

TECHNICAL REPORTS SERIES No. 46

In-Pile Dosimetry



INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1965

IN-PILE DOSIMETRY

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Printed by the IAEA in Austria
June 1965

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IN-PILE DOSIMETRY

REPORT OF A PANEL ON IN-PILE DOSIMETRY
HELD IN VIENNA,
13-17 JULY 1964

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 1965

International Atomic Energy Agency.

In-pile dosimetry. Report of a Panel ... held
in Vienna, 13-17 July 1964. Vienna, the Agency,
1965.

107 p. (IAEA, Technical reports series no. 46)

621.039.564.2

FOREWORD

A serious difficulty in much experimental research work with research reactors is that of measuring accurately doses or neutron fluences in cases where these are an essential parameter. Even if accurate measurements are performed, it is still in most cases impossible to compare experimental results obtained in different types of reactors, because of the inadequacy of present methods and units to reflect the dependence of spectra and radiation composition on the induced processes.

In view of this the International Atomic Energy Agency convened a Panel on in-pile dosimetry which was held from 13 to 17 July 1964 to review the present status of research and development work and to advise the Director General of the Agency on a possible programme in this field.

The results of this Panel are published in the present Report. Although primarily intended as advice to the Agency, many aspects of the discussions and submitted status reports and papers were considered to be of such general interest that the Agency should make them available in published form. The papers have, however, been included only as summaries as they have, to a considerable extent, been incorporated in the Panel's Report and recommendations. For example, the subject matter of the paper submitted by Mr. Y. Droulers* has been used very extensively for this Report but the paper itself is not included.

It is recognized that the International Commission on Radiological Units and Measurements (ICRU) has recommended a terminology for this field, but this is not yet in general use in many laboratories. Although this terminology is used in the Report of the Panel, no effort has been made to introduce it into the submitted status reports and summaries published here as it is felt that there is no major risk of confusion.

This publication should not be taken as a report on the Agency's programme in the field of in-pile dosimetry, although it serves as a basis on which to build such a programme.

The Agency wishes to thank the Panel members and the authors who presented papers and took part in writing and reviewing the Report.

* DROULERS, Y., Dosimétrie sur les dispositifs d'irradiation en pile. Problèmes et solutions, Commissariat à l'Energie atomique, CEN, Grenoble, INT/Pi (NT) 522-23 (10 juin 1964) 18.

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REPORT OF PANEL ON IN-PILE DOSIMETRY

I. INTRODUCTION

For reactor research work in the fields of radiation chemistry, radiation damage to solids and radio-biology, the radiation dose or neutron fluence is a most essential parameter for the evaluation of the results. The determination of dose or fluence in research reactor experiments is extremely difficult. In part, this is because a reactor is a source of mixed radiation. In some reactors, as in the popular swimming pool reactors of intermediate or high power (> 1 MW) there are also pronounced variations with time. Moreover the experiment itself may cause severe flux density perturbations. These well-known perturbations and variations make it almost impossible to estimate doses and fluences from measurements made on cold, clean cores. For this reason it is desirable to measure the radiation parameter in exactly the same core geometry as would be used for each experiment.

At the present time it seems feasible to recommend specific methods, at least for fluence measurements. However, even if unified experimental techniques could be suggested there still exist severe problems in the presentation of experimental results. An example may be found in the field of graphite irradiations. Radiation damage has been expressed as a function of thermal neutron fluence, fission neutron fluence, or fast neutron fluence. Without proper information about the fast neutron spectrum, an intercomparison of results cannot even be attempted in any valid manner. There is a fundamental difficulty because the present incomplete knowledge of the damage-producing mechanism hampers attempts to correlate a fluence parameter with the radiation damage caused in the specimen. As a result, it would be difficult to compare results obtained in different types of reactors even if methods, units and constants were standardized.

Even for thermal neutron measurements where units are defined, there is still no proper guarantee of compatibility of measurements made at different centres if, for instance, different effective cross-sections are being used. Absorbed dose measurements in reactor experiments are currently specific to the problem under investigation. Moreover, defining the irradiation conditions by measurement of the absorbed dose in reference material will not necessarily give the absorbed dose in an experiment. For these reasons, even in principle, standardization seems impossible in this field.

As a result of all these uncertainties, it is at present difficult, if not impossible, to compare experimental results obtained in the same field of research at different research centres. Therefore, there is definite need for a concerted effort for international unified approach to the problem of in-pile dosimetry.

II. IN-REACTOR FLUENCE AND DOSE MEASUREMENTS

A. *Reactor characteristics*

Measurement of the activity induced in an activation monitor, or the measurement of absorbed dose at a particular time, may not be difficult

to accomplish. However, because of the characteristics of reactors, there is no assurance that such measurements are pertinent to the experiment for which these measurements were made. Part of the uncertainty is caused by the known temporal and spatial variations in the reactor radiation field. The spatial perturbations are of three types: gradients, shielding effects and depressions. The gradients are inherent in all reactors whereas shielding effects and depressions may be introduced by the experimental assembly.

Gradients are generally small in the large cores of heavy-water and graphite reactors, at least in positions which are not very close to the fuel elements. In the case of light-water-moderated reactor cores, which are relatively compact, gradients are quite considerable. For irradiation in such reactors uncertainty of only a few millimeters in the sample location may give rise to appreciable errors in dose or fluence estimates.

Shielding effects are noticeable for all samples that are introduced into a reactor in a container or in experimental rigs of any kind. For thermal neutrons and for gamma radiation this effect usually means an attenuation of the fluence or dose-rate, while for fast neutrons it may even cause an increase in fluence rate, especially in reflector positions in light-water reactors.

An irradiation sample generally causes a thermal flux depression within its own volume and around its immediate vicinity due to the absorption of neutrons in the sample and loss of moderator within that volume. A sample may also be placed where the flux perturbation caused by other experiments is noticeable. Generally, however, these perturbations are less pronounced for gamma radiation and may even be reversed for the fast neutron flux.

All these sources of spatial variation of flux density make it necessary to perform measurements at precisely the sample position. Otherwise there is always the danger that the conditions measured by the detector do not apply to the conditions at the experiment.

In addition to the spatial variations in flux density, variations in time also occur. Such time variations are always present since they are caused in large part by factors connected with the control and operation of the reactor. During the first 50 h of steady-state operation in any cycle the core will reach equilibrium poisoning requiring up to 5% $\Delta k/k$ of the excess reactivity built into the clean cold core. This will in many cases require considerable change of the shim rod positions to maintain a stable power level; it will thus also mean considerably changed flux density patterns in the core and reflector.

The burn-up of fuel will in the long term also cause this same effect during the operating lifetime of the core.

During the initial phase of the operating cycle the gamma radiation will also build up to an equilibrium with the establishment of an approximate equilibrium concentration of the fission products.

These effects are due to changing reactivity requirements with consequent need for changed shim and control rod positions which will change flux density and dose-rate patterns continuously while the reactor is operating. They are severe in pool reactors of more than a few megawatts power level but noticeable also in the heavy-water and graphite reactors. The changed flux density patterns may also cause changes in the leakage flux densities and may thus, through interaction on the neutron sensing control

channels, cause disturbances in the operating power level and make the effect even more serious or, in some cases, reduce it for specific experimental locations.

To these inherent effects should also be added effects that are caused by fuel pattern changes, new experiments, etc. that may be introduced during the runs with specific experimental equipment. Moreover, there are cycle-to-cycle variations which would be encountered in any long-term experiment.

Since such time variations occur there is an obvious need to measure the actual fluence or dose that an experiment has received. This may be done by any one of the following approaches:

- (a) continuous measurement of dose-rate or flux density during an experiment by means of calorimeters, ion chambers, gas loops, self-powered detectors, etc;
- (b) the time integration of fast or thermal neutron flux density by means of activation or fission detectors.

These techniques involve making measurements at the full nominal power of the reactor and under the ambient conditions prevailing at the experiment location. Additional problems are posed by irradiations for very long times and to very high fluences.

Radiation damage effects to solids are caused mainly by the displacement of atoms caused by interactions with the intermediate and fast components of the neutron flux. For this reason the measurement of the fast neutron component and the specification of the fast neutron spectrum are considered most important in monitoring for radiation damage studies in the core of a reactor. In positions well outside the core where the thermal-to-fast ratio is 100 or higher, atoms can be displaced by the recoil attending the emission of gamma rays following thermal neutron absorption. However, for in-pile experiments, the neutron component from 0.01 MeV to 2 MeV probably causes the bulk of displacement damage in most materials. This energy range includes portions of both the fast and intermediate range as commonly defined.

B. Fast neutron measurements

1. Routine methods

For the routine measurement of fast neutron fluence rate, the materials and reactions shown in Table I are widely used:

The use of these materials depends on the half-life of the activated nuclide. To obtain an indication of the spectrum in an irradiation facility or for the monitoring of short-term irradiations any of the nuclides may be used.

However, for the monitoring of long-term irradiation, such as is encountered in materials research, the only suitable materials from this list are Ni, Fe and Cu. In the case of Ni, the burn-up of Co^{58} in a thermal flux density of $\sim 10^{14}$ n/cm² s or greater will decrease appreciably the effective half-life of this isotope. Iron and copper are both very useful although a problem can arise in the use of iron because of Fe^{59} which is produced by thermal neutron absorption by Fe^{58} . It is possible to determine Mn^{54} activities in spite of the competing activities and it is also practical to use

TABLE I
THRESHOLD DETECTORS USED ROUTINELY
FOR FLUENCE MEASUREMENTS

Reaction	Half-life
$P^{31}(n, p)Si^{31}$	2.6 h
$Al^{27}(n, \alpha)Na^{24}$	15 h
$S^{32}(n, p)P^{32}$	14.2 d
$Ni^{58}(n, p)Co^{58}$	71 d
$Fe^{54}(n, p)Mn^{54}$	313 d
$Cu^{63}(n, \alpha)Co^{60}$	5.24 yr
$Th^{232}(n, f)$	
$U^{238}(n, f)$	

iron enriched in Fe^{54} . Copper has quite a high threshold energy which may be a handicap, but is otherwise suitable for very long-term studies.

As the materials listed have been used routinely, a large body of experience has been accumulated on their use. It is thus recommended that laboratories embarking on fast neutron fluence measurements include some of the listed materials in their investigations.

2. Energy dependence of activation cross-sections

Ideally the energy-dependent activation cross-sections for each of these materials should be available. For example, the compilation of LISKIEN and PAULSEN presents data [1] for most of these materials. For some cases, however, only the average cross-section in a fission spectrum is available.

Activation data for the isotopes listed, in dps/g of target nuclide, are used to calculate the fluence. There are two common practices. One assumes a fission spectrum in all irradiation facilities and uses the average fission cross-section to calculate an equivalent fission flux density or fluence. The other attempts to calculate a real flux density by using the energy-dependent activation cross-section and the neutron-energy spectrum for the irradiation facility to calculate the effective cross-section in that facility. This effective cross-section is then used to calculate a flux density.

3. Methods under development

There are additional materials which appear to have potential advantages and therefore further investigations of their use as fluence measuring devices should be encouraged. The use of a gas loop containing Freon provides a possible means of continuous measurement of the fast-neutron

fluence. $Ti^{46}(n, p)Sc^{46}$ is a threshold detector in widespread use in Western Europe and the United States. This does not have the burn-up problem of nickel nor the problem of competing thermal neutron activation of iron, but the cross-section information on this isotope is quite scanty and rather inconsistent. There are in addition the detectors under development utilizing the (n, n') process in indium, niobium and rhodium. The perfection of these detectors would be particularly desirable since they offer the possibility of monitoring in the energy range below that for the threshold detectors mentioned in Table I.

C. Intermediate neutrons

1. Methods currently used

The members of the Panel were in agreement that methods currently used for determining the intermediate neutron flux density were all unsatisfactory. In particular, the cadmium difference method suffers from at least two fundamental inadequacies. The effect of the cadmium itself causes a flux density depression and therefore the process of measurement modifies the experimental environment and furthermore, the analysis of the cadmium difference activation normally assumes a strictly $1/E$ distribution of the intermediate flux density. Calculations would indicate that this assumption is not warranted, especially if the energy at the resonance is orders of magnitude different from the energy at which the flux density is required. Therefore, although this section contains a description of methods used for intermediate neutron measurements, the inclusion of these methods should not be construed as a recommendation.

The determination of the flux density in the region of intermediate neutrons may be performed by using the cadmium difference method. In this method the relative activation of a material with and without a cadmium shield for thermal neutrons is used to estimate the intermediate neutron flux density. In this connection attention is called to Section IV. B. (c) of the recommendations with regard to the information on the cadmium shield geometry which should be reported in all cases.

The intermediate neutron energy distribution is generally assumed to be inversely proportional to the energy. A quantity, ϕ_0 , equal to the flux density per unit $\ln E$ interval is defined and if the previous assumption is correct it should be constant for the entire intermediate energy range.

There is a variety of methods by which ϕ_0 may be expressed in terms of the cadmium ratios and monitor activations. Procedures widely used include those of DANCOFF *et al.* [2] and WESTCOTT *et al.* [3]. A review of methods may be found in the document by ZIJP [4] a summary of which is produced in this Report.

Some of the detectors most commonly used are listed in Table II together with the energy of the principal resonance and the half-life of the activation product.

TABLE II

INTERMEDIATE NEUTRON DETECTORS

Isotope	Resonance energy (eV)	Half-life of product
In ¹¹⁵	1.457	54 min
Mn ⁵⁵	337	2.58 h
Au ¹⁹⁷	4.9	64.8 h
Co ⁵⁹	132	5.24 yr

2. Methods under development

There is a variety of methods under development which have potential advantages over the use of the detectors listed above. Two of the methods may eliminate the problem introduced by the cadmium shield while the third permits continuous measurement of the intermediate flux density.

The continuous measurement is made with the so-called self-powered probes. These involve placing a material, such as rhodium, in a cadmium-covered probe and measuring the current flow through the probe from the emission of beta rays accompanying the decay of the activation products.

One of the methods which dispenses with the cadmium-cover requirement involves the irradiation of a sandwich of three foils of the same material. The outer two foils shield the inner foil and the relative activation of the inner and outer foils is used in the calculation of the intermediate neutron fluence.

An alternative method entails the simultaneous irradiation of two foils with cross-sections of different energy dependence. Examples are the pairs Au-Cu, Ag-Co, and Na-P. The activation from one foil is used to estimate the $1/v$ activation in the other foil. Subtracting out the $1/v$ contribution to the total activation permits more accurate estimation of the activation due to the resonance peak. The activation from the resonance peak is then used to calculate the intermediate neutron fluence.

D. Thermal neutrons

1. Routine detectors

For the measurement of fluence during long-term irradiation and for high-flux reactors, the reaction $\text{Co}^{59} (n, \gamma) \text{Co}^{60}$ may be used. It is necessary for fluences greater than 10^{20} to correct for the burn-up of the Co. The cobalt may be in the form of the pure metal or, when necessary to limit the activity of the detector, in the form of aluminium-cobalt alloys.

For the measurement of fluence during short-term irradiations, gold, dysprosium, indium and copper may be used. The use of indium and dys-

prosium is suggested particularly for the measurement of very weak fluences (e. g. zero power reactors).

2. Methods under development

(a) For the measurement of the temperature of the Maxwell spectrum a method should be used which combines in the reactor core or reflector a $1/v$ cross-section detector and a detector with resonance in the thermal energy region (e. g. a combination of Au¹⁹⁷ and Lu¹⁷⁵ detectors);

(b) The suggested method for continuous flux density measurements uses electron-collecting probes employing rhodium, silver, and vanadium, which are chosen for their short activation period and the high energy of the electrons emitted by the isotope formed;

(c) Continuous flux density measurements could also be done with an argon gas loop.

III. DETERMINATION OF ENERGY ABSORBED FROM REACTOR RADIATION

A. Energy absorption processes

Materials irradiated in reactors absorb energy in the following processes:

(a) Absorption of gamma radiation

Detailed spectra of gamma radiation in reactors are seldom known, but the spectra extend over the energy range 10 MeV-keV, with a mean energy of 1 MeV to 2 MeV.

(b) Scattering of fast neutrons

Neutrons with energies in the range 10 MeV to 0.1 MeV contribute significantly to this effect. Spectra in this range may be calculated and can be confirmed by the use of threshold detectors above about 2 MeV.

(c) Nuclear reactions, which occur in the sample irradiated and in surrounding materials

The effect is produced by absorption of prompt radiation emitted (e.g. from (n, p), (n, α), (n, f) and (n, γ) reactions) and of radiations emitted in the decay of radioactive products formed. These reactions may be induced by fast neutrons but reactions of thermal and resonance neutrons usually make a greater contribution to the energy absorbed. Neutron spectra in the thermal and intermediate region are well defined and may be readily measured.

Rates of energy absorption most commonly found lie in the range 10^{-3} to 10 W/g (i. e. 10^{16} - 10^{20} eV/g s or 10^2 - 10^6 rad/s).

B. Definition of irradiation conditions

To define the irradiation conditions used information is always required on (i) the quality of the radiation, and (ii) the quantity of the radiation [5].

1. It is suggested that the information used to define the quality of radiation should be spectra.

2. Two alternative procedures may be adopted to define the quantity of radiation:
 - (a) flux density, or fluence, may be used;
 - (b) absorbed dose or absorbed dose-rate in a reference material may be used.

If absorbed dose is used to define irradiation conditions, then the quantity quoted must refer to the material of the absorber only, and must be independent of its shape and size. The procedure (b) may be used for fast neutrons and for gamma radiations.

('Absorbed dose' may be used to define the quantity of thermal neutron radiation if the energy absorbed in the reference material arises chiefly from thermal-neutron-induced reactions. For this, materials containing B^{10} , Li^6 and U^{235} may be used. However, we consider that this is a special and not a general case).

C. Assessment of radiation heating

It is necessary to determine the total heating from all the processes discussed in section III.A. An accuracy of $\pm 20\%$ is usually satisfactory for this purpose and therefore factors such as secondary radiation loss and self-shielding are not usually important in any measurements made. (Self-shielding however may be important in equipment design). It may be necessary to know the energy absorption from each of the sources listed in Section III.A, and to have some definition of irradiation conditions if an extrapolation of data to different materials is required.

D. Studies in radiation chemistry, radiation biology, corrosion, etc.

Such studies require the determination of the total energy absorbed or, more likely, of the energy absorbed from each of the sources listed in section III.A. An accuracy of at least $\pm 5\%$ is required to match the accuracy of analytical techniques. In this case secondary radiation loss and self-shielding are important effects. It is essential to ensure that the quantity determined is the energy absorbed in the experimental sample used; its size, shape and the nature of the surrounding materials must all be considered. Information is required for experiments on solid, liquid and gaseous systems.

E. Determination of absorbed dose or dose-rate

1. The absorbed dose-rate may be calculated from first principles, that is, from a knowledge of fuel ratings, reactor geometry, prompt gamma spectrum from fission, etc.
2. The absorbed dose-rate may be calculated from other measured data. For example, the dose from fast-neutron scattering may be calculated from known fast-neutron spectra and measured fluence.
3. Measurement of the absorbed dose and/or the dose-rate

(a) Calorimeters

Calorimeters measure the rate of heating in an absorber and thus provide an absolute measurement of the energy absorbed. In adiabatic calorimeters the rate of heating is measured directly. In isothermal calorimeters the rate of heating is determined by measuring the heat transfer coefficient between the sample and the jacket and the steady-state temperature difference. In compensated calorimeters the quantity measured is the electrical heating which has to be added to a suitably designed reference container to produce the same temperature conditions observed in a sample heated by radiation. In all three types it is often possible to reproduce the conditions of the sample irradiation and thus satisfy the requirements in section D. The precision possible for these methods when used in reactors appears to be within $\pm 5\%$.

The advantages of the adiabatic instrument are that low melting-point or unstable absorbers (e. g. hydrogenous materials) may be used without "canning" and that the heat transfer coefficient need not be determined. The advantages of the isothermal and the compensated calorimeters is that they may be used for continuous measurement or monitoring of the dose-rate.

Any one calorimeter, of any of the types considered, is suitable only for a limited range of dose-rates. To find the dose-rates due to the sources listed in section III.A. different absorbers must be used and the results analysed.

The choice of the type and design of calorimeter thus depends on the materials to be irradiated, the length of the irradiations, the dose-rate, the ambient temperature, the relative intensities of gamma rays and fast neutrons and the thermal neutron flux density.

(b) Ionization chambers

An ionization chamber measures the ionization produced in a gas volume which may be accurately defined by correct design. The dynamic range of such a detector is large (greater than six decades in favourable circumstances) and it will respond almost instantaneously to changes in the incident radiation intensity. Chambers may be designed with small dimensions, having a long life in a reactor environment, the main limitation often being set by the characteristics of the electrical connectors. The ionization current produced in the gas is proportional to the absorbed dose-rate in the chamber materials.

For gamma radiation incident on the chamber, the ionization rate in the gas may be made accurately proportional to the absorbed dose-rate in the chamber walls over a wide range of gamma energies by a suitable choice of gas, wall materials and dimensions, conforming with the Bragg-Gray conditions, since W for electrons is known to be independent of energy in many gases. However, although accurate values of W are reported in the literature, it is usually necessary to compare gamma ionization chambers with other dosimeters, making a correction for the difference between the absorption coefficients of the dosimeter and the chamber wall material. Chambers are currently employed which have carbon walls and are filled

with CO₂; designs have also been described which have gas and walls made from carbon-hydrogen materials.

An ionization chamber will also respond to fast neutrons incident on it, due to ionization by recoil nuclei produced both in the gas and the walls. In some designs the contribution of wall recoils can be reduced to a small proportion and then the fast neutron response is essentially that of the free gas. For other carbon-hydrogen materials, the design of homogeneous chambers is possible.

It may be possible to separate the contribution of gamma radiation and fast neutrons in a chamber by comparing its output with that of a similar chamber filled with a gas for which the fast neutron response is much smaller. This can be done, with suitable corrections, if the gamma response of the second chamber is matched to that of the first, for example by the use of external shields to correct an enhanced low-energy response due to photo-electric absorption of the incident radiation in the gas or wall. Where the fast neutron contribution is known to be small it may be possible to dispense with a second chamber.

Ionization chambers having the characteristics mentioned above can be designed with a typical sensitivity of 10^{-6} to 10^{-8} A/mW.g in the wall material. Instruments with graphite or other stable wall materials may be suitable for use at absorbed dose-rates up to 10^3 mW/g and temperatures up to perhaps 500°C. For chambers with carbon-hydrogen walls and gas the absorbed dose-rates and temperatures attainable will be much less. For operations over a wide range of temperatures and/or dose-rates the collector electrode and its external connections should be adequately guarded to minimize electrical leakage effects, since the potential difference between the electrodes may exceed 1 kV.

A major uncertainty in the application of ionization chambers to fast neutron dosimetry concerns the value to be ascribed to W for ionization produced by heavy recoil nuclei in gases which are commonly used e.g. CO₂, argon and carbon-hydrogen gases. More work is required to establish the energy dependence of this parameter over a wide range of neutron energies.

(c) Chemical dosimetry

The amount of chemical change induced by radiation can be used as a measure of the energy absorbed. Chemical dosimeters require calibration, but their advantage is that they can often exactly reproduce the conditions of sample irradiation. The choice of a chemical system which can be used as a chemical dosimeter is fairly limited. In addition to the usual properties needed for a system to be a chemical dosimeter other conditions should be satisfied. The system chosen should have:

- (i) a chemical composition which results in a low level of induced radioactivity;
- (ii) the possibility of measurements in the multi-megarad absorbed dose region, to cover the high dose-rates found in reactors;
- (iii) a radiation yield independent of temperature increase during irradiation, which results from the high dose-rates; and

- (iv) a radiation yield independent of linear energy transfer (LET) over a wide range of energies or, if this is not the case, a known LET dependence.

We can see the method for measuring the total absorbed dose in CH₂ materials (cyclohexane), BOYD, this Report, and a method for water and aqueous solutions (oxalic acid in H₂O or D₂O), DRAGANIĆ, this Report. The latter enables one to assess the dose-rates from (a) and (b) (listed in section III. A. 1) in H₂O and D₂O and hence, by calculation, in other materials of low atomic number.

F. Standardization

Dosimetry problems are specific to the investigation being performed, rather than general in nature. In principle, therefore, they cannot be standardized. However, principles can be distinguished which must be adopted if dosimetry is to be adequate, problems which will usually be encountered can be recognized, and techniques discussed which are likely to produce the information required. Thus it should be possible to produce a handbook similar to the National Bureau of Standards Handbook, NBS No. 85, produced by the International Commission on Radiological Units (ICRU), but dealing specifically with reactor radiation problems.

It is, in principle, possible to introduce some standardization of the measurement of absorbed dose for the definition of irradiation conditions, as discussed in section III. B. 2. (b). Since the method is suitable for defining fast neutrons and gamma radiation, but in general not for thermal neutron radiation, it would be necessary to determine the absorbed dose in materials in which the contribution from nuclear reactions (section III. A. 3.) is negligible. Since all materials placed in a reactor absorb gamma radiation, the use of two absorbers can be considered, one with a high dose-rate from fast neutron scattering and one with a low dose-rate from fast neutron scattering. The use of a hydrocarbon and carbon might be considered. Since absorbed dose can probably be measured more accurately than gamma spectra, absorbers of high atomic number and high spectral sensitivity should not be used. The absorbed dose-rate measured in the selected materials must be a quantity which refers to the material of the absorber and is independent of the size and shape of the sample used. The quantity required is referred to by the ICRU as kerma (NBS No. 84). We should consider ways of measuring this quantity using the methods outlined in section III. E. Exact determination may not be possible because of the very wide range of reactor radiation spectra, and better knowledge of gamma spectra is required before a decision can be reached on this point.

It must be emphasized that the definition of irradiation conditions is not the same thing as the determination of absorbed dose, even if absorbed dose quantities are used in the definition. For example, definition of the irradiation conditions by a knowledge of spectra and measurement of absorbed doses in suitable reference materials will not give the absorbed dose (a) in small samples of KCl irradiated in aluminium containers or (b) in samples of hydrocarbon gases irradiated in quartz vessels.

IV. RECOMMENDATIONS

The Panel's recommendations fell into two categories: (i) those intended for the International Atomic Energy Agency (IAEA), suggesting possible programmes dealing with dosimetry; and (ii) those intended for individual investigators working in this field dealing with practices whose widespread adoption appear desirable for greater consistency and standardization.

