NEUTRON SPECTRUM DETERMINATION BY MULTIPLE FOIL ACTIVATION METHOD

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1. Introduction

In case of determining neutron flux density spectra from multiple foil activation measurements one meets the problem of solving the following type of activation integral equations:

$$A_i = \int_0^\infty \sigma_i(E)\Phi(E)\,dE \qquad i=1(1)n \tag{1}$$

where:

A_i — is the experimentally determined saturation activity per target nucleus of the i^{th} foil detector extrapolated to infinite dilution
$\sigma_i(E)$ — is the energy dependent activation cross section
$\Phi(E)$ — is the neutron flux density per unit energy interval
n — is the number of foil detectors used in the experiment.

In the practice one uses a discrete interval description of the neutron energy range considered rather than a continuous representation by analytical functions. Since the number of equations in this case is generally much smaller than the number of unknowns, that is the freedom of the system is rather high, the solution is not unique and the neutron flux spectrum cannot be derived from these data without a priori assumption on the initial neutron spectrum based on all physical informations available in the given case.

Several neutron spectrum unfolding computer codes [1—6, etc.] based on different philosophies have been used so far in the literature for the solution of this problem. In the present report the program SANDBP developed in our Institute is described [12], and the neutron spectrum in the centre of the core of our nuclear reactor determined with aid of this program is presented.

2. The neutron spectrum unfolding code SANDBP

This code is a version of the well-known program SAND II, being enlarged and modified in its algorithm. The use of this program involves several adventages compared to the old version:

a) the activity and cross section errors can be taken into account in the unfolding procedure;
b) different response data deriving from the solution neutron spectrum can be calculated (e.g. neutron dose rate, neutron material damage characteristics such as dpa, etc.);
c) informations can be obtained about the magnitude of errors involved in the solutions;
d) the so called GOS-values characterizing the goodness of solution can be determined;
e) the covariance and correlation matrices of the solution spectrum can be determined;
f) the variety of output data is greater than that one in case of the old program.

2.1. Principles of the SAND iterative method

The computer code SAND II has been developed to determine the neutron flux density spectrum from activity measurements of foil detectors activated in the neutron environment to be investigated [1]. The mathematical procedure for the method involves the selection of an initial approximation spectrum, and subsequent perturbation of that spectrum by iterative adjustments, to yield a solution spectrum producing calculated activities that agree within a given error limit with the measured activities.

This way the code provides a "best fit" neutron flux density spectrum for a given input set of infinitely dilute foil activities.

The iterative algorithm used in the program may be written as

$$\Phi_j^{k+1} = \Phi_j^k \exp(C_j^k) \qquad j = 1(1)m \tag{2}$$

where

$$C_j^k = \frac{\sum_{i=1}^{n} W_{ij}^k \ln(A_i/A_i^k)}{\sum_{i=1}^{n} W_{ij}^k} \qquad j = 1(1)m \tag{3}$$

and

Φ_j^k — is the k^{th} iterative neutron flux density (assumed constant) over the j^{th} energy interval,

C_j^k — is the k^{th} iterative flux density correction term for the j^{th} energy interval,

A_i^k — is the calculated saturation activity for the i^{th} detector, based on the k^{th} iterative neutron spectrum,

m — is the total number of energy intervals,

n — is the number of detectors used.

The W_{ij}^k is a weighting function, which is in good approximation proportional to the relative response for the i^{th} detector:

$$W_{ij}^k = A_{ij}^k / A_i^k \qquad \begin{matrix} i = 1(1)n \\ j = 1(1)m \end{matrix} \tag{4}$$

$$A_{ij}^k = \Phi_j^k \sigma_{ij}(E_{j+1} - E_j) \qquad \begin{matrix} i = 1(1)n \\ j = 1(1)m \end{matrix} \tag{5}$$

$$A_i^k = \sum_{j=1}^{m} A_{ij}^k \qquad i = 1(1)n \tag{6}$$

and σ_{ij} is the reaction cross section for the i^{th} detector (averaged constant) over the j^{th} energy interval.

The energy range of the solution spectrum is represented in max. 620 intervals depending on the cross section library. The standard library of the program [1] contains 620 energy groups from 10^{-10} MeV to 18 MeV (45 per decade up to 1 MeV and 170 between 1 MeV and 18 MeV). The code includes the provision for calculating flux density attenuation by foil cover materials including cadmium, boron-10 and gold.

2.2. Characteristics of the program SANDBP

2.2.1. Weighting function and solution criteria

Due to the form of the weighting function defined by Eq. (4), the structure of the reponse function for a given foil detector (therefore cross section structure) may be reflected after several iterations in the solution spectrum [1, 7, 8]. This effect may be pronounced especially that case if substantial activity measurement errors or library cross section errors are present in the data set applied, and as a result an extended number of iterations is requested to achieve an appropriate solution.

