

International Atomic Energy Agency

INDC(NDS)-56/U

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**INDC**

**INTERNATIONAL NUCLEAR DATA COMMITTEE**

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PROCEEDINGS OF A CONSULTANTS' MEETING

ON

NUCLEAR DATA FOR REACTOR NEUTRON DOSIMETRY

Vienna, 10-12 September 1973

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**IAEA NUCLEAR DATA SECTION, KÄRNTNER RING 11, A-1010 VIENNA**

PROCEEDINGS OF A CONSULTANTS' MEETING

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<u>Table of Contents</u>	(page)
Introduction	iv
1. Need for Internationally Recommended Cross Sections for Neutron Dosimetry Reactions (U. Farinelli)	1
2. Remarks on Fluence Determination by Activation Methods (W.L. Zijp)	9
3. Fast Reactor Flux-Spectral Characterization (W.N. McElroy)	15
4. Remarks on Response Ranges of Activation Detectors (W.L. Zijp)	37
5. Critical Comparison of Spectrum Unfolding Codes (R. Dierckx)	55
6. Role of Standard Neutron Spectra in Differential Flux Determination (R. Dierckx)	79
7. Methods for Detection of Radiation Monitors Irradiated by Neutrons (K.H. Czock)	89
8. Important Nuclear Reactions and Nuclear Quantities Required (W.L. Zijp)	103
9. Remarks Concerning Cross Sections For Threshold Detectors (H. Liskien and A. Paulsen)	111
10. Conclusions and Recommendations of the Meeting	119
Appendix A Agenda	139
Appendix B List of Participants	141

## INTRODUCTION

In 1971, the International Working Group on Reactor Radiation Measurements stressed the importance of activation dosimetry techniques for the development of fast breeder reactors, particularly for radiation damage studies. This group recommended that "the IAEA take appropriate steps in order to arrive at internationally acceptable reference values for those neutron reactions of primary importance for radiation effects investigations in nuclear reactors".

At the request of this group, the IAEA's Nuclear Data Section undertook a survey of the most important nuclear data required for the use of this technique. The results of this survey are published in INDC(NDS)-47/L (parts I and II). In addition the feeling was strongly expressed by the Euratom Working Group on Reactor Dosimetry, that nuclear data uncertainties cause the largest errors in current dosimetry methods. In response to the expressed need, the IAEA convened a consultants' meeting on this subject on 10 - 12 September 1973. The goal of this meeting is well stated in the introductory paper by Professor Ugo Farinelli. His paper discusses the needs for neutron activation cross sections as well as applications for which activation cross sections are needed and the sensitivity of the final quantities to cross section uncertainties.

This publication contains in unedited form all papers presented at this consultants' meeting\* and the conclusions and recommendations of the experts.

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\* Report by M. Vlasov: "Status of neutron cross section data for some reactions of interest for reactor radiation measurements", published separately as INDC(NDS)-47/L, part II.

NEED FOR INTERNATIONALLY RECOMMENDED CROSS SECTIONS FOR  
NEUTRON DOSIMETRY REACTIONS

Ugo Farinelli

1. Bases for nuclear data requirements - The problems posed by the use of activation detectors for reactor dosimetry and in particular for the determination of neutron spectra are very exacting. In particular the requirements in terms of differential cross sections for detector materials call for a very large programme of new measurements and for a relevant evaluation effort to overcome discrepancies. Quoting Dr. Story, chairman of EANDC, "in real terms, despite great efforts over many years, the fast neutron fission cross-sections are only now reaching the  $\pm 5\%$  level;...we must conclude that if accurate absolute threshold cross-section data are required, to  $\pm 5\%$  or better, it will require a substantial and well-duplicated programme of work". Although there is now general acceptance of the importance of reactor dosimetry and in particular of activation detectors in nuclear technology, it is generally felt that a careful investigation of the needs for detector cross sections in terms of the sensitivity of the quantities of interest to these cross sections is required. Priorities, accuracies and energy resolutions must be well founded and justified in a general context (even if an accurate cost-benefit analysis is not at hand). In other words we must think hard and make a jolly good case of our requests if we want to see results.

I have only mentioned detector cross sections; of course there are other nuclear data of interest in reactor dosimetry (half-lives, decay schemes...) which are also of primary importance. There are in addition other important sources of concern in the area of reactor dosimetry, like unfolding methods, detector technology and material procurement, error correlations and confidence assessment of the results, which are mentioned here only to stress the need for a careful analysis of the actual necessities in this field and for a maximum coordination of the various national and international efforts in order to reduce dispersion.

In the following I shall try to outline an answer to this problem of identifying our necessities in detector cross sections: first by listing the kind of problems for which activation detectors are needed and trying to assess the accuracy with which the final answer should be known; second, by considering the procedure by which these results are obtained from the activation data; finally, by trying to deduce sensitivities of the final quantities to the cross section uncertainties and to discuss the type of information on the cross sections that is most needed. This attempt is very preliminar and should

be regarded mostly as the proposal of a methodology rather than of definite results; in this spirit, I shall not discuss how far is the present information on detector cross sections from the required accuracies.

2. Use of activation measurements - Activation measurements are used in irradiation experiments (in the actual research reactor or on a mock-up); in monitoring irradiation of power reactor components; in reactor physics and in shielding studies.

I will use the following notations:

"Activation rate":  $\int \sigma(E)\phi(E)dE$  refers to a detector used in the measurements";

"Response function",  $F(E)$ , is the contribution per unit flux per unit energy and time to the production of a macroscopic effect (examples: a damage cross section; a relative biological effectiveness or quality factor, etc.)

"Integral response",  $I = \int F(E)\phi(E)dE$ , is the average of the response function over a given spectrum.

The following list of typical applications is certainly not complete, but is meant to be a representative sample from which general schemes and some conclusions may be drawn:

a) Integration of neutron fluxes especially if the intensity is variable with time. Purpose: monitoring of irradiation experiments in experimental reactors or of damage of structural components in a power reactor. No major problem for thermal neutron fluxes. In a thermal reactor, if the spectrum is constant, thermal neutron fluence can also be used to monitor fast fluence if the spectrum is measured or calculated at one time. In the case of radiation damage, if the thermal-to-fast neutron flux ratio is not constant, but the fast spectrum can be considered as fairly constant, fast fluence measurement by a fast neutron integrator and knowledge of the spectrum at one time can be sufficient (thermal neutron fluence is generally also required for corrections). Similar considerations apply to fast reactors, when the spectrum is constant. Generally, there are no practical alternatives to the use of activation detectors, except in some cases the use of well calibrated damage detectors.

b) Measurement of neutron spectra or directly of damage cross sections averaged over spectrum at one time (integral damage response). Purpose: together with the fluence measurements described in a), allow intercomparison of irradiation experiments or evaluation of damage. Very often it is not feasible to use other than activation detectors for this purpose, because of dimensions, presence of high flux gradients, perturbation introduced by other detectors, environment (temperature, coolant...), accessibility etc. As we shall see, it is important to assess how well the spectrum can be calculated (at least a rough theoretical evaluation is always necessary); in many

cases, calculations are unreliable because of extremely complex 3D geometry, transport effects and in some cases insufficient knowledge of the environment or the physical situation.

c) Measurement of time-integrated (or averaged) neutron spectra when they are variable in a fast or unpredictable way, or of the corresponding integral response. Caution should be applied in the interpretation of the results in terms of an "average spectrum" for damage determination.

d) Measurement of the neutron spectrum in which an integral response is measured. Purpose: study  $F(E)$  (the energy dependence of a response function) by a series of integral experiments in different neutron spectra.

e) Measurement of reaction rates of interest for reactor physics. Purpose: in most cases, comparison with calculations; in a few cases, direct determination of reactor parameters of interest (for instance a breeding ratio). Spectrum determinations are generally not necessary; the reaction rates (or their ratios) are either the final purpose of the measurement or are compared with the corresponding calculated quantities.

f) Spectrum measurement in core (generally in a critical facility). Purpose: comparison with calculated spectra; or to have an experimental intermediate step in the calculation of, say, Döppler effects or in the correction of cross sections by a correlation procedure. In general, activation techniques are insufficient for the purpose, and at most complementary to more refined techniques.

g) Measurements on a mock-up for the evaluation of damage functions, of neutron-heating rates (seldom) or of activation rates. Purpose: support to reactor design. Problems are similar to those met in b) or in e).

h) Application to shielding problems. Purpose: either comparison with calculations, or determination of a biological dose (or of damage or activation or heating rates) at the boundary or inside a reactor shield. The measurement can be carried out on a mock-up or in some cases on the actual reactor. More refined or more direct techniques are often applicable, but in some cases only activation detectors can be used.

j) Intercomparison of spectra, detectors, cross sections etc. This point should not be considered as a purpose in itself, but as instrumental to the others listed above. It should be noted the determination of the activation cross section of a nuclide by integral experiments is a particular determination of a response function as considered in point d).

3. Classification of problems - Summarizing, and leaving aside the problem of time-integration vs. instantaneous measurements, the problems in which activation measurements are used can be subdivided, for the purpose of sensitivity studies, into three categories:

i) Measurement of a reaction rate which is of direct interest (for instance, activation rate for a given reactor material) or which is compared with the corresponding calculated value.

ii) Determination of an integral function of the spectrum which is not directly measurable, the integral response  $\int F(E)\phi(E)dE$ .  $F(E)$  is supposed to be known.

iii) Determination of the neutron spectrum.

Of course, once the spectrum has been determined, there is no problem in evaluating its integral functions. However, it may be possible to calculate the integral quantities ii) from the activation of detectors without having to pass through the spectrum. This has been shown in particular cases to be feasible and useful, although it is not a commonly used procedure up to now.

At this point, I shall state some working hypotheses, based on the considerations made up to now, which I consider to be questionable and controversial, but which are essential, in this or some other form, to arrive to some conclusions as to requirements, sensitivities and target accuracies:

- When the object of the experiment is a directly measurable activation rate, the situation is straightforward and no particular difficulties arise (see in the following, point 5.)
- As far as activation detectors are concerned, the evaluation of a neutron spectrum is not interesting in itself (see e)) but only as a step in calculating some integral quantities.
- A possible exception to this statement is the determination of a response function  $F(E)$  (or a cross section) by a set of integral experiments as considered in points d) and j). In this case a detailed knowledge of the spectrum seems necessary in order to arrive to a satisfactory definition of  $F(E)$ . My assumption here will be that activation measurements are insufficient for such a knowledge of  $\phi(E)$  and that such experiments must be carried out in a simplified condition in which either reliable calculations or differential measurement of the spectrum is possible. An alternative to this approach will be mentioned later.
- In all cases, use is made of some a priori information on the system, such as a neutron spectrum calculated in simplified conditions.

4. Target accuracies - In order to assess the accuracies required for the detector cross sections, it is necessary to set target accuracies for the final results of our measurements. Except for the case in which one is investigating a response function itself, target accuracies will concern integral quantities only. The single most important observation to make here is that we have supposed the energy weighting function,  $F(E)$ , to be perfectly known. Although this is certainly not true in a physical sense (it is most likely that the actual uncertainty in the damage

evaluation of a certain steel will derive much more from the lack of information on  $F(E)$  than on insufficient information on the neutron spectrum or fluence!) it is generally a correct assumption from the point of view of the reactor designer. For instance, the life of a reactor component which is approved by a safety analysis is based on a specified damage function and an assigned safety margin (no matter if the one and the other will undergo large changes in the near future) and all the uncertainty in the evaluation of the corresponding damage rate is then attributed to the neutronic evaluation. It was thus considered worthwhile to make a mock-up experiment for the determination of the neutron flux and spectrum on the support grid-plate of the FFTR to reduce an assumed uncertainty of 20% although it is quite clear that the predictions of the actual life of this component are affected by a much larger uncertainty due to our lack of experience on the properties of stainless steel irradiated in a fast flux. In a similar way, the designer must ensure that the dose level in any unrestricted area of a reactor plant is below, say, 2.5 mrem/hr, with no tolerance, even if the precise effect of such a dose is affected by a large uncertainty.

Such considerations, if they make the work of the reactor dosimetrist more demanding, make the assessment of required accuracies easier. It seems justified to assume that the correct order of magnitude for the required accuracy of any of the integral quantities of design or operational interest that have to be determined through activation measurements is between 10% and 20%, in accordance with similar specifications for other design parameters. Such a thumb-rule has to be improved and specified for particular cases, and somebody may want to challenge it, but I think it gives us the guidelines necessary to specify our cross section requirements.

As concerns the experiments aimed to the determination of the response functions themselves, we have assumed that in this case the neutron spectrum is known from other sources.

5. Sensitivities - Let us first consider the simple case of activation rates of direct interest (case i) of point 3). In this case, if the activation rate is the final result desired, there is no need of any knowledge of the corresponding cross section. Since the experiment directly yields the quantity of interest, only a scaling factor between the experimental and the reference conditions is needed.

In case one measures an activation rate of interest (always case i) of point 3) but with the purpose of comparing it with calculations, then the cross section is needed in order to calculate the reaction rate. In this case, the relation between the uncertainty in the reaction rate and the uncertainty in the cross section is straightforward. Using a group notation,

the uncertainty  $\delta A$  in a computed activation rate due to the uncertainties  $\delta \sigma^i$  of the detector activation cross section in each group  $i$  and to the uncertainty in the calculated flux components  $\delta \phi^i$ , is simply

$$\delta A = \sum_i \phi^i \delta \sigma^i + \sum \sigma^i \delta \phi^i$$

and the sensitivity of  $A$  to  $\sigma$  is given directly by the flux:  $\partial A / \partial \sigma^i = \phi^i$ . In general, such comparisons require a rather high accuracy; however, the detectors involved are in most cases those for which the cross sections are best known for their importance in other fields of reactor design, like U-235, U-238, Pu-239 etc.

When the quantity in which we are interested is an integral response  $I$  which is not directly measurable, the situation is much more complicated, and the evaluation of sensitivities is more cumbersome. The integral response is generally obtained from the activation data in two steps: first the application of unfolding codes to obtain the neutron spectrum, then the integration of this spectrum over the response function.

In the first step, one generally starts from a guess flux,  $\phi_g$  which is in most cases the result of a calculation, and arrives to a final flux  $\phi$  subject to two conditions: that the final flux is as close as possible to the original flux  $\phi_g$ , and that the activation rates calculated using the final  $\phi$  flux for the various detectors used agree with the measured activation rates within the assumed a priori uncertainties. The conditions to be satisfied are summed up, in the most general case, in group notations, by the expression:

$$\sum_i \left( \frac{\phi^i - \phi_g^i}{\Delta \phi_g^i} \right)^2 + \sum_j \left( \frac{A_j - \sum_i \sigma_j^i \phi^i}{\Delta A_j} \right)^2 = \text{minimum}$$

where  $\Delta \phi_g^i$  is the assumed uncertainty in group  $i$  of the calculated flux,  $A_j$  is the measured activation rate for detector  $j$ , and  $\Delta A_j$  is the a priori uncertainty in the difference between measured and calculated reaction rate for the "true" flux, which in turn derives from two terms: the experimental uncertainty in the integral measurement,  $\Delta A_j'$ , and the uncertainties  $\Delta \sigma_j^i$  in the cross sections used in the calculation:

$$\Delta A_j = \Delta A_j' + \sum_i \phi^i \Delta \sigma_j^i$$

It is reasonable to assume that in most cases the second term is predominant with respect to the first. Assuming then (to the first order) that  $A_j$  is obtained by the integration of  $\phi$  over the "true" detector cross section and calling  $\delta \sigma^i$  the difference between the true and the assumed cross section for detector  $j$  in group  $i$ , the minimum condition becomes:

$$\sum_i \left( \frac{\phi_i - \phi_j^i}{\Delta \phi_j^i} \right)^2 + \sum_j \left( \frac{\sum_i \delta \sigma_j^i \phi_i}{\sum_i \Delta \sigma_j^i \phi_i} \right)^2 = \text{minimum}$$

First of all it must be noted that in order to make activation measurements of neutron spectra worthwhile, it is necessary that the second term be smaller than the first one. In other words, such measurements add meaningful information only when it is not possible, or not practicable, to calculate the spectrum with an accuracy comparable with the accuracy one can assume for the detector cross sections.

In the next step, the flux so obtained is integrated over the response function to obtain the integral response. Again, in group notations:

$$I = \sum_i \phi^i F^i$$

The sensitivity of  $I$  to  $\phi^i$  is of course  $dI/d\phi^i = F^i/I$ ; but the sensitivity of  $I$  to the original uncertainties in the cross sections is more difficult to calculate and it may not be of great interest in a general case if it does not lend itself to practical results; it seems more useful to apply these considerations to practical cases of interest, if necessary by repeating calculations with unit changes in the most critical cross sections in order to obtain the sensitivities. It should be noted, however, that much depends on the shape of the response function  $F(E)$ . If this is a smooth function (as practically will happen in most cases for lack of adequate information) then all the oscillations in the flux  $\phi$  will be smeared out, and many problems which are currently met in the unfolding of spectra will be of little importance. In other words, errors in flux determination are by no means paralleled by similar errors in the integral response in which we are finally interested; however, as our knowledge of response functions will improve with time, similar refinements will be reflected in the details required for the neutron spectrum and correspondingly more accurate and more detailed detector cross sections will be required.

6. Energy representation - The detail with which detector cross sections have to be known is at least as important as the determination of their accuracy. If the ancient times of the Hughes representation - effective threshold and constant cross section - are definitely gone, it does not seem productive to go to the other extreme, with a number of energy points is beyond practical applicability and details in wiggles and structure which add nothing to a good average value obtained by a broad resolution measurement. The situation is similar to the one met in reactor calculation, where a reasonable group structure (e.g. 26) plus selfshielding factors allow in most cases accurate enough results; in our case, the effect of scattering resonances should not have in most cases

to be represented in detail, at least for the detector, especially as long as the response function smears out most fluctuations in the flux. However, a different complication arises: many of our detectors have a steep rising cross section just where the fission flux has rapid decrease; so that structure in the cross section near the threshold may be more important than the asymptotic value. This is probably the reason for discrepancies observed between differential and integral measurements for some detector cross sections, like in the case of Cu-63(n,a). As Dr. Hannum mentioned, summing up the results of EACRP's discussion on the subject, "the interpretation of dosimetry samples may be more sensitive to uncertainties in the shape of the threshold step than to current uncertainties in plateau levels. Precision requests in this area may be more sensibly interpreted as requirements for effective cross sections (including shape of the threshold) than as requirements on individual points". In this respect, it seems advisable that a good coordination between differential measurements in selected energy regions and with variable energy resolution and integral measurements in well known and specified neutron spectra should supply the bulk of the information needed for reliable evaluated cross sections. Integral measurements of this type are in progress or planned at the  $\Sigma\Sigma$  facility in Mol, at the National Bureau of Standards and at the TAPIRO reactor at Casaccia.

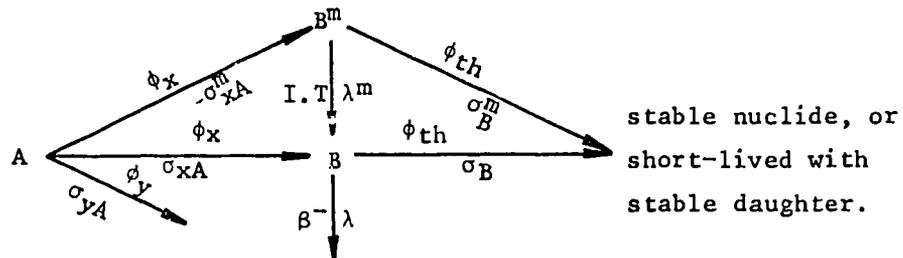
REMARKS ON FLUENCE DETERMINATION  
BY ACTIVATION METHODS

Willem L. Zijp

1. RESPONSE OF AN ACTIVATION DETECTOR.

The reaction rate for the formation of a radionuclide is proportional to the number of target atoms, proportional to the cross section for this reaction, and proportional to the flux density.

Let us consider a general activation reaction of the following type:



For a thermal activation reaction like  $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$  we have  $x = \text{thermal}$  and  $y = \text{fast}$ . For a threshold activation reaction, like  $^{58}\text{Ni}(n,p)^{58}\text{Co}$  we have  $x = \text{fast}$  and  $y = \text{thermal}$ . We will deal with the case that the fluence  $\phi_x$  has to be determined from the activity of nuclide B, after decay of the shorter-lived activity of nuclide  $B^m$ .

One has to deal with effective decay constants:

$$Q = \{(\sigma_{xA} + \sigma_{xA}^m) \cdot \phi_x + \sigma_{yA} \cdot \phi_y\} \quad (1.1)$$

$$R = \lambda_B^m + \sigma_B^m \cdot \phi_{th} \quad (1.2)$$

$$S = \lambda_B + \sigma_B \cdot \phi_{th} \quad (1.3)$$

In case that the flux densities  $\phi_{th}$  and  $\phi_f$  are not constant, but a function of time, while the spectral distribution remains the same, one can proceed as follows:

$$\bar{Q} \cdot t_i = \int_0^{t_i} Q(t) \cdot dt \quad (1.4)$$

$$\bar{R} \cdot t_i = \int_0^{t_i} R(t) \cdot dt \quad (1.5)$$

$$\bar{S} \cdot t_i = \int_0^{t_i} S(t) \cdot dt \quad (1.6)$$

The activity A of an activation detector at the end of the irradiation is given by:

$$A(t_i) = \lambda_B \cdot N_A(0) \cdot \left\{ \sigma_A + \frac{\lambda_B^m}{R} \cdot \sigma_{xA}^m \right\} \cdot e^{-\bar{S}t_i} \int_0^{t_i} e^{(\bar{S}-Q)t} \cdot \phi_x(t) dt \quad (1.7)$$

Here it has been assumed that the irradiation time is long in comparison to the half-life of nuclide  $B^m$  (i.e.  $\lambda_B^m \cdot t_i \gg 1$ , giving  $Rt_i \gg 1$ ). For all reactions of interest one has  $R \gg Q$  and  $R \gg S$ . Also it is assumed that initially there is no activity of B or of  $B^m$  present in the detector.

Introducing the following abbreviations:

$$K = \lambda_B \cdot N_A(0) \left\{ \sigma_{xA} + \frac{\lambda_B^m}{R} \cdot \sigma_{xA}^m \right\} \quad (1.8)$$

and

$$f = \frac{e^{-\bar{S}t_i} \int_0^{t_i} e^{(\bar{S}-\bar{Q})t} \cdot \phi_X(t) \cdot dt}{\int_0^{t_i} \phi_X(t) \cdot dt} \quad (1.9)$$

one has the relation:

$$A = K \cdot \phi_X \cdot t \quad (1.10)$$

or

$$\phi_X = \frac{A}{K} \cdot \frac{1}{f} \quad (1.11)$$

K is a coefficient which is approximately insensitive to variations in flux densities, and may very often be considered as a material constant. The quantity f describes the deviation from linearity in the relation between activity A and fluence  $\phi$ , due to burn-up and decay during irradiation.

If  $\phi(t)$  is constant during the irradiation, and equal to  $\bar{\phi}$ , one obtains:

$$f = \left\{ \frac{e^{-Qt_i} - e^{-St_i}}{(S-Q)t_i} \right\} \quad (1.12)$$

$$\phi = \bar{\phi} \cdot t = \frac{A}{K} \cdot \frac{1}{f} \quad (1.13)$$

## 2. GENERAL CHARACTERISTICS.

In reactor neutron dosimetry one applies activation and fission detectors to determine characteristics of the neutron field: flux density, fluence or spectral distribution.

The response of such an activation or fission detector is related to:

- quantities which characterize the irradiation of the detector in the neutron field;
- quantities which describe the properties of the detector material.

When considering the accuracy of the desired field quantity one has therefore to look at the accuracy of the irradiation parameters, and of the detector properties.

Since the choice of a detector depends also on the expected irradiation conditions, we first list systematically all aspects which influence the selection of an activation detector.

### 2.1. Field characteristics.

1. The flux density to be measured,  $\phi_x$  (e.g.  $\phi_{fast}$ ).
2. The flux density for burn-up of target nuclide by side reactions,  $\phi_y$  (e.g.  $\phi_{thermal}$ ).
3. The flux density for burn-up of product nuclides by secondary reactions ( $\phi_{th}$ ,  $\phi_{epithermal}$ ).
4. The constancy of flux density in time (core changes, operation cycles; control member influences).
5. The constancy of flux density in space (gradients in facilities available for irradiations).
6. The proportionality between flux density  $\phi$  and reactor power  $P$ .
7. Availability of space for samples and detectors.
8. The temperature at the detector position.
9. The chemical reactivity of environment (e.g. Na-K coolant).

### 2.2. Interaction characteristics.

1. Irradiation time (start-up correction).
2. Interaction cross section (see next point).
3. Flux density depression, due to presence of detector.
4. Mutual shielding between two or more detectors.

### 2.3. Detector characteristics.

1. Cross section for the reaction under consideration.
  2. Cross sections for burn-up of target and product nuclides.
  3. Half-life of product nuclide.
  4. Gamma abundance.
  5. Gamma energy.
  6. Self-absorption of radiation emitted by product nuclide.
- } decay scheme.

7. Self-shielding of neutrons inside detector.
8. Purity (minimization of radioactivities produced by side reactions).
9. Melting point of detector (important for high temperature irradiations).
10. Chemical composition of detector.
11. Physical state (metallic, powder, liquid).
12. Dimensions.

#### 2.4. Convenience.

1. Sensitivity of detector (proportional to  $\lambda.N.\sigma/v$ , i.e. proportional to  $\rho.\lambda.\sigma/M$ ).
2. Price.
3. Availability.
4. Ease of fabrication and handling.
5. Availability of counting instruments and spectrometers.

### 3. SOME PROBLEMS IN INTEGRAL ACTIVATION DETECTOR MEASUREMENTS.

#### 3.1. Association of physical quantities.

Sometimes, as in the case of  $^{54}\text{Mn}(n,p)^{54}\text{Fe}$  there is a relation between the observed values for the cross section and the assumed value for the half-life.

In these cases the accuracy in the combination  $(\lambda_x.\sigma)$ , which is of some interest, is not easily derived from the publications.

The response of an activation detector is governed by the product of a cross section and a flux density.

Sometimes "thermal flux density" and "2200 m/s flux density" are incorrectly assumed to be synonymous.

The fast flux density (and also the fast fluence) can be expressed as equivalent fission neutron flux density (fluence) or as flux density (fluence) of neutrons with energies above an energy  $E_L$ . In the latter case one takes often  $E_L = 1 \text{ MeV}$  or  $E_L = 0.1 \text{ MeV}$ .

How can one deal with:  $\langle\sigma\rangle$  and  $\phi(E>E_L)$ ?

First determine the reactor spectrum  $\phi = \phi(E)$ .

Then determine:

$$\langle \sigma \rangle = \frac{E_L \int_0^{\infty} \sigma(E) \cdot \phi_E(E) \cdot dE}{E_L \int_0^{\infty} \phi_E(E) \cdot dE} \quad (3.1)$$

Finally determine absolute magnitude of  $\phi(E > E_L)$ :

$$A_{\text{sat}} = N \cdot \langle \sigma \rangle \cdot \phi(E > E_L) \quad (3.2)$$

### 3.2. The quantity of interest.

People studying radiation damage effects wish to have fluence values above a given limit (e.g. 0.1 MeV or 1 MeV).

The quantity  $\phi^{\text{ef}}$ , the so-called equivalent fission flux density, can rather easily be derived using  $\langle \sigma \rangle^{\text{f}}$ , the cross section averaged over a fission neutron spectrum.

The discrepancy between calculated and measured values of  $\langle \sigma \rangle^{\text{f}}$  is about 10% for the reactions  $^{58}\text{Ni}(n,p)$ ,  $^{46}\text{Ti}(n,p)$ ,  $^{54}\text{Fe}(n,p)$ , but more than 25% for the reaction  $^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$ . One should however realize that the quantity  $\phi^{\text{ef}}$  is difficult to interpret. One should also realize that the quantity  $\phi(E > E_L)$  is not general useful, since the lower limit is different from irradiation experiment to irradiation experiment.

In steel irradiations one prefers another limit than in graphite irradiations. The choice of the limit is very often an arbitrary one.

### 3.3. Constancy of flux density.

During irradiation the flux density may vary (operation schedule, control member movement etc.).

### 3.4. Constancy of spectrum.

How can one determine changes in the spectrum during the irradiation? Is the spectrum in the actual experiment the same as during the mock-up experiment?

### 3.5. Choice of shield in thermal reactors.

The application of a cadmium shield around the detector is not always possible (for reason of space, nuclear heating, etc.). What is experience with application of Gd shield, and with B shields?

### 4. CONCLUSIONS.

Under optimum irradiation conditions the accuracy of the fluence values derived is mainly governed by the nuclear data:

- the cross sections for the main reaction and for the side and secondary reactions;
- the decay scheme data of the product nuclides (half-life, branching ratios).

The normal thermal fluence determinations, e.g. with the reaction  $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$  can be considered as to be accurate enough. Less but acceptable accuracy is obtained in the equivalent fission neutron fluence determinations with  $^{58}\text{Ni}(n,p)^{58}\text{Co}$ ,  $^{46}\text{Ti}(n,p)$  or  $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ , provided that irradiation conditions (burn-up, irradiation time, irradiation temperature) are favourable.

The fluence of neutrons with energies above  $E_L$  can be determined when the average cross section  $\langle\sigma\rangle$  can be derived. This is possible when a knowledge of the spectrum can be combined with energy dependent cross section data.

The accuracy of fast fluences, which are important for radiation damage studies and for fast reactor development studies is therefore dependent on the accuracy of the available  $\sigma(E)$  data. This latter point will however be considered elsewhere.

In fluence determinations one should always report the magnitude of the overall cross section value applied. When this is done adjustment of the final  $\phi$ -value at a later date is still possible.

## FAST REACTOR FLUX-SPECTRAL CHARACTERIZATION\*

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### ABSTRACT

The status and requirements of fast reactor flux-spectral characterization for LMFBR fuels and materials development programs are reviewed. The methods of data reduction, error assignment, and associated nuclear parameters in current use by the Fast Reactor Materials Dosimetry Center at HEDL are discussed. Examples of flux-spectral characterization by the SAND-II multiple foil method are presented for EBR-II dosimetry tests and fuels and materials irradiations.

### I. Introduction

The radiation environment of fast test reactors must be properly characterized for the subsequent correlation and application of the derived irradiation effects data to the development of commercial LMFBR's. Results of controlled high and low power EBR-II "Dosimetry Tests" during reactor runs 31 and 50 have been reported by McElroy et al,<sup>(1)</sup> Dudley et al,<sup>(2)</sup> Sehgal et al,<sup>(3)</sup> and Jackson et al.<sup>(4,5)</sup> This paper presents necessary up-dated state-of-the-art information on the application of the SAND-II multiple foil method of neutron flux-spectral characterization as developed for the USAEC's Fast Reactor Materials Dosimetry Center (FRMDC) at HEDL.<sup>(6)</sup>

In addition to flux-spectral measurements, another important objective of the USAEC's dosimetry program at HEDL is the measurement of reaction rates for the principal fission reactions,  $^{235}\text{U}$ ,  $^{238}\text{U}$ , and  $^{239}\text{Pu}$ , to an accuracy within  $\pm 5\%$  at the 95% confidence level in EBR-II and FTR. Accurate measurement of other fission and non-fission reactions is also required, but to a lesser accuracy, between  $\pm 5$  to 10% at the 95% confidence level. Another objective is improvement in knowledge of other nuclear parameters significant to the measurement of neutron flux, spectra, fluence, and burnup.

In order to help accomplish these objectives, the USAEC has established the Interlaboratory LMFBR Reaction Rate (ILRR) program, which is using a number of different measurement techniques (fission chambers, track etch, radiochemistry, and helium mass spectrometry) in well-established neutron environments of current interest for fast reactor development to establish the required basic technology needed to achieve the stated objectives. Information from EBR-II tests coupled with that from the ILRR program will more clearly define existing capabilities and absolute and relative uncertainties associated with reaction

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\* This paper is based on work performed under United States Atomic Energy Commission Contract AT(45-1)-2170.

rate and flux-spectral determinations. Results of the ILRR program pertinent to the present discussion are briefly reviewed and/or referenced.

Results of SAND-II analysis of multiple foil dosimetry sets being placed routinely in EBR-II fuels and structural materials subassemblies as well as in a few special subassemblies involving measurements for Xe/Kr tag gases and boron carbide control experiments are reported.

## II. Monitor Selection and Reference Data

An optimum set of monitor materials must be selected for proper flux-spectral characterization by the multiple foil method. Details on monitor selection, encapsulation, and placement for fast reactor applications, particularly for EBR-II, are reported by McElroy, et al.(1,6) and Jackson, et al.(4,5) Updated values of nuclear parameters for the more important monitor reactions being used by the FRMDC for EBR-II dosimetry are given in Table I. These values have been up-dated on the basis of ILRR program work which is reported elsewhere.(7-9) The selection of an appropriate set of monitors from those listed in Table I depends on how well a spectrum has been defined by reactor physics calculations and/or specialized dosimetry tests (such as the 31F and 50H tests for EBR-II) and the needs and economics of a particular fuels or materials experiment. More detailed information on the physical form and energy response range for the monitor reactions, Table I, in EBR-II and FTR flux-spectra is presented in references 1, 4, 5, 6, and 11.

Approximately ten monitor reactions are usually selected for most EBR-II fuels and materials dosimetry tests. In some EBR-II tests, however, a single iron gradient monitor [ $^{54}\text{Fe}(n,p)$  and  $^{58}\text{Fe}(n,\gamma)$  reactions] might satisfy the flux-spectral characterization requirements. As demonstrated for the 31F and 50H EBR-II dosimetry tests, when properly combined with other information, the  $^{54}\text{Fe}(n,p)/^{58}\text{Fe}(n,\gamma)$  reaction rate ratio can be used as a sensitive indicator of flux-spectral changes.(1,2,4,5) This also applies for other selected reaction rate ratios, such as  $^{238}\text{U}(n,f)/^{235}\text{U}(n,f)$ .(1)

The dependence of the SAND-II procedure of flux-spectral determination on the number of foil reactions used, input neutron spectrum, and uncertainties in measured reaction rates and evaluated energy dependent foil reaction cross sections has been discussed rather extensively.(1,4,5,6,11-13) Necessary state-of-the-art information on the accuracy of current reaction rate measurements and the status of the USAEC's efforts to develop an ENDF/B evaluated energy dependent cross section file for dosimetry applications is discussed in detail in a special series of papers.(7-10,14)

Here, it is sufficient to indicate that absolute reaction rate uncertainties estimates for EBR-II irradiations are currently in the  $\pm 2$  to  $\pm 7\%$  ( $1\sigma$ ) range for most non-fission reactions. For fission reactions, the range is somewhat higher; at the  $\pm 5$  to  $\pm 10\%$  ( $1\sigma$ ) level, primarily because of fission yield uncertainties. As stated earlier and reported elsewhere, efforts are being made to reduce this uncertainty to the  $\pm 5\%$  ( $2\sigma$ ) level.

At present, evaluated energy-dependent reaction cross section uncertainties are thought to be the largest single source of absolute error for multiple foil derived flux-spectra. On an absolute integral basis, these uncertainties are presently estimated to be at the  $\pm 10\%$  ( $1\sigma$ ) level, although for some reactions like  $^{235}\text{U}(n,f)$ , the value may be considerably less.

Table II lists current up-dated SAND-II evaluated cross section error assignments based on a 15 group energy structure used with the SAND-II Monte Carlo Error Analysis Code.(16) All SAND-II flux-spectral uncertainty estimates given in this and a companion paper(4) are based on these  $\sigma(E)$  error estimates.

