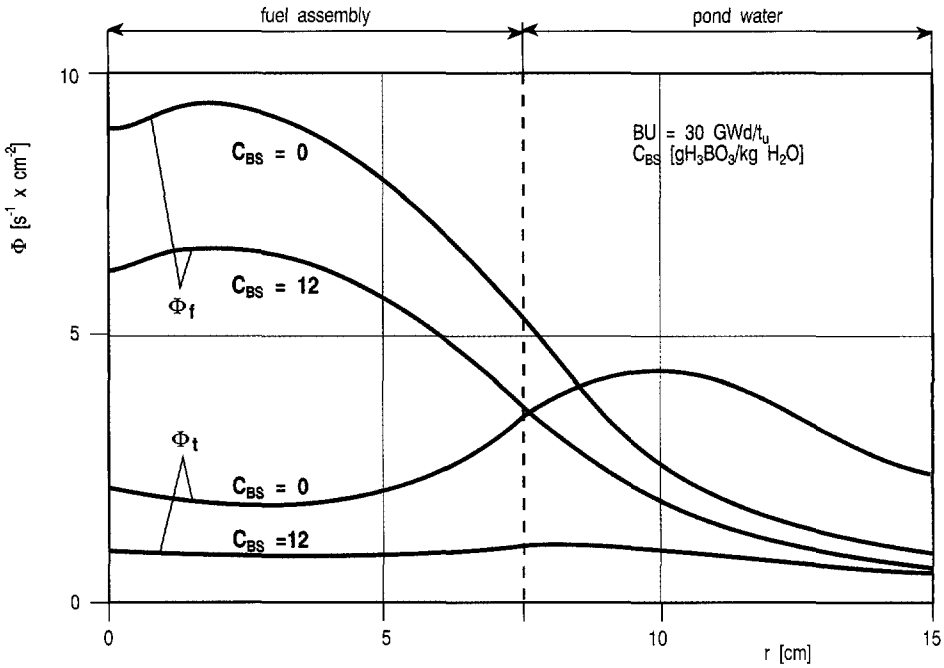


**Fig. 5:** Radial Neutron Flux Distribution for Various Boric Acid Concentrations, Fuel Element Types K16, R16 (WWER-440) ($Q_n$ = 1 n/s x cm$^3$ $UO_2$)