To decrease the role of artificial structures in the solution spectrum the weighting function in Eq. (4) has been modified taken into account the activity and cross section errors, as follows:

$$W_{ij}^k = \frac{A_{ij}^k}{A_i^k}\left(\frac{1}{(\delta A_i)^u} \times \frac{1}{(\delta\sigma_{ij})^v}\right) \qquad \begin{matrix} i=1(1)n \\ j=1(1)m \end{matrix} \tag{7}$$

where (δA_i) and $(\delta\sigma_{ij})$ are the relative standard errors of the activity and cross section values, respectively. The exponents u and v are input parameters and can arbitrarily be chosen. This possibility enables the user to take into consideration the quality of the input data during the unfolding procedure.

If $u=v=0$ no weighting with errors is used, that is the weighting function agrees with that one used in the program SAND II.

As the (positive) value of u and v is increasing the solution spectrum will be determined more and more by the more accurate activity and cross section data. Generally, if the errors of the different cross section values in the library and/or the activity errors within the detector set applied are near each other, a value between 0.5 and 1 for u and v is reasonable. Otherwise a value between 1.5 and 2 is recommended [10]. Too large value of these parameters equals of discarding the reactions having larger errors from the unfolding procedure (the relative weight of these reactions becomes too small, see Eqs (3) and (7)).

The solution criteria of the original program have also been extended. The standard deviation of measured to calculated activity ratios is calculated after each iteration step, expressed in per cents:

$$DEV_{\text{per cent}}^k = \sqrt{\frac{\sum_{i=1}^{n}\left(\frac{A_i - A_{ci}^k}{A_{ci}^k}\right)^2 - \frac{1}{n}\left(\sum_{i=1}^{n}\frac{A_i - A_{ci}^k}{A_{ci}^k}\right)^2}{n-1}} \times 100 \tag{8}$$

and in confidence interval units:

$$DEV_{\text{conf}}^k = \sqrt{\frac{\sum_{i=1}^{n}\left(\frac{A_i - A_{ci}^k}{A_{ci}^k \delta A_i}\right)^2 - \frac{1}{n}\left(\sum_{i=1}^{n}\frac{A_i - A_{ci}^k}{A_{ci}^k \delta A_i}\right)^2}{n-1}} \tag{9}$$

(where A_{ci}^k is the calculated saturation activity of the i^{th} foil in the k^{th} iteration step) taken into account in the latter case the uncertainty of the experimental data. The iteration is then stopped if any of the above defined values is smaller than the one defined as input. The specification of larger u values in Eq. (7) will increase the speed of the convergence defined by Eq. (9).

The iteration procedure is ended also in that case if the standard deviation becomes stable within less that 1% in two successive iterations, or if the maximum number of iterations specified by the input is achieved.

2.2.2. Activity run

The program involves the possibility to calculate detector activities for a given neutron spectrum [1]. This procedure is performed in an ACTIVITY run. The activities per target atom are calculated in units of Bq, consequently they are the integrals of the reponse functions (see point 2.1.).

2.2.3. Calculation of response functions

Any response function (reaction rate, dose rate, dpa-value, etc.) deriving from the solution neutron spectrum can be calculated with aid of the program:

$$R_j = p_j \Phi_j (E_{j+1} - E_j) \qquad j = 1(1)m \tag{10}$$

or

$$R = \sum_{j=1}^{m} p_j \Phi_j (E_{j+1} - E_j) \tag{11}$$

where R_j — is a response function
p_j — is the desired conversion factor.

Using this procedure neutron dose rate values can directly be calculated from the neutron flux density spectrum eliminating the approximation errors deriving from the calculation of average neutron energy over the spectrum regions considered [9].

Nevertheless, the average neutron energy ($\bar{E}$) can also be determined, using neutron density values as weighting functions rather than neutron flux density values being involved in the original SAND II algorithm.

Thus

$$\bar{E}_k = \frac{\sum_{j=k}^{m} \Phi_j \sqrt{E_j}}{\sum_{j=k}^{m} \frac{\Phi_j}{\sqrt{E_j}}} \tag{12}$$

where Φ_j is the neutron flux value in the j^{th} energy group.

2.2.4. Monte-Carlo error analysis

a) Calculation of error limits

With aid of the SANDBP's Monte-Carlo program error analysis can be performed. In the course of the Monte-Carlo runs the input activities and cross sections (assumed that these values follow a normal distribution) are varied in accordance to their random errors:

$$A_i^r = A_i + g \times \mathrm{d}A_i \qquad i = 1(1)n \tag{13}$$

where

A_i^r — is the r^{th} random value of the i^{th} measured activity A_i,

$\mathrm{d}A_i$ — is the standard error of A_i,

g — is a random number from normal distribution.

