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Use of threshold activation technique in estimating energy distributions of accelerator produced neutrons

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Abstract. A study of effective implementation of threshold activation technique for neutron spectral analysis in the environment of an alpha-cyclotron target is presented. The activation data are analysed using LOUHI-82 code. Optimal choices of the regularization parameters of the code are studied and discussed. Energy distribution of neutrons emitted from thick targets of Be, C and Ta irradiated by 40–45 MeV alphas is discussed.

Keywords. Threshold detectors; neutron spectrum; measurement; LOUHI-82 code; accelerator produced neutrons.

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1. Introduction

With the widespread application of medium energy accelerators in radiation therapy, material damage studies, production of radioisotope for nuclear medicine etc, the fundamental importance of accelerator shielding and dosimetry has increased. Therefore, the requirement of a technique for routine measurement of neutron energy spectra in accelerator environment in the presence of intense gamma radiation becomes important. Threshold detectors have been used to measure neutron spectra due to the bombardment of various thick targets with accelerated charged particles (Ringle 1963; Wadman 1967; Shin *et al* 1981; Birattari and Salomone 1985; Khasnabis *et al* 1987). Though the activation technique of threshold detectors has also been widely used to measure neutron energy distribution in reactors (Chiocchio *et al* 1971; Kuijpers *et al* 1977) and gamma energy distribution (Nath and Schulz 1975; Hirayama and Nakamura 1977) the method of unfolding is not well established and the implementation of the technique is still difficult due to the non-availability of the cross-section data and large uncertainties where they are available.

The aim of the present work is to study the implementation of the activation technique in routine measurement of neutron spectra in accelerator environment and to study the suitability of the general purpose unfolding code LOUHI (Routti and Sandberg 1980) in data analysis. Similar studies have been carried out by Birattari and Salomone (1985) with LYRA and SAND codes. The code LOUHI, however, offers more flexibility compared to LYRA and SAND in terms of a priori information input and thus becomes more suitable, at least theoretically, in the analysis of threshold activation data.

2. Experimental details

The experiment was carried out using the variable energy cyclotron at Calcutta. Accelerated alpha particles of energy 40–45 MeV are completely stopped in beryllium, carbon and tantalum targets thicker than the alpha range. Table 1 gives the thickness, the density, the alpha range (for 50 MeV alphas) together with the Q-values for the (α, xn) reactions in the target material.

The beam radius was 8 mm on the target surface. The target was insulated from the cyclotron and the beam transporting tube and connected to a digital scaler through a current integrator to estimate the total number of alpha particles falling on the target. In order to suppress the escape of the secondary emission electrons, a guard ring at a potential of -1600 V was used.

Activation detectors in stacks were kept at a distance of 15 cm from the target and were irradiated with neutrons from the bombardment of the specified targets by 1·5–2·0 μA alpha beam for about an hour. Activities between 100 and 10,000 Bq were allowed to build up to enable estimation of the activity with a statistical uncertainty $\pm$ 5%. Induced activity was determined by measuring the gamma spectrum associated with the activated foils with a Ge(Li) detector. The self-absorption of the neutron beam in the stack of detectors is negligible (Birattari and Salomone 1985) and ignored in the present calculations.

Table 1. Properties of target materials.

Target	Thickness (mm)	Alpha range* (mm)	Reaction Q-value (MeV)			
			(α, n)	$(\alpha, 2n)$	$(\alpha, 3n)$	$(\alpha, 4n)$
Be	3·0	1·18	+ 5·701	− 13·020	− 26·144	− 47·426
C	4·0	0·94	− 8·507	− 21·727	− 44·896	—
Ta	4·0	0·29	− 10·000	− 16·359	− 24·855	− 31·850

* For 50 MeV alphas.

Table 2. Characteristics of the threshold reactions used.

Reactions	Isotopic abundance (%)	Effective threshold (MeV)	Half-life	γ-ray energy (MeV)	Branching ratio (%)	Cross-section reference
$^{27}Al(n, p)^{27}Mg$	100	2·7	9·5 *m*	0·84	71·8	a, b
$^{27}Al(n, \alpha)^{24}Na$	100	5·9	15·0 *h*	1·37	100	a, c
$^{48}Ti(n, p)^{48}Sc$	73·7	5·2	44·0 *h*	1·31	100	b
$^{51}V(n, \alpha)^{48}Sc$	99·75	1·6	44·0 *h*	1·31	100	d
$^{58}Ni(n, p)^{58}Co$	68·3	1·2	71·0 *d*	0·81	99·4	a
$^{58}Ni(n, 2n)^{57}Ni$	68·3	12·5	36·0 *h*	1·38	77·9	c
$^{63}Cu(n, 2n)^{62}Cu$	69·1	10·1	9·8 *m*	0·511	196	b
$^{65}Cu(n, p)^{65}Ni$	30·9	2·8	2·52 *h*	1·48	23·5	d

a, Goldberg *et al* (1966); b, Schett *et al* (1974); c, Bayhurst *et al* (1975); d, Ringley 1963.