A. Recommendations to the IAEA

1. Fluence measurements

Since activation detectors are universally used for fluence measurements, the Panel recommends that the IAEA compile a handbook of practical information, for example,

- (a) A list of sources of supply of very pure detector materials;
- (b) Technical reports on counting procedures for the nuclides of interest;
- (c) Technical reports on procedures for neutron dosimetry and neutron metrology.

Because of the needs of many scientific and technical programmes in the field of radiation damage in solids, the Panel recommends that the IAEA encourage the development of methods for the measurement of neutron flux and fluence in the keV-energy range.

Since nickel is widely used as a fast neutron detector, the Panel recommends that the IAEA encourage the acquisition of accurate cross-section measurements for the thermal and epithermal neutron reactions which are necessary for the evaluation of burn-up of Co^{58} and Co^{58m} .

2. Dosimetry

The Panel recommends:

- (a) that a handbook be produced giving an initial assessment of the applications of methods of measuring absorbed dose in reactor environments;
- (b) that two reference materials be adopted for use in defining irradiation conditions by the determination of absorbed dose, as outlined in Section III.B. One of these materials should be carbon as graphite, as it is obtainable in a sufficiently pure state and is suitable for use in a number of types of instruments. A second reference material should be sought, preferably with a higher dose-rate from fast neutron scattering than carbon, i. e. with a lower atomic number.

Reference methods should be established for the determination of the absorbed dose in these materials, by either

- (i) calorimetry,
- (ii) ionization chambers,
- (iii) chemical methods,

or a combination of these methods.

A bibliography should be compiled to support this work.

3. Standardization

The Panel recommends that the IAEA solicit the co-operation of standards laboratories to prepare and distribute on a world-wide scale some of the radioactive isotopes of relevance to in-pile fluence and spectra measurements. The following standards laboratories should be contacted:

Bureau Central de mesures nucléaires, Euratom, Geel, Belgium

Bureau International des Poids et Mesures, Sèvres, France

Mendeleeff Institute for Metrology, Leningrad, USSR

National Bureau of Standards, Washington, D.C., United States of America

National Physical Laboratory, Teddington, United Kingdom

National Research Council, Ottawa, Canada

Physikalisch-Technische Bundesanstalt, Braunschweig, Federal Republic of Germany

International Atomic Energy Agency Laboratory, Seibersdorf.

As a second step, the Panel recommends the establishment of standard specifications for the standard detectors to be prepared (dimensions, activity, accuracy, etc.)

4. Intercomparison of instruments and techniques

Where close study showed it to be desirable the inter-comparison of instruments and techniques in different reactors should be made. Reactors which do not in general have a heavy loading of high cross-section materials perturbing the spectra will be particularly suitable for such work. These reactors are more likely to be found in the developing countries.

In the case of threshold detectors, the intercomparison should be carried out with simultaneous irradiation of several threshold detectors in spectra comparable to the fission spectrum (converter reactors, gap between plates in the core of an aqueous reactor, etc.) The measurement of their absolute activity then shows the relationship between their cross-sections averaged over the fission spectrum.

5. Catalogues of neutron and gamma spectra

The Panel stresses the importance of knowing the spectral distribution of neutrons in the irradiation facilities. With this in view the Panel recommends the gathering together in a single document of all information currently available on neutron spectra in the various types of reactor. This information might cover the following points: calculated spectra with a description of the code and cross-sections used; and measured spectra with a description of the method used and possibly comparison with the calculated results.

The Panel also recommends that a catalogue of calculated and measured reactor gamma spectra be compiled.

6. Symposium

The Panel recommends that the Agency convene a scientific meeting on in-pile dosimetry and reactor spectra in late 1965 or 1966. (Suggested title: "Calculation and measurement of reactor spectra and their application to in-pile experiments").

7. Terminology

The Panel recommends that close collaboration be maintained with ICRU on terminology, definitions and units used in this field.

B. Recommendations to investigators

The following recommendations are intended for the individual investigator who is exhorted to comply with them wherever possible.

General

Direct measurement of the energy in the actual samples irradiated should be done whenever possible. Also, precise information should be given about the irradiation conditions: fuel elements configuration, loading of the pile, etc.

Fluence measurements

In many instances, as more complete information is acquired, it becomes feasible to re-analyse flux and fluence measurements. To facilitate such a re-analysis by an independent investigator, information should be reported in sufficient detail to permit such a re-analysis. The following specific items should be considered as the very minimum requirement:

- (a) That the original data on the measured saturation activities (disintegration rate/ $1 - \exp(-\lambda t_1) = n\sigma\phi$) of the activation detectors per gram of target nuclide should always be listed;
- (b) That the values for the cross-sections which are used in the calculations (such as resonance integral cross-sections, average fission neutron cross-sections, or effective neutron cross-sections) should always be mentioned, together with a reference to the origin of the value used; and
- (c) That for cadmium ratio measurements the type and dimensions of the cadmium covers used be mentioned, together with the value used for the cadmium cut-off energy.

It was generally agreed that the information about the fast neutron spectrum was of great importance both to the estimation of damage to solids and to the measurement of absorbed dose. In order to secure an experimental indication of the spectral shape, the Panel recommends that, where possible, two or more activation detectors with different threshold energies should be used. It is noted that $P^{31}(n,p)$, Si^{31} , $S^{32}(n,p)P^{32}$, $Ni^{58}(n,p)Co^{58}$ all have

thresholds near 3 MeV and that the reaction $\text{Al}^{27}(\text{n}, \alpha)\text{Na}^{24}$ can monitor the higher energy component.

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STATUS REPORTS

A STATUS REPORT ON IN-PILE DOSIMETRY IN CANADA

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INTRODUCTION

The in-pile dosimetry work in Canada consists of (a) thermal, intermediate and fast-neutron flux measurements by activation methods; (b) thermal-neutron flux monitoring by self-powered detectors; and, (c) calorimetric, ion chamber and chemical measurements of energy absorbed due to neutrons and gamma rays. Work in each of these three fields is summarized in this report.

ACTIVATION MEASUREMENTS OF NEUTRON FLUX

Neutron flux measurements with cobalt

EASTWOOD et al. [1, 2, 3] have investigated the use of cobalt as a thermal-neutron flux monitor. Self-shielding due to both thermal neutrons and resonance capture in cobalt have been determined by irradiations with and without cadmium for different sizes of cobalt wires and cobalt-aluminium alloys. Techniques for rapid and accurate counting of samples over a wide range of activities have been developed. Using the above corrections the effective cross-section for cobalt-59 can be calculated using the Westcott convention [4] and it is found that the accuracy depends mainly on two factors. The first of these is the uncertainty in the 2200 m/s cross-section for cobalt and the second is the accuracy of the absolute disintegration rate of the cobalt standards used for counter calibration. At present values of this cross-section range from 36.5 b to 38 b. It is hoped to reduce this uncertainty by comparisons with gold in the thermal columns of the Chalk River reactors.

Flux surveys of the NRX reactor using cobalt have been carried out by JERVIS [5], BOCK and BOYD [6] and BOYD et al. [7].

Fission neutron flux measurements

ROY et al. [8, 9] determined effective cross-sections for several (n, p), (n, α) and (n, n) reactions in the neutron flux in the NRX and NRU reactors.

Roy also estimated activation cross-sections for a fission neutron spectrum for all the stable isotopes of elements from lithium to bismuth.

BUTLER and SANTRY [10, 11, 12] have used monoenergetic neutrons to determine cross-sections for several elements in the region 1 MeV to 20 MeV. The cross-sections from the threshold energy to 20 MeV have already been determined for the following reactions: $S^{32}(n, p)P^{32}$, $Th^{232}(n, 2n)Th^{231}$, $Al^{27}(n, \alpha)Na^{24}$, $Mg^{24}(n, p)Na^{24}$, $Cu^{65}(n, p)Ni^{65}$, $Cu^{65}(n, 2n)Cu^{64}$, and work is proceeding on the following: $Zn^{64}(n, p)Cu^{64}$, $Fe^{56}(n, p)Mn^{56}$, $Co^{59}(n, \alpha)Mn^{56}$, $Sc^{45}(n, \alpha)K^{42}$, $Sc^{45}(n, 2n)Sc^{44m}$ and $Sc^{45}(n, 2n)Sc^{44}$. The neutrons were obtained by the reactions: $T(p, n)He^3$, $D(d, n)He^3$ and $T(d, n)He^4$. The neutron energy was varied by irradiating samples at various angles to the deuteron beam and varying the energy of the deuteron beam. The accuracy of the cross-sections (relative to $S^{32}(n, p)P^{32}$) in these measurements is generally within 4%.

CROSS [13] has measured cross-sections for the $Ni^{58}(n, p)Co^{58}$ reaction and the $In^{115}(n, n')In^{115m}$ reaction using neutrons of 2 MeV and 14 MeV energy. He has also discussed the use of a number of threshold activation detectors for dosimetric measurements of fast neutrons of energies of 1 to 16 MeV.

Intermediate neutron flux measurements

BIGHAM, GREEN *et al.* [14-17] have measured the relative reaction rates in Mn^{55} , In^{115} and Lu^{176} with and without cadmium to determine neutron temperatures and epithermal indices in the fuel and moderator of various lattices. These measurements were made for the design and burn-up predictions of power reactors, such as NPD and CANDU. Further work on different lattices is being done.

HART, BIGHAM and MILLER [18] have shown that the epithermal flux can be measured by the activities induced in silver and cobalt, thus providing an alternative to irradiations under cadmium.

There are only two research reactors in Canada outside of Chalk River - a 1-MW pool reactor at McMaster University and a subcritical assembly at the University of Toronto. Flux measurements with cobalt and gold have been made at McMaster University. At Toronto, traversing BF_3 counters have been used for flux surveys [19]. Fine structure and thermal utilization in lattices have been determined by thermal and epithermal activation methods [20] and gamma radiation in the reactor core has been measured with a scintillation spectrometer [21].

IN-CORE NEUTRON FLUX MONITORING BY SELF-POWERED DETECTORS

HAWKINGS [22] has used the output of a thermocouple mounted in a block containing uranium to give a continuous measure of thermal neutron flux. The thermocouple (iron-constantan stainless-steel sheathed) was in an insulated zircaloy cylinder containing a natural uranium metal wire. The useful range of this instrument extended from 2×10^{12} to 4×10^{13} n/cm²s and it was operated successfully to an integrated flux of 2.8×10^{20} nvt. The overall size was 1-in diam. by 5 in long.

HILBORN [23] has developed self-powered neutron detectors consisting of a beta emitter along the axis of a coaxial cable surrounded by insulant and a collector. These are made by replacing a short length of the central conductor in a commercial coaxial cable with vanadium or rhodium wire. The rhodium forms 42-s Rh^{104} and the vanadium, 3.8-min V^{52} . Magnesium oxide insulation is used to permit reliable operation in high temperature, high flux environments. These detectors have been used up to temperatures of $360^{\circ}C$ and in fluxes as high as 1×10^{14} for more than a year. A typical rhodium detector, 30 mm long and 1.2 mm O.D. produces $0.3 \mu A$ in a thermal flux of 10^{14} n/cm²s at a response time of 1 min. These monitors are now widely used at Atomic Energy of Canada Ltd. (AECL) for reactor flux mapping and for continuous flux monitoring in loops and other experimental assemblies. Future work is aimed at producing shorter response times by using (n, p) or (n, α) reactions.

CALORIMETRIC, ION CHAMBER AND CHEMICAL MEASUREMENTS

BOYD *et al.* [7, 24] have measured gamma-ray and fast-neutron dose-rates in the NRX reactor using calorimeters, a graphite-CO₂ ion chamber and the hydrogen yield from the radiolysis of cyclohexane.

The calorimeters are adiabatic, simple in construction and give precisions of 0.5 - 3.5% in the range 100 - 500 mW/g.

The ion chamber is sealed in quartz with the electrical leads brought out through molybdenum quartz seals. It has been used over the range 0.1 - 500 mW/g.

The yield of hydrogen from cyclohexane has been found to be constant within $\pm 5\%$ for doses and dose-rates in the ranges $2-10 \times 10^{21}$ eV/g and 1-2000 mW/g. It is also independent within 5% of the ratio of fast neutrons to gamma rays.

The results obtained with the three methods agree within 5 - 10%.

Ion chambers with a greater range are now being developed.

Hilborn has constructed self-powered gamma flux monitors based on the methods of GROSS and MURPHY [25]. A monitor with a 2-in-long by 0.12-in diam. lead core, a polyethylene insulator and an aluminium sleeve gives a current of 10^{-9} at an energy deposition rate of 5 mW/g.

Future work is aimed at developing detectors for high-temperature environments. The major problem is to find suitable materials with low thermal-neutron cross-sections.

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ÉTAT ACTUEL DE LA DOSIMÉTRIE EN FRANCE

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I. CONSIDÉRATIONS GÉNÉRALES

Avant d'aborder le sujet de ce rapport, il est utile de préciser le sens du terme «dosimétrie», ce mot n'étant généralement pas utilisé dans son acception étymologique. Il est devenu d'usage courant d'entendre par dosimétrie tout ce qui concerne la mesure des rayonnements nucléaires: flux neutronique et gamma, radioactivité alpha, bêta ou gamma. Les buts poursuivis correspondent à la détermination des spectres énergétiques, des flux instantanés, des flux intégrés dans le temps (fluence) ou des doses d'énergie absorbées dans les échantillons. En toute rigueur, c'est uniquement à ce dernier type de mesure que devrait s'appliquer le terme «dosimétrie» et c'est d'ailleurs ainsi qu'on l'entend surtout dans certaines spécialités, en biologie notamment. En pratique, la sobriété de l'expression et sa consonance non rébarbative (comme ce serait le cas, par exemple, pour «fluxométrie»), ont conduit de nombreux physiciens à l'utiliser dans le

sens général de « mesure des rayonnements nucléaires ». C'est donc également dans ce sens que nous l'emploierons ici, mais nous nous limiterons surtout aux méthodes utilisées dans et autour des piles.

Les techniques de la dosimétrie intéressent évidemment de nombreux spécialistes, dans des buts différents: physiciens nucléaires, physiciens des piles, physiciens de la protection contre les rayonnements, biologistes, physiciens du solide et métallurgistes, etc. Cette situation a provoqué une certaine dispersion des efforts et une multiplication d'études voisines sinon semblables, sans que les échanges et les liens indispensables en pareil cas aient toujours été suffisamment étudiés. L'inconvénient majeur d'une telle dispersion, outre les doubles emplois qu'elle provoque, est de rendre extrêmement difficile la comparaison des résultats expérimentaux, chacun utilisant ses propres méthodes et « tours de main » et les constantes de base (sections efficaces, par exemple) qu'il juge les meilleures. Les complications auxquelles il fallait s'attendre n'ont pas tardé à apparaître, et de nombreuses tentatives de coordination se sont développées depuis quelques années, tant à l'intérieur de certains pays que sur les plans européen et international.

Les moyens d'information et d'échanges qui en résultent, pour incomplets qu'ils soient encore, permettent d'avoir une vue d'ensemble assez précise de l'état actuel de la dosimétrie dans les piles [1]. Nous ferons donc ici le point de ces problèmes, en distinguant ce qui est considéré comme acquis de ce qui présente encore des difficultés et fait actuellement l'objet d'études. Pour cela, nous diviserons le sujet de la manière suivante:

neutrons thermiques:	$0 < E < 0,55 \text{ eV}$;
neutrons intermédiaires:	$0,55 \text{ eV} < E < 1 \text{ MeV}$;
neutrons rapides:	$E > 1 \text{ MeV}$;
rayonnement gamma.	

A l'intérieur de chaque catégorie, nous examinerons ce qui a trait à la mesure des spectres, des flux instantanés, et des doses intégrées dans le temps.

II. NEUTRONS THERMIQUES

La mesure des flux instantanés de neutrons thermiques a été très étudiée depuis de nombreuses années, et les sections efficaces dans ce domaine sont assez bien connues. La définition généralement adoptée pour ce flux est celle du flux « conventionnel », dit encore « flux à 2200 m/s »: $\phi_{th} = n v_0$. n est la densité neutronique entre 0 et 0,55 eV; il sera donc nécessaire d'effectuer une correction si la mesure sous cadmium, destinée à séparer l'effet des neutrons intermédiaires, est réalisée de manière telle que la coupure effective n est pas exactement à 0,55 eV.

v_0 est, par convention, la vitesse de 2200 m/s; c est la vitesse qui correspond au maximum d'un spectre de Maxwell à 20°C.

La détermination de la densité est immédiate avec un détecteur en $1/v$ irradié nu et sous cadmium. On obtient ainsi la densité réelle, indépendam-

ment de la température d'équilibre du spectre de Maxwell. Si la section efficace des détecteurs utilisés n'est pas en $1/v$, il faut effectuer une correction qui dépend de la température.

Les détecteurs les plus couramment utilisés pour les mesures relatives sont Cu, Au, In, Co, Mn, Dy. Leur préparation sous forme de détecteurs épais ou de dépôts minces est maintenant bien au point. Cu et Au sont parfois également utilisés, sous forme de bandes ou de fils pour le premier, sous forme de fils pour le second, lorsqu'il est nécessaire d'étudier de manière fine la répartition spatiale d'un flux. Pour les mesures absolues on emploie surtout Au, Cu et Co. La détermination absolue de l'activité se fait généralement à l'aide d'un compteur $4\pi - \beta$ pour Cu, en coïncidences $\beta - \gamma$ pour Au, et en coïncidences $\gamma - \gamma$ pour Co.

Il faut également citer pour la mesure des flux instantanés les moyens classiques mais dont l'utilisation est généralement limitée à des niveaux de flux assez faibles ($< 10^{10}$ ou 10^{11}): chambres au bore, compteurs à BF_3 , chambres à fission. Une mention particulière doit être faite des systèmes qui permettent une mesure continue des flux à niveau élevé: boucle à argon [2], sonde à rhodium [3]; l'intérêt de ces dispositifs est très grand si l'on veut suivre en permanence la valeur du flux en un point quelconque d'un réacteur, et ceci explique que plusieurs laboratoires poursuivent actuellement des études de mise au point dans ce domaine.

En ce qui concerne les substances dont la section efficace n'est pas en $1/v$, le calcul d'un taux de réaction ne peut pas être fait à partir de la seule donnée du flux « conventionnel », qui ne constitue qu'une information sur la densité neutronique globale au-dessous d'une certaine énergie. Il faut, dans ce cas, connaître en plus la répartition énergétique des neutrons, c'est-à-dire la température du spectre de Maxwell. Cette situation est celle, en particulier, de la plupart des corps fissiles du fait de l'existence de résonances se traduisant par une déformation de la section efficace dans le domaine thermique. La détermination de la température du spectre de Maxwell peut être faite, en principe, à l'aide de deux détecteurs, l'un ayant une section efficace non en $1/v$, l'autre étant en $1/v$; le rapport des activités prises par les deux détecteurs varie en fonction de la température du spectre et peut donc servir à déterminer celle-ci [1]. Il faut dire toutefois que l'interprétation de telles mesures est, dans l'état actuel des études, souvent délicate. Les méthodes employées ou en cours de mise au point utilisent surtout, comme détecteur en $1/v$, ou proche de cette loi, l'un des corps suivants: ^{175}Lu , Mn, Au; et comme détecteur non en $1/v$ ^{176}Lu , Eu, ou Gd. Le couple des deux isotopes de Lu semble devoir donner de bons résultats et présente l'avantage de permettre la mesure à partir d'un seul détecteur. L'intérêt du ^{176}Lu est d'ailleurs généralement reconnu, car il présente une forte résonance à basse énergie (0,142 eV); sa section efficace s'éloigne donc très nettement de la forme en $1/v$ dans le domaine du spectre de Maxwell. Le couple U-Pu semble également très intéressant puisque l'on peut ainsi obtenir des résultats directement exploitables pour les combustibles, c'est-à-dire dans le domaine où la connaissance du spectre aux basses énergies est primordiale.

Nous examinerons maintenant le problème de l'intégration des doses de neutrons thermiques (mesure de la fluence). Le choix s'est jusqu'à pré-

sent porté très souvent sur Co parce que cette méthode a fait l'objet de très nombreuses études; elle est par conséquent bien connue et se prête parfaitement à une utilisation généralisée. En outre, la période de 5,3 a convient à la très grande majorité des applications, et Co peut être fourni facilement dans un état de pureté convenable, et être préparé sans difficulté sous des formes diverses. Bien que l'emploi de Co tende à se généraliser, il est intéressant de dire quelques mots des autres méthodes possibles [4], et particulièrement de celles qui sont susceptibles d'être utilisées là où Co ne convient plus très bien. Les études menées actuellement dans divers laboratoires concernent surtout:

a) L'utilisation des alliages Al-Co. L'intérêt est ici de pouvoir disposer, sous une forme aisément manipulable, de très faibles quantités de Co, afin de ne pas avoir à travailler sur des détecteurs de très grande activité comme cela se produit lorsqu'on doit effectuer l'intégration de doses très élevées. Le principe d'utilisation est évidemment le même que pour Co pur, mais une difficulté pratique intervient pour la détermination précise de la quantité de Co contenue dans l'alliage Al-Co.

b) La mesure de la disparition de noyaux d'Au. La réaction (n, γ) en neutrons thermiques sur ^{197}Au donne naissance à ^{198}Hg qui, étant stable et ne présentant qu'une très faible section efficace, s'accumule. La mesure de la quantité d'Au disparue (par réactivation ultérieure du détecteur, par exemple), ou de la quantité de Hg formée (par analyse spectroscopique), permet en principe de déterminer la dose intégrée au niveau du détecteur. La méthode est bien adaptée aux doses très élevées, supérieures à 10^{21} n/cm²; elle peut donc constituer un relais intéressant pour Co dans cette gamme des très hautes doses. Il reste cependant de nombreux essais à effectuer pour vérifier que les impuretés contenues dans Au ne perturbent pas la mesure.

c) La spectrométrie alpha sur U irradié. Le grand intérêt de cette méthode sera de permettre une détermination directe du taux de fission et de la production de Pu dans un élément de matière fissile. Le dispositif de mesure utilise comme détecteur une diode semi-conductrice spéciale; la résolution obtenue lors des premiers essais permet d'espérer une bonne discrimination des rayonnements alpha provenant de ^{235}U , ^{238}U et de ^{239}Pu . L'un des problèmes actuellement en cours d'étude a trait à la préparation de dépôts convenables d'U irradié.

d) L'utilisation de détecteurs fissiles et comptage gamma sur la raie d'un produit de fission. Le comptage est réalisé généralement sur la raie Cs, ou globalement au-dessus d'une certaine énergie située vers 500 keV; la difficulté réside essentiellement dans la détermination absolue de l'activité.

Un dernier point à signaler à propos des neutrons thermiques concerne la réalisation des empilements étalons. Leur utilité apparaît de manière évidente dès que l'on considère la simplification qu'ils apportent pour la détermination absolue des flux. En l'absence d'empilement étalon, la mesure absolue d'un flux implique la réalisation de détecteurs très étudiés (nature du corps utilisé, connaissance des impuretés, dépôts minces dans certains cas, connaissance précise des sections efficaces et des schémas de désintégration, etc.) et l'utilisation d'un matériel de comptage souvent complexe. Il est nécessaire, en outre, d'effectuer de nombreuses corrections pour tenir compte de l'autoabsorption des rayonnements émis par

le détecteur, et des erreurs systématiques introduites par le système de comptage. On conçoit qu'avec toutes ces conditions à remplir, la mesure absolue d'un flux soit une opération délicate et qu'un petit nombre seulement de détecteurs permettent d'obtenir une précision convenable. Si l'on dispose d'un empilement étalon, il devient très facile d'effectuer l'étalonnage de détecteurs très divers quelles que soient, dans une large mesure, leur nature et leur géométrie. La mesure de l'activité elle-même peut alors être faite à l'aide d'un appareillage relativement simple. Il est évidemment nécessaire que le flux soit parfaitement connu aux différents points-repères choisis à l'intérieur de l'empilement, mais cette détermination peut être faite une fois pour toutes, ou tout au moins seulement contrôlée de temps en temps. La valeur du flux dans l'empilement peut être déterminée soit par le calcul (la géométrie est volontairement choisie simple) soit par mesure (en utilisant un détecteur dont la section efficace, très bien connue, est prise comme référence). La seconde méthode doit en principe donner de meilleurs résultats, mais ceci n'est pas aussi évident qu'il peut paraître à première vue car la mesure absolue de l'activité devient difficile à réaliser avec précision lorsque cette activité est faible; une telle situation correspond bien au cas qui nous intéresse ici puisque les empilements étalons ne sont alimentés que par des sources de Ra-Be ou de Pu-Be, et ne délivrent que des flux neutroniques faibles, de l'ordre de 10^4 n/cm².s. Dans l'état actuel des techniques, et en utilisant Au comme détecteur de référence, on peut espérer obtenir une précision de l'ordre de 1% sur la connaissance des flux dans l'empilement. Récemment, plusieurs projets ont été étudiés en France pour l'installation d'empilements étalons dans les centres de Saclay, Fontenay-aux-Roses, Grenoble et Cadarache; les centres de Fontenay-aux-Roses et de Cadarache sont dès maintenant équipés. L'utilisation, que l'on prévoit journalière, de ces dispositifs justifie leur installation dans chacun des centres importants du Commissariat à l'énergie atomique. Ces divers empilements n'auront pas tous la même structure; celui de Cadarache utilise le graphite et quatre sources Ra-Be; celui de Grenoble, non encore entièrement défini, utilisera le graphite et deux ou quatre sources de Ra-Be; celui de Fontenay utilise le graphite et l'eau lourde, et deux sources de Ra-Be; celui de Saclay utilisera le graphite et une seule source (le projet initial prévoyait une source de Pu-Be, mais des publications récentes font état d'une évolution dans le temps importante des sources Pu-Be; il sera donc fort probablement nécessaire de revenir au Ra-Be). L'empilement de Saclay, dont la géométrie est la plus simple, servira sans doute d'étalon primaire.

III. NEUTRONS INTERMÉDIAIRES

Dans le domaine des neutrons intermédiaires, les travaux actuels ont surtout pour but de préciser les idées directrices pouvant servir de base sérieuse à la mesure du spectre réel entre 0,55 eV et 1 MeV environ. A l'intérieur de cette gamme énergétique très étendue où les mesures présentent un certain nombre de difficultés particulières, la situation est la suivante.

