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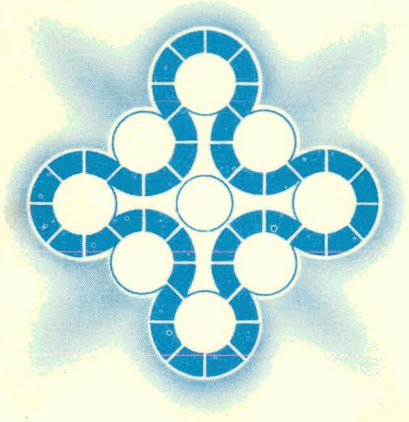
SELF-SHIELDING IN STACKED FOILS

R. G. Nisle
Y. D. Harker



Aerojet Nuclear Company

NATIONAL REACTOR TESTING STATION

Idaho Falls, Idaho — 83401

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BY

R. G. Nisle
Y. D. Harker

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SUMMARY

Both theoretical and experimental work on self-shielding in a stack of 5 mil gold foils is reported. The theoretical work assumes first flight encounters of neutrons from an isotropic flux in a thin infinite slab. Formulas for the perturbed flux and for the induced activity are derived. Calculated activities are then fitted to the observed activities. This procedure is thus an extension of the sandwich foil technique.

A significant conclusion to be drawn from these results is that this technique provides a means for measuring flux spectra by the use of stacks of foils of relatively few absorbers. The technique does not depend on the existence of an isolated, high-intensity resonance and no correction for background cross sections is needed. A large number of foils of a small number of absorbers replaces a few foils of a limited number of suitable absorbers. Thus, the number of points in a spectrum can be increased beyond the limit fixed by the availability of suitable absorbers.

This report forms the basic reference for additional work with other absorbers and mixtures of absorbers.

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SELF-SHIELDING IN STACKED FOILS

R. G. Nisle and Y. D. Harker

I. INTRODUCTION

The problem of flux-spectrum evaluation in fast breeder reactors is somewhat more complicated than in the case of thermal reactors. The flux variation with energy follows no simple division into Maxwellian and 1/E functions. A large variety of computer codes have, therefore, been devised to solve this problem. The experimental confirmation of results calculated by these codes can be accomplished by two possible methods: instrumental methods, of which the proton-recoil technique is one, and activation methods such as the sandwich foil and threshhold detector techniques. Time-of-flight methods, while theoretically possible, are subject to practical difficulties.

The problem of deriving flux spectrum information by unfolding integral data based on activation measurements can be well handled by such computer codes as INSPECT[1] or SAND II[2].

At energies above 1 keV, cross sections are generally small enough that self-shielding is not usually thought to be a problem. Results reported here, however, show that self-shielding does exist in gold foils as thin as 5 mils for energies in the keV range. Thus, self-shielding, i.e. shielding of the inner layers of a foil by surface and near surface layers, can be used to measure the flux at these energies. The sandwich foil technique[3] is one example in which self-shielding is used to obtain flux spectrum information.

The work discussed in this report is a generalization of the self-shielding problem as it applies to its use in the measurement of flux spectra. The theory that is developed applies to the entire neutron energy range from thermal to fission. Within the limitations of the theory, the self-shielding can be calculated for any energy.

II. THEORY

1. Assumptions and Limitations

The theory is based on the following assumptions and is subject to the consequent limitations.

(1) Only first flight encounters are considered. The theory is thus limited to thicknesses less than those in which second encounters can be expected to be significant.

(2) The flux is assumed to be isotropic and scattering events are also assumed to be isotropic. Thus the isotropy of the flux is not destroyed by scattering.

(3) Inelastic scattering is assumed to be negligible. This assumption limits the applicability to relatively heavy elements and to those with no strong inelastic scattering resonances.

(4) Infinite slab geometry is assumed. This assumption limits the theory to foils that are thin relative to their other dimensions.

