NEUTRON SPECTRUM ANALYSIS FROM DOSIMETRY EXPERIMENTS

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ABSTRACT

A fairly good approximation to a neutron spectrum can be obtained commensurable with the accuracy of the activity and cross section measurements. However, these approximations are valid only in regions where the coverage is determined by the cross sections and spectrum used and may be locally disturbed by poor resolution. Coverage and resolution are two properties which determine the goodness of a given monitor set. The trial spectrum influences the outcome of the unfolding only to the extent that the monitor set fails to provide the necessary coverage and resolution.

Key Words: reaction rate, cross sections, trial spectrum, coverage, resolution, neutron flux, number of iterations.

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The purpose of this paper is to present the type of analysis that is currently being done with neutron dosimetry data, specifically foil activation data to extract information for use in materials study. The objective is to give you an insight into the limitations or qualifications from this type of analysis.

The dosimetry experiments for materials application generally involve the irradiation of monitors in a reactor environment, and the analysis involves the mathematical procedure which transforms the measured data into a neutron flux distribution. While this determination of the neutron flux distribution may be of prime importance to the dosimetrist, it is usually of secondary importance to the material investigator. Their principal concern could be the intercomparison of radiation effects results on a wide variety of different materials irradiated at different reactor sites; but whatever is their concern, at least one additional step is required with the neutron flux distribution.

The type of analysis done here is not unique.¹ An experimentalist is constantly faced with the problem of measuring a distribution function $f(t)$; but due to distortions introduced by the measuring device, he observes a different distribution function $F(x)$. Following the usual procedure he denotes the operation of the measuring device by an operator O and writes the statement of his problem symbolically as

$$O[f(t)] = F(x) \quad (1)$$

Although the operator O can assume many forms, the experiment of irradiating monitors can be characterized by a linear operator so that equation (1) can be written as

$$F(x) = \int K(x,t)f(t)dt \quad (2)$$

where

$K(x,t)$ = response function of the monitors;

$f(t)$ = desired distribution; and

$F(x)$ = observed experimental distribution.

In the set of integral equations given by (2), the operator $\mathcal{O}$ does not always yield the same distribution $F(x)$ when applied to the same spectrum $f(t)$. The reason may be from noise in the measuring device, from the statistics associated with $f(t)$, or because the operator itself may be stochastic. Thus, in any experiment only a member of the many possible distributions is measured. Thus, in addition to the purely mathematical aspect of solving the set of integral equations, there are statistical considerations that must be considered. In all further discussions the response functions represent activation cross sections $\sigma_i(E)$, $f(t)$ is the neutron spectrum $\phi(E)$, and $F(x)$ is measured reaction rates α_i so that equation (2) takes the form

$$\alpha_i = \int \sigma_i(E)\phi(E)dE \quad i=1,2, \dots, n. \quad (3)$$

Here n represents the number of measured reaction rates in the experiment. The determination of $\phi(E)$ from the observed α_i 's is referred to as unfolding or inverting.

The main difficulty in any unfolding procedure originates from the fact that the problem is mathematically ambiguous. There is an infinity of solutions from which by some intuitive rules a physically reasonable or most likely one is selected. Even though some bounds could theoretically be

established from the condition that all flux values are positive, these bounds are too large to be of practical value at least for the differential spectrum. Consequently, the presently used unfolding methods do not attempt and are generally unable to give such bounds but rather present a single solution which for one reason or another is a reasonable candidate.

There are several broad classes of unfolding procedures. The first attempts in this field used finite linear combinations of suitable functions to represent the spectrum. Such functions may be of histogram type, or piecewise linear, or polynomials, perhaps in combination with exponential functions. The disadvantage of this class of methods is that a small set of functions is unable to approximate well enough a given spectrum whereas a large set tends to introduce oscillations and numerical instabilities into the solution.

A seemingly quite different approach uses the statistical method of linear estimation for obtaining the spectrum. Loosely speaking, one tries to find the most likely solution based on the information given by the reaction rate measurements and some a priori information which may include closeness to a first guess of the real spectrum. Like in the first class of methods the spectrum solution is also a linear combination of a fixed set of functions, but these functions are imposed by the problem and not by some outside criteria like simplicity or ease of computation. Consequently, the problem of numerical instability is less severe for these methods.