TABLE I - Monitor Reactions and Associated Parameters

Fission Reaction	Half-Life of Products <sup>a</sup>				Fission Yields <sup>b</sup>				Required Target <sup>c</sup> Isotope Abundance, %, and Physical Form	
	<sup>95</sup> Zr	<sup>140</sup> Ba	<sup>137</sup> Cs	<sup>148</sup> Nd	<sup>95</sup> Zr	<sup>140</sup> Ba	<sup>137</sup> Cs	<sup>148</sup> Nd		
<sup>239</sup> Pu (n, f) F.P.....	64.6d	12.79d	29.94y	Stable	{4.5-4.8}	{5.1-5.3}	{6.2-6.4}	{1.6-1.7}	>99.0	Metal or Oxide
<sup>235</sup> U (n, f) F.P.....					{6.3-6.5}	{5.8-6.0}	{6.0-6.1}	{1.6-1.7}	>93.0	
<sup>237</sup> Np (n, f) F.P.....					{5.4-5.6}	{5.4-5.7}	{5.9-6.4}	{1.6-1.7}	>99.98	
<sup>238</sup> U (n, f) F.P.....					{5.0-5.5}	{5.9-6.1}	{5.9-6.3}	{2.0-2.3}	>99.995	
Nonfission Reaction	Half-Life of Products				Fission Yields			Target Isotope Abundance, %		
<sup>109</sup> Ag (n, γ) <sup>110m</sup> Ag..	260d				....			48.18	~0.1% alloy	
<sup>181</sup> Ta (n, γ) <sup>182</sup> Ta....	115.1 d				....			99.988	~0.1% alloy	
<sup>197</sup> Au (n, γ) <sup>198</sup> Au....	2.696 d				....			100.0	~0.1% alloy	
<sup>45</sup> Sc (n, γ) <sup>46</sup> Sc.....	83.85				....			100.0	Metal foil or wire	
<sup>59</sup> Co (n, γ) <sup>60</sup> Co.....	5.268 y				....			100.0	~0.1% alloy	
<sup>58</sup> Fe (n, γ) <sup>59</sup> Fe.....	44.6 d				....			0.33	Metal foil or wire	
<sup>54</sup> Fe (n, p) <sup>54</sup> Mn.....	312.6 d				....			5.84	Metal foil or wire	
<sup>63</sup> Cu (n, γ) <sup>64</sup> Cu.....	12.78 h				....			69.09	Metal foil or wire	
<sup>63</sup> Cu (n, α) <sup>60</sup> Co.....	5.268 y				....			69.09	Metal foil or wire	
<sup>58</sup> Ni (n, p) <sup>58</sup> Co.....	71.3 d				....			67.88	Metal foil or wire	
<sup>46</sup> Ti (n, p) <sup>46</sup> Sc.....	83.85				....			7.99	Metal foil or wire	
<sup>47</sup> Ti (n, p) <sup>47</sup> Sc.....	3.39				....			7.32	Metal foil or wire	
<sup>48</sup> Ti (n, p) <sup>48</sup> Sc.....	43.8 h				....			73.99	Metal foil or wire	
<sup>27</sup> Al (n, α) <sup>24</sup> Na.....	14.99 h				....			100.0	Metal foil or wire	
<sup>127</sup> I (n, 2n) <sup>126</sup> I.....	13.2 d				....			100.0	Powder, KID	
<sup>55</sup> Mn (n, 2n) <sup>54</sup> Fe....	312.6 d				....			100.0	Metal pieces.	

<sup>a</sup> h=hours, d=days, y=years. Selected gamma-ray energies and branching intensities and associated error estimates are given elsewhere. (8-10)  
<sup>b</sup> The uncertainty associated with these values of fission yield is discussed elsewhere (6-8). Here, only a range of values is given.  
<sup>c</sup> Fission foils meeting these requirements are available through the Isotope Target Center, Oak Ridge National Laboratory (15).  
<sup>d</sup> Encapsulated in vanadium at ORNL, see references (4-6).

TABLE II  
SAND-II EVALUATED CROSS SECTION ERROR ASSIGNMENT<sup>a</sup>

Reaction	1-162 <sup>b</sup>	162-226	226-361	361-406	406-440	440-455	455-463	463-471	471-481	481-491	491-501	501-521	521-551	551-571	571-621
	1-10-4-7 <sup>0</sup>	4-7-1-5	1-5-1-2	1-2-1-1	1-1-6-1	6-1-1.4	1.4-2.2	2.2-3.0	3.0-4.0	4.0-5.0	5.0-6.0	6.0-8.0	8.0-11.0	11.0-13.0	13.0-18.0
	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)
<sup>23</sup> Na(n,γ)	1	5	5	5	5	5	5	5	5	5	5	5	5	10	10
<sup>24</sup> Mg(n,p)	0	0	0	0	0	0	0	0	0	50	50	10	10	10	10
<sup>27</sup> Al(n,α)	0	0	0	0	0	0	0	0	0	30	20	6	6	10	10
<sup>27</sup> Al(n,p)	0	0	0	0	0	0	0	30	10	8	8	8	8	20	20
<sup>28</sup> Si(n,p)	0	0	0	0	0	0	0	0	0	50	50	15	15	15	15
<sup>31</sup> P(n,p)	0	0	0	0	0	0	0	50	6	12	12	10	10	10	10
<sup>32</sup> S(n,p)	0	0	0	0	0	0	100	20	5	5	5	5	5	8	8
<sup>34</sup> S(n,α)	0	0	0	0	0	0	0	0	0	50	50	25	15	10	10
<sup>35</sup> Cl(n,α)	0	0	0	0	0	0	0	30	20	20	30	30	30	30	30
<sup>45</sup> Sc(n,γ)	2	5	5	6	10	10	11	11	11	11	11	11	11	11	11
<sup>46</sup> Ti(n,p)	0	0	0	0	0	0	50	50	25	20	20	10	10	20	20
<sup>47</sup> Ti(n,p)	0	0	0	0	50	50	50	50	15	15	15	15	15	15	15
<sup>48</sup> Ti(n,p)	0	0	0	0	0	0	0	0	50	50	25	15	15	15	15
<sup>54</sup> Fe(n,p)	0	0	0	0	30	30	30	10	10	8	8	7	5	10	10
<sup>55</sup> Mn(n,γ)	0.8	5	5	5	5	5	5	8	8	8	8	8	10	10	10
<sup>56</sup> Fe(n,p)	0	0	0	0	0	0	0	0	8	8	8	6	6	15	15
<sup>56</sup> Fe(n,γ)	8	8	28	12	12	12	12	12	12	12	12	12	12	12	12
<sup>59</sup> Mn(n,p)	0	0	0	0	0	30	30	10	10	5	5	4	6	10	10
<sup>59</sup> Mn(n,2n)	0	0	0	0	0	0	0	0	0	0	0	0	0	30	20
<sup>59</sup> Co(n,α)	0	0	0	0	0	0	0	0	0	30	15	8	8	10	10
<sup>59</sup> Co(n,γ)	4	5	5	5	6	7	7	7	7	7	7	7	7	10	10
<sup>60</sup> Ni(n,p)	0	0	0	0	0	0	50	50	50	50	15	8	8	8	8
<sup>63</sup> Cu(n,α)	50	50	50	50	50	50	50	50	50	50	25	8	8	10	10
<sup>63</sup> Cu(n,γ)	5	5	5	5	5	5	5	5	7	7	7	7	7	7	7
<sup>63</sup> Cu(n,2n)	0	0	0	0	0	0	0	0	0	0	0	0	0	8	8
<sup>64</sup> Zn(n,p)	0	0	0	0	50	50	50	50	15	15	15	15	15	15	15
<sup>90</sup> Zr(n,2n)	0	0	0	0	0	0	0	0	0	0	0	0	0	30	15
<sup>113</sup> In(n,n')	0	0	0	0	30	20	10	10	10	5	5	5	5	10	10
<sup>113</sup> In(n,γ)	2.5	5	5	5	5	10	17	17	17	17	17	17	17	17	17
<sup>127</sup> I(n,2n)	0	0	0	0	0	0	0	0	0	0	0	0	30	30	15
<sup>197</sup> Au(n,γ)	0.5	4	5	5	5	5	5	5	5	5	5	5	5	7	7
<sup>232</sup> Th(n,γ)	1.4	5	5	5	5	8	8	8	8	8	8	8	8	8	8
<sup>232</sup> Th(n,f)	0	0	0	0	0	30	25	20	10	10	10	10	10	10	10
<sup>235</sup> U(n,f)	2	8	8	8	7	6	5	3	6	6	6	6	6	10	10
<sup>237</sup> Np(n,f)	16	10	10	10	20	5	3	2	5	5	5	10	10	10	10
<sup>238</sup> U(n,f)	0	0	0	0	30	30	4	2	3	3	3	4	4	10	10
<sup>238</sup> U(n,γ)	1.5	5	5	5	5	5	5	5	5	5	5	5	5	10	10
<sup>238</sup> U(n,f)	10	10	10	10	12	10	10	10	10	10	10	10	10	10	10
<sup>239</sup> Pu(n,f)	0.5	8	8	8	7	6	5	5	6	6	6	6	6	10	10

- a. One standard deviation in percent.  
b. SAND-II group numbers.  
c. SAND-II group energy bounds. Note: 1<sup>-10</sup> = 1 x 10<sup>-10</sup>, etc.

### III. Neutron Flux-Spectra - Multiple Foil Method

#### A. General Discussion

Neutron flux-spectra discussed in this paper were determined from measured reaction rates using the SAND-II code(17,18) with input spectra from typical physics calculations. The code calculated reaction rates are compared with the measured reaction rates and the input spectrum is adjusted iteratively to secure the best fit between calculated and measured reaction rate values.

The philosophy adopted by the FRMDC is that for the correlation and subsequent application of fuels and materials irradiation effects data, the basic energy-dependent neutron correlation parameter is a set of measured reaction rates. If properly selected, these measured reaction rates permit an accurate interpolation or transfer of EBR-II test reactor derived neutron induced property change data to planned FTR or LMFBR reactor operating conditions. To affect this transfer, the SAND-II code is used to derive flux-spectral data directly from the measured reaction rates. The derived flux-spectral and associated test reactor property change data are then combined with predicted flux-spectra to complete the transfer for fast reactor engineering and design studies.

Although the absolute accuracy of SAND-II multiple foil derived flux-spectra is of concern and will be briefly considered in this paper, it must be emphasized that it is the relative consistency of the reactor physics and SAND-II derived flux-spectra that is of primary concern for LMFBR fuels and materials development programs. That is, given the capabilities to measure reaction rates accurately, in both test and operating power reactors, it is only necessary to use a reference set of integrally consistent evaluated energy-dependent cross sections for the flux-spectral definition. The SAND-II reference cross section file currently being used by the FRMDC was a first attempt to establish and use such a set and the procedures used to develop the necessary consistency are documented elsewhere.(19) Currently, the SAND-II  $\sigma(E)$ 's make use of the ENDF/B-III fission cross sections for  $^{235}\text{U}$ ,  $^{238}\text{U}$ , and  $^{239}\text{Pu}$ , and the capture reaction for  $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ .

To further improve the relative consistency of SAND-II derived flux-spectra, a number of improvements have been made in the SAND-II algorithm which place limitations on the amount of spectral structure which can be generated. To accomplish this, Oster and Simons(20) developed an improved SAND-II procedure which reduces the amount of artificial structure allowed in an iterative solution by smoothing and subsequent modulation of reactor physics calculated structure back into the solution. Figure 1 shows the results of the application of this new SAND-II smoothing procedure for a typical EBR-II spectrum. Of particular interest is a major flux depression around  $3 \times 10^{-3}$  MeV due to resonance absorption of neutrons by the sodium coolant. The smoothing procedure eliminated most of this depression because the relative activation monitor responses were not sufficiently different in this energy region. Using the reactor physics calculated spectrum, the modulation procedure reshaped the SAND-II solution to include this structure while still maintaining the necessary integral consistency between calculated and measured reaction rates for the 14 monitor reactions shown in the figure.

#### E. Multiple Foil Method Error Analysis

One major step in the quantification of the uncertainty of multiple foil derived flux-spectra is the development of the SAND-II Monte Carlo error analysis code.(16) Basically, the procedure combines uncertainties in the reaction rates and in reaction cross sections (Table II) to provide estimates of uncertainty for the derived flux-spectrum. The Monte Carlo procedure selects values of reaction rates and cross sections within assigned uncertainties for a pre-selected

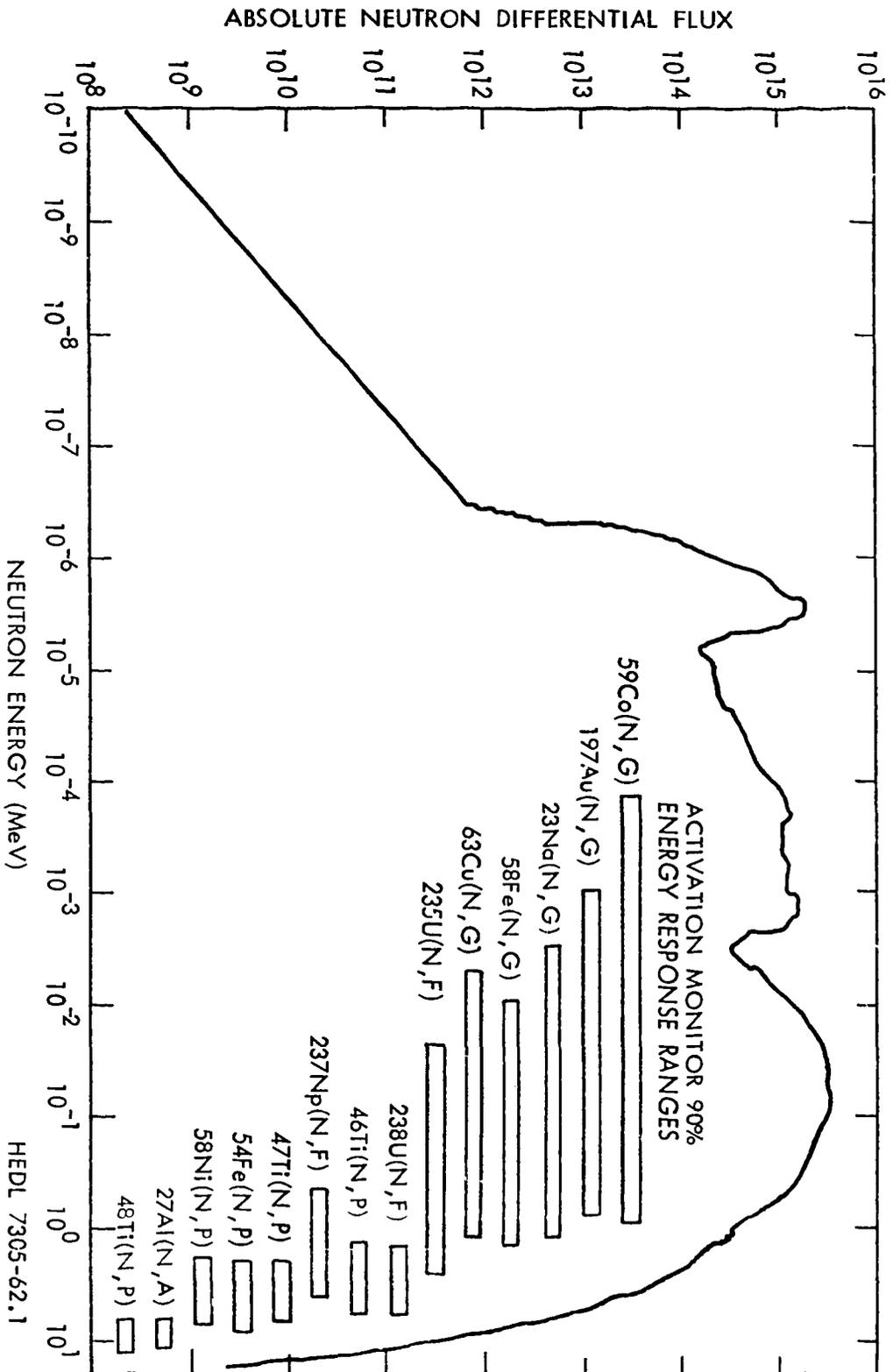


Figure 1. SAND-II Smoothed and Moduled Solution Using 14 Measured Reaction Rates at an EBR-II Location of R=5.2 cm. Z=12.3 cm. Reactor Run 31F at 50 Mw.

number of regular SAND-II runs. These sets of selected values are then used to generate sets of solution spectra and error estimates are derived from these sets of spectra. This process provides realistic absolute and relative error estimates for flux-spectra or spectral parameters such as average neutron energy or neutron flux above any energy.

#### IV. Applications

##### A. Well Established Reference Neutron Fields

Using the ILRR program measured reaction rates for ten foils (see the top part of Figure 2), the SAND-II multiple foil derived flux-spectrum is compared with the available proton recoil measurements of Rogers<sup>(21)</sup> for the Coupled Fast Reactivity Measurement Facility (CFRMF) central core region. If the proton-recoil results have an accuracy of ~5-10% in the 20 keV to 1 MeV range, as might be inferred on the basis of recent intercomparisons between Karlsruhe (KFK), Reactor Centrum Nederland (RCN), and Mol (CEN/SCK),<sup>(22)</sup> then these results strengthen confidence in the SAND-II method of analysis for absolute flux-spectral determination and error estimation for EBR-II fast reactor core spectra with mean energies in the 0.7 to 0.9 MeV range. Confidence in the method has also been established by similar studies for harder fast reactor spectra in the 1.5 to 2.0 MeV range<sup>(13)</sup> and for softer fast reactor spectra in the 0.4 to 0.5 MeV range.<sup>(11)</sup> LMFBR core spectra will have mean energies in the 0.4 to 0.5 MeV range.

##### B. EBR-II Neutron Fields

Knowledge of the EBR-II neutron environment is essential for interpreting fuels and materials experiments conducted at different reactor locations.<sup>(1-6,23-26)</sup> Integral flux and neutron average energy results of the HEDL analysis of the reactor run 50H test, which represents the most recent large-scale, high-power measurements of the EBR-II neutron environment are reported in another paper.<sup>(4)</sup> Here, consideration is given to SAND-II multiple foil derived group flux results for the 50H test and integral flux results for an out-of-core Xenon/Krypton tag gas experiment. Flux-spectral results associated with a boron carbide materials experiment are also presented to complete a brief review of the status of multiple foil flux-spectral characterization for EBR-II.

##### 1. Measured and Calculated Flux-Spectral Comparisons

The SAND-II Monte Carlo code yields estimates of the uncertainties in differential flux, integral flux, and the spectrum average energy. Also available are the uncertainties associated with group fluxes for selected group structures. Using this code, a detailed analysis was performed for two positions in row 2, midplane and 60.4 cm below midplane.<sup>(5)</sup> Table II provides an updated listing of the SAND-II cross section error assignments used for both these analyses.

For the row 2 ( $r = 5.2$  cm) midplane position, Table III lists the values of measured reaction rates and corresponding assigned ( $1\sigma$ ) uncertainties. A typical calculated EBR-II spectral shape (in this case, a 26 group 2DB diffusion code calculation)<sup>(27)</sup> was used as the reference starting approximation for this study.\* The spectrum together with the 14 measured reaction rates, their assigned uncertainties, and the cross section error assignments were input to the SAND-II Monte Carlo code to derive a readjusted multiple foil representation of the reactor spectrum at this location. The results are shown in Figure 3.

\* This code was developed at HEDL for fast reactor engineering and design studies, particularly for FTR.

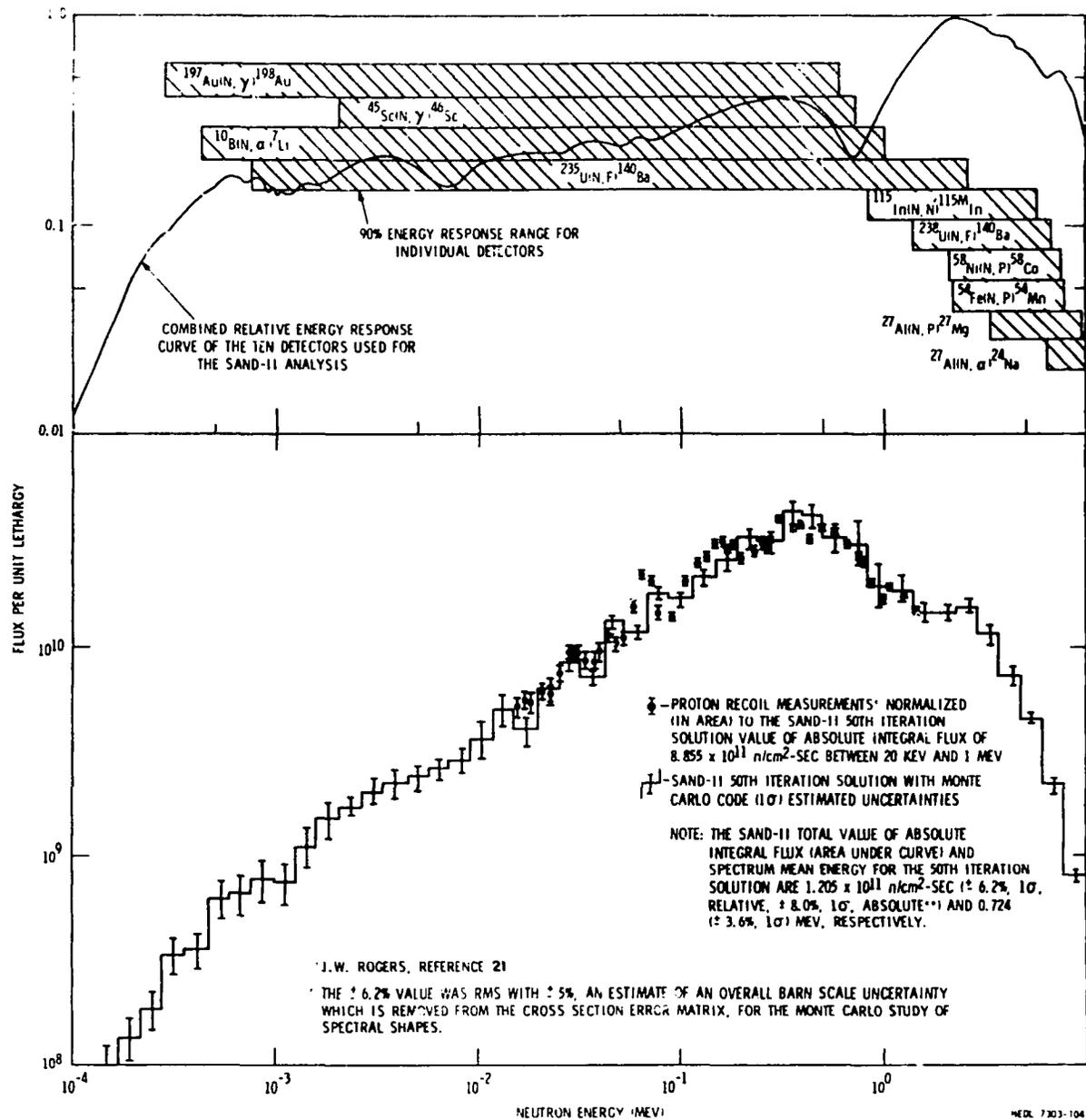


Figure 2. Comparison of SAND-II and Proton Recoil Group Fluxes Per Unit lethargy.

TABLE III  
50H MEASURED REACTION RATES AND ASSIGNED ERROR AT ROW 2 - MIDPLANE

<u>Reaction</u>	<u>Structural - W-5 Reaction Rate Per Second Per Target Nucleus</u>
$^{235}\text{U}(n,f)\text{F.P.}$	3.730 -09 ( $\pm 6.4\%$ )*
$^{238}\text{U}(n,f)\text{F.P.}$	2.376 -10 ( $\pm 6.4\%$ )
$^{237}\text{Np}(n,f)\text{F.P.}$	1.769 -09 ( $\pm 6.2\%$ )
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	3.994 -11 ( $\pm 3.1\%$ )
$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	2.778 -13 ( $\pm 3.4\%$ )
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	5.791 -11 ( $\pm 5.3\%$ )
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	6.240 -12 ( $\pm 3.0\%$ )
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	1.611 -13 ( $\pm 3.4\%$ )
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	6.825 -11 ( $\pm 3.4\%$ )
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	4.968 -11 ( $\pm 3.0\%$ )
$^{239}\text{Pu}(n,f)\text{F.P.}$	4.773 -09 ( $\pm 6.4\%$ )
$^{127}\text{I}(n,2n)^{126}\text{I}$	6.005 -13 ( $\pm 4.1\%$ )
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	3.916 -11 ( $\pm 3.9\%$ )
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	3.850 -13 ( $\pm 5.0\%$ )

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\* Values in parentheses are estimated absolute uncertainties ( $1\sigma$ ). Uncertainties contain counting, fission yield and weight uncertainty estimates as appropriate.

GROUP ABSOLUTE FLUX PER UNIT LETHARGY COMPARISON

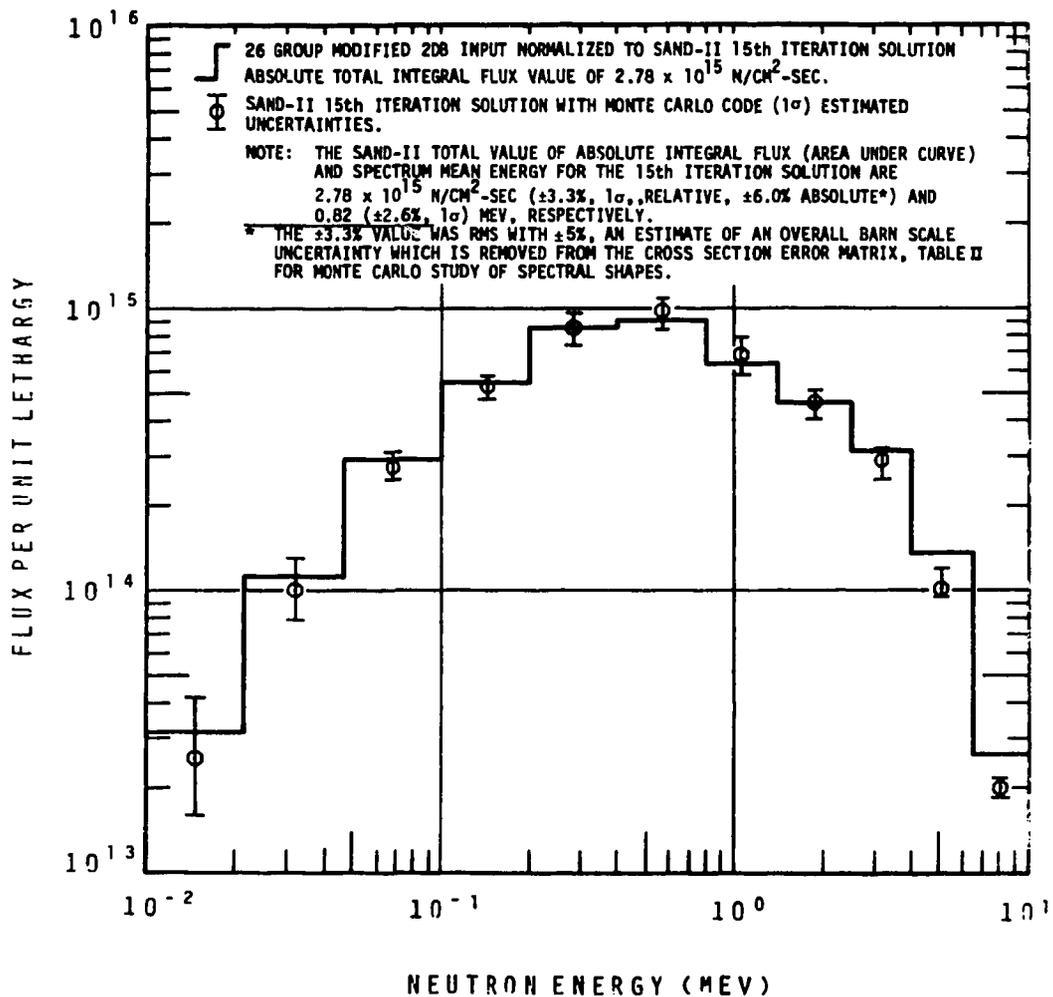


FIGURE 3. Group Absolute Flux per Unit Lethargy Comparison at  $r = 5.2$  cm, Midplane, for Run 50H

The small circles with attached ( $1\sigma$ ) absolute error bars are the SAND-II results. The input spectrum was normalized to the SAND-II total integral flux value of  $2.78 \times 10^{15}$  n/cm<sup>2</sup>-sec and is plotted as a solid line. As can be seen in Figure 3, the shape of the input spectrum was changed in some regions by up to 20 to 30% by the SAND-II process. This is seen better in Figure 4 where the ratio of the calculated to SAND-II group flux values are compared.

Uncertainties due to solution uniqueness are expected to be small and are not included in the error estimates of Figure 3. Based on previous studies,<sup>(1)</sup> the absolute error from this source would be expected to contribute mostly in the energy region below  $\sim 5 \times 10^{-2}$  MeV, where there is less than  $\sim 5\%$  of the total flux and the foil response is low.

As previously stated, for fuels and materials development programs, the relative error between measured-to-measured and measured-to-calculated flux-spectra is more important than the absolute errors. Using the same 2DB calculation, relative group flux comparisons are given in Figure 4 for three other axial locations in row 2. The figure is a composite of comparisons made at four different axial positions, all at the same radius of 5.2 cm. These positions are midplane, -18.7 cm, -41.2 cm, and -60.4 cm. For this figure, we have taken the ratio of the input group absolute flux per unit lethargy normalized to the total SAND-II integral flux and divided it by the SAND-II derived group absolute flux per unit lethargy. This normalization removes any absolute flux-fluence scale differences so that spectral shapes alone can be compared. Ratios have been plotted for the four different locations and exact spectral agreement would produce a ratio of 1. Also provided for reference are bars showing the energy regions that encompass both 99% and 90% of the total flux at each location.

In this comparison, the differences between the typical physics calculated spectra and SAND-II multiple foil derived spectra are obvious. In the region from  $10^{-6}$  MeV to 10 MeV, some calculated group fluxes are a factor of  $\sim 1.7$  higher than the SAND-II values while some are a factor of  $\sim 2.0$  lower. In view of the many sources of error and for certain applications, these differences might not be considered too significant; however, for most LMFBR fuels and materials development programs, they must be accounted for to achieve proper correlation of property change data for engineering and design studies.<sup>(28)</sup>

By improved modeling and more sophisticated reactor physics analysis, ANL has significantly improved the quality of calculated EBR-II flux-spectra.<sup>(3,23-26)</sup> By modeling various axial regions in RZ geometry and each sub-assembly in XY geometry, the effect of subassembly-wide and core-wide spectral variations are considered. Future routine use will be made of these ANL calculations to improve the quality of the agreement between calculated and SAND-II derived flux-spectra, such as those shown in Figures 3 and 4.

## 2. Relative Flux-Spectral Comparisons

That spectral changes can occur within a specific subassembly or between different subassemblies in EBR-II has been recognized for some time. Previous calculations by Miller and Jarka<sup>(24-26)</sup> have indicated that the <sup>235</sup>U averaged fission rate can increase by  $\sim 4\%$  from the center of a structural to the center of a fuel subassembly.\* Part of the 50H dosimetry test was designed to measure such changes so that appropriate corrections could be made to experimental results. For the following comparisons, the Monte Carlo error analysis code

\* A value of  $6(\pm 2)\%$  in total flux measured in the 50H test is equivalent to  $\sim 6\%$  measured <sup>235</sup>U fission rate difference, which is consistent with Miller and Jarka's value of  $\sim 4\%$ , when modeling effects and uncertainties are considered.<sup>(4,5)</sup>

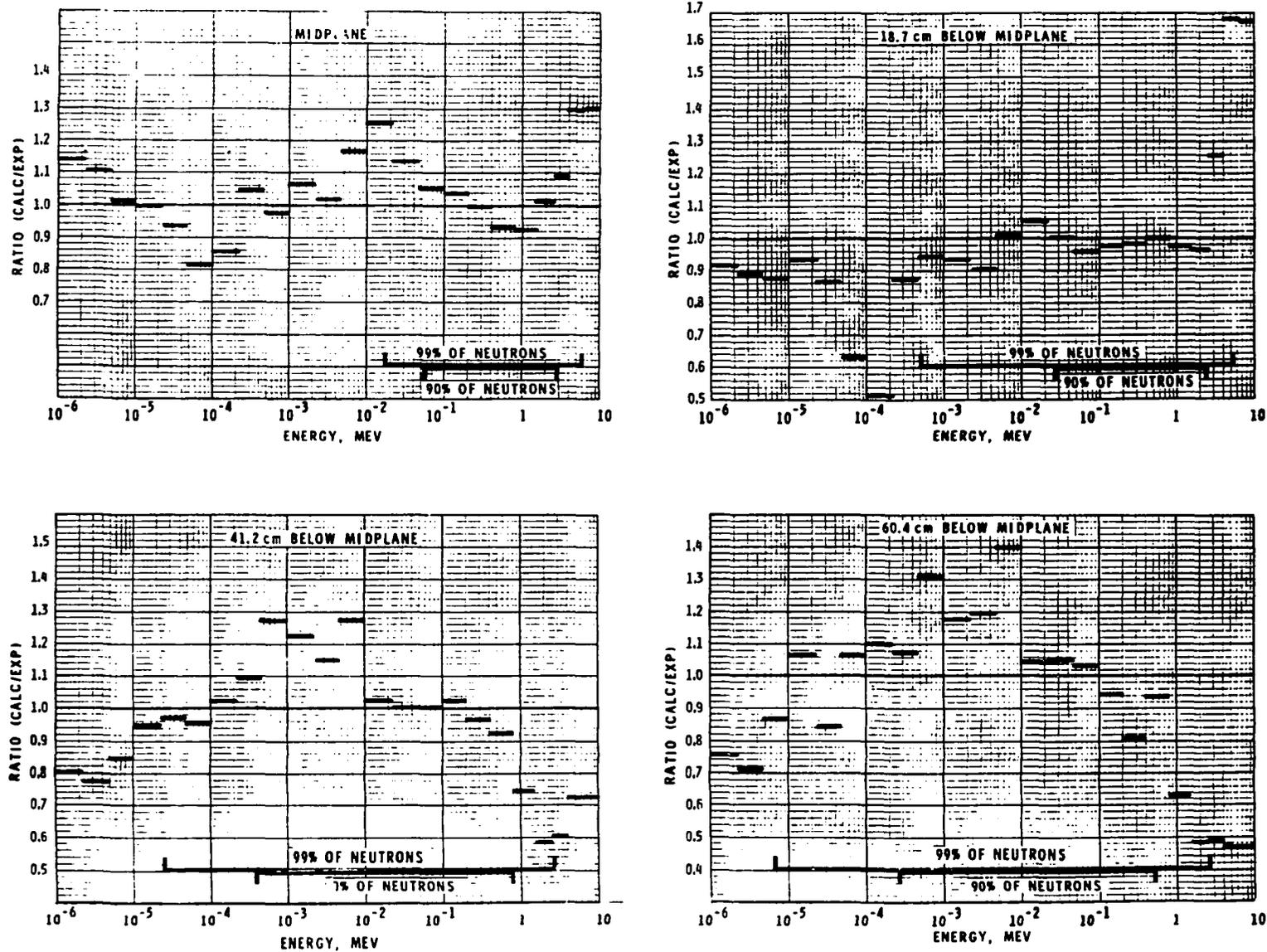


FIGURE 4. Measured to Calculated Spectral Comparison at Four Axial Locations in Row 2 - Run 50H

was run assuming no errors in cross sections, since for relative comparisons of similar spectra one need only consider reaction rate errors; that is, only relative uncertainties in reaction rates between different locations are important for this analysis.

Three different locations, shown in Table IV, were selected for comparison: a set of monitors from set W-5 at a radius of 5.2 cm from the core center on the edge of a structural subassembly; W-8, the same axial location at a radius of 5.9 cm in the center of the same structural subassembly, X125 in reactor position 2B1; and W-53, a set located at a radius of 10.2 cm in the center of the driver subassembly, X126 in reactor position 3B2. Estimates of relative uncertainties for the different reaction rate measurements in each of the three sets were made. Comparisons of the relative uncertainties showed very little difference in values. For this analysis, the uncertainties given in Table V are the largest associated with any reaction from the three different sets. Using the reaction rates given in Table V for set W-8 and the maximum relative uncertainties shown, a Monte Carlo run gave an estimate of the uncertainties associated with the group fluxes for all three sets.

Group averaged fluxes per unit lethargy were derived by the multiple foil method for these two locations. Ratios of the group fluxes, W-5 to W-8, are plotted in Figure 5. The uncertainties derived for the group fluxes from the W-8 Monte Carlo analysis were root mean squared with themselves to provide an estimate of the uncertainty in the ratio. Using these uncertainties, the uncertainty bands shown as error bars on the ratios in Figure 5 were calculated. The ratios were computed only for the 15 highest energy groups, since this region encompasses 94% of the flux. The ratios presented in Figure 5 show a trend indicating that the spectrum is softer in the center of a structural subassembly than at the edge.

The same procedure was employed to compare spectral differences between the center of the driver subassembly (W-53) and the center of the structural subassembly (W-8). The ratios of group fluxes from the driver to the structural subassembly are plotted in Figure 6 along with the ( $1\sigma$ ) error bars obtained as before. In this case, a more pronounced difference in the two spectra is seen, with a significantly harder spectrum in the center of the fuel than in the structural subassembly. The spectrum mean energy changes associated with these and other spectral differences for EBR-II are discussed elsewhere.(1,4,5,29,30) When knowledge of such spectral changes is needed for subsequent data correlation and neutron damage function analysis,(28-30)the dosimetry procedures described previously will provide this information.

### 3. Materials Subassembly Flux-Spectral Characterization

Flux-spectral and fluence determinations are made routinely by the FRMDC for materials subassemblies irradiated in EBR-II. A brief discussion of the results obtained for two tests will complete this discussion of flux-spectral characterization. The procedures used by the FRMDC to correct for varying power levels and to make necessary burn-in and -out corrections for individual monitors for long term exposures are described elsewhere.(6,31,32)

Subassembly X135 for an FTR Xenon/Krypton tag gas experiment was irradiated in EBR-II reactor position 4F1 from run 51A through run 53E for a total exposure of 4655 MWD. The cycle time history of the irradiation was used as input to the time-history code (TIMH)(6,31) with a set of measured activation rates to obtain a set of measured reaction rates for SAND-II analysis.