2.1 *Selection of activation foils*

The material for activation detectors was chosen based on the following criteria: (i) the material should be easily available and no enrichment is necessary; (ii) the material should be chemically stable and solid at room temperature (20–40° C); (iii) the foil material should be available either with very high purity or with an accurately known percentage of impurity; (iv) the half-life of the produced radionuclide should be longer than a few minutes, and shorter than 100 days; (v) the produced radionuclide should be a gamma emitter and interference from competing reactions should be negligibly small; (vi) detailed knowledge about the utilized reactions, e.g. cross-section data, branching ratios and gamma ray emission energy of the produced radionuclides must be known; (vii) the measurable energy range covered by the threshold energies should be as wide as possible. Based on the above considerations, Ni, Al, Cu, Ti and V were chosen as threshold detectors and foils of purity greater than 99% were procured. Table 2 gives the threshold reactions used in the present measurements, along with the effective thresholds (Ringle 1963) of the reaction, half-lives of the residual nuclei, gamma ray energies used for the measurements and the branching ratios.

3. Data analysis

The measured total activity of a threshold activation foil is related to the cross-section and the unknown flux distribution by

$$A_i = \int_0^\infty [\sigma_i(E)\phi(E)\,dE]N_i \exp(-\lambda_i t_w)[1-\exp(-\lambda_i t_I)] \times [1-\exp(-\lambda_i t_c)]p_i/(\lambda_i t_c) + \varepsilon_i, \quad (1)$$

where A_i is the activity of the residual nucleus after the ith threshold reaction, $\sigma_i(E)$, the energy dependent ith reaction cross-section, $\phi(E)$, the unknown differential flux density spectrum of neutrons, ε_i, the experimental error of the measured activity, λ_i, the decay constant of the ith isotope, p_i, the number of photons per decay in the ith reaction; t_w, the time between end of exposure and beginning of counting, t_I, the irradiation time, t_c, the counting time and N_i, the number of nuclei in the foil for the ith reaction. Equation (1) can be written as

$$A_i = \int_0^\infty \sigma_i(E)\phi(E)\,dE + \varepsilon_i, \quad i = 1,\ldots,m, \quad (2)$$

where A_i absorbs the normalizing constant which relates measured activity to flux density and m is the number of activation reactions used.

The numerical solution of (2) requires discretizing the equation. A good representation of the spectrum can be obtained by defining a number n of flux intensity intervals that greatly exceeds m, the number of activation reactions employed. Thus the system of equations to be solved becomes under-determined in nature. It is impossible to solve such a system of equations without any a priori information about the solution. The quality of the solution depends on the quality and quantity of the information used. Even with some a priori information, the group of equations (2) does not have

a unique solution. The solutions can be grouped into three classes: exact, approximate and appropriate (Gold 1964). The exact solution satisfies the equations exactly but may have an unphysical shape; an approximate solution matches the measured activities with a reasonable accuracy. A physically acceptable approximate solution is called an appropriate solution, which generally is not unique either. This is because an m-dimensional vector A is transformed by the unfolding procedure to an n-dimensional vector ϕ, where the information about ϕ in the m-n dimensional subspace is completely arbitrary. This leads to non-uniqueness and bias due to approximation. There are several unfolding codes like LYRA (Nakamura and Hirayama 1976), SAND-II (Draper 1971) and LOUHI (Routti and Sandberg 1980), that can be used for activation data analysis. All these codes differ by the manner in which they obtain and utilize the a priori information. Among the codes, LOUHI offers the maximum flexibility in controlling the amount and type of information that can be supplied to unfold the spectrum and therefore is suitable for use in different situations.