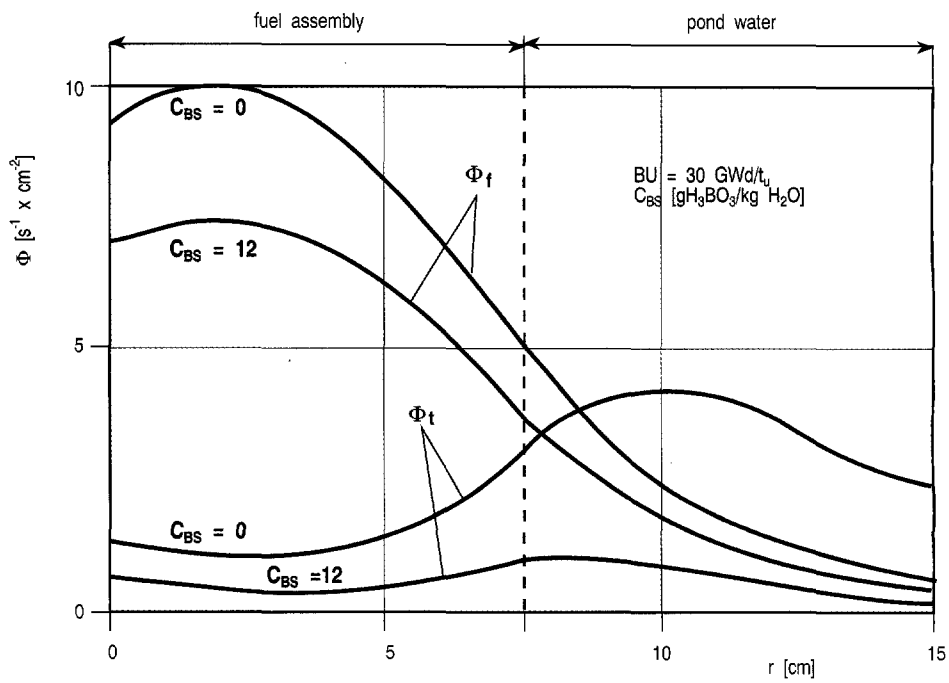


**Fig. 6:** Radial neutron Flux Distribution for Various Boric Acid Concentrations, Fuel Element Type R36

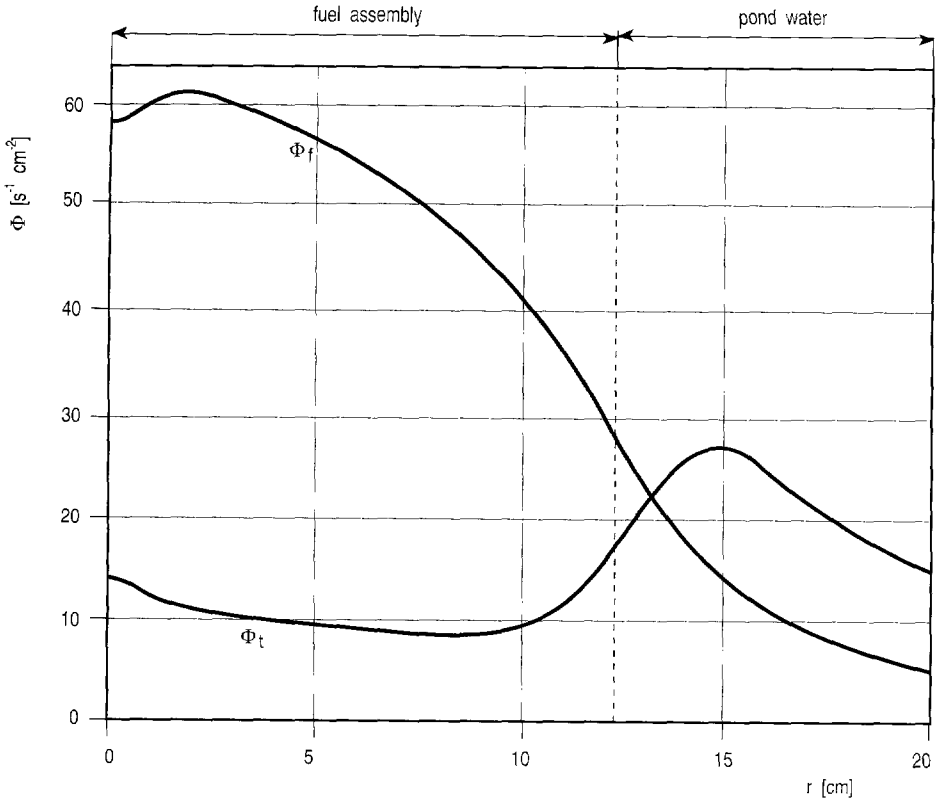


**Fig. 7:** Radial Neutron Flux Distribution for Various Boric Acid Concentrations, Fuel Element Type 9 (WWER-1000) ($c_{BA}$ = 0, BU = 0, $Q_n$ = 1 n/s x cm$^3$ $UO_2$)

### Parametrization of Neutron Source Strength

The underlying physical mechanism of passive neutron emission suggests to choose a parametrization method which combines an exponential law for the burnup dependence of $Q_n$ at $t_c = 0$ with a time function whose behaviour should be based on the decay of the main neutron emitters Cm-242 and Cm-244:

$$Q_n (BU, t_c) = \alpha \cdot BU^{\beta} \cdot g (BU, t_c)$$
$$g(BU, 0) = 1 \qquad (4)$$

A detailed examination results in the following representation

$$Q_n (B, t_c) = \alpha \cdot B^{\beta} \cdot g (B, t)$$

$$g (B, t) = \frac{\exp (-\lambda_{244} \cdot t) + k \cdot \exp (-\lambda_{242} \cdot t) + r_t}{1 + k + r}$$

$$k(B) = c_0 + 2c_1 \cosh[c_2 (B\text{-}BF)] \qquad (5)$$

$$r = r_0 \exp[-\gamma (B\text{-}B_0)]$$

$$r_t = r(1 + 0.02\, t)$$

Accordingly, a coefficient set (K) of 7 parameters is required per fuel element type to describe the neutron emission for all occurring combinations of burnup and cooling time:

$$K = \{\alpha, \beta, c_0, c_1, c_2, r_0, \gamma\}$$

In addition, there are the data for the burnup interval ($B_0$, BF) across which the parametrization was performed.

Tables VII and VIII list the coefficients for the above formula. With the numerical values stated there, expression (5) reproduces the numerically calculated tabular values /1, 3/ within an accuracy of ≤ 5% (WWER-1000) and ≤ 4% (WWER-440) respectively.

### Parametrization of Neutron Multiplication

The neutron signal measured is proportional to the number of fast neutrons emitted per second and per element of volume from the spent fuel; through that, neutron multiplication is included into the measurement signal and will vary according to the actual conditions of storage.

Therefore, it seems to be advantageous to derive a parametrization formula also for the multiplication factor $k_{eff}$

$$k_{eff} = f (BU, c_{BA})$$

The simple equation base

$$k_{eff} = \gamma \cdot \frac{1 - \alpha \cdot BU}{1 + \beta \cdot c_{BA}}$$

was tested for this purpose. It results, that $k_{eff}$-values are reproduced with a relative accuracy of < 4%. The numerical values of the parametrization factors and the maximum deviations between parametrized values and original values are listed in Tables IX and X. The parametrization factors were obtained by a calculus of observation using the least squares method.

## 8. Conclusions

For the evaluation of measurements of spent fuel with cooling times $t_c \geq 2a$ the application of the numerical results obtained for the nominal state is possible without any restrictions. In this period of time the data presented in this article can be used for the whole range of technologically possible operational states of the reactors, especially for the different types of irradiation histories. For cooling time $t_c < 2a$ only extreme irradiation histories and operational states, which considerably deviate from the nominal state parameters over long periods of time, require a detailed consideration of the individual situation.