Integral flux results of the SAND-II analysis are given in Table VI as reported to the experimenter for an extreme out-of-core position at  $r=17.7$  cm and  $z=69.8$  cm.(33) The SAND-II derived spectrum mean energy of 0.163 MeV and total fluence value of  $1.18 \times 10^{21}$  n/cm<sup>2</sup> are also given. A SAND-II Monte Carlo and solution uniqueness(1,6,12,13) study was required for this test because the

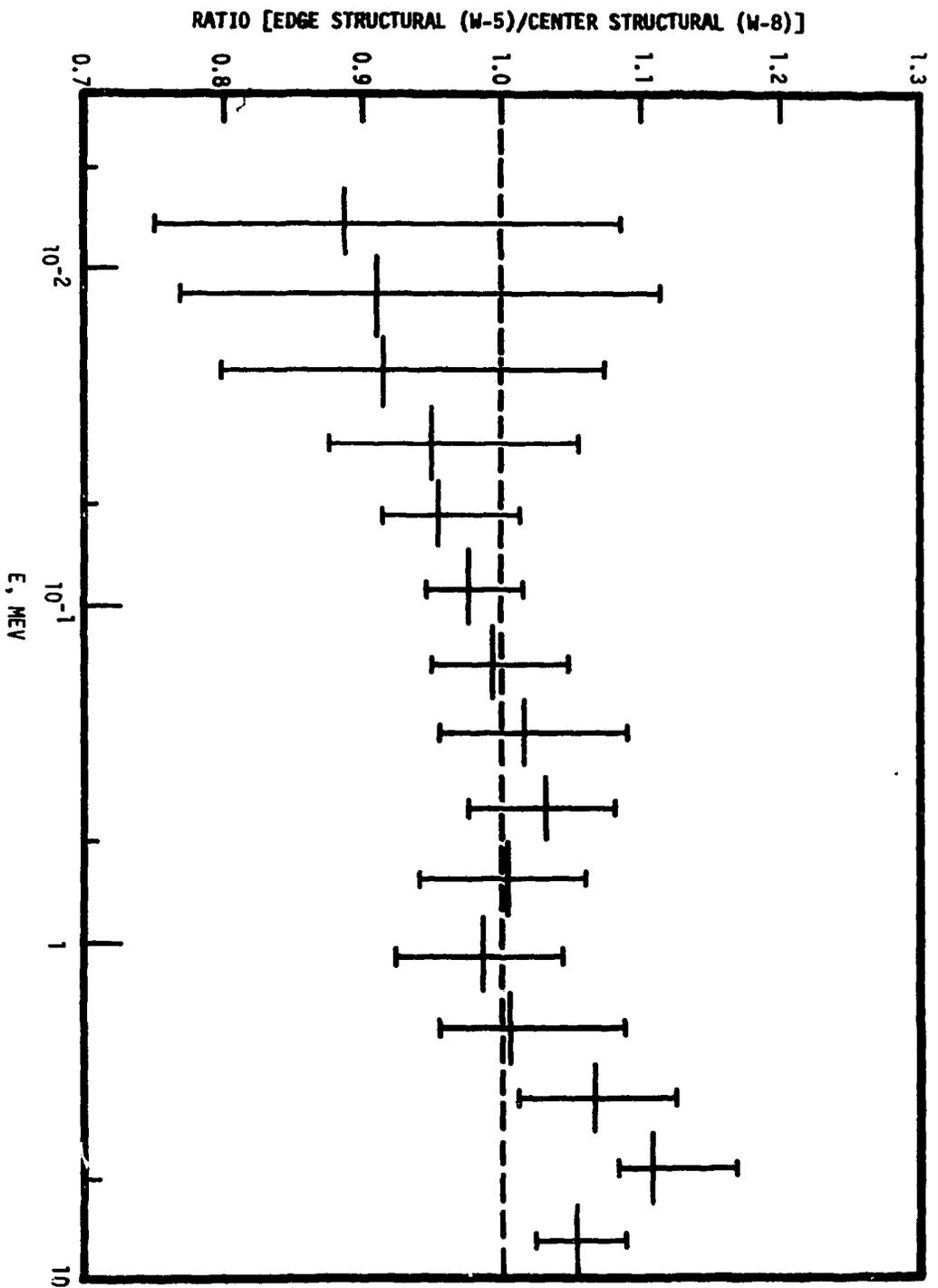


FIGURE 5 Structural Subassembly - Edge to Center Spectral Comparison, 50H, Row 2, Midplane

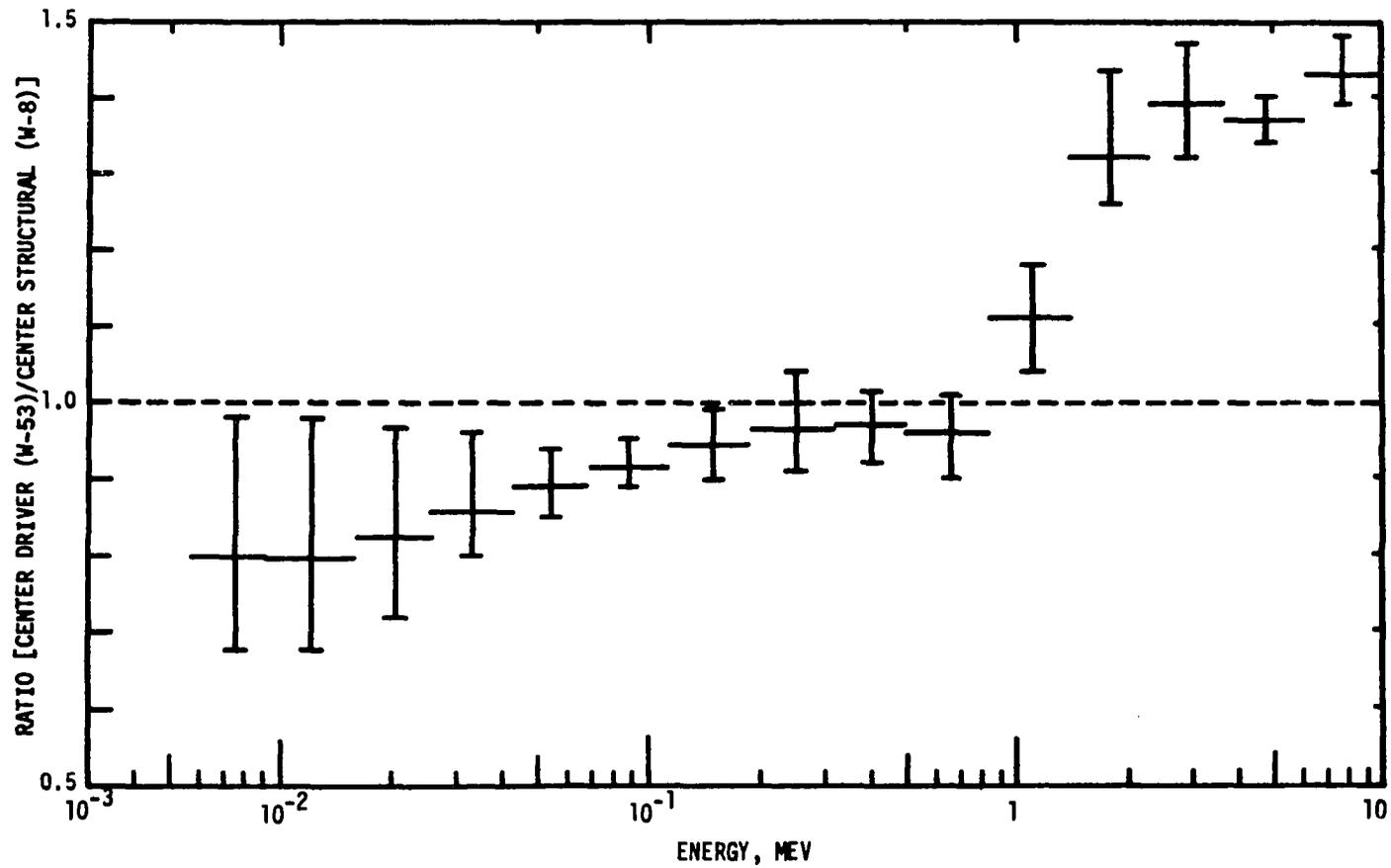


FIGURE 6 Center of Driver to Center of Structural Spectral Comparisons at Reactor Midplane, Run 50H

TABLE IV  
LOCATIONS OF MONITOR SETS USED FOR SPECTRAL COMPARISONS

<u>Monitor Set</u>	<u>Subassembly</u>	<u>Position</u>	<u>Radius</u>
W-5	X125	Edge	5.2 cm
W-8	X125	Center	5.9 cm
W-53	X126	Center	10.2 cm

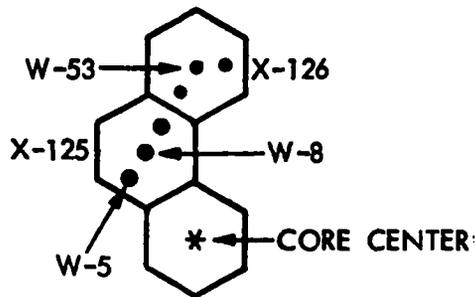


TABLE V  
50H MEASURED REACTION RATES AND ASSIGNED RELATIVE ERRORS  
R = 5.9 CM AT MIDPLANE (W-8) AND R = 10.2 CM AT MIDPLANE (W-53)

<u>Reaction</u>	<u>Structural - W-8</u> <u>Reaction Rate per Second</u> <u>per Target Nucleus</u>	<u>Fuel - W-53</u> <u>Reaction Rate per Second</u> <u>per Target Nucleus</u>
$^{235}\text{U}(n, f)^{140}\text{Ba}$	3.762 -09 (2.0%)*	3.881 -09 (2.0%)*
$^{238}\text{U}(n, f)^{140}\text{Ba}$	2.050 -10 (2.4%)	2.803 -10 (2.4%)
$^{237}\text{Np}(n, f)^{140}\text{Ba}$	1.773 -09 (2.6%)	2.017 -09 (2.6%)
$^{45}\text{Sc}(n, \gamma)^{46}\text{Sc}$	4.093 -11 (1.4%)	3.851 -11 (1.4%)
$^{63}\text{Cu}(n, \alpha)^{60}\text{Co}$	2.708 -13 (2.3%)	3.867 -13 (2.3%)
$^{46}\text{Ti}(n, p)^{46}\text{Sc}$	5.716 -12 (1.3%)	7.981 -12 (1.3%)
$^{58}\text{Ni}(n, p)^{58}\text{Co}$	6.356 -11 (2.0%)	8.759 -11 (2.0%)
$^{54}\text{Fe}(\gamma, p)^{54}\text{Mn}$	4.575 -11 (1.3%)	6.269 -11 (1.3%)
$^{239}\text{Pu}(n, f)^{140}\text{Ba}$	4.520 -09 (1.4%)	4.779 -11 (1.4%)

\* Values assigned are estimated relative  $1\sigma$  uncertainties. These values include only counting and weight uncertainties.

TABLE VI

XENON/KRYPTON TAG GAS ABSOLUTE INTEGRAL FLUXES AND ESTIMATED UNCERTAINTIES  
EBR-II EXPERIMENT X135, RADIUS 17.7 CM, AXIAL DISTANCE FROM MIDPLANE 69.8 CM

Energy (MeV)	Absolute Integral Flux <sup>(a)</sup> n/cm <sup>2</sup> -sec	Estimated Foil Set Solution Uniqueness Uncertainties (1 Std. Dev. % Error)	Monte Carlo Foil Reaction Rate and Cross Section Uncertainties (1 Std. Dev. % Error)		Estimated Total <sup>(b)</sup> Absolute Uncertainties (1 Std. Dev. % Error)	
			+	-	+	-
>10 <sup>-10</sup>	1.84 +14	4	12	10	13	11
>10 <sup>-6</sup>	1.84 +14	4	12	10	13	11
>10 <sup>-5</sup>	1.82 +14	4	12	11	13	12
>10 <sup>-4</sup>	1.75 +14	4	13	10	14	11
>10 <sup>-3</sup>	1.59 +14	4	14	12	15	13
>10 <sup>-2</sup>	1.44 +14	3	14	13	14	13
>10 <sup>-1</sup>	8.21 +13	16	15	9	22	18
>1	1.69 +12	26	19	16	32	30
>2	3.84 +11	6	10	12	12	13
>3	1.34 +11	5	8	10	9	11
>4	5.79 +10	3	11	12	11	12
>5	2.99 +10	2	11	10	11	10
Spectrum Mean Energy (MeV)	0.163					
Total Fluence n/cm <sup>2</sup>	1.18 x 10 <sup>21</sup>					

(a) At a stated reactor power of 62.5 MW and based on a selected best set of 9 foil reaction rates.

(b) Combined uncertainties, columns 5 and 6.

uncertainties associated with out-of-core reactor physics calculations are large and the experimenter had requested absolute flux values.

The estimated total absolute uncertainties (plus and minus one standard deviation) listed in the table are the root mean square values of the estimated foil set solution uniqueness uncertainties and the Monte Carlo analyses of foil reaction rate and cross section uncertainties.

The solution uniqueness uncertainty is based on the difference between the best solution and the SAND-II solution derived from an extreme, but smoothed, input. The extreme was taken as an "E" form from  $10^{-10}$  to  $10^{-5}$  MeV, a constant from  $10^{-5}$  to 3 MeV, with a fission form above 3 MeV. The best solution input form is based on a 26 group, 2DB, two-dimensional diffusion code calculation.<sup>(27)</sup> Uncertainties derived by the Monte Carlo analysis are associated with errors in cross section and reaction rates, including errors from interpolation of measured activation data obtained from monitors placed adjacent to (above and below) the Xenon/Krypton capsules.

As the second example, subassembly X140 for an FTR boron carbide experiment was irradiated during EBR-II runs 51C through 53E for a total of 4009 MWD. Total flux, fluence, and predicted helium production rates for infinitely dilute  $^{10}\text{B}$  at spectral set locations adjacent to boron carbide pellets were provided to the experimenter, Table VII.<sup>(34)</sup> The predicted infinitely dilute helium production rates were calculated using the SAND-II derived flux-spectra at each location and a recently up-dated SAND-II energy-dependent cross section for total helium production. The helium production results of Table VII were correlated with measured  $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$  reaction rates from iron gradient wires included in the subassembly. The ratio of the calculated helium production rate to the measured  $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$  reaction rate was found to be constant to within  $\sim 3\%$  for all spectral locations.\* Consequently, a direct transfer of predicted infinitely dilute helium production rates could be reliably made to other axial locations using iron gradient wire results.

Measured total infinitely dilute helium production rates are being determined by Farrar by high sensitivity helium mass spectrometry at Atomic International (AI) from small ( $\sim 0.1$ " long by  $\sim 0.050$ " diameter) vanadium encapsulated  $^{10}\text{B}$  specimens that were fabricated at ORNL<sup>(15)</sup> and were included with the spectral sets, Table VII.<sup>(35)</sup> Comparison of the Table VII predicted and AI measured values of helium for the X140 subassembly as well as other test reactor results are presented elsewhere.<sup>(6,29,36)</sup> Based on the results of these comparisons, the SAND-II evaluated energy dependent total helium production cross section will be readjusted, if necessary, to improve the reliability of predicted helium generation rates<sup>(6,36)</sup> for FTR and LMFBR engineering and design studies.

## V. Conclusions

The SAND-II method of flux-spectral characterization for fast reactor fuels and materials irradiation experiments now includes methods of error propagation and uncertainty analyses. The flux-spectral information is merely an intermediate step in the correlation and application (or transfer) of test reactor derived irradiation effects data to LMFBR operating conditions. To effect this transfer the SAND-II procedure is used to derive consistent flux-spectral data directly from the measured reaction rates and reactor physics calculations. The derived neutron flux-spectra and associated property change data are then

\* Radial flux distributions from the run 50H dosimetry test indicated that the midplane total flux ( $E > 10^{-10}$  MeV) depression caused by the X140 assembly was  $\sim 30\%$  compared to a structural subassembly in the same position.

TABLE VII  
FLUX, FLUENCE AND HELIUM PRODUCTION RATES FOR  $^{10}\text{B}$

<u>Spectral Set Locations, cm</u>	<u>Total Flux (<math>E &gt; 10^{-10}</math> MeV) <math>\times 10^{-15}</math> n/cm<sup>2</sup>-sec</u>	<u>Fluence <math>\times 10^{-22}</math> n/cm<sup>2</sup></u>	<u>Helium Production Rate atom/sec-nucleus <math>\times 10^9</math>(a)</u>
R = 14.3 Z = -2.5	2.03	1.13	2.17
R = 16.3 Z = -2.5	1.90	1.05	2.13
R = 16.3 Z = -10.8	1.72	0.95	1.95
R = 16.3 Z = -16.1	1.50	0.83	2.05
R = 18.3 Z = -2.5	1.91	1.06	2.07

(a) This column is the reaction rate based on the unfolded spectra and the energy dependent cross section for helium production in the SAND-II library, coded B10(N,A)LI7FS.

combined with calculated flux-spectra based on predicted LMFBR operating conditions to complete the transfer for fast reactor engineering and design studies.

The neutron flux and integral flux spectrum can be determined to an accuracy within  $\pm 10$  to 30% ( $1\sigma$ ) in most fast reactors using the monitors and analytical methods presented. However, relative uncertainties which are of primary concern for irradiation experimentation in fast reactors are substantially smaller. To reduce this relative uncertainty, an ENDF/B file of evaluated energy-dependent reaction cross sections is being developed. The goal accuracy for measured fission reaction rates in EBR-II and FTR for  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{238}\text{U}$  is  $\pm 5\%$  at the 95% confidence level. For other fission and non-fission reactions the goal is better than  $\pm 10\%$  at the 95% confidence level. These goal accuracies are expected to be achieved by the end of fiscal year 1975. Improved cross sections and improved reaction rate measurements will increase the consistency and relative accuracy of neutron environment characterization for future EBR-II and FTR fuels and materials irradiation experiments.

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REMARKS ON RESPONSE RANGES OF ACTIVATION DETECTORS

Willem L. Zijp

1. Looking at the responses of activation detectors in various spectra, one observes an influence of the presence of a Cd cover on the magnitude of the total response of the threshold reactions. This influence is about 2%, i.e. the response of Cd covered threshold detectors is about 2% less than the response of bare detectors.
2. For a typical thermal reactor spectrum there is no bare detector with a 90% response range from about 600 eV (the end of the response of  $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$ ) to about 475 keV (the beginning of the response of  $^{237}\text{Np}(n,f)\text{FP}$ ).
3. For such a spectrum there is no cadmium covered detector with a 90% response range from about 10 keV (the end of the response of  $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$ ) to about 475 keV (begin point for  $^{237}\text{Np}(n,f)\text{FP}$ ).
4. When for a certain spectrum two detectors have the same 90% range, then this does not hold automatically also for other neutron spectra, since the cross section curves may have clearly different shapes (like the reactions  $^{55}\text{Mn}(n,f)^{56}\text{Mn}$  and  $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ ).

Table 1:

RESPONSE RANGES FOR BARE FOILS  
without cadmium cover

reaction	spectrum 12	spectrum 59	spectrum 36	spectrum 1
$^{164}\text{Dy}(n,\gamma)^{165}\text{Dy}$	4.50 MeV...0.19 eV	110 eV...0.048 MeV	47.5 keV...2.7 MeV	150 keV...3.7 MeV
$^{239}\text{Pu}(n,f)\text{F.P.}$	5.25 MeV...0.425 eV	27 eV...1.8 MeV	84 keV...4.2 MeV	270 keV...5.1 MeV
$^{151}\text{Eu}(n,\gamma)^{152}\text{Eu}$	4.00 MeV...0.48 eV	0.60 eV...0.066 MeV	11.5 keV...1.2 MeV	22 keV...2.3 MeV
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	4.75 MeV...0.50 eV	36 eV...0.32 MeV	17 keV...1.6 MeV	45 keV...3.2 MeV
$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	4.75 MeV...1.05 eV	340 eV...0.12 MeV	3 keV...2.6 MeV	55 keV...4.4 MeV
$^{235}\text{U}(n,f)\text{F.P.}$	4.50 MeV...1.42 eV	45 eV...1.2 MeV	55 keV...4.0 MeV	190 keV...5.1 MeV
$^{115}\text{In}(n,\gamma)^{116}\text{In}^m$	11 MeV...1.7 eV	1.35 eV...0.36 MeV	30 keV...2.4 MeV	100 keV...3.2 MeV
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	10.5 MeV... 18 eV	5 eV...0.06 MeV	12 keV...1.4 MeV	36 keV...2.5 MeV
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	55 MeV...100 eV	20 eV...0.22 MeV	25.5 keV...2.1 MeV	92 keV...2.8 MeV
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	5.5 MeV...135 eV	115 eV...4.5 MeV	4.5 keV...1.4 MeV	18 keV...2.4 MeV
$^{232}\text{Th}(n,\gamma)^{233}\text{Th}$	11.5 MeV...180 eV	24 eV...0.4 MeV	34 keV...1.9 MeV	80 keV...3.0 MeV
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	50 MeV...320 eV	255 eV...0.011 MeV	8.8 keV...2.1 MeV	32 keV...3.8 MeV
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	5.0 MeV...340 eV	230 eV...0.32 MeV	47.5 keV...2.3 MeV	160 keV...3.3 MeV
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	5.0 MeV...600 eV	575 eV...0.10 MeV	7.6 keV...2.5 MeV	25.5 keV...4.0 MeV

Spectrum 12: 293 K Maxwellian matched at 0.14 eV to 1/E matched at 0.5 MeV to Watt fission spectrum.

Spectrum 59: ECEL core 14-13 calc. + Godiva below 0.00926 eV + fission spectrum above 3 MeV.

Spectrum 36: EBR-hole 1 + fission below 0.05 MeV and above 3 MeV.

Spectrum 1: Watt fission neutron spectrum.

Table 2:

RESPONSE RANGES FOR FOILS WITH CD COVER  
with cadmium cover

reaction	spectrum 12	spectrum 59	spectrum 36	spectrum 1
$^{164}\text{Dy}(n,\gamma)^{165}\text{Dy}$	0.42 eV...0.16 keV	115 eV...0.052 MeV	47.5 keV...2.7 MeV	150 keV...2.3 MeV
$^{239}\text{Pu}(n,f)\text{F.P.}$	0.38 eV...3.4 keV	47.5 eV...1.9 MeV	8.4 keV...4.3 MeV	270 keV...5.1 MeV
$^{151}\text{Eu}(n,\gamma)^{152}\text{Eu}$	0.40 eV...0.060 keV	12.8 eV...0.069 MeV	11.5 keV...1.2 MeV	22 keV...2.3 MeV
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	0.48 eV...0.58 keV	42.5 eV...0.32 MeV	17 keV...1.6 MeV	45 keV...3.2 MeV
$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	0.50 eV...3.2 keV	380 eV...0.12 MeV	3 keV...2.7 MeV	57.5 keV...4.5 MeV
$^{235}\text{U}(n,f)\text{F.P.}$	0.55 eV...6.6 keV	47.5 eV...0.12 MeV	55 keV...4.0 MeV	190 keV...5.1 MeV
$^{115}\text{In}(n,\gamma)^{116}\text{In}^m$	1.00 eV...0.002 keV	1.35 eV...0.38 MeV	30 keV...2.4 MeV	105 keV...3.2 MeV
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	4.25 eV...0.069 keV	5 eV...0.06 MeV	12 keV...1.4 MeV	36 keV...2.5 MeV
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	6.03 eV...1.00 keV	20 eV...0.23 MeV	25.5 keV...2.1 MeV	92 keV...2.8 MeV
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	0.84 eV...0.15 keV	115 eV...0.0045 MeV	4.5 keV...1.5 MeV	18 keV...2.4 MeV
$^{232}\text{Th}(n,\gamma)^{233}\text{Th}$	1.50 eV...1.05 keV	24 eV...0.42 MeV	34 keV...2.0 MeV	80 keV...3.1 MeV
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	0.63 eV...1.05 keV	255 eV...0.011 MeV	8.8 keV...2.1 MeV	32 keV...3.8 MeV
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	0.63 eV...2.8 keV	230 eV...0.32 MeV	47.5 keV...2.3 MeV	160 keV...3.3 MeV
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	0.60 eV...10 keV	575 eV...0.10 MeV	7.6 keV...2.5 MeV	25.5 keV...4.0 MeV

Spectrum 12: 293 K Maxwellian matched at 0.14 eV to 1/E matched at 0.5 MeV to Watt fission spectrum.

Spectrum 59: ECEL core 14-13 calc. + Godiva below 0.00926 eV + fission spectrum above 3 MeV.

Spectrum 36: EBR-hole 1 + fission below 0.05 MeV and above 3 MeV.

Spectrum 1: Watt fission neutron spectrum.

Table 3: COMPARISON OF RESPONSE RANGES  
for activation and fission detectors

reaction	energy range comprising 90% response in fission neutron spectrum
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	18 keV ... 2.4 MeV
$^{151}\text{Eu}(n,\gamma)^{152}\text{Eu}$	22 keV ... 2.3 MeV
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	26 keV ... 4.0 MeV
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	32 keV ... 3.8 MeV
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	36 keV ... 2.5 MeV
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	45 keV ... 3.2 MeV
$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	55 keV ... 4.4 MeV
$^{232}\text{Th}(n,\gamma)^{233}\text{Th}$	80 keV ... 3.0 MeV
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	92 keV ... 2.8 MeV
$^{115}\text{In}(n,\gamma)^{116}\text{In}^m$	100 keV ... 3.2 MeV
$^{164}\text{Dy}(n,\gamma)^{165}\text{Dy}$	150 keV ... 3.7 MeV
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	160 keV ... 3.3 MeV
$^{235}\text{U}(n,f)\text{F.P.}$	190 keV ... 5.1 MeV
$^{239}\text{Pu}(n,f)\text{F.P.}$	270 keV ... 5.1 MeV
$^{237}\text{Np}(n,f)\text{F.P.}$	0.69 MeV ... 5.6 MeV
$^{115}\text{In}(n,n')^{115}\text{In}^m$	1.2 MeV ... 5.8 MeV
$^{238}\text{U}(n,f)\text{F.P.}$	1.5 MeV ... 6.7 MeV
$^{232}\text{Th}(n,f)\text{F.P.}$	1.5 MeV ... 7.2 MeV
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	2.1 MeV ... 7.0 MeV
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	2.1 MeV ... 7.5 MeV
$^{31}\text{P}(n,p)^{31}\text{Si}$	2.2 MeV ... 7.0 MeV
$^{64}\text{Zn}(n,p)^{64}\text{Cu}$	2.3 MeV ... 7.8 MeV
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	2.3 MeV ... 7.8 MeV
$^{32}\text{S}(n,p)^{32}\text{P}$	2.5 MeV ... 7.5 MeV
$^{60}\text{Ni}(n,p)^{60}\text{Co}$	2.7 MeV ... 9.6 MeV
$^{35}\text{Cl}(n,\alpha)^{32}\text{P}$	3.2 MeV ... 8.0 MeV
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	3.4 MeV ... 9.1 MeV
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	3.5 MeV ... 9.3 MeV
$^{34}\text{S}(n,\alpha)^{31}\text{Si}$	5.1 MeV ... 10.4 MeV
$^{28}\text{Si}(n,p)^{28}\text{Al}$	5.4 MeV ... 10.1 MeV
$^{56}\text{Fe}(n,p)^{56}\text{Co}$	5.5 MeV ... 11.0 MeV
$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	6.1 MeV ... 11.3 MeV
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	6.4 MeV ... 11.9 MeV
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	6.5 MeV ... 11.5 MeV
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	6.6 MeV ... 12.8 MeV
$^{127}\text{I}(n,2n)^{126}\text{I}$	10.0 MeV ... 14.6 MeV
$^{63}\text{Cu}(n,2n)^{62}\text{Cu}$	11.9 MeV ... 16.4 MeV
$^{90}\text{Zr}(n,2n)^{89}\text{Zr}$	12.5 MeV ... 16.7 MeV
$^{58}\text{Ni}(n,2n)^{57}\text{Ni}$	13.2 MeV ... 17.0 MeV

List of figures:

- Fig. 1: Watt fission neutron spectrum. Indicated are the 90% response ranges of bare detectors.
- Fig. 2: Watt fission neutron spectrum. Indicated are the 90% response ranges of cadmium covered detectors.
- Fig. 3: Watt fission neutron spectrum. Indicated are the 90% response ranges of detectors covered by a 8 mm layer consisting of 90%  $^{10}\text{B}$  + 10%  $^9\text{B}$ .
- Fig. 4: EBR hole 1 spectrum + fission spectrum below 0.05 MeV and above 3 MeV. Indicated are the 90% response ranges of bare detectors.
- Fig. 5: EBR hole 1 spectrum + fission spectrum below 0.05 MeV and above 3 MeV. Indicated are the 90% response ranges of cadmium covered detectors.
- Fig. 6: EBR hole 1 spectrum + fission spectrum below 0.5 MeV and above 3 MeV. Indicated are the 90% response ranges of detectors covered by a 8 mm layer consisting of 90%  $^{10}\text{B}$  + 10%  $^9\text{B}$ .
- Fig. 7: Calculated ECEL core 14-13 spectrum + Godiva spectrum below 0.00926 eV + fission spectrum above 3 MeV. Indicated are the 90% response ranges of bare detectors.
- Fig. 8: Calculated ECEL core 14-13 spectrum + Godiva spectrum below 0.00926 eV + fission spectrum above 3 MeV. Indicated are the 90% response ranges of cadmium covered detectors.
- Fig. 9: ECEL 14-13 spectrum + Godiva spectrum below 0.00926 eV + fission spectrum above 3 MeV. Indicated are the 90% response ranges of detectors covered by a 8 mm layer consisting of 90%  $^{10}\text{B}$  + 10%  $^9\text{B}$ .
- Fig. 10: 293 K Maxwellian spectrum matched at 0.14 eV to 1/E spectrum, matched at 0.5 MeV to Watt fission spectrum. Indicated are the 90% response ranges of bare detectors.

Fig. 11: 293 K Maxwellian spectrum matched at 0.14 eV to 1/E spectrum, matched at 0.5 MeV to Watt fission spectrum. Indicated are the 90% response ranges of cadmium covered detectors.

Fig. 12: 293 K Maxwellian spectrum matched at 0.14 eV to 1/E spectrum, matched at 0.5 MeV to a Watt fission spectrum. Indicated are the 90% response ranges of detectors covered by a 8 mm layer consisting of 90%  $^{10}\text{B}$  + 10%  $^9\text{B}$ .



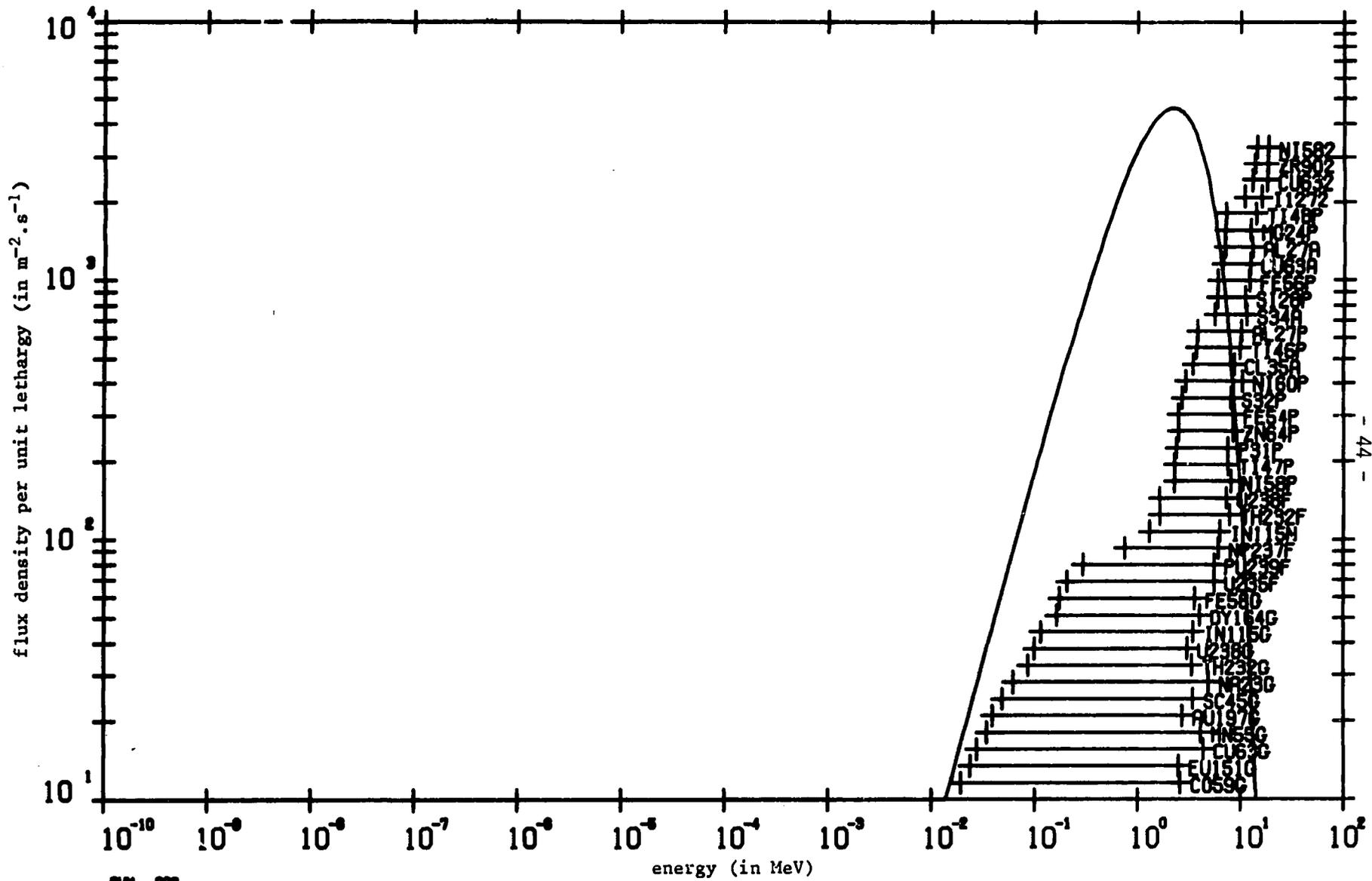
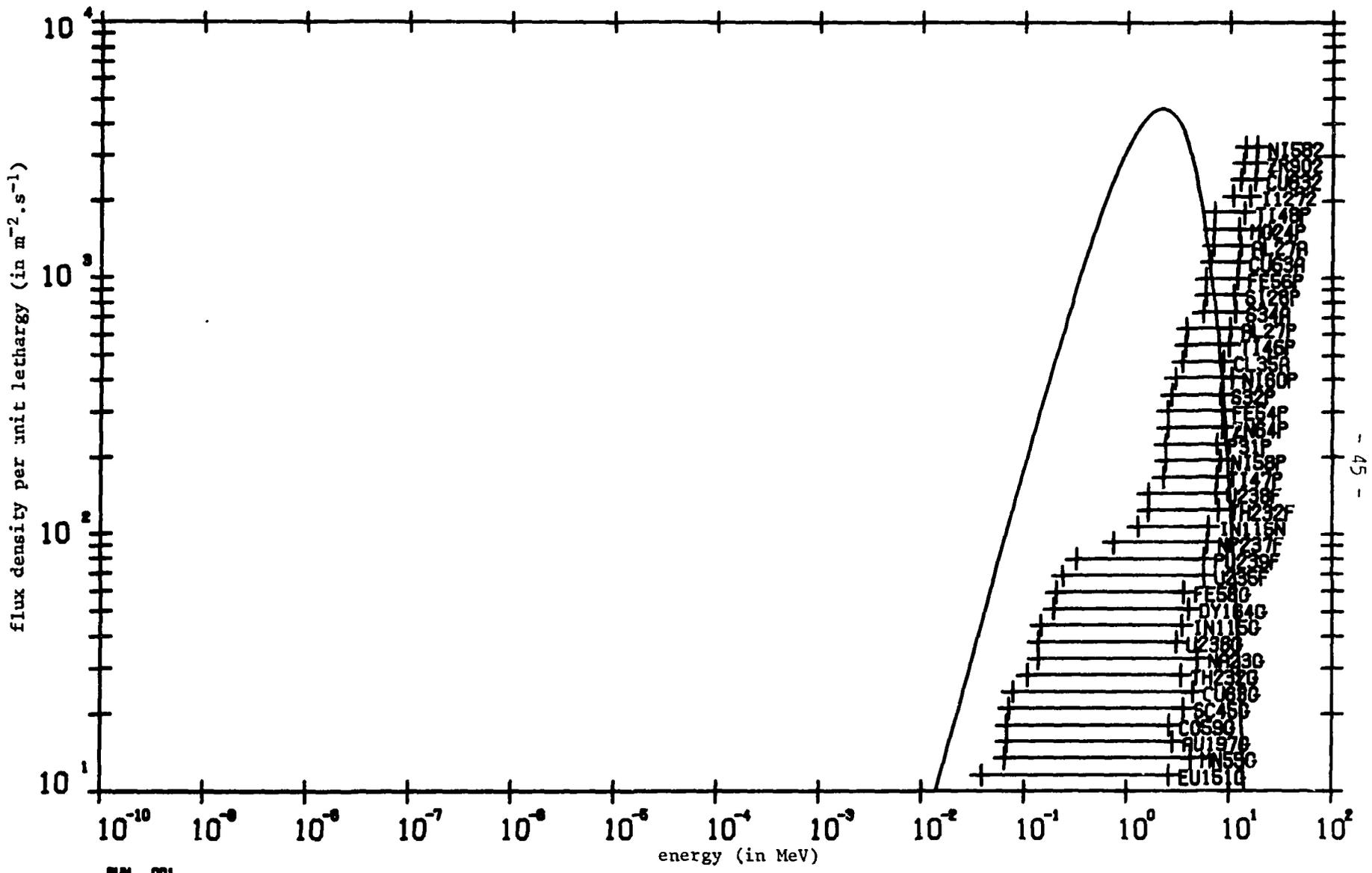


Fig. 2: Watt fission neutron spectrum. Indicated are the 90% response ranges of cadmium covered detectors.