Les méthodes utilisées dans le passé étaient en réalité très rudimentaires. Basées sur l'emploi des détecteurs résonnants et sur la technique

des rapports cadmium, elles ne pouvaient constituer qu'un contrôle sommaire de la validité de l'hypothèse du spectre en $1/E$. La théorie classique montre en effet que le rapport des flux thermique et intermédiaire peut être relié aux mesures du rapport cadmium par l'expression:

$$\frac{\Phi_{th}}{\Phi_{int}} = \frac{I_R}{\sigma_{th}} (R_{Cd} - 1), \text{ où}$$

Φ_{th} est le flux thermique;

Φ_{int} est le flux intermédiaire par intervalle logarithmique d'énergie (c'est-à-dire le $q/\xi\Sigma_s$, de la théorie du ralentissement);

I_R est l'intégrale de résonance du détecteur: $\int_{E_{Cd}}^{\infty} \sigma(E) \frac{dE}{E}$ (E_{Cd} étant la coupure effective de Cd);

σ_{th} est la section efficace du détecteur en neutrons thermiques; R_{Cd} est le rapport cadmium mesuré à partir des irradiations du détecteur, nu et sous cadmium.

Si les mesures effectuées au même endroit avec des détecteurs divers donnent la même valeur pour l'expression ci-dessus, on peut effectivement en conclure que le spectre au point de mesure est bien en $1/E$. Dans le cas contraire, il apparaît difficile de déduire des mesures quelque chose de précis concernant la forme réelle du spectre. Il est vrai que les mesures courantes donnent souvent des résultats qui se recoupent, mais ceci provient surtout du fait que les marges d'erreurs sont importantes par suite de la connaissance imprécise des sections efficaces et des intégrales de résonance. De telles mesures sont par conséquent peu significatives, autant par leur principe que par leur imprécision. La connaissance des sections efficaces devenant maintenant plus précise, et le nombre de détecteurs utilisables s'accroissant, il devient nécessaire et possible d'envisager des méthodes plus élaborées.

La détermination expérimentale d'un spectre réel quelconque serait très facile si l'on disposait d'un nombre suffisant de détecteurs dont les réponses respectives en énergie soient situées dans diverses bandes très étroites. Il semble que certains détecteurs résonnants puissent répondre avec une bonne approximation à cette condition; il faut pour cela que

- l'activation due à la partie en $1/v$ de la section efficace soit faible vis-à-vis de l'activation due aux résonances;
- l'activation due aux résonances secondaires soit négligeable vis-à-vis de l'activation due à la résonance principale.

Ces deux conditions sont évidemment d'autant moins impératives que le spectre à mesurer est plus voisin de la forme en $1/E$. Mais si l'on veut définir une méthode valable dans la plupart des cas réels, il est indispensable de sélectionner les détecteurs qui remplissent les conditions citées ci-dessus. Il faut pour cela, non seulement connaître la section efficace différentielle du corps dans la gamme énergétique utile, mais aussi évaluer la contribution de chaque partie de cette section efficace (partie en $1/v$, résonance principale, résonances secondaires) dans l'activation globale du

détecteur. Ce second point nécessite, en toute rigueur, la connaissance du spectre à l'endroit de la mesure, alors que c'est précisément cela que l'on cherche à déterminer expérimentalement; la difficulté peut cependant être levée dans la plupart des cas, par une estimation à partir de la forme approchée donnée par la théorie ($1/E$, par exemple, dans les piles thermiques). Toutes ces considérations conduisent à sélectionner un certain nombre de détecteurs résonnants utilisables pour la détermination du spectre intermédiaire; à l'heure actuelle, les plus intéressants semblent être surtout In, Au, Co et Mn. Il faut noter d'ailleurs que plus la résonance principale d'un corps s'éloigne vers les hautes énergies, plus l'activation due à la partie en $1/v$ de la section efficace prend de l'importance. Ainsi pour Mn dont la première résonance est située à 337 eV, l'activation due à cette résonance est du même ordre de grandeur que celle qui est due à la partie en $1/v$ (dans l'hypothèse approchée du spectre en $1/E$). Ceci montre bien que le principe même de l'utilisation des détecteurs résonnants pour la détermination expérimentale du spectre intermédiaire limite cette technique à une gamme ne s'étendant pas très loin en énergie. Cette limite théorique est située vers 400 eV.

Entre quelques centaines d'eV et quelques centaines de keV, il n'existe pratiquement pas de méthodes permettant la mesure du spectre. Il est nécessaire de faire ici un effort d'imagination afin de dégager des principes valables. Cet effort est d'ailleurs certainement justifié, car la gamme énergétique considérée présente une importance réelle pour de nombreuses expériences. Certaines études récentes montrent qu'il est sans doute possible d'obtenir des informations significatives dans cette gamme par l'utilisation de détecteurs présentant une résonance principale à haute énergie et qui seraient irradiés sous une épaisseur convenable de bore. Le bore permet, en supprimant une grande partie des neutrons de basse énergie, de rendre à la résonance principale un rôle prépondérant dans le bilan d'activation du détecteur. Ainsi, avec Na irradié sous 450 mg/cm^2 de B naturel, l'activation obtenue devrait être due pour 90% aux neutrons situés dans la gamme 0,75-5 keV. Avec F [5] irradié sous $1,2 \text{ g/cm}^2$ de ^{10}B , l'activation devait être due pour environ 60% aux neutrons situés entre 20 et 70 keV. De tels travaux sont prometteurs mais de nombreux essais restent à faire avant que les méthodes correspondantes soient du domaine de l'utilisation courante.

Au-delà de 350 keV, et jusqu'aux énergies les plus élevées, les émulsions nucléaires peuvent rendre de grands services pour la détermination du spectre [6]. Leur inconvénient majeur, très important dans la pratique, réside dans leur grande sensibilité au rayonnement gamma, ce qui limite leur emploi aux piles ne fonctionnant qu'à basse puissance (maquettes critiques par exemple). Pour les émulsions couramment utilisées, le flux gamma au point de mesure ne doit pas dépasser 0,5 à 1 r/h. Il faut noter toutefois que de nouvelles émulsions, mises récemment sur le marché par Gevaert, présentent une sensibilité gamma environ 10 fois plus faible; en outre, ces nouvelles émulsions permettent de porter la limite inférieure des mesures à 150 ou 200 keV, avec une résolution de $\pm 30 \text{ keV}$. Le progrès n'est donc pas négligeable, mais il est encore insuffisant pour permettre

l'utilisation des émulsions nucléaires dans les réacteurs de recherche où le bruit de fond gamma est très important.

Dans cette même gamme des énergies supérieures à quelques centaines de keV, les chambres à fission deviennent utilisables (^{237}Np , ^{241}Am , par exemple). Leur mise en œuvre n'est pas toujours très facile, surtout pour les mesures absolues, mais il s'agit là de difficultés techniques et non plus de difficultés de principe.

Il apparaît, en définitive, que la détermination expérimentale des spectres de neutrons intermédiaires est une opération difficile, et l'on peut se demander si les méthodes de calcul purement théoriques ne devraient pas constituer ici un support indispensable. C'est à partir de ces considérations que diverses études ont été menées et sont actuellement poursuivies, afin de préciser la forme théorique du spectre intermédiaire dans différents types d'emplacements expérimentaux. Ces études ont déjà permis de préciser quelques points importants; nous les examinerons au chapitre V puisqu'ils ont trait à la fois aux neutrons intermédiaires et aux neutrons rapides.

On peut conclure en disant que, dans l'état actuel des techniques, la détermination des spectres intermédiaires doit faire appel à la fois aux considérations théoriques et aux procédés expérimentaux. Les résultats du calcul permettent ainsi de guider l'interprétation des mesures. On trouvera d'ailleurs au chapitre V ci-dessous un bref exposé de la méthode qui permet actuellement d'utiliser les résultats de mesures même assez sommaires, dans les domaines intermédiaire et rapide, pour l'interprétation des expériences de dommages.

Nous avons surtout évoqué les problèmes liés à la détermination du spectre intermédiaire parce qu'ils constituent une question importante et difficile. La valeur du flux intermédiaire, sur toute l'étendue de la gamme énergétique considérée ou sur une partie seulement de celle-ci, se déduit directement des mesures décrites puisqu'en fait chaque détecteur résonnant, traité de la façon décrite, permet d'obtenir le flux dans un intervalle logarithmique d'énergie situé autour de sa résonance principale.

L'intégration des doses dans le domaine intermédiaire ne fait pas l'objet d'études à l'heure actuelle, car le problème n'est pas d'une importance primordiale. Pour l'interprétation de la plupart des expériences, il est d'abord nécessaire de connaître le spectre dans la zone d'irradiation, puis d'effectuer la mesure relative des flux dans les différentes gammes énergétiques utiles, et enfin de réaliser l'intégration des doses reçues au cours de l'irradiation en opérant sur l'une des gammes; pour cette dernière opération on choisissait surtout jusqu'à présent les neutrons thermiques, mais il est maintenant également possible, ainsi que nous le verrons au chapitre suivant, d'opérer dans la gamme des neutrons rapides. Toutefois, lorsqu'il est nécessaire d'effectuer l'intégration dans le domaine des neutrons intermédiaires, l'utilisation du cobalt sous bore ou sous cadmium est toujours possible.

IV. NEUTRONS RAPIDES

Comme pour les autres gammes énergétiques, la mesure des flux de neutrons rapides est relativement aisée dès que l'on connaît la forme du

spectre. Il suffit alors, en principe, d'un seul détecteur dont la section efficace différentielle est bien connue. La première préoccupation sera donc, ici encore, de déterminer le spectre réel dans la gamme énergétique considérée.

Les techniques possibles pour la mesure d'un spectre rapide sont assez diverses:

- émulsions nucléaires;
- dispositifs à semi-conducteurs (jonctions au lithium);
- verres scintillateurs au lithium;
- détecteurs à seuil (activation ou fission).

Il faudrait également citer les compteurs à protons de recul et les spectromètres à ^3He , mais l'expérience acquise en France pour la mise en œuvre de ces appareillages dans les piles est pratiquement négligeable à l'heure actuelle.

L'utilisation des émulsions nucléaires est connue depuis longtemps; la méthode qui consiste à mesurer les traces laissées par les protons éjectés sous l'impact des neutrons rapides est longue et fastidieuse. La tendance actuelle est d'automatiser au maximum le dépouillement des émulsions; de nombreux travaux sont en cours dans ce sens. Comme nous l'avons déjà vu au paragraphe précédent, l'emploi des émulsions n'est pas limité à la partie du spectre que nous avons dénommée «rapide», mais il s'étend aussi à une partie importante du spectre intermédiaire.

Dans les dispositifs à semi-conducteurs et les scintillateurs utilisant Li, le principe consiste à effectuer la spectrométrie des particules émises lors de la réaction $^6\text{Li}(n,\alpha)^3\text{H}$ afin d'en déduire le spectre d'énergie des neutrons incidents. Les diodes au Li ont déjà fait l'objet de nombreuses études de mise au point; il semble que leur emploi ne tardera pas à passer dans le domaine de la pratique courante. La gamme couverte s'étend d'environ 200 ou 300 keV à 7 ou 8 MeV, avec une résolution de l'ordre de 70 keV. En ce qui concerne les scintillateurs au Li, les études n'en sont encore qu'à leur début; il est donc trop tôt pour se prononcer sur les possibilités réelles de cette méthode qui, si l'on veut l'employer pour les mesures à l'intérieur des piles, nécessite par ailleurs la mise au point de conduits de lumière dont la technique est délicate.

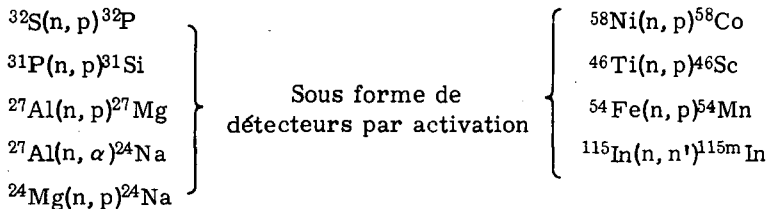
Les détecteurs à seuil peuvent également être utilisés pour la détermination expérimentale des spectres de neutrons rapides [7, 8]. La méthode de dépouillement est ici très différente de celles qui viennent d'être mentionnées; il est nécessaire de mettre en œuvre tout un ensemble de détecteurs à seuil, chacun d'eux fournissant une information «intégrale» qui se rapporte à une zone relativement large du spectre, et les zones couvertes par les différents détecteurs se recouvrant partiellement. Il y a donc là une certaine ambiguïté, et le problème est de savoir comment coupler les diverses informations recueillies de façon à retrouver la forme du spectre. Les méthodes possibles sont nombreuses, mais elles ne donnent pas toutes la même précision; en particulier, le nombre de détecteurs disponibles a une influence directe sur la validité de telle ou telle méthode. Parmi les procédés qui ont été jusqu'à présent proposés, on peut citer surtout:

- l'approximation du spectre par une fonction en escalier;
- l'approximation du spectre par une fonction polygonale linéaire;

- l'approximation par une fonction polygonale exponentielle (Dierckx);
- l'approximation par un polynôme simple;
- la méthode polynomiale de P. M. Uthe (spectre de fission multiplié par un polynôme);
- la méthode «semi-empirique» de O. W. Dietrich;
- la méthode des «indices de spectres» de J. Grundl et A. Usner, dans laquelle le spectre est approximé par une fonction $\varphi(E) = KE^{\frac{1}{2}} \exp(-\beta E)$;
- la méthode du changement de variable, de P. M. Uthe;
- la méthode du développement en séries de fonctions orthonormales, proposée indépendamment par S. R. Hartmann et J. B. Irice.

Au cours des dernières années, de nombreux laboratoires se sont efforcés d'accumuler les résultats expérimentaux dans le but de préciser les domaines de validité de ces différentes méthodes. Les résultats obtenus, encore très incomplets, semblent montrer l'intérêt des méthodes polygonales et polynomiales. Il semble également que l'approximation du spectre par une expression de la forme $\varphi(E) = KE^{\frac{1}{2}} \exp(-\beta E)$, dans laquelle le facteur β peut être ajusté pour rendre compte des déformations du spectre, constitue une méthode élégante et assez souvent valable; les possibilités de cette méthode sont encore accrues si l'on adopte l'expression $\varphi(E) = KE^{\nu} \exp(-\beta E)$ dans laquelle les deux paramètres ν et β peuvent être ajustés pour trouver une forme analytique correcte du spectre réel.

La difficulté majeure rencontrée dans ces études expérimentales réside, en général, dans le fait que les détecteurs disponibles sont en trop petit nombre. Les réactions les plus couramment utilisées sont les suivantes:



ainsi que les réactions de fission sur ^{238}U , ^{232}Th , ^{237}Np , ^{241}Am . Ces derniers détecteurs peuvent être utilisés, soit sous forme de chambres à fission, soit sous forme de dépôts; dans ce dernier cas, la mesure du taux de fission est effectuée par comptage spectrométrique du rayonnement émis par l'un des produits de fission, généralement Cs.

Parmi les détecteurs cités, il faut noter que certains font double emploi, à cause de leurs seuils trop voisins; ils ne fournissent donc qu'une duplication de la même information. C'est le cas de S et de P, d'une part, de Al et de Mg, d'autre part. Ceci est un argument supplémentaire pour qu'un effort soit fait en vue d'accroître le nombre de détecteurs à seuil utilisables.

Par suite d'une recommandation du Groupe de dosimétrie de l'EURATOM, les études expérimentales actuellement en cours utilisent les valeurs de sections efficaces effectives et de seuils effectifs déterminés par la méthode de GRUNDL et USNER [9]. On sait en effet que ces deux grandeurs varient en fonction de la forme du spectre; la méthode préconisée, qui permet de minimiser ces variations, est donc en principe préférable à celle qui fut naguère proposée par Hughes.

En ce qui concerne l'intégration des doses de neutrons rapides, les possibilités offertes par la technique d'activation ont surtout trait aux réactions suivantes [1, 10]: $^{58}\text{Ni}(n, p)^{58}\text{Co}$, $^{63}\text{Cu}(n, \alpha)^{60}\text{Co}$, $^{46}\text{Ti}(n, p)^{46}\text{Sc}$, $^{54}\text{Fe}(n, p)^{54}\text{Mn}$. Les études les plus avancées concernent Ni et Cu, qui sont maintenant couramment utilisés, mais il est encore nécessaire d'effectuer des essais dans la gamme des hautes doses (supérieures à 10^{20} n/cm²) avant de se prononcer définitivement sur la validité de ces méthodes. En effet, l'un des problèmes majeurs pour l'intégration des doses est celui des impuretés contenues dans le détecteur, celles-ci pouvant entraîner après une longue irradiation des activités parasites qui masquent complètement la réaction que l'on veut utiliser. Pour Cu, par exemple, le produit Johnson-Matthey déjà très pur cependant, contient encore trop d'Ag; il a fallu effectuer de nombreux essais avant de sélectionner pour cette application le cuivre américain «Asarco» qui a donné de bons résultats.

Les réactions d'activation proposées pour l'intégration en neutrons rapides présentent aussi, pour la plupart d'entre elles, l'inconvénient d'une section efficace mal connue. Des demandes de nouvelles mesures ont été déposées auprès de l'EANDC, par l'intermédiaire de l'EURATOM, et l'on peut espérer que cette situation s'améliorera petit à petit au cours des prochaines années. Pour le moment les études effectuées par les physiciens des piles ont surtout pour but, en premier lieu, de sélectionner des produits suffisamment purs, et en second lieu d'établir l'intercalibration relative des sections efficaces.

Toujours pour l'intégration des doses de neutrons rapides, d'autres recherches sont également effectuées selon des principes différents de l'activation. Des essais sont en cours, par exemple, sur les variations de densité ou de fréquence de résonance des quartz, en fonction de la dose de neutrons rapides reçue. Il faut également citer les études concernant les variations de résistivité de W qui ont déjà donné lieu à des travaux approfondis mais dont l'interprétation n'apparaît pas très facile [11]. Enfin, l'utilisation de Th avec dosage du Cs formé semble très prometteuse; il se pourrait que l'on dispose là, dans un proche avenir, d'une méthode d'application assez simple [12].

Nous citerons enfin l'emploi de boucles à gaz forane utilisant la réaction $^{19}\text{F}(n, \alpha)^{16}\text{N}$ [2] qui permet simultanément la mesure continue des flux et leur intégration dans le temps.

V. UTILISATION DES RÉSULTATS DE MESURES EN NEUTRONS INTERMÉDIAIRES ET RAPIDES POUR L'INTERPRÉTATION DES EXPÉRIENCES DE DOMMAGES [13, 14, 15]

Nous avons vu dans les deux chapitres précédents que les techniques expérimentales pour la détermination des spectres intermédiaire et rapide sont loin d'être parfaitement au point. Les informations recherchées étant d'une grande importance pour l'interprétation des expériences de dommages, il est apparu nécessaire d'étudier les moyens qui permettraient d'utiliser le mieux possible les données fragmentaires fournies par les méthodes expérimentales dans leur état actuel. Pour cela, les considérations théoriques

doivent permettre d'établir une formulation à paramètres destinée à lever, dans certaines gammes énergétiques, l'indétermination introduite par l'insuffisance des résultats expérimentaux. Nous résumerons ici brièvement l'état actuel de cette question, en nous basant sur les travaux qui paraissent les plus complets.

L'étude systématique des spectres de neutrons intermédiaires et rapides dans les piles montre la possibilité de représenter ces spectres par une expression générale de la forme

$$\varphi(E) = K_0[\varphi_0(E) + h\varphi_e(E)].$$

La composante $\varphi_0(E)$, dite «composante homogène» du flux, peut s'écrire

$$\varphi_0(E) = \Gamma(V - E) \frac{\exp(b\sqrt{E})}{E} + \Gamma(E - V)FE''\exp(-\beta E).$$

La fonction $\Gamma(V - E)$ est égale à 1 pour $E < V$ et à 0 pour $E > V$. Les paramètres V et F sont déterminés de manière que l'on obtienne la continuité des flux et de leurs dérivées pour l'énergie V . Le terme b caractérise la nature du modérateur de la pile. La composante $\varphi_e(E)$, dite «composante hétérogène» du flux, peut s'écrire sous la forme $\varphi_e(E) = E''\exp(-\beta E)$.

Selon la nature du modérateur, les composantes $\varphi_e(E)$ et $\varphi_0(E)$ peuvent être représentées par des formes quelque peu différentes, parfois plus simples, mais toujours voisines de celles qui ont été données ci-dessus. Par exemple, dans la pile au graphite, la composante $\varphi_e(E)$ se réduit à $\varphi_e(E) = \exp(-0,775 E)$.

Au-delà de 2 ou 3 MeV, l'expression de $\varphi(E)$ peut parfois être également représentée plus correctement par une forme analytique différente de celle qui a été adoptée dans la formulation générale donnée ci-dessus. C'est ainsi que l'on peut être amené à prendre pour $\varphi_e(E)$, aux hautes énergies, des formes telles que $E^{0,5}\exp(-\beta E)$ pour les piles à eau lourde, ou $E^{0,5}\exp(-0,775 E) + dE^5\exp(-\beta E)$ pour l'eau légère; dans ce dernier cas, les paramètres β et d sont liés une fois pour toutes par un résultat de calcul.

La formulation générale adoptée, qui peut s'appliquer à l'analyse de tous les phénomènes relatifs à la cession d'énergie par chocs neutroniques, n'est valable que pour des énergies supérieures à 3 keV environ, mais cette limite peut dans certains cas être abaissée à quelques eV (eau légère, par exemple).

L'expression générale de $\varphi(E)$ montre que les composantes $\varphi_0(E)$ et $\varphi_e(E)$ sont déterminées a priori d'après la nature de la pile, c'est-à-dire en fait d'après la nature de son modérateur. Les réponses de deux détecteurs suffisent en principe pour déterminer les coefficients K et h :

- un détecteur sensible vers 3 keV [12] détermine K puisque la composante $\varphi_e(E)$ est négligeable à cette énergie;
- un détecteur à seuil, S par exemple (seuil à 2,7 MeV), permet de déterminer h .

Si besoin est, un troisième détecteur à seuil plus élevé, (Al par exemple, dont le seuil est à 7,5 MeV), permettra de préciser la formulation aux hautes énergies.

En définitive, la méthode décrite ici revient pratiquement à fixer la forme générale du spectre par des considérations théoriques, et à préciser ensuite les valeurs absolues en deux ou trois points du spectre à partir des résultats de mesures fournis par des détecteurs dont l'utilisation est bien connue. Nous avons donc là un moyen de pallier les insuffisances actuelles des techniques de mesures.

VI. RAYONNEMENTS GAMMA [1]

La mesure des rayonnements gamma peut être abordée par des techniques très diverses parmi lesquelles il faut citer surtout les chambres d'ionisation, la dosimétrie chimique, les verres doseurs, les matières plastiques (variations colorimétriques), les calorimètres (isothermes ou adiabatiques), les semi-conducteurs.

Toutes ces méthodes n'en sont pas au même point de développement, et l'intérêt de chacune d'elles dépend dans une large mesure de l'application que l'on veut en faire. Nous récapitulerons donc, sommairement, les possibilités de ces diverses techniques.

Les chambres d'ionisation sont utilisées depuis très longtemps, et peuvent être considérées comme parfaitement au point. Dans leur version classique, ces chambres ont généralement une grande sensibilité; cela exclut leur emploi dans les flux gamma très intenses qui règnent dans les piles. Etant donné l'intérêt des mesures par chambre d'ionisation à air — elles sont d'une mise en œuvre simple et donnent une grandeur directement liée à la définition même du roentgen — on a réalisé des chambres de très petites dimensions, sous basse pression, de manière à réduire leur sensibilité. Avec de telles chambres il est possible de mesurer des flux gamma atteignant $5 \cdot 10^7$ ou 10^8 r/h.

La dosimétrie chimique, principalement par la transformation sulfate ferreux-sulfate ferrique, a été naguère en vogue. Bien qu'elle ne soit pas complètement abandonnée aujourd'hui, cette méthode est quelque peu délaissée pour les raisons suivantes: d'une part, sa mise en œuvre n'est pas très facile (préparation et dosage des solutions, analyse spectrophotométrique nécessitant un appareillage coûteux), d'autre part, chaque fois que le rayonnement au point de mesure est complexe, et ce cas est fréquent, les neutrons rapides peuvent perturber le phénomène dû aux rayonnements gamma. Il est possible, en principe, de séparer les deux effets en utilisant un mélange de sulfate ferreux et de sulfate cérique, mais l'interprétation des résultats devient alors plus difficile.

Les verres doseurs et les matières plastiques peuvent être traités sous la même rubrique puisqu'il s'agit, dans les deux cas, d'étudier avant et après irradiation les variations colorimétriques subies par les détecteurs, ou plus précisément les variations d'opacité sur certaines longueurs d'ondes bien déterminées. Les premiers essais effectués en France, il y a quatre ans environ, avaient donné de grands espoirs par suite de la simplicité de

la méthode et du coût peu élevé des détecteurs. Malheureusement le phénomène d'autoguérison en fonction de la température est mal connu et complique considérablement l'interprétation des mesures. Il y a là une difficulté réelle chaque fois que l'on désire effectuer une mesure absolue ou même simplement une comparaison entre des mesures faites à des températures différentes. Si ce problème pouvait être résolu de manière satisfaisante, le grand intérêt des matières plastiques serait de permettre la mesure des flux gamma dans la gamme des intensités comprises approximativement entre 10^6 et 10^{10} r/h; les verres, eux, permettraient de couvrir la gamme de 10^4 à $5 \cdot 10^6$ r/h.

L'application des techniques calorimétriques aux mesures dans les piles a été très étudiée au cours des dernières années, et l'on peut considérer qu'il existe à l'heure actuelle plusieurs types de calorimètres (isothermes [16, 17, 18], adiabatiques [19]) parfaitement adaptés à ce genre de mesures. Le principe de leur utilisation pour la dosimétrie gamma correspond à une mesure indirecte de ce rayonnement par l'intermédiaire de l'échauffement produit dans certains matériaux. Les phénomènes qui provoquent l'échauffement sont multiples et les rayonnements gamma ne sont pas seuls à intervenir lorsque les matériaux sont placés dans le rayonnement complexe des piles. Il est toutefois assez facile, par un choix judicieux du matériau constituant le noyau du calorimètre, d'obtenir un échauffement dû presque uniquement aux rayonnements gamma; il faut pour cela utiliser une substance de densité élevée et de faible section efficace de capture en neutrons thermiques. Les différents types de calorimètres réalisés jusqu'à présent permettent en pratique de couvrir la gamme des flux gamma comprise entre $5 \cdot 10^5$ et 10^9 r/h. Ce sont des appareils robustes et de mise en œuvre relativement simple; les calorimètres isothermes présentent en outre l'avantage, après un étalonnage préalable, de donner le résultat de mesure en lecture directe et instantanée, et permettent ainsi de faire des mesures continues dans le voisinage des dispositifs expérimentaux en pile.