A definite assessment of the reliability of the presented results is only feasible on the basis of a comparison between calculated data on nuclide concentration and data obtained from destructive analysis of spent fuel. For WWER-440 fuel, this comparison demonstrated the good applicability of the used calculational method /2/. This allows expecting a good agreement be-

**Table VII: Parametrization Coefficients for the Primary Passive Neutron Source Strength of WWER-440 Fuel Elements**

| Element Type | $\alpha$ | $\beta$ | $r_o$ | $\gamma$ |
|---|---|---|---|---|
| K16, R16 | 1.3279-1 | 3.3122+0 | 2.7622-2 | 2.6044-1 |
| K24, R24 | 4.9990-2 | 3.4724+0 | 5.9489-1 | 2.7808-1 |
| R36 | 4.8055-2 | 3.3136+0 | 1.0642-1 | 1.3572-1 |

| Element Type | $c_o$ | $c_1$ | $c_2$ |
|---|---|---|---|
| K16, R16 | -9.4892 -2 | 3.9722-1 | 1.2180-1 |
| K24, R24 | -2.4973 -1 | 6.8634-1 | 1.1800-1 |
| R36 | -1.2046+0 | 9.8385-1 | 6.3160-1 |

**Burnup intervals for parametrization**

| Element Type | $B_o$ | BF |
|---|---|---|
| K16, R16 | 10 | 30 |
| K24, R24 | 10 | 30 |
| R36 | 20 | 45 |

**Table VIII: Parametrization Coefficients for the Primary Passive Neutron Source Strength of WWER-1000 Fuel Elements**

| Element Type | $\alpha$ | $\beta$ | $r_0$ | $\gamma$ |
|---|---|---|---|---|
| 1 | 4.9894-2 | 3.4721+0 | 2.5238-1 | 2.3340-1 |
| 2 | 7.8060-2 | 3.3684+0 | 2.0023-1 | 2.1836-1 |
| 3 | 3.4010-2 | 3.4514+0 | 1.3099-1 | 1.6085-1 |
| 4 | 5.0358-2 | 3.3387+0 | 1.0967-1 | 1.5104-1 |
| 5 | 2.4315-2 | 3.4769+0 | 1.5808-1 | 1.6240-1 |
| 6 | 3.9080-2 | 3.3750+0 | 1.3173-1 | 1.5318-1 |
| 7 | 2.5731-2 | 3.4703+0 | 1.5074-1 | 1.6197-1 |
| 8 | 4.2042-2 | 3.3646+0 | 1.2464-1 | 1.5242-1 |
| 9 | 1.6659-2 | 3.4815+0 | 9.5060-2 | 1.2073-1 |
| 10 | 2.6966-2 | 3.3808+0 | 8.1055-2 | 1.1343-1 |

| Element Type | $c_0$ | $c_1$ | $c_2$ |
|---|---|---|---|
| 1 | -1.4386+0 | 1.0886+0 | 8.4894-2 |
| 2 | -1.6714+0 | 1.2068+0 | 7.3944-2 |
| 3 | -1.6760+0 | 1.0920+0 | 5.3512-2 |
| 4 | -2.1576+0 | 1.3493+0 | 4.4893-2 |
| 5 | -2.1048+0 | 1.3443+0 | 5.1442-2 |
| 6 | -2.5996+0 | 1.6033+0 | 4.3807-2 |
| 7 | -1.9927+0 | 1.2784+0 | 5.1923-2 |
| 8 | -2.4829+0 | 1.5350+0 | 4.3934-2 |
| 9 | -3.1395+0 | 1.8300+0 | 3.7121-2 |
| 10 | -4.2655+0 | 2.4030+0 | 3.0114-2 |

**Burnup intervals for parametrization**

| Element Type | $B_0$ | BF |
|---|---|---|
| 1, 2 | 12 | 30 |
| 3 - 8 | 18 | 45 |
| 9, 10 | 24 | 54 |

t cooling time in a
B unloading burnup in GWd/tU
$Q_n$ passive neutron strength in n/cm$^3$s

**Table IX: Parametrization Coefficients for $k_{eff}$ (BU, $c_{BA}$) for WWER-440 Fuel Elements**

| Element Type | $\alpha$ | $\beta$ | $\gamma$ | $\Delta_{max}$ (%) |
|---|---|---|---|---|
| R16, K16 | 5.897-3 | 5.85-2 | 4.940-1 | 3.0 |
| R24, K24 | 5.913-3 | 5.47-2 | 5.542-1 | 3.7 |
| R36 | 6.004-3 | 4.95-2 | 6.252-1 | 3.1 |

**Table X: Parametrization Coefficients for $k_{eff}$ (BU, $c_{BA}$) for WWER-1000 Fuel Elements**

| Element Type | $\alpha$ | $\beta$ | $\gamma$ | $\Delta_{max}$ (%) |
|---|---|---|---|---|
| 1 | 7.27461-1 | 6.77313-3 | 4.69007-2 | -2.0 |
| 2 | 5.63273-1 | 2.81896-3 | 4.28224-2 | 3.4 |
| 3 | 8.14309-1 | 6.52533-3 | 4.10335-2 | 2.2 |
| 4 | 6.43346-1 | 4.28870-3 | 3.85238-2 | -2.4 |
| 5 | 8.35244-1 | 6.42598-3 | 3.97403-2 | 2.1 |
| 6 | 6.62056-1 | 4.42670-3 | 3.76134-2 | -2.1 |
| 7 | 8.30859-1 | 6.46522-3 | 3.98663-2 | 2.1 |
| 8 | 6.56243-1 | 4.38760-3 | 3.76735-2 | -2.2 |
| 9 | 8.78574-1 | 5.83652-3 | 3.57971-2 | -2.4 |
| 10 | 7.03134-1 | 4.36647-3 | 3.48204-2 | 2.3 |

