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**Determination of particle flux spectra with multireaction activation
detectors**

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Finland

Thesis for the degree of Doctor of Technology to be presented with due permission for public examination and criticism in Auditorium F 1 at Helsinki University of Technology, on the 15th of June, 1984, at 12 o'clock noon.

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ABSTRACT

Activation detectors constitute one method for measuring particle flux spectra at reactors and accelerators. The determination of the energy spectrum from a few discrete measurements leads to an underdetermined system of integral equations which does not have a unique mathematical solution, but by using some prior information on the spectrum and special unfolding programs physically relevant solutions can be obtained. The general purpose, least-squares regularization type unfolding program LOUHI78 is described and it is applied to unfolding high-energy secondary hadron spectra measured with multireaction spallation detectors at the CERN Super Proton Synchrotron and reactor neutron energy spectra measured with multicomponent activation detectors. The design, fabrication and testing of multicomponent detectors, where a number of component materials is irradiated and measured together, are also described.

PREFACE

This thesis deals with the use of multireaction activation detectors for particle flux spectrum measurements at reactors and accelerators. The main emphasis is on the computational methods used for analysing the measurements.

The work related to nuclear reactors has been carried out at the Nuclear Engineering Laboratory of Helsinki University of Technology in a research group lead by Professor Routti, whom I wish to thank for an interesting subject and encouraging advice. I also wish to thank all members of the group, especially Dr. Markku Koskelo, Mr. Pertti Aarnio and Mr. Peter Lund.

The spallation detector irradiation was carried out at CERN, Geneva. I wish to thank Dr. Klaus Goebel for the opportunity to work at the CERN Radiation Protection Group, Drs. Graham Stevenson and Alberto Fasso for arranging the spallation detector irradiation, and Professor Ranft for advice on hadron cascade simulations.

The financial support by the Emil Aaltonen foundation is gratefully acknowledged.

Helsinki, March 1984
Jorma Sandberg

CONTENTS

Preface	3
List of publications	5
The author's contributions	6
1 Introduction	7
2 Multicomponent activation detectors for reactor neutrons	11
2.1 The multicomponent principle	11
2.2 Comparison with separate activation foils	19
2.3 Limitations of multicomponent detectors	23
3 Multireaction spallation detectors for high energy hadrons	26
4 Neutron spectrum unfolding	30
4.1 The few-channel unfolding problem	30
4.2 LOUHI-type unfolding programs	31
4.3 Other unfolding programs	35
5 Conclusion	48
References	50
Corrigenda	57

LIST OF PUBLICATIONS

This thesis consists of this introductory report and the following eight publications.

- I P.A. Aarnio, M.J. Koskelo, P.D. Lund, V.K. Maunula, J.T. Routti, J.V. Sandberg and H.J. Takala, "Multicomponent Activation Detectors for Reactor Neutron Spectroscopy", Nucl. Technol. 57 (1982) 318-328.
- II J.V. Sandberg and P.D. Lund, "Determination of Reactor Neutron Spectra with Multicomponent Activation Detectors", J. Radioanal. Chem. 76 (1983) 151-170.
- III J.T. Routti and J.V. Sandberg, "General Purpose Unfolding Program LOUHI78 with Linear and Nonlinear Regularizations", Comput. Phys. Commun. 21 (1980) 119-144.
- IV J.V. Sandberg and J.T. Routti, "Unfolding Triga Reactor Neutron Spectra from Multicomponent Activation Detector Data with LOUHI82", Nucl. Technol. 63 (1983) 170-175.
- V J.V. Sandberg, "Application of the Program LOUHI82 to Reactor Neutron Spectrum Unfolding", Helsinki University of Technology Report TKK-F-A524 (1983).
- VI J.V. Sandberg, "On the Feasibility of Multicomponent Activation Detectors for Fusion Reactor Neutronics Measurements", Nucl. Instr. Meth. 206 (1983) 227-234.

- VII J.V. Sandberg, "Angular and Energy Distribution Measurements of Secondary Hadron Fluxes with Multireaction Spallation Detectors at CERN SPS", Nucl. Instr. Meth. 200 (1982) 211-218.
- VIII J.T. Routti and J.V. Sandberg, "FORTRAN Program SPALL for Computing Spallation Reaction Cross Sections", Comput. Phys. Commun. 23 (1981) 411-426.

In the introduction these publications are referred to with Roman numerals.

THE AUTHOR'S CONTRIBUTIONS

The author's contribution to publications I and II includes participation in the design and testing of multicomponent detectors, development of analysing methods and evaluation of the results, whereas the actual measurements were carried out by other members of the group.

The contribution to publications III-V comprises revising, documenting and testing the LOUHI-type unfolding programs and performing application runs.

The work described in publications VI-VIII was carried out mainly by the author.

Publications II-VIII and parts of publication I were written by the author of this thesis.

1 INTRODUCTION

Neutrons are electrically uncharged elementary particles which can be detected through their nuclear interactions with atomic nuclei. Two types of interactions can be used to measure neutron energy distributions. In activation reactions a radioactive product nucleus is formed, and the number of reactions can be determined by measuring the intensity of radiation from the decay of product nuclei. The number of activation reactions is related to the density and energy distribution of the neutron field. In scattering interactions, part of the neutron kinetic energy is transferred to a proton or a heavier nucleus. Since they are electrically charged particles, their energy distribution can be measured and it can be used to determine the neutron energy distribution.

Intense neutron fields are found in nuclear reactors and at neutron generators and isotope neutron sources. The neutron field determines, e.g., the power production in a reactor and the generation of radioactive nuclides, and it makes an important contribution to radiation damage and biological effects. Neutrons are also present in the stray radiation fields at high-energy particle accelerators, together with other hadrons, mainly protons and pions. Activation reactions can also be used to detect these particles.

The intensity of a neutron field at a point is characterized by the energy dependent flux, also called differential flux, flux spectrum or briefly flux /1,2,3,4/

$$\phi(E) = n(E) v(E), \quad (1)$$

where

E is the neutron energy,

$v(E) = \sqrt{2E/m_n}$ is the neutron speed,

m_n is the neutron mass,

$n(E)$ is the neutron density per volume unit and energy unit at energy E .

The integral of $\phi(E)$ over the energy range is the total flux. The integral of the total flux over a time interval is called neutron fluence. The nomenclature is not quite uniform. For example ICRU /5/ and ASTM /4/ recommend the term flux density or fluence rate instead of flux. The term flux density has been lately used interchangeably with flux, but the term fluence rate is seldom used in reactor physics literature.

The rate R_x of a given reaction x is related to the neutron flux through the equation

$$R_x = \int_0^{\infty} \sigma_x(E) \phi(E) dE, \quad (2)$$

where $\sigma_x(E)$ is the cross section of reaction x , describing the probability of the reaction. If the microscopic cross section is used, R_x is normalized per one target nucleus, and if the macroscopic cross section is used, the normalization is per unit volume.

The reaction rate of interest may be the fission rate determining the power generation in a nuclear reactor, or the activation rate, or it may describe radiation damage or biological effects due to the neutron field.

For the design and operation of nuclear reactors and other neutron producing facilities, the neutron flux should be known as a function of position and time. It can be solved with reactor physics calculations, but validating measurements are also required.

Neutron flux measurements can be made with activation detectors, which are based on the production of radioactive nuclides through nuclear transmutations induced by neutrons in suitable detector materials. /6/

The saturation activity per one detector nucleus is equal to the reaction rate and it is related to the flux through eq. (2). When several activation reactions with different energy dependence of cross section are used, a group of linear integral equations is obtained for the flux $\phi(E)$. This group is

underdetermined, since a finite number of measurements cannot define uniquely a continuous distribution. The solution of these integral equations is commonly called spectrum unfolding, and it can be carried out e.g. by the program LOUHI78 described in publication III.

The type of activation detectors used in each application depends on the neutron energy range of interest. In Fig. 1 is shown the neutron energy spectrum in a typical thermal reactor. At high energies, above about 1 MeV, the spectrum resembles the fission spectrum, i.e. the energy distribution of neutrons produced in fission reactions. At the intermediate energies, down to about 0.5 eV the spectrum has an energy distribution of the $1/E$ -type. At low energies the neutrons are approximately in thermal equilibrium with their environment and they have a Maxwellian energy spectrum /6/.

Threshold-type activation reactions have cross sections which rise rapidly to a maximum value above a threshold energy and then remain approximately constant. Many (n, α) , (n, p) , (n, n') , and $(n, 2n)$ reactions have threshold energies in the range 1-15 MeV and they can be used to measure the fast neutron spectrum.

Radioactive capture or (n, γ) -reactions generally have at low energies cross sections with a $1/v$ -dependence on neutron speed. At higher energies the cross sections may have very narrow peaks corresponding to resonance absorption at discrete energies. The resonance energies of interest for activation measurements vary from about 1 eV to about 3 keV. Resonance reactions can be used to measure the intermediate neutron flux while the thermal flux is measured with $1/v$ -absorbers.

The detectors can be irradiated in a cover made of e.g. cadmium which effectively absorbs all thermal neutrons. It is then possible to separate the resonance and $1/v$ -contribution to activities. The spectrum above the cadmium cut-off energy is called the epicadmium spectrum.