In the present study we have used the LOUHI-82 version (Sandberg 1983) of the LOUHI code. The details of the LOUHI code have been reported by Routti and Sandberg (1980). However, we describe briefly the relevant portions of the data analysis procedure and input specification for completeness and to facilitate discussion of our results. For numerical solution of (2) by LOUHI, E, $\sigma_i(E)$ and $\phi(E)$ are made discrete and the integrals are replaced with quadrature forms. Once a set of n energy values E_j have been assigned, (2) can be described as:

$$A_i = \sum_{j=1}^{n} \sigma_{ij}\phi_j + \varepsilon_j, \quad i = 1, \ldots, m \tag{3}$$

or in matrix form

$$A = \sigma \cdot \phi + \varepsilon.$$

The unfolding equations are often ill-conditioned in the sense that small changes in the responses may cause relatively large changes in the solution. The ill-conditioning and the unphysical oscillations in the solution can be avoided by using a priori knowledge of the solution in a manner which is known as the regularization of the problem. In this method the discrete equation (2) is replaced with the least square problem (Routti and Sandberg 1980):

$$\text{minimize } Q = Q_0 + Q_p = Q_0 + \gamma \sum_{k=1}^{4} W_k Q_k$$

where γ is known as the regularization parameter and controls the overall importance of the a priori information in the minimization problem, W_k specifies the importance of the term Q_k, and

$$Q_0 = \sum_{i=1}^{m} r_i^{\varepsilon} \varepsilon_i^2 = \sum_{i=1}^{m} r_i^{\varepsilon} \left[A_i - \sum_{j=1}^{n} \sigma_{ij}\phi_j \right]^2$$

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

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

= the weighted square sum of deviations of the solution from a trial solution p,

$$Q_2 = \sum_{j=1}^{n} r_i^f \phi_j^2$$

= is the weighted square sum of the solution values,

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

= the weighted square sum of first differences of the solution,

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

= the weighted square sum of second differences of the solution.

The non-negativity of the solution is imposed by setting

$$\phi_i = X_i^2$$

and performing the minimization with respect to X_i. The equations defining the minimum are nonlinear and the minimization is carried out using an iterative algorithm. The code LOUHI has provisions for using different modes of weighing like, linear, relative and logarithmic, for the terms $Q_1, \ldots Q_4$. In the present study we have used the logarithmic mode of weighting in which case ϕ_j in the terms of Q replaced by $\ln X_j^2$, for example the term Q_4 becomes:

$$Q_4^{\log} = \sum_{j=2}^{n-1} r_j^d(\ln X_{j-1}^2 - 2\ln X_j^2 + \ln X_{j+1}^2)^2.$$

The weights W_i are used to specify the individual weights of different prior conditions. The weights r_j's are weights defined for each quadrature node E_j. In the present study we have used relative weighting for the responses which corresponds to

$$r_i^e = 1/(mA_i^2), \quad i = 1, \ldots, m$$

and this mode of weighting has been found to be more effective compared to the statistical weighting where

$$r_i^e = 1/(mA_i)$$

and for other cases we have set,

$$r_j^p = r_j^f = r_j^t = r_j^d = 1/n, \quad j = 1, \ldots, n.$$

The trial solution p in the present study is set as

$$P(E_j) = aE_j^{-1.7}$$

which approximately gives the expected shape of the solution spectra, where a is a normalizing constant.

A guess spectrum is required as the starting value in LOUHI to start the iteration. In the present case we have set the trial spectrum as the initial guess spectrum.

4. Results and discussion

At first the dependence of the unfolded spectrum on the initial guess spectrum, the trial spectrum P and the regularization parameter γ is studied using the experimental data on carbon. The following two initial guess and trial spectrum were used in the test:

(i) $\phi_j = P_j = 1{\cdot}18 \times 10^{-6}$,

(ii) $\phi_j = P_j = 1{\cdot}68 \times 10^{-4} E_j^{-1.7}$.

Figure 1 shows the neutron spectrum for carbon target at zero degree unfolded by the LOUHI-82 code for the initial and trial spectra 1 for different values of γ (2·0, 0·5, 0·05). It is observed that the unfolded spectra almost merges with the initial spectra down to a γ value of 0·1. At $\gamma = 0{\cdot}05$ the unfolded spectra show unnatural oscillations and thus cannot be taken as the appropriate spectrum. For comparison we have taken the neutron spectra due to 65 MeV alphas on carbon measured by Shin *et al* (1984) using the NE-213 scintillation technique.