Similarly,

$$\sigma_{ij}^r = \sigma_{ij} + g \times \mathrm{d}\sigma_{ij} \qquad \begin{matrix} i = 1(1)n \\ j = 1(1)m \end{matrix} \tag{14}$$

where σ_{ij}^r — is the r^{th} random value of the i^{th} reaction cross section in the j^{th} energy group.

The cross section values are varied independently from each other in m energy groups. No correlation is assumed between cross section values of different energy groups: $r_{jl} = 0$ for all j and l.

Performing the neutron spectrum unfolding procedure with these modified input data, the relative standard error in the j^{th} energy group of the solution spectrum can be written as:

$$\delta\Phi_j = \sqrt{\frac{\sum_{r=1}^{k} (\varphi_j^r)^2 - \frac{1}{k}\left(\sum_{r=1}^{k} \varphi_j^r\right)^2}{(k-1)\times(k-2)}} \times \frac{1}{\frac{1}{k}\sum_{r=1}^{k} \varphi_j^k} \qquad j = 1(1)m \tag{15}$$

where k is the number of Monte-Carlo runs. $k \geqq 10$ is allowed by the program.

The relative errors of the response data are derived on the analogy of the Eq. (15).

As it is seen from this equation the error limits calculated by the program are simple standard errors (67.5% confidence level).

b) Goodness of solution

In separate Monte-Carlo runs the input spectrum (Φ^{inp}) is varied:

$$\varphi_j^{\text{inp},\,r} = \Phi_j^{\text{inp}} \times (1 + 0.1\ \text{g}) \qquad j = 1(1)m \tag{16}$$

and the change of the output spectrum in the function of the input spectrum is used to characterize the *g*oodness *o*f solution (GOS):

$$\text{GOS}_j = \left. \frac{\partial \varphi_j^{\text{out}}}{\partial \varphi_j^{\text{inp}}} \right|_{\varphi_j^{\text{inp}} = \Phi_j^{\text{inp}}} \qquad j = 1(1)m \tag{17}$$

Let the functionality between the point pairs (φ_j^{out} and φ_j^{inp}) be approximated by polynomial regression:

$$\varphi_j^{\text{out}} = \sum_{l=0}^{M} a_l \times (\varphi_j^{\text{inp}})^l \qquad j = 1(1)m \tag{18}$$

$$\text{GOS}_j = \sum_{l=0}^{M-1} (l+1) \times a_{l+1} \times (\Phi_j^{\text{inp}})^l \qquad j = 1(1)m \tag{19}$$

where M is the degree of the polynom. $M \leqq 3$ is used in the curve fitting procedure.

GOS = 0 means that the output spectrum does not depend on the input spectrum. A GOS value differing from zero indicates a "worse" solution.

The latter may happen in the energy regions poorly or not covered by the response of the detector set applied. In this case the algorithm of the program performs an interpolation, and the solution spectrum may simply reflect the input spectrum.

c) Covariance and correlation matrices

As a result of the Monte-Carlo runs one has a series of solution spectra, which can then be used to determine the covariance and correlation matrices of the nominal solution spectrum.

The value of covariance between the neutron flux density values of the j^{th} and l^{th} energy groups can be expressed as:

$$s_{jl} = \frac{\sum_{r=1}^{k} \varphi_j^r \varphi_l^r}{k} - \frac{\left(\sum_{r=1}^{k} \varphi_j^r\right) \times \left(\sum_{r=1}^{k} \varphi_l^r\right)}{k^2} \qquad \begin{matrix} j = 1(1)m \\ l = 1(1)m \end{matrix} \tag{20}$$

where k is the number of Monte-Carlo runs.

The value of the correlation coefficient for the same relation:

$$r_{ji}=\frac{\sum_{r=1}^{k}\varphi_j^r\varphi_l^r-\frac{\left(\sum_{r=1}^{k}\varphi_j^r\right)\times\left(\sum_{r=1}^{k}\varphi_l^r\right)}{k}}{\sqrt{\sum_{r=1}^{k}(\varphi_j^r)^2-\frac{\left(\sum_{r=1}^{k}\varphi_j^r\right)^2}{k}}\times\sqrt{\sum_{r=1}^{k}(\varphi_l^r)^2-\frac{\left(\sum_{r=1}^{k}\varphi_l^r\right)^2}{k}}}\qquad\begin{matrix}j=1(1)m\\ l=1(1)m\end{matrix}\qquad(21)$$

2.2.5. Code input

The program requires as input

a) the measured (input) activity values of the foil detectors corrected for neutron selfshielding,

b) cross section library for the detector set applied,

c) a guess neutron spectrum (input spectrum),

d) relative errors of the activity and cross section values.