Les techniques utilisant les semi-conducteurs sont d'une mise en œuvre simple puisqu'il suffit, en principe, de mesurer l'évolution du courant de court-circuit d'une photodiode en fonction des doses gamma reçues. Elles présentent toutefois un inconvénient majeur par le fait que la température d'irradiation doit être limitée à une valeur assez basse; pour les diodes au silicium, par exemple, la température ne doit guère excéder 60°C . Sous réserve d'observer cette précaution, de telles photodiodes peuvent être utilisées pour mesurer des flux gamma compris entre 10^5 et 10^7 r/h.

VII. CONCLUSIONS

Les conclusions qui s'imposent à l'issue de ce tour d'horizon sur l'état actuel de la dosimétrie peuvent être résumées de la manière suivante:

Dans le cas des neutrons thermiques, presque tous les problèmes importants ont été abordés, et des solutions satisfaisantes ont été généralement trouvées. Il est cependant nécessaire que la précision sur certaines sections efficaces soit encore améliorée. Parmi les difficultés qui subsistent, dans ce domaine d'énergie, il faut citer surtout la mesure de la

température d'équilibre du spectre de Maxwell; ce problème, lié à la détermination précise de la jonction entre spectres thermique et intermédiaire, présente une grande importance pour les corps qui possèdent de fortes résonances aux basses énergies, c'est-à-dire en particulier pour les combustibles. Dans un autre ordre d'idées, les études concernant les méthodes par «disparition de noyaux», pour l'intégration des doses de neutrons thermiques doivent être poursuivies afin de préciser leurs possibilités réelles.

Dans le domaine des neutrons intermédiaires, les efforts devraient surtout consister maintenant à préciser les principes de mesure valables dans la gamme comprise entre quelques centaines d'eV et quelques centaines de keV. Pour cela, les études expérimentales entreprises devraient être poursuivies et approfondies. Une meilleure connaissance des intégrales de résonance apparaît également indispensable.

Pour les neutrons rapides, les principes de mesure sont maintenant bien établis. Il reste à faire un important travail expérimental avant de choisir, parmi toutes les méthodes possibles, celles qui présentent le plus d'intérêt. Les sections efficaces, dans ce domaine, sont en général beaucoup trop mal connues; ainsi qu'il a été précisé dans la requête faite auprès de l'EANDC par le Groupe de dosimétrie de l'EURATOM, il serait très utile que quelques équipes de physiciens nucléaires acceptent d'entreprendre une campagne de mesures précises des sections efficaces pour un certain nombre de réactions (n, n') , (n, p) ou (n, α) particulièrement intéressantes en dosimétrie.

Dans les domaines intermédiaire et rapide, il apparaît indispensable de mener de front les études de dosimétrie proprement dites et les études de dommages causés par les rayonnements dans les matériaux. Il se peut en effet que, dans certains cas particuliers tout au moins, la forme du spectre n'ait pas d'influence sensible sur les efforts observés; mettre en évidence de manière certaine un tel phénomène entraînerait évidemment une grande simplification des problèmes de dosimétrie.

Nous avons dit souvent, au cours des pages précédentes, que la diversité des détecteurs disponibles n'est pas assez grande, surtout pour les mesures mettant en jeu des neutrons intermédiaires et rapides. Les efforts consacrés à la technologie des détecteurs (élaboration de matériaux très purs, préparation des détecteurs de types divers) doivent donc être poursuivis, et même accrus. Il est également nécessaire d'améliorer les méthodes de spectrométrie quantitative sur détecteurs irradiés.

En ce qui concerne les rayonnements gamma, les méthodes valables sont d'ores et déjà assez nombreuses et diverses, à la fois pour la mesure des flux et pour la mesure des doses; il reste surtout à apporter quelques améliorations aux appareillages réalisés et à préciser quand cela est nécessaire les phénomènes annexes qui risquent de perturber les mesures (autoguérison, par exemple). Il serait sans doute nécessaire de développer également les techniques de spectrométrie en pile, ce problème n'ayant encore été que très peu étudié.

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STATUS REPORT ON IN-PILE DOSIMETRY IN THE FEDERAL REPUBLIC OF GERMANY

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In the past routine detection methods for flux, fluence and dose measurements were developed in every reactor centre. Except for some unimportant technical details most of them are identical in all stations. Besides these

routine procedures special detection methods were developed and utilized, often at considerable expense for equipment, to meet specific experimental requirements. In the following a brief survey is given of the methods of in-pile dosimetry employed in the reactor stations of the Federal Republic of Germany.

1. DETERMINATION OF THERMAL AND INTERMEDIATE NEUTRON FLUX

1.1. *Activation detectors for flux measurements*

Routine measurements of thermal and epithermal neutron flux are carried out by means of activation detectors. The detector materials used are Au and In (in some cases also Mn) in the low-power range (kW), and Co in the high-power range (MW).

Generally the activation caused by thermal neutrons is separated from the activation of the bare irradiated detectors by a Cd-difference measurement [1, 2, 3, 4]. The thickness of the Cd sheath may differ. In most cases it is 0.5 mm; in rarer instances Cd containers of 1-mm thickness are employed. The activity measurements are interpreted by taking into account self-shielding in the detector and flux depression in the neutron field [1, 4, 5, 6, 7].

The thermal neutron flux quoted is the product of neutron density n and the most probable neutron velocity $v_0 = 2.200$ m/s. The intermediate neutron flux (also called resonance flux) is defined as the flux per lethargy interval.

In addition to the Cd-difference method the sandwich method with two foils is routinely used for measuring thermal and intermediate neutron fluxes (Au + Cu at low power and Co + Ag at full power and long-time irradiation) [8, 9]. Compared with the Cd-difference method the latter procedure is advantageous especially when a major set of foils is to be irradiated simultaneously in a reactor for recording flux profiles.

Flux profiles may be determined in a reactor by irradiation and subsequent activity measurement of small, circular disc-like detectors as well as of long, tape or wire-like probes. This latter shape of detectors has certain advantages. It may serve for determining flux profiles in the area of large flux gradients, e. g. near strong neutron absorbers [10].

For measurements of thermal neutron flux in the FR2 reactor, electrolytic copper tapes 10 mm wide and 50 μ m thick were used. They permit the measurement of fluxes between 10^7 and 10^{12} n/cm² s. Due to its decay scheme, the half-life of 12.8 h for the Cu⁶⁴ isotope and the main resonance at higher energies (600 eV), copper is particularly suited for thermal flux measurements. In the reactor FR2 the epithermal activation of Cu⁶³ can be neglected in these measurements, as the product of the Westcott factors r and s is below 0.03. The tape probes were evaluated in a converted commercial radiochromatograph. The tapes were passed discontinuously in equal steps along a lead collimator. The activity of the screened-out portion of the tape was measured with a methane flow counter.

1.2. Activation detectors for spectrum measurements of intermediate neutrons

The intermediate neutron spectrum of a reactor can be determined in principle by the same resonance detectors which are usually employed for measuring the epithermal neutron flux. Spectrum measurements have been carried out in the FR2 with In, Au, W, La, and Mn as foil materials [11]. When measuring spectra by means of resonance detectors, it is necessary to be careful not to attribute probe activation to one specific neutron energy because of the secondary resonances of the detector materials. To be able to eliminate the major part of secondary resonance as a contribution to the total activation, sandwiches of three foils of the same material were employed. Principally this ensures that neutrons of the main resonance are mainly absorbed in the two external foils, whereas the interior foil is activated almost as highly as the external ones by the $1/v$ portion and the secondary resonances. The difference between external and internal probe activities then is a measure of neutron flux at the energy of the main resonance.

This method was used to determine the epithermal neutron spectrum in the centre of a lattice cell in the reactor FR2. The measurements showed that the decrease of epithermal neutron flux with energy is less than $1/E$.

1.3. Beta current detector

According to a proposal by HILBORN [12] β -current Rh-detectors were employed in the reactor FR2 as monitors for thermal neutron flux and for the measurement of flux profile.

In the first design the detector had the dimensions of about $20 \times 10 \times 2$ mm and was diminished in size in a later design. Between two zirconium plates (0.5 mm), a plate of Rh (0.2 mm), was placed.

The Rh plate was insulated from the zirconium plates by a layer of Scotch Tape. The zirconium and rhodium plates were connected to a Keithley electrometer by an amphenol cable [13]. The detector was calibrated by simultaneous irradiation of gold detectors. The linear relationship between current and neutron flux was checked over a wide range ($1:10^5$) by increasing the reactor power in steps. The time response of the detector was investigated. Any sudden increase in flux was indicated immediately (delay < 1 s). Immediately after the flux change the linearity between flux and β -current is lost temporarily due to the finite half-life of Rh^{104} and Rh^{104m} .

1.4. Thermopile

The thermopile was employed as another detecting instrument for determining flux profiles in a reactor [14]. With it the rise in temperature of a small sample of fissile material (in this case about 1 mg U^{235}) during irradiation is measured by a thermocouple. In the equilibrium state the temperature increase is proportional to the thermal flux (in preliminary experiments the gamma sensitivity of the thermopile was proved to be negligible). The

time response of the thermopile was < 5 s, so that even rapid flux changes could be detected.

2. DETERMINATION OF FAST NEUTRON FLUX

The routine measurements of the fast neutron flux in reactors are performed exclusively by threshold detectors; Al, P, S, Mg and Ni are the most frequently used detector materials. As is well known, the interpretation of the activity measurements offers certain difficulties. On the one hand, these difficulties are caused by the dependence of the spectrum of fast neutrons on the reactor type and on the irradiation position, and on the other hand, by the energy dependence of threshold detectors' cross-sections. The most convenient, though very dubious, evaluation procedure consists of calculating the "equivalent integral fission flux", which would induce the same activity in the detector as the real flux [1, 3, 13].

More precise flux values may be evaluated if the neutron spectrum is known by calculations. The integral flux of fast neutrons may be calculated from the activity measurements by means of cross-sections averaged over the calculated spectrum. The assumed spectrum is verified by several threshold detectors of different materials [15, 16].

3. SPECIAL DETECTION METHODS FOR NEUTRON FLUX AND NEUTRON FLUENCE

In addition to the activation detectors, catcher foils have been tested for the determination of fast neutron flux [17]. Foils of fissile materials (e.g. U^{238} and Th^{232}) are surrounded by so-called catcher-foils (e.g. Al or polyethylene). The fast neutron flux is determined by the activity of fission fragments, which precipitate on the catcher foils. Furthermore, neutron dose measurements on the basis of density changes of quartz during reactor irradiation are being prepared (Research Reactor Munich).

4. DOSIMETRY OF REACTOR IRRADIATION WITH CALORIMETERS

The absorption of radiation energy in materials which are inserted in a reactor generally causes a material heating. Also, changes of the physical and chemical properties of the irradiated materials may occur. They depend on the radiation quantity and quality. In addition to the total absorbed dose it is desirable also to know the contributions of the different types of reactor radiation to the dose.

Different calorimeters have been developed for the determination of material heating and the neutron and γ dose-rate respectively. The material heating in C, Al, Bi and Fe have been measured as a function of irradiation position in the reactor FR2 [21]. The contribution of photons to the total heating in Al, C and Fe has been estimated on the basis of heating measurements in Bi and the calculated photon spectrum.

Similar measurements have been performed in the FRM reactor with an isothermal calorimeter, with Al as calorimeter material [18]. The contributions of fast and thermal neutrons to the total dose-rate could be neglected. An estimate showed them to be less than 1.5% of the total dose-rate.

For the analysis of irradiation effects on predominantly organic substances an isothermal and an adiabatic calorimeter have been developed. The isothermal calorimeter has been designed in such a way that the calorimeter vessel (made of quartz) was nearly similar to the irradiation vessel used for the real radiochemical analysis; with this calorimeter it was also possible to change conditions for heat transfer by variation of the pressure [19]. Thus it became possible to control the equilibrium temperature of the calorimeter in a large range, the reactor power being constant. Diphenyl served as calorimeter material. The dose contribution caused by fast neutrons has been estimated by measuring the fast neutron flux with nickel foils. With the aid of an adiabatic calorimeter in the FRG reactor, measurements of the gamma and neutron dose-rate have been performed [20]. The calorimeter contained samples of Al, polystyrene and polyethylene. Aluminium had been vaporized upon the organic samples. The three samples, insulated by vacuum, were surrounded by an adiabatic jacket which could be heated.

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A STATUS REPORT ON IN-PILE DOSIMETRY IN HUNGARY

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INTRODUCTION

In-pile dosimetry at the Central Research Institute of Physics is concerned with the following problems:

- (a) Thermal, epithermal and fast neutron flux measurements by the activation method;
- (b) Calorimetric and chemical measurements of energy absorbed from reactor radiation.

The flux measurement technique applied in the WWR-S research reactor [1] which has been in operation for five years, is determined primarily by aspects of isotope production. Microflux distribution and neutron temperature measurements are carried on within the scope of the reactor physical experiments [2] performed on the ZR-2 critical assembly built at the Institute.

In-pile irradiation experiments on organic liquids required the development of methods for measuring the energy absorbed.

NEUTRON FLUX MEASUREMENTS BY ACTIVATION METHODS

Thermal and epithermal flux measurements.

In the WWR-S reactor the flux distribution of thermal and epithermal neutrons is routinely measured in the core as well as in the irradiation channels adjacent to the core [3]. In the standard procedure 0.7-mm diam. aluminium wire containing 10% dysprosium or 0.5-mm diam. copper wire is used. The activity distribution over the wire is measured by a scintillation detector coupled to a semi-automatic counter assembly. The thermal flux within the core is usually measured by a dysprosium wire, while in the irradiation channels, where the epithermal neutron contribution to the flux is relatively small and nearly constant, a copper wire is used.

The absolute values of the thermal neutron flux are determined generally by measuring the absolute activity of gold foils, using the beta-gamma coincidence technique [4].

In the ZR-2 critical assembly, the power of which is limited to a few watts, dysprosium is preferred because of its high activation cross-section and relatively short half-life. Dysprosium-aluminium wire was used, for instance, to determine the neutron density distribution in neutron traps of various diameters for cores of different lattice pitches [2].

To characterize the relative magnitude of the epithermal contribution to the spectrum the index r is used, which is determined by measuring the cadmium ratio for gold foils in the WWR-S reactor and for manganese foils in the ZR-2 critical assembly.

FAST NEUTRON FLUX MEASUREMENTS

The absolute and relative values of the fast neutron flux are determined by means of sulphur threshold detectors. The measured data are evaluated by making use of the value $G_{\text{eff}} = 114$ mb measured in the Rossendorf reactor of a similar type [10].

MICRO-FLUX AND NEUTRON TEMPERATURE MEASUREMENTS

In the ZR-2 critical assembly the flux distribution over the unit cell is measured, using manganese, dysprosium, lutetium and europium foils. The foils are prepared from a mixture of the oxide of the element in question and polyethylene powder by hot pressing to thicknesses at which the self-screening of the foils becomes negligible. The foil thicknesses (in mg/cm^2) are: 2, 2, 10 and 0.4 for lutetium, dysprosium, manganese and europium, respectively. After irradiation the foils are cut into suitable pieces and the activities of the individual pieces are measured by GM tube or by a 2π methane gas-flow counter. Both the flux distribution and the spectrum variation in fuel elements and moderator are determined from these measurements [5].

The method applied to the determination of the neutron temperature in the ZR-2 critical assembly is based on the measurement of reactivity variations induced by dilute solutions of $\text{Cd}(\text{NO}_3)_2$; H_2BO_3 ; $\text{Dy}(\text{NO}_3)_3$; AgNO_3 [6]. This technique has the great advantage of yielding absolute values for the neutron temperature. The drawback of the method is that the measured value of the neutron temperature is an average over the solution volume and the spectrum gets harder in the vicinity of the solutions. These disadvantageous effects call for further investigations. No such problems arise in measuring the neutron temperature by means of foils, but the foils should be calibrated using a neutron flux with known spectrum [7].

MEASUREMENT OF ABSORBED RADIATION ENERGY

For the determination of absorbed energy in organic liquids a calorimetric technique was chosen as the basic method, since it permits the direct determination of the radiation energy absorbed in the sample [8]. Because of the small G-values of the materials involved in our experiments the heat energy determined by the calorimeter may be considered as the total energy absorbed.

The calorimeter used is a 20-mm diam., very thin-walled (0.2 mm) quartz flask, which is filled with the organic liquid (a mixture of 28% diphenyl and 72% diphenyl methane) investigated in the loop experiment. The flask is surrounded by a double-walled aluminium mantle. The advantage of this calorimeter is that it contains a minimum amount of structural materials, but its disadvantage is that it can be considered adiabatic only for a short term (~ 35 s). The calorimeter can be used satisfactorily in the range from 0.1 to 0.4 W/g.

It has been found that, in order to overcome the uncertainty due to the delayed gammas from the fission products, it is most convenient to perform the calorimetric measurements at relatively high reactor power. This power level in our case is 1 MW, which corresponds to a maximum dose-rate value of 0.4 W/g. By means of this calorimeter the axial distribution of the dose-rate in the loop channel is routinely measured.

Investigations have begun for the determination of the gamma contribution to the dose by using a lead-filled calorimeter of similar design. The measurements show the results to be quite sensitive to the size of the lead spheres.

For monitoring the dose, purified diphenyl samples were irradiated and the G_{gas} values were measured. For this material the G_{gas} value is nearly independent of the irradiation temperature and proportional to the dose up to 10^9 rad. In the near future other materials (also of CH_2 type) will be investigated.

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STATUS REPORT ON IN-PILE RADIATION METROLOGY IN THE NETHERLANDS

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In the Netherlands the following research reactors are in operation:

1. The HFR (High Flux Reactor) at Petten, which is very similar to the ORR reactor at Oak Ridge in the United States and the Swedish R2 reactor. The maximum power is 20 MW. It is owned by Euratom and is operated on its behalf by the Reactor Centrum Nederland (RCN).
2. The LFR (Low Flux Reactor) at Petten, very similar to the Argonaut and Jason reactors. Its maximum power is 10 kW, and it is operated by RCN.
3. The KRITO reactor in Petten, a critical facility to test the possibility of a specially designed ship propulsion reactor. Operated at a few watts by RCN.
4. The HOR (Hoger Onderwijs Reactor) in Delft, a swimming-pool-type reactor with a maximum power of 100 kW. It is operated by the Reactor Institute Delft.
5. The BARN (Biological and Agricultural Reactor, Netherlands) at Wageningen, operated at 100 kW.

The experimental work on in-pile dosimetry is mainly concentrated in the reactor centre at Petten, although the Institute for Nuclear Physics Research at Amsterdam has given valuable contributions to the subject of fast-neutron activation detectors [1, 2, 3]. In the centre of Petten, Euratom and RCN co-operate, for this subject, in a radiation metrology group.

THERMAL NEUTRON MEASUREMENTS

For the measurement of thermal neutron flux densities Dy, Mn, In, Au, Cu and Co are used in several forms, that is, in the form of foils and wires with different dimensions. For absolute measurements Au and Co foils are used widely, because of the ease of absolute activity measurements with the coincidence method. The other materials mentioned are sometimes used for the measurements of relative flux density distributions.

For the determination of the fluence of thermal neutrons only cobalt-foils are applied, because of cobalt's long half-life. For the absolute measurements of cobalt activities in the millicurie range, we rely on a set of Co⁶⁰ sources, calibrated and provided by the Euratom Central Bureau for Nuclear Measurements in Geel, Belgium [4, 5]. Measured thermal-flux densities are expressed as conventional flux densities (i. e. as 2200 m/s flux densities).

FAST NEUTRON MEASUREMENTS

Fast-neutron flux densities are routinely measured using the reactions $\text{Ni}^{58}(\text{n}, \text{p})\text{Co}^{58}$ and $\text{Al}^{27}(\text{n}, \alpha)\text{Na}^{24}$. For the determination of the fast-neutron fluence the following reactions are used $\text{Ni}^{58}(\text{n}, \text{p})\text{Co}^{58}$, $\text{Ti}^{46}(\text{n}, \text{p})\text{Sc}^{46}$, $\text{Fe}^{54}(\text{n}, \text{p})\text{Mn}^{54}$ and $\text{Cu}^{63}(\text{n}, \alpha)\text{Co}^{60}$. The reaction with nickel is considered as a standard reaction, although it has the following disadvantages:

the half-life of 71 d is, in particular for long-term irradiations, too short; and

corrections for the burn-up of the Co^{58} and $\text{Co}^{58\text{m}}$ by thermal neutrons must be applied, and cannot be neglected;

the cross-sections for the important reactions $\text{Ni}^{58}(\text{n}, \text{p})\text{Co}^{58\text{m}}$ $\text{Ni}^{58}(\text{n}, \text{p})\text{Co}^{58}$, $\text{Co}^{58\text{m}}(\text{n}, \gamma)\text{Co}^{59}$ are not very well known.

For short irradiations the reaction with titanium seems a promising alternative [6]. For longer irradiations only the reactions with iron and copper are applied, although not all nuclear data are available with the desired accuracy. Iron might be applied in enriched composition to minimize the Fe^{59} activity produced by thermal neutrons. Full attention is given to other reactions with low effective thresholds, e.g. the reactions $\text{Rh}^{103}(\text{n}, \text{n})\text{Rh}^{103\text{m}}$, $\text{In}^{115}(\text{n}, \text{n})\text{In}^{115\text{m}}$ and $\text{Np}^{237}(\text{n}, \text{f})$.

Experiments for the determination of the fast-neutron spectrum in reactors using threshold activation detectors have been performed [7] and this effort will be increased. From the observed specific saturation activities spectral indices [8] are derived and compared with theoretically calculated spectral indices assuming a distribution in the form $N(E) = CE^{\nu} \exp(-\beta E)$.

While in many cases the neutron spectrum at irradiation positions is not fully known, and while therefore reliable values for effective cross-sections are not always available, we generally use the average cross-sections for a fission neutron spectrum in the calculations, thus obtaining "equivalent fission flux densities" or "equivalent fission fluences".

Details on the calculation methods are given elsewhere [9]. Compilations [10, 11] on energy-dependent cross-sections for threshold reactions are used to determine average cross-sections and effective thresholds. A report to the Euratom Working Group deals with the influence of the inaccuracy in the cross-section curves on the determination of the fast-neutron flux density [12].

INTERMEDIATE NEUTRON MEASUREMENTS

The normal procedure for measuring the intermediate flux densities with the cadmium ratio for gold foils is also practised in the Netherlands. The Westcott convention for representation of flux densities and cross-sections is generally used.

Some time ago experimental work was started to obtain information on the spectrum of intermediate neutrons, using several resonance detectors. An extensive literature review on methods and techniques applied elsewhere has been written [9]. Experiments have been performed using Co, Mn, Au, Cu, As and Ga detectors. The triple foil technique, as mentioned by EHRET

[13] has been applied. Work was started using relatively thick foils, where the self-shielding was no longer negligible. Tables with correction functions [14] have been made following the assumptions made by MANNER and SPRINGER [15].

NUCLEAR HEAT GENERATION MEASUREMENTS

In the HFR reactor the nuclear heating has been measured in several positions applying the following two methods [16]:

determination of the heat current in a sample material (aluminium or stainless steel) by measuring the temperature difference with thermocouples between two appropriately chosen points;

determination of the equilibrium temperature of a sample, the heat transfer from the sample to the surroundings in the instrument being determined previously in laboratory experiments.

The first method has been applied for measurements within the reactor vessel and also in the so-called pool-side facility, while the second method has been used for special measurements in this pool-side facility and in the pneumatic post system.

The measurements within the reactor vessel have been performed using a plug fitting in the normal beryllium reflector elements. This plug consists of a series of cylindrical samples (about 12), separated by filler samples, welded together (see Fig.1). All samples have a central cylindrical hole, through which pass the thermocouple wires from samples below. Each sample contains two thermocouples, one near the central hole, and one near the outside.

The heat transfer occurs in radial direction, as the axial heat transfer is diminished by small gaps between the successive samples. The temperature difference between the two thermocouples is a measure for the nuclear heating. The heat generation was calculated, using the following equation:

$$H = \frac{2\lambda \Delta T}{\rho \left\{ \frac{1}{2}(r_2^2 - r_1^2) - r^2 \ln r_2 / r_1 \right\}}$$

where H = heat generation (in W/g)

λ = heat conductivity (in W/cm °C)

ΔT = temperature difference between the two thermocouples (in °C)

ρ = mass density of the sample material (in g/cm³)

r_2 = radius to the outer thermocouples (in cm)

r_1 = radius to the inner thermocouples (in cm).

r = radius of the central hole in the plug (in cm)

From the results it follows that the position of Cd-fuel connection in the control members has a great influence on the heat generation. The measuring device 10 cm above the core centre line especially shows a great variation between the beginning of an operating cycle and the end of the cycle (at the start about 60% of the heat generation at the end). The variations of the maximum heat generations for various fuel core configurations amount to about 20%.

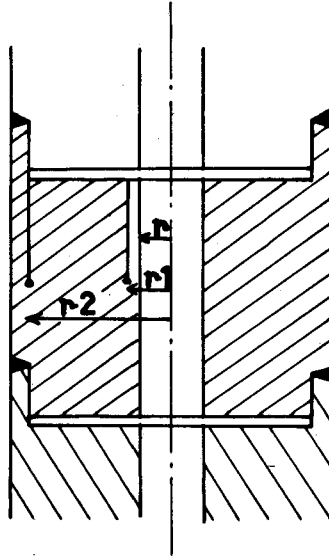


Fig. 1

Detail of measuring plug

Outside the core a device was used, consisting of an aluminium cylindrical pin, mounted on one side on the base of a can. By applying a vacuum in this can the radial heat transfer can be neglected with respect to the axial heat transfer. The heat generated will therefore be transferred via the base plate to the surrounding cooling water. The temperature difference between the two ends of the cylindrical pin is measured with thermocouples. The heat generation in this case is calculated with the formula:

$$H = \frac{2\lambda\Delta T}{c(x_1^2 - x_2^2)}$$

where x_1 and x_2 denote the positions of the thermocouples.