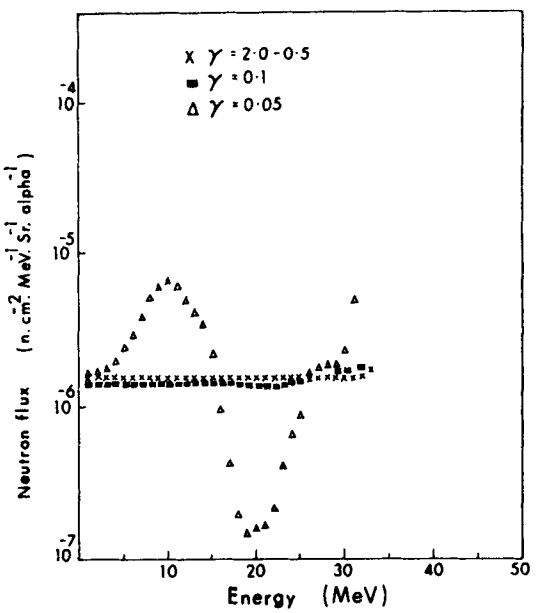

Figure 1. Neutron spectrum at 0° from thick carbon target bombarded with 45 MeV alphas, unfolded with initial and trial spectra $P_j = 1{\cdot}18 \times 10^{-6}$ with different values of γ.

Figure 2 shows the same neutron spectrum unfolded with the initial and trial spectra 2. Here, for $\gamma = 2{\cdot}0 - 0{\cdot}5$ the unfolded spectra almost coincide with the initial spectrum. At $\gamma = 0{\cdot}1$, the unfolded spectrum shows independent structures in the distribution though the mean trend remains almost the same as that of the initial guess. When the value of γ is reduced to 0·05, the unfolded spectrum shows unusual fluctuations.

Table 3 gives the values of Q_0 and Q_p for the two initial guess spectra 1 and 2, for

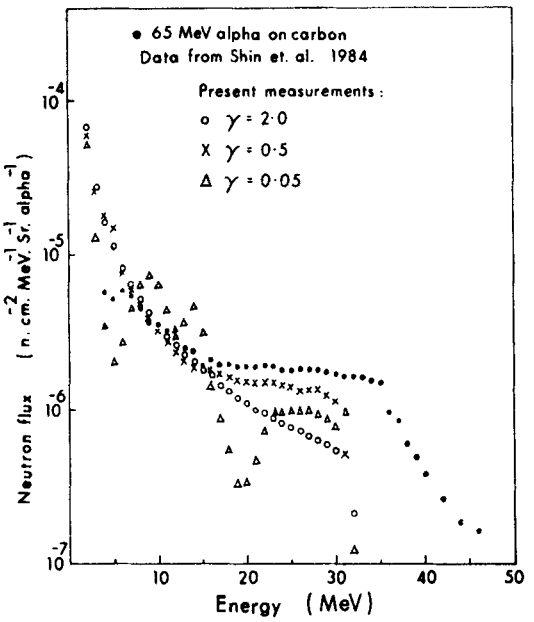

Figure 2. Neutron spectrum at 0° from thick carbon target bombarded with 45 MeV alphas, unfolded with initial and trial spectra $P_j = 1{\cdot}68 \times 10^{-4} E_j^{-1{\cdot}7}$ and with different values of γ, along with neutron spectrum as measured by Shin *et al* (1984) due to 65 MeV alphas on carbon.

Table 3. Values of Q_0 and $\gamma \sum_{k=1}^{4} W_k Q_k$ for activation data analysis of neutrons due to bombardment of 45 MeV alphas on carbon for two different initial and guess spectra (i) $\phi_i = p_j = 1{\cdot}18 \times 10^{-6}$ and (ii) $\phi_j = p_j = 1{\cdot}68 \times 10^{-4} E_j^{-1{\cdot}7}$ and for different values of γ.

	$\phi_j = p_j = 1{\cdot}18 \times 10^{-6}$				$\phi_j = p_j = 1{\cdot}68 \times 10^{-4} E_j^{-1{\cdot}7}$			
	Initial		Final		Initial		Final	
γ	Q_0	$\gamma \sum_{k=1}^{4} W_k Q_k$	Q_0	$\gamma \sum_{k=1}^{4} W_k Q_k$	Q_0	$\gamma \sum_{k=1}^{4} W_k Q_k$	Q_0	$\gamma \sum_{k=1}^{4} W_k Q_k$
2·00	3·398	3305·0	3·435	570·2	2·432	3178·0	2·447	515·3
1·00	3·398	1652·5	3·428	287·6	2·432	1589·0	2·446	253·5
0·50	3·398	826·3	3·422	137·3	2·432	794·5	2·445	129·1
0·20	3·398	330·5	3·406	50·88	2·432	317·8	2·441	47·34
0·10	3·398	165·3	3·38	27·9	2·432	158·9	2·164	11·36
0·05	3·398	82·64	2·032	6·47	2·432	79·45	1·949	5·06

different values of γ. In the table the column "initial" indicates the values obtained with the initial guess spectrum, and the column "final" indicates the values obtained after the optimization is carried out.