The SAND II program package contains a subroutine (VIF), which allows the recording of user's problem data with a maximum freedom of restrictive format. The input format and the input data deck sequence description can be found in the user's guide [11].

2.2.6. Code output

The program can supply the following outputs:

a) In case of ITERATION (spectrum unfolding) run:

— listing of all cards of the input data set;

— if the solution is achieved a table summarizing the final iterative results is given, including the tabulation of the solution neutron spectrum.

b) In case of MONTE-CARLO run:

— all the above items are given followed by their relative errors;

— the GOS-values are written;

— covariance and correlation data of the solution spectrum are written on a magnetic tape and they can also be printed.

c) In case of ACTIVITY run items similar to those ones given in ITERATION run can be obtained.

d) Output to plotter: Due to corresponding specifications in the input data set the solution neutron spectrum and GOS-values can be plotted.

2.2.7. Coding language and computer

The program is written in IBM FORTRAN IV (G) language for an 1024 Kbyte R-40 type computer under the control of the operating system IBM OS-MVT. The code needs a memory region of 300 Kbyte.

3. Neutron spectrum determination

The Budapest Technical University's nuclear reactor is a swimming pool type light-water reactor with a maximum power of 100 kW [15]. The neutron spectrum in the core position D5 (Fig. 1) was determined. For this purpose sets of activation detectors were irradiated and the neutron spectrum was unfolded using the computer program SANDBP.

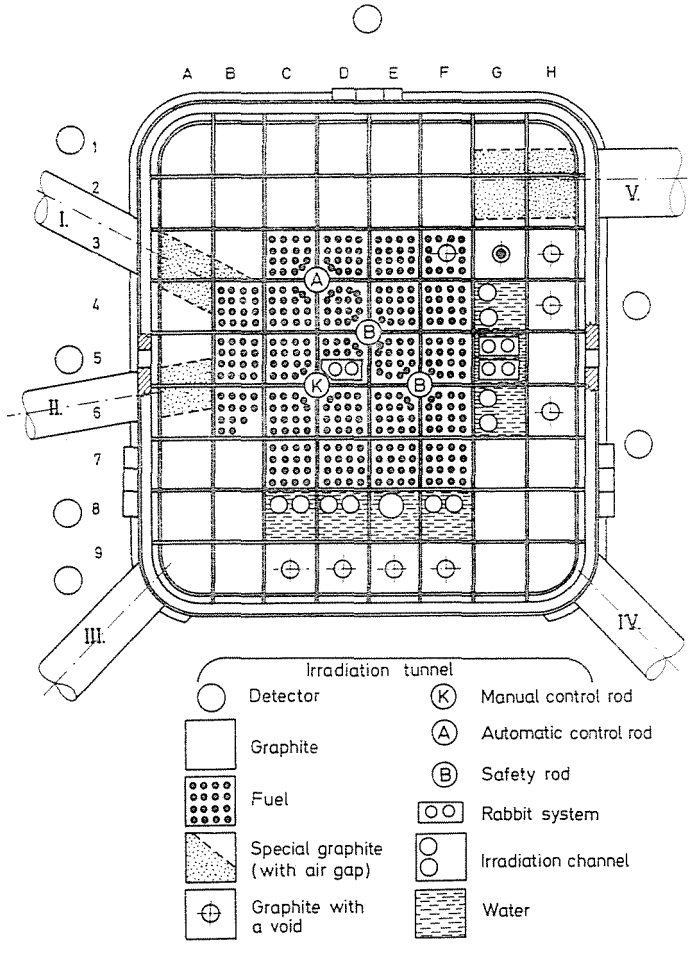

Fig. 1. Horizontal cross section of the 10 kW reactor core

3.1. *Experimental procedure*

The activation detectors were used in form of thin foils, irradiated in two separate runs at a reactor power level of 10 kW [9]. The foils were covered by aluminium or cadmium, respectively. Three separate foils (Au, Co, Ni) with different responses were used to monitor the reactor power level.

The irradiated foils were counted by means of a Ge(Li) semi-conductor detector and a 4096 channel pulse amplitude analyser system. The detection peak efficiency curve of the spectrometer had previously been established.

The saturation activities of the radionuclides of interest, which serve as the input of the spectrum unfolding program, are shown in Table 1.