A serious drawback of activation detectors is their lack of sensitivity in the range 3 - 100 keV, where there are no suit-

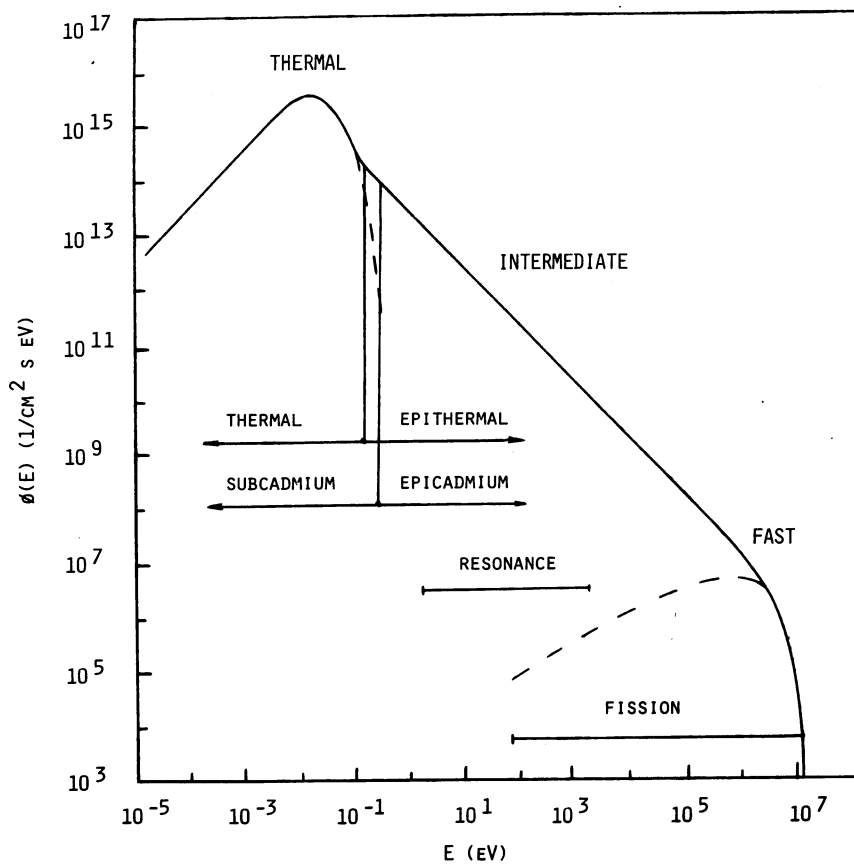


Figure 1. Neutron flux energy spectrum in a typical thermal reactor /6/.

able resonances or threshold energies. In the region 100 keV - 1 MeV the choice of detectors is very limited and the properties of possible reactions are not very well known.

Activation measurements in a reactor environment are most often carried out with thin metal activation foils or wires which are irradiated in a sandwich arrangement and then taken apart for activity measurement /7/. The use of thin foils is advantageous, because they do not significantly disturb the flux to be measured and the self-shielding effects in activation and activity measurement are small and they can be easily estimated.

2 **MULTICOMPONENT ACTIVATION DETECTORS FOR REACTOR NEUTRONS**

2 . 1 **The multicomponent principle**

High-resolution gamma spectroscopy with Ge(Li) detectors has made it possible to measure simultaneously tens of gamma-emitters. In earlier studies Routti /8-10/ has shown that it is possible to identify and measure quantitatively tens of gamma-active spallation reaction products in targets of medium heavy elements irradiated with high-energy hadrons. The feasibility of these multireaction spallation detectors has given rise to the question whether similar principles would be useful for reactor applications. In reactor neutron spectroscopy the number of suitable detector reactions per element is small, at most three. The multireaction concept must therefore be accomplished by mixing several elements into multicomponent detectors.

One of the purposes of this work is to examine possibilities to design multicomponent detectors for reactor applications and to describe test measurements and the techniques used

in their analysis. The advantages and limitations of multicomponent detectors are also discussed.

The potential advantages of multicomponent detectors as compared to separate foils are

- reduced number of activity measurement,
- small size, especially for resonance detectors,
- possibility to produce cheap, standardized detectors,
- reduced handling of radioactive materials.

The significance of these properties depends on the application. They would be desirable in a research environment where large numbers of measurements with short durations are made. On the other hand, in power reactor measurements with durations of about one year and with ample space for foil packages the advantages would be less significant.

The multicomponent principle sets the following additional requirements on the detector materials

- significant amounts of each product nuclide must be produced in one reaction only,
- the component materials should be pure from interfering impurities,
- the total size of the detectors should be small enough to avoid excessive self-shielding and attenuation of the softest gamma-rays to be measured.

For neutron spectrum unfolding it is also desirable that the resonance or threshold energies cover the energy range of interest as evenly as possible. The requirements set on the component reactions are discussed in publication I.

In the selection of detector reactions, a compromise must be made between conflicting requirements. The masses of the selected components and the irradiation, waiting and measuring

times must be chosen so that for each product nuclide at least one gamma peak can be measured with good accuracy. To facilitate the design of multicomponent detectors, Aarnio and Koskela have coded the simulation and optimization program OPTO /11/, which has been revised by the author. The design of a detector is generally not very sensitive to changes in the flux level and time parameters, and it is possible to design a detector applicable for a fairly wide range of flux levels and irradiation times. A block diagram of the phases involved in the design and use of detectors is shown in Fig. 2.

The gamma spectra of multicomponent detectors are measured with Ge(Li) detectors. Though the spectra have tens of peaks, there is generally no serious overlapping of important peaks. The gamma spectrum analysis and the calculation of saturation activities can be carried out, for example, with the program SAMPO80 /12/. At Helsinki University of Technology it has been installed on a Data General Eclipse S/140 minicomputer which is connected to a 4096-channel multichannel analyser /13/.

To demonstrate the multicomponent concept, separate cadmium covered detectors were designed for the fast and intermediate energy regions of a 250 kW Triga research reactor. For the fast region a detector with six elements and nine threshold type reactions was designed, and for the intermediate region eight resonance reactions in as many elements were used. The composition of the detectors is given in Tables 1-2 and their gamma spectra are shown in Figs. 3 and 4.

The resonance detectors were fabricated by evaporating solutions of detector materials in quartz glass ampoules. All other components except tungsten could be mixed in a single solution. In the threshold detectors a mixture of metal powders or small pieces of wire was used except for indium which was evaporated from a solution due to its small mass. Both types of detectors were enclosed in cylindrical cadmium covers about 1 mm thick. In this measurement polyethylene vials could have been used instead of quartz ampoules. However, quartz was used to gain experience with an encapsulation technique also suitable for more severe irradiation conditions.

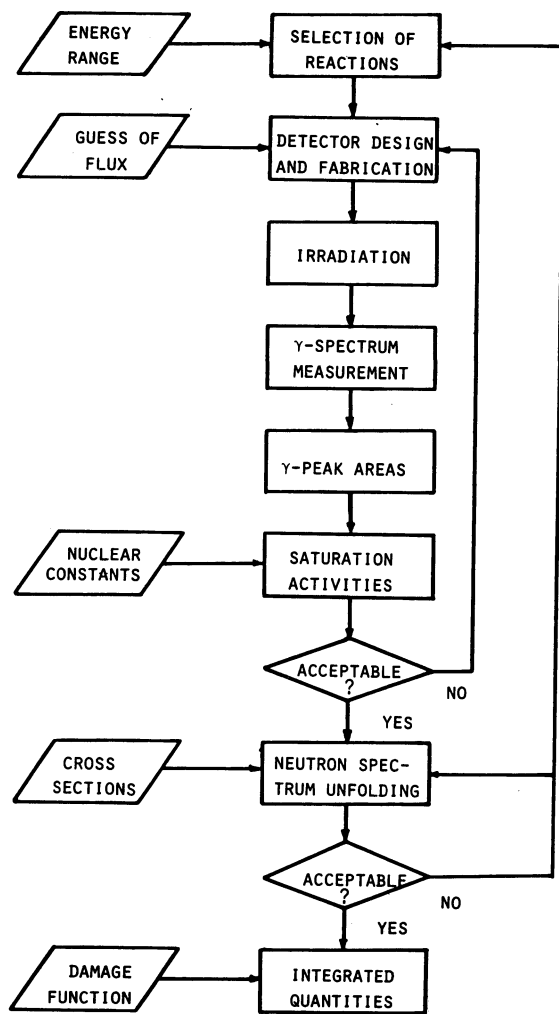


Figure 2. Block diagram of the phases of the design and use of multicomponent activation detectors.