BU unloading burnup in GWd/tU
$c_{BA}$ boric acid concentration in the pond water in g$H_3BO_3$/kg $H_2O$

$$\Delta_{max} = \frac{k_{eff}^{par} - k_{eff}^{cal}}{k_{eff}^{cal}}$$

tween calculational and experimental data also for WWER-1000 spent fuel. However, an exact evaluation of the data obtained for this type of fuel will only be possible if destructive analysis data are available.


## Acknowledgement

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# A Quality Control Approach to Near Real Time Material Accounting

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## Abstract

A procedure is suggested whereby a sequence of values of material unaccounted for (MUF) obtained from a near real time materials accountancy (NRTMA) can be analysed using standard statistical process control methods. Though this analysis may not be the most powerful, it has the advantage of using widely implemented techniques and can prove to be more powerful in practice due to the fact that it requires fewer assumptions about the variability of the process measurements.

## Introduction

The idea of using Near Real Time Materials Accountancy (NRTMA) for monitoring the throughput of plutonium at nuclear reprocessing facilities has been widely discussed (Goldman et al., 1982). The basic idea is that rather than waiting till the end of a reprocessing campaign to decide whether plutonium has been diverted, one can monitor sequentially the material unaccounted for (MUF) over a number of short balance periods.

The approach taken has generally been to search for a most powerful statistical test of predetermined size (usually 0.05). The test chosen will depend on the diversion strategies one wishes to detect and a number of tests may have to be carried out cojointly in order to ensure simultaneous high power against different alternatives (Jones, 1988). In fact, there have been many tests suggested for use in NRTMA. Speed and Culpin (1985) report on a comparison of twelve different ones. It should be noted though that all of the tests proposed are based on some model for the variability in the data and require knowledge of the extent of that variability.

However, the problem of monitoring plutonium throughput is a classical problem of quality monitoring, complicated by the fact that the sequence of observed MUFs are highly correlated due to having common inventory measurements when they are contiguous. In this paper we suggest how the MUF sequence can be decomposed and normalized to allow it to be analysed by standard statistical process control (SPC) techniques. Such an approach allows one to use well known and widely implemented control procedures as well as avoiding one of the critical assumption about the variance of MUF values.

Some data obtained from the Dounreay campaign PR 7/8 are analysed using these ideas and the results compared with other analyses on the same data.

## NRTMA

For simplicity we shall consider a single materials balance area for which an inventory is taken at the beginning of each materials balance period and for which a record of all receipts and shipments is kept. Let

$N$= the total number of balance periods considered,
$I(0)$= the estimated inventory at the beginning of the first balance period,
$I(n)$= the estimated inventory at the end of period $n$,
$R(n)$= the estimated receipts during period $n$,
$O(n)$= the estimated outgoings during period $n$,

where $n = 1,2, ..., N$. The material unaccounted for (MUF) in period $n$ is defined by

$$MUF(n) = I(n\text{-}1) + R(n) - O(n) - I(n)$$

If all measurements were precise, then the MUF sequence would be identically zero when there are no diversions and equal to the amount of material diverted otherwise. But no measurements are precise; they are subject to both systematic and measurement errors. To take account of these errors a probability model is superimposed on the sequence of inventory and transfer measurements, resulting in a probability model for the sequence of MUFs.

The models in the literature (Avenhaus et al., 1984) for the inventory, receipt and shipment measurement errors:

- usually assume they are independent both amongst themselves and over time;
- often assume they are normally distributed;
- sometimes assume that their variances are constant over time.

Thus, in the simplest possible model the $I(n)$ are normally distributed with variance $\sigma_I^2$, the $R(n)$ are normally distributed with variance $\sigma_R^2$ and the $O(n)$ are normally distributed with variance $\sigma_O^2$ and all measurement errors are independent.

## Set

$T(n)$ = the estimated net transfer into the balance area during period $n$, so that

$$T(n) = R(n) - O(n).$$

Then a consequence of the simplest model is that the $T(n)$ are also normally distributed with variance

$$\sigma_T^2 = \sigma_R^2 + \sigma_O^2 ;$$

the MUF sequence is multivariate normal with

$$var(MUF(n)) = \sigma_M^2 = 2\sigma_I^2 + \sigma_T^2$$

and

$$\text{cov}(MUF(m), MUF(n)) = \begin{cases} -\sigma_I^2 & \text{if } |m-n| = 1 \\ 0 & \text{if } |m-n| > 1. \end{cases}$$

In this simple model, the variance-covariance structure of the MUF sequence is constant over time; the difference between a no diversion and a diversion regime is modelled by the sequence of MUF means. When there is no diversion (and no systematic bias in the measurements)

$$E(MUF(n)) = 0, \quad n = 1, \ldots\ldots, N.$$

When there is a diversion,

$$E(MUF(n)) \geq 0, \quad n = 1, \ldots\ldots, N,$$

with strict inequality for at least one $n$.