Table 1

Saturation activities per target atom for detectors used in the unfolding procedure

Reaction	Cover	Half-life	A_{sat}/target atom [Bq]	$\frac{\Delta A_{sat}}{A_{sat}}$ [%]
^{164}Dy(n, γ)^{165}Dy*	—	139.2 min	6.35 E-10	5.82
^{164}Dy(n, γ)^{165}Dy*	Cd	139.2 min	6.32 E-12	1.89
^{27}Al(n, p)^{27}Mg	Cd	9.46 min	9.01 E-16	12.49
^{27}Al(n, α)^{24}Na	Cd	15.03 h	1.60 E-16	4.88
^{45}Sc(n, γ)^{46}Sc*	Cd	84.0 d	3.45 E-12	0.63
^{56}Fe(n, p)^{56}Mn*	Cd	2.587 h	1.35 E-17	5.78
^{58}Ni(n, p)^{58m}Co	—	71.3 d	2.47 E-14	2.80
^{58}Ni(n, p)^{58m}Co	Cd	71.3 d	2.48 E-14	7.17
^{115}In(n, γ)^{116m}In	—	53.34 min	1.75 E-11	1.40
^{115}In(n, γ)^{116m}In	Cd	53.34 min	1.22 E-11	0.82
^{55}Mn(n, γ)^{56}Mn	—	155.2 min	5.73 E-12	8.62
^{55}Mn(n, γ)^{56}Mn	Cd	155.2 min	2.79 E-13	8.20
^{197}Au(n, γ)^{198}Au	—	2.695 d	5.86 E-11	2.94
^{197}Au(n,γ)^{198}Au	Cd	2.695 d	3.22 E-11	1.60
^{115}In(n, n')^{115m}In	Cd	4.5 h	4.56 E-14	1.30
^{47}Ti(n, p)^{47}Sc	Cd	80.4 h	4.29 E-15	1.13
^{48}Ti(n, p)^{48}Sc	Cd	44.1 h	3.92 E-17	0.74

* Deleted from the unfolding procedure

3.2. *Cross section library and neutron selfshielding*

The calculations were performed for 620 energy groups using the cross section library DOSCROS77 [13]. For all the detectors the neutron self-shielding correction was obtained by applying modified cross section

values in the cross section library, correcting each of the 620 group cross section values of the reactions as follows:

$$(\sigma_j)_{\text{corr}} = \sigma_j G(\tau_j) \tag{22}$$

where:

$$G(\tau_j) = \frac{1 - 2E_3(\tau_j)}{2\tau_j} \tag{23}$$

and $\tau_j = N\sigma_j t$, is the thickness of the detector measured in units of mean free path for absorption;

N is the number of target atoms per unit volume;

σ_j is the total cross section for goup j;

t is the thickness of the detector;

E_3 is the exponential integral of the third order.

This approach enables us to introduce group cross sections corrected for selfshielding. In absence of appropriate total cross section data capture cross sections were used. To minimize the selfshielding effects thin dilutions of detector materials were applied, except the threshold detectors.

3.3. *Input neutron spectrum*

The input neutron spectrum was a LWR type WWR reactor spectrum available from reactor physics calculations in 49 energy groups [14] modified in accordance with our former measuring results. Using the special interpolation and extrapolation procedure included in the unfolding program this information was converted into a 620 groups spectrum. The joining point between the Maxwellian and intermediate neutron energy region was determined at energy $E \sim 2 \times 10^{-7}$ MeV. The input spectrum is seen in Fig. 2.

3.4. *Results*

The reaction rates of 17 activation detectors were determined, however 4 reactions had to be deleted from the analysis showing incompatibility with respect to the other detectors.

The aim of the investigations was, besides determining the actual form of the neutron spectrum, to show the effect of the weighting function defined by the program SANDBP on the unfolding procedure. For this purpose two different runs were performed in conditions shown by Table 2.

In case of $u = v = 0$ no weighting with error values was used, while the conditions $u = 2$, $v = 0$ took the activity errors of the detector set into account