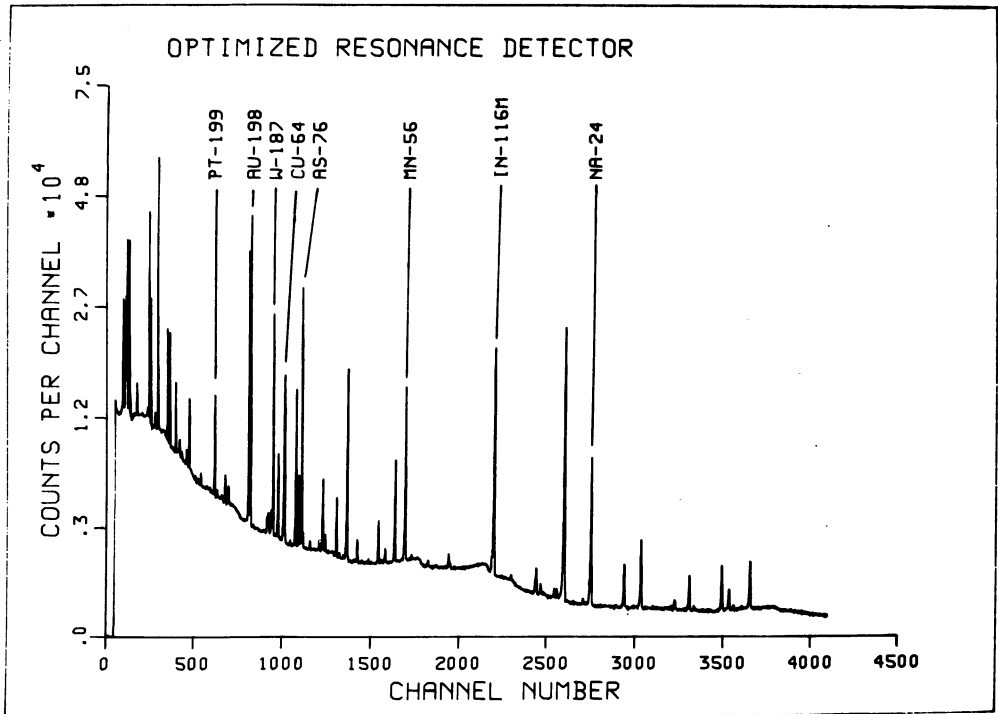


Figure 3. The gamma spectrum of the optimized multi-component resonance detector of Table 1.

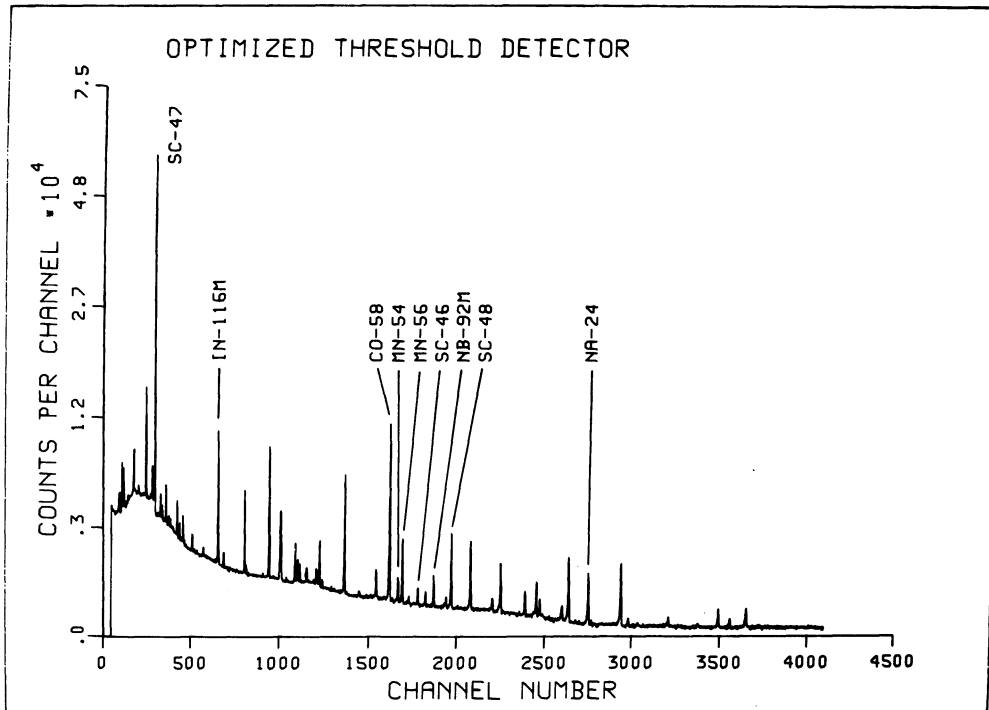


Figure 4. The gamma spectrum of the optimized multi-component threshold detector of Table 2.

Table 1 Composition and nuclear properties /6,17,52/ of a multicomponent resonance detector for a Triga reactor. The optimized irradiation time is 150 min, waiting time 35 min and measuring time 120 min.

Reaction	Resonance energy (eV)	Excess resonance integral I' (barn)	$\sigma(2200 \text{ m/s})$ (barn)	Half-life	γ -energy used in this work (keV)	Branching ratio (%)	Isotope fraction in natural element (%)	Mass of natural element (μg)
$^{115}\text{In}(n,\gamma)^{116}\text{In}^m$	1.46	2440	160	54.3 min	1097	55.7	95.72	0.4
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	4.906	1490	98.8	64.7 h	411.8	95.5	100	2
$^{186}\text{W}(n,\gamma)^{187}\text{W}$	18.8	486	34	23.9 h	479.5	26.6	28.9	27.4
$^{75}\text{As}(n,\gamma)^{76}\text{As}$	47	58	5.4	26.4 h	559.1	44.6	100	25.2
$^{198}\text{Pt}(n,\gamma)^{199}\text{Pt}$	97	48.1	3.7	31 min	317	5.6	7.21	1016
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	337	9.4	13.3	2.58 h	846.6	99	100	6
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	580	3.17	4.51	12.7 h	511	37	69.1	165
$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	2850	0.075	0.525	15 h	1368.6	100	100	352

Table 2 Composition and nuclear properties /52,53/ of a multicomponent

threshold detector for a Triga reactor. The optimized irradiation time is 180 min, waiting time 960 min and measuring time 180 min.

Reaction	Threshold energy (MeV)	σ_{eff} (mb)	Half-life	γ -energy used in this work (keV)	Branching ratio (%)	Isotope fraction in natural element (%)	Mass of natural element (mg)
$^{27}\text{Al} (n, \alpha) ^{24}\text{Na}$	7.0	49.7	15 h	1368.5	100	100	10
$^{46}\text{Ti} (n, p) ^{46}\text{Sc}$	4.0	107	84.2 d	889.3	100	7.99	145
$^{47}\text{Ti} (n, p) ^{47}\text{Sc}$	2.2	48.8	3.4 d	159.4	70	7.32	145
$^{48}\text{Ti} (n, p) ^{48}\text{Sc}$	7.6	30.1	1.84 d	983.5	100	73.99	145
$^{54}\text{Fe} (n, p) ^{54}\text{Mn}$	3.3	404	313 d	834.8	100	5.89	200
$^{56}\text{Fe} (n, p) ^{56}\text{Mn}$	6.1	42.5	2.58 h	846.7	99	91.68	200
$^{58}\text{Ni} (n, p) ^{58}\text{Co}$	2.8	470	71.3 d	810.6	99.4	67.76	50
$^{93}\text{Nb} (n, 2n) ^{92}\text{Nb}^{\text{m}}$	10.2	317	10.2 d	934.5	99	100	150
$^{115}\text{In} (n, n') ^{115}\text{In}^{\text{m}}$	1.3	294	4.48 h	336.3	44.5	95.77	1

The publications of this thesis include a more detailed description of the design and fabrication of the detectors /I/ and of test measurements and their analysis /II,IV,V/. It is shown that it is possible to design and optimize multicomponent detectors for a research reactor so that they can be fabricated with simple methods, their gamma spectra can be measured and analysed with good accuracy and the results can be used to fit or unfold physically sound neutron spectra with acceptable fitting errors.

There are a number of potential applications of multicomponent detectors, and with the design methods available it is possible to estimate their feasibility for different neutron fields. As an example, the suitability of multicomponent detectors for fusion reactor blanket neutronics measurements is studied in publication VI. When the fusion reactor material testing programs are started, great numbers of flux measurements have to be made, and standardized multicomponent detectors might decrease the amount of routine work in them.

2.2 Comparison with separate activation foils

Multicomponent detector measurements are subject to similar error sources as conventional activation measurements. Some types of uncertainties are more serious for multicomponent detectors than for separate foils. The differences in the uncertainties are usually rather small and may in some cases also be in favour of multicomponent detectors. To compare the measuring accuracy of multicomponent detectors and separate foils we give here a qualitative or a rough quantitative discussion of various error sources.

The following sources contribute to uncertainties in multicomponent detector measurements

- impurities and errors in component masses,
- self-shielding effects in the detectors,

- errors in gamma spectrum measurement and analysis,
- errors in cross section data,
- errors in neutron spectrum fitting or unfolding.

Partly the error sources are the same as in foil activation measurements. The impurity effects are more severe in MADs than in conventional foils, since for each activity to be measured the possible interference from the impurities of all components must be considered. To find out the interfering impurities, activation analysis was carried out for each component material. When readily available high-purity, materials are used, the effects of impurities are small but not negligible. When quartz glass ampoules were used, sodium and manganese present in the ampoule material were the most significant, and corrections of 2 to 11 % were required. The correction factors can be found in publication II. Since the impurity corrections are small and they can be estimated with activation analysis, impurities do not constitute a severe problem in the multicomponent concept, the uncertainties from this source are typically less than 1 %.