In order to construct a test for diversion (against the null hypothesis of no diversion), the probability model is used in three ways:

- to suggest sensible, if not optimal, test statistics;
- to calculate the appropriate critical values for the test statistics;
- to calculate the power of the test under different diversion scenarios.

When the calculations cannot be made explicitly they can be estimated as closely as desired by Monte Carlo techniques. These calculations are always made under the assumptions of the model.

Almost all the tests suggested in the literature are sequential tests; i.e. ones which at the end of each balance period look at the sequence of MUFs already obtained and decide whether there is evidence of a diversion up to the present. Speed and Culpin (1985) compared twelve different sequential tests constructed from the MUF sequence. Their comparisons were based on the associated power curves under different diversion strategies. They found that for the simplest model (independence of measurement errors, normality and variances constant over time) two tests outperformed the others, namely the tests they called the corrected Page test and an extended power one test. The definition of the most important of these tests can be found in Speed and Culpin (1986).

All the work that has been carried out to construct different tests, sensitive to different diver-

sion strategies, and to compare these tests on the basis of estimates of power is, of necessity, founded on explicit probabilistic models. Though the model assumptions may be wrong, at lest a comparison between different possible tests can be made and a number of them removed from further consideration.

Of the three basic model assumptions, the independence of measurement errors seems not unreasonable, the asssumption of normality is usually not essential but the third assumption, knowledge of the variances, is critical. Misspecification of the error structure can lead to either over or under sensitive tests (Beedgen and Seifert, 1988).

It is very easy to show that the powers of the tests usually considered are dramatically increased when the variability of the measurement errors is reduced. It is worthwhile remarking that while there is general agreement on the optimality of one or two test procedures, future improvements in the tests are likely to be marginal in comparison with the improvements which could be obtained by tightening the control of the underlying processes. The fact is that the detection of diversion is a quality control problem and some insight can be gained by looking at it in that light.

## Statistical Process Control

In monitoring any process we are interested in two types of disturbance: the odd unusual observation and systematic changes in the target values or the variability. This is a classical problem which has been extensively studied and for which there are standard and well understood procedures when the data are generated by a simple model.

The usual model in the statistical process control of continuous measurements (such as MUFs) is that the variables are independently, normally distributed with constant variance. Normality is often assured by analysing averages of contiguous measurements. Independence is essential and is certainly not true for a sequence of MUFs.

However, if the assumption of independence amongst the measurement errors is valid for the quantities being used to calculate the MUFs, they will be independent if they are not contiguous. That is, MUF($m$) and MUF($n$) are independent if m-n > 1. So under this assumption, the sequence of even numbered MUFs (and the sequence of odd numbered MUFs) is a sequence of independent random variables. By splitting the MUF sequence into two, we obtain two sequences of independent random variables which might be monitored by standard statistical process control techniques.

The MUFs however, are not identically distributed unless the variances are constant over time. If the variances are not constant but at least their ratios are known or can be estimated, the MUF sequence can be normalized to have approximately constant variance. Thus, for example, if MUF($n$) has an estimated standard deviation of SD(n), then {MUF($n$) / SD($n$); n = 1,..., N} should have approximately constant variance.

The normality requirement for statistical process control is the basis of the calculation of appropriate control limits. The limits usually calculated are appropriate even when the normality assumption is violated provided that the underlying distribution is symmetric or close to symmetric.

Our suggestion is, therefore, that as well as monitoring the complete sequence of MUFs on the basis of an appropriate test, one should also monitor the odd and even subsequences using standard statistical process control procedures. In both cases some assumptions about the variances have to be made. All the NRTMA tests require knowledge of the absolute values of the variances; with the statistical process control tests, only the relative values need be known.

More precisely, if estimates of the standard deviation, SD(n), for each MUF are known (if they are not we can only assume that the variances are constant over time, or equivalently SD(n) ≡1 for all n), set

$$X1(n) = MUF(n)/SD(n),\ n = 2,4..., [\frac{N}{2}] \times 2$$

and

$$X2(n) = MUF(n)/SD(n),\ n = 1,3,..., [\frac{N-1}{2}] \times 2 + 1$$

Plot the values of X1(n) and X2($n$) over time on control charts whose control limits are calculated from the variance of the process (Nelson, 1984). In this way, the estimated standard deviations are used to estimate the ratios of variances, but one allows for the fact that they could be inaccurate by a common scale factor. The estimated standard deviations are often taken from some computer simulation of the reprocessing system. We shall see later that for the Dounreay data there is evidence that the estimated standard deviations are too high. These control charts are then monitored for gross and systematic changes using the standard rules.

A warning that the process may be out of control is given by the occurrence of one of the following events. Each event will occur with a probability of less than one in a hundred if the process observations are normally distributed, independently, with constant mean and variance. (The definitions of the events can, of course, be altered to make the criteria more or less stringent with an appropriate change in the associated probabilities.) The eight standard warning signals we have used are:

- an observation outside the control limits;
- eight successive observations on one side of the mean;
- six consecutively increasing (decreasing) values;
- fourteen values alternating up and down, i.e. oscillating;
- two out of three values more than 2 S.D.s away from and on the same side of the mean;
- four out of five values more than 1 S.D. away from and on the same side of the mean;
- fifteen successive observations within plus or minus 1 S.D. of the mean;
- eight successive observations more than plus or minus 1 S.D. away from the mean.