The two classes of methods discussed so far are "one-shot" procedures, meaning that the solution is obtained in a single step. This results quite often in physically unacceptable negative values in the unfolded spectrum.

To avoid such an outcome, iterative methods are used which start with an arbitrary trial spectrum and change it gradually until it conforms to the conditions imposed by the reaction rates, always avoiding negative flux values. The SAND-II code² uses this principle applying a mathematically somewhat unpredictable nonlinear transform for each approximating step. The method seems to be stable and working well in practice.

The linear estimation method can also be converted into an iterative procedure changing the a priori information contained in the guessed spectrum gradually at each iteration step. The CRYSTAL BALL code³ which will be used in this investigation is based on this principle. Other linear estimation methods like SPECTRA⁴ have iterative modes too.

All methods discussed above and possibly all feasible methods of unfolding have the common drawback that it is impossible to predict on theoretical grounds how good the unfolded spectrum approximates the true spectrum found in the reactor. In addition, one would like to know whether there are any ways and means to improve this approximation under specific conditions. To accomplish this goal a numerical simulation of the problem was designed with a modified version of CRYSTAL BALL to investigate the effects of parameters which influence the unfolded spectrum. The parameters investigated in this report are experimental errors, number and selection of monitors, and the trial spectrum.

NUMERICAL SIMULATION OF PROBLEM

Three neutron spectra and a set of forty reaction cross sections form the basis for the numerical experiments. The spectra and cross sections are taken from the reference spectrum library and the reference cross section library of the SAND-II code.

The three spectra (Figures 3, 5, and 7) are labeled in SAND-II as "Fission Form 2, Frye", "Non-equilibrium 'Thermal' (5.3×10^5 c) + Spectrum 7", and "AE-6 (AIM-6/CAESAR-II) + Thermal Below 0.414 eV + Fission Above 3 MeV". These were chosen arbitrarily as representatives of a fast, an intermediate, and a thermal spectrum, respectively. In the study to follow these spectra will represent the true spectra from which all comparisons will be made.

The SAND-II cross sections (of Table 1) and the three spectra are used to compute the reaction rates α_i^T according to the integral (3). The coverage of the cross sections for each of the three spectra are plotted, and from these plots a set of 20 cross sections is chosen to make the study.

The cross sections were distorted by adding random errors to each of the listed energy points E_k

$$\sigma_i^*(E_k) = \sigma_i(E_k)[1 + c(r - 0.5)], \quad (4)$$

where r is an equally distributed random variable in the interval (0,1) and c is a constant varying from experiment to experiment. From these distorted cross sections the corresponding distortion of the activities was computed by the same formula as (3)

$$\alpha_i^* = \int_0^\infty \sigma_i^*(E) \phi(E) dE. \quad (3a)$$

The difference $|(\alpha_i - \alpha_i^*)|/\alpha_i$ gives a measure of the change in the activities corresponding to the change in cross sections.

The effect of errors in the activity measurements is accomplished by distorting the exact activities using a similar random distortion as in formula (4).

The unfolding is carried out for each of the three reference spectra until the error between the given activities and the ones computed from the unfolded spectrum has the same order of magnitude as the change in activities from the distortion added to the exact activities. The influence of different trial spectra is treated by distorting the original spectrum in a way similar to formula (4). In several cases a constant spectrum is used.

RESULTS AND DISCUSSION OF RESULTS

The results must be viewed as preliminary in that not all questions receive a definite answer; that is, no attempt is made to pin down exact numerical values to each result. Rather the results indicate where further research would be needed and fruitful. Table 2 represents a few of the cases which were performed to answer the questions in the preceding sections. Figures 1-21 correspond to cases 1-21 of Table 2, respectively.

Input Errors (Figures 1-8)

Errors in cross section and activity measurements are equivalent as far as the outcome of the unfolding procedure is concerned. More specifically, α_1^* instead of α_1 should be used if the faulty cross section values $\sigma_1^*(E_k)$ are the basis for the unfolding procedure and the difference between the two is an additional change in the activity value which would compensate for the cross section values.

The computer experiments show that this difference between α_1^* and α_1 is smaller by approximately a factor 4, meaning that an average 10 percent error in the cross section values is equivalent to an error of about 2.5

percent in the activity values. The factor of 4 is related directly to the distribution function (4) so that a re-evaluation of this factor should be made with a more realistic representation of the cross section errors.