Weighing of detector materials can be carried out with errors less than 1 %. If a mixed solution is used for fabricating resonance detectors, it should be made sure that no precipitation takes place if the solution is stored for longer times. In our experiments no precipitate could be seen, nor were there any differences between detectors made after different storing times. With the mixed solution method good weighing accuracy can be reached, especially the ratios of component masses can be kept constant. This is an advantage in accurate monitoring of the spectrum shape. On the other hand, a weighing error may lead to a systematic error in all masses, which is propagated to the normalization of the spectrum.

Self-shielding errors can be further divided into flux depression effects due to the detector, especially with cadmium covers, absorption of neutrons in the detector materials and attenuation of the emitted gamma-rays in the detector. The

first of these effects is common to both multicomponent detectors and separate foils. The neutron self-shielding problem is slightly different for foils and multicomponent detectors. In resonance MADs the masses can be made very small without difficulties in handling, and the self-shielding problems can be reduced. On the other hand, the geometry of separate foils is well-defined and the self-shielding corrections can be calculated with good accuracy.

For threshold detectors neutron self-shielding effects are very small, if the masses are well below one gramme. The rough estimates described in publication II give corrections of less than one per cent.

The attenuation of gamma-rays in multicomponent resonance detector materials is negligible, but a quartz ampoule may cause up to 2 % attenuation. For a polyethylene vial the attenuation is smaller. In multicomponent threshold detectors the attenuation of gamma-rays is significant. It is less than 10 % for most product nuclides, but for the 159 keV line of ^{47}Sc the calculated value is 27 %. For so large values possible errors in the attenuation correction become important, and the use of the $^{47}\text{Ti}(n,p)^{47}\text{Sc}$ reaction becomes questionable.

For separate foils the gamma-ray attenuation does not constitute a problem. The foils are usually so thin that attenuation is insignificant. In addition, the geometry is better defined for foils than for multicomponent detectors and the attenuation can be estimated with greater accuracy. For separate foils the attenuation is a problem only with X-ray counting, which is practically impossible with the multicomponent concept.

If we assume a ± 50 % error in the correction factors presented in Tables 3 and 4 of publication II, the uncertainty introduced in the saturation activities is typically about 5 % and for ^{47}Sc about 15 %. It should be noted, however, that if smaller masses are used in longer irradiations or higher fluxes, these uncertainties are reduced.

The errors in the gamma spectrum counting and analysis are due to statistical variations in the pulse counts, errors in

efficiency calibration, errors in peak area calculation, and errors in branching ratios and half-lives. These errors are mainly the same for multicomponent detectors and separate foils. In our measurements the net peak areas were typically more than 10 000 which means statistical errors of less than 1 %. The smallest peak areas were about 2 000 leading to about 2 % statistical error. The peak area fitting error given by SAMPO80 is typically less than 2 %. The efficiency calibration involves typically 3-5 % errors. If the multicomponent threshold detector is not quite homogeneous there may be some uncertainty of the position of the mass centroid of each component. An error of 2 mm with a 30 cm source to detector distance, which we have used, leads to an additional 1.3 % error in efficiency calibration, but with a 10 cm distance the error would be 4 %.

The uncertainties of branching ratios and half-lives quoted in references are usually quite small, but the differences of values in different references may be 1-2 %.

The total uncertainty in the activity determination can be obtained as the root-mean-square value of the different contributions and it is typically about 6 % or less. Typically the largest error source seems to be efficiency calibration. Together with the uncertainties in shielding and impurity correction, the uncertainties in multicomponent detector saturation activities are, according to these rough estimates, 6-8 %, with the smaller values typically applicable for resonance detectors. These values are about 1-2 percentage points higher than for thin separate foils.

The errors in cross sections and spectrum fitting or unfolding are the same for multicomponent detectors and separate foils. Typical uncertainty estimates of cross sections are 5-20 % /14/. The cross section errors and the errors due to the spectrum fitting or unfolding procedure should be treated together. If spectrum fitting with a simple theoretical form, e.g. fission spectrum or $1/E$ spectrum, leads to activity fitting errors which are about the same as or smaller than the

cross section uncertainties, the fine structure of the spectrum unfolded with a more sophisticated method should be analysed critically.

The **a priori** information used in fitting or unfolding affects the spectrum, and its effect is the most significant in the energy regions where the resolution of the detectors is poor. Qualitative information on these effects can be obtained by using different **a priori** conditions and by carrying out simulation unfoldings using activities generated with a given test spectrum. Examples can be found in publications V and VI. As a rule of thumb it can be said that the uncertainties of integrated quantities are about the same as the average fitting errors of activities. Locally the errors of the flux values may be much larger.

Recently efforts have been made for more rigorous treatment of uncertainties. Such methods are used by Perey in the program STAY'SL /15/ briefly described in section 4.3. Such methods require covariance matrices for measured activities, input spectrum and cross sections. As the importance of reactor dosimetry is increasing, covariance data files for dosimetry reactions are being compiled /16/. In this work, however, such methods have not been used.

2.3 Limitations of multicomponent detectors

Activation detectors in general have some serious shortcomings especially as flux and fluence monitors in radiation damage studies. These are due to the physical properties of available detector reactions. Naturally these problems are not alleviated by the multicomponent principle, on the contrary, the interferences may exclude some potentially useful reactions.

Of the commonly used resonance reactions $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$ has the highest resonance energy, 2850 eV, /17/ and the lowest threshold energies are about 1 MeV. Activation detectors do not yield information on the flux in this energy interval. But if

the neutrons in this interval do not induce reactions, accurate knowledge of the flux is not very important in such applications as activity calculations. With radiation damage the situation is quite different. Damage is usually measured with the number of displacements per atom (DPA) in the lattice, and the displacement cross section is significant from about 10 keV upwards.

The contribution to the total DPA rate by neutrons with energies in the range 0.01-1 MeV may be more than 20 %. Some values for the Triga reactor are given in papers IV and V. At this interval the neutron energy spectrum in a thermal reactor changes from the fission spectrum to the 1/E-type moderation spectrum, and its behaviour cannot be interpolated very accurately. Consequently it would be most desirable to have an activation detector with a high cross section in this interval. Since the interest in radiation damage problems has lately increased, detector reactions with low threshold energies have been sought after and their cross sections have been measured.

The reaction $^{93}\text{Nb}(n,n')^{93}\text{Nb}^m$ has a cross section resembling the DPA cross section and there is considerable interest in using it for long-term fluence monitoring /18,19/. Unfortunately the product nuclide emits only low-energy X-rays and special processing is required for a niobium detector. It cannot be used in a multicomponent detector due to the high absorption of low-energy radiation. The same is true also for the reactions $^{103}\text{Rh}(n,n')^{103}\text{Rh}^m$ /6/ and $^{107}\text{Ag}(n,n')^{107}\text{Ag}^m$ /20/. Fission detectors are not convenient for use in multicomponent detectors due to the multitude of radioactive fission products which would interfere with other reaction products.

The use of the reaction $^{199}\text{Hg}(n,n')^{199}\text{Hg}^m$ for flux measurements has been studied recently /19,21/. The threshold energy is about 0.53 MeV and the product gamma energies are 158 and 374 keV. The use of this reaction in multicomponent detectors would be worth further studies, though its usefulness in long-term measurements is reduced by the relatively short product half-life 42.6 min.

In spite of the limitations mentioned above, multicomponent detectors can be used, for example, to monitor neutron flux in radiation damage measurements with durations up to about one day. Such short measurements are used in the study of microscopic phenomena in materials due to radiation exposure. The methods used include electron microscopy, measurements of internal friction or residual resistivity and positron annihilation techniques, which are also being developed at Helsinki University of Technology /22/. The slight uncertainty of the flux below 1 MeV is not too serious, if the same estimates are consistently used in all measurements.

In power reactors the situation is different. The irradiation time would be one loading period, i.e. about one year. One of the current problems of importance is fluence monitoring at the pressure vessel wall for radiation damage studies. Here the contribution by the flux below 1 MeV is significant and the main problem is to find suitable detectors with low threshold energies. In this respect the multicomponent concept would not offer significant advantages. It is, however, possible that applications are found for multicomponent detectors also in power reactor studies, for example in short-term measurements outside the pressure vessel. It might also be possible to develop a multicomponent detector with a long-lived fluence monitor and several shorter-lived detectors for measuring the shape of the flux spectrum, which can be assumed to remain reasonably constant despite variations in the flux level.

An application of the multicomponent principle has been reported by Mehner et al. /23/. In their method a multicomponent metal wire is inserted in the core of a 440 MW pressurized water reactor (VVER) in which a dry channel is available. Four or five reactions are sufficient to give the axial distribution and rough spectral information on the thermal, intermediate and fast flux.

3 MULTIREACTION SPALLATION DETECTORS FOR HIGH-ENERGY HADRONS

Activation detectors can also be used for measuring stray radiation fields around particle accelerators. In the stray field of a high-energy proton synchrotron also protons and pions are important in addition to neutrons. The energy range of interest goes up to the GeV region. At these energies spallation reactions constitute one possible method for particle flux measurements.