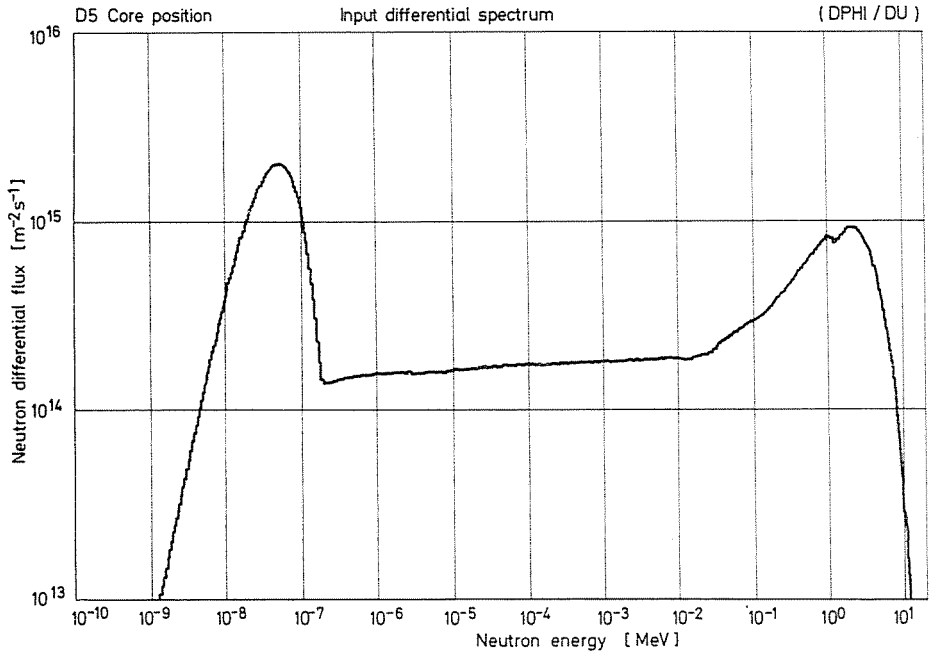

Fig. 2. The input neutron spectrum

Table 2

Unfolding conditions and integral parameters of the solution neutron spectra

	$u=v=0$	$u=2,\ v=0$
No. of iterations	3	3
Ratio of measured to calculated activities in confid. interval units	4.50	3.79
Φ_{total} [$m^{-2}s^{-1}$]	1.004E16	1.008E16
$\Phi_{>1\,\text{MeV}}$ [$m^{-2}s^{-1}$]	1.792E15	1.817E15
$\Phi_{>.1\text{MeV}}$ [$m^{-2}s^{-1}$]	3.404E15	3.562E15
$\Phi_{>.2\,\text{eV}}$ [$m^{-2}s^{-1}$]	6.185E15	6.596E15

during the unfolding procedure. As the difference between the error values of the detector activities was rather large in the present case (see Table 1.), $u=2$ was chosen to ensure a bigger role for the more accurate reaction rates in determination of the solution neutron spectrum [10]. The neutron spectra obtained and some integral parameters referring to them are presented in Figures 3 and 4 and in Table 2.

From the figures it is seen that the solution only slightly affects the thermal part of the input spectrum, but it changes the intermediate and fast