While some of these rules do not apply until there is a relatively long sequence of balance periods, nevertheless they can be used as indicators of lack of control in the MUF sequence. Another drawback is that a long sequence of balance periods is needed to obtain a reasonably accurate estimate of the variance of the process. As an alternative one can calculate control limits sequentially from the data. To do this robustly we propose using limits based on a boxplot of the data so far collected.

The boxplot is a simple graphical summary of a data set showing the median, lower and upper quartiles of the observations as well as indications of outlying observations. (For further details see Tukey, 1977). The spread of the data set is assessed from the upper and lower quartiles and used to determine reasonable limits beyond which one would not expect to see observations. Its importance here is that its estimate of spread is based on the middle half of the data and so is unaffected by the odd outlying observation. Note that in this case the control limits cannot be calculated till there have been at least five material balance periods. Of course, the control charts for the even and odd MUFs can also be complemented by standard CUSUM charts and range charts (Goel, 1982).

An obvious objection to using the above approach is that one is presumably losing information and hence power by splitting the MUF sequence into two. However, these tests should be seen as an addition and not an alternative to the other tests already suggested in the literature. As a consequence they have the potential to indicate that the process is out of control when the standard approach may not do so. Used this way they can only improve the sensitivity of the overall analysis, though care must be taken with the interpretation of any results as well as we are suggesting using extra tests.

On the other hand, use of statistical process control methods carries with it the advantage that one is using standard, well understood methods. As well, seeing the problem as one of quality monitoring leads one to think about the modern management philosophy of quality management with its emphasis on knowing and constantly improving the underlying processes.

In the next session we shall apply these methods to one of the few available data sets to see what insight is provided.

## The Dounreay Data

One of the few publicly available NRTMA data sets comes from the Dounreay fast reactor fuel processing plant. The plant is divided into two material balance areas: a head end MBA and a solvent extraction and export MBA. During a campaign which took place between September 1985 and February 1986, the material balances were determined in both areas at approximately daily intervals.

The measurements of inventory levels, receipts and shipments of plutonium in both areas and of uranium in the head end MBA are presented and analysed in a report by Jones and Gordon (1986). The Dounreay plant has a sophisticated software package, CIMACT (Gale)

which, amongst other tasks, is able to combine data on the precision of component measurements to give estimates of the standard errors of the calculated MUFs. The estimates for this campaign were far from constant over time and were used to normalize the MUF values. We are thus assuming that the output of CIMACT represents the most accurate estimate of error variance or, at the very least, estimates of the relative weights to be attached to the MUFs.

The data sets are quite large (about 100 observations each). We shall take the opportunity of analysing all three sets, even though one relates to uranium; the procedures suggested here do not depend on the material being measured. As Jones and Gordon (1986) point out, the benefit in analysing the uranium data as well as the plutonium is that such analysis can confirm the interpretation of the plutonium data.

Before any analysis was begun, the data were studied for unusual features. Given that we have suggested using robust control techniques, the aim was not to detect outliers in the data but values which might not fit the basic model. Thus, for example, the first MUF in all three data sets was identically zero since the only activity during that period was the receipt of material and the amount of the receipts was taken to be the inventory at the end of the period. Also at the end of the campaign, the inventory levels are comparatively small as are the error estimates. It was decided, therefore to remove the following observations from the three data sets:

| | |
|---|---|
| Set 1: Plutonium Head End | 1, 98, 99, 100 |
| Set 2: Uranium Head End | 1, 98, 99, 100 |
| Set 3: Plutonium Export End | 1, 108 |

The MUF sequences were then divided by their corresponding estimates of standard deviation from the CIMACT output and split up into odds and evens. If the independence assumptions are true, this should produce two sequences of independent, identically distributed random variables. These subsets were then drawn as boxplots. A boxplot is a simple graphical representation of data, sensitive to non-symmetries and unusual observations. The six boxplots are shown in Figure 1.

The boxplots show the standardised data to be approximately symmetrically distributed, except maybe for the odd numbered subsequences of Set 1 and Set 3. The plots also indicate the existence of unusual observations (indicated by an asterisk) in five of the six data sets.

Note that the boxplots are drawn from the complete data sets. In an application of NRTMA this luxury would not be available though boxplots can be drawn once there are at least five observations.

Proceeding with the complete data sets we can calculate control limits on the basis of the data observed. These are shown in Figure 2, together with an indication of points (shown as large dots) where the standard tests, listed in the previous section, are violated. Thus for set1-odd, test 2 indicates there is an unusual event: for set2-odd, tests 1 and 7 are invoked; for set2-even, tests 2 and 1; and for set3-even, tests 1, 5, 4 and 4. Using the same data to both calculate the control limits and assess process stability is very conservative - any observations which are unusually far from the process mean contribute to an increase in the sample variance which makes the control limits wider and hence less likely to pick up extreme readings. That this has happened here can be seen from the fact that some of the boxplots indicate outliers which are within the control limits.