If a high energy hadron impinges on a nucleus, a number of nucleons may be knocked out or evaporated from the nucleus. Depending on the number of emitted nucleons, a variety of product nuclides can be formed. The spallation yield cross section for each product has a threshold-type energy dependence. The threshold energies are typically from about ten to a few hundred MeV.

It is possible to measure the activity of just one spallation product or use several products formed in one detector material. For example, the production of ^{149}Tb in gold or mercury has been used to measure hadron fluxes above 600 MeV /24/.

With high-resolution gamma spectroscopy it is possible to identify tens of radioactive spallation products from a medium heavy target. Routti /8-10/ has shown that copper is a suitable spallation detector material, since the number of reaction products is large, but still the gamma spectrum is not too complicated. It is worth noting that the measurement of a large number of activities is feasible even though the optimization possibilities available with multicomponent detectors do not exist.

Particle flux measurements at accelerators are required for radiation protection purposes. The stray fields cause doses to personnel, radiation damage and activation in instruments and structures, and background pulses in physical experiments. In addition, measurements of secondary fluxes from targets constitute good integral tests of high-energy hadron production calculations and of the underlying particle production models.

An experiment where spallation detectors were used to measure the angular distribution of hadrons above different threshold energies is described in paper VII. The experiment was carried out at the CERN Super Proton Synchrotron (SPS) with 225 GeV/c primary protons and an extended copper target. The measurement was part of a project on experimental and computational determination of hadron fluxes.

The results were compared with earlier measurements using 22 GeV/c primary protons, carried out by Routti /10/, and also with the results of hadron cascade simulations with the program MAGKO /25,26/. It was found out that the angular distributions do not significantly differ from those measured at lower primary energies. Some results are shown in Fig. 5. The agreement with theoretical computations was also found satisfactory. The methods used in the radiation protection design of the SPS and its experimental facilities are based on the experiences with lower-energy proton synchrotrons. It is therefore of great importance that no significant deviations from the expected behaviour of the secondary particle fields can be found.

The unfolding of the secondary hadron energy spectrum at different angles was carried out with LOUHI78. The results, however, are not very accurate, since the uncertainties of the spallation yield cross sections are rather large and the spectrum extends well beyond the highest threshold energies of a few hundred MeV.

The energy dependent spallation yield cross sections were calculated with semiempirical Rudstam /27/ and Silberberg-Tsao formulas /28/. These have been implemented in the FORTRAN program SPALL which is described in publication VIII of this thesis. The program was developed by adding the more complicated Silberberg-Tsao formulas to a program based on the straightforward Rudstam formula used by Routti /8-10/.

The analysis of spallation detector measurements involves large uncertainties from several sources. The accuracy of the semiempirical cross section formulas is about $\pm 30\%$ at its best, but the lower energy limit of their region of applicability is about 50-100 MeV. Below that the errors may be much larger. In addition, the formulas do not make any difference between

different types of hadrons. Since experimental data exists mainly for proton irradiations, the formulas are most reliable for protons.

Detailed experimental or calculated data for the dependence of spallation yield cross sections on the type of incident hadron are not available. It is however, possible to give some rough characteristics of differences in cross sections and of the relative abundances of different hadrons in the flux. The low energy particles, up to a few hundred MeV, are protons and neutrons generated in the intranuclear cascade and evaporation, with slightly more neutrons than protons /54/. At higher energies charged pions also become important. If the target is large, the fractions of different hadrons are distorted by slowing down of charged particles. In our measurement the target was relatively small and the contribution of nucleons was by far more important than that of pions.

According to the Rudstam model /27/ the differences of spallation yields for different incident hadrons, would be caused by differences in the total inelastic cross sections. According to Ranft /55/, at energies above 50 MeV the neutron-nucleus (σ_{nN}) and pion-nucleus ($\sigma_{\pi N}$) cross sections are slightly smaller than the proton nucleus cross section (σ_{pN}) with values $\sigma_{nN} = \sigma_{pN}/1.07$ and $\sigma_{\pi N} = \sigma_{pN}/1.20$. These differences are not important as compared with other uncertainties.

At low energies, 10-50 MeV, the Coulomb barrier, about 3 MeV for copper, is significant and the lowest threshold energies may be somewhat different for protons and neutrons. At these energies pions are not significant. The semiempirical formulas are not reliable at 10-50 MeV, but better cross section values are not readily available. In principle, they could be obtained with Monte Carlo simulations using intranuclear cascade and evaporation models. Fortunately at the lowest energies, where the relative uncertainties are large, the yield cross sections are small and the effect of cross section errors to the total yield is not excessive.

The slowing down of charged particles in the spallation detectors and attenuation of the gamma rays of the product

nuclides also increase uncertainties if the detectors are thicker than a few millimeters.

Due to the difficulties described above the results of multireaction spallation detectors must be interpreted carefully. It is, however, worth noting that the variations of secondary radiation fields at high-energy particle accelerators may vary several decades and results with accuracy within a factor of two or three must often be considered satisfactory. The accuracy requirements are generally much less stringent than, for example, in reactor flux measurements. Spallation detectors can also be used conveniently in cases where measurements carried out under roughly similar conditions have to be compared with each other.

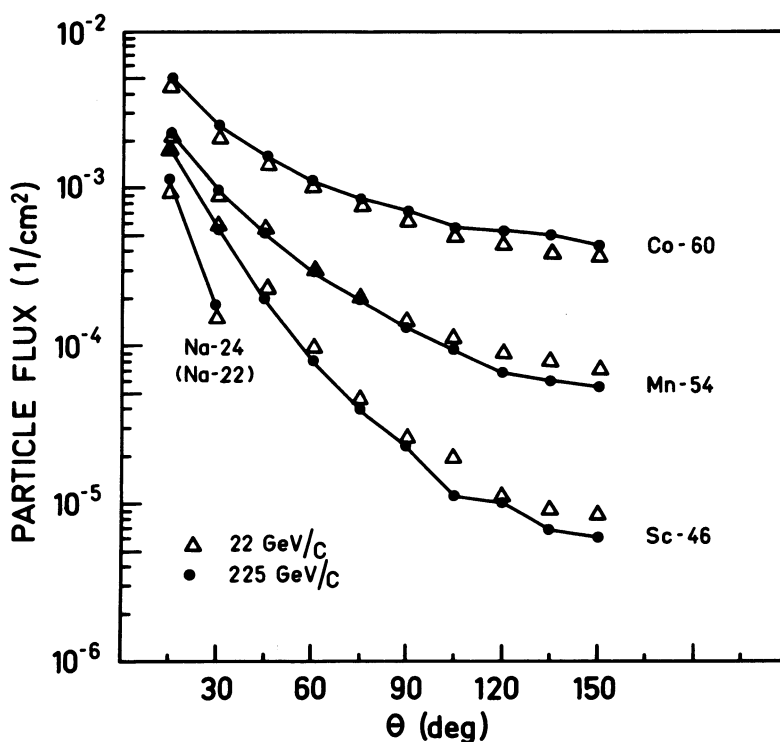


Figure 5. Comparison of the angular distribution of the secondary hadron flux measured for 22 and 225 GeV/c primary protons. The 22 GeV/c results have been multiplied by 8.8 to match the curves of ^{46}Sc at 150° angle. The particle flux is given above the spallation yield threshold energies which are 12.5 MeV for ^{60}Co , 50 MeV for ^{54}Mn , 126 MeV for ^{46}Sc and 400 MeV for ^{22}Na and ^{24}Na .

4 NEUTRON SPECTRUM UNFOLDING

4.1 The few-channel unfolding problem

The unknown neutron spectrum and the saturation activities are related through the activation equations

$$A_i = \int_{E_{\min}}^{E_{\max}} \sigma_i(E) \phi(E) dE + \epsilon_i, \quad i = 1, \dots, m, \quad (3)$$

where

A_i is the saturation activity of detector i normalized per detector nucleus,

$\sigma_i(E)$ is the cross section of reaction i at energy E ,

$\phi(E)$ is the spectrum to be solved,

ϵ_i is a random error reflecting experimental uncertainty in reaction i ,

E_{\min} is the lower limit of the energy region of interest,

E_{\max} is the upper limit of the energy region of interest,

m is the number of detector reactions.

This group of equations is a degenerate case of a linear Fredholm integral equation of the first kind.

The solution of eq. (3) is called spectrum unfolding or deconvolution. Even if the experimental errors ϵ_i were neglected, the problem would not have a unique solution. This is

physically evident, since a finite number of measurements cannot define uniquely a continuous distribution. To obtain physically relevant solutions some kind of **a priori** information must be introduced in the unfolding procedure. Conditions can be set on the nonnegativity, smoothness and shape of the spectrum.

The older unfolding methods include series expansions with orthogonal functions /29-31/, but these have given way to iterative and least-squares methods. Of the iterative methods SAND-II /32/ is the best known. The least-squares programs include among others SPECTRA /33/, CRYSTALL BALL /34/, STAY'SL /15/ and the LOUHI-type codes described in this report. Most unfolding programs have been developed for reactor neutron spectroscopy, whereas the LOUHI-type programs were originally coded as flexible general purpose unfolding programs applicable to many types of unfolding problems.