The second method mentioned above is applied with a different device. An aluminium sample is housed in a water-tight jacket of stainless steel. In the middle of the sample and on the inner side of the jacket the temperature is measured with thermocouples. As the jacket is very thin, it is assumed that the measured temperature difference is approximately equal to the temperature difference between sample and outer side of the jacket, while the radiation heating of the jacket can be neglected.

In this case we have the following equation:

$$\Delta T(t) = PR(1 - \exp(-t/RMC_0))$$

where ΔT = temperature difference between sample and jacket (in °C)

P = heat production of sample (in W)

R = heat resistance between sample and jacket (in °C/W)

M = mass of sample (in g)

C_0 = specific heat of sample (in $W/^\circ C$ g)

When equilibrium has been reached

$$\Delta T = PR = MHR.$$

The quantity R is determined in laboratory experiments from cooling curves.

The total accuracy with which the nuclear heating is determined by these methods amounts to about 15%. The main sources of error originate from the thermocouple signal, the position of the thermocouples and the values of the thermal conductivity of the material. For the design of irradiation capsules or loops the accuracy obtained for the materials investigated is quite satisfactory.

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НЕКОТОРЫЕ ВОПРОСЫ ДОЗИМЕТРИИ НА РЕАКТОРАХ*

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Проблему дозиметрии на реакторе, очевидно, можно подразделить на 2 частные задачи:

1. Измерение доз и потоков вокруг реактора, создаваемых излучением, выходящим из защиты и рассеянным полом, потолком, стенами и т. д.
2. Измерение доз и потоков излучения в каналах реактора.

Каждая из этих задач имеет свои особенности и свои пути решения.

Дозиметрические измерения вокруг реактора, как правило, проводятся комплексом приборов, состоящим из детекторов гамма-излучения, быстрых нейтронов всеволновых дозиметров [1]. Примером такого комплекса приборов является Рентгенометр Универсальный Сцинтилляционный — РУС, разработанный И. Б. Кеирим-Маркусом, Л. Н. Успенским и др.

Радиометром быстрых нейтронов является сцинтилляционный счетчик с дисперсным сцинтиллятором $ZnS(Ag)$ в полистироле. Всеволновой дозиметр представляет собой сферу диаметром 30 см, внутри которой помещена еще одна сфера диаметром 15 см. Наружное сферическое кольцо заполнено смесью парафина и карбида бора для исправления "хода с жесткостью". Внутренний парафиновый шар имеет полость для помещения фотоумножителя с тонким боросодержащим сцинтиллятором для регистрации замедлившихся нейтронов. Прибор отличается хорошей изотропностью. Кривая чувствительности прибора выбором размеров и количества борного поглотителя подгонялась под кривую биологической эффективности нейтронов. Ошибка при измерениях этим прибором нейтронного излучения, выходящего из водных, графитовых, бериллиевых, бетонных барьеров, не превышает $10 \div 15\%$ (исключая статистическую ошибку).

При измерении биологических доз после слоев из весьма чистого железа ошибка в измерении может значительно увеличиться за счет большего количества промежуточных нейтронов.

Использование комплекта РУС позволило составить более правильную карту дозного поля возле реактора и выделить группу промежуточных нейтронов, под которой понимался поток нейтронов с энергией от 1,5 Мэв до 1 эв.

Наряду с выше описанным дозиметром используются и другие модификации приборов с предварительным замедлением. Дозиметр, разработанный Андросенко и Смирениным [2], представляет собой цилиндр диаметром 34 см, внутри которого помещается борный счетчик диаметром $3,5 \div 3,8$ см. Изодозный детектор, как он назван авторами, имеет меньшую зависимость чувствительности, выраженной в бэр, от энергии, но худшую изотропность, чем прибор РУС.

Существуют и другие способы и приборы, позволяющие весьма точно определить дозу от нейтронов.

* Copies of this paper may be obtained in English upon application to the Agency (Ref. PL-121/18).

Из сказанного выше видно, что большое внимание уделяется измерению доз как от быстрых, так и промежуточных нейтронов. В целом проблема измерения промежуточных нейтронов считается важной и по сей день.

Если за верхнюю границу интервала промежуточных нейтронов принять 0,5 Мэв, то оценки показывают [3], что после водного барьера доза промежуточных нейтронов составляет 11% от дозы быстрых, после бетона — около 50% и после графита — около 60–70%. Однако, если за верхнюю границу промежуточных нейтронов принять 1 Мэв, то, естественно, вклад промежуточных нейтронов возрастает.

Выделим 3 группы нейтронов: 1 эв — 10 кэв, 10 кэв — 1 Мэв и больше 1 Мэв. В этом случае, после Ве толщиной 30 см доза от I группы — 1%, от II группы — 18%, от III группы — 81%; после графита толщиной 90 см: от I группы — 10%, II — 42%, III — 48% и, наконец, после воды — 2; 27,5 и 70,5% соответственно.

Если же детектор быстрых нейтронов обладает нижним порогом около 1,5 Мэв, то тогда на долю II группы (т.е. от 10 кэв до 1,5 Мэв) приходится 44% после 30 см Ве, 62% после 90 см графита, 50% после воды.

Из этих данных видно, что нельзя не учитывать вклад промежуточных нейтронов и ограничиваться измерением только быстрых нейтронов.

Часто делаются попытки измерения потока промежуточных нейтронов борным счетчиком в кадмиевом фильтре с последующей экстраполяцией данных по равновесному спектру Ферми. Однако такой подход неоправдан, несмотря на привлекающую простоту метода из-за большой ошибки.

Так, для воды ошибка этого метода составляет 100% при верхней границе 0,5 Мэв и 300% при верхней границе 1,5 Мэв [4] (такие измерения занижают дозу).

При измерениях доз и потоков внутри реактора имеется свой круг специфических задач. Требования к величине детектора, наличие высоких потоков радиации не позволяют использовать детекторы с замедлителем. С другой стороны, круг задач, решаемых в этом случае, значительно шире: облучение материалов, биологических объектов и т.д. Поэтому представление данных в бэр'ах вряд ли будет достаточным. Очевидно, что знание спектра нейтронов в различных точках реактора является предпочтительным.

Спектры быстрых нейтронов внутри реактора, на выходе экспериментальных каналов, как правило, определяются пороговыми индикаторами.

Наиболее употребительными являются: $\text{Th}^{232}(nf)$ $E = 1,0$ Мэв, $\text{U}^{238}(nf)$ $E_{\text{пор}} =$, $\text{P}^{32}(np)\text{Si}^{32}$ $E_{\text{пор}} = 3$ Мэв, $\text{S}^{32}(np)\text{P}^{32}$ $E_{\text{пор}} = 2$ Мэв, $\text{Al}^{27}(np)$ Mg^{27} $E_{\text{пор}} = 5,3$ Мэв, $\text{Si}^{28}(np)\text{Al}^{28}$ $E_{\text{пор}} = 6,1$ Мэв, $\text{Fe}^{56}(np)\text{Mn}^{56}$ $E_{\text{пор}} = 7,6$ Мэв, $\text{Al}^{27}(n\alpha)\text{Na}^{24}$ $E_{\text{пор}} = 8,6$ Мэв.

Кроме этих, широко известных детекторов, используются изотопы, у которых в результате неупругого рассеяния образуется длительный метастабильный уровень [5]:

In^{115m} — эфф. порог 1,5 Мэв; $T = 4,5$ часа ($\sigma_{\text{эф}} = 0,25$ б);

Hg^{199m} — эфф. порог 1,9 Мэв; $T = 44$ мин ($\sigma_{\text{эф}} = 0,12$ б).

При использовании индия-115 и ртути-199 гамма-излучение продуктов реакции регистрируется, как правило, большим кристаллом NaJ(Tl) и сцинтилляционным спектрометром со 100-канальным анализатором.

Примером такого использования может служить работа по измерению спектров в экспериментальном канале реактора ВВР-2 [5].

Сразу следует подчеркнуть, что использование пороговых детекторов, несмотря на сравнительную простоту измерения, может привести к значительным ошибкам как из-за ограниченных знаний точных сечений, так из-за несоответствия измеряемого спектра нейтронов "пробному" — обычно спектру деления.

Из-за такого несоответствия ошибка измерения может достигать 100% и более. Для увеличения точности измерений был предложен ряд способов. Один из них — метод последовательных приближений — не нуждается в особом пояснении [6]. Другой метод заключается в подборе "пробного" спектра нейтронов, близкого к исследуемому.

Для правильности контроля вводится параметр ошибки:

$$\epsilon = \frac{\Phi_{0,k} \sigma_{0,k,\text{эф}} \int_0^{\infty} \sigma_k \psi_m dE}{\Phi_{0,i} \sigma_{0,i,\text{эф}} \int_0^{\infty} \sigma_i \psi_m dE}; \quad E_{k,\text{эф,пор}} > E_{i,\text{эф,пор}}$$

Если измеряемый и "пробный" спектры совпадают, то ϵ близко к 1. Если $\epsilon < 1$, то экспериментальный спектр мягче, при $\epsilon > 1$, экспериментальный спектр жестче теоретического "пробного" спектра, взятого для расчета.

Если иметь набор спектров, то можно подобрать такие, что ϵ не сильно отличается от 1 как в ту, так и в другую сторону. В этом случае можно определить интегральный поток нейтронов, как среднее между этими "вилючными" спектрами, а именно:

$$\Phi_k = \frac{\sigma_{0,k,\text{эф}} \Phi_{0,k} \cdot 1 / \sigma_{m,k,\text{эф}} + \sigma_{0,k} \Phi_{0,k} \cdot 1 / \sigma_{m,k,\text{эф}}}{2}$$

Примером комплексного исследования спектральных распределений является также исследование реактора БР-5 [7]. Изучение спектров проводилось в каналах, проходящих через никелиевый отражатель и защиту от поверхности активной зоны.

В активной зоне спектр довольно жесткий и значительная его часть лежит выше 100 кэв. Никелиевый отражатель деформирует спектр нейтронов так, что их средняя энергия лежит в области десятков кэв. Детальные измерения проводились для нейтронов с энергией выше 50 кэв. В остальных случаях использовались реакции $U^{235}(n,f)$, $Th^{232}(n,f)$, $Al^{27}(n,\alpha)$, Na^{24} , $Pu^{239}(n,f)$. Кроме того, спектр нейтронов при малых энергиях исследовался детекторами $Au^{197}(n,\gamma)$, методом резонансной блокировки [8].

Камера деления с Pu^{239} была всеволновым счетчиком для диапазона энергий от 50 кэв до 6 Мэв и использовалась только в тех местах, где спектр лежит в области постоянства сечения.

Интересный способ был предложен для измерения спектров промежуточных нейтронов в каналах реактора [9]. Использовался метод пропускания через "п-гексан" в хорошей геометрии. Детекторами были борные счетчики. Кривая пропускания — изображение — подвергалась обратному преобразованию Лапласа. Полученный оригинал после учета энергетиче-

ческой зависимости чувствительности детектора и учета сечения п-гексана позволил установить спектр нейтронов. Кроме того, в одном из каналов спектр быстрых нейтронов измерялся также по трекам протонов отдачи в фотоэмульсии. Было обнаружено, что только при энергиях больше 3 Мэв спектр совпадает со спектром деления, и хорошо согласуется с расчетным спектром, определенным по многогрупповому методу (18 групп).

В ряде случаев, при облучении биологических объектов используют полиэтиленовые камеры с водородосодержащим наполнителем. Вклад гамма-излучения определяется по графитовой камере, наполненной углекислым газом и вычитается.

При больших мощностях дозы, наряду с химическими дозиметрами, употребляются термолюминисцентные стекла [10]. Они отличаются чрезвычайно широким диапазоном регистрируемых доз от 0,02 до $2 \cdot 10^8$ рад, причем их показания не зависят от мощности дозы вплоть до 10^8 рад/сек.

Здесь, конечно, упомянуты далеко не все методы употребляемые в Советском Союзе, а отражены наиболее распространенные.

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A STATUS REPORT ON IN-PILE DOSIMETRY IN THE UNITED KINGDOM

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1. INTRODUCTION

This brief report will review some problems of in-pile dosimetry currently being examined in the United Kingdom under the two main headings of:

- (i) measurement of the reactor radiations, including fluence, fluence rates and spectrum characterizations;
- (ii) measurement of absorbed dose in materials by reactor radiations.

These divisions are convenient not only because of the widely different techniques necessary for the assessment of the various parameters but also because they underline the basic difference between absorbed dose and exposure. Confusion between these two items has long resulted in misunderstandings in radiological research, but a substantial measure of agreement has recently been reached by the International Commission on Radiological Units and Measurements (ICRU) and it is hoped that their findings may be accepted by reactor physicists.

Almost all the work at present being conducted in the United Kingdom is done at the various establishments of the Atomic Energy Authority (UKAEA); almost no effort is expended on this subject by the Universities, Central Electricity Generating Board or the commercial reactor construction consortia, but this is inevitable since almost all the United Kingdom research reactors are operated by the (UKAEA).

The necessity for standardization of techniques and the use of generally acceptable values for important constants (cross-sections, etc.) is recognized as a matter for urgent consideration, and under each heading there is given some indication of progress which has already been made towards this end.

2. MEASUREMENT OF REACTOR RADIATIONS - EXPOSURE

A most important factor is the need for agreement on the conventions to be adopted in measurements of neutron flux or fluence, and this is considered first.

2.1. Neutron flux conventions for use in reactor irradiation experiments

There is a considerable variation in the neutron energy spectrum in the various types of research reactors in use and from place to place in any one reactor and a single measurement of a "neutron flux" with no reference to how this flux is defined is not adequate for a reactor experiment. One of the most urgent needs in the field of reactor experiments is for a wider appreciation of the importance of the neutron energy spectrum and for more precision and care in the presentation of experimental flux measurements. Unfortunately, the techniques at present available for the measurement of neutron flux and spectrum are not adequate for all the necessary data to be obtained, particularly in the range of energies of interest in radiation damage studies. These techniques are improving all the time however and much of the value of some very expensive irradiation experiments can be lost if the flux measurement results are presented in such a manner that they cannot be re-interpreted as better data becomes available.

Ideally, data should always be presented in such a manner that the method of measurement is obvious and the original measured data can be reconstructed. Also it is desirable that any convention should present the data in a direct and simple manner with as few assumptions and approximations as possible. Unless this is so there is always a danger of misinterpretation or misapplication and the convention will soon become outdated.

2.1.1. Thermal and epithermal flux conventions

The convention in use for the presentation of thermal and epithermal fluxes is that described by Westcott. It has the advantage of being simple and straightforward in application and does not introduce any arbitrary cut-offs in continuous energy spectra. Its main disadvantages are, firstly, that the fluxes obtained are not immediately applicable to fuel irradiation experiments and, secondly, that rather arbitrary joining functions are assumed to join the Maxwellian spectrum to the slowing down spectrum. However, these disadvantages are likely to be inherent in any convention which does not fully specify the energy spectrum over the range of energies being considered.

The introduction of an arbitrary cut-off into any convention should be avoided especially if the use of some heavily absorbing material such as cadmium is implied. Such a cut-off in what is essentially a smoothly varying function can only be a source of inaccuracy and error, and the use of cadmium, which does not produce a sharp cut-off in any case, leads to distortion of the spectrum being measured.

It is likely that the best convention for thermal and epithermal neutrons would be a measure of the total neutron density, with the neutron temperature and an epithermal parameter such as the Westcott 'r' value quoted where necessary. However, for the sake of uniformity the straightforward Westcott convention is used and a neutron temperature is quoted in addition for those cases for which it is required.

2.1.2. High energy neutron flux conventions

In the range of energies above a few tens of keV the neutron flux conventions are less well resolved than in the thermal and epithermal range. Over this energy range the neutron spectrum varies much more than in the thermal region, and so far no satisfactory method of neutron spectrometry has been found. These two points make it extremely important that any conventions used to express flux measurements should be clearly stated and understood, and that all data should be capable of re-analysis as new information on the neutron spectrum becomes available.

The quantity usually measured in fast neutron flux determination is the activity induced in a material due to a threshold reaction. Ideally, the quantity which should be quoted is the reaction rate for the threshold reaction concerned, as this is unambiguous and involves no arbitrary assumptions. There is considerable resistance to results quoted in this form and most people prefer some conventional flux even if this is not realistic.

The main criterion that any convention has to satisfy is that it should be useful, scientifically realistic, and should not imply more than it actually means. Also it is desirable that it should not impose artificial divisions or limitations on the quantity being defined; some of the conventions at present in use for fast neutron flux do not satisfy these conditions.

The use of "flux above 1 MeV" for radiation damage results is one example of a misleading convention. Its use implies that in some way the integral of the flux spectrum above 1 MeV has been determined, but in practice it is usually assumed that the spectrum above 1 MeV is a fission spectrum

and the flux is calculated from a single threshold reaction-rate measurement. This can easily be demonstrated to be wrong by measuring two or more threshold detectors in several different reactors or in several positions in the same reactor. Another shortcoming of this convention, especially in connection with radiation damage work, is the introduction of an arbitrary cut-off with its implication that radiation damage is caused only by neutrons with energies above 1 MeV. The spectrum is continuous in this region, with considerable variations just below 1 MeV; neutrons of this energy have a substantial effect on the radiation damage for most materials. Thus, calculations for steel and graphite show that at least as much damage is done by the part of the spectrum below 1 MeV as is done by the part above this energy.

Other flux conventions which introduce an artificial cut-off in the neutron spectrum are equally undesirable and the use of any such convention must be viewed with suspicion until adequate methods of measuring the neutron spectrum are available.

At the present state of fast neutron flux measurement it is very difficult to define a convention which satisfies all requirements, but if it is required to quote results in the form of a flux the "equivalent fission flux" has advantages. It is defined as the flux of fission neutrons which would produce the same reaction rate in the detector used. The detector and the cross-section used to calculate the flux should also be quoted. This convention is not ideal as it can be taken to imply the presence of a fission spectrum which is certainly not intended. It does however have the advantage that there is no doubt about the spectrum used to calculate the results and the original data can easily be re-constructed from the results. A general agreement to quote reaction rates would be the best solution to fast flux monitoring, but failing this the equivalent fission flux has a lot to recommend it in spite of its limitations.

2.2. Detectors for monitoring neutron flux

2.2.1. Thermal flux

For thermal flux monitoring cobalt wire is most commonly used. The wire is 0.25 mm diam. and monitors are cut with a hand guillotine, producing detectors of weight $1 \text{ mg} \pm \frac{1}{2}\%$. The method suffers from the disadvantage that the neutron self-shielding is appreciable, particularly in the resonance region. However, the fact that the foils are robust, easily handled and easier to reproduce than dilute alloys of cobalt in aluminium saves considerable time in an extensive monitoring programme. The thermal neutron self-shielding factor for the foils is approximately 0.92 and the correction for epithermal activation in the core of DIDO where r is 0.1 is 7%.

Individual foils are compared with a set of cobalt standards in an ion chamber or scintillation counter and, with care, a relative scale for thermal neutrons can be maintained to about $\frac{1}{2}\%$ over very long periods. Several sets of standard foils were prepared and very carefully intercompared at Harwell. These sets are now in use in several laboratories in the United Kingdom and also at Risø in Denmark, with good agreement. An extension of this type of scheme on an international basis would have much to recom-

mend it if agreement could be reached on cross-sections and self-shielding factors.

When it is necessary to obtain continuous information on thermal neutron flux variations recourse is made to miniature fission chambers, and designs are becoming available which are 6 mm diam. and contain mixed coatings of U^{238} and U^{235} to minimize the effect of burn-up on current sensitivity. The use of carefully purified argon/ CH_4 gas mixtures enables pulse counters to be constructed with current rise-times of less than 2×10^{-8} s, so that mean counting rates of at least $2 \times 10^6/s$ may be attained with small counting losses. Pulse counters may usefully supplement mean current chambers when activation effects become serious.

Interest is also growing in the self-powered neutron activation detector, in which the beta activity produced in one electrode is collected at another electrode, the resultant current being measured directly. This type of instrument may be made smaller than 3 mm diam., and can be little more than an extension of a standard mineral-insulated flexible cable. It requires no extensive power supplies and makes much less stringent demands on the electrical insulation properties of connecting cables.

2.2.2. Fast flux

For monitoring fast neutron flux a similar foil technique is used to that described for thermal neutron monitoring, but the choice of technique is more difficult. The most common reaction is $Ni^{58}(n,p)Co^{58}$, but the high thermal cross-section of Co^{58} and Co^{58m} limits its usefulness in high thermal fluxes due to shortening of the effective half-life. In a thermal flux of 10^{14} n/cm² s the effective half-life of these detectors is reduced to about 30 d.

For long-term irradiation experiments in high thermal fluxes the $Fe^{54}(n,p)Mn^{54}$ reaction is proving to be a useful flux monitor, but this introduces difficulties due to the Fe^{59} activity produced with thermal neutrons. Unless a separated isotope can be used the analysis involves a rather tedious chemical separation to remove the Fe^{59} activity, and thus whenever possible nickel is preferred.

For some experiments requiring the measurement of fast neutron flux it is difficult to find a suitable threshold detector, and monitoring is often done using thermal detectors. Careful calibrations are required and it is not likely that such a technique could form the basis of an internationally-acceptable method, as it is applicable only in special circumstances.

3. MEASUREMENT OF ABSORBED DOSE AND DOSE-RATE

The main technique in use for the continuous measurement of absorbed dose and dose-rate in materials under irradiation in reactors has been the calorimeter of an essentially isothermal design, discussed in the companion paper.

Calorimeters of a number of different designs, to measure absorbed dose-rates in different materials in different reactors, have been produced by a number of workers in different laboratories, although all have used the same basic design principles. There has as yet been no conscious effort to intercompare these instruments, although a number have been indirectly

compared by gamma irradiation where the observed gamma dose-rate has been measured by the Fricke dosimeter.

The two main research programmes for which these techniques have been developed are

- (i) to confirm nuclear heating calculations and to provide other data required in the design of experimental in-pile rigs; and
- (ii) to study the methods by which reactor radiations modify or induce chemical changes in materials.

This last programme has special relevance to the study of reaction rates between the graphite moderator and coolant gases in power reactors forming part of the United Kingdom civil power programme.

Calorimeters suffer from a number of disadvantages, mainly because their dynamic range is limited, and they do not respond instantaneously to changes in dose-rate.

For these reasons they are being supplemented in the graphite/CO₂ programme by measurements made with mean current ionization chambers. The chambers are about 22 mm diam., with thick graphite electrodes, fully guard-ringed, and have a volume of about 1.36 cm³. Pairs of chambers filled with CO₂ and argon may be used to evaluate the energy deposition rates due to fast neutrons and gammas. The sensitivity is about 4.32×10^{-8} A/mW g.

A programme of inter-comparisons between ion chambers and calorimeters is currently in progress and agreement within a few per cent has been obtained in gamma dose-rates up to 10⁶ rad/h. Similar measurements are in progress in the core of BEPO reactor to compare the response due to fast neutrons. The validity of the ion chamber results depends on the assumption that W , the energy/ion pair, is the same in CO₂ for recoil nuclei due to fast neutrons as for electrons.

A programme of work is being carried out to measure W as a function of neutron energy over the range 50 keV-4 MeV. Preliminary results at 3.5 MeV neutron energy give a value for W of 36.3 eV/ion pair compared with the accepted value for electrons of 36.9 ± 0.3 , but the first figure carries an uncertainty of perhaps as much as $\pm 5\%$. Over the range 0.4 - 3.5 MeV neutron energy (equivalent to carbon recoils between 0.110 + 1.0 MeV) there is no evidence of any large change in W , but below 100 keV it seems likely that it may be appreciably higher.

The present ionization chambers have a maximum operating temperature of about 130°C, but other designs are under development to operate at up to 600°C.

IN-PILE DOSIMETRY TECHNIQUES AND STANDARDIZATION IN THE UNITED STATES OF AMERICA.

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INTRODUCTION

Various methods for the dosimetry of neutrons and gamma rays in reactors are reviewed. Generally the emphasis is on novel methods which may not yet be perfected, inasmuch as more conventional techniques have already been reported in some detail at previous meetings of the International Atomic Energy Agency, such as the one held at Harwell in 1962 [1].

Standardization programmes in the field of in-pile dosimetry are being conducted by two groups in the United States of America. The Irradiation Standards and Procedures Committee (ISPC), formed under the auspices of the United States Atomic Energy Commission (USAEC) Program, Radiation Effects to Reactor Structural Materials, is primarily concerned with neutron flux measurements for radiation damage studies. The American Society for Testing and Materials (ASTM), E-10 committee, broader in scope and national in membership, has distributed several tentative procedures for measurement of neutron flux. The tentative procedures are under active consideration and appear close to acceptance.

THERMAL NEUTRONS

The radiochemical determination of the activation products in such materials as gold, cobalt, indium or dysprosium is a well-established technique for the measurement of thermal neutrons. Some alternative methods, now under development, do not require radiochemical analyses.

The change in ratio of selected isotopic pairs is related to the integral neutron exposure received. Isotopic ratios can be determined with accuracies of one part in a thousand using a mass spectrometer and are not subject to limitations encountered in specific radioactivity analyses.

An example of the method is the use of the isotopic ratio U^{236}/U^{235} [2]. For exposures causing little depletion of U^{235} , the ratio of U^{236} to U^{235} is directly proportional to the thermal neutron exposure times the cross-section for the production of U^{236} from U^{235} . The equations describing the formation of U^{236} have been solved more exactly and exposures in the range 10^{19} - 10^{22} may be monitored by this method. Measurements yielded consistent results for a series of incremental exposures ranging from 2 to 10×10^{20} n/cm².

The chief limitation on the accuracy of this method in known spectral environments is the accuracy of the cross-sections involved in the analysis. For that reason, the precision measurement of cross-sections for suitable isotopic pairs could be a useful programme for consideration by the Agency.

Another possible isotope pair for this type of measurement is B^{10} and B^{11} . During exposure to a neutron flux, B^{10} is consumed and therefore the $B^{10} - B^{11}$ ratio is changed. It should be possible to monitor exposures of $10^{18} - 10^{21}$ using the naturally-occurring isotopic mixture. If the material is enriched in B^{10} , a larger range could be measured.