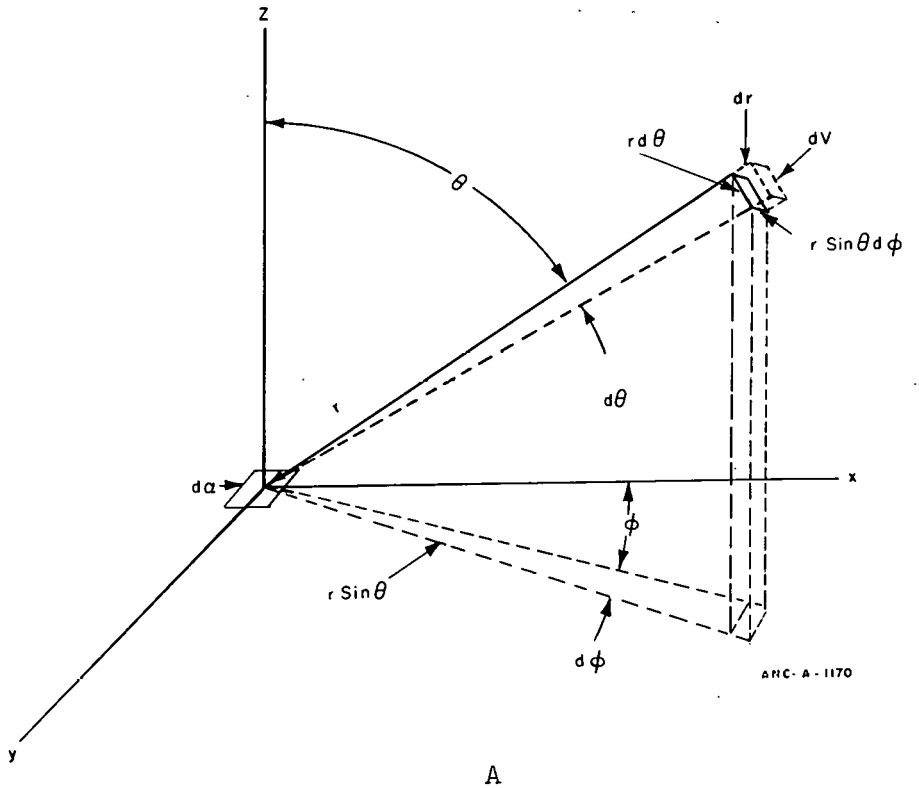
2. Derivations of Equations

The geometry of the problem is shown in Figure 1a. Consider the neutron density, n, in an element of volume, dV and calculate the number at the element of area, da in the surface of the slab, arriving from the direction specified by the angles θ and φ. The number of neutrons in the volume dV that cross the element of area da is $ndV\cos\theta da/4\pi r^2$. But $dV = r^2\sin\theta d\theta d\varphi dr$. By substituting dV and integrating over the half-sphere, one gets the neutron current crossing da from one side in time dt.

$$J = \frac{nda}{4\pi} \int_0^{vdt} dr \int_0^{2\pi} d\theta \int_0^{\pi/2} \sin\theta\cos\theta d\theta = \frac{nv}{4} da, \qquad (1)$$

where r = vdt is the distance all neutrons having the speed, v, travel in the time dt.

These neutrons enter the foil and some are absorbed. The number arriving at da' from the plane source at the surface of the foil is calculated in a similar manner. The geometry is shown in Figure 1b. Of the neutron current given by Eq. 1, those moving in the direction Θ are given by the solid angle fraction as before, namely, $da'\cos\theta/2\pi(z/\cos\theta)^2$. Those absorbed in a slab of thickness dz are $\Sigma_a dz/\cos\theta$, and the absorption from the surface to z is $\exp(-\Sigma_a z/\cos\theta)$.