The reduction in errors for the cross section is of course due to partial cancellation of errors in the integral (3a). This works both ways. Since the unfolded spectrum can be considered a distortion of the real spectrum leading to distorted activity values, we can expect that an increase in the experiment errors of the activity measurements leads to an about four-fold increase in the errors of the unfolded spectrum, whereas increase in cross section errors are about the same magnitude as the unfolded spectrum. This is of course only true if a reliable unfolding can be obtained and to this extent the experiments seem to bear out the prediction.

Number and Selection of Monitors (Figures 9-21)

In all experiments a fairly good approximation (within about 10 percent of the true spectrum) could be obtained for one or more intervals in the full energy range. These intervals coincide roughly with the energy ranges covered by the monitors where the covering is determined both by the cross section and the spectrum used. The results from the 10 foil set are obviously poorer than the ones from the 20 foil set but no significant difference for the energy intervals could be found. A much more extensive study would be needed to find something like a "best" set of monitors.*

A closer look reveals the following situation: There are energy ranges which are not covered by the monitors. In these regions the unfolded spectrum

*The set of 14 reactions suggested by W. N. McElroy appears to be a good set for a fast spectrum (Figures 19-20).

is solely determined by the trial spectrum. This does not necessarily mean that within these regions the unfolded spectrum equals the trial spectrum. It may be a multiple of the trial spectrum or changed in some other way to get a smooth transition from the trial spectrum to the true spectrum. In addition, one can find some oscillations of the unfolded spectrum in respect to the true spectrum. This reflects the tendency to compensate for values which are too small in one part of the spectrum with larger values in another part to obtain the correct value for the integral (1). Heavy oscillations are excluded by the unfolding procedure, but small ones can be generated if the true and the trial spectrum have markedly different slopes at some energies. These oscillations must of course be consistent with the activity values for the given monitor set. The ability of a given set to prevent such oscillations may be called resolution of the set. Covering and resolution are the two properties which determine the goodness of a given monitor set.

Effect of Different Trial Spectra (Figures 9-14)

The discussions of the Section 3.2 give an immediate answer to the question of trial spectra. The trial spectrum influences the outcome of the unfolding only to the extent to which the monitor set fails to provide the necessary covering and resolution. On the other hand, these properties can be checked by unfolding the same spectrum with several different trial spectra. Energy regions in which the different unfolded spectra differ from each other are necessarily unreliable, whereas regions which give consistent results can be expected to be close to the true spectrum.

A trial spectrum which is closer to the true spectrum does not necessarily lead to a better approximation in the unfolded spectrum, unless it is already so close that the unfolding procedure introduces no further refinement. In the experiments a constant spectrum frequently gave better results than a distorted true spectrum.

SUMMARY AND APPLICATION

The results of the numerical simulation indicate the following facts:

1. The choice of the monitor set is essential to obtain a good approximation to a neutron spectrum by the method of unfolding irradiation data. More attention should be directed to the two properties, coverage and resolution, in the selection of a set. The number of foils is important to the extent that more coverage and resolution is possible with more foils.
2. The trial spectrum influences the outcome of the unfolding in regions which the monitor set fails to provide the necessary coverage and resolution. Although the choice of a trial spectrum which is closer to the true spectrum does not necessarily lead to a better approximation in the unfolded spectrum, a good trial spectrum will reduce the number of iterations considerably to obtain a solution.
3. The relative errors for cross section and reaction rate measurements are roughly additive and are of the same order of magnitude. If the cross section errors are truly random, their influence may be somewhat less since they tend to cancel each other out in the integral (3).

The preceding sections concern mainly the differential spectrum. A considerable interest exists in obtaining reliable values for the integral spectrum

$$\phi(E_1) = \int_{E_1}^{\infty} \phi(E) dE \quad (5)$$

or perhaps more general

$$\Phi(E_i, E_k) = \int_{E_i}^{E_k} \phi(E) dE. \quad (6)$$

One of the principal applications of dosimetry is the determination of damage to materials subject to irradiation in reactor environments. This is expressed by the integral

$$I = \int F(E) \phi(E) dE. \quad (7)$$

This formula is used in two ways. Knowing several spectra $\phi_i(E)$ and associated damages I_i an unfolding of the function $F(E)$ is obtained. If $\phi_i(E)$ is obtained by unfolding, the unfolding procedure is applied twice and its uncertainties compounded. In addition, the results of the preceding sections relating to coverage and resolution apply to the ϕ_i as used here. It seems desirable to subject this process to a more detailed mathematical analysis and to develop a reliable compound procedure.