The numerical solution of eq. (3) is calculated at a finite number of discrete energy points or groups. If the number of energy points is larger than the number of measured activities or other responses, it is customary to speak of a few-channel unfolding problem. This is the normal situation with activation detectors.

With proton recoil measurements the number of measured responses may be larger than the number of energy points, which leads to a many-channel unfolding problem. It can be solved, for example, with the programs FERDOR or COOLC /35,36/. The many-channel unfolding problem is not considered in this report.

4.2 LOUHI-type unfolding programs

Many unfolding programs include prior constraints imbedded in the solution methods so that they cannot be controlled by the user. One of the objectives of designing the LOUHI-type programs has been to make all such conditions controllable by the user. Two different solution algorithms were originally used in two distinct programs KIEPPI and LOUHI coded by Routti /8,37/ for the CDC 6600 computers of Lawrence Berkeley Laboratory of

the University of California. The methods were combined and extended in LOUHI78, as explained in publication III, and further developed in LOUHI82 described in publication IV.

The LOUHI-type programs are general purpose unfolding programs and they are not inherently restricted to any specific problems. They have been used for unfolding spectra measured with activation detectors, spallation detectors, Bonner spheres and proton recoil emulsions /8,9,37,38,39/. They can also be used for other types of problems involving the solution of linear Fredholm integral equations, such as the determination of energy dependent photofission cross sections from the measured fission rates induced by electrons or bremsstrahlung /40/.

For numerical solution eq. (3) is put in the discrete form

$$A_i = \sum_{j=1}^n K_{ij} \phi_j + \epsilon_i, \quad i = 1, \dots, m, \quad (4.a)$$

or in matrix form

$$\underline{A} = \underline{K} \underline{\phi} + \underline{\epsilon}, \quad (4.b)$$

where

ϕ_j = $\phi(E_j)$ is the spectrum value at the energy point E_j ,

m is the number of activities,

n is the number of energy points,

\underline{K} is the discretized kernel matrix with elements K_{ij}
 $= w_j \sigma_i(E_j)$,

w_j is a quadrature weight for the numerical integration of eq. (3).

In LOUHI78 and LOUHI82 prior information on the solution is introduced by transforming the group of linear equations into the minimization problem

$$\min Q = Q_0 + \nu \sum_{k=1}^5 W_k Q_k, \quad (5)$$

where

$$Q_0 = \sum_{i=1}^m r_i^{\epsilon} \epsilon_i^2 = \sum_{i=1}^m r_i^{\epsilon} \left[A_i - \sum_{j=1}^n K_{ij} \phi_j \right]^2, \quad (6)$$

$$Q_1 = \sum_{j=1}^n r_j^p (\phi_j - p_j)^2, \quad (7)$$

$$Q_2 = \sum_{j=1}^n r_j^f \phi_j^2, \quad (8)$$

$$Q_3 = \sum_{j=2}^n r_j^t (\phi_j - \phi_{j-1})^2, \quad (9)$$

$$Q_4 = \sum_{j=2}^{n-1} r_j^d (\phi_{j-1} - 2\phi_j + \phi_{j+1})^2, \quad (10)$$

$$Q_5 = \sum_{j=n_0}^{n_u} r_j^s (\phi_j - z_j \phi_{j+1})^2, \quad (11)$$

Here r_i^{ϵ} are weights defined for each measured response r_j^p , r_j^f, r_j^t, r_j^d and r_j^s are weights defined for each energy point E_j . $\underline{p} = (p_1, \dots, p_n)^T$ is a trial solution and z_j are coefficients defining the assumed shape of the solution. ν is an overall weight for the **a priori** criteria and W_k are their individual weights which are selected by the user.

The terms Q_k have the following physical meaning

Q_0 is the weighted square sum of the differences of measured and calculated responses,

- Q_1 is the weighted square sum of deviations of the solution from a trial solution P at the energy points E_j ,
- Q_2 is the weighted square sum of the solution values at the energy points,
- Q_3 is the weighted square sum of first differences of the solution,
- Q_4 is the weighted square sum of second differences of the solution,
- Q_5 describes the deviation on the solution from a shape defined by the relation

$$\phi_j / \phi_{j+1} = z_j \quad (12)$$

in the energy interval $n_l \leq j \leq n_u$.

For evenly spaced energy points, the square sums are approximately proportional to corresponding square integrals. Q is a positive definite function and reaches its minimum at the zero of its gradient.

In some publications the notation $Q_1 = Q_p$, $Q_4 = Q_d$ and $Q_5 = Q_s$ is also used.

In the program KIEPPI the zero point of the gradient was found by solving a group of n linear equations with n unknowns. In the program LOUHI nonnegativity condition was implemented by setting $Q_j = X_j^2$, $j = 1, \dots, n$. This leads to a nonlinear minimization problem which is solved by the iterative Davidon algorithm with a variable metric /41/. In the original KIEPPI and LOUHI programs only the Q_1 and Q_4 conditions were used.

In LOUHI78 both the linear and nonlinear methods were implemented and all the prior conditions can be used. In LOUHI82 the number of energy points was increased from the earlier 40

to 100. The linear problem is also solved with the iterative minimization algorithm, which leads to savings in memory requirements and improves numerical stability.

The LOUHI-type programs include also options for internal testing and error simulation with generated activities.

As an example, the results of unfolding the epithermal spectrum of a Triga reactor from measurements with the multi-component detectors of Tables 1 and 2 are shown in Fig. 6.

LOUHI78 has been used in the international REAL-80 inter-comparison of evaluation methods for activation measurements /42/ and it has done reasonably well though it suffers from the small number of energy points. The REAL-80 data was also used to test LOUHI82 and the results can be found in publication V.

The LOUHI-type programs are unnecessarily sophisticated for typical thermal research reactors where the fast flux has approximately the fission neutron spectrum and the intermediate spectrum a $1/E$ -behaviour. Therefore we have also coded the program MADAP described in publication II. It can be used to fit the epithermal spectrum with the said functions. Fitting programs of this type are easy to use and the interpretation of results is straightforward, and they are quite adequate for many purposes in which the higher energy resolution of the more sophisticated methods is not necessary.

4.3 Other unfolding programs

A large number of unfolding programs have been published during the past 15 years. We describe below some of the best-known programs and compare their methods with LOUHI. SAND II serves as an example of iterative programs, SPECTRA, CRYSTALL BALL and STAY'SL are different types of least-squares programs, and WINDOWS can be used to compute integral quantities directly from the measured activities. The code SWIFT serves as an example of an unfolding code with Monte Carlo minimization.

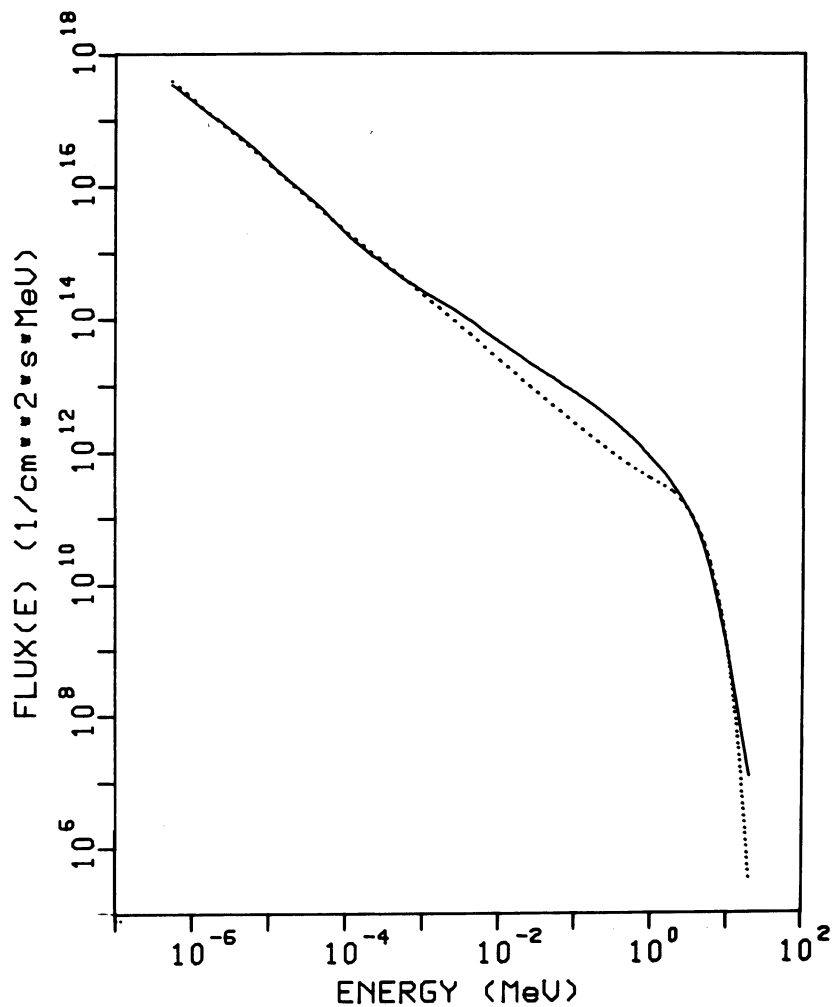


Figure 6. The epithermal spectrum in a Triga reactor, unfolded from multicomponent activation detector measurements with LOUHI82.