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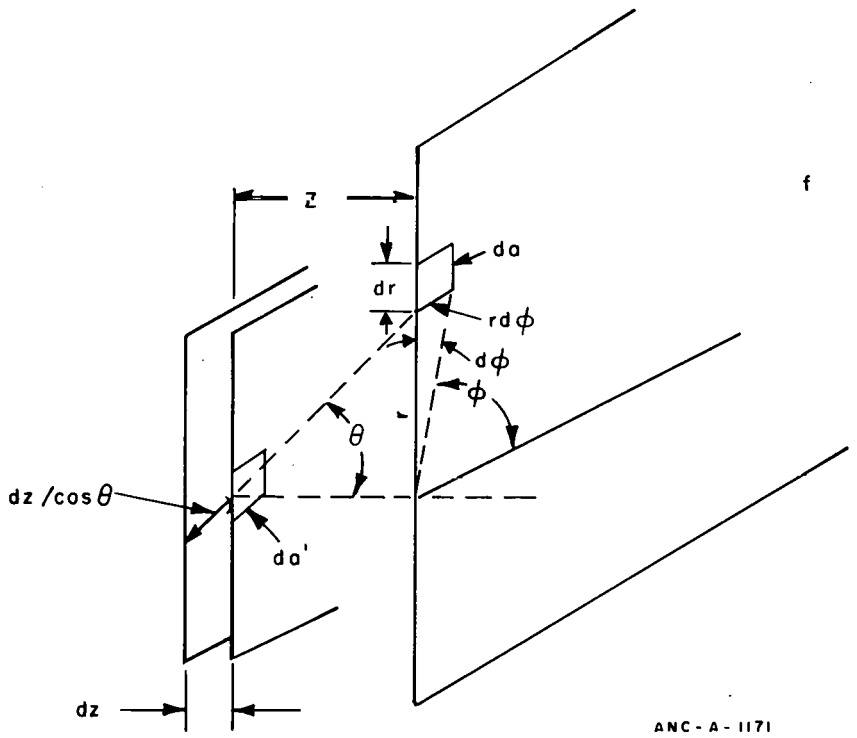

B

Figure 1. Infinite slab geometry for foils in an isotropic flux.

Thus the neutron absorption rate in the slab dz is

$$\frac{dA}{dt} = \frac{nv}{4} \Sigma_a \exp(-\Sigma_a t/\cos\theta) da da' dz / 2\pi (z/\cos\theta)^2 .$$

But $da = r d\varphi\, dr$, $r = z \sin\theta/\cos\theta$, $dr = z\, d\theta/\cos^2\theta$.
Hence, the absorption in dz is

$$\left(\frac{dA}{dt}\right)_{dz d\theta d\varphi\, da'} = \frac{nv\Sigma a}{8\pi} da' (\sin\theta/\cos\theta) \exp(-\Sigma_a z/\cos\theta) dz d\theta d\varphi. \quad (2)$$

Integration of Eq. 2 over the right half-sphere and the thickness $z_2 - z_1$ may be carried out thus

$$\left(\frac{dA}{dt}\right)_{da'} = nv\Sigma_a da'/8\pi \int_0^{2\pi} d\varphi \int_0^{\pi/2} \sin\theta/\cos\theta \int_{z_1}^{z_2} \exp(-\Sigma_a z/\cos\ ) dz d\theta$$

$$= \frac{nv da'}{4} \left[\int_0^{\pi/2} \sin\theta \exp(-\Sigma_a z_1/\cos\theta) d\theta - \int_0^{\pi/2} \sin\theta \exp(-\Sigma_a z_2/\cos\theta) d\theta \right] .$$

By making the substitution $u = 1/\cos\theta$, the two integrals are converted to a form that is recognizable as an exponential integral of the second order. Hence

$$\left(\frac{dA}{dt}\right)_{da'} = nv\, da'/4 \left[E_2(\Sigma_a z_1) - E_2(\Sigma_a z_2) \right] . \quad (3)$$

It will be remembered that the neutron absorption rate given by Eq. 3 was calculated for neutrons arriving from the right half-sphere. The total absorption rate due to an isotropic flux will then be the sum of absorption of neutrons arriving from both right and left half-spheres.

Thus

$$\left(dA/dt\right)_{tot.} = nv\ da'/4 \left\{E_2(\Sigma_a z_1) - E_2(\Sigma_a z_2) + E_2(\Sigma a[b-z_2]) - E_2(\Sigma a[b-z_1])\right\}, \quad (4)$$

where b = the total thickness of the stack of foils.

As written, Eq. 4 refers to an element of area da'. Eq. 4 may be summed over the entire area of a foil of thickness $\Delta z = z_2 - z_1$ and expressed in terms of the weight, m of the foil.

$$m = \rho A_r \Delta z \quad (5)$$

where ρ = density, and A_r is the area of the foil. By substitution, Eq. 4 becomes

$$dA/dt = \left[m(nv)/4\rho\Delta z\right]\left\{z_2 - z_1\right\}, \quad (6)$$

where $\left\{z_2 - z_1\right\}$ has been substituted as an abbreviation for the bracketed term in Eq. 4.