If the damage function $F(E)$ is known the integral (7) is a generalization of the integral (5) and (6) and can be computed from the unfolded spectrum. The accuracy of these integrals is of course determined by the accuracy of the spectrum. Usually the accuracy of the integral spectrum will be higher since errors due to poor resolution will be washed out and poorly covered parts of the spectrum may not contribute substantially to the result.

It is also possible using linear programming techniques to obtain bounds for the integrals (5), (6), and (7) which in some favorable cases may be small enough to be of practical value. It can be suspected that favorable bounds for the integral (7) can be obtained exactly for those intervals in which consistently good results are obtained by unfolding procedures.

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Table 1. Reaction Cross Sections in SAND-II Library

Cover Name	Cover Symbol	Abbrev. Reaction Name	Complete Reaction Name	Decay Constant s^{-1}	No. of Points	Comments
Cadmium	CD				195	
Boron	B				148	
Gold	AU				621	
		CU63AM	CU63(N,A)CO60M	4.161E-9	621	1. The letters enclosed in parenthesis of the complete reaction name have the following meaning:
		AG109G	AG109(N,G)AG110M	3.171E-8	621	
		CO59A	CO59(N,A)MN56	7.463E-5	28	
		FE58G	FE58(N,G)FE59	1.783E-7	621	
		NI60P	NI60(N,P)CO60	4.161E-9	40	N = neutron A = alpha G = gamma P = proton.
		CU63A	CU63(N,A)CO60	4.161E-9	621	
		NI58P	NI58(N,P)CO58	1.119E-7	621	
		PU239F	PU239(N,F)FP	1.	445	
		SC45G	SC45(N,G)SC46	9.559E-8	621	2. FP = fission product.
		NA23G	NA23(N,G)NA24	1.286E-5	621	
		TH232G	TH232(N,G)TH233	1.	200	
		U238G	U238(N,G)U239	1.	200	
		AU197G	AU197(N,G)AU198	2.974E-6	621	3. The product nuclides of fissionable atoms were arbitrarily assigned values of 1 for the decay constant.
		CO59G	CO59(N,G)CO60	4.161E-9	621	
		IN115G	IN115(N,G)IN116M	2.139E-4	339	
		MN55G	MN55(N,G)MN56	7.483E-5	279	
		CU63G	CU63(N,G)CU64	1.500E-5	621	4. M = metastable state.
		TH232F	TH232(N,F)FP	1.	139	
		ZN64P	ZN64(N,P)CU64	1.500E-5	97	
		FE54P	FE54(N,P)MN54	2.559E-8	621	
		S32P	S32(N,P)P32	2.567E-7	108	
		SI28P	SI28(N,P)AL28	5.067E-3	58	
		S34A	S34(N,A)S131	7.349E-5	128	
		MG24P	MG24(N,P)NA24	1.284E-5	117	
		CL35A	CL35(N,A)P32	5.567E-7	37	

Table 1. (continued)

Cover Name	Cover Symbol	Abbrev. Reaction Name	Complete Reaction Name	Decay Constant s^{-1}	No. of Points	Comments
		NI582	NI58(N,2N)NI57	5.247E-6	21	
		I1272	I127(N,2N)I126	6.123E-7	25	
		ZR902	ZR90(N,2N)ZR89	2.456E-6	24	
		TI46P	TI46(N,P)SC46	9.574E-8	35	
		TI47P	TI47(N,P)SC47	2.339E-6	621	
		TI48P	TI48(N,P)SC48	4.384E-6	58	
		CU632	CU63(N,2N)CU62	1.169E-3	10	
		P31P	P31(N,P)SI31	7.349E-5	42	
		FE56P	FE56(N,P)MN56	7.463E-5	32	
		AL27P	AL27(N,P)MG27	1.217E-3	37	
		NP237F	NP237(N,F)FP	1.	222	
		U238F	U238(N,F)FP	1.	134	
		U235F	U235(N,F)FP	1.	891	
		AL27A	AL27(N,A)NA24	1.286E-5	135	
		IN115N	IN115(N,N)M	4.268E-5	66	