NUM 001

Fig. 3: Watt fission neutron spectrum. Indicated are the 90% response ranges of detectors covered by a 8 mm layer consisting of 90%  $^{10}\text{B}$  + 10%  $^9\text{B}$ .



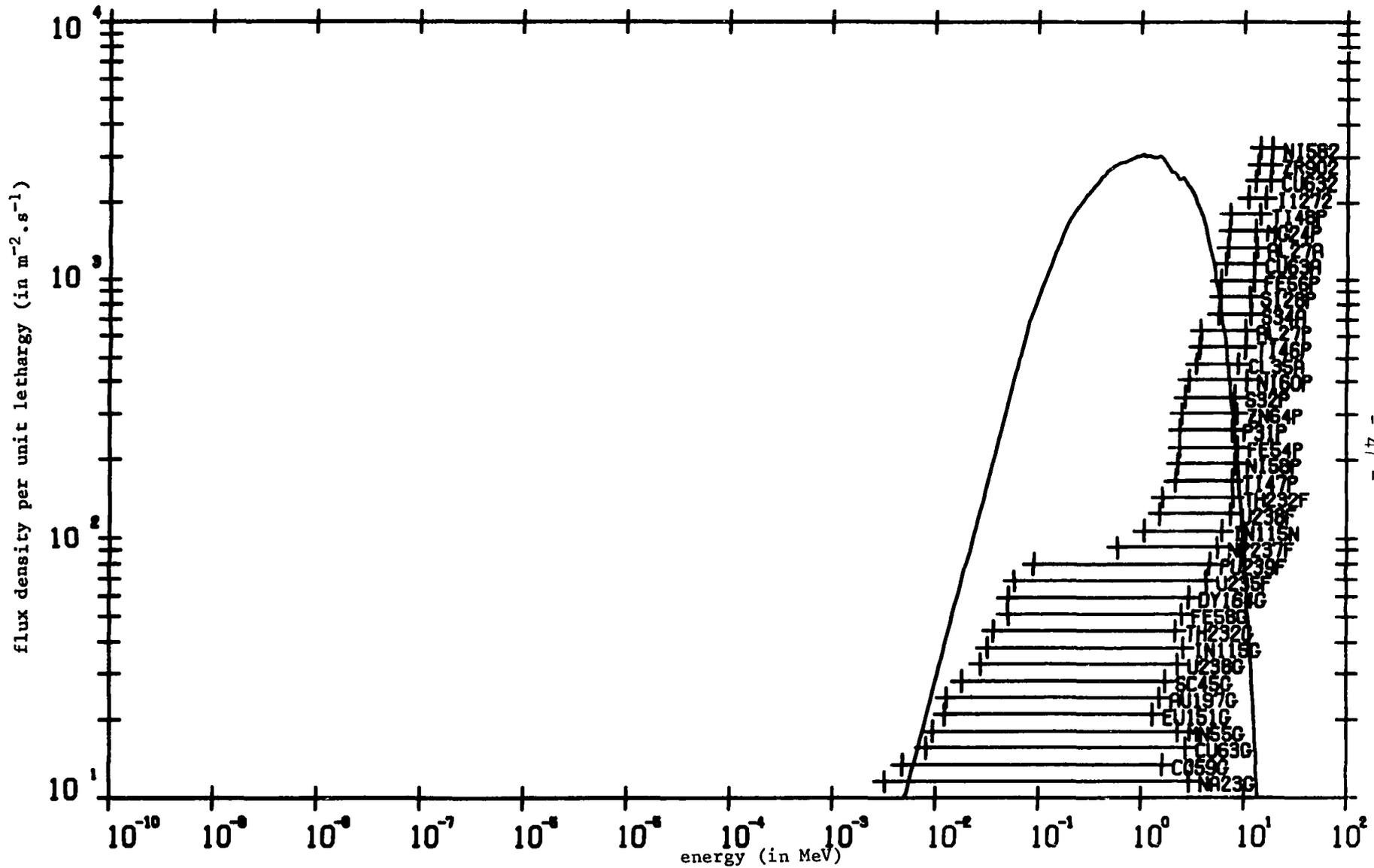
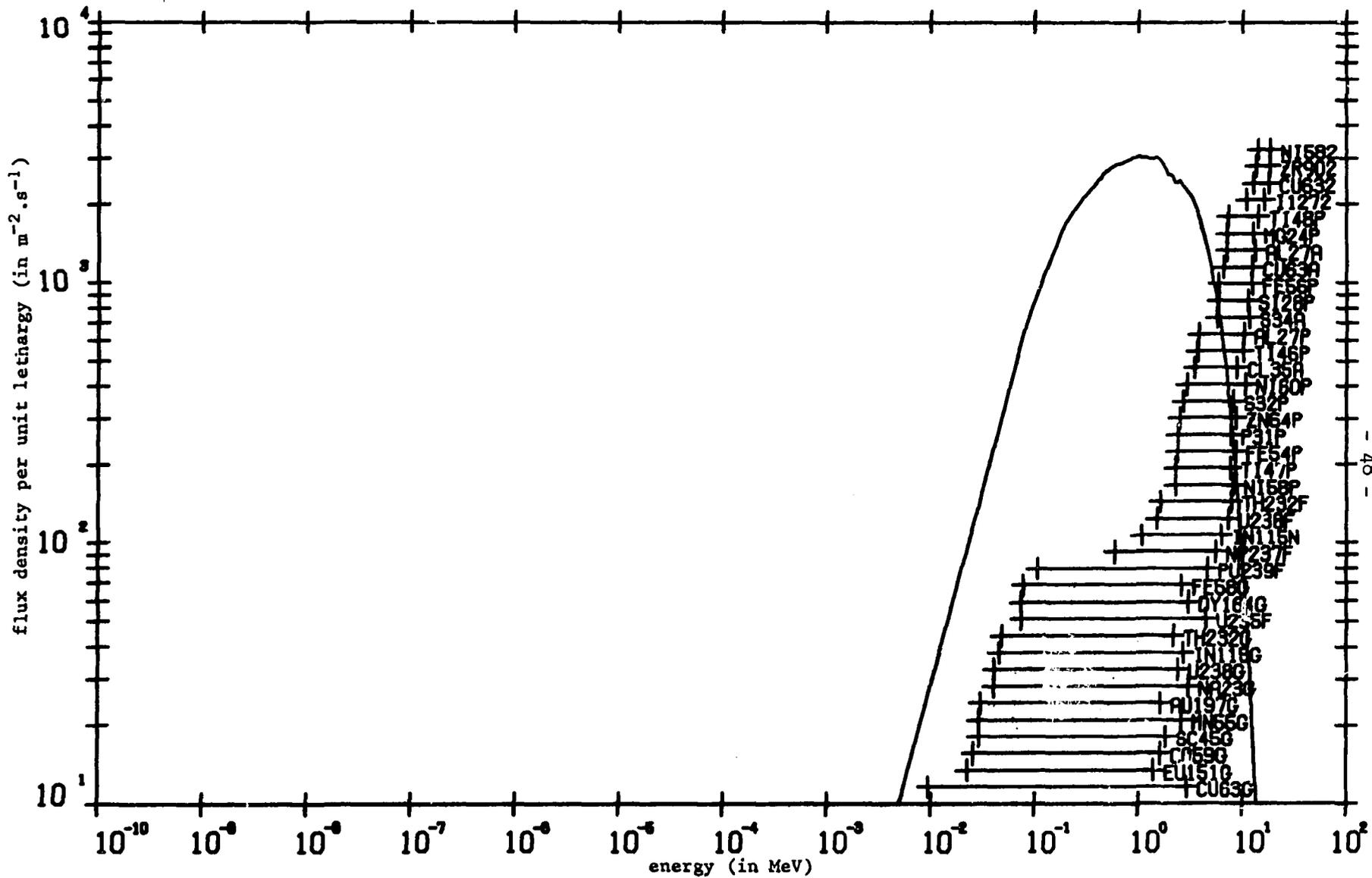
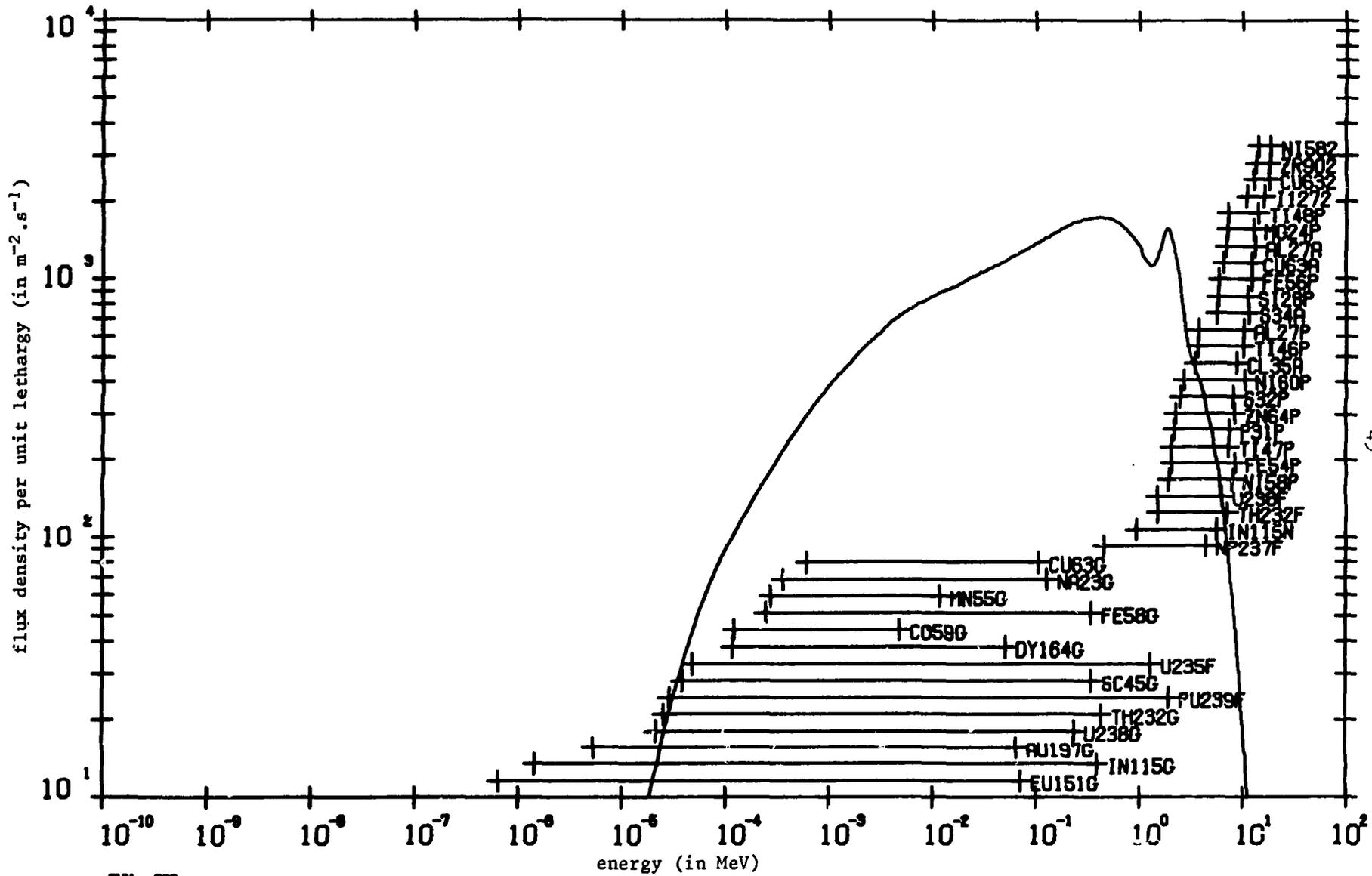


Fig. 5: EBR hole 1 spectrum + fission spectrum below 0.05 MeV and above 3 MeV. Indicated are the 90% response ranges of cadmium covered detectors.



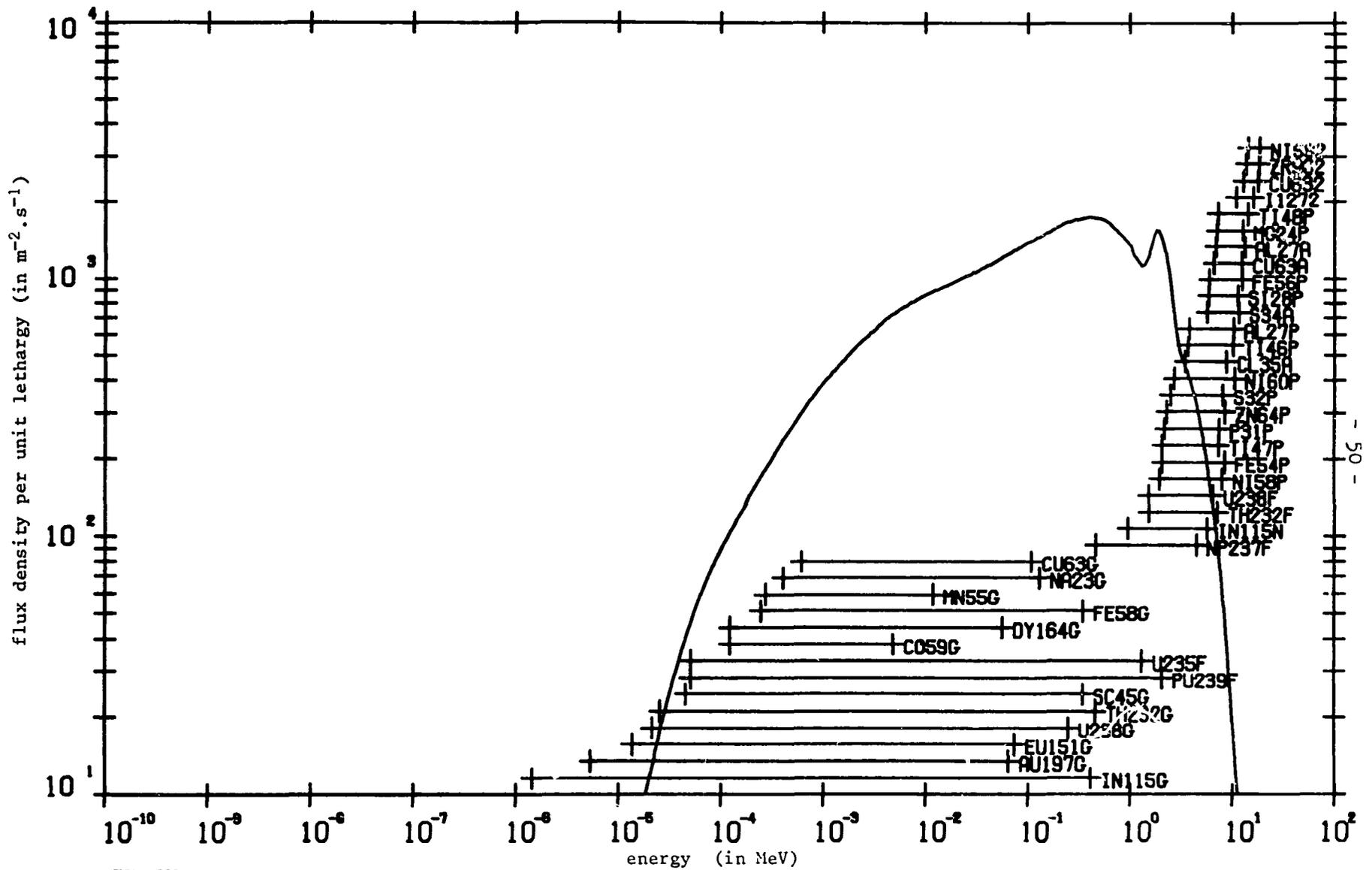
RM 008

Fig. 6: EBR hole 1 spectrum + fission spectrum below 0.5 MeV and above 3 MeV. Indicated are the 90% response ranges for foil covered by a 8 mm cover consisting of 90% <sup>10</sup>B + 10% <sup>9</sup>B.



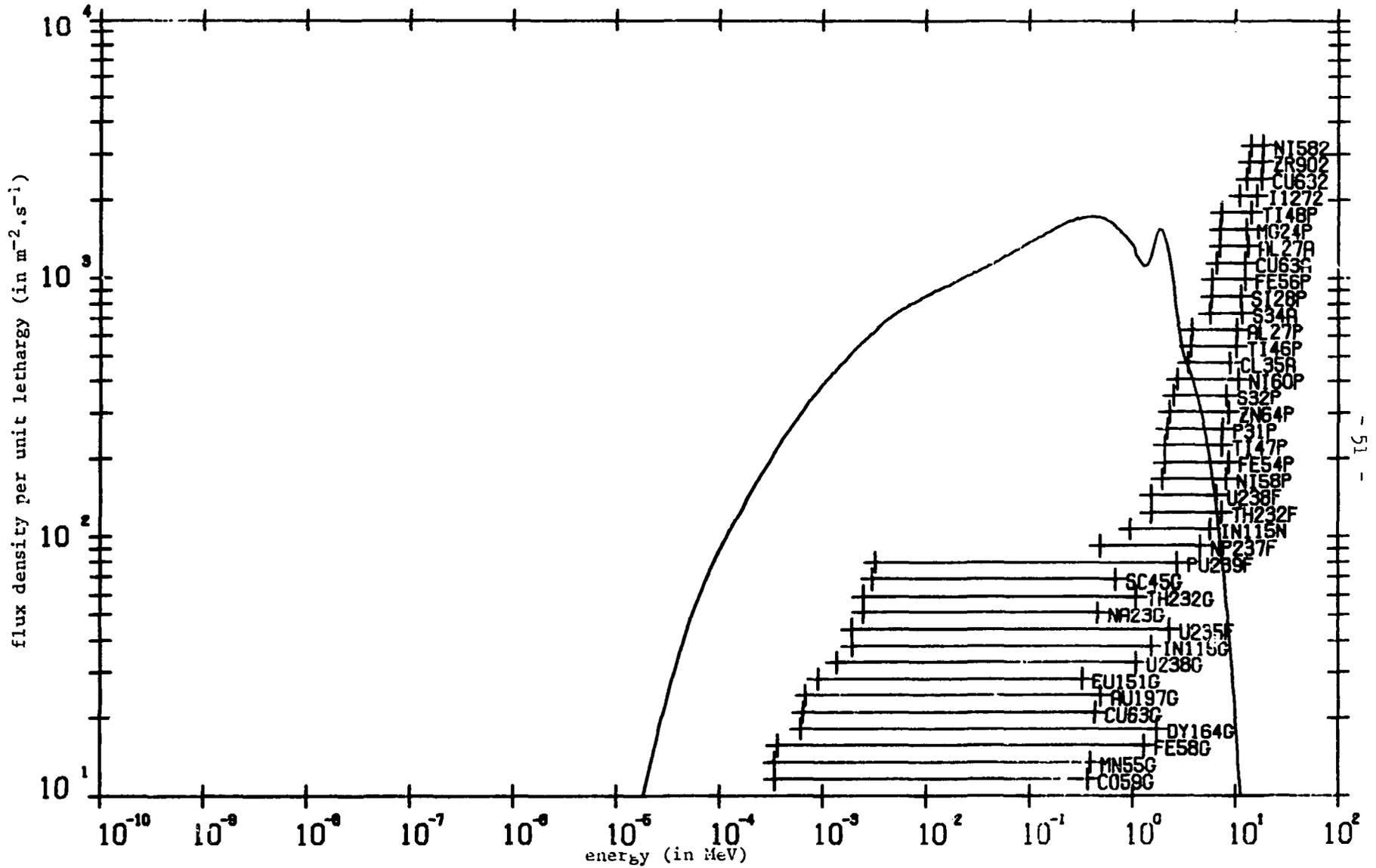
RUN 003

Fig. 7: Calculated ECEL core 14-13 spectrum + Godiva spectrum below 0.00926 eV + fission spectrum above 3 MeV. Indicated are the 90% response ranges of bare detectors.



RUN 003

Fig. 8: Calculated ECEL core 14-13 spectrum + Godiva spectrum below 0.00926 eV + fission spectrum above 3 MeV. Indicated are the 90% response ranges of cadmium covered detectors.

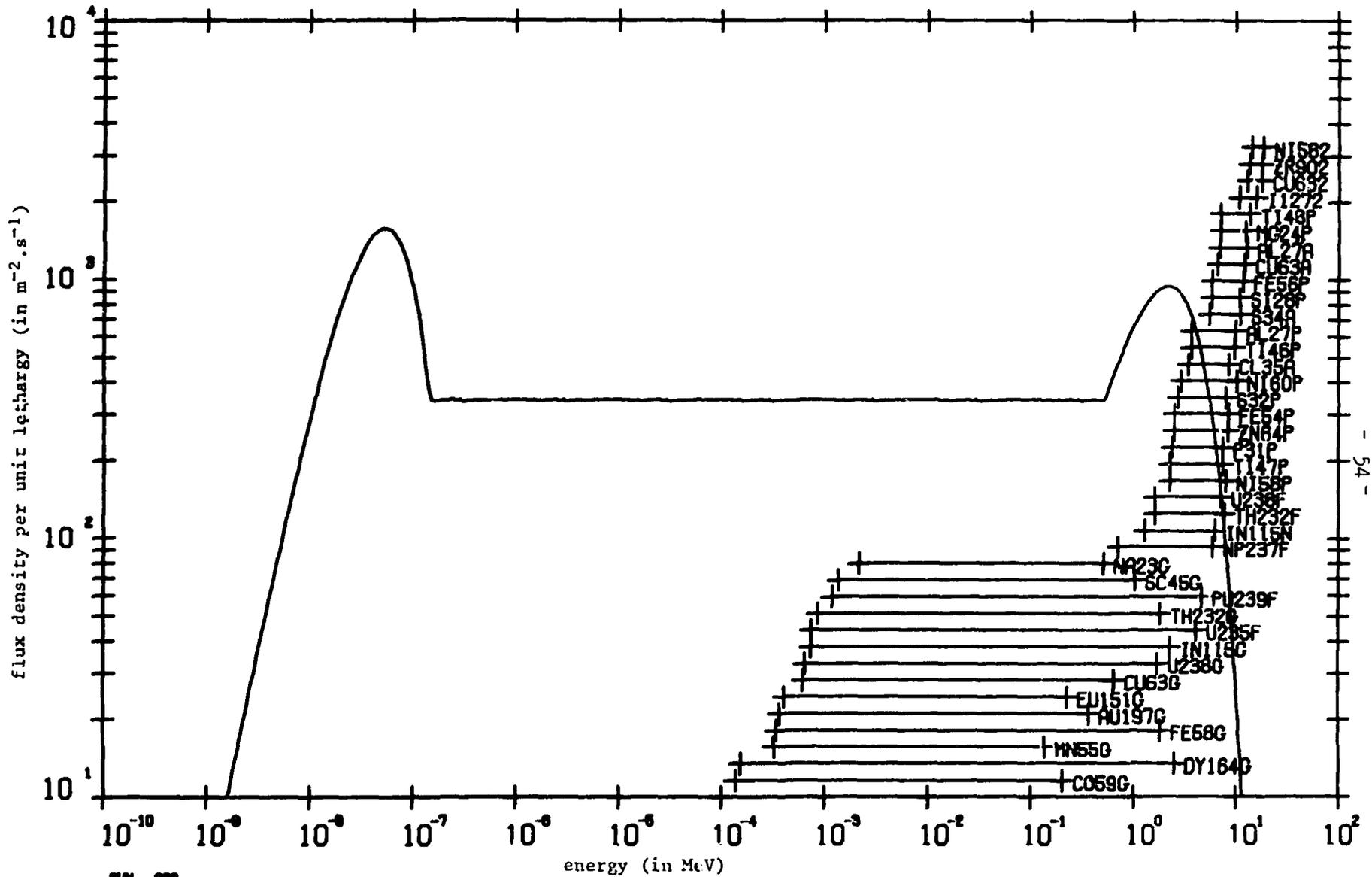


RUN 004

Fig. 9: ECEI 14-13 spectrum + Godiva spectrum below 0.00926 eV + fission spectrum above 3 MeV. Indicated are the 90% response ranges of detectors covered by a 8 mm layer consisting of 90%  $^{10}\text{B}$  + 10%  $^{11}\text{B}$







NUM 002

Fig. 12: 293 K Maxwellian spectrum, matched at 0.14 eV to 1/E spectrum, matched at 0.5 MeV to a Watt fission spectrum. Indicated are the 90% response ranges of detectors covered by a 8 mm layer consisting of 90%  $^{10}B$  + 10%  $^9B$ .

CRITICAL COMPARISON OF SPECTRUM UNFOLDING CODES

R. Dierckx

ABSTRACT

Different unfolding methods of foil activation data are compared with each other. The recommended codes are either SPECTRA (REPETE mode) either SAND II. The errors which affect the measured differential neutron flux are analysed and estimated.

1. INTRODUCTION

Unfolding methods are a useful mathematical tool to get a maximum of information out of a limited amount of foil activation data. The problem arises to derive from integral data the differential neutron spectrum. This problem cannot be solved without any additional knowledge of the differential spectrum to be measured. In most practical cases, theoretical calculations may predict with enough precision the differential neutron spectrum. The problem now is transformed to find deviations from this first approximation of the spectrum to be measured. Different unfolding methods were proposed in the last years, and the most promising ones are confronted with each other. This discussion is based on a paper confronting RDMN, SPECTRA, and SAND II published as information by the "American Society for Testing and Materials" (1) and on the work of myself and my collaborators (2) (3) (4) (5) comparing PARAMETER, RDMN, MESCO, SPECTRA and SAND II.

I have to apologize that these are not the only codes written but they are thought to be the most versatile ones of those available to us.

After a short description of the codes and the results of the intercomparison, an error analysis on SPECTRA and SAND II is added.

As general conclusion may be stated that the recommended codes to be used are SPECTRA (REPETE mode of operation) and SAND II with a slight superiority of SAND II due to its computing velocity and less sensitivity to the trial spectrum.

## 2. DESCRIPTION OF THE UNFOLDING CODES

Due to the nature of this discussion I will limit myself here to a short description of the codes, their solution and critics. A complete list of references is added at the end for those who want a complete knowledge of these codes.

### 2.1. PARAMETER Method (6) (7)

This method is based on two principles : the total energy range is divided in energy bands, and in each energy band a spectral shape with some free parameters is assumed, predicted by theoretical calculations and considerations. The number of parameter is lower or equal the number of detectors.

Insertion of this algebraic expressions into the activation integrals permits to obtain a solution for the free parameters which characterise the unknown spectrum.

The solution is always a physical one, bound however at the mathematical representation utilised. For fast reactor type spectra f.c. de formula of ref.7 are valid. A computer program solve the integral equations by an iterative procedure which starts with first approximation values for the free parameters on which the final solution is not sensitive.

It is not a sophisticated code, its advantage lies in the fact that with only the knowledge that it is a fast reactor type spectrum you find a physically valid solution. The so-obtained spectrum may be used as first approximation or trial spectrum in the more sophisticated codes which follow.

## 2.2. RDMN (8) (9) (10)

In this "Relative Deviation Minimization Method" the unknown flux is approximated by the first approximation of the unknown spectrum multiplied by a series of linearly independent polynomials in energy with unknown coefficients. (f.e. Laguerre polynomials). The number of expansion terms has to be smaller or equal the number of detectors. The solution is found by a least square fit procedure. The solution to be retained depends on the minimization of the least squares activity error function as function of the number of terms, combined with a personnel judgment of physically acceptable spectra (oscillations and negative fluxes are likely to appear).

This method is very influenced by the first approximation of the spectrum utilized and often the retainable solution is not much better than the trial spectrum.

An advantage is the possibility to calculate the error on the differential and integral spectrum due to errors in the cross sections and the activation integrals by a Monte Carlo method.

### 2.3. MESCO (11) (12)

In this method the unknown flux is expressed as the sum of a first order flux approximation with unknown amplitude and of an energy dependent flux deviation.

In as many energy bands as detectors available the mean value of the flux deviation is determined, minimizing the integral of the squared flux deviations and the integral of its squared first derivative. The so-obtained mean flux deviations are transformed into a continuous function using pieces of polynomes of the third order with a continuous first derivative and maintaining the integral fluxes in each energy band.

MESCO shows the greatest sensitivity to the first spectrum approximation and sometimes no acceptable physical solution is found.

### 2.4. SPECTRA (13) (14) (15)

This method uses N flux points with unknown flux value. For our problem the number of flux point is normally much greater than the number of detectors (50 flux points in the original version; 100 point in the version I used). The solution is found minimizing a function including a least squares error term for the differences between measured and calculated detector activities and a least squares error term for the deviations of the solution spectrum from the trial spectrum.

Two operating modes are available, REPETE searches by an iterative procedure the solution until the function to be minimized has reached a certain value, LIMIT searches directly the solution which fits exactly the activation input data.

The LIMIT code of operation can provide and normally does provide unrealistic solutions (oscillations and negative fluxes) due to the fact that the solution found fits exactly (in the least square sense) the experimental data. As a consequence the experimental errors cause unrealistic solutions. It has indeed no sense to fit the measured data to better than their experimental accuracy. For this reason LIMIT mode should never be used.

In the REPETE mode a personnel judgment is necessary, to retain this solution which fits the measured data to within the assumed experimental accuracy, eliminating these solutions which presents unrealistic oscillations. In the REPETE mode an error in the measured activities greater than the assumed accuracy may be detected. It is up to a personnel judgment whether to accept a less good fit solution or to eliminate the bad detector(s) and restart the calculation.

In Ref.15 L.TURI describes an advanced version of the SPECTRA code which should require less computing time than the original version.

The code is not very sensitive to first approximation spectrum. A criterion for the trial spectrum could be formulated like this : The normalized activity ratios\* should lie for all detectors between 1.3 and 0.7.

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The normalised activity ratio is the ratio of the calculated activity in the trial spectrum to the measured activity of the same detector, normalised to the mean activity ratio of all detectors.

This means : a 30% deviation between the normalised calculated activity in the trial spectrum and the measured activity is tolerable. Under these conditions the solution has a great chance to be unique (within the error limits) and does not depend on the trial spectrum.

### 2.5. SAND II (16) (17) (18) (19) (20)

This code used even more flux points than SPECTRA, up to 621 in the original version. The codes changes a trial function by each iteration point by point until the least squares difference between the measured and calculated activation data reached a prefixed limit.

What is said above for SPECTRA, REPETE mode of operation is valid too for SAND II.

It has no sense to fit the measured data to better than there experimental accuracy. Here too a personnel judgment is necessary to retain the solution which is physically acceptable.

SAND II is even less influenced by the trial spectrum than SPECTRA REPETE : Larger deviations may be tolerated between the normalised calculated activities in the trial spectrum and the measured activities.

### 3. INTERCOMPARISON OF THE UNFOLDING CODES

In Table 1 the different codes are compared with each other. We did not include SPECTRA, LIMIT mode of operation, as in our mind SPECTRA LIMIT should never be used. As already mentioned above it has no sense to fit activation data to better than their experimental accuracy.

TABLE 1 : COMPARISON OF THE DIFFERENT CODES

	PARAMETER	RDMN	MESCO	SPECTRA REPETE	SAND II
energy range coverable	thermal to fast (14 MeV)				
computing time***	0.6 m	0.8 m	10 m	5 m 8 m(U)	1 m (U)
solution model	simple	matrix equations (sensitive to ill conditioning)			simple no matrix equations
solution	physical bound to mathematical expressions	not always physically acceptable	sometimes no solution is found	lowest Q* avoiding oscillations and negative fluxes	
Q attainable	moderate bound to solution imposed			as low as one likes to have it	
sensitivity to trial spectrum	not	great	greatest	small	smallest
spectral details	few, bound to solution imposed			moderate bound to broad resolution of detectors	
error in the** integral spectrum	+2-3% relative to the standard spectrum				
error in the** differential spectrum	bound to solution imposed			+5% in each point	

- \*Q is the least squares difference between the measured and calculated activities  
 \*\* in the energy range covered by the detectors for detector activity errors of better than ±2% (intercalibrated in a standard spectra)  
 \*\*\* on IBM 360/65 for a typical case, except(U) on UNIVAC 1108

Comments on Table 1

- 3.1. Each code may treat the whole energy range from thermal to fast. Minor but not substantial modification at the original versions may be necessary (as f.e. changing the weighing function in RDMN, increase of the number of points in SPECTRA, a.s.o.).
- 3.2. The computing time is greatest for MESCO and SPECTRA. SAND II converges very rapidly.
- 3.3. The mathematical treatment is the simplest for SAND II and PARAMETER. The matrix equations made that the other codes are more complex, specially SPECTRA with 100 flux points, and more sensitive to all conditioning of the matrix.
- 3.4. The shape of the solution itself is imposed in the three codes PARAMETER, RDMN and MESCO; only slight and well defined deviations are permitted from the trial spectrum. Only SPECTRA and SAND II have the freedom to change the flux values point by point in order to arrive at a best fit within experimental errors for all detectors. Consequently the Q value, least squares difference between the measured and calculated detectors activities, reachable in SPECTRA and SAND II is as low as one likes to have it.  
For the other codes the Q value obtainable is defined by the solution imposed.  
SAND II is the less influenced by the trial spectrum even much less than SPECTRA and converges faster.  
In RDMN and MESCO the final solution spectrum depends very much on the first approximation spectrum.

3.5. Only SPECTRA and SAND II are able to give moderate spectral details. The number of spectral details does not depend on the number of flux points but is defined by the broad energy resolution of the detectors, the number of detector and how the detectors cover the energy range to be measured.

### 3.6. Errors in the resulting spectra

Unfolding codes are only to be used with activation data with small errors (a few %), obtainable by intercalibration in standards spectra (22). It is clear that with errors of 10% or more (error already present due to the inaccuracy of the cross sections) quasi every trial spectrum fit the measured activation data.

Keeping the relative errors of the activation data one versus the other below 2% the error in the integral of the flux is of the same order, relative to the standard spectrum, in the energy range covered by the detectors. In these conditions the errors in the point fluxes by SPECTRA and SAND II are of the order of  $\pm 5\%$ .

The three other codes results in a mean spectrum shape around which the real spectrum oscillates.

### Conclusion

As a general conclusion I would forward :

1. The codes recommended to be used are either SPECTRA-REPETE, either SAND II.
2. A trial spectrum has to be used for which the normalised activity ratios do not deviate by more than 30% from 1.
3. The obtained results are equivalent for both codes.
4. SAND II is slightly superior due to its single mathematical treatment, computing velocity, and its smaller sensitivity to the trial spectrum.

#### 4. ERROR ANALYSIS ON SPECTRA-REPETE AND SAND II

In the foregoing sections it is discussed how to get a differential spectrum from measured activation data and a trial spectrum. It is important to know the resulting spectrum, but it is as important to know the errors by which it is affected.

For a fast case as RDMN, a MONTE CARLO program permits the calculation of the errors due to the inaccuracy in cross sections and measures activities.

For SAND II a MONTE CARLO program is written permitting a limited calculation of the errors (23). Such codes as SAND II and SPECTRA-REPETE are, due to their nature, not very well adapted for a complete error analysis of each spectrum determined.

D.GUIDETTI made an approximate error analysis on SPECTRA-REPETE based on simulated experiments (5), applied on a practical spectrum measurement with 9 threshold detectors. The generality of the so found error estimation is not proven, but the extrapolation of its found errors to other measured spectra and to broader energy zones is thought to be acceptable.

In a simulated experiment, activation data are calculated in the exact spectrum and it is looked for the differences between the resulting spectrum after analysis of these activation data by the code and the exact spectrum.

These differences are defined as errors. Following sources of error are investigated :

- intrinsic errors due to the mathematical treatment
- inaccuracy of the detector cross sections
- influence of the number of flux points
- errors in the activation data
- influence of the trial spectrum.

#### 4.1. Intrinsic errors due to the mathematical treatment

Each code has two inputs, the activation data and a trial spectrum, and tries by means of mathematics to change the trial spectrum, in order to fit the activation data.