But dA/dt is simply the production term, $N_1\sigma_1(nv)$ in the equation

$$dN/dt = N_1\sigma_1(nv) - \lambda_2 N_2 . \quad (7)$$

Also remember that $N_1 = mN_o/Awt$, where m = weight of foil

N_o = Avogadro's number

Awt = atomic weight of the isotope in question.

Then let $\hat{\sigma} = A_{wt}\left\{z_2 - z_1\right\}\Big/4N_o\rho\Delta z$.

Eq. 7 may therefore be written

$$dN_2/dt = N_1\hat{\sigma}(nv) - \lambda_2 N_2,$$

which has the well known solution

$$N_2 = \frac{N_{1o}\hat{\sigma}(nv)}{\lambda - \hat{\sigma}(nv)}\left[\exp\left[-\hat{\sigma}(nv)t\right] - \exp\left(-\lambda_2 t\right)\right].$$

$\hat{\sigma}$ is of the order of 10^{-23}

nv is of the order of 10^9

t is of the order of 10^4,

hence, $\hat{\sigma}(nv)\,t$ is of the order of 10^{-10},

and $\exp\left[-\hat{\sigma}(nv)t\right] \simeq 1.0$.

For gold $\lambda_2 = 2.967 \times 10^{-6}$ sec^{-1}.

Therefore,

$$N_2 \approx \frac{N_{1o}\hat{\sigma}(nv)}{\lambda_2}\left[1 - \exp(-\lambda_2 t)\right].$$

But, since λ_2 is also small, the exponential can be approximated by the first two terms in its expansion.

Therefore,

$$N_2 \approx N_{1o}\hat{\sigma}(nv)t,$$

and the activity at the end of the irradiation time, t_e, is

$$N_2\lambda_2 = N_{1o}\lambda_2\hat{\sigma}(nv)t_e,$$

which reduces to

$$N_2\lambda_2 = \frac{m_1 e \lambda t_e}{4\rho\Delta z}\quad (nv)\quad \left\{z_2 - z_1\right\}. \tag{8}$$

Eq. 8 expresses the activity at the end of an irradiation of t_e seconds duration for a neutron density, n of neutrons having a speed, v. We now define the integrated flux in a interval $v_2 - v_1$, thus

$$(nv)_i = \phi_i \equiv \left[\int_{v_1}^{v_2} (nv)dv\right]_i = \left[\phi_o \int_{v_1}^{v_2} \varphi(v)dv\right]_i = \phi_o \varphi_i$$

where φ_i is defined by

$$\sum_i \varphi_i \equiv 1.$$

The index, i, refers to the i-th interval $(v_2 - v_1)_i$. Although the derivation just given has been in terms of the variable, v, the actual calculations were made, for practical reasons, by use of φ_i in terms of lethargy units. The conversion is well known and need not be repeated.

After making the above substitutions and summing over all groups the calculated activity of an irradiated foil as given by Eq. 4 becomes:

$$D_c = \frac{m\lambda te}{4\rho\Delta z}\ \phi_o \sum_i \varphi_i \left\{z_2 - z_1\right\}_i \tag{9}$$

where D_c refers to a calculated disintegration rate, which in the following will be equated to a measured disintegration rate D_m to find a value for ϕ_o.

3. Calculational Procedure

The code used to calculate the exponential integrals used a series expansion[1] as follows:

$$E_n(x) = (-x)^{n-1}/(n-1)!\left[\psi(n)-\ln x\right] - \sum_{\substack{m=0 \\ m\neq n-1}}^{m=\infty} (-x)^m/m!\,(m-n+1)$$

$$\psi(1) = -\gamma,\quad \psi(n) = -\gamma + \sum_{m=1}^{m=n-1} 1/m$$

$$\gamma = 0.577215665.$$

For large values of the argument, the series expansion becomes unsatisfactory since the value of the function approaches the convergence criterion. In such cases, a continued fraction[4] expansion gives values to several significant figures with relatively few terms. The expression used is

$$E_n(x) = \exp(-x)\left[\frac{1}{x+}\,\frac{n}{1+}\,\frac{1}{x+}\,\frac{n+1}{1+}\,\frac{2}{x+}\,\frac{n+2}{1+}\,\frac{3}{x+}\,\cdots\cdots\frac{n+c}{1+}\,\frac{c}{x'}\right]. \qquad (10)$$