Table 2. Numerical Experiments

Case No.	Reference Spectrum*	Parameter	Comments
CROSS SECTION:			Number of foils = 20; trial spectrum is constant.
1	1	$\leq 10\%$	The distortions of the cross sections are equal to or less than 10% for CASE 1 and 5% for CASE 2.
2	1	$\leq 5\%$	
REACTION RATE:			Number of foils = 20; trial spectrum is constant.
3	1	$\leq 1\%$	The distortions of the reaction rates are equal to or less than 1% for CASES 3, 5, and 7 and 15% for CASES 4, 6, and 8.
4	1	$\leq 15\%$	
5	2	$\leq 1\%$	
6	2	$\leq 15\%$	
7	3	$\leq 1\%$	
8	3	$\leq 15\%$	
TRIAL SPECTRUM:			Number of foils = 20.
9	1	≤ 3.0	$\phi(E_k) \leq 3.0 * \phi_I^1(E_k) $
10	1	CONSTANT	$\phi(E_k) = \text{constant.}$
11	2	≤ 3.0	Where I is reference spectrum No. 1, 2 or 3.
12	2	CONSTANT	
13	3	≤ 3.0	
14	3	CONSTANT	
NUMBER OF FOILS:			
15	1	10	$\phi(E_k) \leq 3 * \phi_1(E_k) $
16	2	10	$\phi(E_k) \leq 3 * \phi_2(E_k) $

*Reference spectra:

1 = "Fission Form 2, Frye".

2 = "Non-equilibrium 'Thermal' (5.3×10^5 c) + Spectrum 7".

3 = "AE-6 (AIM-6/CAESAR-II) + Thermal Below 0.414 eV + Fission Above 3 MeV".

Table 2. (continued)

Case No.	Reference Spectrum*	Parameter	Comments
17	2	10	$\phi(E_k) = \text{constant.}$
18	3	10	$\phi(E_k) \leq 3 * \phi_2(E_k) .$
19	3	10	$\phi(E_k) = \text{constant.}$
20	1	14	$\phi(E_k) = \text{constant; reaction rates distorted 1\%.}$
21	1	14	$\phi(E) = \text{constant/ reaction rates distorted 15\%}$

APPENDIX B

ORNL-DWG 74-9026

Figure 1. Cross Sections Distorted (+10%),
Spectrum 1

Figure 2. Cross Sections Distorted (+5%),
Spectrum 1

ORNL-DWG 74-9030

Figure 3. Reaction Rates Distorted ($\pm 1\%$),
Reference Spectrum 1

ORNL-DWG 74-9031

Figure 4. Reaction Rates Distorted (+15%),
Reference Spectrum 1

Figure 5. Reaction Rates Distorted (4.1%), Reference Spectrum 2

ORNL-DWG 74-7023

Figure 6. Reaction Rates Distorted ($\pm 15\%$),
Reference Spectrum 2

Figure 7. Reaction Rates Distorted (+1%),
Reference Spectrum 3

Figure 8. Reaction Rates Distorted (+15%),
Reference Spectrum 3

Figure 9. Trial Spectrum Distorted (+300%),
Reference Spectrum 1

Figure 10. Trial Spectrum Constant,
Reference Spectrum 1

Figure 11. Trial Spectrum Distorted (+300%),
Reference Spectrum 2

Figure 12. Trial Spectrum Constant,
Reference Spectrum 2

Figure 13. Trial Spectrum Distorted ($\pm 300\%$),
Reference Spectrum 3

Figure 14. Trial Spectrum Constant,
Reference Spectrum 3

Figure 15. Trial Spectrum Distorted (+300%),
Reference Spectrum 1 (Number of
Foil = 10)

Figure 16. Trial Spectrum Distorted ($\pm 300\%$),
Reference Spectrum 2 (Number of
Foils = 10)

Figure 17. Trial Spectrum Constant
 Reference Spectrum 2 (Number of
 Foils = 10)

Figure 18. Trial Spectrum Distorted (+300%)
Reference Spectrum 3 (Number of
Foils = 10)

Figure 19. Trial Spectrum Constant
Reference Spectrum 3 (Number of
Foils = 10)

Figure 20. Reaction Rates Distorted (+1%),
 Reference Spectrum 1 (Number of
 Foils = 14)

Figure 21. Reaction Rates Distorted (+15%),
Reference Spectrum 1 (Number of
Foils = 14)