In a simulated experiment without errors in the activation data and a trial spectrum identical with the exact spectrum the resulting spectrum is identical with the exact spectrum. This is obvious. If however the trial spectrum is different from the exact spectrum the resulting spectrum will show differences from the exact spectrum.

In the typical case treated here three deviations,  $p_i(E)$  between the exact spectrum and trial spectrum were investigated and the percentual error on the resulting deviation calculated. Results are shown in Fig.1, 2 and 3.

Between 0.5 MeV and 11 MeV the errors are smaller than  $\pm 5\%$  even for deviations up to 30-40%. Below 0.5 MeV and above 11 MeV the code gives no valuable result. As the information coming from the activation data covers only the energy range 0.5-11 MeV (Fig.4), the code is unable to change the trial spectrum outside this energy range. It may be stated that the code is able to give a good spectrum unfolding with small errors only in the energy range covered by the detectors.

#### 4.2. Influence of the cross section curve as function of the energy

Two detectors were chosen ( $\text{In}^{115}$  and  $\text{Ti}^{46}$ ) and their cross section as function of the energy changed as in Fig.5 and 6. Intercalibration (22) with the perturbed cross sections ( $\sigma'(E)$ ) in the standard spectrum, the difference between the exact spectrum (different from the standard spectrum) and the resulting spectrum is smaller than a few percent and shows as  $\mathcal{E}_1(E)$  in Fig.4.

If however intercalibrated data with  $\sigma(E)$  are analysed in the code with  $\sigma'(E)$ , (this means : no intercalibration in a standard spectrum) the calculated spectrum is wrong by more than 60% at 6 MeV and 35% at 3 MeV. This proves that even with wrong cross sections curves but intercalibration in a standard spectrum, the errors in the resulting spectra are within the limit of 5%.

#### 4.3. Influence of the number of flux point

In Fig.8 the errors calculating with 29 ( $\xi_1(E)$ ) and 45 ( $\xi_2(E)$ ) flux points are shown accepting the solution with 88 flux points as exact. As logic the errors increase with decreasing number of flux points.

#### 4.4. Errors in the activation data

As in our mind, the unfolding codes should only be used when the relative errors on the activation data are small (1-2% obtainable by intercalibration in standard spectra) the activities of the two detectors were changed by 1%. We have chosen Rh and  $Ti^{46}$ . The errors on the calculated spectra are found in Fig.9.

Rh lies on the lower limit of the covered energy range, consequently the error on the calculated spectra lies below 2 MeV but is limited to 3%. The error due to the erroneous Ti activity is spread in a larger region but also limited to 2%.

#### 4.5. Influence of the trial function

Several trial functions were taken, changing from a pure fission spectrum to a combination of a  $E^{-1}$  or a  $E^{-0.55}$  spectrum up to joining the fission spectrum at 2, 3 or 4 MeV. These are reasonable trial spectra for our typical case.

Fig.10, 11, 12 shows the differences between the different resulting spectra. As can be seen the differences do not exceed 5% in general.

#### 4.6. Difference between SPECTRA and SAND II

Although the calculation above is done for SPECTRA some of them were repeated for SAND II and identical results were found.

I will just present here the differences in our case between the spectrum calculated with SPECTRA and SAND II (Fig.13). The two spectra do not differ by more than 5%.

#### CONCLUSION

Under the following conditions :

1. small relative errors on the activation data  
(intercalibration technique)
2. within the energy range covered by the detectors
3. trial spectra for which the normalised activity ratios lies within 1.3 and 0.7.

The unfolding codes SPECTRA (REPETE mode) and SAND II are able to determine the differential flux spectrum to within +5%, and the integral of the spectrum to within 1-2% relative to the standard spectrum.

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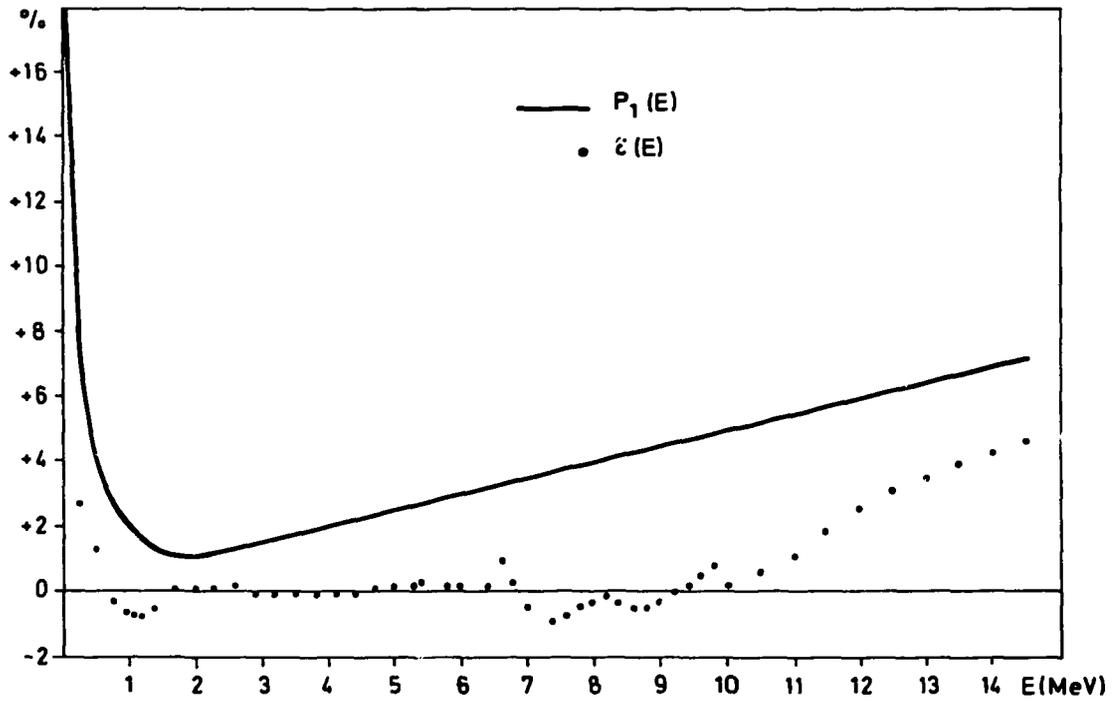


FIG. 1 DEVIATIONS,  $P_i(E)$ , BETWEEN EXACT SPECTRUM AND TRIAL SPECTRUM RESULTING ERROR,  $\epsilon_i(E)$ , ON THESE DEVIATIONS.

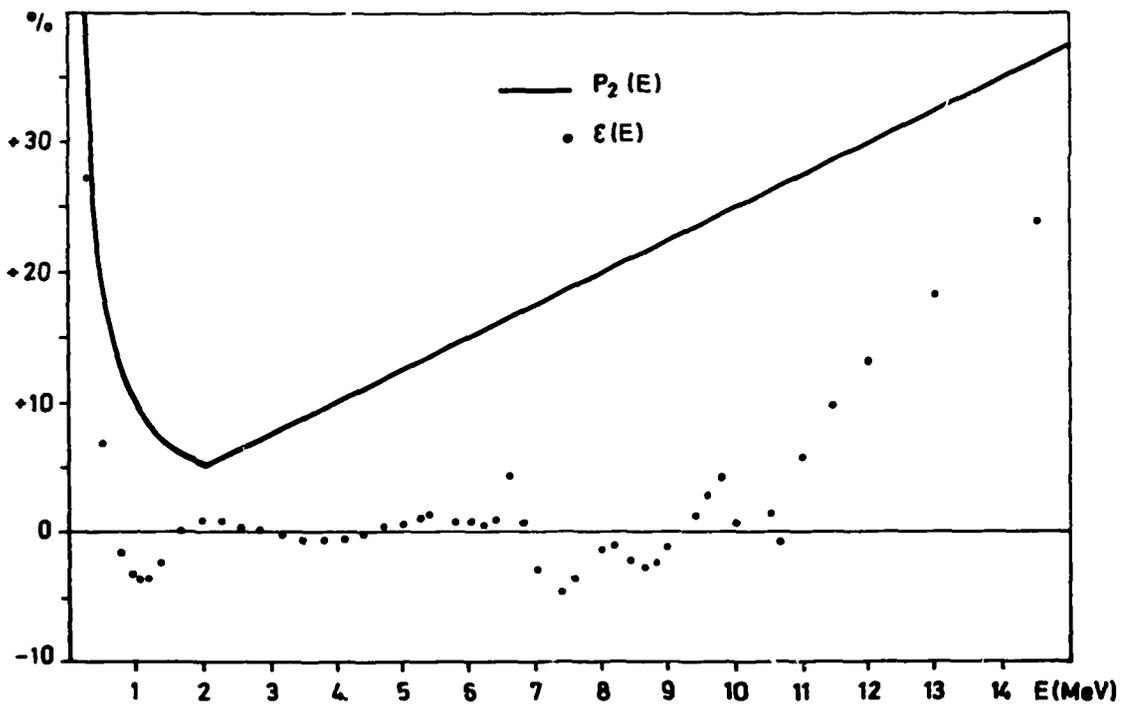


FIG. 2 SEE EXPLANATION UNDER FIG. 1

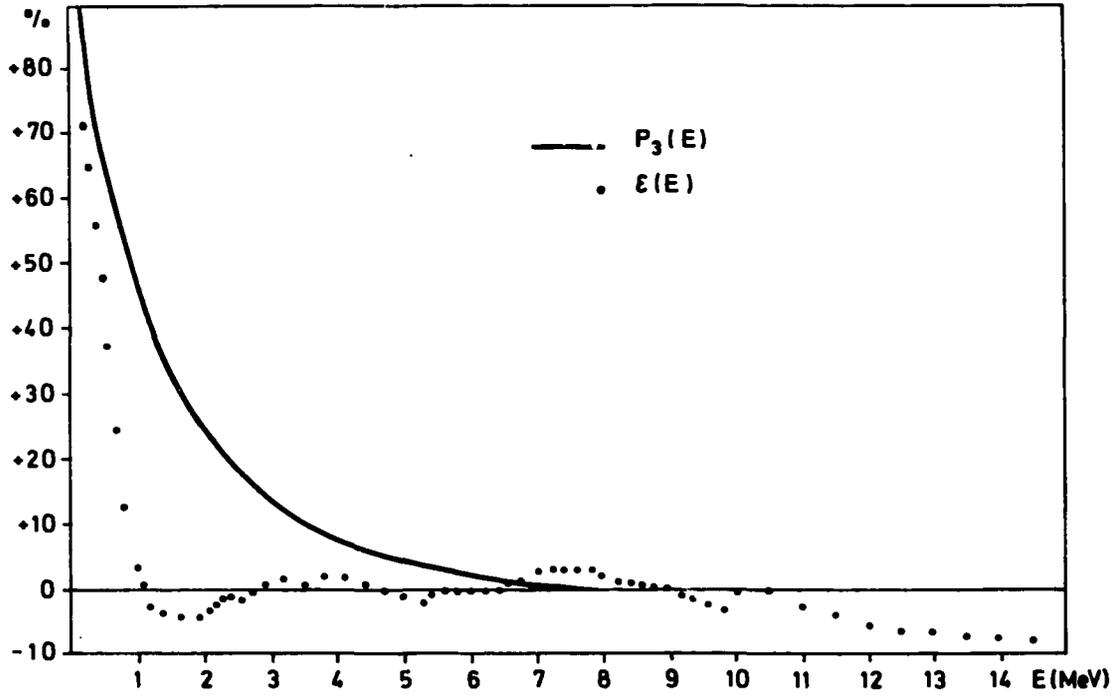


FIG. 3 SEE EXPLANATION UNDER FIG. 1

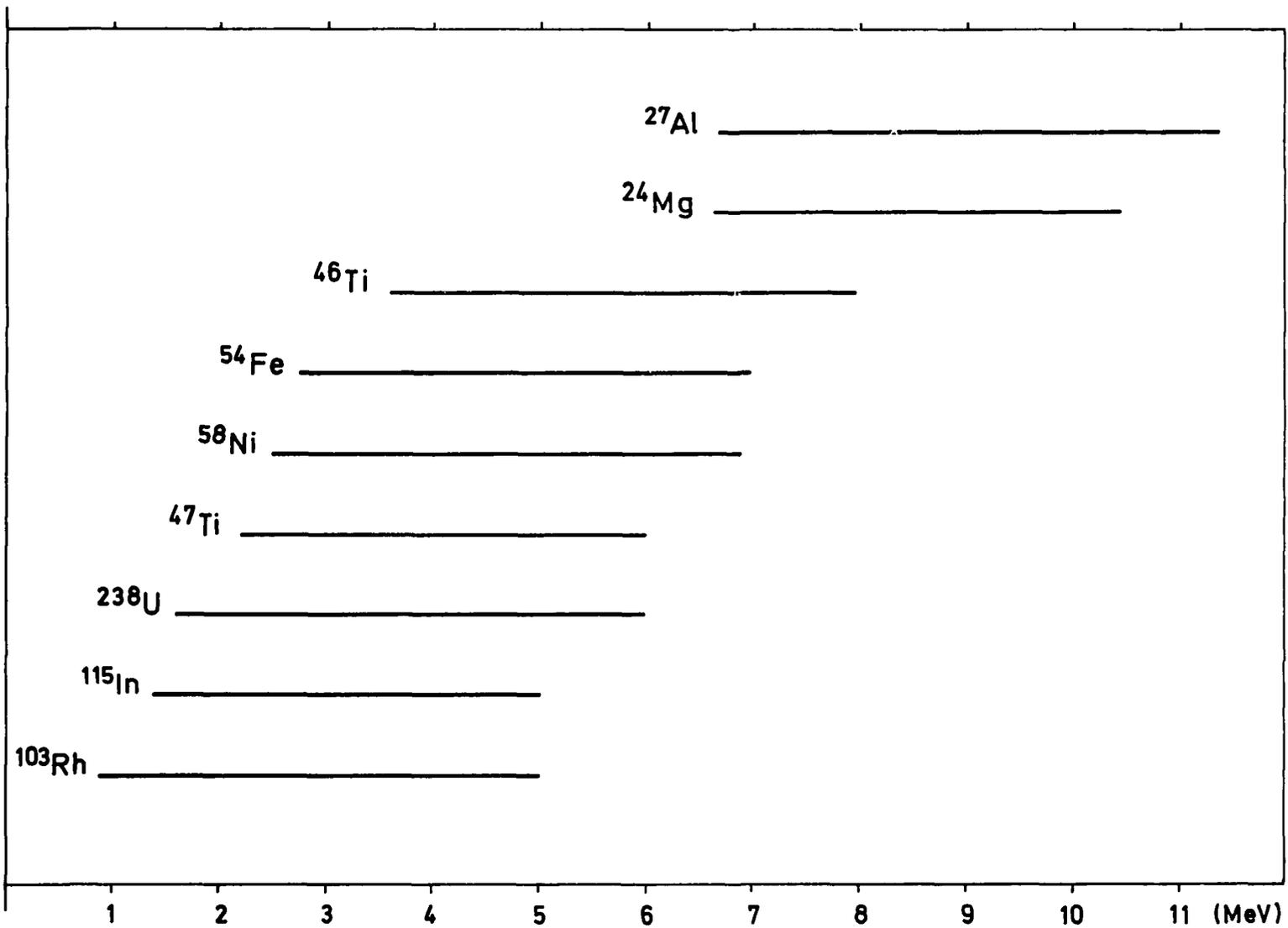


FIG. 4 10-90% ENERGY LIMITS OF ACTIVITY RESPONSE

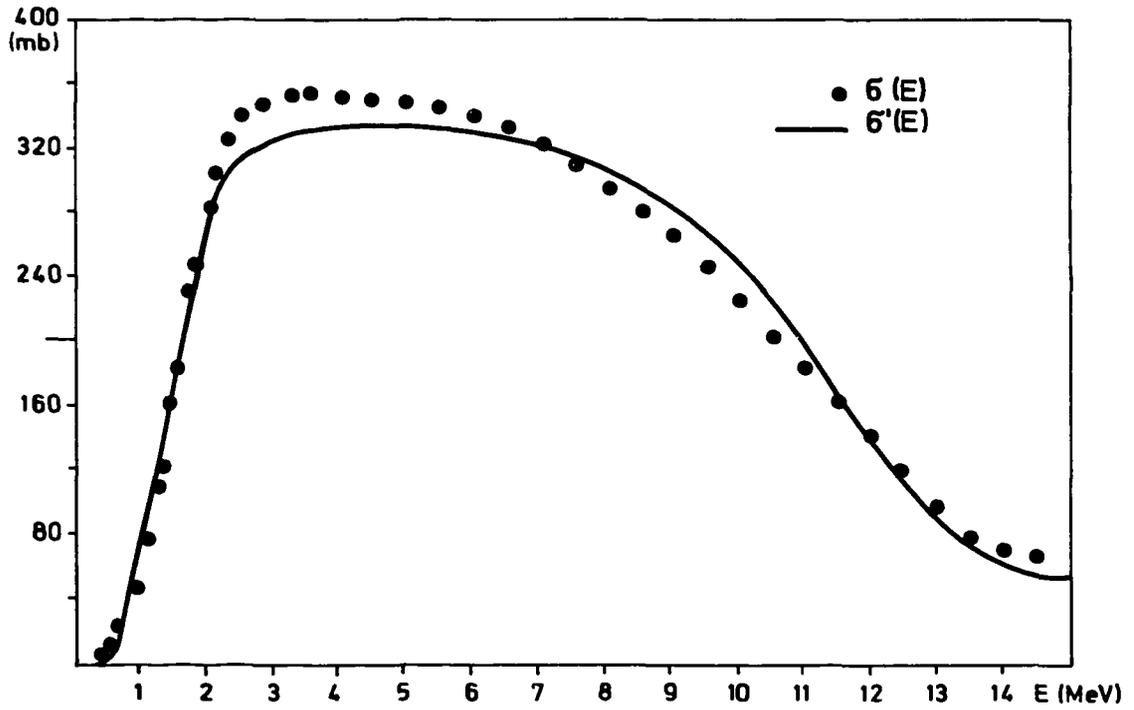


FIG. 5 CROSS-SECTION OF  $\text{In}^{115}(n, n')\text{In}^{115m}$

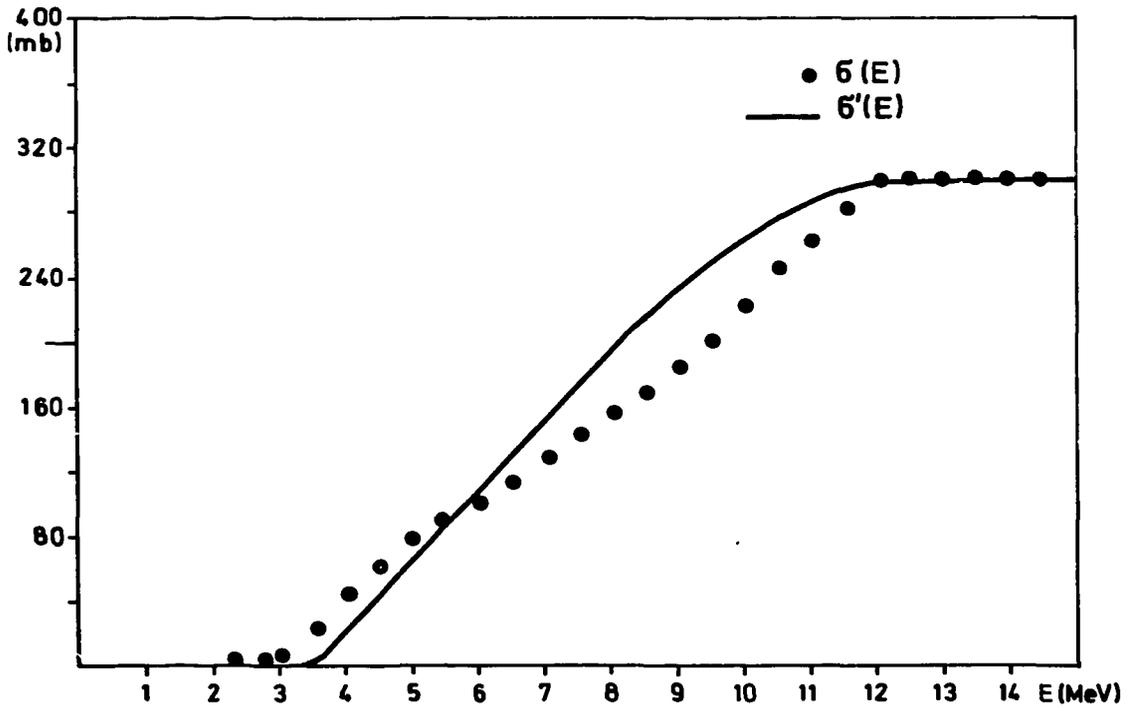


FIG. 6 CROSS-SECTION OF  $\text{Ti}^{46}(n, p)\text{Sc}^{46}$

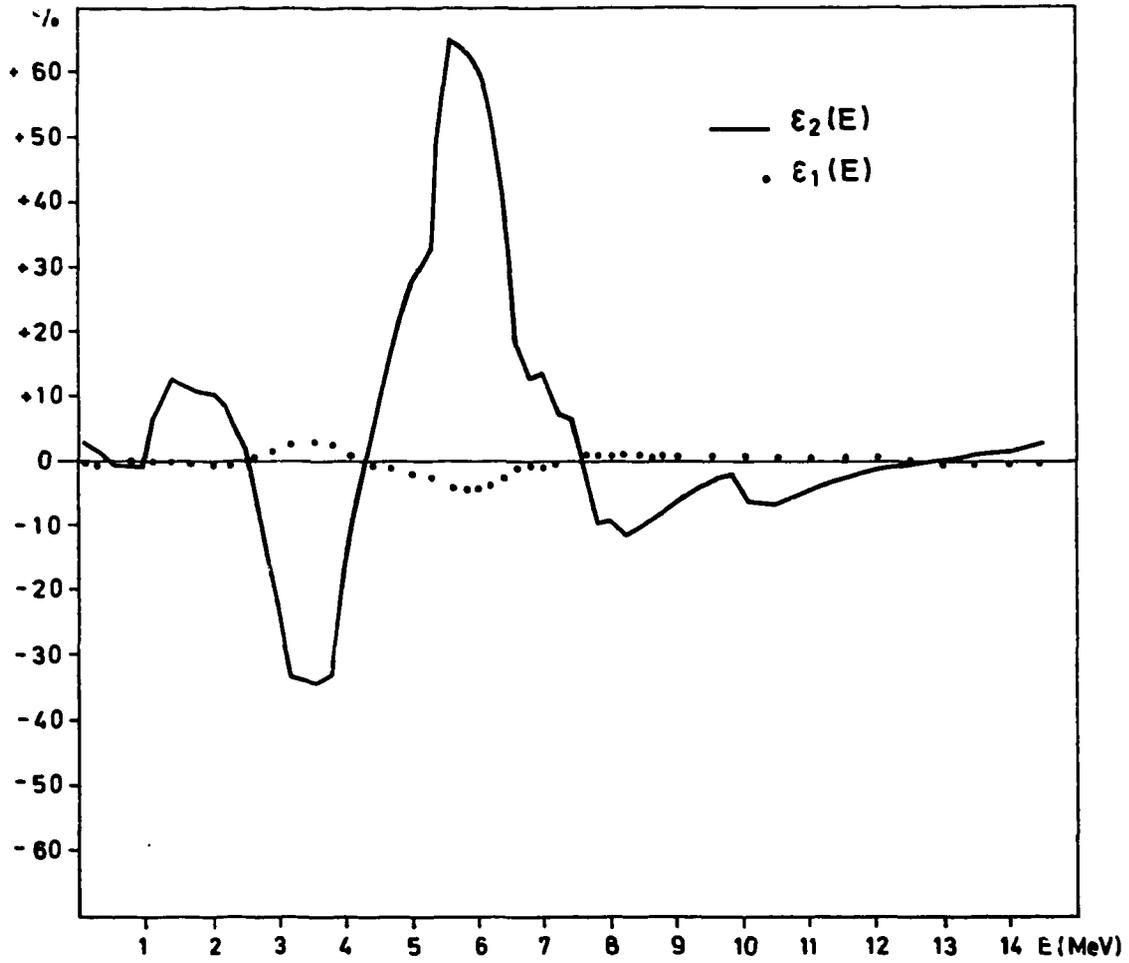


FIG.7 ERRORS DUE TO INACCURACY OF CROSS-SECTION DATA  
 $\epsilon_1(E)$  WITH INTERCALIBRATION  
 $\epsilon_2(E)$  WITHOUT INTERCALIBRATION

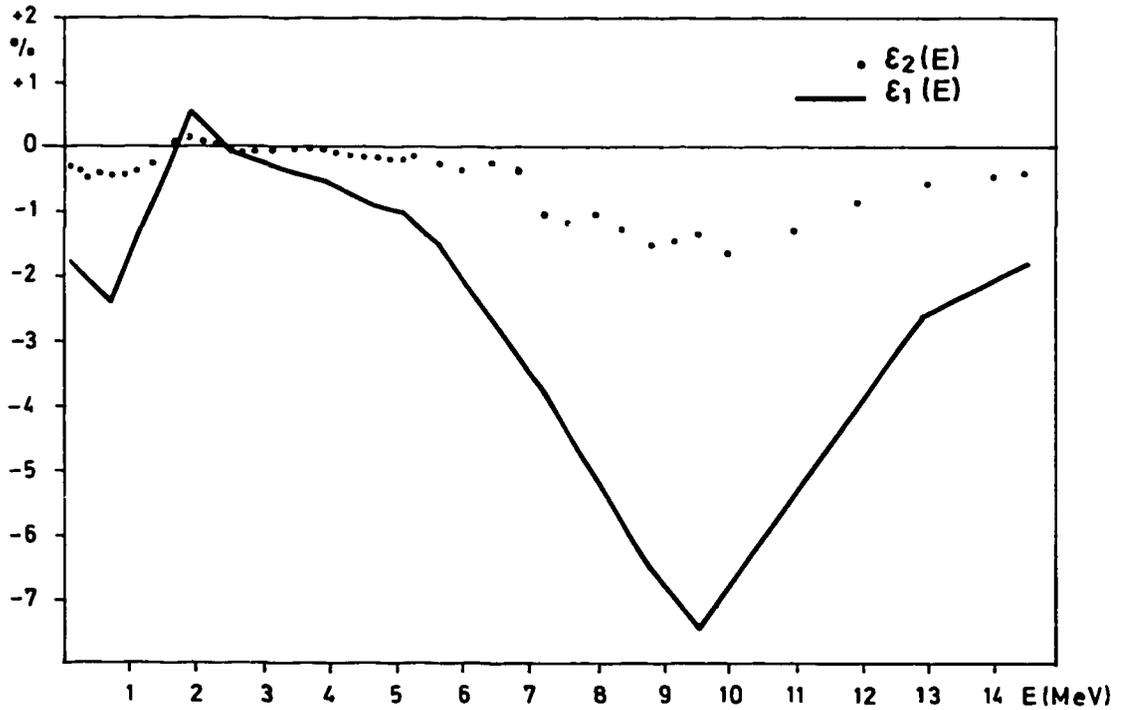


FIG. 8 ERRORS ON THE SPECTRUM AS FUNCTION OF NUMBER OF FLUX POINTS.

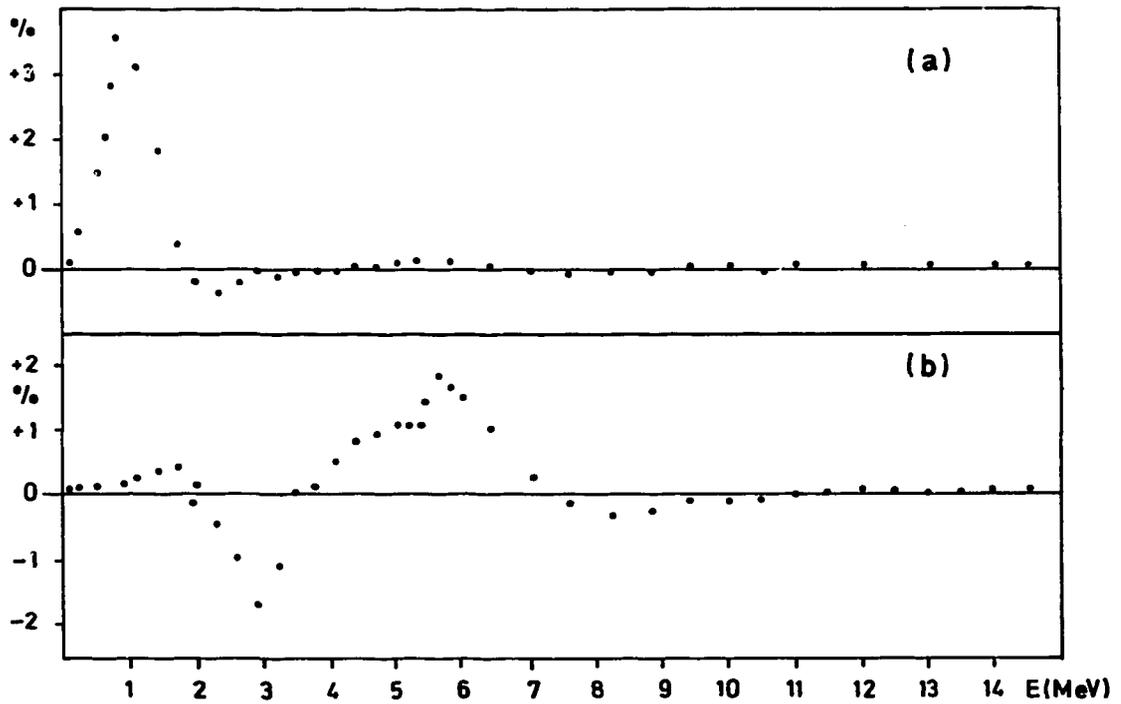


FIG. 9 ERRORS ON THE SPECTRUM DUE TO  
a. RH ACTIVITY INCREASED BY 1%.  
b. Ti ACTIVITY INCREASED BY 1%.

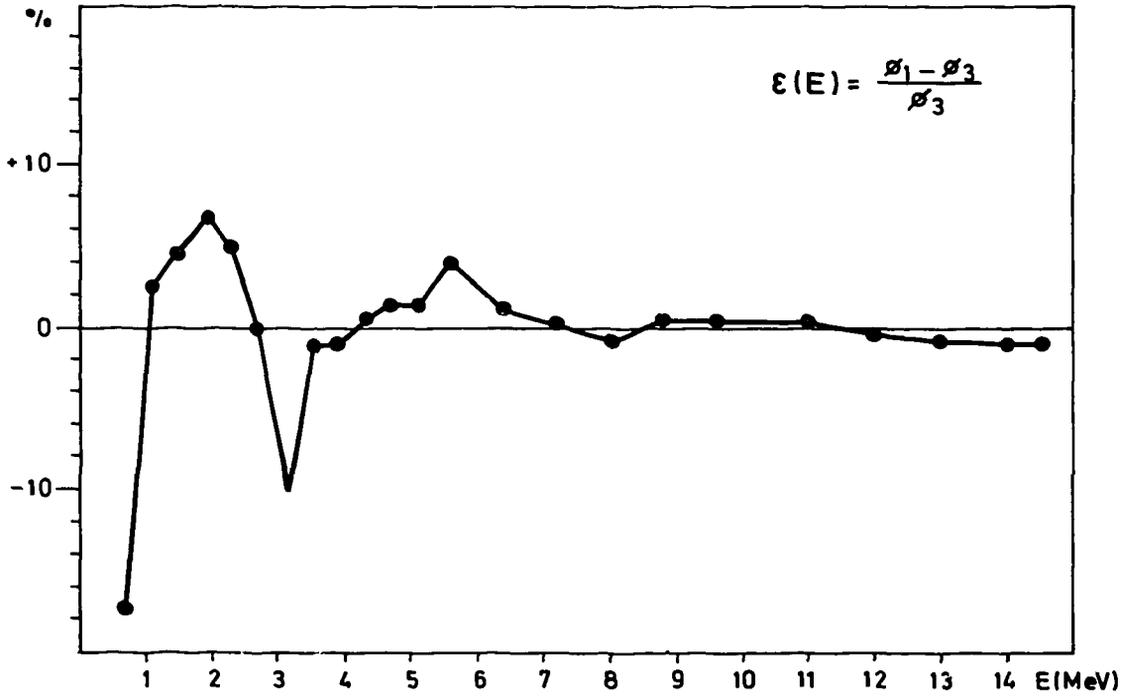


FIG.10 DIFFERENCE BETWEEN CALCULATED SPECTRA STARTING FROM DIFFERENT TRIAL SPECTRA

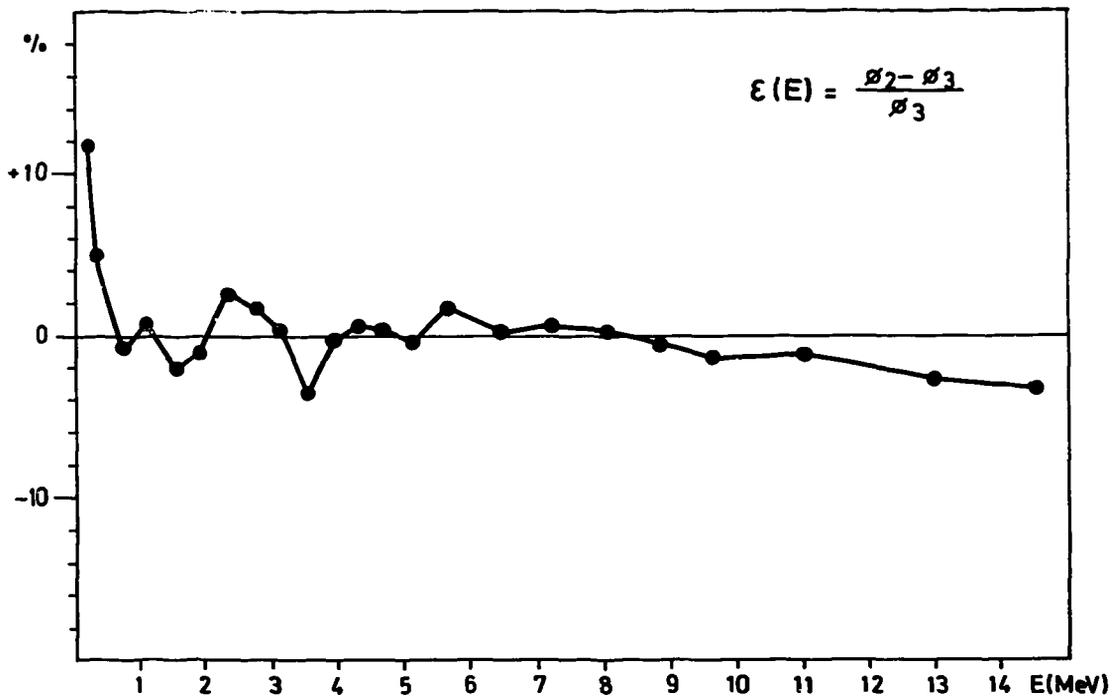


FIG.11 DIFFERENCE BETWEEN CALCULATED SPECTRA STARTING FROM DIFFERENT TRIAL SPECTRA

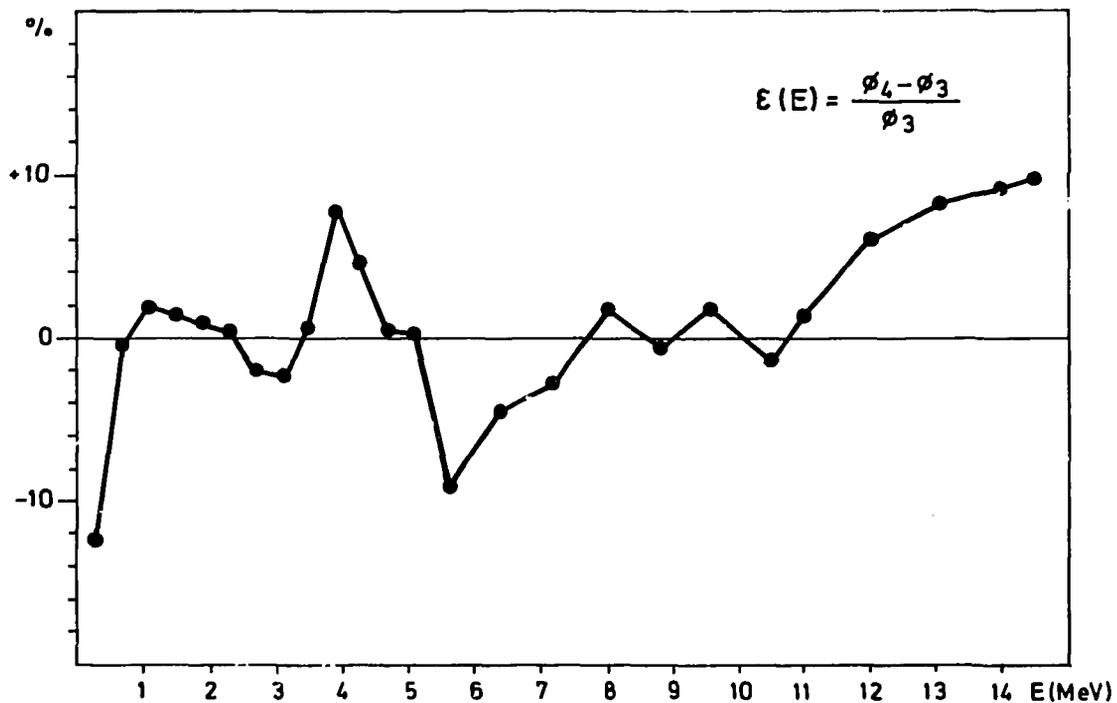


FIG. 12 DIFFERENCE BETWEEN CALCULATED SPECTRA STARTING FROM DIFFERENT TRIAL SPECTRA

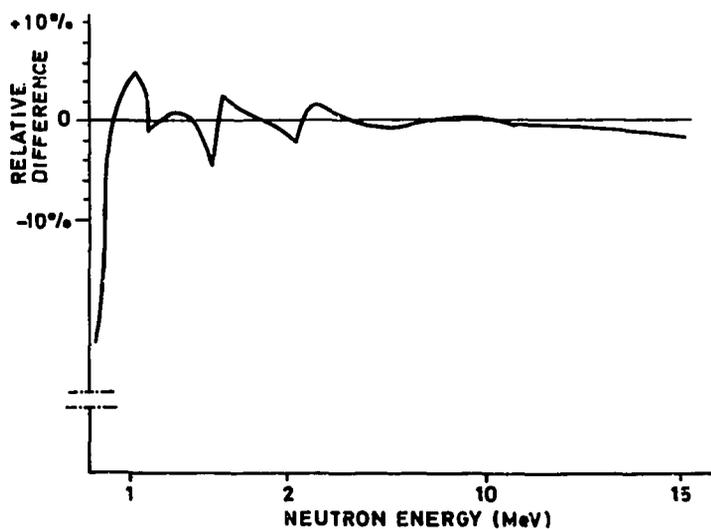


FIG. 13 DIFFERENCE IN THE SPECTRUM AS CALCULATED BY SPECTRA-REPETE AND SAND II.