It is evident from table 3 that at high values of γ there is no reduction in the value of Q_0. In fact, the value of Q_0 increases after optimization. This is because by setting a high value of γ, more importance is given to the a priori information and thus minimization is carried out with respect to the a priori information part only, neglecting the Q_0 part completely. The situation changes as we lower the value of γ and at relatively low values (0·1 and 0·05) the final value of Q_0 is smaller than the initial value. But for $\gamma = 0{\cdot}05$ where the final value of Q_0 is appreciably smaller than the initial value, the unfolded spectrum shows unrealistic fluctuations.

Now, if we examine the two different choices of the initial spectra then we see that spectrum 2 has a lower initial values of Q_0 and its overall convergence both in terms of Q_0 and Q_p is better than spectrum no. 1. It is evident that the unfolded spectrum has a strong dependence on the initial guess spectrum. This will be more evident if we examine the unfolded spectra for $\gamma = 0{\cdot}05$ in both the cases. Here the values of final Q_0 or Q_p are not very different from each other, but the unfolded spectra do not show a similar overall trend and specially in the low energy portion of the spectrum, though there are similarity in the unrealistic fluctuations present in both the cases. Finally, it is evident that the spectra unfolded with spectrum 2 as the initial guess and trial spectrum resemble more closely the shape of the spectrum obtained by Shin *et al* (1984).

The above study reveals that it is profitable to choose spectra 2 as the initial and trial spectra and the value of γ should be kept between 0·5 and 0·1. However, this area needs careful and systematic study, since the dependence of the final spectrum

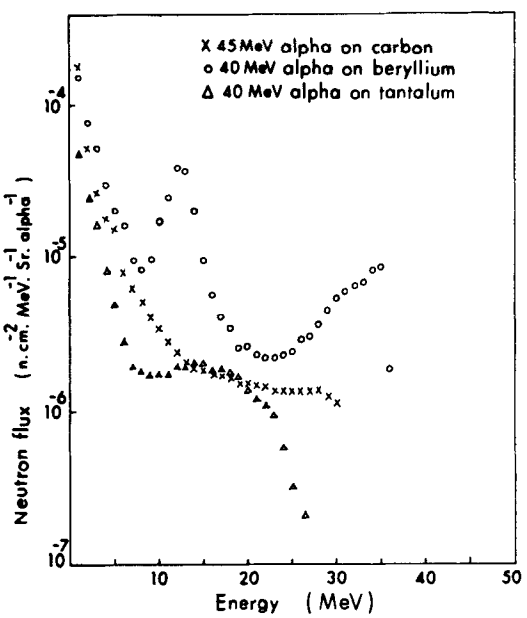

Figure 3. Neutron spectrum at 0° from thick targets of beryllium bombarded by 40 MeV alphas, carbon bombarded by 45 MeV alphas, and tantalum bombarded by 40 MeV alphas.

on the initial spectrum indicates that the objective function Q becomes multiextremal with respect to ϕ and the variable metric minimization technique used in LOUHI-82 code (or for that matter any deterministic minimization technique used in other existing codes) is only a local minimizer and thus has the disadvantage of getting trapped in any local minima close to the initial guess.

Figure 3 shows the energy distribution of neutrons at 0° from thick targets of Be, C and Ta bombarded with alphas. These are unfolded from the activation data using spectrum 2 as initial guess and trial spectra and $\gamma = 0{\cdot}5$. The energy distribution of neutrons in all the three cases can be divided into two regions e.g. low energy region (< 8 MeV) and high energy region (> 8 MeV). The major contribution in the low energy region comes from the Weisskopf-Ewing evaporation and multistep compound processes. This also explains the maxima of the distribution around 1–2 MeV. The high energy neutrons are due to single step and multistep direct processes, where the probability of particle emission decreases rapidly with increasing energy.

Except for Be which has a positive Q value for the (α, n) reaction and is a prolific neutron emitter, and therefore should be treated as a special case, the shape of the energy distribution of neutrons for C and Ta targets agrees well with the published results (Shin *et al* 1984).

5. Conclusion

Threshold detectors can be used as a routine procedure for obtaining neutron energy distributions in an accelerator environment. LOUHI-82 code can be used for unfolding the activation data. However, a detailed study of the regularization parameters is necessary to arrive at an universally optimal set of values, since the present study is restricted to a particular type of spectra and specific set of threshold reactions.

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