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FOR DETERMINING NEUTRON SPECTRA
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ABSTRACT

This report describes the activation method used in our laboratory for experimental determination of neutron spectra and fluences. The list of foils is given and the codes for the evaluation of measured values are described.

1. INTRODUCTION

The activation methods belong at present to the classic measuring techniques of experimental reactor physics. Though they have many imperfections there is a number of applications we cannot manage without them. This is especially true for the high-flux experimental and power reactors. In any case it is necessary to bear in mind that they are highly time-consuming and need an accurate recording of the whole procedure. The measurement is always absolute which means that we must work with a carefully prepared apparatus and accurately write down all time regimes of the irradiation and measurement. In order to use fully the parity of the foil and the differences in half-lives of induced activities, it is necessary to find the optimum time regime for each foil and each time of experiment.

It is therefore not possible to determine any universal procedure suitable in any situation and for any set of activation materials. At first we must study the spectra of induced activities /1/. From the viewpoint of the method itself it is possible to divide the measurement into two stages. In the first one we take notice of the irradiation of foils and the measurement of their activities. In the second stage we shall deal with the evaluation of measured data and its physical interpretation. One of the weakest points of the activation method is that during a short time we must perform a great number of activity measurements of irradiated foils and usually only after the experiments are finished it is possible to evaluate all results. The object of the first stage is to obtain, as soon as possible, at least the activities per nucleus which gives us the first physical information. In practice it requires to have either an analyser-computer system working in on-line mode, or to have a quick and ready software for a suitable minicomputer or programmable calculator working in off-line mode.

2. MEASURING FOIL ACTIVITIES

In our laboratory we specialised in the foils emitting gamma-rays and having good mechanical and chemical properties in extreme conditions (high temperature, pressure, etc.).

Therefore, we do not use such well-known materials as sulphur and phosphorus whose nuclear properties are very advantageous. For the gamma ray activity measurements the following methods are used:

1. The scintillation detector SKG with NaI(Tl) crystal with dimensions ϕ 45 x 50 mm.
2. The scintillation detector SKG with NaI(Tl) crystal ϕ 40 x 1 mm.
3. A Ge-Li spectrometer with an active volume of 53 ccm.

Our foils have a diameter of 4 mm, 17 mm or 1/2", and thickness up to 3 mm. For each radionuclide we measure the characteristic photopeak or a certain part of the integral spectrum. In the case of scintillation measurements (simple spectra) the chosen part of the spectrum is integrated by a single channel analyser, while the spectra from the Ge-Li detector are punched and evaluated using the code PIKAR.

The apparatuses are calibrated with the set of standard sources having the same geometrical shape as the foils. The photopeak efficiency in the energy region 0.2 - 1.6 MeV is described by the formula $\eta = A.E^B$, where A and B are constants determined experimentally. Where possible, a direct comparison with a corresponding standard source is made (^{24}Na , ^{60}Co , ^{54}Mn). The reproducibility of measurements is very good, repeating the calibration gives the efficiency values within $\pm 2\%$. For SKG and Ge-Li detectors two distances of the foils are used, for which the efficiency differ approximately ten times.

2.1 The code PIKAR

The code PIKAR serves for automatic evaluation of simple gamma ray spectra obtained from a semiconductor detector on the computer ADT 4100. It searches for peaks and determines their positions, areas and relative standard deviations. In choosing the algorithm we started from the fact that the peaks searched for are sufficiently expressed and isolated in the spectrum so that we may adopt a relatively simple way of their location. In searching for the beginning and the end of the peak we assume that the background is linear. We do not make

any assumptions as to its shape, only the magnitude of the pulse rate change is studied in comparison with the mean value obtained from five preceding channels. Before the searching procedure starts, the code allows to smooth the spectra with a parabola. The code is very simple and reliable enough for evaluating the simple spectra. False peaks are easily eliminated both in accordance with their positions in the spectrum and very high value of the error of evaluated area. The code PIKAR is written in the language BASIC FORTRAN for the computer ADT 4100. It is not suitable for complex spectra or spectra obtained by a scintillation spectrometer.

3. THE SPECTROMETRY

To evaluate the measured values the code FACT is used which provides the foil activity. Neutron spectra unfolding is made by iterative codes SAND II and RFSP.

3.1 The code FACT

The code FACT makes all time corrections for the activation and decay and provides the foil activity normalised for one nucleus. The decay data are taken from /3/ and /4/. The data needed for the evaluation are on the so called tape of constants. It is not a part of the code but it is read each time before the first calculation. It is advantageous from the viewpoint of making the corrections or supplements into it. The constant field dimension is 40 and for each reaction it contains the following informations:

1. The name (code) of the foil,
2. The number of target isotope nuclei per one gramm of foil,
3. The detection efficiencies for foil activity measurements.

The numbers of nuclei and the efficiencies are corrected for the isotope abundance in the natural mixture and the gamma line branching ratio in the spectrum.