To avoid this problem and to construct control limits sequentially as data is collected we adopted the following procedure:

a) wait till 5 observations are collected;

b) calculate the boxplot for the current amount of data and use its limits as the control limits;

c) increase the number of observations by one and repeat b).

The corresponding control charts are shown in Figure 3. Note how the control limits vary at the beginning of a sequence and then settle down to a stable pattern unaffected by the odd outlier.

The control charts show (with a large dot) those points which are outside the robust control limits together with the points which violate one of the other seven standard tests. Notice that the points where the process is shown to be out of

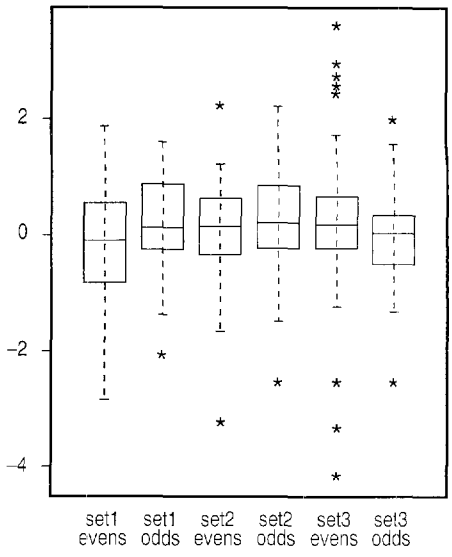


Fig. 1

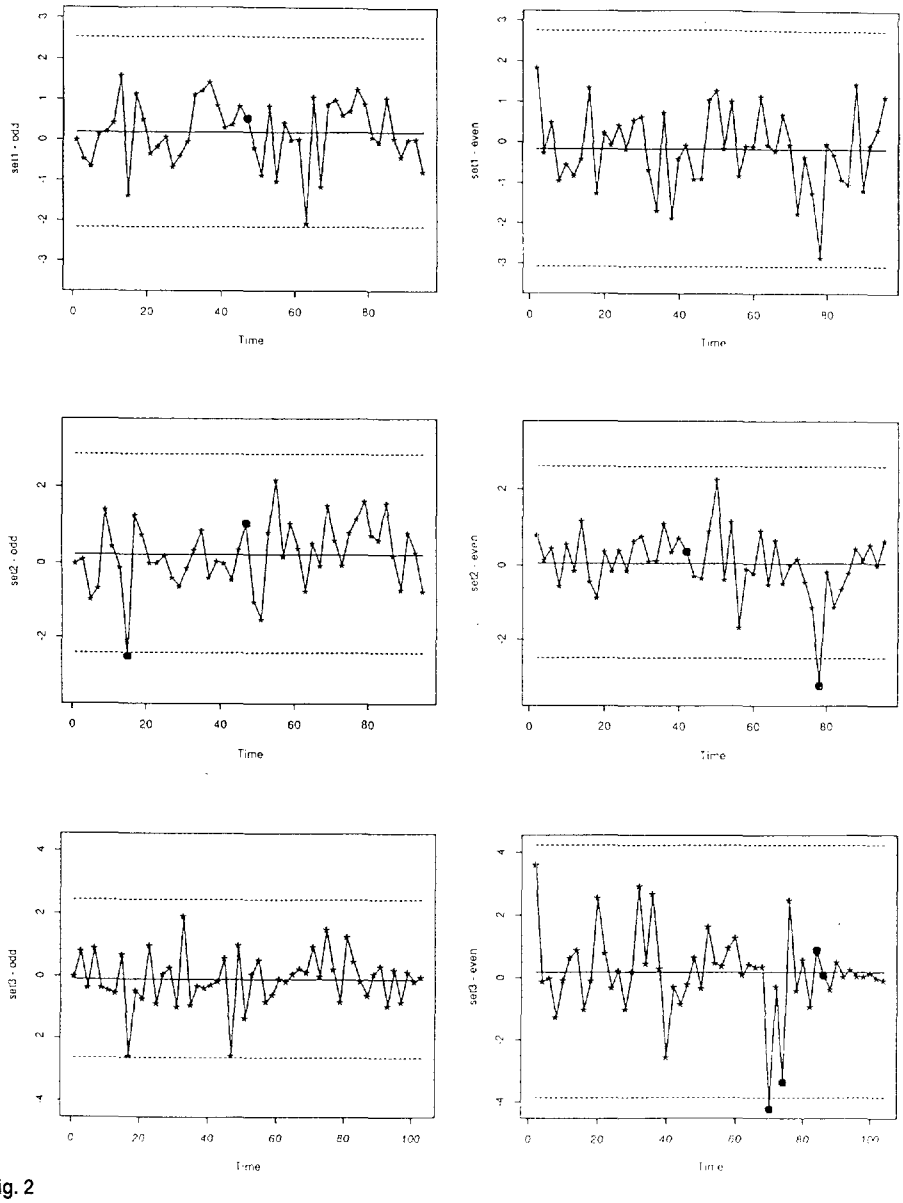


Fig. 2

control are often when the MUF is negative, a reflection that there must be problems with the measuring process.

We are not able to speculate on the interpretation of the unusual points or patterns indicated - that must be done by the engineers involved in the particular plant and campaign. The important point to note here is that the use of standard statistical process controls on these data sets has indicated a number of points of concern, most of which were not picked up by the earlier analysis of the data (Jones and Gordon, 1986).