Counting etch tracks produced in mica by the disordering action of fission fragments is a convenient and very inexpensive method of detecting neutrons [3]. The minimum requirements for this method are mica, etching acid, and an optical microscope. Such modest equipment requirements recommend this technique to programmes operating on small budgets. This method has been demonstrated for reactor exposures to 10^{20} n/cm², but the upper limit is larger, possibly 10^{22} n/cm².

EPITHERMAL NEUTRONS

The epithermal neutron flux is usually measured by resonance detectors. However, it is necessary to assume a $1/E$ flux distribution, and in many cases calculations indicate that the assumption is questionable.

A useful type of computation has been proposed by McELROY [4] to aid in the measurement of this neutron flux component. A tabulation of effective cross-sections in a variety of spectra is made. By comparing the activation of a number of resonance detectors, one could identify the spectral shape most compatible with the observed data.

At the present time the practice in the United States is to assume a $1/E$ spectrum despite its inadequacy and to analyse the activation of bare and cadmium-covered resonance detectors. A description of the procedure is found in the ASTM Tentative E 199-62T, issued in 1962.

FAST NEUTRONS

The measurement of fast neutrons appears to be standardizing in the United States on the basis of threshold or fission monitors. Nickel and iron are most extensively used. Fissionable materials such as Np^{237} and U^{238} have the notable advantage of lower threshold energy. However, Np^{238} is transmuted by thermal neutrons. Fission products from the transmuted species are a source of difficulty. For U^{238} , fission of contaminant U^{235} by thermal or epithermal neutrons may be troublesome. If materials are shielded from thermal neutrons, these problems are alleviated.

The calculation of neutron exposure actually introduces variability. Past practice has been to report data on the assumption of a fission spectrum even for positions where the spectrum is known to be well moderated. There are two objections to this procedure. First, the activation rate is dependent on the spectral shape. Second, the extrapolation to neutron energies below the activation threshold energy is also dependent on the spectral shape. For these reasons, investigators at several sites [5] advocate the use of calculated spectra in the reduction of activation data to neutron exposures.

For much of the radiation-effects data, the unit expressing exposure to reactor neutrons is nvt $E > 1$ MeV assuming a fission spectrum. However, there are recent proposals to modify the exposure unit based on a more detailed analysis of the displacement process. ROSSIN [6] has introduced a Radiation Damage Unit (RDU) which is proportional to the average effect of a fission neutron. The gross displacement production would be calculated using appropriate displacement models to determine the effectiveness of the particular spectrum in the experimental position.

A similar analysis [7] has shown that a neutron exposure unit may be defined which is proportional to gross displacement production with little dependence of typical reactor spectra or displacement model. This type of analysis has been applied to results of STEELE and HAWTHORNE [8] on the shift in nil ductility temperature of steel with irradiation. Data from a graphite reactor and from the core of a water-moderated test reactor were statistically distinct as originally reported. Through the use of this exposure unit, these results could be reconciled to constitute a single set of data.

From experiments reported on semi-conductors [9], a radiation exposure unit has been proposed based on the relative activation of U^{238} and Pu^{239} . Uranium-238 is a fast-neutron threshold detector, whereas the Pu^{239} is sensitive to neutrons of all energies. From the activations produced in these two materials, an average neutron energy is calculated which is related to the effectiveness of various reactor spectra in causing damage to semi-conductor materials.

GAMMA RADIATION

The measurement of gamma radiation intensity in an operating high flux reactor such as the MTR is a formidable problem. Therefore, the technique developed by LEWIS at the National Reactor Test Station is notable [10]. The calorimeter consists of three units of the same geometrical shape and with identical thermal transfer paths to a common heat sink but incorporating two different materials. The materials are chosen to be quite different in their response to the nuclear heating component. During reactor operation, the electric power required to maintain identical temperature in each unit is measured from which both gamma and nuclear heating may be calculated.

Another technique measures total heat generation in a device without discriminating between gamma and nuclear heating. This device consists of an aluminium block with a known heat transfer to a heat sink. During reactor operation, the block temperature is measured by thermocouples to determine the heating rate. Examples of such routine measurements may be found in [16]. The lack of discrimination between gamma and nuclear heating is desirable when determining the heating rate for an experiment designed to run at a particular temperature in a reactor.

A similar apparatus has been used at Oak Ridge by BOPP and TOWNS [11]. A cylinder is suspended within an isothermal shield and exposed to radiation in a reactor. The isothermal shield is heated several degrees above the ambient temperature and its cooling curve is recorded for use in calculating the rate of heating of the cylinder material.

Nitrous oxide has been advocated as a dosimeter material by SIMPSON [12] of the US Naval Radiological Defense Laboratory and also used by FLORY [13]. This dosimeter is based on the property that nitrous oxide decomposes linearly with dose into the constituents N_2 , O_2 , and NO_2 in the ratio 1.0:0.14:0.48 respectively. The range of $10^6 - 10^{10}$ ergs/g-carbon may be measured in both pure gamma and mixed radiation fields. Little temperature dependence (<10%) is noted from -90 to $120^\circ C$ and the response appears independent of pressure. Rate effects have not been observed, but less than an order of magnitude difference was studied.

STANDARDIZATION

Steps toward greater standardization have been accomplished through the efforts of the Irradiation Standards and Procedures Committee (ISPC) of the Radiation Effects to Reactor Structural Materials Program. The overall programme, sponsored by the USAEC Division of Reactor Development, is conducted at a number of laboratories in the United States*. To insure greater consistency in data from diverse sites, several committees were appointed to co-ordinate the programme and assure compatibility within the programme.

At the first meeting of the ISPC in April 1962, a list of recommended flux-monitor materials was compiled. It was the intention of the committee not to displace traditional methods used by the participating laboratories, but to assure that all experimenters used some common materials to monitor neutron flux. The materials chosen were nickel, iron and titanium for fast-neutron monitoring and cobalt for thermal neutrons. Because the overall programme deals with radiation damage, the emphasis is on the monitoring of the fast-neutron component.

As a second step, the ISPC decided upon a counting intercalibration programme to insure that the counting of the radioactive isotopes used in monitoring did not introduce significant errors. The details of that intercalibration have been reported [14]. Inconsistencies up to 100% were found in the counting of Co^{58} , Mn^{54} and Sc^{46} upon the first distribution of samples. The individuals responsible for the radiochemistry at the several sites met to consider these differences and in many cases unsuspected sources of error were discovered.

A second sample distribution was made and the results then indicated that counting was being accomplished within a range of 5% among the sites. Additional samples of Co^{60} and Co^{58} have been exchanged with the United Kingdom Atomic Energy Authority (UKAEA), Harwell Laboratories and with the EURATOM standards laboratory at Geel. Satisfactory agreement was noted with both sites. As a result, there is now reasonable assurance that many laboratories in Europe and the United States are consistent in their counting of these isotopes.

* Argonne National Laboratory, Battelle Memorial Institute, Brookhaven National Laboratory, General Atomics, Hanford Laboratories - General Electric Co., Naval Research Laboratory, Nuclear Materials and Propulsion Operation - GE Co., Oak Ridge National Laboratory, Phillips Petroleum Co., Southwest Research Institute.

A more extensive intercalibration programme in the United States is now under way under the auspices of the ASTM E-10 Committee. The central site for that intercalibration will be the Hanford Laboratories which served as the central site for the ISPC Program. Thus the two counting intercalibration programmes will be very closely related.

The activities of the ASTM committee are probably much more familiar since they have been reported previously. The problem of neutron-flux measurement finally appears to be achieving some degree of resolution. Specific tentative procedures have been written for neutron flux measurement by the activation of sulphur, nickel, iron, aluminium, and a general tentative procedure has been written on the subject of neutron-flux measurements. All tentative procedures, currently under consideration, have been rewritten several times. With each rewriting, the degree of support has increased and therefore it appears that some of the tentative procedures will be approved soon.

It should be borne in mind that ASTM standards do not prescribe the sole acceptable method for performing a measurement. Rather they define methods whereby different investigators should obtain consistent results. In this sense they constitute a means for intercomparing techniques.

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A STATUS REPORT ON IN-PILE DOSIMETRY TECHNIQUES IN YUGOSLAVIA

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Work in the field of reactor dosimetry at the Boris Kidrič Institute has been concentrated on the following problems:

determination of integral values of thermal, epithermal and fast neutron fluxes;

measurements of absorbed doses of reactor radiation in various materials.

The five years' experience with experiments on the RA 10-MW research reactor have resulted in the standardization of the most frequently used techniques for routine neutron flux measurement (foil technique or ionization chamber technique) and in the development of some new methods, for example, semiconductor counters for neutron flux measurement and for measurements of absorbed doses of mixed reactor radiation, specially constructed calorimeters and the aqueous oxalic acid dosimeter.

The measurements are performed in the core or reflector of the heavy water 2% enriched uranium 10-MW RA research reactor of the Boris Kidrič Institute at Vinča [1].

1. NEUTRON DOSIMETRY

1.1. *Foil technique measurements*

This technique is most frequently used for absolute and relative measurements of neutron flux integral values. The main advantage is a quick preparation of the experiments and the irradiation usually does not interfere with the normal reactor operation.

The standard procedure for the determination of absolute thermal neutron flux values is absolute measurement of the activity of gold and cobalt foils. Gold is mostly used for low power reactor operation up to 50 kW, and activity measurements are made by the beta, gamma coincidence technique. Cobalt foils are used for a somewhat higher reactor power, 500 kW and above, and the measurements are made by the gamma, gamma coincidence technique [2].

Relative measurements are made in places with low neutron flux values, such as the beam ports or the thermal column. The applied method consists of the comparison of dysprosium or indium foil activities with the same foil activities obtained by irradiation in a graphite standard pile.

The thermal neutron flux mapping in experimental holes is performed by the activation of copper, dysprosium or gold foils. Relative measurements are made on a set of four intercalibrated GM counters. The flux mapping is used as a routine check of the distribution changes in the course of the reactor core life, or caused by the influence of some strong absorber.

Epithermal neutron fluxes are determined through the epithermal index r . Owing to the fact that for most irradiations it was necessary to estimate the induced activity, the Westcott convention for effective cross-sections was used, and the knowledge of the 2200 m/s thermal neutron flux and the epithermal index r were sufficient for most calculations. To determine the r -factor distribution in the reactor core and reflector the cadmium ratios for Au and In were measured. The self-shielding factors for epithermal neutrons are determined experimentally.

Fast neutron measurements are standardized with threshold detectors of S, P, Ni, Al and Mg. The average effective cross-sections over the fast neutron spectrum are obtained by graphical integration of the $\delta(E)$ curve. For measurements of the absolute fast flux values inside the hollow fuel element, the fission neutron spectrum is estimated using the fact that the hollow fuel element is a sort of fast neutron converter. When measurements inside experimental holes are made, the fast neutron spectrum is determined by calculation and checked indirectly on the basis of the experiment. It has been assumed that the spectrum inside the hole has the form of a virgin fast neutron flux from the neighbouring fuel elements. If the neutron spectrum form is correctly estimated, the results for the integral flux values obtained by various threshold detectors will be consistent. This is usually the case, as is shown by the experimental results [3, 4].

1.2. Fission chamber measurements

In the early period of reactor exploitation fission chambers were used for neutron flux distribution measurements [5]. The fission chambers were argon-filled [3 atm] with the addition of 6% CO₂. For thermal neutron fluxes enriched uranium layers inside the chamber are used and for fast neutron fluxes a thorium coating is used. For measurements inside the reactor core the chambers are mounted on a long aluminium tube and by a special technique moved along the vertical experimental holes. Preamplifiers are kept outside the reactor holes. The results obtained are consistent with foil measurements. The measurements are quick and easy but their preparation takes a long time and low power operation of the reactor is required.

1.3. Semiconductor counter measurements

Semiconductor detectors allow the detection of fission fragments and heavy particles from various reactions such as (n, alpha), (n, p), etc. Semiconductor counters using both fission and n, alpha reactions are broadly used for neutron detection and measurements at the Vinča Institute. In addition to the standard procedure where the counter is in direct contact with the target and the whole system placed inside the reactor core, a special procedure has been developed where the counter is separated from the target. In this method, usually called the long tube method, the target is placed at the bottom of a long evacuated Al-tube immersed in the reactor. A semiconductor counter is placed at the other end of the tube outside the reactor core. In this way the measurements of thermal and fast neutron fluxes are possible even if the reactor operates at a very high power and high flux values. This method is used for a quick determination of the

neutron flux distributions inside the experimental holes of the RA reactor without stopping or even changing the power level. The long tube method, in principle, permits the determination of absolute values of the neutron flux if the solid angle in the system target-detector and the target efficiency are known. The preliminary results show that absolute values of the thermal neutron flux can be determined if the sources of errors are eliminated as, for example, the fission fragments scattered from the tube walls, self-shielding of the target, etc. [6, 7].

The Li^6 target with a semiconductor counter has been developed as a neutron spectrometer for fast neutron spectra measurements. Its practical use encounters difficulties because of the fact that the resolution is poor due to the background gamma radiation of the reactor core and the sensitivity of the Li^6 detector to thermal neutrons. Using the fact that the fragments of the $\text{Li}^6(n, \alpha)\text{H}^3$ reaction with thermal neutrons form an angle of 180° , the geometry of the counter is adjusted so that these fragments cannot reach the detector. The thermal neutron background is thus greatly reduced. Application is proposed for fast reactors or low power thermal reactors. The method is being successfully used for fast neutron spectra determinations inside the RB zero power reactor, with a specially closely-spaced lattice core configuration [8].

2. MEASUREMENT OF PILE RADIATION ABSORBED DOSES

2.1. Calorimetric measurements

For the period of experimental pile operation at low powers (up to 100 kW) an isothermal calorimeter with thermistors was constructed [9]. It measured the absorbed dose-rates of mixed radiation from 10^4 to 10^6 rad/h. The measurements were made in a few different materials (H_2O , D_2O , graphite) as calorimetric bodies, so that the gamma and neutron contributions to the absorbed dose could be calculated [10]. The accuracy of direct measurements was within 2-5%. It was shown that pile radiation accompanied by $(nvt)_{\text{th}}$ of the order of 10^{12} n/cm².s, and total $(nvt)_{\text{th}}$ up to 10^{16} n/cm².s had no influence on the characteristics of the thermistors used. However, the values calculated from experimental results were not quite in good agreement with those estimated from neutron flux measurements. A high proportion of gamma radiation contribution to the absorbed dose (about 80%) pointed to the presence of delayed gamma radiation from fission products which could not be neglected at low powers. Some later measurements with the Fricke dosimeter proved this, indicating a possible contribution of the order of 20-30% to the total energy absorption measured calorimetrically. Therefore, this delayed gamma radiation was probably the source of the discrepancies at low reactor powers. Owing to the uncertainty of such corrections at low powers we constructed another system for measurements at high reactor power (up to 1 MW).

A differential calorimeter with three bodies [11] permits the simultaneous determination of the energy absorption and the contributions of the different components of the pile radiation. Besides, the estimation of the different corrections necessary for isothermal or, in general, one-body

calorimeters was avoided here. In such a device one body was empty (reference) and the other two contained materials with different cross-sections for neutron elastic scattering. Measurements were made in different materials as calorimetric bodies: H_2O , D_2O , graphite, polystyrene, polyethylene. This construction is specially suitable for experiments in the pile radiation chemistry of water and aqueous solutions; a calorimetric body can be a sample in which, after irradiation and energy absorption measurement, chemical analyses are made. At the same time the contributions of neutron and gamma radiation can be obtained. Results obtained with this device are in good agreement with those derived from other physical measurements [12].

2.2. Aqueous oxalic acid as a chemical dosimeter

Following an earlier suggestion [13] for the use of the decomposition of aqueous solutions of oxalic acid in chemical dosimetry, subsequent studies have led to the development of a dosimetric procedure [14, 15]. Compared with existing chemical dosimeters, the oxalic acid dosimeter has many advantages: induced radioactivity is negligible; the dose-rate range and the upper limit that can be measured are considerably greater and permit measurements during long irradiations or at fairly high dose-rates; the system is neither sensitive to impurities nor photosensitive. A good reproducibility in routine work can be achieved [16]. Although it seems to be the only aqueous chemical system at present available which can be used for measuring large radiation doses within the reactor, it is certainly not an ideal dosimeter. One of the reasons is the fact that the amount of the decomposed acid used as a measure of the absorbed dose must be determined from the concentration difference before and after irradiation; the smaller the difference, the higher the possible error. Because of the non-linearity of high conversion, this system requires lengthy calculations and previous selection of the conversion ranges is desired so as to permit dose measurements in the multi-megarad region at levels of conversion at which linear behaviour is encountered. Other inconveniences are more or less the same as those of other aqueous systems - undesirable heating of the samples at very high dose-rates and the formation of large amounts of gases (H_2 and CO_2) at high absorbed doses which produce considerable pressure in the dosimetric vessel. The radiation-chemical yield is LET-dependent and in the calculations the relative contribution of the different LET components (f_{gamma} and f_p) have to be taken into account,

$$G(-H_2C_2O_4)_{\text{pile}} = f_{\text{gamma}} \times G_{\text{gamma}} + f_p \times G_p,$$

where f_{gamma} and f_p are derived from physical measurements.

$G_{\text{gamma}} = 4.91 \pm 1.5\%$ is measured calorimetrically [17]. G_p depends on LET values of protons in given conditions. As derived from recent measurements in the core centre of the RA reactor it is fairly high (3.3) and indicates a low proton energy under the given circumstances. Further experiments are in progress to get more information about the low-energy-particles component in the mixed pile radiation and the corresponding decomposition yields of the oxalic acid dosimeter.

2.3. Solid oxalates for in-pile measurements

Our present work refers also to the possibility of the use of some solid oxalate in the chemical dosimetry of pile radiation. The preliminary results indicate that as a measure for the absorbed dose not only could the decomposition of the solid samples be used, but also spectrophotometric and luminescence measurements of the products formed during irradiation [18].

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ACTIVITIES OF THE EURATOM DOSIMETRY WORKING GROUP FROM 1962 TO 1964

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1. INTRODUCTION

The present report of the Euratom Dosimetry Working Group supplements the first one* and refers to the activities of the group from September 1962 to June 1964. In this period the 7th to 12th plenary meetings, as well as several meetings of the sub-groups, were held. Sub-groups have been formed for the following fields:

the use of cobalt for integral flux measurements of thermal neutrons; sample technology; cross-section compilations; and terminology.

The sample technology sub-group held two meetings at Saclay and Geel and tried to obtain a first survey on the possibilities and requests of sample preparation. For example, several thousands of samples are requested at the Bureau Central de Mesures Nucléaires (CBNM), so that some organization and normalization is a worthwhile task.

The terminology sub-group has directed its efforts into two main channels, one to unify the use of symbols and neutron physics expressions [114, 143, 144, 145, 169] and the other to determine a common language in the field of errors and corrections of measured values [155, 156].

2. MEASUREMENTS ON THERMAL NEUTRONS AND LOW-ENERGY INTERMEDIATE NEUTRONS

For more details on this subject the references to this paper should be used. We mention here only those subjects which are considered as most important. These can be summarized as follows:

(a) The use of cobalt to integrate thermal neutron fluxes. Working Group members have been advised to use this method systematically to facilitate comparison between the experiments carried out in the different reactors of the European Community countries. Publication of the text giving details of application of the method has been delayed and the final document was only issued for the 13th Working Group meeting in October 1964. This

* DELATTRE, P. and PROSDOCIMI, A., Les activités du groupe de travail Dosimétrie d'Euratom, EUR 88, f (1962).

delay was caused by the fact that new comparative measurements of gold and cobalt cross-sections needed to be done in order to choose a coherent value for the latter compared to the one for gold, taken as a reference at 98.8 barns [132, 133]. For a valid standardization it has also been necessary to make a first intercalibration of the counting equipment belonging to the laboratories of the Community countries concerned [113]. To perform this task, standards of cobalt have been distributed to the different laboratories by the CBNM.

(b) Investigation of the best values of resonance integrals for detectors of interest and study of spectrometry problems in the fields of low-energy intermediate neutrons [106, 120, 134].

(c) Study of methods of continuous measurement of instantaneous fluxes to follow their variations during reactor operations, particularly in the neighbourhood of experimental facilities [125, 167, 168].

3. MEASUREMENTS ON HIGH-ENERGY INTERMEDIATE NEUTRONS AND FAST NEUTRONS

For details on this subject the reference list to this paper should be used. The most important subjects can be summarized as follows:

(a) Choice of cross-section values. Differential curves versus energy and comparative values of cross-sections for the main reactions of interest averaged on a given spectrum [101, 102, 104, 105, 112, 115, 123, 129, 163]. The Working Sub-Group for the compilation of cross-sections met at Geel in April 1963 to discuss the most useful reactions and the method of compilation [128]. Meanwhile, the CBNM published a second volume of the compilation in August 1963 [129], giving the differential curves as a function of energy. It is intended to keep this compilation up-to-date by yearly additions and amendments.

(b) Determination of spectra by experimental methods only (threshold detectors, fission chambers, nuclear emulsions, and lithium spectrometer) or by mixed methods, i. e. theoretical and experimental [92, 107, 109, 118, 135, 137, 148, 149, 150, 151, 152, 159, 165].

(c) Integration of fast neutron doses [108, 119, 130, 153] by using threshold detectors or quartz detectors.

(d) Definition of a standard spectrum for fast neutrons [126, 136, 147, 154].

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ACTIVITÉ DU BUREAU INTERNATIONAL DES POIDS ET MESURES DANS LE DOMAINE DES RAYONNEMENTS IONISANTS

A. ALLISY

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Le Bureau International des Poids et Mesures (BIPM) est l'agent de liaison entre les laboratoires de métrologie internationaux ou nationaux et son rôle est de promouvoir une meilleure uniformité des mesures dans le monde. Dans le domaine des rayonnements ionisants, le BIPM effectue des mesures et organise des comparaisons internationales portant sur les grandeurs suivantes:

Exposition des rayons X et gamma

- a) Des chambres d'ionisation de transfert permettant la comparaison d'étalons primaires d'exposition dans le domaine compris entre 50 keV et 1 MeV circulent entre les laboratoires nationaux de métrologie.
- b) A Sèvres, un générateur de rayons X ultra-stable a été monté et des sources intenses de ^{60}Co et ^{137}Cs vont être mises en place pour la comparaison des instruments de transfert qui nous sont envoyés.

Activité de radionuclides

- a) De nombreuses comparaisons internationales ont été organisées depuis 1961 et des rapports analysant les résultats et décrivant les techniques ont été rédigés. Les nuclides suivants ont été traités: ^{32}P , ^{131}I , ^{60}Co , ^{198}Au , ^{204}Tl , ^{35}S , ^{241}Am , ^{90}Sr et sources solides de ^{60}Co . Cette dernière comparaison, en particulier, a été réalisée avec le concours du laboratoire de mesures nucléaires de l'EURATOM et le «National Physical Laboratory» (Teddington); elle trouve une application très directe aux mesures effectuées dans les réacteurs nucléaires.
- b) Une installation de comptage $4\pi\beta\text{-}\gamma$ de très haute précision est en fonctionnement quasi permanent à Sèvres et le laboratoire du BIPM prend part régulièrement à toutes les comparaisons internationales de radionuclides qui sont organisées.

Mesures neutroniques

- a) Une source de neutrons Ra-Be (α, n) prêtée par le National Research Council du Canada a circulé dans plusieurs laboratoires qui en ont déterminé le taux d'émission. Un rapport analysant les résultats est en préparation.
- b) Le laboratoire de mesures neutroniques de Sèvres a mesuré absolument et relativement cette source, en particulier par la méthode du bain de manganèse. Un étalon Ra-Be (α, n) est maintenu en permanence au

BIPM et son taux d'émission est en voie d'être déterminé avec la précision la plus grande possible dans l'état actuel de la technique. Une source Ra-Be (γ, n) est acquise pour servir également d'étalon de source. Un accélérateur d'ions positifs à 150 keV est installé; utilisé comme générateur de neutrons rapides, il est destiné en particulier à l'étude d'instruments de transfert dans ce domaine d'énergie.

SUMMARIES OF SOME PAPERS PRESENTED TO THE PANEL

IN-PILE DOSIMETRY FOR RADIATION DAMAGE STUDIES*

SUMMARY**

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For the design of a reactor it is of utmost importance to know the amount of radiation damage produced in structural materials under a given irradiation of neutrons. Properties such as strength, dimensions or thermal conductivity are subject to change as a result of exposure in a reactor. Perhaps the greatest difficulty in interpreting radiation damage data is the lack of a widely accepted system for reporting exposure. Radiation exposures are reported in units of time-integrated neutron flux, neutrons per square centimeter (nvt). It is thus necessary to have a consistent scale of exposure in order to interpret experimental measurements properly and also to compare results from different test reactor facilities. Results of work done in this direction (in-pile dosimetry) at Hanford are reported in this paper and a suitable exposure unit (in terms of radiation damage or "gross displacement" produced) is proposed.

As a first step, it is necessary to have an accurate idea of the fast-neutron spectrum at different positions in the reactor lattice under consideration. This can be achieved with the help of several available multi-group computer codes of different degrees of sophistication, e. g. GNU-II, SFN, GE-HAPO-SX, etc., of which the last one has been used in the present work because of its greater refinement, flexibility and applicability. One should note that the assumption of an unmoderated fission spectrum in a reactor is, in general, not justified. Having obtained the spectrum $\phi(E)$, we can compute the effective cross-section σ_{eff} for activation of a monitor material defined as

$$\sigma_{\text{eff}} = \frac{\int_0^{10} \phi(E)\sigma(E)dE}{\int_{E_1}^{10} \phi(E)dE}$$

where $\sigma(E)$ is the energy-dependent activation cross-section and E_1 is the reference energy chosen to correlate radiation effects obtained in different situations. σ_{eff} can now be used to calculate neutron exposures from monitor activities over the chosen energy range. Most threshold monitors are activated by neutrons with energies above 3 MeV, whereas exposures are calculated to limits of 1 MeV or lower. This involves a sizeable extrapolation. Furthermore, it is known that a major fraction of vacancy production is

* Work performed under United States Atomic Energy Commission Contract No. AT(45-1)-1350.

** The paper forming the basis for this summary will appear in Nuclear Science and Engineering.

caused by neutrons of less than about 1.5 MeV. Hence it becomes evident why the spectral shape should be known to discuss exposure.