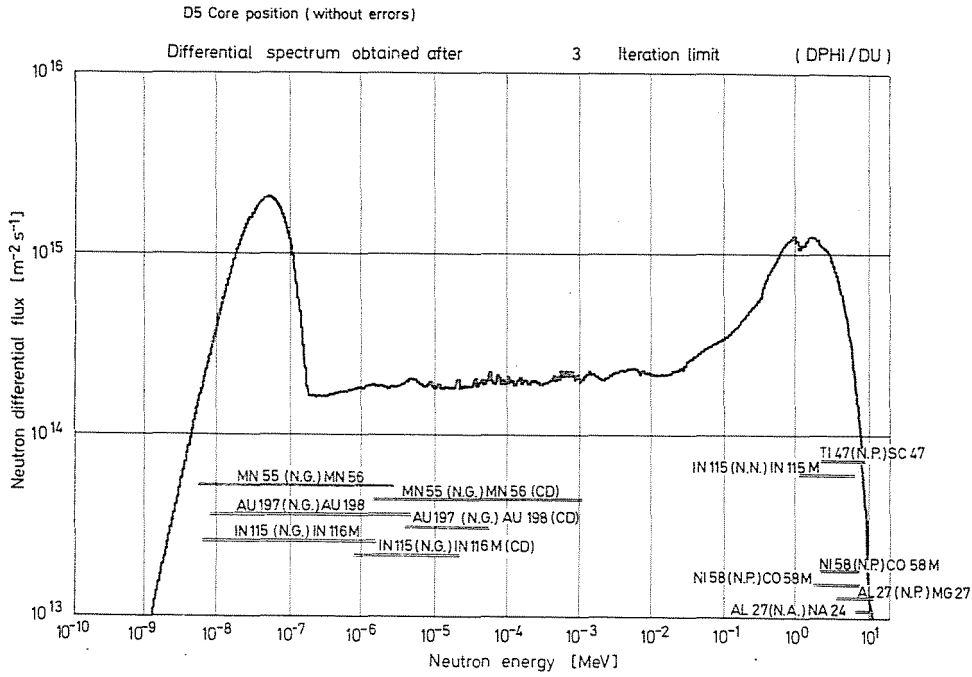

Fig. 3. Solution neutron spectrum obtained after 3 iterations in case of $u=v=0$

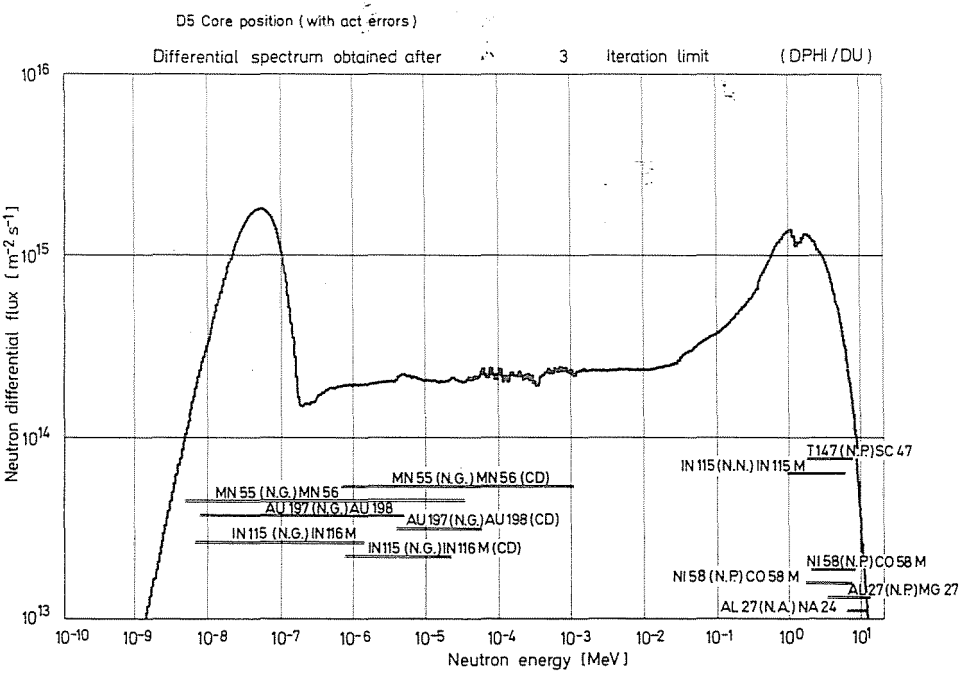

Fig. 4. Solution neutron spectrum obtained after 3 iterations in case of $u=2$, $v=0$

energy region to a higher degree. Both solution spectra have some—probably largely artificial— structure in the intermediate neutron energy region, but in case of $u=2$ the shape of the spectrum is smoother showing the effect of weighting with the activity errors. This solution seems to be more realistic. In the energy region (between 10^{-3} and about 1 MeV) poorly (or not) covered by the response of the detector set applied the solution obtained with error weighting reflects the input spectrum. However, in the other case some artificial structure is introduced also in this part of the spectrum. In the fast energy region, in the vicinity of 1 MeV, the effect of the oxygen resonance scattering can be seen.

The intergral data in Table 2 also show the difference between the two solutions.

The correlation matrices for the solution neutron spectra are given in Figures 5 and 6 in perspective representation. Strong correlation can be observed between the flux density values