## Conclusion

The application of standard quality monitoring tests to the Dounreay data has provided indications of unusual events which need to be explained or investigated. It has shown the value in contemplating rough but simple ideas which avoid some of the assumptions about variances. It is particularly valuable if it convinces of the need for paying attention to variability, the power that can be achieved by reducing variability and, hence, the importance of process improvement to this as to any quality problem.

We finish though on a word of warning. Our decision to look at the application of statistical process control techniques to the Dounreay data arose from a belief that there was some evidence of patterns in the MUF sequences. This evidence came from an analysis of the odd and even MUF sequences based on cubic smoothing splines (Silverman, 1985). Cubic splines are non-parametric regressions which require choice of a smoothing parameter (similar to the choice of degree when fitting polynomials). For these data sets the smoothing parameters were chosen by looking at the behaviour of plots of variance estimates against smoothing parameters as outlined in Eagleson (1989). These plots are sensitive to correlation in the error structure and in all six data sets showed evidence of correlations still being present despite considering only odd and even MUFs. It turns out (T. Jones, personal communication) that at Dounreay the measurements that are added together to calculate the inventories are not repeated if the particular contribution has not changed since the last balance period. As a consequence quite long range correlations can exist between the MUFs and the models typically used to generate and assess tests are incorrect. It would not be impossible to devise a finer accounting system which tracked the component measurements and which could be more realistically modelled. The lesson to be learnt is that any sensible model is likely to be plant specific and should be developed in close cooperation with those who know operations intimately.

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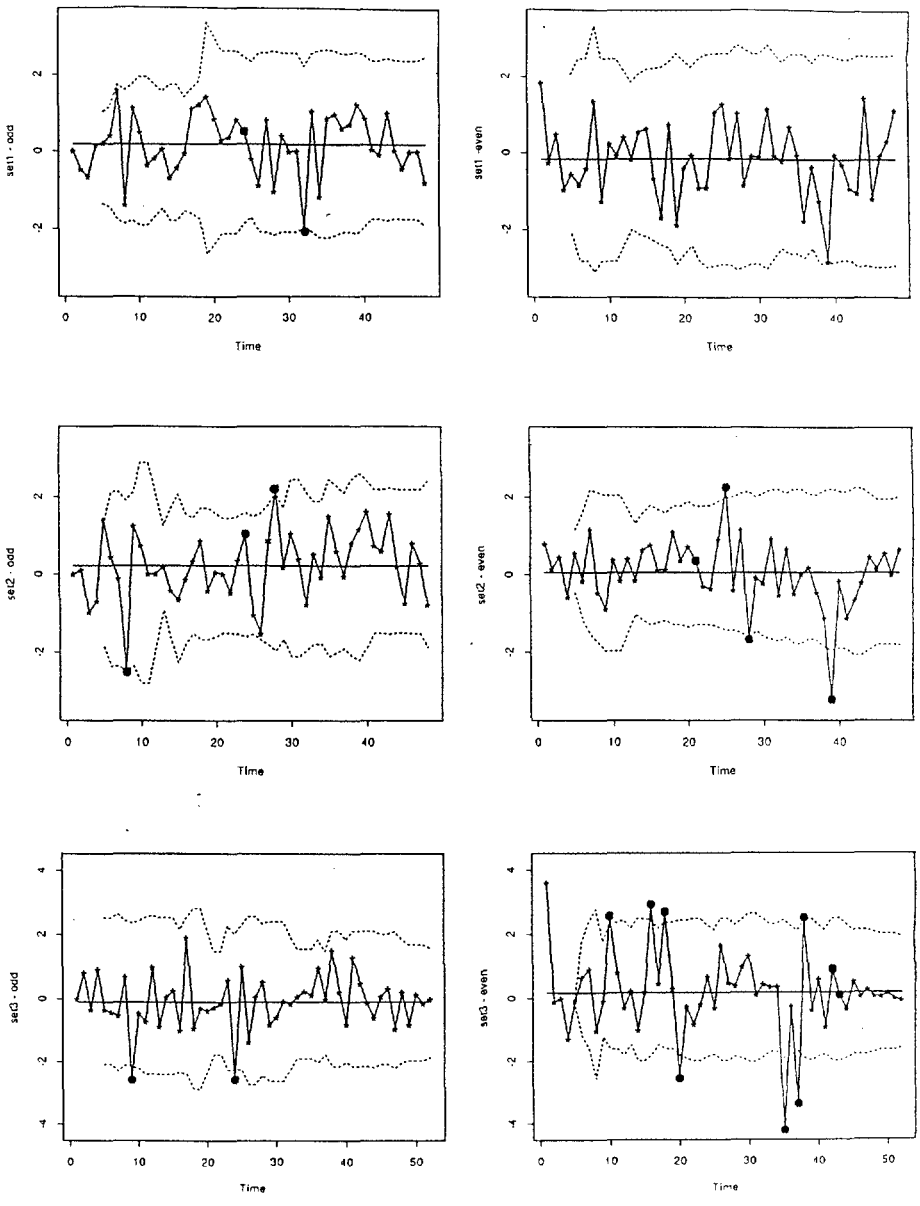


Fig. 3

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