ROLE OF STANDARD NEUTRON SPECTRA IN  
DIFFERENTIAL FLUX DETERMINATION

R. Dierckx

ABSTRACT

It is discussed in this paper how the intercalibration technique in standard neutron spectra minimizes the errors on foil activation data.

## 1. INTRODUCTION

- Standard neutron spectra are used throughout the world,, from the graphite standard pile, the uranium 235 converters, to more complicated intermediate neutron spectrum standards.
- The graphite pile and the thermal column of a reactor have a well defined thermal neutron spectrum used often for the intercalibration of thermal detectors. The step to the intermediate  $1/E$  spectrum of well moderated reactors is not great.
- In the fast energy range the uranium 235 converter is widely used mostly for the measurement of threshold detector cross sections averaged over a fission spectrum and for the intercalibration of threshold detectors. When known, the core spectrum of a reactor is very useful for these purposes. The use of californium neutron sources is forwarded many times in order to become independent of a reactor. Neutron spectra of the fast reactor type are more difficult to construct. A simple device is a  $B^{10}$  capsule irradiated by plane converter located in a cavity and surrounded by moderating material (1). More complicated is the use of a block of multiplying or non multiplying materials fed by a big converter or the fast leakage flux of a reactor. A sophisticated design is realised by FABRY (2) in a spherical cavity inside the thermal column of a reactor a spherical converter is clad by moderating materials. Standard spectra of the fast reactor type are necessary to intercalibrate detectors which have also a thermal response. Although not new and described several times (3) (4) (5) in literature I feel it necessary to repeat in a meeting like this the advantage of the use of standard neutron spectra for differential flux determination.

## 2. ERROR ELIMINATION BY THE USE OF STANDARD SPECTRA

- For the determination of differential neutron spectra by multiple foil methods it is necessary to know the absolute activity or activity ratios for many foils.
- The absolute counting of foils is time consuming and is not an easy procedure; it is normally affected by rather great errors. Further not all laboratories have at their disposition a specialized team for absolute counting. Secondly the activation integrals  $A_i$  have to be interpreted. We have

$$A_i = \int_0^{\infty} \Sigma_i(E) \phi(E) dE \quad (1)$$

with

$A_i$  : absolute activity of foil i

$\Sigma_i(E)$  : macroscopic cross section of the foil material

$\phi(E)$  : unknown spectrum

- As the differential cross sections are normally affected by error in the order of 10-20% it is clear that they will affect the resulting spectrum  $\phi(E)$ .

The effect on the resulting spectrum due to two main sources of error : absolute counting and inaccurate cross sections are minimized using the intercalibration technique.

- 2.1. As the foils are counted relative to the one irradiated in the standard spectrum, errors due to absolute counting do not effect the measured activities.

The error in the activity determinations in a relative counting may be kept easily to smaller than 1%.

2.2. Error effect of the inaccuracy of the cross sections

The calibration of the detectors is done by irradiating them in a calibration spectrum ( $\Psi(E)$ ). For a foil "i" the efficiency ( $\epsilon_i$ ) of a counter is given by

$$\epsilon_i = \frac{a_i}{z_i} = \frac{\int_0^{\infty} \Sigma_i(E) \Psi(E) dE}{z_i} \quad (2)$$

with

$a_i$  : the calculated activity (dis/sec)

$z_i$  : the counting rate of foil (counts/sec)

Irradiation of the foil in the unknown spectrum ( $\phi(E)$ ), gives a counting rate  $Z_i$  which is related to the activity ( $A_i$ ) by

$$A_i = \epsilon_i Z_i = \int_0^{\infty} \Sigma_i(E) \phi(E) dE \quad (3)$$

Putting (2) into (3) we find

$$A_i = \int_0^{\infty} \Sigma_i(E) \phi(E) dE = \frac{Z_i}{\epsilon_i} \int_0^{\infty} \Sigma_i(E) \Psi(E) dE \quad (4)$$

As the unknown quantity we have  $\phi(E)$  which will be in error due to :

- 1) the errors in  $Z_i$  and  $\epsilon_i$   
(these are counting rates and the relative errors are small)
- 2) the error in the cross section which is normally 10 to 20%.

Rewriting (4) taking into account the error in  $\Sigma_i$  we find

$$\begin{aligned} \int_0^{\infty} \phi(E) \left\{ \Sigma_i(E) + \Delta \Sigma_i(E) \right\} dE &= \frac{Z_i}{\epsilon_i} \int_0^{\infty} \Psi(E) \left[ \Sigma_i(E) + \Delta \Sigma_i(E) \right] dE \\ \int_0^{\infty} \phi(E) \Sigma_i(E) dE &= \frac{Z_i}{\epsilon_i} \int_0^{\infty} \Psi(E) \Sigma_i(E) dE \\ &+ \int_0^{\infty} \Delta \Sigma_i(E) \left[ \frac{Z_i}{\epsilon_i} \Psi(E) - \phi(E) \right] dE \end{aligned} \quad (5)$$

Now, if the spectra are fully equal in shape, the error in the cross section plays no role. If there is a difference in the spectra, the effect of the error in the cross section is diminished by the difference  $(\frac{Z_i}{Z_i} \psi(E) - \phi(E))$  between the two spectra.

Let us write the error integral in another form :

$$\Delta = \int_0^{\infty} \Delta \Sigma_i(E) \left[ \frac{Z_i}{Z_i} \psi(E) - \phi(E) \right] dE = \int_0^{\infty} \frac{\Delta \Sigma_i(E)}{\Sigma_i(E)} \left\{ \frac{Z_i}{Z_i} \psi(E) \Sigma_i(E) - \phi(E) \Sigma_i(E) \right\} dE \quad (6)$$

- The total error in the cross section  $\Delta \Sigma_i(E)$  is composed of two parts

$$\Delta \Sigma_i(E) = k \Sigma_i(E) + \Delta \Sigma_s(E)$$

- $k \Sigma_i(E)$  is proportional to  $\Sigma_i(E)$ . For this part of the error the spectrum difference has no effect since due to the calibration

$$\int_0^{\infty} \left( \frac{Z_i}{Z_i} \psi(E) \Sigma_i(E) - \phi(E) \Sigma_i(E) \right) dE = 0 \quad (7)$$

- $\Delta \Sigma_s(E)$  is due to statistical errors and oscillated around zero. The effect of  $\Delta \Sigma_s(E)$  will not be reduced to zero but due to its oscillatory nature the error integral

$$\Delta = \int_0^{\infty} \Delta \Sigma_s(E) \left\{ \frac{Z_i}{Z_i} \psi(E) - \phi(E) \right\} dE \quad (8)$$

will be small; even for rather great difference between  $\psi(E)$  and  $\phi(E)$ . The errors of 10-20% may be reduced in each case by an order of magnitude, to smaller than 1%.

This is proven by a simulated experiment of a practical case measuring a spectrum with nine intercalibrated threshold detectors (5). From two of them the differential cross sections were changed within the error limits as shown in Fig.1 and 2.

- One calculation was done reintercalibrating the two detectors with the changed cross-sections. The error between the resulting spectrum and the exact one stays within  $\pm 5\%$ , curve  $\xi_1(E)$  on Fig.3, which are the errors due to the mathematical treatment of the code SPECTRA (6) used in this case.
- However not reintercalibrating the two detectors, which means full effect of the cross section inaccuracy, the difference between the exact and the resulting spectrum amounts to 40-60% in the energy zones influenced mainly by the two detectors (curve  $\xi_2(E)$  on Fig.3).

This simulated experiment shows clearly the error minimizing effect of the intercalibration technique.

### 3. CONCLUSION

It is proven in the above discussion that the intercalibration technique is very advantageous for differential flux determination.

This technique permits to keep to relative error in the activation integrals to within 1-2%, minimizing on the resulting spectrum the effect of the errors of the absolute counting, and of the cross section inaccuracy.

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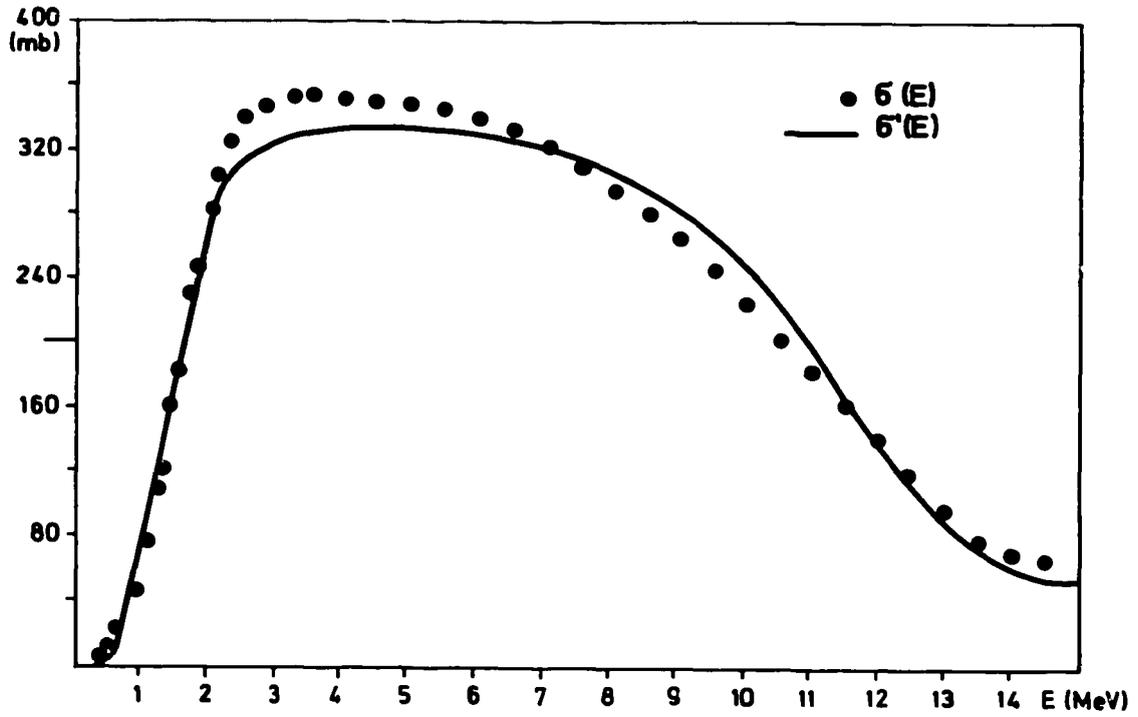


FIG. 1 CROSS-SECTION OF  $\text{In}^{115} (n, n') \text{In}^{115m}$

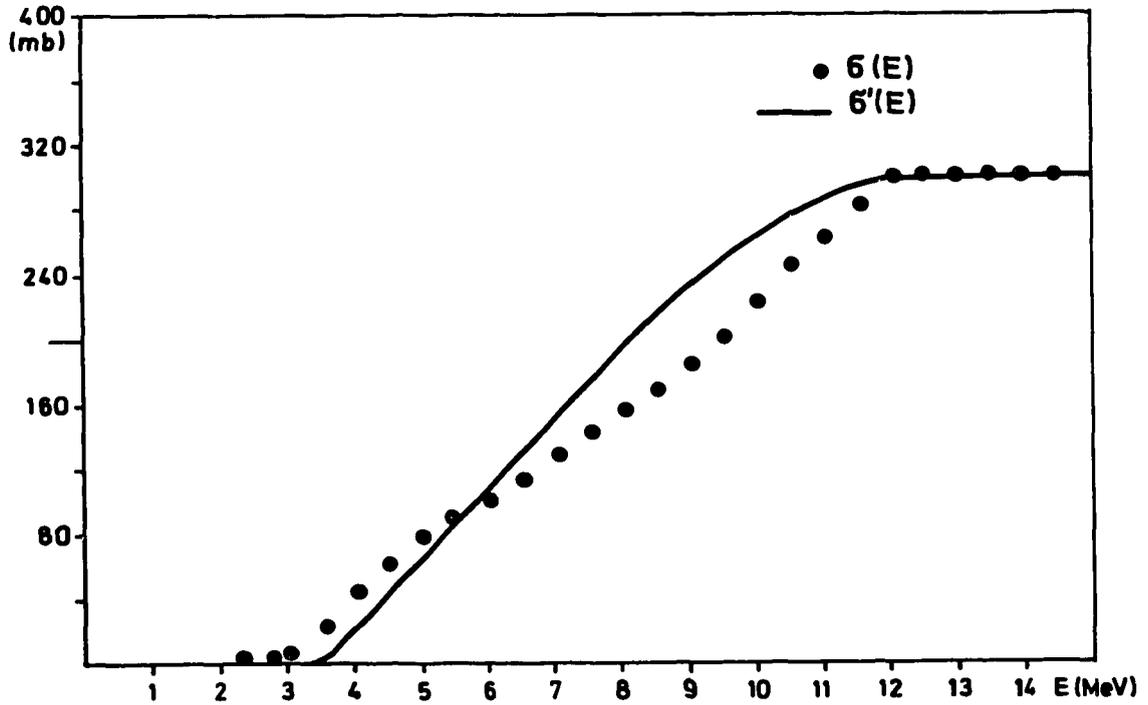


FIG. 2 CROSS-SECTION OF  $\text{Ti}^{46} (n, p) \text{Sc}^{46}$

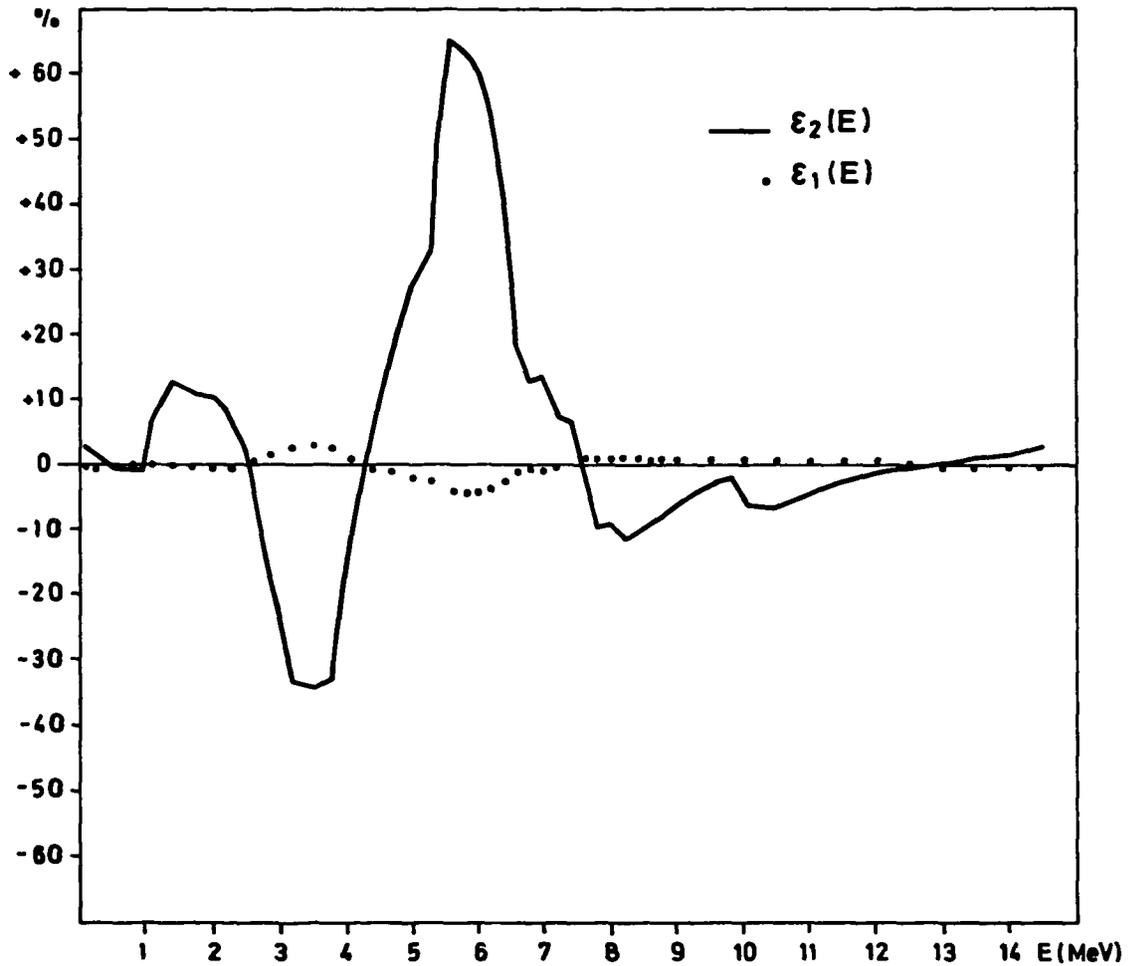


FIG. 3 ERRORS DUE TO INACCURACY OF CROSS-SECTION DATA  
 $\epsilon_1(E)$  WITH INTERCALIBRATION  
 $\epsilon_2(E)$  WITHOUT INTERCALIBRATION

METHODS FOR DETECTION OF RADIATION IN MONITORS  
IRRADIATED BY NEUTRONS

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ABSTRACT

For many purposes the use of threshold reactions still represents the most convenient technique for fast neutron monitoring in a reactor. The use of this type of monitors requires both a convenient method for measuring the activity induced in a sample and sufficiently accurate information concerning the cross section as a function of energy.

The aim of this report is to describe counting methods used in activity determinations.

As a specific example, neutron monitoring using rhodium foils and rhodium activity determinations are described.

## I. Introduction

A well known method to determine the fast neutron spectrum in a reactor is the multiple foil activation technique using  $(n,p)$ ,  $(n,\alpha)$ ,  $(n,n')$  and  $(n,f)$  threshold reactions <sup>(1)(2)(3)</sup>. The neutron spectrum is determined from the measured activities of a set of detectors irradiated simultaneously. The method consists of unfolding a set of Fredholm integral equations of the first order:

$$R_i = \int_0^{\infty} \sigma_i(E) \cdot \varphi(E) dE = R_i(\sigma_i, \varphi)$$

- $R_i$  - experimental saturation activity per atom of the  $i$ th detector  
 $\sigma_i$  - cross section of the  $i$ th detector  
 $\varphi(E)$  - neutron spectrum to be determined

Computer programmes for solving the equations are available <sup>(4)</sup>. These codes need extra physical spectrum information as input, which will to some extent influence the final output spectrum. Inconsistent data will result in a series of peaks and valleys in the spectrum. A good knowledge of the cross section curves and high precision and accuracy of the measured foil activities are required. Such knowledge is needed for the energy distribution of neutrons, the overall instantaneous flux in the specific energy region and the total dose received by samples during various exposure times.

Especially important are the cross section data of such reactions, which are frequently used as reference standards, e.g.  $^{32}\text{S}(n,p)^{32}\text{P}$ ,  $^{58}\text{Ni}(n,p)^{58}\text{Co}$  and  $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ .

Not only flux and spectrum determinations but also most of the cross section measurements, absolute or relative, are performed by the activation techniques.

- (a) If the cross section is desired, the foils have to be irradiated in a known neutron flux:

$$\sigma = \sigma(R, \phi)$$

- (b) If the cross section is known and the neutron flux is desired:

$$\phi = \phi(R, \sigma)$$

In any case, the activity induced in the monitor foil has to be known. The accuracy with which the activity is measured determines to a large extent the accuracy of the results.

## II. Activity determination

One can measure the induced activity by absolute calibration or compare it directly with a known standard source of the same material. In case the calibration of a radionuclide presents special difficulties (no long-lived standard available), the detector efficiency is determined by interpolation between values obtained from the calibration of other radionuclides.

Absolute counting of activities entails knowledge of the overall counter efficiency which involves such factors as:

- (a) Effective solid angle subtended by the counter at the source
- (b) Loss of particles in the source and in the counter window
- (c) Particles which pass through the counter without detection
- (d) Electric pulses which are smaller than a preset discriminator level and do not get recorded

The only routine counting method that has been described which does not require the specific knowledge of these factors is the  $4\pi$  counter. Absolute counting is, however, possible within an error of 1-2% in suitable cases (where there exist easily separable  $\gamma$ -ray peaks) by the  $\gamma$ -ray spectrometer and by the  $\beta$ - $\gamma$  coincidence counter.

At present the determination of radioactivity in some threshold detectors has become relatively easy. Some of the radioactive nuclides are available as standards (average uncertainties are  $\pm 1\%$ ) from national laboratories and from commercial sources.

These standards are calibrated either by  $4\pi\beta$  counting or by  $4\pi\beta$ - $\gamma$  coincidence counting. In all cases the sample used for absolute calibration should have negligible self-absorption<sup>(5)</sup>.

If standard sources can be obtained from specialized laboratories, it does not pay to set up apparatus for absolute calibration at the institute performing the cross section or neutron flux measurements.

For the comparison it is essential that the geometric construction of the monitor and the standard source are as similar as possible so that the counter efficiency remains the same. If the comparison is done directly, the efficiency of the detection apparatus may

fluctuate with time, but have no effect on the results. If the detector efficiency had been determined earlier or no suitable long-lived sources are available, then the efficiency must remain constant. For routine counting, it is very convenient to use standards of long half-lives.

The radiation characteristics of the most used neutron monitors are given in Table 1.

### III. Detectors and counting corrections

Since most of the radioactive nuclides emit both  $\beta$ - and  $\gamma$ -radiation, the advantage of counting only one type of radiation or both together has to be considered. Both types of radiation may be detected by Geiger-, proportional-, scintillation and solid state-counters or ionization chambers. Which type of detector is used depends on the source strength, kind of radiation, purity, stability, etc.

Scintillation counters are preferred because of their high sensitivity to  $\gamma$ -rays, rapid response time and relatively high resolution. The most frequently used scintillation counter is the inorganic crystal of NaI activated with Tl whose high density ( $3,678 \text{ g/cm}^3$ ) and atomic number make it particularly efficient for counting  $\gamma$ -rays. NaI(Tl) crystals have the disadvantage that they are hygroscopic and require a hermetic seal. Since larger and larger Ge(Li) detectors are becoming available, solid state detectors have come into common use for  $\gamma$ -radiation counting. A large Ge(Li) crystal of about  $90 \text{ cm}^3$  has an efficiency of about 20% that of an 3"x3" NaI(Tl) crystal at 1,33 MeV  $\gamma$ -energy. The advantage of the Ge(Li) crystal is its excellent resolution, which is about 50 times better than the NaI(Tl) crystal. It is mostly used for impurity checks of neutron monitors.

Organic materials such as plastic scintillators are used for electron counting and are not hygroscopic. The decay time of plastic scintillators ( $\sim 1-3 \cdot 10^{-9} \text{ s}$ ) is shorter than that of a NaI(Tl) ( $2.5 \cdot 10^{-7} \text{ s}$ ) and their density ( $\sim 1 \text{ g/cm}^3$ ) is lower. However, the plastic scintillators are less efficient in converting the kinetic energy of the electron to light energy than NaI(Tl) crystals.

The  $\beta$ -spectrum from an active source is usually degraded in energy as a result of absorption of the  $\beta$ -particles in the source, the source holder and the counter window.

As the majority of the  $\beta$ 's emitted originate from the outer layers of the source, the count rate will change as a function of the surface area of the active source. Slight variations in shape and surface conditions will cause large changes in counting efficiency. These effects are much smaller when counting  $\gamma$ -rays, and except for low-energy  $\gamma$ - or X-rays, there is a uniform contribution from the whole volume of the source. Also, if flux monitors are used repeatedly, any deterioration in the quality of the source will thus have a smaller effect on the counting efficiency of  $\gamma$ -rays than of  $\beta$  rays.

In  $\beta$  counting, since the  $\beta$  spectrum is degraded it is not easy to use differential spectrometry methods to separate individual activity contributions from two or more active nuclides. If integral methods are used in these cases, the half-life discrimination is more simple than energy discrimination. In contrast, in  $\gamma$ -counting both differential and integral methods can be used.

The necessary correction factors for determination of the disintegration rate of a monitor from the measured count rate are:

- (a) loss of counts due to dead time
  - (b) natural background
  - (c) counting stability
  - (d) geometrical corrections due to counter efficiency and the solid angle subtended by the detector at the counter window
  - (e) absorption of the ionising particles in the detector itself and in the counter window
- (a) The type of dead time which occurs in the counting methods used is the time required by the electronics to recover after registering a pulse. The recovery time normally needed by the scaler is longer than the pulse which enters the scaler, and, therefore, the counting channel will not record any other pulse until the required dead time has elapsed. In scintillation counting with non-overload amplifiers and double delay line pulse shaping where pulse lengths of  $2\ \mu\text{sec}$  are used, recovery time is between  $4-7\ \mu\text{sec}$ . In practice, dead times are determined either by the double-source method<sup>(6)</sup> or by using a double-pulse generator. The relationship between

the dead time and observed count rate is:

$$A_T = \frac{A_0}{1 - A_0 T_D}$$

- (b) If good counting statistics are required, the count rate due to natural background should be small compared with the monitor count rates. Since the major proportion of the background is due to cosmic showers, it is possible if required to isolate this by using coincidence or anti-coincidence counting methods.
- (c) The stability of counting systems is especially important if "spectrum stripping" techniques are to be employed. A pulse height analyzer records the energy spectrum of the radiation. The photo-multiplier, in addition to the amplifiers and PHA, suffers complex gain variations; hence it is necessary to stabilize the gain and zero drifts of the whole spectrometer. This is particularly true when spectra have to be recorded for sources of widely divergent activities<sup>(7)</sup>, especially when high count rates are involved. There are many methods of gain stabilization established, and since 1964 stabilizers are available commercially.

Two further considerations limit the accuracy at high counting rates. These are overlapping of the pulses in time (pile-up) and the fact that the peak amplitude of any pulse will be affected by the average rate of pulses passing through the amplifier (baseline shift). These difficulties are partially overcome with suitable pulse shaping and anti-pile-up circuits<sup>(7)</sup>.

- (d) + (e) Geometric corrections and absorption effectiveness are usually considered under-counter efficiency, which can vary from 1% for a  $\gamma$ -ray spectrometer to nearly 100% for  $4\pi$  counters. Direct calculation of the overall efficiency of a counter is difficult<sup>(8)</sup>.

Another correction which normally comes under this heading arises from the self-absorption in the source. With  $\beta$ -counting and X-ray counting, unless thin electro-deposited sources are used, this correction can be large<sup>(5)</sup>, but for  $\gamma$ -ray counting it is usually negligible.

In  $\gamma$ -counting the loss due to absorption in the counter window is small. When comparison methods are used, it is not necessary to know the counter efficiency. But in all counting methods a fixed

geometry is required to obtain reproducible counting conditions.

Looking at the examples (Table 1), one can see that the activity determinations of  $^{58}\text{Co}$ ,  $^{54}\text{Mn}$ ,  $^{59}\text{Fe}$  are straight-forward.

The monitors are  $\beta$  and  $\gamma$  emitters, and long-lived standards for comparison are readily available.

But for  $^{24}\text{Na}$  and  $^{115}\text{In}^m$  long-lived standards are not available. Here one has to use the indirect comparison method where the efficiency curve of the  $\gamma$ -spectrometer first has to be determined with a set of long-lived standards. By interpolation of the curve one obtains the desired efficiency. The area of the photo-peak must be defined to obtain the induced foil activity. Some conventions for peak definition is needed here.

For sulfur activity determination, there are well established counting techniques described elsewhere<sup>(9)(10)</sup>

#### IV. Neutron monitoring with rhodium foil

Among the various threshold reactions, the  $^{103}\text{Rh}(n,n')^{103}\text{Rh}^m$  reaction would be expected to be the most useful for overall determination of fast neutrons, and as a fast neutron radiation damage monitor<sup>(11)(12)</sup>. This reaction would partially fill the gap between 0.01 MeV and 1 MeV. The threshold energy is much lower than that of other non-fission reactions. The reaction cross section is very large as the majority of the excited levels decay to the 40 keV isomeric state.

But, this reaction still has a series of problems:

- (a) A poor knowledge of the cross section data
- (b) Poorly established decay scheme
- (c) The calibration of  $^{103}\text{Rh}^m$  is difficult
- (d) No activity standards of the same material are available due to the short half-life of  $56,116 \pm 0,009$  min., recently determined by E. Günther<sup>(13)</sup>
- (e) The strong self-absorption of the K-X-rays of about 20 keV within the rhodium foil
- (f) The metal foil cannot be obtained in a pure enough form (99,99). The main impurity is usually iridium which has a high sensitivity to thermal and epithermal neutrons and results in the production of  $^{192}\text{Ir}(T_{1/2} = 72 \text{ d})$  and  $^{194}\text{Ir}(T_{1/2} = 19 \text{ h})$ . Even when its abundance is as low as a few ppm, a correction has to be made for its contribution to the rhodium count rate.

A correction method has been described by Trebilcock<sup>(14)</sup>.

(g) As rhodium is likely to contain unwanted activities from the short-lived ( $T_{1/2} = 4,4$  min.) thermally activated  $^{104}\text{Rh}^m$  and also activities of long half-lives due to iridium, the time of counting of the rhodium activity is very critical. The optimum time for which to count the foils lies between 80-100 min.

The uncertainty 20 to 40% in the differential cross section data and the large discrepancies in the measured fission spectrum averaged activation cross section between 403 and 1100 mb make this reaction unsuitable for use at this time. It seems that the cross section problem is strongly related to the difficulties encountered in the activity determination.

In spite of all the difficulties described above it is important to be able to use this reaction for neutron monitoring. The technique described below shows how one can partially overcome these problems.

As counting equipment a 1-2 mm thick NaI(Tl) crystal with a beryllium window (0,1 mm) is normally used. One has to calibrate the detector efficiency (indirect comparison method) by means of a series of low energy photon radiations with known abundance emitted by calibrated reference sources<sup>(12)</sup>. But, it is not easy to find reference sources for this low energy region. Another problem is how to determine the strong self-absorption of the X-rays within the foil. Geometric efficiency and self-absorption of Rh-activity for a thin NaI(Tl) crystal was recently calculated and verified by experiments<sup>(8)</sup>. Another calibration of  $^{103}\text{Rh}^m$  is possible by making use of the secular equilibrium between the parent  $^{103}\text{Ru}$  which decays (almost 100%) with a half-life of 40 d to its daughter  $^{103}\text{Rh}^m$ .

$^{103}\text{Ru}$  can be standardized by means of the 498 keV  $\gamma$ -ray which is emitted in 88% of the decays<sup>(11)(15)</sup>. To determine the X-ray self-absorption, Rh powder is mixed with irradiated Ru powder and samples of different weight are counted. One could determine the Rh activity in the following way:

$^{103}\text{Pd}$  has a half-life of about 17 days and decays with 99,95% efficiency by electron capture to  $^{103}\text{Rh}^m$  (Fig. 1). Therefore, the K-X radiation of the  $^{103}\text{Pd}$ - $^{103}\text{Rh}^m$  equilibrium and that of  $^{103}\text{Rh}^m$  are equal, and a  $^{103}\text{Pd}$  source is an ideally long-lived reference source for  $^{103}\text{Rh}^m$ . Although the longer half-life of  $^{103}\text{Ru}$  (40 days) is more attractive, the presence of several high energy  $\gamma$ -rays gives disturbingly high background in the region of the 20 keV peak.  $^{103}\text{Pd}$  has a lower background in this region (Fig. 2).

The  $^{103}\text{Rh}^m$  activity induced by neutron irradiation of a foil is obtained by simultaneous irradiation of the foil and soluble rhodium oxide<sup>(16)</sup>. From the solution, aliquot sources are prepared with practically no self-absorption for the 20 keV X-rays. The X-ray emission of these sources is compared with that of calibrated electrolytically  $^{103}\text{Rh}^m$  sources. The very thin  $^{103}\text{Rh}^m$  sources (with  $4\pi$ -efficiency of  $99,5 \pm 0,3\%$ ) prepared for calibration by the  $4\pi$ -X coincidence method were obtained by electro-deposition on a film of  $8 \mu\text{g}/\text{cm}^2$  V.Y.N.S. covered with gold ( $10 \mu\text{g}/\text{cm}^2$ ). Because the mass of rhodium in rhodium oxide source is known, the activity per gram of rhodium is obtained. This activity and the mass of the simultaneously irradiated foil give the rhodium activity  $A_{\text{Rh-foil}}$  present in the foil. The X-ray emission of the foil is now compared with that of the  $^{103}\text{Pd}$  reference source. The  $^{103}\text{Rh}^m$  equivalent which would when contained in the rhodium foil give the same counting rate as the  $^{103}\text{Pd}$  reference source, is

$$A_{\text{eq}} = \frac{N_{\text{Pd-ref}}}{N_{\text{Rh-foil}}} A_{\text{Rh-foil}}$$

where the  $N_i$  are the counting rates.

Because  $A_{\text{eq}}$  decays with the half-life of  $^{103}\text{Pd}$ , this number has to be known very well. Large discrepancies between 17 d - 18,4 d in half-life data of  $^{103}\text{Pd}$  make a new measurement very important (this measurement is presently being made in our lab). As counting equipment, one can use either a 1 mm thick NaI(Tl) crystal with a beryllium window or a  $0,5 \text{ cm}^3$  Ge(Li) crystal with a beryllium window (Fig. 3).

### Conclusions

The activities induced in activation foil detectors are usually determined by  $\gamma$ -ray spectrometry, either with NaI(Tl) crystals or with Ge(Li) crystals. The accuracy of activity determinations is profoundly affected by the knowledge of the decay scheme of the product nuclide formed in the activation detector. Therefore, it is important to make further measurements to extend the precision of the nuclear data.

While these measurements may not be of great interest in the field of theoretical nuclear physics, nevertheless such data are of importance to physicists and technologists studying the phenomena occurring in reactors due to fast neutrons.