The method of activity measurement is defined by a so called number of spectrometer, NS (NS = 1 - 7).

- NS = 1, 2 - the scintillation detector SKG
- NS = 3 - the scintillation detector SKX
- NS = 4, 5 - the Ge-Li spectrometer
- NS = 6, 7 - these numbers are reserved for further photopeak or for other method of measuring.

A survey of all methods for single foils is given in table I.

For several radionuclides we do not have a good calibration and therefore we perform relative measurements with them. We set $NS = 0$, and the program computes the time-corrected counting rates. It is often suitable to know the decay time before the measurement. This value is then printed as DEC (in hours) among results.

3.2 Calculating the neutron spectrum

The unfolding of neutron spectra is made by codes SAND II and RFSP. If we are interested only in the fast part of the spectrum, we use the code SAND ADT. It is a simplified version of SAND II. The libraries of cross-sections as well as input spectra are from SAND II. If there is a possibility, our own experimental spectra obtained by differential methods (stilbene, hydrogen counters) are used.

In evaluating the activation methods as spectrometric methods it is necessary to bear in mind several important facts. First of all, they are not spectrometric methods but integral ones. The only objective criterion here is the correspondence of the measured activity with that calculated from obtained neutron spectrum. From the method of calculation it is obvious that the obtained solution is only the case which is physically most probable, its trustworthiness depending on many circumstances. The activation method offers therefore only the information about the character of the neutron spectrum or the verification of correctness of the assumptions obtained in some other way. Obviously, the activation method must always be combined with differential measurements, or reliably calculated spectra have to be used. The set of activities may to a certain extent modify the shape of the zero approximation and to shift it nearer to the actual spectrum. Notwithstanding this, the influence of the input information on the calculated spectrum is very significant and the choice of a good zero approximation is one of the basic prerequisites of satisfactory work.

4. DETERMINING THE NEUTRON FLUENCE

During a long-term experiment, e.g. in material testing, it is also necessary to know the neutron fluences. From the view-

point of the method the way of their determining is very similar to the spectroscopical method. For foil activity measurements the same apparatuses are used as for neutron spectrometry. Because the number of integrators is small, we use, if it is possible, a direct comparison with the corresponding standard. It is more accurate and effective than reading the detection efficiencies from the calibration curve.

Evaluating the integrators requires to know the neutron spectrum. This spectrum, measured or calculated before the irradiation, is then used to calculate the effective cross section of all integrators.

4.1 The choice of integrators

For the majority of irradiation experiments the most important role plays the fast neutron region. The set of integrators for this region is the best one and enables to obtain relatively important information concerning the neutron spectrum. Demanding requirements are imposed on material purity and thermal endurance.

For standard monitoring during material testing the following reactions are recommended by IAEA:

$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ - owing to a very high cross section of the reaction, only Al-Co alloys with cobalt content up to .1 percent may be used. Aluminium must have the purity 5N to suppress parasitic activities.
 $T_{1/2} = 5.27 \text{ a}$
 $E_{\gamma} = 1.17 \text{ MeV}$
 1.33 MeV

$^{93}\text{Nb}(n,n')^{93\text{m}}\text{Nb}$ - serious troubles are involved due to relatively large impurity of Ta presented everywhere in basic material. When metallic niobium is used, foils of thickness up to .02 mm are produced. To reduce the admixture it is advantageous to use Nb_2O_5 . The niobium activity measurement, however, has not yet been solved.
 $T_{1/2} = 11.4 \text{ a}$
 $E_{\gamma} = .018 \text{ MeV}$

$^{54}\text{Fe}(n,p)^{54}\text{Mn}$ - the foils enriched with the ^{54}Fe isotope are often used but also the natural iron with the purity 5N is applicable. The activity measurement is easily made, comparing with the ^{54}Mn standard.
 $T_{1/2} = 312 \text{ d}$
 $E_{\gamma} = .845 \text{ MeV}$

^{60}Co - this reaction has a very low cross section which implies a strong requirement on target material purity. First of all the content of cobalt must not exceed 1 ppm. For the activity measurement we use the standard ^{60}Co .

All activity measurements must be carried out with the β -Li detector. Practically in all cases parasitic activities are present due to impurity activation. During the stay of foils inside the irradiation rigs and during their manipulation in hot chambers, the foils are considerably contaminated. For niobium foils the optimum method consists in the combination of chemical separation with the measurement using planar silicon detector.