It was found that thirty terms were sufficient to give better than nine-place accuracy for arguments as small as one. For arguments larger than one, fewer terms are required. For example, ten terms are sufficient to give better than nine-place accuracy for arguments of fifteen and larger. That is, the exponential integral is calculated to nine significant figures though the absolute value is of the order of 10^{-11} for an argument of twenty. Hence c was set to thirty.

x' represents all subsequent terms of the form

$$x' = x + x''\,(n + c)/(x'' + c + 1).$$

To get the program started, x'' is taken equal to the argument, x. x' is then calculated from the recursion relation above, and c is decremented by unity until c = 0. The result is the bracketed term in Eq. 10.

A Digital Equipment Corporation PDP-15 was programmed by means of

the FOCAL system to calculate D_c by Eq. 9. The input consists of the group averaged cross sections for the absorber in question, the measured activities, a first-guess flux spectrum, foil thickness and number, irradiation exposure time, other absorber parameters such as density and atomic weight, and the decay constant of the resulting radioactive isotope. The output consists of the calculated activities of half of the foils in the stack (ideally the activities are symetrical about the center), the integrated flux, ϕ_o in each foil, the average integrated flux and its standard deviation, and the measured to calculated activity ratios for each of the foils. The normalized flux spectrum used to make these calculations may be printed out if desired.

To get the integrated flux, ϕ_o from Eq. 9, the calculated value of all terms on the right except ϕ_o is equated to the measured activity of the foil in question. A value for ϕ_o is thus obtained for each foil. These ϕ_o's are averaged and the average is used to calculate a D_c for each of the foils. If a good choice of φ_i's has been made and if there are no errors in any of the other parameters then all the ϕ_o's should be equal to one another. Errors in the parameters and bad choices of the φ_i's show up in unequal ϕ_o's and large differences between D_m and D_c. The initial guess is then modified in any manner that seems appropriate to cause the ϕ_o's to be equal and the D_m and D_c's to be equal.

In this work an evaluated composite flux derived from other sources[5] was used for energies above 10 keV. The flux below this energy was then adjusted until an acceptable match was obtained.

III. RESULTS AND DISCUSSION

Table I contains the experimental data and Table II lists the gold cross-sections that were used. Sample numbers 1, 2, 3, 10, and 13 consisted of stacks of 20 each of 5 mil gold foils, 3/8 inch in diameter. These were irradiated under various conditions in the CFRMF as described below.

TABLE I

EXPERIMENTAL DATA -- 20 FOIL STACK

	Sample Numbers				
No.	1	2	3	10	13
	(cts/s/mg)	(cts/s/mg)	(cts/s/mg)	(cts/s/mg)	(cts/s/mg)
1	3.08	3.00	3.08	3.15	3.19
2	2.81	2.82	2.83	2.93	2.94
3	2.79	2.70	2.72	2.77	2.82
4	2.72	2.65	2.68	2.75	2.75
5	2.71	2.55	2.64	2.75	2.70
6	2.66	2.61	2.61	2.71	2.68
7	2.68	2.53	2.60	2.65	2.65
8	2.62	2.53	2.59	2.68	2.65
9	2.61	2.55	2.60	2.66	2.63
10	2.61	2.55	2.61	2.61	2.61
11	2.61	2.57	2.57	2.66	2.65
12	2.61	2.58	2.57	2.63	2.58
13	2.64	2.56	2.63	2.66	2.60
14	2.60	2.60	2.60	2.66	2.65
15	2.65	2.60	2.61	2.70	2.66
16	2.70	2.65	2.65	2.73	2.68
17	2.72	2.68	2.68	2.74	2.69
18	2.79	2.72	2.76	2.79	2.73
19	2.85	2.80	2.80	2.93	2.84
20	3.06	3.00	3.05	3.11	3.08