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TABLE 1

Radiation characteristics of some detectors used for fast neutron monitoring

Reaction	Nuclear data of produced isotope <sup>(17)(18)</sup>		
	Half-life	Type of decay and energy of detected radiation [keV]	Percentage (of photons) per decay
$^{103}\text{Rh}(n,n')^{103}\text{Rh}^m$	56,116 min	X-ray $E_x = 20$	7,4
$^{115}\text{In}(n,n')^{115}\text{In}^m$	4,5 h	$\beta^-$ , $\gamma$ $E_\gamma = 335$	46,3 - 50
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	71,3 d	$\gamma$ $E_{\gamma 1} = 511$ $E_{\gamma 2} = 811$ $E_{\gamma 3} = 864$	30 99,5 0,64
		EC $\beta^+$ $E_{\beta^+} = 470$	85 15
$^{32}\text{S}(n,p)^{32}\text{P}$	14,3 d	$\beta^-$ $E_\beta = 1710$	100
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	312,6 d	EC $\gamma$ $E_\gamma = 835$	100 100
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	45,1 d	$\beta^-$ $\gamma$ $E_\beta = 1570$ $E_\beta = 1099$ $E_\gamma = 1291$	56 44
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	15 h	$\beta^-$ $\gamma$ $E_\beta = 1390$ $E_\beta = 1369$ $E_\gamma = 2754$ $\gamma$	100 100 100

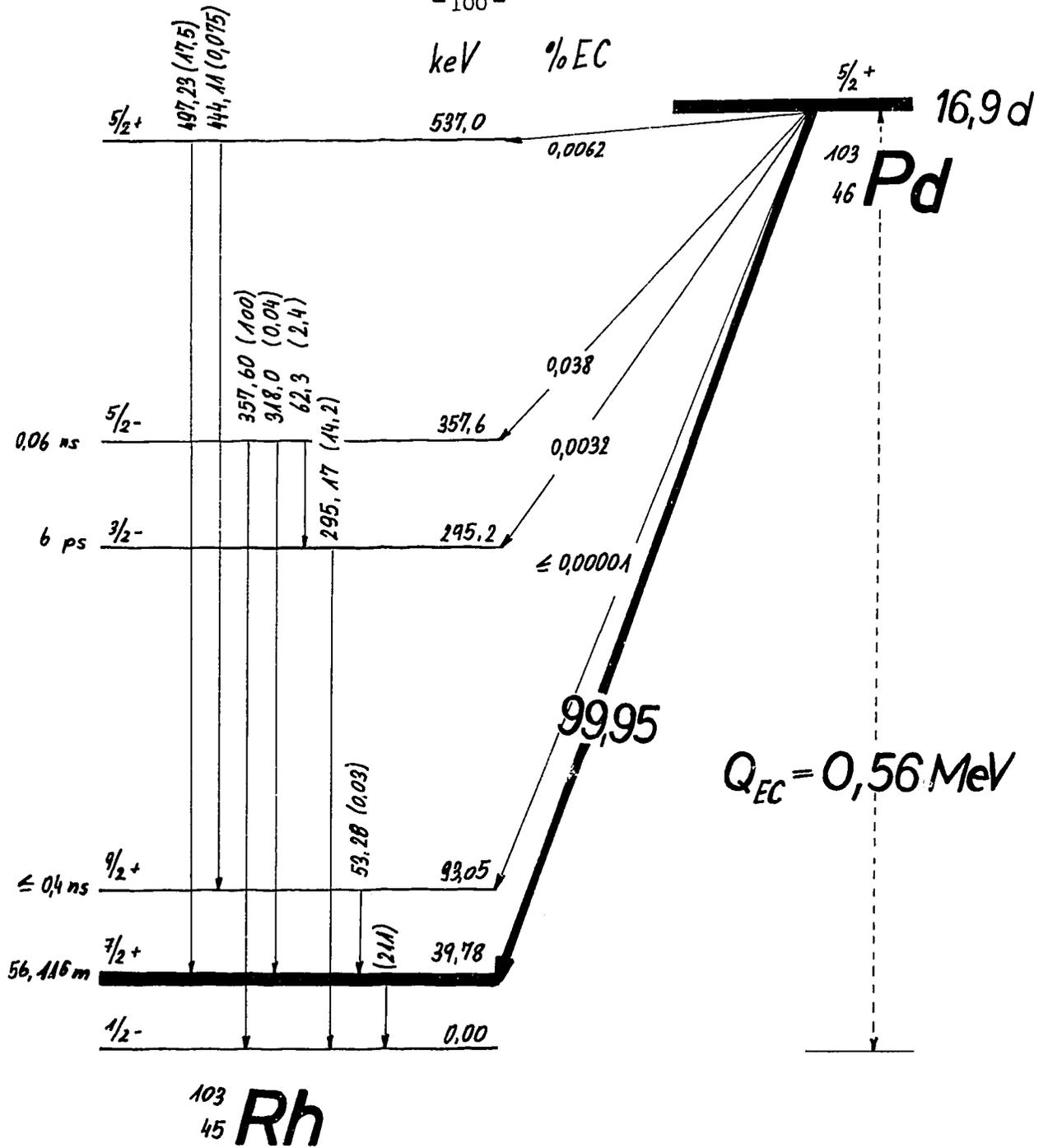


Fig.1. Decay scheme of  $^{103}\text{Pd}$

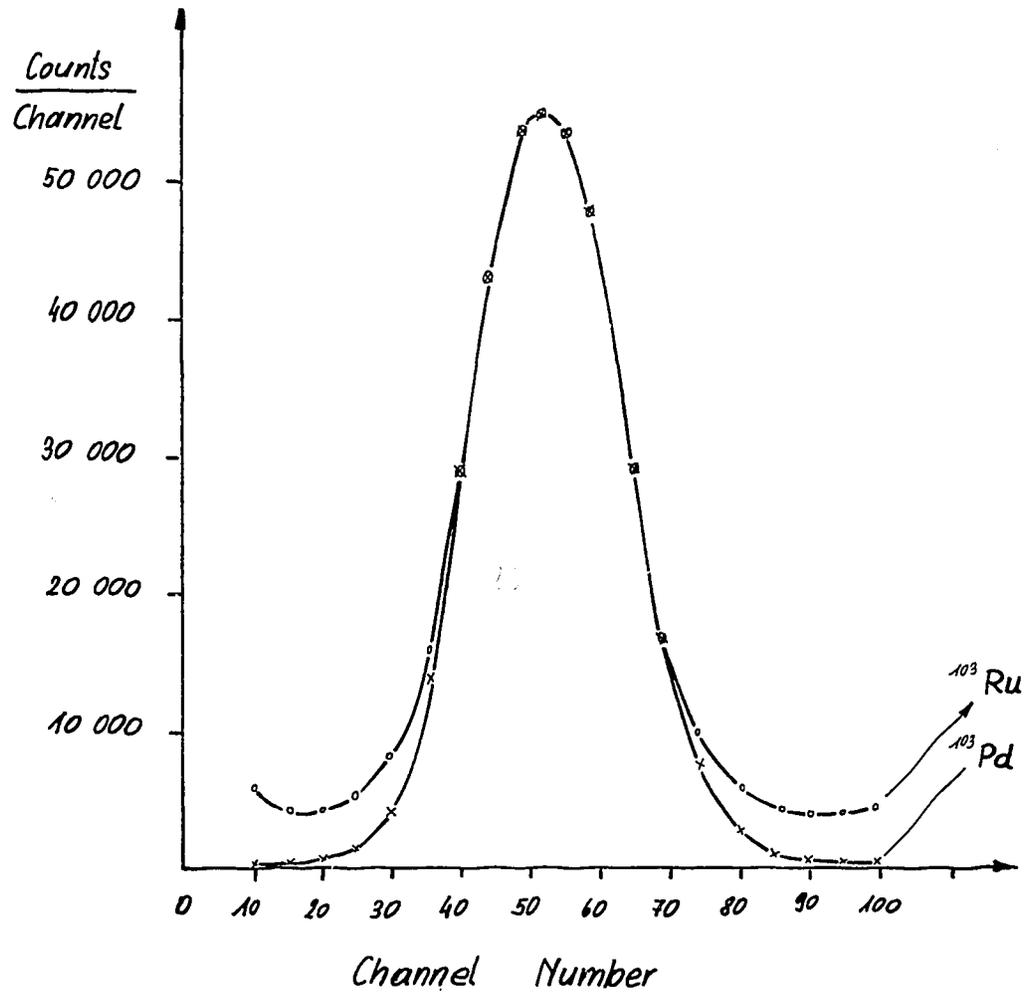
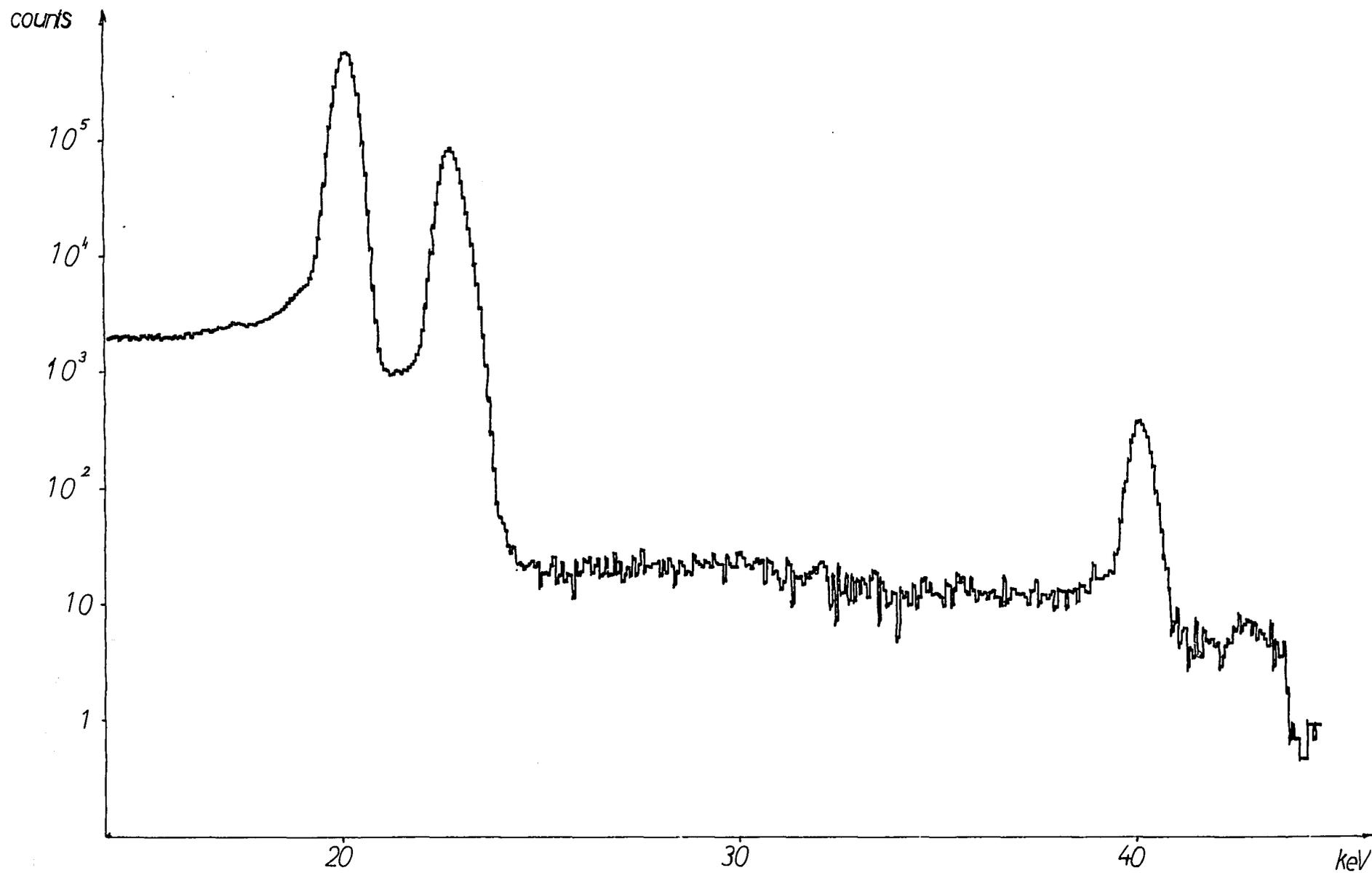


Fig. 2. Pulse height spectra of the  $\sim 20$  keV K-X-rays from

a)  $^{103}\text{Pd}$  (+  $^{103}\text{Rh}^m$ ): —x— line

b)  $^{103}\text{Ru}$  (+  $^{103}\text{Rh}^m$ ): —o— line

registered with a thin NaI(Tl)-(Be-window) detector



**Fig. 3** Photon spectrum of irradiated palladium recorded with an 0,5 cm<sup>3</sup> Ge(Li) counter

IMPORTANT NUCLEAR REACTIONS AND  
NUCLEAR QUANTITIES REQUIRED

Willem L. Zijp

1. INTRODUCTION.

In neutron dosimetry literature about 75 reactions of interest have been mentioned. Within the framework of an introduction to the subject of important nuclear reactions and nuclear quantities required to be treated in this document, it is impossible to make a systematic review of all these reactions.

People interested in "nuclear data for neutron metrology" can be referred to a paper [1] with this title, which has been presented at the IAEA Symposium on Applications of Nuclear Data in Science and Technology, held in Paris from 12-16 March 1973.

A compilation [2] of evaluated cross section data of interest to neutron metrology is distributed during the Consultants' meeting, and will soon be available as RCN report.

In this contribution some remarks are made on nuclear data problems in a few categories.

2. DISCREPANCY IN  $\langle\sigma\rangle^f$  VALUES.

When we compare evaluated average cross sections for a fission neutron spectrum, one observes some large discrepancies. The following reactions, taken from reference [1], show a discrepancy larger than 10% between integral experimental value (as given by Fabry) and the integral calculated value (as derived from SAND-II cross section library):

reaction	discrepancy in $\langle\sigma\rangle^f$
$^{32}\text{S}(n,p)^{32}\text{P}$	13%
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	15%
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	>25%
$^{55}\text{Mn}(n,2n)^{54}\text{Mn}$	>25%
$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	>25%
$^{63}\text{Cu}(n,2n)^{62}\text{Co}$	>25%
$^{64}\text{Zn}(n,p)^{64}\text{Cu}$	20%
$^{93}\text{Nb}(n,2n)^{92}\text{Nb}^m$	>25%
$^{127}\text{I}(n,2n)^{126}\text{I}$	>25%
$^{232}\text{Th}(n,f)$	15%
$^{238}\text{U}(n,f)$	15%

### 3. THE (n,n') REACTIONS [1].

Since (n, $\gamma$ ) reactions have their main response in the thermal energy range and to a lesser extent in the intermediate energy region below 1 keV, and since (n,p), (n, $\alpha$ ) and (n,f) reactions are normally threshold reactions with a threshold above 1 MeV, there are often gaps in the response in the region between say 0.01 MeV and 1 MeV.

To a certain extent (n,n') reactions might be used to partly fill in the gaps, but these reactions have a series of problems:

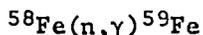
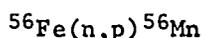
- a poor knowledge of the cross sections;
- not very well established decay schemes (half-life and gamma abundances);
- the difficulty of measuring absolutely low energy gamma rays and X-rays (i.e. from  $^{103}\text{Rh}^m$  and  $^{93}\text{Nb}^m$ ).

The low effective threshold make these (n,n') reactions very attractive for neutron spectrum characteristics.

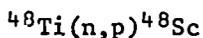
### 4. MULTI-RESPONSE DETECTORS.

Sometimes more than one reaction of interest is occurring in the detector. In order to minimize the number of foils needed for the spectrum unfolding technique, it might be advantageous to apply multi-response detectors.

Examples are:



and:  $\text{Ti}(n,x)^{46}\text{Sc}$ , comprising  $^{46}\text{Ti}(n,p)$ ,  $^{47}\text{Ti}(n,d)$ ,  $^{47}\text{Ti}(n,np)$



### 5. ACCOMPANYING REACTIONS.

When more than one reaction occurs in a detector, there is often only one reaction of main interest. Although in principle such a detector may be a multi-response detector, in practice the accompanying reactions may perturb the response for the main reaction, especially when gross counting methods (integral counting, ionization chambers) are applied.

Examples:

Main reaction	$^{58}\text{Ni}(n,p)^{58}\text{Co}$	$(T_{1/2} = 71.3 \text{ d})$
Accompanying reactions	$^{58}\text{Ni}(n,2n)^{57}\text{Ni}$	$(T_{1/2} = 36.0 \text{ h})$
	$^{58}\text{Ni}(n,\alpha)^{55}\text{Fe}$	$(T_{1/2} = 2.60 \text{ a})$
Main reaction	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	$(T_{1/2} = 5.27 \text{ a})$
Accompanying reactions	$^{59}\text{Co}(n,p)^{59}\text{Ni}$	$(T_{1/2} = 7.5 \text{ ka})$
	$^{59}\text{Co}(n,\alpha)^{55}\text{Fe}$	$(T_{1/2} = 2.60 \text{ a})$
	$^{59}\text{Co}(n,2n)^{58}\text{Co}$	$(T_{1/2} = 71.3 \text{ d})$
Main reaction	$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	$(T_{1/2} = 5.27 \text{ a})$
	$^{65}\text{Cu}(n,p)^{65}\text{Ni}$	$(T_{1/2} = 2.56 \text{ h})$
	$^{65}\text{Cu}(n,2n)^{64}\text{Cu}$	$(T_{1/2} = 12.8 \text{ h})$

When the energy response of such accompanying reactions are not required for spectrum unfolding, one needs less accurate cross section data in order to evaluate their contributions to the response.

6. SECONDARY REACTIONS.

In some cases the product nuclide produced by fast neutrons suffers burn-up by thermal neutrons.

Examples:

Primary reaction	$^{58}\text{Ni}(n,p)^{58}\text{Co}^m + ^{58}\text{Co}$
Secondary reaction	$^{58}\text{Co}^m(n,\gamma)$
	$^{58}\text{Co}(n,\gamma)$

There are only the following data available (see Hogg and Weber, report IDO 16744, and Kondurov, Energie Atomique 24 (1968), 38):

$$\sigma_0(^{58}\text{Co}^m) = 176000 \text{ barn}$$

$$\sigma_0(^{58}\text{Co}) = 1650 \text{ barn}$$

$$I'(^{58}\text{Co}^m) = 760000 \text{ barn}$$

Primary reaction	$\text{Ti}(n,x)^{46}\text{Sc}$
Secondary reaction	$^{46}\text{Sc}(n,\gamma)$
Primary reaction	$^{54}\text{Fe}(n,p)^{54}\text{Mn}$
Secondary reaction	$^{54}\text{Mn}(n,\gamma)$

Data on these secondary reactions have not been collected or measured in a systematic way. It seems that in any case the secondary reactions in the nickel detector, because of its importance, need confirmation by new measurements.

Laboratories which have not the facilities for accurate high resolution measurements could possibly contribute in this field to increase our knowledge.

### 7. THERMAL CROSS SECTIONS.

Some thermal neutron capture reactions have 2200 m/s cross sections which are not known very well. From the list given in reference [1] the following values are taken:

$^{151}\text{Eu}(n,\gamma)^{152}\text{Eu}^m$	$\sigma_0 = 3100 \pm 400$ barn
$^{164}\text{Dy}(n,\gamma)^{165}\text{Dy}$	$\sigma_0 = 2700 \pm 200$ barn
$^{176}\text{Lu}(n,\gamma)^{177}\text{Lu}$	$\sigma_0 = 2100 \pm 150$ barn
$^{186}\text{W}(n,\gamma)^{187}\text{W}$	$\sigma_0 = 38 \pm 2$ barn

### 8. RESONANCE INTEGRALS.

The reaction  $^{109}\text{Ag}(n,\gamma)^{110}\text{Ag}^m$  which together with  $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$  in the double foil method can be used to determine fluences of thermal and intermediate neutrons, has the disadvantage that literature values of the resonance integrals (and also for the 2200 m/s cross section) show a large spread. The recent values as reported by Sims and Juhnke (J. Inorg. Nucl. Chem. 30 (1968), 349) should be confirmed by new measurements.

### 9. CAPTURE REACTIONS WITH ISOMERIC STATES.

There are two capture reactions with europium:

$^{151}\text{Eu}(n,\gamma)^{152}\text{Eu}^m$	$(T_{\frac{1}{2}} = 9.3 \text{ h})$
$^{151}\text{Eu}(n,\gamma)^{152}\text{Eu}$	$(T_{\frac{1}{2}} = 12.5 \text{ a})$

If the nuclear data files, such as the ENDF/B file give only the total activation cross sections, one wishes to have data on the isomeric yield in activation, and its energy dependence.

In the case considered there is no isomeric transition, and for reasons of counting statistics one prefers to count the  $^{152}\text{Eu}^m$  nuclide, whose activity is much larger than the activity of the ground state. The gamma ray spectra of both nuclides show some common peaks.

For the reactions  $^{176}\text{Lu}(n,\gamma)^{176}\text{Lu}^m$  ( $T_{1/2} = 155$  d) and  $^{176}\text{Lu}(n,\gamma)^{177}\text{Lu}$  ( $T_{1/2} = 6.7$  d) we have the same situation that only the total activation cross section is known.

With respect to the reactions  $^{115}\text{In}(n,\gamma)^{116}\text{In} + ^{116}\text{In}^m$  one may remark that the data for the total cross section for In, needed for the self-shielding correction is not present in the evaluated data files.

#### 10. (n,p) REACTIONS.

Here one should mention the discrepant values for the average cross section for the  $^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$  reaction. Here one needs to investigate in detail the energy dependent response in the region near the threshold. The problem of a possible sub-threshold activation should be solved.

#### 11. LONG TERM FLUENCE DETECTORS | 1 |.

For purposes of determining the radiation damage e.g. to the reactor pressure vessel one should like to apply long term neutron fluence detectors over a period of 10 years or more.

Activation technique have the advantage of simplicity, the limitations being in the choice of suitable reactions. Also one might think of formation of  $^{137}\text{Cs}$  produced in a fission detector.

With respect to the activation detectors for this purpose it has been suggested that the reactions  $^{93}\text{Nb}(n,\gamma)$  and  $^{94}\text{Mo}(n,p)$ , both giving rise to  $^{94}\text{Nb}$  with a half-life of 20 ka, should be investigated in more detail, not only with respect to material purity and absolute measurements, but also with respect to cross section data.

## 12. HELIUM PRODUCTION REACTIONS.

With respect to radiation damage processes, or within the framework of long term fluence measurements one should pay more attention to the nuclear reactions in which by virtue of  $(n,\alpha)$  reactions helium is produced, and the helium quantity produced can be determined.

Here the  $(n,\alpha)$  reactions with nuclides present in structural materials (such as Ni, Fe, Cr, Mn) should be considered.

## 13. INACTIVE NUCLIDE PRODUCTION.

The case of a reaction of the type  $Me(n,\alpha)$  total He is a particular case of a general fast neutron reaction, in which an inactive nuclide is produced and is to be measured by advanced techniques such as mass spectrometry.

It might be worthwhile to study possibilities for such reactions.

## 14. FISSION PRODUCT YIELDS.

At the IAEA Symposium on Nuclear Data in Science and Technology, held in Paris from 12-16 March 1973, some important compilations on fission product yield have been presented.

Since at the IAEA panel meeting on fission product nuclear data, to be held in Bologna, 26-30 November 1973, this topic will be considered in detail, we only mention here that for neutron metrology work the fission yields of  $^{95}\text{Zr}$ ,  $^{131}\text{I}$ ,  $^{132}\text{I}$ ,  $^{137}\text{Cs}$ ,  $^{140}\text{Ba}$  and  $^{148}\text{Nd}$  are of importance.

## 15. DECAY SCHEMES OF PRODUCT NUCLIDES.

Nearly always the activities of product nuclides in activation (or fission reactions) are determined with gamma ray spectrometers.

For accurate absolute activity determinations one has to know the decay schemes (half-lives; transition energies) and especially the gamma abundances of dominant gamma transitions. It is reasonable to ask for well established and evaluated decay schemes. As examples could be mentioned  $^{27}\text{Mg}$ ,  $^{64}\text{Cu}$ ,  $^{110}\text{Ag}$ ,  $^{116}\text{In}^m$ ,  $^{152}\text{Eu}$ ,  $^{165}\text{Dy}$ ,  $^{170}\text{Lu}$ ,  $^{182}\text{Ta}$  and  $^{187}\text{W}$ .

#### 16. GAMMA RAY CALIBRATION SOURCES.

Some years ago the IAEA laboratory at Seibersdorf made regularly available sets of calibrated solid radionuclide sources which could be used for absolute activity determinations and/or for preparing detection efficiency curves for gamma ray spectrometry.

For gamma ray calibration work especially for low energies one could think of the following radionuclides:  $^{133}\text{Ba}$  ( $T_{1/2} = 7.2$  a),  $^{166}\text{Ho}^m$  ( $T_{1/2} = 26.7$  a),  $^{182}\text{Ta}$  ( $T_{1/2} = 115$  d),  $^{226}\text{Ra}$  ( $T_{1/2} = 16000$  a) which all have a series of low energy gamma ray transitions.

The gamma abundances should however be known accurately.

Especially the  $^{166}\text{Ho}^m$  nuclide (see N. Lavi, Nucl. Instr. Methods, 109 (1973), 265), seems to be very promising. Therefore it is suggested that the IAEA Laboratory at Seibersdorf considers a distributions of calibrated  $^{166}\text{Ho}^m$  sources.

New determination of the gamma abundances are in this respect very valuable.

#### 17. CONCLUSIONS (from |1|).

In the last years one can observe that there is an increasing need for better and more reliable neutron spectrum data. Advanced computer codes such as SAND-II which are able to unfold the responses from activation and fission detectors, require consistent cross section data sets and accurate activity measurements which in turn require well known decay schemes (half-lives and gamma abundances).

There still remain an appreciable number of discrepancies between evaluated measured average cross sections and integral values derived from evaluated differential cross section data.

#### 18. SUGGESTED RECOMMENDATIONS.

The IAEA Nuclear Data Section should continue reporting on the status of cross section data for reactions of interest. Studies for comparing and checking the available cross section data files in reference spectra should be promoted.

The IAEA Nuclear Data Section should prepare a compilation of recommended decay schemes (inclusive of error estimates) for all nuclides produced in important activation reactions.

The IAEA should publish and keep up-to-date recommended nuclear data sets.

19. REFERENCES.

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RCN-73-083 (Reactor Centrum Nederland, Petten, September 1973).

REMARKS CONCERNING CROSS SECTIONS  
FOR THRESHOLD DETECTORS

H. Liskien and A. Paulsen

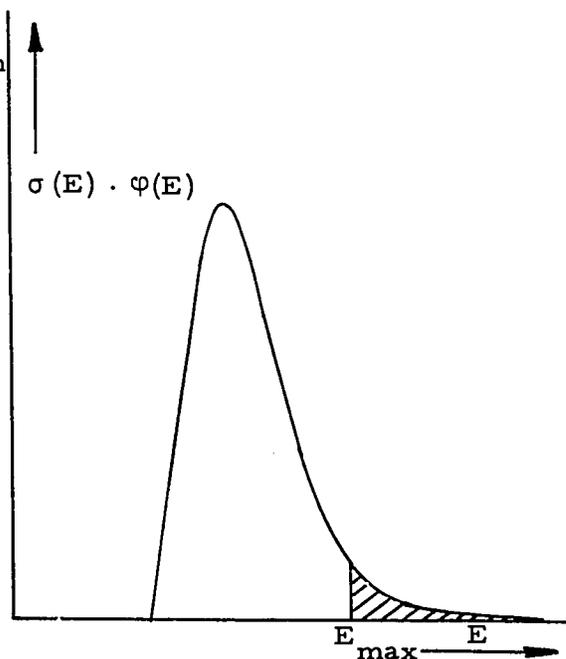
Most Important Energy Range

Concerning differential cross section data of interest for fast reactor neutron dosimetry WRENDAs contains requests concerning as much as 30 reactions for which new measurements are requested covering neutron energies often up to 15 to 18 MeV, although the corresponding thresholds are in the few MeV region. It seems to be indispensable to concentrate the anyway weak activities in the field of differential activation cross section measurements, if a rapid improvement is expected. Consequently an agreement on a smaller set of well chosen reactions should be found and a limitation of the energy range in which new measurements are requested should be discussed.

A typical response function of a threshold reaction in a  $^{235}\text{U}$  thermal fission spectrum is given in the figure. Using the Watt expression, we have calculated values for  $E_{\text{max}}$  defined by

$$\frac{\int_0^{E_{\text{max}}} \sigma(E) \cdot \varphi(E) dE}{\int_0^{\infty} \sigma(E) \cdot \varphi(E) dE} = 0.04$$

for a number of threshold reactions. The choice of the constant 0.04 was based on the following consideration.



Typical accuracies for careful differential cross section determinations with monoenergetic neutrons produced with charged particle accelerators are around 5 to 7 %. Even for very rough measurements a 25% accuracy

can be achieved. If the constant discussed is set to 0.04, then an uncertainty of 25% in the cross section corresponds to a 1% contribution in activity uncertainty which at the same time is believed to be a realistic estimate for activity determinations of irradiated threshold detectors.

The following table summarizes the results :

Reaction	$E_T$ (MeV)	$E_{max}$ (MeV)	$E_{max}-E_T$ (MeV)
$^{103}\text{Rh}(n, n')$	0.1	6.3	6.2
$^{115}\text{In}(n, n')$	0.4	6.0	5.6
$^{238}\text{U}(n, f)$	0.9	6.8	5.9
$^{232}\text{Th}(n, f)$	1.1	7.3	6.2
$^{58}\text{Ni}(n, p)$	1.2	7.9	6.7
$^{32}\text{S}(n, p)$	1.7	7.6	5.9
$^{54}\text{Fe}(n, p)$	1.8	8.0	6.2
$^{46}\text{Ti}(n, p)$	3.3	9.6	6.3
$^{56}\text{Fe}(n, p)$	5.0	11.4	6.4
$^{59}\text{Co}(n, \alpha)$	5.5	12.0	6.5
$^{63}\text{Cu}(n, \alpha)$	5.8	11.5	5.7
$^{27}\text{Al}(n, \alpha)$	6.0	12.0	6.0
$^{65}\text{Cu}(n, 2n)$	10.1	15.9	5.8
$^{63}\text{Cu}(n, 2n)$	11.0	17.0	6.0

Although  $E_{max}$  has been calculated for all kinds of relevant reactions and covering the whole range of threshold (or effective) energies it is always found roughly 6 MeV above threshold. We think that this result is also valid for actual spectra deviating from a pure fission spectrum and would recommend to concentrate new measurements of differential cross sections in the energy range  $E_T$  to  $E_T+6$  MeV, if there are already results outside this range.

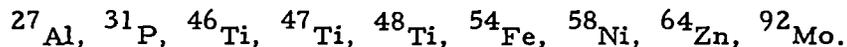
#### Neutron Energy Coverage

Concerning the coverage of neutron energies for which differential cross section measurements have been published, two observations can be made :

The relative large number of results around 14 MeV and the lack of information for the range 6 to 12 MeV.

The many results around 14 MeV are simply due to the fact that here monoenergetic neutrons can easily be produced with low energy deuterons employing the  $T(d, n)^4\text{He}$  reaction. However, the activity contribution from neutrons of this energy is already very small in a typical reactor spectrum for all reactions except  $(n, 2n)$ . Therefore these results are often of limited value although they may allow a point-check if other measurements exist which cover a wider energy range which includes the 14 MeV region.

Employing the  $D(d, n)^3\text{He}$  reactions monoenergetic neutrons up to 6.3 MeV may be produced if a 3 MV electrostatic accelerator is available and this energy may be brought up to 8.7 MeV for 5.5 MV machines. The lack of differential cross section data in the 6 to 12 MeV range is due to the rareness of accelerators with more than 3 MV which are available for neutron work. Requests should be directed especially to laboratories equipped with such machines where measurements should be performed for the following  $(n, p)$  reactions:



#### Theoretical Cross Section Calculations

Theoretical calculations for cross sections of threshold detectors or for capture cross sections are based on the compound nucleus theory and yield results with accuracies around 50 to 100%, this means one order of magnitude more uncertain than results of carefully executed experiments. This is due to the neglect of the direct reaction mode and pre-equilibrium emission and to the insufficient knowledge of used parameters (level structure, transmission coefficients). Such calculations, therefore, are only of value when normalized to existing results and used for inter- or extrapolation. Promising results have been obtained only near threshold by Hauser-Feshbach calculations, especially for  $(n, n')$  reactions.

Subthreshold Response ?

The threshold of excitation functions for reactions used as "threshold detectors" is determined by the Q-values of the reaction, the masses involved and the height of the Coulomb wall. (n, 2n) and (n, n') reactions have a priori a negative Q-value and therefore no response to thermal and epithermal neutrons. (n, α)-reactions used in reactor dosimetry have the following Q-values :

$^{27}\text{Al}(n,\alpha)$	Q = -3.1 MeV
$^{59}\text{Co}(n,\alpha)$	Q = +0.3 MeV
$^{63}\text{Cu}(n,\alpha)$	Q = +1.7 MeV
$^{58}\text{Ni}(n,\alpha)$	Q = +2.9 MeV.

Although the results of integral measurements for  $^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$  are widely scattered they are on average 40 % higher than the value which can be calculated from the published experimental excitation function<sup>2)</sup>.

A cross check of the differential data at 8 MeV (maximum of the response function) confirmed the published experimental cross sections<sup>3)</sup>.

Other values around 14 MeV do not allow a clear conclusion :

14.05 MeV	$39.1 \pm 2.7$ mb	excit. function ref. 4)
14.42 MeV	$35.7 \pm 2.5$ mb	
14.99 MeV	$31.6 \pm 2.2$ mb	
14.2 MeV	$26.1 \pm 5.0$ mb	ref. 5)
14.6 MeV	$49.5 \pm 10.0$ mb	ref. 6)
14.7 MeV	$33.8 \pm 2.4$ mb	ref. 7)

To clarify the situation more reliable integral measurements for  $^{63}\text{Cu}(n,\alpha)$  are needed. The two last recent results differ by a factor 1.7<sup>8, 9)</sup>.

Recently McELROY in a SAND-II analysis found solutions which evidenced a low energy component of the  $^{63}\text{Cu}(n,\alpha)$  cross section<sup>10)</sup>. Such an effect is undetectable with monoenergetic neutrons but demands reactor irradiations with high thermal to flux ratios for verification. This effect, if it exists, should increase with the Q-value of the reaction. This would explain that no discrepancy

(between measured and calculated average cross section) has been observed for  $^{59}\text{Co}(n,\alpha)$ . No data exist for  $^{58}\text{Ni}(n,\alpha)$  where this effect should be even stronger. A less sensational explanation of a sub-threshold response for  $^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$  is the reaction  $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$  in cobalt impurities. A set of irradiations of copper detectors (preferable relative to  $\text{Al}(n,\alpha)$ ) containing different but well-known cobalt impurities and extrapolation to zero should allow to decide which explanation is correct.

#### Accuracy of Integral Data

The extensive evaluation of a consistent set of integral values for cross sections of detectors used in reactor neutron dosimetry by FABRY <sup>11)</sup> allows to point out relatively inaccurate data for which new measurements are urgently needed, namely  $^{27}\text{Al}(n,p)$ ,  $^{47}\text{Ti}(n,p)$ ,  $^{63}\text{Cu}(n,\alpha)$ ,  $^{63}\text{Cu}(n,2n)$ .

The remaining results yield in average an error of  $\pm 3.5\%$  not including the uncertainty of the absolute scale. This is supported by a second set of recommended values published by ZIJP <sup>12)</sup> from which one may deduce an average error of  $3.8\%$ . If one adjusts the two sets to the same absolute scale, then one finds an average difference of  $\pm 2.8\%$  also confirming the above mentioned numbers and permitting the statement that the relative accuracy of evaluated integral cross section is between  $\pm 3$  and  $\pm 4\%$ . FABRY estimates the uncertainty in absolute scale to  $\pm 5\%$  to  $\pm 7\%$  again consistent with the difference of  $6.6\%$  in absolute scale which exists between the sets of FABRY and ZIJP.

#### Availability of Existing Data

There is no longer a necessity for evaluators and reactor physicists to waste time in scanning literature and compiling experimental differential cross sections for reactions relevant to reactor neutron dosimetry. Nowadays the four Neutron Data Centers linked together by the EXFOR exchange have well established files for such data : A recent comparison between the file of CBNM Geel and the NDCC/NEA Sacaly revealed satisfactory agreement. Consequently the CBNM compilation <sup>13)</sup> will no longer be continued. Instead,

NDCC plans the issue of computer outputs from their NEUDADA files which will contain data in numerical and graphical form for the following reactions :

$^{24}\text{Mg}(n, p)$	$^{48}\text{Ti}(n, p)$	$^{63}\text{Cu}(n, 2n)$	$^{103}\text{Rh}(n, n')$
$^{27}\text{Al}(n, p)$	$^{54}\text{Fe}(n, p)$	$^{63}\text{Cu}(n, \alpha)$	$^{115}\text{In}(n, n')$
$^{27}\text{Al}(n, \alpha)$	$^{55}\text{Mn}(n, 2n)$	$^{64}\text{Zn}(n, p)$	$^{127}\text{I}(n, 2n)$
$^{31}\text{P}(n, p)$	$^{56}\text{Fe}(n, p)$	$^{65}\text{Cu}(n, 2n)$	$^{232}\text{Th}(n, f)$
$^{32}\text{S}(n, p)$	$^{58}\text{Ni}(n, p)$	$^{92}\text{Mo}(n, p)$	$^{237}\text{Np}(n, f)$
$^{46}\text{Ti}(n, p)$	$^{58}\text{Ni}(n, \alpha)$	$^{93}\text{Nb}(n, 2n)$	$^{238}\text{U}(n, f)$
$^{47}\text{Ti}(n, p)$	$^{59}\text{Co}(n, \alpha)$	$^{93}\text{Nb}(n, n')$	

Storing and distributing evaluations for this class of data also belongs to the responsibility of the four Neutron Data Centers. However, those stemming from the ENDF/B file are at present only distributed within the OECD area.