Except the reactions mentioned above there are other ones suitable for flux integrating. Their utilisation is however not so frequent as there are several troubles with nuclear data and with the availability of materials. Our code therefore contains information concerning the following reactions:

-) $^{93}\text{Nb}(n,n')^{93\text{m}}\text{Nb}$
-) $^{54}\text{Fe}(n,p)^{54}\text{Mn}$
-) $^{58}\text{Ni}(n,p)^{58}\text{Co}$ - this reaction is suitable for short-timed experiments; the correction for the $^{58\text{m}}\text{Co}$ burn-up is necessary.
-) $^{56}\text{Ti}(n,p)^{56}\text{Sc}$ - involves troubles with the purity of material.
-) $^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$
-) $^{60}\text{Ni}(n,p)^{60}\text{Co}$ - a very promising reaction, but it needs experimental testing.
-) $^{55}\text{Mn}(n,2n)^{54}\text{Mn}$ - involves troubles with material purity.
-) $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$
-) $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ - reliable data are missing.
-) $^{109}\text{Ag}(n,\gamma)^{110\text{m}}\text{Ag}$ - silver may be used only in an alloy with aluminium (too high cross section).

.2 The code FLUE

The code FLUE /5/ serves for the calculation of the mean value of the neutron flux density and the fluence in the chosen energy interval. The actual time course of the irradiation may be approximated by the following scheme (fig. 1), the power being constant over each cycle lasting T_1 days. The foil activity is then calculated as the sum of partial activities and it is re-

lated to the date of the measurement. For the i-th cycle it holds

$$A_i = P_i \phi_{eff} G_{eff} (1 - e^{-\lambda t_i}) e^{-\lambda t_i}$$

where t_i is the decay time and ϕ_{eff} and G_{eff} are in the following relation

$$G_{eff} = \frac{\int (\eta(E) G(E)) dE}{\phi_{eff}} ; \quad \phi_{eff} = \int \eta(E) dE$$

The foil activity after n cycles is then given as

$$A = \phi_{eff} G_{eff} \sum_{i=1}^n P_i (1 - e^{-\lambda t_i}) e^{-\lambda t_i}$$

As the fluence is defined by a formula after irradiation we get

$$F_{eff} = \phi_{eff} \sum_{i=1}^n P_i$$

$$F_{eff} = \frac{A}{G_{eff}} \cdot \frac{\sum P_i t_i}{\sum P_i (1 - e^{-\lambda t_i}) e^{-\lambda t_i}}$$

The power of the reactor (value P_i) is measured in arbitrary units (usually in kW or MW), so that ϕ_{eff} refers to the neutron flux density per power unit. The scheme of FLUS is very similar to that of FACT. It again contains the tape of constants with the data for each foil. Other informations are used as input data. First of all it is the information about irradiation (the code permits 30 cycles at the utmost). For the fluence evaluation the knowledge of the neutron spectrum is necessary. The spectrum serves also for calculating the effective values of G_{eff} and ϕ_{eff} . As the results, the following values are printed:

FLUX - the mean value of the neutron flux density per power unit; it has the character of flux density averaged over the whole interval of irradiation.

FLUENCE - the fluence above the chosen energy threshold.

5. CONCLUSIONS

This report describes briefly the activation method as it is used in our laboratory for determining neutron spectra and fluence. It is obvious that each laboratory modifies the general principles according to its requirements and conditions. The most important role play the experiments and measurements to be performed and the apparatuses available. As already mentioned hereinbefore it is necessary to obtain reliable information about neutron spectra because this is one of the main prerequisites for a successful use of this method. The well-known troubles with nuclear data and the imperfection of codes were supposedly neglected. These matters are under intensive study in world-wide scale.

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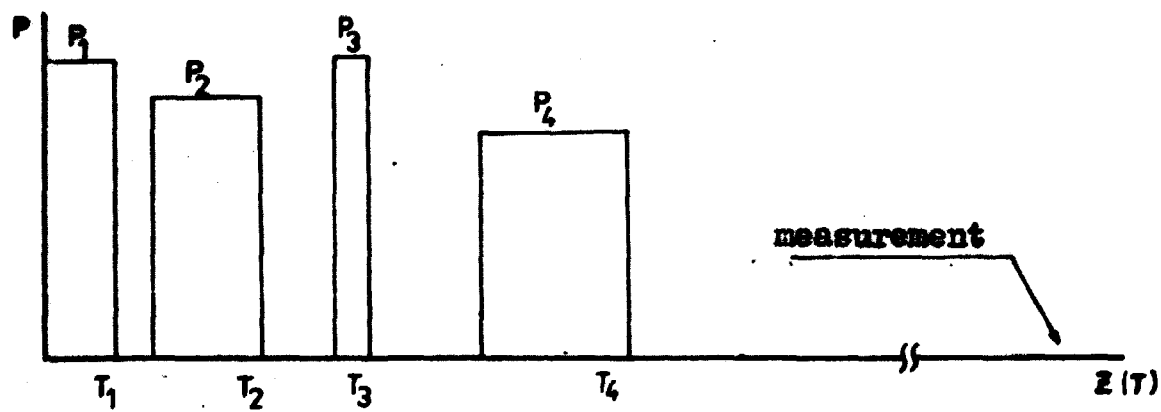


Fig. 1. Time diagram of irradiation.