Solid line: results with the smoothness condition, average fitting error 10.2 %.

Dotted line: results with the shape condition, average fitting error 10.7 %.

SAND II

One of the most often referred unfolding programs is SAND II developed by McElroy et al. /32/. In SAND II a trial spectrum is modified iteratively until acceptable fitting errors of activities are reached.

The solution spectrum $\phi_j^{(k)}$, $j = 1, \dots, n$, of iteration step k is obtained from the trial spectrum or the solution of the previous step according to the following procedure.

The ratio $R_i^{(k)}$ of measured and calculated activity at iteration step k is determined for each reaction,

$$R_i = A_i/A_i^C = A_i / \sum_{j=1}^n \sigma_{ij} \phi_j^{(k-1)}, \quad i = 1, \dots, m, \quad (13)$$

where

σ_{ij} is the cross section of reaction i in group j ,

$\phi_j^{(k-1)}$ is the group flux in energy group j from the previous step, $\phi_j^{(0)}$ is given as input.

The spectrum is then modified by setting

$$\phi_j^{(k)} = M_j^{(k)} \phi_j^{(k-1)}, \quad j = 1, \dots, n, \quad (14)$$

where the modifying factor is defined by the equation

$$\ln(M_j^{(k)}) = \frac{\sum_{i=1}^m w_{ij}^{(k)} \ln R_i^{(k)}}{\sum_{i=1}^m w_{ij}^{(k)}}, \quad j = 1, \dots, n. \quad (15)$$

The energy dependent weights W_{ij} are determined by the relative contribution of energy group j in the i^{th} activity

$$W_{ij}^{(k)} = \sigma_{ij} \phi_j^{(k-1)} / \sum_{j=1}^n \sigma_{ij} \phi_j^{(k-1)}. \quad (16)$$

The relationship of the SAND II algorithm and least-squares methods used e.g. in the program STAY'SL has been discussed by Guthrie and Simons /43/ and Matzke /44/. They have pointed out that the iteration algorithm is equivalent to solving the minimization problem

$$\min \sum_{i=1}^m (\log A_i - \log A_i^c)^2 \quad (17)$$

where the logarithms of the flux values, $\log \phi_j$, are used as independent variables. The prior information introduced by the trial spectrum can be treated formally by considering each given group flux a measured response with a nonzero cross section in one group only. The SAND II algorithm can be interpreted so that the flux is a random variable with a lognormal distribution, which has the advantage that negative values are not encountered.

STAY'SL

One of the most recent and best-known unfolding programs is STAY'SL by Perey /15,45/, in which the uncertainties of input data are treated with statistical rigour. This program uses least-squares methods and covariance matrix representation of uncertainties of measured activities, trial spectrum and cross sections.

The problem solved by STAY'SL can be formulated as finding the most likely value of the neutron spectrum and its uncer-

tainty when the following input data are given

- detector reaction cross sections with their uncertainties,
- a trial spectrum with its estimated uncertainties,
- results of activation measurements in the actual neutron spectrum with their uncertainties.

We use the following notation, which is different from that used by Perey

$$\underline{A} = \begin{bmatrix} A_1 \\ \vdots \\ A_m \end{bmatrix} \quad \text{is the vector of measured saturation activities,}$$

\underline{V}_A is the $m \times m$ covariance matrix of \underline{A} ,

\underline{P} is the vector of trial spectrum group flux values,

\underline{V}_P is the $n \times n$ covariance matrix of \underline{P} ,

$$\underline{\Sigma}^i = \begin{bmatrix} \Sigma_1^i \\ \vdots \\ \Sigma_n^i \end{bmatrix} \quad \text{is the group cross section vector of reaction } i,$$

$$\underline{\Sigma} = \begin{bmatrix} \Sigma_1^1 \\ \vdots \\ \Sigma_n^1 \\ \Sigma_1^2 \\ \vdots \\ \Sigma_n^2 \\ \vdots \\ \Sigma_n^m \end{bmatrix} \quad \text{is a vector with the group cross section of all the } m \text{ reactions consecutively, this form is used to make its covariance matrix two-dimensional.}$$

\underline{V}_Σ is the $nm \times nm$ covariance matrix of $\underline{\Sigma}$. Transpose of a matrix or vector is denoted with the superscript T.

In the solution method both flux and cross section values are formally considered free parameters. They are combined into the parameter vector

$$\underline{X} = \begin{bmatrix} \underline{\phi} \\ \underline{S} \end{bmatrix}, \quad (18)$$

where $\underline{\phi}$ is the group flux vector and \underline{S} is the adjusted cross section vector. With \underline{A}^c we denote the vector of activities calculated with parameter vector \underline{X} . The input value of \underline{X} is denoted \underline{X}^0 ,

$$\underline{X}^0 = \begin{bmatrix} \underline{P} \\ \underline{\Sigma} \end{bmatrix}, \quad (19)$$

and its covariance matrix is

$$\underline{V}_{X^0} = \begin{bmatrix} \underline{V}_P & 0 \\ 0 & \underline{V}_\Sigma \end{bmatrix}. \quad (20)$$

Mathematically the problem can be formulated as minimizing the function

$$\chi^2 = \begin{bmatrix} \underline{X}^0 - \underline{X} \\ \underline{A} - \underline{A}^c \end{bmatrix}^T \begin{bmatrix} \underline{V}_{X^0} & 0 \\ 0 & \underline{V}_A \end{bmatrix}^{-1} \begin{bmatrix} \underline{X}^0 - \underline{X} \\ \underline{A} - \underline{A}^c \end{bmatrix} \quad (21)$$

with respect to \underline{X} . Perey /15,45/ has derived the solution

$$\underline{X} = \underline{X}^0 + \underline{V}_X^0 \underline{G}^T (\underline{V}_A^c + \underline{V}_A)^{-1} (\underline{A} - \underline{A}^c), \quad (22)$$

with the covariance matrix

$$\underline{V}_X = \underline{V}_X^0 - \underline{V}_X^0 \underline{G}^T (\underline{V}_A^c + \underline{V}_A)^{-1} \underline{G} \underline{V}_X^0. \quad (23)$$

Here \underline{G} is a sensitivity matrix defined through the equation

$$\Delta \underline{A} = \underline{G} \Delta \underline{X}, \quad (24)$$

\underline{G} transforms changes in the parameter vector into changes of the activities. \underline{G} can be expressed explicitly as

$$\underline{G} = \begin{bmatrix} \underline{\Sigma}^1 T & \underline{P}^T & 0 & 0 \dots \\ \underline{\Sigma}^2 T & 0 & \underline{P}^T & 0 \dots \\ \vdots & & & \\ \vdots & & & \\ \underline{\Sigma}^m T & \dots & & \underline{P}^T \end{bmatrix}. \quad (25)$$

\underline{V}_A^c is the covariance matrix of \underline{A}^c and it can be calculated with \underline{G} as

$$\underline{V}_A^c = \underline{G} \underline{V}_X \underline{G}^T. \quad (26)$$

It should be noted that the expression (25) for \underline{G} linearizes the otherwise nonlinear problem, since the trial spectrum \underline{P} is used rather than $\underline{\phi}$. If $\underline{\phi}$ were used, it would be possible to calculate \underline{G} with iterations. Perey has, however, pointed out that if \underline{P} can be used as a trial spectrum, eq. (25) can also be used for \underline{G} .

In the program STAY'SL the parameter vector \underline{X} and its covariance matrix are solved explicitly only for the $\underline{\phi}$ -component. This can be interpreted so that cross section values are treated as free parameters only formally in order to introduce in the solution their uncertainties.

In order to compare STAY'SL and LOUHI82 the χ^2 -function in eq. (21) can be put in the form

$$\chi^2 = (\underline{P}-\underline{\phi})^T \underline{V}_P^{-1} (\underline{P}-\underline{\phi}) + (\underline{\Sigma}-\underline{S})^T \underline{V}_\Sigma^{-1} (\underline{\Sigma}-\underline{S}) + (\underline{A}-\underline{A}^C)^T \underline{V}_A^{-1} (\underline{A}-\underline{A}^C), \quad (27)$$

The first of these terms describes the deviation of the solution from a given trial spectrum \underline{P} as does the term Q_1 of eq. (7) used in LOUHI-type programs. If the matrix \underline{V}_P^{-1} is diagonal and the weights r_j^P in LOUHI82 are set equal to $(\underline{V}_P^{-1})_{jj}$, then the first term of eq. (27) and Q_1 of LOUHI are identical. The second term of eq. (27) has no counterpart in LOUHI. If \underline{V}_A^{-1} is diagonal and the weights r_i^ϵ of LOUHI are set equal to these diagonal elements, then the last term of eq. (27) is identical to the term Q_0 of LOUHI.

The LOUHI-type programs and STAY'SL have been designed for use in different circumstances. STAY'SL is at its best in cases where good trial spectra and reliable cross sections with uncertainty estimates are available, and activation measurements are used to adjust the trial spectrum. According to Perey, however, STAY'SL gives good results even with crude estimates of covariance data /45/. On the other hand, LOUHI-type programs can be used in cases where little prior information is available and large uncertainties are involved. At least rough solutions can be obtained with very general prior conditions, such as nonnegativity and smoothness.