The files of the Neutron Data Centers are less complete with respect to  $^{235}\text{U}$ -fission neutron averaged cross sections. FABRY in his recent evaluation on integral cross sections <sup>11)</sup>, for example, is using 13 references from which only six are included in the last NDCC/NEUDADA Index <sup>14)</sup>. Similarly, PEARLSTEIN compares his semiempirical cross section calculation <sup>15)</sup> with literature values. Concerning threshold reactions in use for reactor neutron dosimetry he is referring to nine references, but only five of them are contained in the index. A special effort by the Neutron Data Centers should be made to complete their files also with respect to these data, although in many cases it may be difficult for the compiler to decide if the used spectrum is near enough to a pure fission spectrum to justify a compilation.

There existed no compilation problem as long as one described the fission neutron spectrum of  $^{235}\text{U}$  by a one or two (Maxwellian or Watt form) parameter formula. However, the IAEA Consultants' Meeting on Prompt Fission Neutron Spectra <sup>16)</sup> recognized that such simple forms do not satisfactorily fit all observed fission spectra and are poorly grounded in theory. As it is premature to suggest more complex representations it was recommended that the Neutron Data Centers compile these data in numerical form as measured. The next step, the careful evaluation of these experimental fission spectra, will be a very important future task.

A list of relevant references of compilations for half-lives and decay data are given in the Conclusions and Recommendations of the First Meeting of the International Working Group on Nuclear Structure and Reaction Data (IWGNSRD) <sup>17)</sup>. These data are not specific neutron data and therefore do not belong to the scope of the Neutron Data Centers. Evaluation activities in this field normally cover the whole field of radionuclides with accordingly long delays between the publication of new experimental results and its inclusion in a new re-evaluation. It would be highly desirable if for the small number of radionuclides of interest for reactor neutron dosimetry a specific solution could be found. To this end the Euratom Working Group for Reactor Dosimetry (EWGRD) recently asked the Euratom group which is engaged in evaluation of decay data of radio-isotopes, for assistance <sup>18)</sup>.

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- 3) H. LISKIEN, A. PAULSEN and R. WIDERA,  
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- 4) H. LISKIEN and A. PAULSEN, Nuclear Physics 63, 393 (1965)
- 5) M. BORMANN et al. , Nuclear Physics A 186, 65 (1972)
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- 7) A. PAULSEN, Z. f. Phsyik 205, 226 (1967)
- 8) F. NASYROV, Atom. Energ. 25, 437 (1968)
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- 10) W. N. McELROY et al. , Proc. 3rd Conf. on Neutron Cross  
Section and Technology, Knoxville, p. 120 (1971)
- 11) A. FABRY, Report BLG-465 (1972)
- 12) W. L. ZIJP, Report RCN-71-083 (1971)
- 13) H. LISKIEN and A. PAULSEN, Report EUR 119. e.
- 14) Newsletter Bulletin CCDN-NW/13 (1972)
- 15) S. PEARLSTEIN, Journ. Nucl. Energy 27, 81 (1973)
- 16) Proceedings of a Consultants' Meeting on Prompt  
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- 17) L. HJÄRNE, Report INDC(NDS)-46/U+W (1972)
- 18) H. LISKIEN, Report EWGRD-482"L" (1972)

Conclusions and Recommendations

of the Meeting

## I. General Conclusions and Recommendations

1. The problem of cross sections for the detectors used in reactor dosimetry (damage studies, irradiation experiments, shield assessment, reactor performance studies) is a very critical one that may appreciably influence the development of competitive nuclear power. This fact has recently been recognized by many organizations, and a certain amount of systematic work has been started in several countries and agencies.
2. Much remains to be done before differential cross sections and other nuclear data are adequate for neutron dosimetry purposes. At present, evaluated energy-dependent reaction cross section uncertainties are thought to be the largest single source of error for multiple foil derived flux-spectra. Large discrepancies and gaps exist in differential measurements. Furthermore unacceptable discrepancies exist between integral and differential measurements of neutron cross sections.
3. It should be emphasized that it is the relative consistency of reactor physics and multiple foil derived flux-spectra that is of primary concern to reactor development programs for application in the correlation of radiation effects in such areas as materials damage. That is, given the capability to measure reaction rates with sufficient accuracy, in both test and operating power reactors, it is then necessary to use a reference set of integrally consistent evaluated energy-dependent cross sections for the flux-spectral definition.
4. The role of international cooperation in the effort to develop a reference set of neutron dosimetry cross sections is essential. The IAEA and its Nuclear Data Section should act as a focus in promoting this international cooperation. One vital aspect of this cooperation is the necessity for the free unrestricted circulation of detector cross sections, evaluated differential

files, adjusted fine group or multi-group sets, integral values and other nuclear data as has been done already for neutron standard cross sections.

5. The inconsistencies between integral data and differential data make necessary new measurements of differential cross section data for a few reactions to better than 5%. Experiments in well defined standard spectra, benchmark experiments, should be used to improve consistency between differential and integral data and to complement our knowledge of detector cross sections.
6. It is important that the effort to improve our knowledge of detector cross sections should be paralleled by an effort to arrive at a set of reference values for differential neutron data to be recommended for use in different laboratories, so that reactor experiments, and in particular irradiation experiments, can be directly compared and exchanged, thereby increasing the amount of useful available information and reducing the duplication of efforts. The acceptance of such a set of reference values on an international basis, and the internal consistency of such values, are considered to be potentially more important than the improvement of the knowledge of the differential cross sections.
7. In practical applications of activation measurements the neutron spectrum in itself is of limited importance except for benchmark or special applications. The final purpose of activation measurements is in general to arrive at values for integral quantities which are not directly measurable for which purpose an accuracy of 10-20% is required. In special cases like fuel irradiations and graphite irradiations in high temperature gas cooled reactors, accuracies to 5% or better may be needed.
8. In order to specify better the requirements on the basic differential nuclear data, the sensitivity to these data of integral quantities of interest should be studied for practical cases.

9. The cooperative effort for improvement of reactor dosimetry should include the exchange of detectors, the intercalibration of facilities and measuring equipment, and the distribution of calibrated sources. The IAEA Seibersdorf Laboratory and other "standards" laboratories could play an important role in this area. It would be desirable to arrive at some convergence among the various national and international groups working in reactor dosimetry on the selection of dosimeters for the various uses in order to facilitate assignment of priorities.

## II. Conclusions and Recommendations Concerning Nuclear Data

10. In spite of efforts during the past 15 years, the target goal of  $\pm 5\%$  accuracy for differential neutron cross sections for reactor dosimetry purposes has been reached for a rather limited number of reactions. These reactions are mostly those also of interest in other fields like reactor design and neutron standards.

Reasons for this unsatisfactory situation are:

- a) the lack of agreement on a limited set of reactions to which all measuring efforts are concentrated;
  - b) the failure to concentrate on the most sensitive energy region for dosimetry purposes;
  - c) the lack of sufficient laboratories equipped with accelerators which can produce monoenergetic neutrons in the 6 - 12 MeV region and which are available for neutron measurements.
11. In view of the integral nature of the quantities of practical interest for dosimetry purposes, the detailed structure in the energy dependence of the cross sections of the detectors is not important except near the threshold or for resonances. Unnecessary detail should not be requested. Of critical concern in the threshold region and in resonances is an accurate energy definition. In many cases it may be possible to combine some good resolution

differential measurements in restricted energy areas with broad resolution measurements in others.

12. When practical, one should recommend measurements of cross sections for the production of the particular radiation that will be used in the final activation measurements rather than measurements for the production of the particular radionuclide. This would remove the uncertainty contributed by insufficient knowledge of decay schemes. The knowledge of decay schemes remains useful for corrections or for the selection of methods.
13. The comparison between averaged cross sections calculated from differential data and determined on integral experiments suffers from the insufficient knowledge of the neutron spectrum. In particular, typical accuracy achieved for the fission neutron spectrum averaged cross sections is about  $\pm 10\%$  and  $\pm 4\%$  if normalization to the absolute scale is not taken into account.
14. Information on the neutron cross section data used in dosimetry consists of three types:
  - a) measured differential cross sections
  - b) benchmark spectra (measured or calculated)
  - c) integral measurements made in these benchmark spectra

The correlation between measured integral data and calculated integral data (that is data calculated from differential cross sections and known spectra) are at present inconsistent.

15. The first aim of a benchmark experiment is to obtain additional information to aid in producing consistency among a limited number of reactions to be called CATEGORY I reactions (see item 18 below) by supplying the necessary data which together with new differential measurements will permit a re-evaluation of the differential cross sections for these reactions.

16. The second aim of a benchmark experiment is to test the data for reactions which are called CATEGORY II reactions (see item 18 below). The results will be used to normalize or adapt the differential neutron cross sections for these reactions to obtain consistency. The adjustments should be within the errors of existing differential data and/or consistent with theoretical nuclear reaction model considerations.
17. The following benchmarks are identified:

Benchmark	Approximate Data Testing Energy Range
1. Thermal spectrum	$< 4 \times 10^{-7}$ MeV
2. Epithermal $1/E$ spectrum	$\sim 4 \times 10^{-7} < E < 10^{-3}$ to $10^{-2}$
3. Intermediate energy standard neutron field (ISNF) a)	$10^{-3} < E < 1$
4. TAPIRO b)	$10^{-3} < E < 10$
5. BIG 10 c)	$\bar{E} = 0.6$
6. $\Sigma \Sigma$ d)	$\bar{E} = 0.7$
7. CFRMF e)	$\bar{E} = 0.7$
8. Fission spectrum f)	$\bar{E} = 2$
9. Fusion spectrum g)	$10^{-3} < E < 14$

a) ISNF-Intermediate-energy Standard Neutron Field being developed at the National Bureau of Standards, Washington, D.C., USA, and at MOL, Belgium (J.A. Grundl and A. Fabry, Private Communication, 1973)

b) A. d'Angelo, Mr. Martini, M. Salvatores, "<sup>235</sup>U Compact Cu-Reflected TAPIRO Reactor Integral Experiment Results and a Check of Some High-Energy ENDF/B-111 Data", Trans.Am.Nucl.Soc., 17, 498, November 1973.

- c) BIG 10 - A 10% enriched bare metal (cylindrical core) fast assembly located at Los Alamos Scientific Laboratory, Los Alamos, New Mexico, USA. (L.J. Sapir, H.H. Helmick, and J.D. Orndoff, "Big Ten, a 10% Enriched Uranium Critical Assembly: Kinetic Studies", Trans. Am. Nucl. Soc. 15, 312, June 1972 and E.J. Lozito and E.J. Dowdy, "A Measurement of the Central Neutron Spectrum of "BIG-10" Critical Assembly", Trans. Am. Nucl. Soc. 17, 529, November, 1973.)
- d)  $\sum\sum$ - Secondary Intermediate Standard Spectrum in Cavity located at CEN/SCK, Mol, Belgium. (A. Fabry and G. and S. Deleeuw, "The  $\sum\sum$  Secondary Intermediate-Energy Standard Neutron Field", Trans. Am. Nucl. Soc. 17, 527, November, 1973)
- e) CFRMF - Coupled Fast Reactivity Measurement Facility located at Aerojet Nuclear Company, Idaho Falls, Idaho, USA. (J.W. Rogers, D.A. Millsap, Y.D. Harker, "CFRMF Neutron Field Flux Spectral Characterization", Trans. Am. Nucl. Soc., 17, 527, November, 1973.)
- f) Fission Spectrum Cavity Facilities located at CEN/SCK, Mol, Belgium, National Bureau of Standards, Washington, D.C., USA, and  $^{252}\text{Cf}$  source at NBS. (J.A. Grundl and A. Fabry, private communication, 1973 and J.A. Grundl, V. Spiegel, and C. Eisenhouer, "Measurement of  $^{235}\text{U}$  and  $^{238}\text{U}$  Fission Cross Sections for  $^{252}\text{Cf}$  Spontaneous Fission Neutrons", Trans. Am. Nucl. Soc. 15, 945, November 1972.) U-235 Converter at Ispra, Italy (EURATOM) (O. Chiochio et al., "Measurement of Fast Reactor Type Neutron Spectra by Foil Activation Techniques", Nucl. Inst. Meth. 91, 45, 1971)
- g)  $\text{B}_4\text{C}$ -Al block fed by 14 MeV neutrons under construction at CCR Euratom. Li block at GKF Jülich fed by 14 MeV neutrons in experimental phase.

These benchmarks are selected so that their spectral characterization is NOT based on integral measurements, but on more refined methods, e.g. neutron spectrometry and detailed reactor physics calculations. Other appropriate benchmarks should be identified.

- 18. With a view to recommending a practical coordinated approach to the improvement of neutron dosimetry data by using benchmark experiments, two categories of neutron dosimetry reactions are defined. The reactions contained in these two categories are

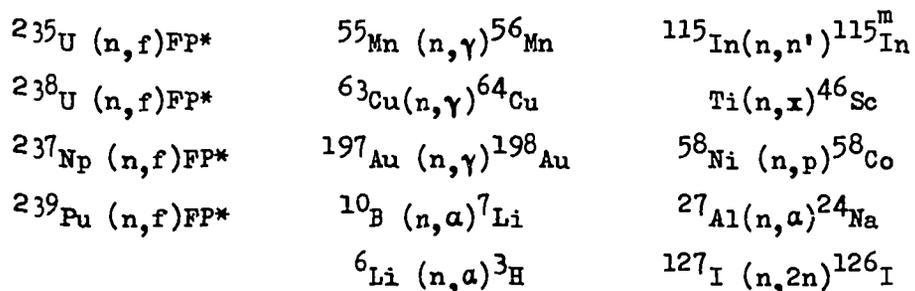
not necessarily chosen with respect to their practical importance or priority for actual routine dosimetry, but rather with the aim of improving dosimetry cross section data files.

The set of reactions in CATEGORY I is selected according to the following criteria:

- a) in terms of energy response, the set provides a reasonable coverage of neutron spectra in most reactor environments;
- b) insofar as possible the differential nuclear data for these reactions are among the best known ones today as they are either standards for differential cross section measurements or have been already extensively studied with monoenergetic neutrons.

Most other useful dosimetry reactions are placed in CATEGORY II. The energy dependent cross section data required for these reactions should be adjusted with respect to the CATEGORY I cross sections by a correlation scheme involving precise (and preferably interlaboratory) integral measurements in a limited set of benchmark spectra.

19. Tentative list of CATEGORY I reactions.



\* yields of  $^{95}\text{Zr}$ ,  $^{137}\text{Cs}$ ,  $^{140}\text{Ba}$ ,  $^{148}\text{Nd}$  belong to the second category

20. Tentative list of CATEGORY II reactions

(See Annex I.)

21. With respect to secondary and/or accompanying reactions in activation detectors, it was felt that cross section data with broad energy resolution and moderate accuracy are useful. Available data should be collected. New measurements for this category of reactions could be performed by laboratories, which do not possess the specialized facilities which are required for accurate measurements at high energy resolution on the main activation reactions.
22. A recommended consistent set of detector cross sections meeting the requirements for reactor neutron dosimetry can be reached only by an iterative procedure including evaluations, differential measurements and integral experiments. In order to speed up this process by making optimum use of the efforts carried out in various laboratories and countries, it is essential that different experimental results can be compared and interpreted.

It would be desirable that as the first step of the effort to establish a set of internationally accepted neutron dosimetry cross sections, i.e. the setting up of a consistent set of cross sections for CATEGORY I reactions, that all groups involved in this effort could have available the same "zero approximation" evaluated data. This would speed up the procedure for arriving at a unique internally consistent set of detector cross sections and at a better characterization of benchmark spectra.

23. The most effective way to obtain this "zero approximation" international set of dosimetry cross sections would be through the general availability of the ENDF-B/IV Dosimetry file, at least that part relative to our CATEGORY I reactions. ENDF-B/IV is recommended because of its completeness, the flexibility of its format and the general availability of processing codes for it.

We therefore recommend that these files should be made available through the Four Neutron Data Centers as soon as possible, regardless of the performance of such cross sections, since the aim of the recommended international effort would be to explore the consistency and adequacy of such data and to suggest improvements. All producers of integral data and benchmark experiments

having access to these files would be asked to transmit to the original evaluators of the CATEGORY I reactions the results of their measurements, their interpretation in terms of ENDF-B cross sections and the identification of possible discrepancies. No modification of the CATEGORY I cross sections should be applied or distributed except through those responsible for the ENDF-B data file. All modifications should be based on a re-evaluation of the differential measurements.

24. New evaluations of CATEGORY II cross sections, based on corrections of original evaluations by means of careful benchmark experiments, should be encouraged. The IAEA at a future date should consider these results and issue a set of recommended values for CATEGORY II reactions.
25. The assessment of errors is of primary importance for the utilization of the nuclear data. We recommend that all evaluated data files for dosimetry should include an evaluation of errors, if possible in the complete form foreseen for the ENDF-B/IV error files.
26. The accuracy of half-lives and decay schemes of relevant radionuclides contributes to the accuracy of reactor dosimetry measurement. However these data are outside the scope of the Four Neutron Data Centers. Most of the present evaluation activities for this kind of data cover all of the known radionuclides resulting in long delays between the publication of new experimental results and their inclusion in an updated evaluation. Therefore, this Consultants' Meeting recommends an IAEA evaluation for the small number of radionuclides of interest for reactor neutron dosimetry making use of the expertise in the Agency's Nuclear Data Section and the Seibersdorf laboratory. This evaluation should be kept current.

27. The use of fission detectors requires a knowledge of yields for selected fission products, as a function of the energy of the neutron inducing fission and for several nuclides, and the nuclear data and decay schemes for such fission products.
28. These questions have not been discussed in detail, in view of the IAEA Panel on Fission Product Nuclear Data to be held in Bologna, 26-30 November 1973. It would be desirable that this Panel give advice concerning recommended data for use in dosimetry problems and the uncertainty that should be attributed to that data. In particular, it would be important to have recommendations

for the nuclides  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{237}\text{Np}$ ,  $^{239}\text{Pu}$

for the yield of the fission products  $\text{Zr}^{95}$ ,  $\text{Cs}^{137}$ ,  $\text{Ba}^{140}$ ,  $\text{Nd}^{148}$

for incident neutron energies from thermal to fission.

Possible competing neutron absorption cross sections in the chains containing these fission products and yields of fission products decaying to the same final products should also be considered if important for high flux and/or long term irradiations.

### III. Miscellaneous Conclusions and Recommendations

29. Intercalibrations of detectors in standard neutron fields (when available), reduce the relative errors in reaction rate determinations between different foil materials. The influence of the two main sources of error - the reaction rate determination and the cross section - is minimized in this way. Relative errors in reaction rate determination may be reduced to one or two percent. The results (fluence or spectra) are then relative to the standard spectrum measured so that the error then depends on the precision to which the standard (fluence or spectrum) is known.

30. The consultants' meeting recommends that a programme of inter-calibration between fission spectra obtained in different laboratories be undertaken. If standard spectra are not available the reaction rate must be determined by direct comparison with a known standard source of the same material.
31. If standard sources can be obtained from specialized laboratories, it is not appropriate to set up apparatus for absolute calibration at the reactor station.
32. Materials used as neutron monitors must be accurately defined and contain a minimum of impurities. A pool of such materials could be established by the IAEA at its Seibersdorf laboratory. This laboratory should promote the establishment of a close working relationship between different centers (CBNM, NBS, IAEA, CEA, PTB, ORNL (Target Isotope Center) which fabricate and provide such detector materials. This cooperative effort should establish the necessary procedures that are needed to maintain a uniform level of overall standardization of the necessary physical and chemical properties of the materials and fabrication of the detectors.
33. Most dosimetry measurements require spectrum unfolding codes for their interpretation. Some useful computer codes of this type are available as evidenced by a recent intercomparison sponsored by the IAEA. Some additional activity in the area of computer programmes seems necessary. The areas of interest are:
  - a) the processing codes needed to prepare libraries for unfolding codes from nuclear data files;
  - b) the assessment of the minimum number of energy groups needed for a satisfactory unfolding;
  - c) the possibility of the selection of standard group structure, in which the exchange of detector data could be more convenient than in the complete file form, especially for institutions with limited computing capability;

- d) further efforts in the assessment of rules for a satisfactory "objective" use of unfolding codes and the appropriate preservation of the physical information contained in the "trial" spectrum when this is a reliable calculated spectrum.
34. Interpretation of dosimetry measurements would profit from an increased exchange and a possible standardization of methods and codes. Among the problems to be considered are corrections for flux perturbations, self-shielding and self absorption of detectors, and interpretation of fluence measurements with variable flux.
35. Although of no immediate concern, problems connected with dosimetry in controlled thermonuclear experiments should be considered. The first measurements in this area have already been started. This additional field of activity requires at present a better characterization of the detectors in the energy region between  $\sim 8$  and 14 MeV and a good calibration at 14 MeV.
36. Some dosimetry methods alternative to activation detectors present interesting features and should be studied further. These include in particular the total helium production methods for high fluence measurements, and the use of accurately calibrated damage detectors.

Tentative list of category 1 reactions:

The "response remarks" refer for the (n,γ) reactions to the energy  $E_r$  of the main resonance, and for the other reactions to the energy range comprising 90% response in a Watt fission neutron spectrum.

Numbers and letters in the column "general remarks" are explained in notes at the end of the list of category 2 reactions.

The reactions are listed in order of increasing proton number.

reaction	response remarks	general remarks	special remarks
${}^6\text{Li}(n,\alpha){}^3\text{H}$		4 6	
${}^{10}\text{B}(n,\alpha){}^7\text{Li}$		4	
${}^{27}\text{Al}(n,\alpha){}^{24}\text{Na}$	6.4 ... 11.9 MeV	3 4 5 7 8 9 10 a d	
$\text{Ti}(n,x){}^{46}\text{Sc}$	3.4 ... 9.1 MeV	1 3 4 5 7 8 9 10 a d	Comprises ${}^{46}\text{Ti}(n,p)$ and also ${}^{47}\text{Ti}(n,d)$ and ${}^{47}\text{Ti}(n,np)$ .
${}^{55}\text{Mn}(n,\gamma){}^{56}\text{Mn}$	$E_r = 337$ eV	2 3 4 7 b d	
${}^{58}\text{Ni}(n,p){}^{58}\text{Co}$	2.1 ... 7.0 MeV	1 3 4 5 6 7 8 10 a d	Includes ${}^{58}\text{Ni}(n,p){}^{58}\text{Co}^m$
${}^{63}\text{Cu}(n,\gamma){}^{64}\text{Cu}$	$E_r = 580$ eV	2 3 4 5 7 a b d	
${}^{115}\text{In}(n,n'){}^{115}\text{In}^m$	1.2 ... 5.8 MeV	1 3 4 5 6 7 8 10 a d	Low threshold; of particular importance /4/, /7/.
${}^{127}\text{I}(n,2n){}^{126}\text{I}$	10.0 .. 14.6 MeV	1 3 5 7 8 10 a d	High threshold.
${}^{197}\text{Au}(n,\gamma){}^{198}\text{Au}$	$E_r = 4.90$ eV	2 3 4 5 6 7 a b d	
${}^{235}\text{U}(n,f)^*$	0.19 ... 5.1 MeV	3 4 5 6 7 8 a c d	} * The yields for the fission products ${}^{95}\text{Zr}$ , ${}^{137}\text{Cs}$ , ${}^{140}\text{Ba}$ and ${}^{148}\text{Nd}$ belong to the second category.
${}^{238}\text{U}(n,f)^*$	1.5 .... 6.7 MeV	1 3 4 5 6 7 8 10 a c d	
${}^{237}\text{Np}(n,f)^*$	0.69 ... 5.6 MeV	3 5 6 7 8 10 a c d	
${}^{239}\text{Pu}(n,f)^*$	0.27 ... 5.1 MeV	3 4 5 6 7 a c d	

Tentative list of category 2 reactions

Thermal and intermediate energy range:

reaction	response remarks	general remarks	special remarks
$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	$E_r = 2850 \text{ eV}$	2 3 4 7 b d	
$^{30}\text{Si}(n,\gamma)^{31}\text{Si}$		4 7	
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$		3 d	
$^{51}\text{V}(n,\gamma)^{52}\text{V}$	$E_r = 4162 \text{ eV}$	2 4 7 b	
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$		3 4 7 c d	
$^{58}\text{Co}^m(n,\gamma)^{59}\text{Co}$			) Important to derive burn-up ) correction for nickel as ) fast neutron detector.
$^{59}\text{Co}(n,\gamma)^{59}\text{Co}$			
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	$E_r = 132 \text{ eV}$	2 3 4 6 7 a b c d	
$^{64}\text{Ni}(n,\gamma)^{65}\text{Ni}$		4 7	
$^{71}\text{Ga}(n,\gamma)^{72}\text{Ga}$	$E_r = 95 \text{ eV}$	2 b	
$^{75}\text{As}(n,\gamma)^{76}\text{As}$	$E_r = 47 \text{ eV}$	2 b	
$^{80}\text{Se}(n,\gamma)^{81}\text{Se}$	$E_r = 1965 \text{ eV}$	2 b	
$^{81}\text{Br}(n,\gamma)^{82}\text{Br}$	$E_r = 101 \text{ eV}$	2 b	
$^{93}\text{Nb}(n,\gamma)^{94}\text{Nb}$		4 9	Suggested as possible long term fluence detector /7/.
$^{98}\text{Mo}(n,\gamma)^{99}\text{Mo}$	$E_r = 12 \text{ and } 480 \text{ eV}$	2 4 7 b	
$^{100}\text{Mo}(n,\gamma)^{101}\text{Mo}$	$E_r = 97.3 \text{ and } 364 \text{ eV}$	2 b	
$^{103}\text{Rh}(n,\gamma)^{104}\text{Rh}$	$E_r = 1.257 \text{ eV}$	2 4 b	
$^{108}\text{Pd}(n,\gamma)^{109}\text{Pd}$	$E_r = 2.96 \text{ eV}$	2 b	
$^{109}\text{Ag}(n,\gamma)^{110}\text{Ag}^m$		4 7 c d	Together with $^{59}\text{Co}(n,\gamma)$ important in double foil technique to determine fluence of thermal and intermediate neutrons. Long $t_{1/2}$ replacement for $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$
$^{114}\text{Cd}(n,\gamma)^{115}\text{Cd}$	$E_r = 120 \text{ eV}$	2 b	
$^{115}\text{In}(n,\gamma)^{116}\text{In}^m$	$E_r = 1.46 \text{ eV}$	2 3 4 5 7 a b d	
$^{121}\text{Sb}(n,\gamma)^{122}\text{Sb}$		2 b	
$^{133}\text{Cs}(n,\gamma)^{134}\text{Cs}$	$E_r = 5.9 \text{ eV}$	2 b	
$^{139}\text{La}(n,\gamma)^{140}\text{La}$	$E_r = 72.4 \text{ eV}$	2 4 7 b	
$^{151}\text{Eu}(n,\gamma)^{152}\text{Eu}^m$		4 7	
$^{152}\text{Sm}(n,\gamma)^{152}\text{Sm}^m$	$E_r = 8.01 \text{ eV}$	2	
$^{164}\text{Dy}(n,\gamma)^{165}\text{Dy}$		4 7 a	
$^{175}\text{Lu}(n,\gamma)^{176}\text{Lu}$		4 7	
$^{176}\text{Lu}(n,\gamma)^{177}\text{Lu}$		4 7	
$^{181}\text{Tm}(n,\gamma)^{182}\text{Tm}$		d	Long $t_{1/2}$ replacement for $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$
$^{186}\text{W}(n,\gamma)^{187}\text{W}$		2 4 7 b	
$^{187}\text{Re}(n,\gamma)^{188}\text{Re}$	$E_r = 18.8 \text{ eV}$	2	
$^{191}\text{Ir}(n,\gamma)^{192}\text{Ir}$		2	
$^{198}\text{Pt}(n,\gamma)^{199}\text{Pt}$		2	
$^{232}\text{Th}(n,\gamma)^{233}\text{Th}$		3	
$^{238}\text{U}(n,\gamma)^{239}\text{U}$		3 4 6 7	Of particular importance /4/.

Fast energy range:

reaction	response remarks	general remarks	special remarks
$^{23}\text{Na}(n,2n)^{22}\text{Na}$		1	Very high threshold = 12.5 MeV
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	6.5...11.5 MeV	1 3 5 7 10	d
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	3.5... 9.3 MeV	1 3 4 5 6 7 8 10	d Of particular importance [4].
$^{28}\text{Si}(n,p)^{28}\text{Al}$	5.4...10.1 MeV	3	
$^{31}\text{P}(n,p)^{31}\text{Si}$	2.2... 7.0 MeV	1 3 4 5 7 8 9 10	d
$^{32}\text{S}(n,p)^{32}\text{P}$	2.5... 7.5 MeV	1 3 4 5 7 8 9 10 a	d
$^{34}\text{S}(n,\alpha)^{31}\text{Si}$	5.1...10.4 MeV	3	
$^{35}\text{Cl}(n,\alpha)^{32}\text{P}$	3.2... 8.0 MeV	3	
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	3.4... 9.1 MeV	1 3 4 5 7 8 9 10 a c	d
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	2.1... 7.0 MeV	1 3 4 5 7 8 9 10	d
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	6.6...12.8 MeV	1 3 4 5 7 8 9 10	d
$^{55}\text{Mn}(n,2n)^{54}\text{Mn}$		1 4 5 7 8 9 10	d Possible long term fluence monitor.
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	2.3... 7.8 MeV	1 3 4 5 6 7 8 10 a c	d Of particular importance [4].
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	5.5...11.0 MeV	1 3 4 5 7 8 9 10	d
$^{59}\text{Co}(n,p)^{59}\text{Fe}$		1	Might be of interest.
$^{59}\text{Co}(n,\alpha)^{56}\text{Mn}$		1 4 5 7 8 9 10	
$^{59}\text{Co}(n,2n)^{58}\text{Co}$		1	
$^{58}\text{Ni}(n,\alpha)^{55}\text{Fe}$		4 6 10	Of particular importance [4].
$^{58}\text{Ni}(n,2n)^{57}\text{Ni}$	13.2...17.0 MeV	1 3	Very high threshold.
$^{60}\text{Ni}(n,p)^{60}\text{Co}$	2.7... 9.6 MeV	3	
$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	6.1...11.3 MeV	1 3 5 6 7 8 10 a c	d Of particular importance [4].
$^{63}\text{Cu}(n,2n)^{62}\text{Cu}$	11.9...16.4 MeV	1 3 5 7 8 10	
$^{65}\text{Cu}(n,p)^{65}\text{Ni}$		1 3	d
$^{65}\text{Cu}(n,2n)^{64}\text{Cu}$		1 7 8 10	
$^{64}\text{Zn}(n,p)^{64}\text{Cu}$	2.3... 7.8 MeV	1 3 5 7 8 10	d
$^{64}\text{Zn}(n,2n)^{63}\text{Zn}$		1	
$^{90}\text{Zr}(n,2n)^{89}\text{Zr}$	12.5...16.7 MeV	3	d
$^{93}\text{Nb}(n,n')^{93}\text{Nb}^m$		4 6 7 10	Low threshold; of particular importance [4], [7].
$^{93}\text{Nb}(n,2n)^{92}\text{Nb}$		4 5 7 8 10	
$^{92}\text{Mo}(n,p)^{92}\text{Nb}$		5 7 10	
$^{94}\text{Mo}(n,p)^{94}\text{Nb}$			Possible long term fluence monitor.
$^{103}\text{Rh}(n,n')^{103}\text{Rh}^m$		4 6 7 8 10	Low threshold; of particular importance [4], [7].
$^{169}\text{Tm}(n,2n)^{168}\text{Tm}$			
$^{175}\text{Lu}(n,2n)^{174}\text{Lu}$			
$^{232}\text{Th}(n,f)$	1.5... 7.2 MeV	1 3 4 5 6 7 8 10	d Of particular interest [4]; fission product activities contain information on irradiation history.

Footnotes:

The numbers refer to data reviews; the letters refer to methods.

1. Energy dependent cross section data present in report EUR-119 by Liskien |1|.
2. Resonance parameters and resonance integrals given by Conolly, Kruyf and Schmidt |2|.
3. Energy dependent cross section data present in SAND-II cross section library (partly given in |3|).
4. Reactions recommended by IWGRRM for consideration by IAEA Nuclear Data Section |4|.
5. Evaluated integral cross sections given by Fabry |5|.
6. Status of energy dependent cross sections discussed by Vlasov |6|.
7. Status of nuclear data (such as  $T_{1/2}$ ,  $\sigma_0$ ,  $I'$ ,  $\langle\sigma\rangle^f$ ) given by Zijp |7|.
8. Evaluated cross section data compiled by Zijp |8|.
9. Status of energy dependent cross sections discussed by Vlasov |9|.
10. Energy dependent cross section data to be made available by the four Neutron Data Centres, as continuation of |1|.
  - a. Often used for flux density determinations (here a knowledge of integral cross sections and decay scheme data is required).
  - b. Often used in triple foil ("sandwich") techniques (here a knowledge of resonance activation integral and decay scheme data is required, and also supplementary data to calculate self-shielding factors).
  - c. Often used for fluence determinations (here a knowledge of integral cross sections and decay scheme data is required).
  - d. Often used in spectrum unfolding techniques using computer codes like SAND-II and SPECTRA (here a knowledge of energy dependent cross section data is required).

References:

- |1| Liskien, H., Paulsen, A.; "Compilation of cross sections for some neutron induced threshold reactions"  
Vol. I and II (Central Bureau for Nuclear Measurements, Geel, 1963)  
with supplementary sheets up to 1968.

- |2| Conolly, T.J., De Kruyf, F.; "An analysis of 24 isotopes for use in multiple foil (sandwich) measurements of neutron spectra below 10 keV" and Schmidt, J.J.; "Recommended resolved and statistical resonance parameters for 24 isotopes"  
KFK 718, EUR 3716e (Gesellschaft für Kernforschung, Karlsruhe, 1968).
- |3| Simons, R.L. and McElroy, W.N.; "Evaluated reference cross section library"  
BNWL-1312 (Battelle Memorial Institute, Pacific North-West Laboratories, Richland Washington, 1970).
- |4| Serpan Jr, C.Z. (compiler); Annual report 1971 for IAEA Working Group on Reactor Radiation Measurements (Naval Research Laboratory, Washington DC, 1972).
- |5| Fabry, A.; "Evaluation of microscopic cross sections averaged in the uranium 235 thermal fission neutron spectrum (for 29 nuclear reactions relevant to neutron dosimetry and fast reactor technology)"  
BLG-465 (Centre d'étude de l'énergie nucléaire, Mol, 1972).
- |6| Vlasov, M., Dunford, C., Schmidt, J.J., Lemmel, H.D.; "Status of neutron cross section data for reactor radiation measurements"  
INDC (NDS) - 47/L (IAEA, Vienna, 1972).
- |7| Zijp, W.L.; "Nuclear data for neutron metrology", RCN-73-017;  
Proc. Symposium on Applications of Nuclear Data in Science and Technology, held in Paris, 12-16 March 1973. (IAEA, Vienna, to be published).
- |8| Zijp, W.L.; "Compilation of evaluated cross section data used in fast neutron metrology"  
RCN-73-083 (Reactor Centrum Nederland, Petten, to be published).
- |9| Vlasov, M.F.; "Status of neutron cross section data for some reactions of interest for reactor radiation measurements"  
Working paper for the consultants meeting, 1973 (IAEA, Vienna, to be published).
- |10| Liskien, H.; Private communication (1973).

AGENDA

Consultants' Meeting on  
Nuclear Data for  
Reactor Neutron Dosimetry

Vienna, 10-12 September 1973

MONDAY, 10 September

- |      |  |           |
|------|--|-----------|
| I.   | Opening of Meeting   | Dunford   |
|      | A. Agency welcome  | Dunford   |
|      | B. Need for internationally recommended cross sections for neutron dosimetry reactions | Farinelli |
| II.  | Fluence Determination by Activation Methods  | Zi jp     |
|      | A. Critique of method(s) including accuracy, selection of reactions, limitations, etc. | Zi jp     |
| III. | Neutron Spectrum Determination by Activation Methods                                   | McElroy   |
|      | A. Critique of method(s) including accuracy, selection of reactions, limitations, etc. | McElroy   |
|      | B. Critical comparison of spectrum unfolding codes                                     | Dierckx   |
|      | C. Role of Standard Spectra in Differential Flux Determination                         | Dierckx   |
|      | D. Special problems at low energies $\leq 1$ MeV                                       | Najzer    |
|      | E. Determination of activity induced in monitors irradiated by neutrons                | Czock     |

- continued

TUESDAY, 11 September

- |     |   |         |
|-----|---|---------|
| IV. | Nuclear Data Assessment                                     | Liskien |
| A.  | Important nuclear reactions and nuclear quantities required | Zijp    |
| B.  | Status of knowledge of important nuclear data               | Vlasov  |
| C.  | Integral Measurements and Benchmarks                        | Fabry   |
| D.  | Needed data measurements                                    | Liskien |

WEDNESDAY, 12 September

- |     |  |           |
|-----|--|-----------|
| V.  | Selection of Standards and Reactions for which Internationally Accepted Values Are Desired | Farinelli |
| A.  | Thermal and intermediate energies  | Zijp      |
| B.  | Fast energies  | McElroy   |
| VI. | Recommendations to IAEA for Future Activities  | Vlasov    |

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