The values of σ_{eff} for Fe⁵⁴ and Ni⁵⁸ monitors placed at different locations, for two reactor irradiation facilities GETR and ETR have been calculated. For greater accuracy in the complex geometry, a two-dimensional transport code 2DXY was used here for computation. The data lead to entirely consistent results for neutron exposure as obtained from nickel and iron monitors in both the reactors. This also holds true for large graphite-moderated reactors. However, one should note that it is necessary to locate quite accurately the positions of both flux monitors and samples as the flux intensity can change from one position to another within the same irradiation facility. Best available values for $\sigma(E)$ for Ni and Fe were used for the above calculations, but it is necessary to secure a more reliable neutron activation curve for iron than the one available now.

NEUTRON EXPOSURE UNIT

For much of the radiation effects data, the unit expressing exposure to reactor neutron has been nvt $E > 1$ MeV assuming a fission spectrum. In consideration of the displacement process in the irradiated material, the following definition of the unit, which is proportional to the gross displacement production with little dependence on typical reactor spectra or displacement model, is being put forward here.

It is proposed that E_L , the lower limit of energy integration in the definition of neutron exposure, should be such that the integrated flux above E_L is proportional to gross displacement production for different spectra, i. e. the quantity

$$D(E_L) = \int_0^{\infty} N(E)\sigma(E)\phi(E)dE / \int_{E_L}^{\infty} \phi(E)dE,$$

where $N(E)$ is the number of displacements produced by a neutron energy E , $\sigma(E)$ is the differential elastic scattering cross-section, and $\phi(E)$ is the neutron spectrum, should be constant in different spectra. For example, using the Kinchin and Pease displacement model it was found that for iron, $D(E)$ calculated on the basis of a Watt fission spectrum, an ETR spectrum and a fully-moderated graphite reactor spectrum came out to be the same if one took $E_L = 0.4$ MeV. It was also found that change in models caused only relatively small changes in E_L . Calculations have been made using the Kinchin and Pease model for elements ranging in atomic weight from beryllium to tungsten and it was possible to choose an E_L (which is different for each material) which gives less than 5% variation in the calculated number of displaced atoms in different spectra.

As an application of exposure unit to experimental data, results obtained by Steel and Hawthorne [1] on the shift in nil ductility temperature of stainless steel with irradiation, from a graphite reactor (BGR) and from the core of a water-moderated test reactor (LITR) were analysed. The data were statistically distinct as originally reported. Through the use of this exposure unit and $E_L = 0.5$ MeV, these results could be reconciled to constitute a single set of data. Even closer agreement between the results can

be achieved by using the limit 0.18 MeV. Thus the improvement obtained through the use of calculated spectra is evident.

The exposure unit used in this analysis is recommended for further study to determine its suitability as a general exposure unit for radiation damage studies.

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REVIEW OF METHODS FOR NEUTRON AND GAMMA MEASUREMENTS AT THE FRM REACTOR, MUNICH

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A description of the FRM reactor at Munich and the experimental facilities is given by H. Maier-Leibnitz elsewhere [18].

A. ROUTINE MEASUREMENTS OF

(a) Flux density

thermal neutrons $0 < E < 0,5 \text{ eV}$

- (α) Gold foils: 20 μm thick, area = 1 cm^2 [1]

The thermal flux ϕ_{th} is evaluated from the difference of the induced activities $A_{\text{th}} = A_{\text{bare}} - A_{\text{Cd}}$, $\delta_{\text{th}} = 98.8 \text{ b}$

- (β) intermediate neutrons $0.5 \text{ eV} < E < 0.1 \text{ MeV}$

$$\phi_{\text{res}} = \int_E^{CE} \phi'(E) dE \quad \phi'(E) \approx 1/E$$

$$\phi_{\text{int}} = \int_{0.5 \text{ eV}}^{0.1 \text{ MeV}} \phi'(E) dE$$

Cd-covered gold foils (0.5 mm thick)

Effective activation integral (for β -counting) [2]: IG = 358 b

Within the core of the light-water-moderated reactor [3]: $\phi_{\text{int}} = 12.2 \phi_{\text{res}}$

(γ) fast neutrons $0.1 \text{ MeV} < E < \infty$

For the reaction $P^{31}(n, p) Si^{31}$ the effective threshold energy was determined from the calculated flux density distribution [4] and $\delta(E)$ [5]:

$$E_{\text{eff}} = 2.9 \text{ MeV} \quad \delta_0 = 0.140 \text{ b.}$$

The calcium metaphosphate-disc $Ca(PO_3)_2$ ($\phi = 10 \text{ mm}$, $d = 1 \text{ mm}$) is covered with Cd to reduce the thermal activation of P^{31} to P^{32} . An effective cross-section is also calculated:

$$\hat{\delta} = \frac{\int_0^{\infty} \delta(E) \phi'(E) dE}{\int_{0.1 \text{ MeV}}^{\infty} \phi'(E) dE}$$

$$P^{31}(n, p) Si^{31}: \hat{\delta} = 0.020 \text{ b}$$

$$S^{32}(n, p) P^{32}: \hat{\delta} = 0.040 \text{ b}$$

(b) *Integrated flux density* [6]

(α) thermal neutrons

Co-detectors (with correction for epithermal activation)

$$\delta_0 = 38 \text{ b [7]}$$

(β) intermediate neutrons

Co-detector covered with Cd (not yet used)

(γ) fast neutrons [8]

$$Ni^{58}(n, p) Co^{58} \hat{\delta} = 59 \pm 10 \text{ mb}$$

$$Ti^{46}(n, p) Sc^{46} \hat{\delta} = 6 \pm 1 \text{ mb}$$

(c) *Gamma radiation*

There are no routine measurements of gamma radiation in irradiation positions.

B. SPECIAL MEASUREMENTS OF

(a) *Energy distribution of the non-thermal flux density*

The calculated flux density distribution was checked by several activation and two fission detectors [9, 10].

	$\delta_0(\text{b})$	$E_{\text{eff}}(\text{MeV})$
$Th^{232}(n, f)$	0.140	1.40
$U^{238}(n, f)$	0.600	1.55
$P^{31}(n, p)$	0.140	2.9
$S^{32}(n, p)$	0.350	3.2
$Mg^{24}(n, p)$	0.060	6.3
$Fe^{56}(n, p)$	0.110	7.5
$Al^{27}(n, \alpha)$	0.113	8.1

If the measured flux densities above the effective threshold energies agree within the margins of error with the calculated distribution, the flux density per energy interval is fixed by the measured intensities, and an interpolation is carried out between this fast region and the measured resonance flux [9, 11].

For the irradiation positions in the core and in the graphite reflector elements adjoining the core the two extrapolations (= interpolation) coincided within 10 to 15%. For most of the experiments (e.g. solid state physics [12]) this accuracy is sufficient.

(b) Gamma flux

The gamma flux in a certain position near the centre of the core was determined by direct measurement of the heating of an aluminium rod [13]. The rod was cooled at one end by pool water. At 1 MW the gamma flux was 2.4×10^8 r/h.

Another measurement of gamma flux was done with chemical dosimetry (ferrous sulphate [14]) by SCHÖN [15]. In the pneumatic irradiation facility the gamma flux is 3.7×10^7 r/h.

(c) Distribution of thermal neutron flux density

The heating of about 1 mg U^{235} was used for measurements of the flux distribution within the fuel elements [16]. In another set-up the changes of the core flux distribution caused by the different settings of the shim rods were detected in order to find a core arrangement which ensures a good constancy of neutron fluxes at all locations in and around the core [17].

(d) Fast neutron measurements

by the density change of quartz (SiO_2) are in preparation.

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FAST NEUTRON FLUX DENSITY MEASUREMENTS

SUMMARY

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Comparison of irradiation experiments with each other and with theoretical calculations is not possible without knowledge of the intensity and the energy distribution of the radiation field within the reactor. An international agreement about the data to be measured and the methods of their measurement would be very useful.

The determination of the neutron-flux density in the thermal region is comparatively easily done by the use of a $1/v$ detector, since the energy distribution may be assumed to be a Maxwellian. The definitions (e. g. the conventional thermal flux density) and the methods of measurement are well known and generally used [1]. The evaluation of the flux density in the region of (n, γ) resonances is also well known [2].

To calculate the activation by (n, p) , (n, α) , or $(n, 2n)$ reactions, the use of the "equivalent fission" flux density [3] may be sufficient, but not for physical investigations in connection with the irradiation of metals or graphite. The radiation damage is mainly caused by neutrons with an energy of between 0.1 MeV and 5 MeV (Fig. 1) [4, 5, 6, 7]. Such irradiations should therefore be characterized by the fast-flux density, $\phi(0.1 \text{ MeV})$, and the differential flux density, $\phi'(E)$ for $0.1 \text{ MeV} < E < 5 \text{ MeV}$.

The energy distribution of fast neutrons in reactors has been investigated by many authors during the last three years [8-19]. To conclude I should like to suggest the following ways for its determination:

- (a) Calculation of energy distribution. For thermal reactors many methods are already described in the literature [13-19];
- (b) Test of the assumed distribution. In some cases it might be possible to use differential methods [20, 21], but usually only the activation detectors (e. g. comparison of calculated and measured activities [19, 22, 23], determination of parameters [10, 13, 24]). Each method has some

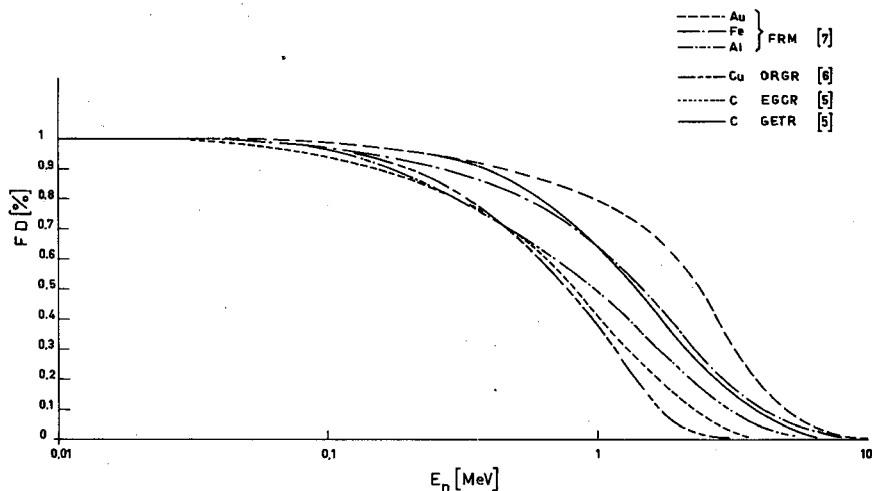


Fig. 1

Damage by neutrons above the energy $E(\%)$

advantages and some disadvantages; a recommendation for standardization of testing is not possible at the present time;

- (c) Interpolation between the measured resonance-flux density and the measured fast-flux density according to the calculation or by the semi-empirical formulation of GENTHON [14].

Figure 2 shows the evaluation of $\phi(0.1 \text{ MeV})$ for some measurements in different reactors.

Figure 3 gives the interpolation for a position within the core and for one position in the graphite reflector.

In the same manner a determination may be possible for other irradiation facilities.

Since such measurements have already been done in many reactors, standardization might be obtained by the recommendation of the "best" differential cross-section for those reactions normally used. Before the recommendation of such a set of "best" $\sigma(E)$ values, better data of $\sigma(E)$ are necessary, especially for reactions with a low effective threshold energy (e. g. (n, n') reactions of In^{115} , Nb^{93}).

Further assistance in many experiments would be provided by a compilation of calculated spectra with the activation rate of several detectors.

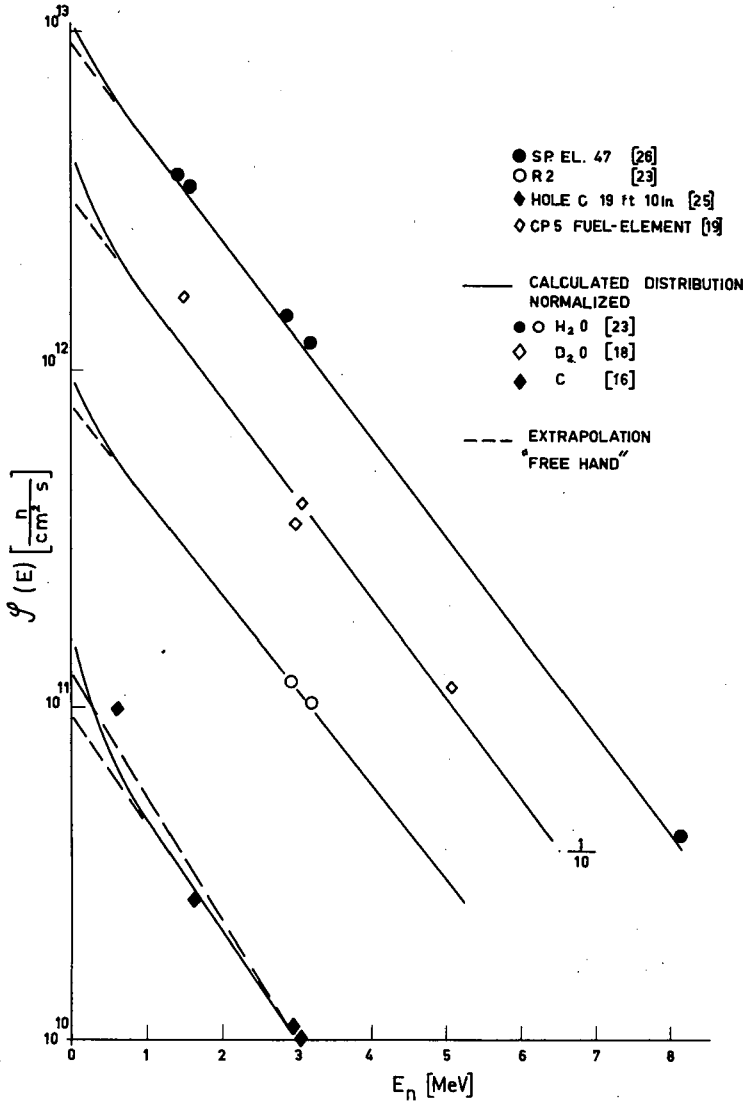


Fig. 2
Fast flux density

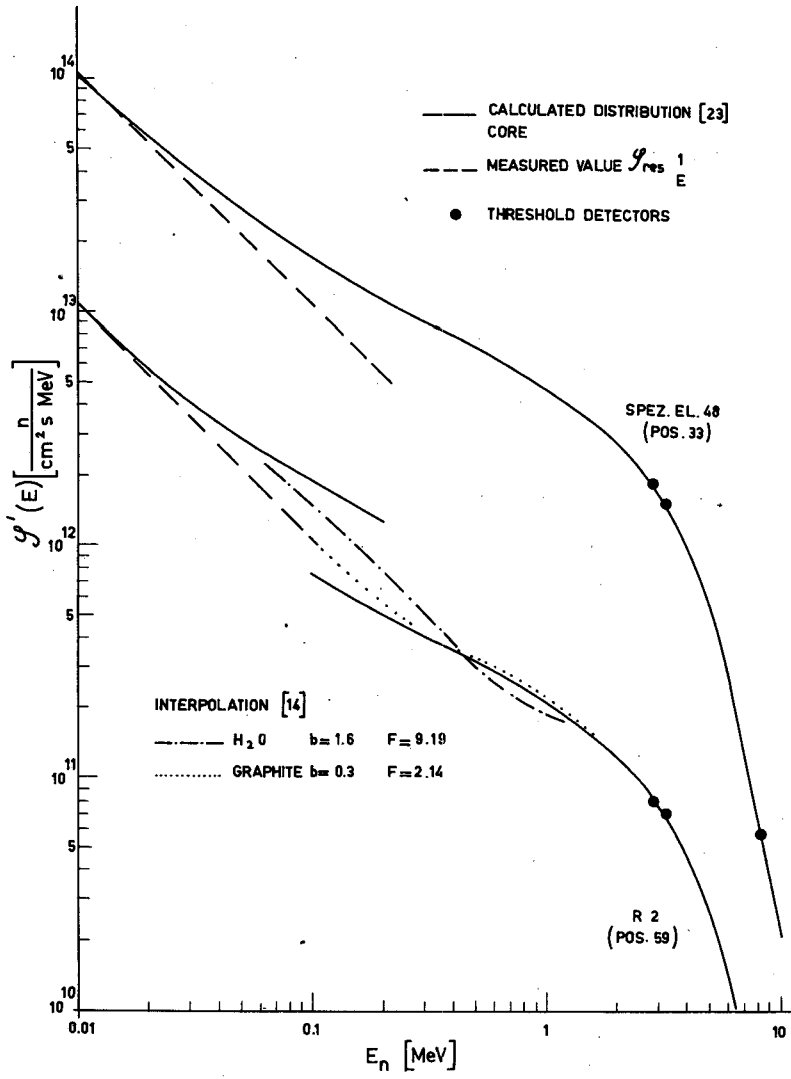


Fig. 3

Light-water-moderated reactor

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REVIEW OF ACTIVATION METHODS FOR THE DETERMINATION OF NEUTRON SPECTRA

SUMMARY*

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PART I. INTERMEDIATE NEUTRON SPECTRA

This review article gives attention to the conventions for flux density and cross-sections, and to the concepts of cadmium cut-off and of resonance integral. After a more detailed examination of the self-shielding correction factors, different methods for the determination of the intermediate flux density and the slowing down density are considered. Resonance reactions of interest are mentioned, together with important nuclear data.

The general conclusions given in the review are:

1. When using or presenting resonance integral cross-sections it is necessary to state the lower energy limit, the $1/v$ contribution, the correction factors applied and the nuclear data used. The recommendations of the European American Nuclear Data Committee (EANDC) should be followed.
2. In cadmium ratio measurements, the type and the dimensions of the cadmium cover used should be specified. These procedures should be internationally standardized.
3. At present there is a preference for the Westcott convention for flux densities and cross-sections. The unified formalism as proposed by Nislé, but not yet discussed in the literature, requires full attention.
4. There is general need for more accurate values for resonance parameters and for activation resonance integral cross-sections. The following resonance detectors have been used in several laboratories: indium, gold, manganese, cobalt, copper, tungsten and sodium.
5. Self-shielding correction functions are important when thin detectors cannot be used. More experimental data for several resonance reactions are very welcome. They have to be compared with theoretical relations, and with computer results for those reactions in which scattering cannot be neglected.
6. It is not yet justified to make recommendations for standard procedures for measuring intermediate neutron flux density, because for many resonance reactions, the required resonance data are not sufficiently well known.
7. With respect to radiation damage studies, methods have to be elaborated for the measurement of intermediate flux densities and fluence in the keV region. Because of space and temperature limitations inside a reactor the use of covers or shields for the activation detectors is not always acceptable.

* The full Review contains 74 references and three tables, with nuclear data for several resonance reactions.

Here we have severe experimental difficulties and much development work has still to be performed.

8. From comparison of the experimental data and techniques with activation detectors for measuring flux densities and fluences of fast neutrons with those of intermediate neutrons, it is obvious that the whole field of intermediate neutron measurements needs development.

PART II. FAST NEUTRON SPECTRA

A short review is given of the possibilities of determining the shape of the fast-neutron spectrum from the counting results obtained with threshold detectors.

The general conclusions given in the review are:

1. The accuracy of absolute fast-flux values is determined by the accuracy of the cross-section data used, and by the accuracy of the measurement of the absolute activity. For routine work it is an advantage if the induced activities in the samples can be counted without chemical separation. The samples must, therefore, be as pure as possible.
2. Bearing in mind the large discrepancies between the values for the average cross-section for fission neutrons from different laboratories and the need for calibration or intercalibration of threshold detectors, it is worthwhile to consider the possibility of establishing a reference flux of fast neutrons, for example, by means of a converter device in a thermal column of a reactor.
3. There is a growing need for threshold detectors with an effective threshold lower than that of the $S^{32}(n,p)P^{32}$ reaction. Especially the range from 1 MeV down to the resonance region is of interest, because the radiation damage effects here are still quite important.
4. As more experimental data on the spectral distribution of the fast neutrons become available, these data have to be compared with the results from multi-group calculations.
5. When presenting experimental data on fast-neutron distributions it is necessary to mention the cross-section data used. It is recommended to state the measured specific saturation activities of the threshold detectors, and the material composition of the detector, so that the spectral indices can be derived from the data.
6. Although many mathematical methods for the analysis of the data and for the calculation of the spectral shape have been proposed in the literature, there is not yet a generally-accepted best method. The most promising methods at this moment are (a) the method of spectral indices; and (b) the method in which the spectrum is approximated with exponential functions in suitably chosen energy ranges.

MEASUREMENT OF THERMAL AND EPITHERMAL NEUTRON FLUXES WITH THE DOUBLE DETECTOR FOIL TECHNIQUE

SUMMARY

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Usually measurements of the thermal and epithermal fluence components are made with the Cd-difference method. This, however, has several serious disadvantages, above all the flux perturbation that is introduced and the fact that the irradiations have to be performed either at two different times or in two different positions.

In the double detector foil method these drawbacks can be avoided as two closely-packed foils of different materials are irradiated in the reactor in the same location at the same time. It is advantageous, if the materials can be chosen so that the ratio between the resonance integral and the thermal activation cross-section is large, and that cross-sections and induced activity half-lives are fairly similar for the two detector materials. This has led to the choice of Au + Cu as detectors for flux measurements at low reactor power and Co + Ag at higher powers (same MW).

The counting rates measured on the two probes A and C after the activation (Z_A and Z_C) can be described by two linear equations,

$$Z_A = a_A \phi^M + b_A \phi^{epi} = Z_A^M + Z_A^{epi}$$

$$Z_C = a_C \phi^M + b_C \phi^{epi} = Z_C^M + Z_C^{epi}$$

where

ϕ^M = integral thermal flux,

ϕ^{epi} = integral epithermal flux.

The factors a_i and b_i can be calculated from the thermal cross-section, the resonance integral, the counting geometry, the time of irradiation and the β self-shielding in the detector foil. The resonance integral and the self-shielding are not well enough known for copper and silver, but it is possible to avoid this difficulty.

With

$$a = \frac{a_C}{a_A} = \frac{Z_C^M}{Z_A^M}$$

$$b = \frac{b_C}{b_A} = \frac{Z_C^{epi}}{Z_A^{epi}}$$

the following relations are obtained for Z_A^M and Z_A^{epi}

$$Z_A^M = \frac{Z_C - bZ_A}{a - b} = a_A \phi^M$$

$$Z_A^{epi} = \frac{aZ_A - Z_C}{a - b} = b_A \phi^{epi}$$

Z_A and Z_C are the measured total count-rates for the probes A and C, Z_A^M and Z_A^{epi} are obtained from activation of the probe A and C by thermal and epithermal neutrons.

Here the factors a_A and b_A can be calculated for one of the foils (Au or Co respectively) and a and b can be determined experimentally; a can thus be determined through irradiation in a pure Maxwellian spectrum, for instance in a thermal column; b is the ratio of the count-rates for the two foils irradiated in Cd sheath; a and b are dependent on the irradiation time and the thickness of the foils, but these functions can be determined accurately.

ACTIVITIES OF EURATOM IN THE FIELD OF IN-PILE DOSIMETRY

SUMMARY

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1. In October 1960 Euratom set up a Dosimetry Working Group to facilitate the exchange of information between the countries of the European Community in the subject of in-pile dosimetry, and for standardization in this field*.
2. The activities of the working group are described in two papers [1, 2].
2. The Laboratories of Euratom's Central Bureau for Nuclear Measurements (ECBNM), Geel, Belgium, contribute directly to the standardization effort in the following ways:
 - (a) Fabrication and distribution of pure cobalt detectors (foils and wires), corresponding to the specifications laid down by the working group in

* The group is presided over by Mr. Delattre (Centre d'études nucléaires (CEN), Saclay) and the secretariat is at Euratom's Central Bureau for Nuclear Measurements, Geel, Belgium, Dr. K. Gubernator being its present secretary.

- a standard sheet on thermal flux integration. Studies of flux perturbation have been performed [3].
- (b) Fabrication and distribution of Co^{60} sources for calibration of counting equipment used for thermal flux integration. A description of some of the work done in this respect is given in [4, 5].
 - (c) Re-measurement of the Co^{59} thermal-neutron activation cross-section is under way.
 - (d) Improvement of absolute counting methods for radionuclides of interest in neutron flux and spectra measurements. An example is given in [6].
 - (e) A study, in collaboration with CEN, Grenoble and CEN, Mol, on the use of small quartz crystals as fast-neutron flux monitors is in progress.
 - (f) Data on most of the neutron-induced threshold reactions, used in in-pile measurements, have been compiled, critically reviewed and graphically represented [7, 8]. Those compilations, which are in loose-leaf form, are kept up to date by issuing new graphs when new data become available.
 - (g) Measurements of cross-sections of neutron-induced threshold reactions have been performed and others are in progress [9].

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DOSIMETRY REQUIREMENTS IN SUPPORT OF AN IN-REACTOR RADIATION CHEMISTRY PROGRAMME

SUMMARY

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Reactors are now used as radiation sources in a wide range of studies. In some, the changes observed result from the interaction of the irradiated material with only one type of radiation present in the reactor; in others, a number of different radiations may produce the observed effect. However, any material placed in a reactor has some interaction with all the radiations present, absorbing energy from them in scattering processes or nuclear reactions.

In any investigation of the effects of radiation on matter, it is necessary to know the spectrum of the radiation used. This information should be available in the form of the variation of photon or particle flux density with photon or particle energy, that is, we should know $\phi(E)dE$. The amount of change observed in an irradiated material will always be the product of the radiation flux density and an interaction function, integrated over the radiation spectrum. The nature of this interaction function will vary with the type of study being performed.

The radiation spectra in reactors are complex in form and cover wide ranges of photon and particle energies. Given sufficient information about the fission reaction, and about the geometry and materials of construction of a reactor, it is possible, in principle, to calculate the spectra of the reactor radiations. It is then possible, in principle, to calculate the rates of production of change in irradiated materials. Such calculations are very complex, but with modern fast computer techniques they are feasible. There have been some calculations of spectra and some of changes in materials.

The problem of the experimenter studying changes produced in reactor irradiations is to make measurements of the radiation treatment which are adequate for three purposes,

- (a) to correlate experiments at different irradiation positions and in different research reactors;
- (b) to extrapolate the information obtained to projected reactor systems, power production reactors, etc.;
- (c) to compare the results obtained with those of other irradiation experiments using isotope and machine sources.