TABLE II

GROUP AVERAGED CROSS SECTIONS FOR GOLD

Group No.	$\sigma(n,\gamma)$ barns	Group No.	$\sigma(n,\gamma)$ barns
1	2.261 E-2	35	5.020
2	2.726 E-2	36	6.134
3	3.306 E-2	37	9.924
4	4.010 E-2	38	11.52
5	4.917 E-2	39	15.79
6	6.026 E-2	40	14.23
7	7.355 E-2	41	16.89
8	8.607 E-2	42	16.58
9	9.563 E-2	43	14.69
10	0.1026	44	66.36
11	0.1127	45	22.34
12	0.1374	46	7.249
13	0.1740	47	0.9047
14	0.2123	48	16.96
15	0.2535	49	131.6
16	0.2917	50	0.8222
17	0.3140	51	0.3257
18	0.3218	52	0.3388
19	0.3251	53	0.5858
20	0.3287	54	1.067
21	0.3464	55	2.338
22	0.4067	56	6.417
23	0.4998	56	26.24
24	0.6160	58	723.4
25	0.7603	59	5004.00
26	0.9418	60	102.8
27	1.141	61	46.44
28	1.368	62	32.33
29	1.628	63	26.99
30	1.933	64	24.87
31	2.315	65	24.24
32	2.790	66	24.60
33	3.381	67	25.78
34	4.114	68	27.55

Sample density = 19.32 g/cc

Atomic weight = 196.957

Decay constant = 2.9757×10^{-6} sec^{-1}

Group structure is that of the PHROG reactor code, and the group-averaged cross sections are those obtained by use of that code with ENDF/B cross sections.

Sample 1. During the irradiation of this sample, the ends of the experiment hole were closed with boral and U-238 to eliminate any thermal flux that might enter from an axial direction. The foils were oriented perpendicular to the reactor vertical axis.

Sample 2. The irradiation conditions were similar to those for Sample 1, except that a boral and lead plug was used in the ends of the experiment hole.

Sample 3. This stack of foils was irradiated in the CFRMF with the ends of the experiment hole open. In each of these first three irradiations, the stacks were placed in an aluminum holder.

Sample 10. This sample was also irradiated in a horizontal position, but was mounted in a paper holder rather than in an aluminum one.

Sample 13. This sample was oriented parallel to the vertical axis of the reactor during irradiation, and the ends of the experiment hole were closed with a boral and U-238 plug.

Figures 2 through 6 show a comparison between measured activities and those calculated as described above. The average ϕ_o's for each stack of foils are given in Table III. The error limits are standard deviations of twenty values about the averages.

TABLE III

INTEGRATED FLUXES

Sample No.	ϕ_o (nv)
1	$4.28 \pm 0.08 \times 10^9$
2	$4.18 \pm 0.08 \times 10^9$
3	$4.23 \pm 0.07 \times 10^9$
10	$4.33 \pm 0.07 \times 10^9$
13	$4.30 \pm 0.07 \times 10^9$

The average cross section for this flux distribution was found to be 0.252 b. The resulting normalized flux is shown in Figure 7. It will be noted that the flux level drops off sharply below 10 keV.