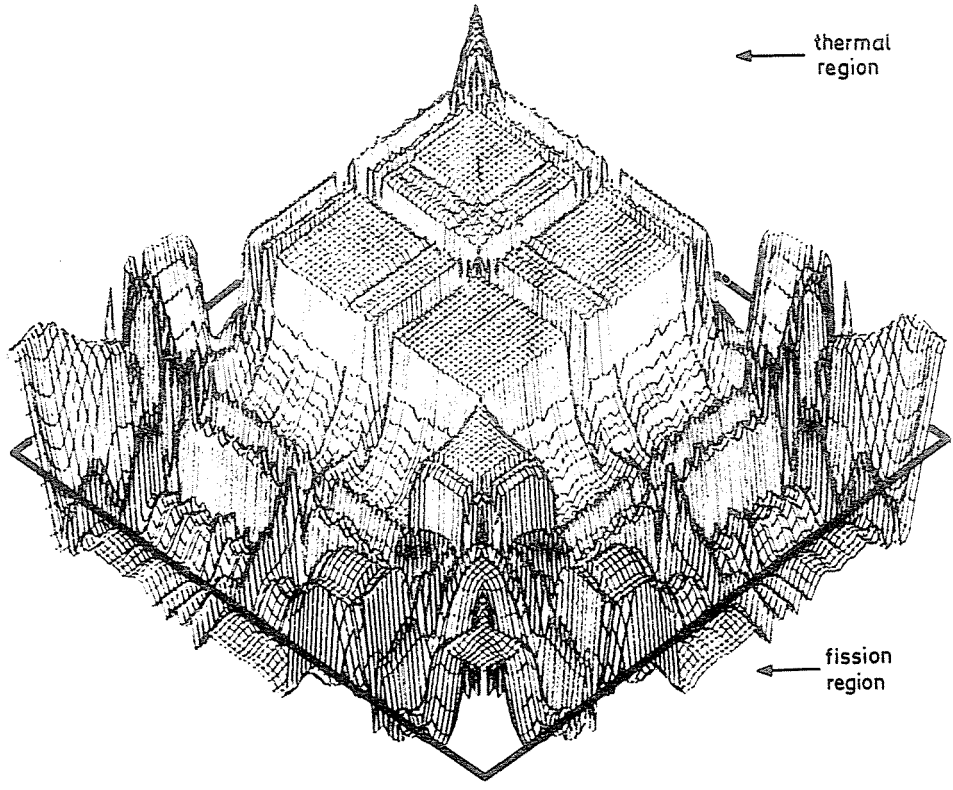

Fig. 5. Correlation matrix for the solution neutron spectrum in case of $u=v=0$

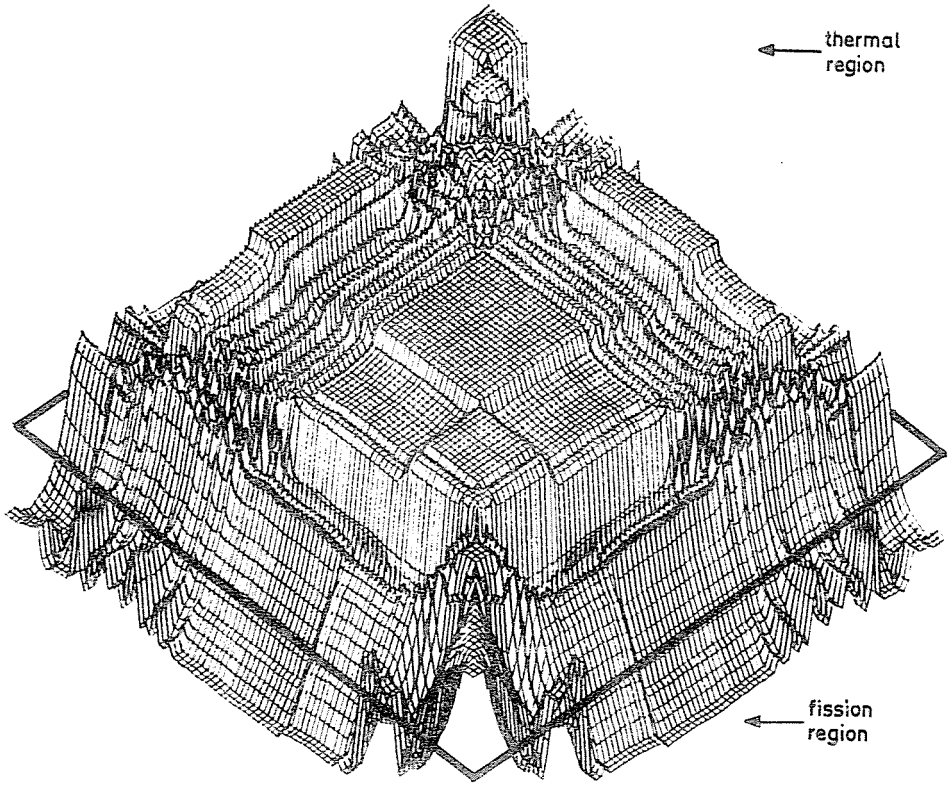

Fig. 6. Correlation matrix for the solution neutron spectrum in case of $u=2$, $v=0$

— in the thermal
— in the intermediate and
— in the fast

neutron energy regions.

If the activity errors are taken into account in the unfolding procedure ($u=2$, $v=0$) the solution is determined by the more accurate activity values. As a result, the correlation matrix in this case has a less perturbed smoother character (compare Figures 5 and 6). The investigation of the structures in the correlation matrices and their interpretation is presently in progress.

Summary

The main features of the neutron spectrum unfolding code SANDBP are described and some results obtained using this code are presented.

The program determines the neutron flux density spectrum from multiple foil activation measurements using an iterative method. Response functions deriving from the unfolded neutron spectrum can also be calculated, furthermore Monte-Carlo error analysis can be performed.

The neutron spectrum in the centre of the core of the Budapest Technical University's nuclear reactor was measured and unfolded with aid of this program.

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