But the changes observed, if expressed in fundamental terms, are integrals of complex multiples. The experimenter generally seeks to introduce some convention, to enable such a function to be expressed as the multiple of two quantities, a radiation term, and an interaction term. Experiments are then correlated by measuring the radiation term. However, since reactor radiation spectra cannot be conveniently measured at flux densities required for experiment, it is often necessary to adopt a convention which gives a

radiation term which is readily measurable, rather than one which is scientifically desirable.

In this paper the conventions used in radiation chemistry are discussed. This is followed by a discussion of the various types of calorimeters which can be used for dosimetry, with some remarks about their limitations.

The reasons for choosing an isothermal-type calorimeter for the work at the Atomic Energy Research Establishment (AERE) are given, followed by a description of the construction, characteristics, and calibration of the calorimeters in use. It is shown that under the proper conditions these calorimeters measure the ideal absorbed dose-rate, or kerma rate. By the use of three calorimeters, as described, containing respectively graphite, anthracene, and an empty can, measurements were made of the dose-rates in graphite and in anthracene, and in the reactor BEPO at AERE. The problem of correlating the calorimetric experiment with the chemical experiment is discussed.

Ionization chambers are also being used at AERE and a description of their construction, characteristics, and calibration is given. The ionization chambers described measure the ideal absorbed dose-rates in graphite. A comparison is made of calorimeters and ionization chambers, and it is concluded that both probably have a place in routine dosimetry. By inter-comparison of the data obtained in the two ways something is learned about the loss of secondary radiation across a boundary between two materials.

DOSIMETRY AND FLUX MEASUREMENTS IN THE NRX REACTOR, CHALK RIVER

SUMMARY

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The NRX is a heavy-water-moderated reactor with a power of 42 MW using enriched and natural uranium. The core is 2.66 m in diameter and 3.05 m high and is surrounded by a graphite reflector. Energy deposition rates in carbon and hydrocarbons and thermal- and fast-neutron fluxes have been measured in the core and the reflector and inside annular uranium rods.

Three methods of dosimetry were used - calorimeter, graphite-CO₂ ion chambers and the hydrogen yield from cyclohexane radiolysis.

The adiabatic calorimeters are shown in Fig. 1. The energy deposition rate in the sample is calculated from the rates of rise of sample and jacket temperatures. At dose-rates from 50-500 mW/g the accuracy is estimated to be within $\pm 5\%$.

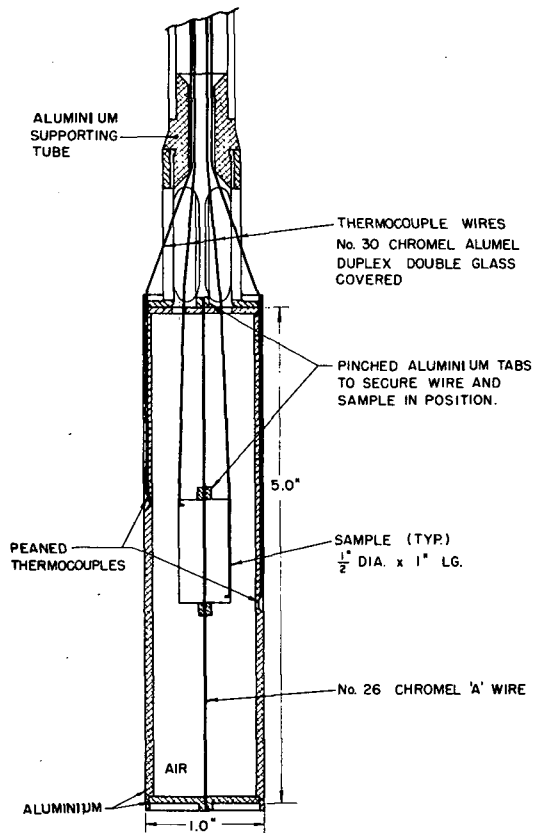


Fig. 1

Adiabatic calorimeter

The ion chamber, shown in Fig. 2, can be used to measure dose-rates from 0.1 to 500 mW/g. The applied voltage is 900-1300 V and the current is measured with an electrometer.

Spectrographic grade cyclohexane was used, and after removal of dissolved gases, was sealed in quartz capsules. Work done elsewhere has shown the hydrogen yield to be the same within 5% for fast neutrons and gamma rays. The dependence on dose and dose-rate is shown in Fig. 3.

Values from these three methods agree within $\pm 5\%$.

The thermal-neutron flux was determined using cobalt (assuming a cross-section of 37 b for 0.0013 cm diam. cobalt wire). The fast-neutron flux was determined by assuming a cross-section of 90 mb for the reaction $\text{Ni}^{58}(n, p)\text{Co}^{58}$.

The values obtained for energy deposition rates and neutron fluxes in various regions of NRX are shown in Table I. The vertical distribution of gamma rays and neutron fluxes in the core is shown in Fig. 4.

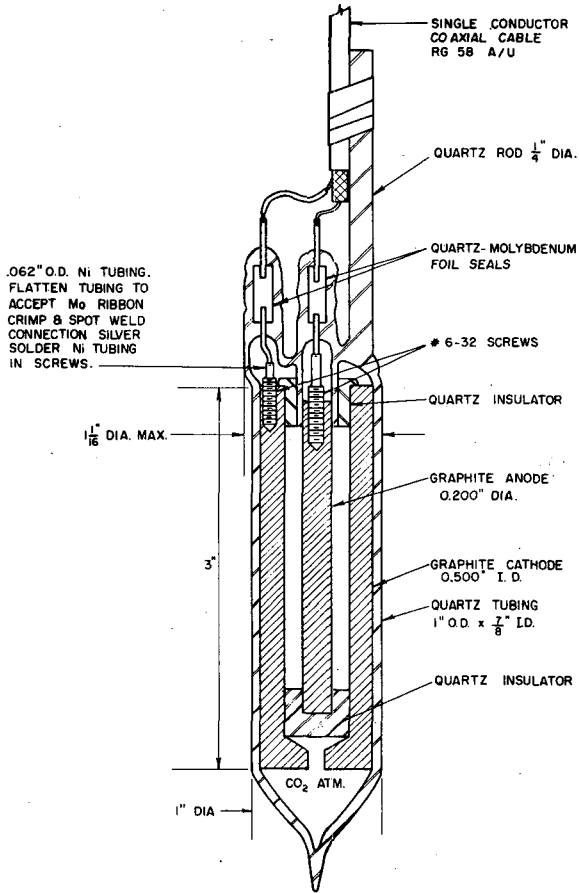


Fig. 2

Ion chamber

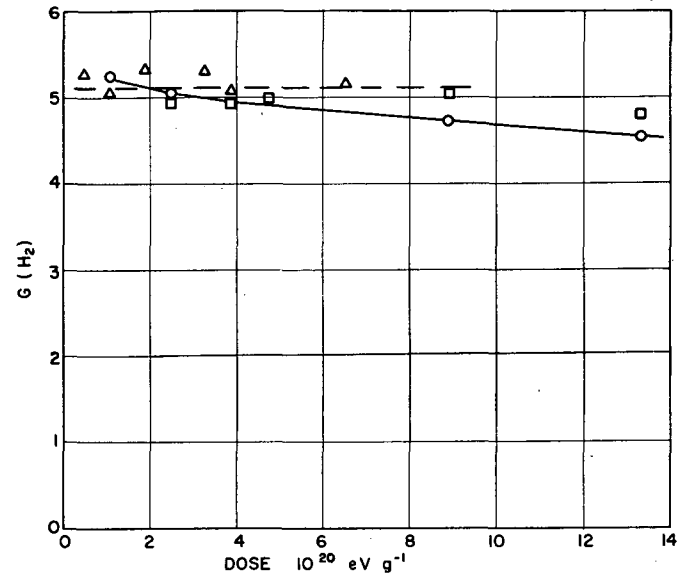


Fig. 3

$G(\text{H}_2)$ dependence upon dose for cyclohexane irradiations

- Dose rate 1 m Wg^{-1} Co^{60} γ -rays
- △ Dose rate 0.2 Wg^{-1} } $\sim 70\%$ fission neutrons
- Dose rate 2 Wg^{-1} } 30% reactor γ -rays

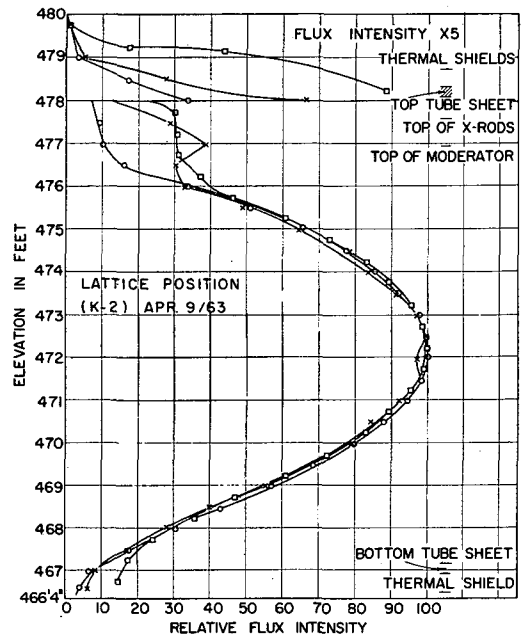


Fig. 4

Flux distribution in cone

TABLE I

SUMMARY OF ENERGY DEPOSITION RATES AND FLUXES IN NRX

Position	Typical values		Deposition in C_6H_{12} mW/g		Ratio Fast flux: Thermal flux	Ratio Fast neutrons in C_6H_{12} : Fast flux mW/g 10^{12}
	Thermal neutrons 10^{13} n/cm ² .s	Fast neutrons 10^{12} n/cm ² .s	Gamma rays	Fast neutrons		
Empty lattice	4.0	0.80	111	39	0.02	49
Reflector annulus	1.0	0.05	18	4	0.005	80
Nat. U Annulus in horizontal hole	0.47	2.6	70	130	0.55	50
Enriched U Annulus in horizontal hole	0.63	9.1	210	540	2.3	59
U Annulus in lattice	8.1	26	600	1200	0.32	46

IN-PILE DOSIMETRY MEASUREMENTS IN THE HUNGARIAN WWR-S REACTOR.

SUMMARY

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In the Central Research Institute for Physics irradiation experiments on organic liquids have been in progress for some time. Both static and loop experiments are carried out in the WWR-S reactor. (Core: light-water-moderated, 10% enriched uranium fuelled; power: 2-2.5 MW). Evaluation of radiation damage required development of in-pile dosimetry.

A calorimetric technique was chosen as the basic method, since it permits the direct determination of the radiation energy absorbed in the specimen. Because of the small G-values of the materials involved in our experiments the heat energy determined by the calorimeter may be considered as the total energy absorbed by the sample.

The calorimeters used in the measurements are very thin-walled (0.2 mm) 20 mm diam., spherical quartz flasks filled with the organic liquid to be analysed, in the present case with a mixture of 28% diphenyl and 72% diphenyl methane, called DDM. The copper-constantan thermocouple is located in the centre of the sphere. The flask, which is suspended at its neck, is surrounded by two thin-walled aluminium mantles separated from each other and from the flask by pressboard spacers. The temperature is recorded by a recording galvanometer.

Calorimeters of similar design but with lead filling are used for determining the gamma contribution to dose-rate.

According to control measurements performed in a thermostat for a temperature difference of 30°C, the DDM and the lead calorimeters can be considered as adiabatic for 35 and 15 s, respectively.

For the correct evaluation of loop experiments it was necessary to determine the spatial dose-rate distribution along the loop channel. During the calorimetric measurements the actual loop conditions were simulated.

Measurements with a DDM calorimeter show the dose-rate distribution to be slightly asymmetric; the maximum occurs a little below the horizontal mid-plane of the core. The dose-rate contribution of the reflector region must be taken into account when considering the total dose received by the material circulating in the loop.

In choosing the operating power level during calorimetric measurements, two contradictory aspects must be taken into account. At high power levels the duration of the measurements is strictly limited because of the high energy absorption rate. (At 2.5 MW full power the temperature rise in the sample is more than 20°C/min.). At low power levels, on the other hand, the results of measurements can be strongly disturbed by the "background dose-rate", arising from the radiation field of the shut-down reactor, which depends very much on the operational history of the core. Its level is too low for direct calorimetric measurements, but introduces nevertheless a significant error if neglected in the extrapolation procedure to full power.

In order to minimize the effects of the background dose-rate, two methods for compromise seem available.

The first is to perform the measurement at the highest power level at which the rate of the temperature rise of sample is yet acceptable. In the present case 1 MW was found to be a convenient power level.

In the second method the background is determined at a single point of the channel (e. g. at the mid-plane) by the extrapolation to zero power.

Assuming then the spatial distribution of the background dose-rate to be of the same shape as that of the dose-rate for the reactor on power, the necessary corrections can be made. The results given by this method agree rather well with those of the first method.

The fast flux distribution was measured with S^{32} pellets. The effects of neutrons having energies less than 1 MeV cannot be neglected. By calculation of the neutron component of the total dose, assuming that the slowing down part of the spectrum merges into the fission spectrum at 100 keV, the maximum value of the neutron dose-rate is obtained as 88 Mrad/h, i. e. 27% of the total. By assuming the spectra to be joined at 1 MeV, the fast neutron part of the dose-rate is found to be 97 Mrad/h.

Our lead calorimeters cannot be considered adiabatic; therefore they are not suitable for absolute dose value determination. Measurements were performed with four lead calorimeters of different diameters (5, 10, 15, 20 mm, respectively). Ambient temperature was kept constant. The results show that the diameter has a strong influence on absorbed energy, i. e. self-screening and build-up cannot be neglected, even in the case of small specimens.

TABLE I
THE G_{gas} VALUES

G_{gas}	Reference	Reactor
0.07 - 0.08	FISCHER, E. <u>et al.</u> , Atomkernenergie <u>11</u> (1962) 405	Reaktorstation Geesthacht
0.112	BURNS, W. G., VI, Rassegna internazionale elettronica e nucleare (giugno 1959)	Reactor BEPO
0.19	BURNS, W. G. <u>et al.</u> , "The effect of fast electrons and fast neutrons on polyphenyls at high temperatures," 1st UN Int. Conf. PUAE <u>29</u> (1958) 266.	Reactor BEPO
0.041	This paper	Reactor WWR-S

The irradiations of diphenyl were carried out in a vertical channel of the reactor. The radiolytic gases were separated, collected, and measured. The doses absorbed during the irradiations were in the range from $5 \cdot 2 \times 10^8$ to $8 \cdot 4 \times 10^9$ rad.

Initially, the gas production was proportional to the absorbed dose, while upon absorption of a dose of approximately 1×10^9 rad it began to decrease tending towards a limit. For calculating the G_{gas} value, only the first part of the curve was used. The G_{gas} values found by different authors are listed in Table I.

A CALORIMETER FOR DETERMINATION OF HEATING OF MATERIALS IN THE IRRADIATION TUBES OF THE REACTOR FR 2

SUMMARY

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For technical and economic reasons it is desirable to know the rate of absorption of energy by different materials in different places in a reactor. It is often hard to calculate this rate because data on the flux and spectra of the radiation are not sufficiently well known. However, the rate can be measured with a calorimeter. By choosing different absorbing materials, some success can be achieved in determining the energy contribution of the different kinds of radiation. Thus, for example, bismuth and lead have high absorption coefficients for γ -rays, the scattering of fast neutrons can be ignored and their activation cross-sections are low, so they are suitable for measuring the energy contribution of the γ -rays. On the other hand, for neutron dosimetry the lighter elements, such as beryllium or hydrogen, must be used. In this case, however, the effect of γ -rays cannot be ignored, which makes interpretation of the results more difficult.

For the measurements of the heating of materials in the irradiation tubes of the FR 2 reactor an isothermal type of calorimeter was chosen. The mechanical construction of the calorimeter is shown in Fig. 1. The calorimeter foot (5) and the inner (2) and outer (1) cans are made from aluminium (AlMg_3). The rod holder (4) is screwed into the calorimeter foot, into which three small Al_2O_3 tubes (8), 50 mm in length, are cemented. The calorimeter materials, as round rods 7 mm in diameter and 70 mm long, with an axial hole (3), are pressed onto the free ends of these tubes and secured with a screw. The hole accepts the heating element used for calibra-

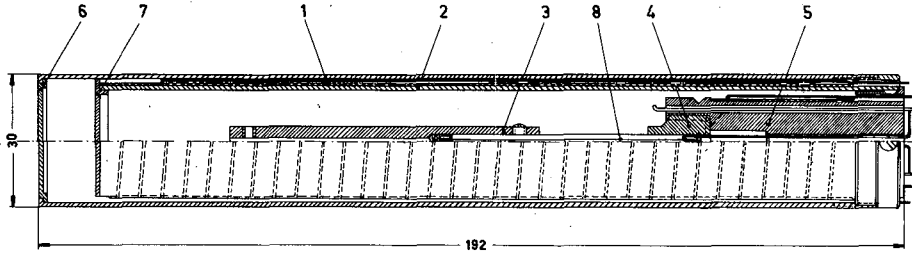


Fig.1

Mechanical construction of calorimeter

tions. Bismuth, iron, aluminium, and graphite are used as calorimeter materials. The design is such that the rods can be changed easily. The temperatures of the aluminium can, and the calorimeter foot, are regulated independently by electrical heating and forced draft cooling, and can be held at $50 \pm 0.02^\circ\text{C}$.

The calibration of the calorimeter was achieved by heating the rod electrically, turning off the current, and observing the exponential cooling rate from which the cooling time constant could be determined. The aluminium can must be kept at the same temperature during calibration as during measurements in the reactor. Temperatures were measured with iron-constantan thermocouples.

Measurements were made in some of the channels of the FR 2 using this calorimeter. The measured energy absorption rates at the core centre are between approximately 9 and 12 mW/gMW. This corresponds to dose-rates of some 3 to 4 Mrad/hMW.

The relative error due to uncertainties in the measured quantities is estimated at about 8%. However, this does not take account of changes in the thermocouple output because of high neutron dose, or heating of the thermocouple in the rod by the reactor radiation. However, because of the way the calorimeter was constructed and because of the short time of irradiation at relatively low reactor power, these sources of error are not believed to be very large. The total error is estimated at $\pm 15\%$.

ABSORBED DOSE MEASUREMENTS OF MIXED PILE RADIATION IN AQUEOUS RADIATION CHEMISTRY

SUMMARY

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To use a nuclear reactor as a radiation source in the radiation chemistry of water and aqueous solutions, reliable routine dosimetry techniques are of basic importance. For this purpose we have tried to develop a calorimetric device and a chemical system.

The differential calorimeter described here permits simultaneous measurements of energy absorption in different materials. From these values the relative contributions from gammas and non-thermalized neutrons to the total absorbed dose can be calculated. The possibility of inserting a liquid sample into the calorimeter makes it very convenient for radiation chemical studies of aqueous solutions or, generally, liquid systems. For a period of about two years, reliable values for the absorbed doses in different materials have been obtained, which are in good agreement with other physical measurements in the RA research reactor at Vinča.

The chemical system described is an aqueous solution of oxalic acid. Its advantages are: the possibility of measurements in the multi-megarad region and negligible induced radioactivity. The G-value is not sensitive to dose-rates even up to 10^{10} rad/s (for low LET radiation), or to temperature increase during irradiation (up to 80°C). Its disadvantages are mainly the same as for other aqueous systems: LET dependence of the G-value, fast heating due to high dose-rates; a considerable gas production (H_2 and CO_2), and the necessity of calculating the absorbed dose from concentration differences before and after irradiation.

The results of calorimetric and chemical measurements are presented in Table I.

Calorimetric data have been obtained by the least-square method from 25 energy absorption measurements in light and heavy water, graphite and polystyrene. All measurements have been performed in five series within a rather long period of about two years. In some cases the energy absorption was measured in 600 mM solutions of oxalic acid, but no difference was found on comparison with the energy absorption measurement in pure water as the calorimetric body. The mean values for oxalic acid decomposition presented were also obtained from a series of irradiations performed simultaneously with calorimetric measurements or under identical irradiation conditions.

Gamma and neutron contributions to the total absorbed dose have been calculated from calorimetric measurements as described in the literature.

The standard deviations are $\pm 10\%$ and $\pm 4\%$ for the calorimetric and chemical results respectively.

Using data from Table I, we obtained the G-values for oxalic acid decomposition as presented in Table II.

TABLE I
RESULTS OF CALORIMETRIC AND CHEMICAL MEASUREMENTS

Oxalic acid solution in	Energy absorbed (eV/g MWh)	Contributions from (%)		Oxalic acid molecules decomposed (mol./g MWh)
		gamma	neutron	
H ₂ O	1.38×10^{21}	67	33	6.08×10^{19}
D ₂ O	1.03×10^{21}	80	20	3.80×10^{19}

TABLE II
G-VALUES FOR OXALIC ACID DECOMPOSITION

Oxalic acid solution in	G _{gamma}	G _{pile} * (total)	G _{pile} (deuteron)	G _{pile} (proton)
H ₂ O	4.91	4.39	-	3.3
D ₂ O	3.9	3.67	2.8	-

* For the absorbed doses composed of 67% from gammas and 33% from neutrons in light water, and 80% from gammas and 20% from neutrons in heavy water.

It seems that the dependence of the radiation yield on the composition of the reactor radiation is rather weak, which is certainly attractive for the routine use of the oxalic acid aqueous dosimeter.

The G-values for gammas in light water solution have been determined calorimetrically, earlier, with an accuracy of about $\pm 1\%$. The corresponding G-value in heavy water solution is the result of a small number of irradiations and has to be confirmed and determined more precisely.

The G-values for decomposition induced by pile protons and deuterons (resulting from elastic scattering of non-thermalized neutrons) have been deduced from G_{gamma} and $G_{\text{total}}^{\text{pile}}$, and known gamma and neutron fractions in the total absorbed dose.

Further experiments are in progress in order to obtain more information on the low-energy particle component in mixed pile radiation.

The results shown in Table II show that the oxalic acid solutions in H₂O and D₂O enable one to assess the dose-rate from the gamma and the fast-neutron components of mixed pile radiations in H₂O and D₂O, and hence by calculation in other materials of low Z number. Further work is going on

in order to develop the procedure for routine use of this system in chemical dosimetry, as well as to get some more basic information on its radiation chemical behaviour.

NEUTRON DOSIMETRY IN BIOLOGY¹

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Neutrons are becoming increasingly important for the irradiation of biological material in the fields of fundamental radiobiology and for the more practical purpose of inducing mutations in crop plants with subsequent selection of beneficial mutants for the establishment of improved varieties. More knowledge is needed of the effects of neutron irradiation on biological material, not only for the latter fields but also for the more accurate evaluation of radiation hazards, improved applications in radiotherapy and for space flights.

Before meaningful assessments can be made on a world-wide scale of the effects of reactor-generated neutrons on living material, it is essential that reliable and universally standardized dosimetry be established.

The accurate measurement of reactor-generated neutron radiation absorbed by exposed biological material is a complex and difficult problem. It is affected by: (1) the energy spectrum and intensity of the neutrons; (2) the angular distribution of neutrons over the whole surface of the irradiated object; (3) the additional unwanted radiation accompanying the neutrons; and (4) the physical state and chemical composition of the irradiated material. Added to these complexities are differences in reactors and the varying output from a single reactor during different periods in the fuel element cycle. It seems clear that, in addition to conventional methods of physical and chemical measurement, the use of a biological dosimeter system will be required. Biological dosimetry would necessarily be carried out with single-source seed of one or two crop species and tested under uniform post-irradiation conditions for growth.

If an international project were to be organized by the Agency the objectives of the dosimetry phase would be primarily to determine accurately the dose received by exposed biological material and to select a group of reactors on the basis of their neutron-dose potential. Additional benefits to be derived from such a project are: (1) to make biologists aware of the requirements for accurate physical data in order to interpret biological results; (2) to acquaint reactor physicists with dosimetry problems peculiar

¹ Assistance was rendered by Professor H. Glubrecht of the Institut für Strahlenbiologie of the Technische Hochschule, Hanover, Federal Republic of Germany, and Dr. G. Ahnstrom of The Institute of Radiobiology of the University of Stockholm.

to experimentation with biological material; (3) to determine reactor characteristics best suited for biological experimentation; and (4) to increase the number of reactors carefully tested and approved on the grounds of dosimetry standards agreed as being acceptable for accurate biological research.

To study adequately the biological effects of different energy neutrons it is necessary to have high-intensity sources which are not contaminated by other radiations, the most serious of which are gamma rays. An effective dosimetry must provide an accurate measure of the absorbed dose, in biological materials, of each type of radiation at any reactor facility involved in radiobiological research. A standardized biological dosimetry, in addition to physical and chemical methods, may be desirable.

The ideal data needed to achieve a fully documented dosimetry has been compiled by Dr. H. Glubrecht (see footnote¹) and is set out below:

- (1) Energy spectrum and intensity of neutrons.
- (2) Angular distribution of neutrons on the whole surface of the irradiated object.
- (3) Additional undesired radiation accompanying the neutrons.
- (4) Physical state and chemical composition of the irradiated object.

It is not sufficient to note only an integral dose value (e. g. in "rad") as the biological effect depends on the above data.

Since all these data will not be available in many cases, it is recommended to note those listed below, which should also be presented for comparison and control even if the ideal data can be given.

SUBSTITUTE PRACTICAL DATA

I. Type of source

For a sufficient definition the following will be needed:

- (a) Reactor
 1. Configuration and materials
 2. Geometrical dimensions of core
 3. Power or mean flux
- (b) Accelerator
 1. Neutron generating reaction
 2. Geometrical dimensions and chemical composition of the target
 3. Energy, intensity and geometry of the ion beam in the accelerator
- (c) Sources with radionuclides
 1. Radionuclide and neutron generating reaction
 2. Present activity of the radioactive source
 3. Geometrical dimensions and structure of the source

II. Geometry of irradiation facility

1. Distance from the source
2. Angular position of the object with respect to the source

3. Absorbing and scattering media between source and object
4. Arrangements and type of materials surrounding the object

III. Irradiated object

1. Geometry of the object itself
2. Isotopic composition of the object with special respect to all nuclides relevant for nuclear reaction with neutrons (e.g. B¹⁰, H¹, N¹⁴).

IV. Duration of irradiation (dose-rate)

If such dosimetry could be established, used in all biological neutron research, and reports published of biological experiments, a significant step would be taken in the integration and advance of knowledge in this field.

A main interest of the Agency is to foster international recognition of the importance of an accurate dosimetry and the world-wide adoption at reactor centres of recommended principles and practice of this dosimetry.

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VIENNA, 1965**

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