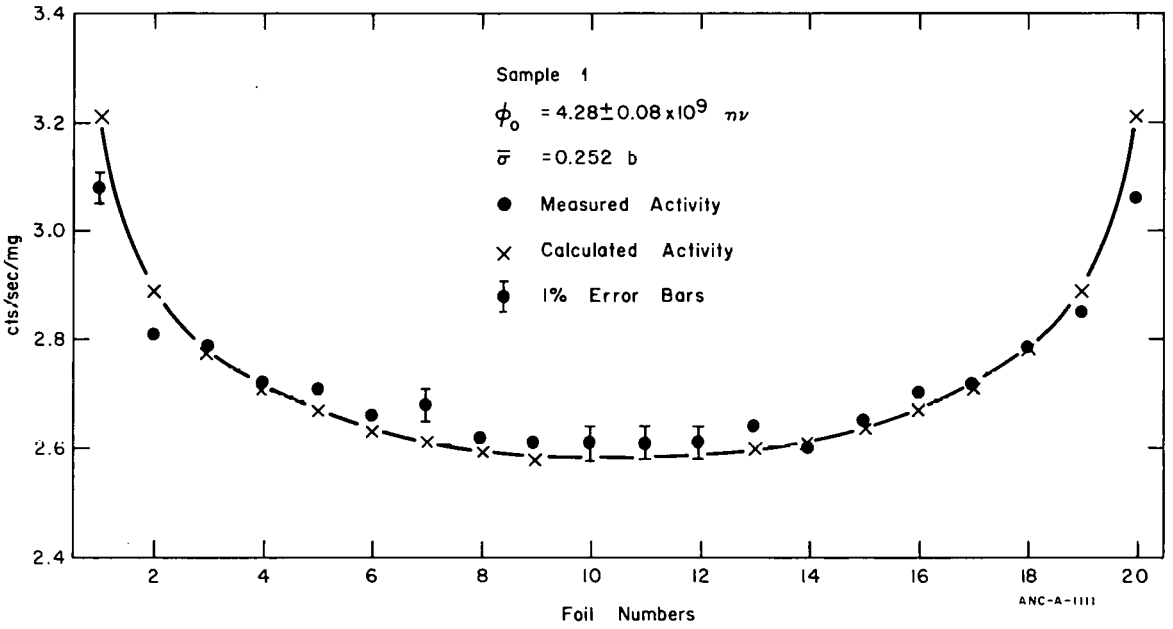

Figure 2. Comparison of calculated with experimental activities for Sample 1.

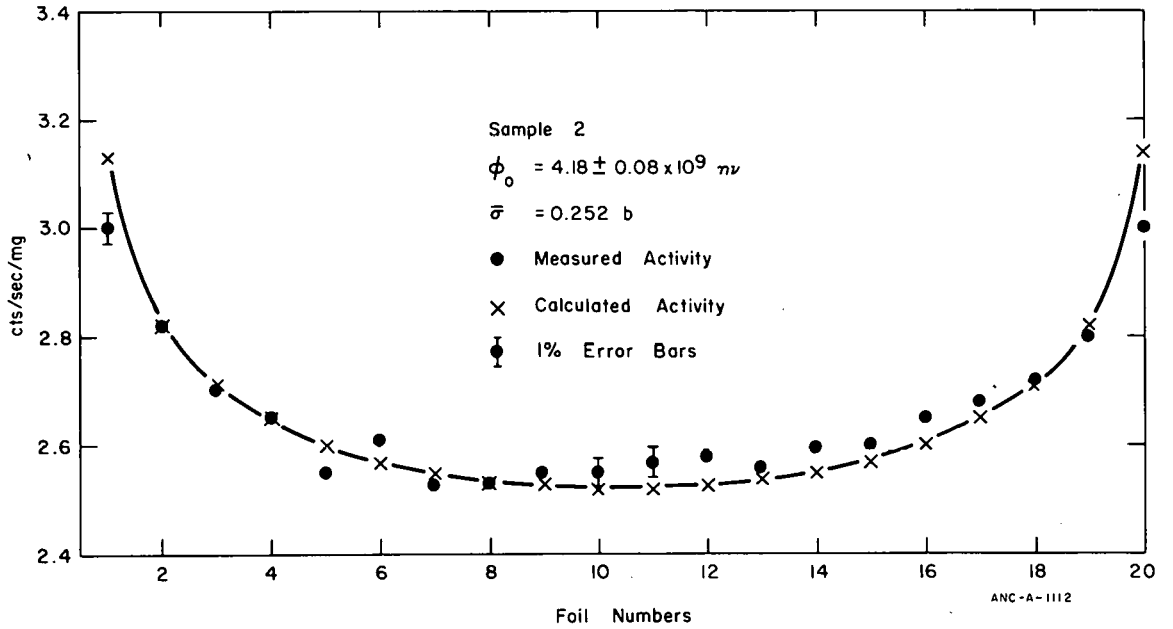

Figure 3. Comparison of calculated with experimental activities for Sample 2.