In principle, STAY'SL may give negative spectrum values, since the spectrum is considered a random variable with a multivariate normal distribution. This could be avoided by using lognormal distributions as suggested by Matzke /43/ and Schmittroth /46/.

STAY'SL has been designed for reactor neutron spectroscopy. For the most generally used dosimetry reactions cross section covariance data has been compiled in the ENDF/B-V dosimetry files /16/ and new information is being gathered.

SPECTRA

One of the older least-squares unfolding programs is SPECTRA by Greer, Halbleib and Walker /33/, where the solution is tied to a trial spectrum. This can also be done iteratively so that the solution of one step is used as the trial spectrum for the next one. The problem is formulated as

$$\begin{aligned} \min Q = & \sum_{i=1}^m \left[\frac{A_i - A_i^c}{A_i} \right]^2 \\ & + \sum_{j=1}^n \left[\frac{\phi_j^{(k)} - \phi_j^{(k-1)}}{\phi_j^{(k-1)}} \right]^2, \end{aligned} \quad (28)$$

where

$$A_i^c = \sum_{j=1}^n \sigma_{ij} \phi_j^{(k)},$$

$\phi_j^{(k)}$ is the solution flux in group j at iteration step k ,

$\phi^{(k-1)}$ is the solution at step $k-1$ and it is used as the trial spectrum at step k ,

$\phi_j^{(0)}$ is the initial guess of the spectrum.

The function Q is minimized by setting

$$\frac{\partial Q}{\partial \phi_j} = 0, j = 1, \dots, n, \quad (29)$$

and solving the resulting linear group of equations. In principle the solution of the iteration procedure could be obtained in closed form, but in practice numerical difficulties are encountered in solving the resulting equations.

SPECTRA has been designed for use with up to 100 energy points. It has been used mainly in the keV and MeV regions, though it has no inherent limits of the energy scale.

The program RFSP by Fisher and Turi /47/ resembles SPECTRA, but in it $E\phi(E)$ is used as the unknown instead of ϕ . In this way numerical difficulties are reduced, since $E\phi(E)$ generally varies much less than $\phi(E)$.

In comparison with the LOUHI-type programs it can be seen that SPECTRA is a special case of the linear method of LOUHI78. The first term of eq. (27) is clearly equivalent to the term Q_0 of LOUHI in eq. (5) if $r_i^\epsilon = 1/A_i^2$. The second term is equivalent to the term Q_1 of eq. (5), if $p_j = \phi_j^{(k-1)}$, $r_j^p = 1/p_j^2$ and $v = W_1 = 1$. In LOUHI78 it is also possible to use the solution of one step as the trial spectrum $P(E)$ for the next step.

CRYSTALL BALL and WINDOWS

CRYSTALL BALL is a more recent unfolding code developed by Kam and Stallmann /34/. As in SPECTRA, the prior condition is expressed by tying the solution to a trial spectrum. The problem is formulated as minimizing the functional

$$S^2 = \int_0^\infty \left[\frac{d}{dE} \frac{\phi(E)}{P(E)} \right]^2 W(E) dE \quad (30)$$

subject to the constraint

$$f = \sum_{i=1}^m \left[\frac{A_i^c - A_i}{\Delta A_i} \right]^2 \quad (31)$$

Here P is a trial spectrum, $W(E)$ is a weighting function, usually of the $1/E$ -type, ΔA_i is the uncertainty of the measured activity A_i , and A_i^c is the calculated activity

According to Kam and Stallmann /34/ methods used in CRYSTALL BALL give smooth solutions which are not sensitive to the selection of the trial spectrum.

The program WINDOWS /48/ is an extension of CRYSTALL BALL for calculating upper and lower limits of integrated quantities using a linear programming scheme suggested by Rust and Burrus /49/.

SWIFT

O'Brien and Sanna have used a Monte Carlo unfolding method in the program SWIFT /49,50/. The principle is quite straightforward, solution spectra are generated randomly until convergence is reached.

The unfolding problem is put in the form

$$A_i = \sum_{j=1}^n w_j \sigma_{ij} \phi_j, \quad i = 1, \dots, m, \quad (32)$$

where

A_i = the response of detector i ,

ϕ_j = unknown differential flux in group j ,

σ_{ij} = the response function of detector i in energy group j ,

$w_j = E_{j+1} - E_j$ is the width of group j .

The unfolding is started by selecting n random numbers over the interval $(0,1)$ and possibly weighting them according to one of several schemes. The random numbers R_j , $j = 1, \dots, n$, are used as "group fluxes"

$$w_j \phi_j = R_j \quad (33)$$

to calculate activities

$$A'_i = \sum_{j=1}^m \sigma_{ij} R_j. \quad (34)$$

A normalization factor is then calculated as

$$N = \sum_{i=1}^m A_i / \sum_{i=1}^m A'_i, \quad (35)$$

and the Monte Carlo estimate of the flux is

$$\phi_j = N R_j / w_j, \quad j = 1, \dots, n, \quad (36)$$

and the corresponding responses are

$$A_i^C = \sum_{j=1}^n w_j \sigma_{ij} \phi_j, \quad i = 1, \dots, m. \quad (37)$$

The goodness of the random solutions is compared using the relative fitting errors of responses

$$H_i = (A_i^C - A_i) / A_i, \quad i = 1, \dots, m. \quad (38)$$

Any choice of ϕ , say the k^{th} choice, is considered a better estimate than the l^{th} choice if

$$\left[\max(H_i) \right]_k < \left[\max(H_i) \right]_l, \quad (39)$$

To smooth statistical oscillations the mean value of, e.g., the four best solutions is used.

The rate of convergence of the method can be estimated as follows. If there are s "important" groups contributing to each H_i , then it will take at least

$$k = (1/f_k)^s \quad (40)$$

trials to produce a solution yielding

$$\left[\max(H_i) \right]_k \leq f_k, \quad (41)$$

where f_k is a given convergence limit.

The Monte Carlo method is very time consuming as it takes 1 000 - 10 000 seconds of CDC 6600 time to produce a realistic spectrum. However, since the time goes mainly to sampling random numbers, several sets of measured data can be unfolded simultaneously with very little extra computing time.

The advantage of the method is that no prior information of the solution is required, though it could be used to accelerate convergence. The method has been tested with measured and simulated Bonner sphere data in reactor and accelerator applications using 25 energy groups. It has given good results when the experimental errors have been less than about 20 %, with larger errors the method fails in an obvious way indicating the presence of large uncertainties.

The method seems to be best suited for applications where the response functions cover the whole energy range of interest.

Though the Monte Carlo method is quite different from other unfolding techniques it can still be interpreted as one method

of minimizing a certain norm of the fitting errors of measured and calculated responses.

Sanna /51/ has also developed the faster iterative program BON for unfolding Bonner sphere data.

5 CONCLUSION

Multicomponent activation detectors have been shown feasible for reactor neutron spectroscopy with test measurements in a Triga reactor and their composition and fabrication principles have been published. The suitability of multicomponent detectors for 14 MeV fusion neutron measurements has also been studied with simulation calculations. At this stage, however, it is difficult to say whether multicomponent detectors will find a significant role in practical work. Though the concept is physically elegant, it is not yet clear in which applications its advantages can outweigh its limitations to such a degree that changes in established methods are justified. The prospects would be the most promising in applications where large numbers of routine measurement are required.

To promote the multicomponent principle, designs and test measurements should be made for different applications. Multicomponent detectors should also be made readily available for potential users so that they could offer a competitive alternative to commercially available activation foil sets.

Multireaction spallation detectors have been used for secondary hadron measurements at very high primary energies. The results can be used in radiation protection design and provide test material for hadron cascade simulation studies.

A large part of the work is concerned with the interpretation of activation detector measurements. A relatively simple method has been installed on minicomputer for the fast fitting of reactor neutron spectra.

The more sophisticated unfolding program LOUHI has been revised and extended. The new versions LOUHI78 and LOUHI82 have been used for both reactor neutron spectrum and high energy hadron spectrum unfolding. Though these problems are mathematically similar, they differ considerably in the amount of *a priori* information of the spectrum and in the accuracy of cross section data. LOUHI78 has been made publicly available at the Computer Physics Communications Program Library.

Though the applications described in this work are connected with multicomponent or multireaction detectors, the programs can be used also with conventional activation detectors as well. The LOUHI-type programs are independent of the physical application and can be used in various problems involving the solution of a group of linear integral equations of the first kind.

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CORRIGENDA

Publication II

p. 164 eq. (40) V_{ii} should be $\sqrt{V_{ii}}$

p. 165 eq. (47) V should be V_{ii}

Publication III

p. 123 eq. (2.15) r^e should be R_o

p. 124 eq. (2.27) Q_o/Q should be Q_o

p. 129 line 28 charged should be changed

p. 130 eq. (3.20) $\sigma_{ik} \frac{E_b - E_{b-1}}{2}$ should be $\sigma_{ik} + \frac{E_b - E_{b-1}}{2}$

p. 130 line 19 components of the gradient should be
componets of the step

Publication VIII

p. 423 Table 9 A^{meas} should be $A^{\text{meas}} \cdot 10^{24}$

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