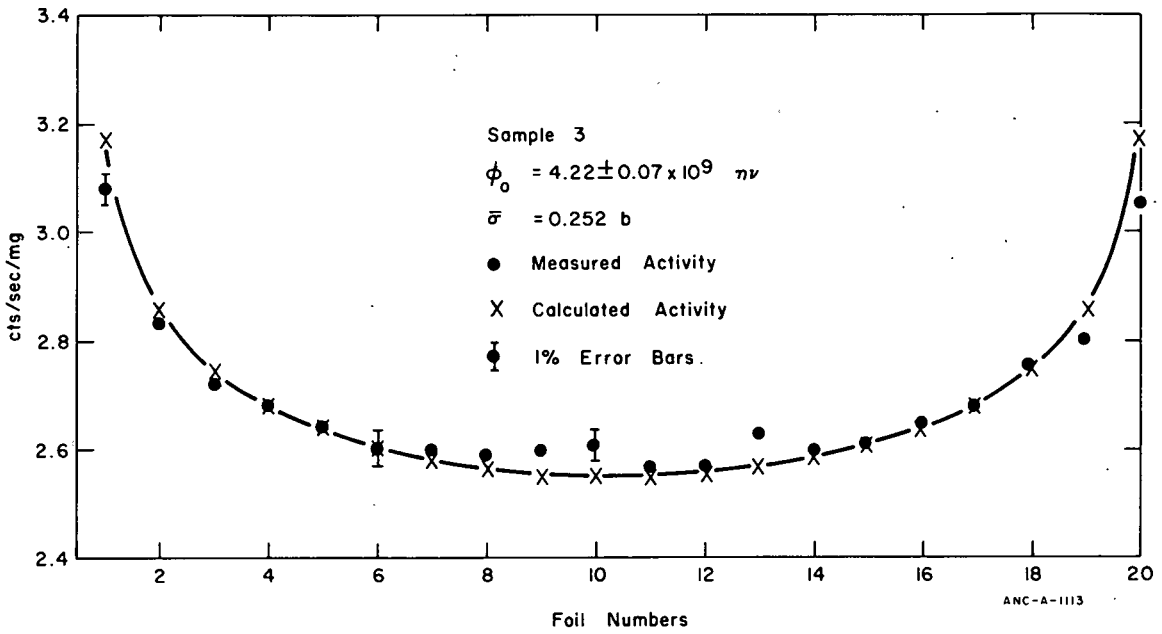

Figure 4. Comparison of calculated with experimental activities for Sample 3.

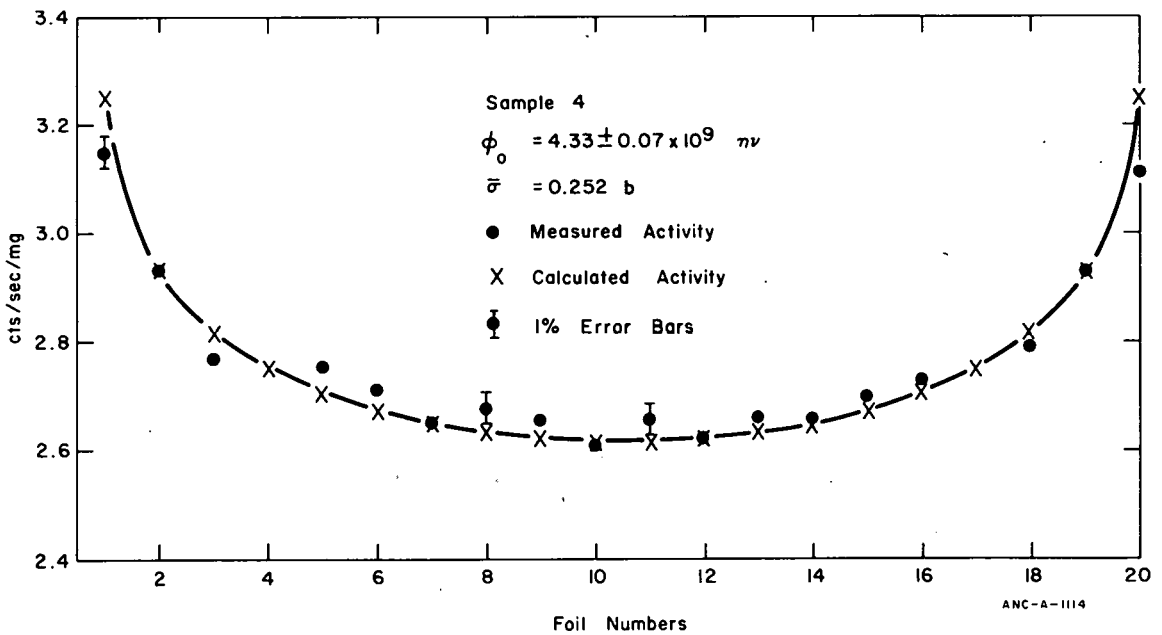

Figure 5. Comparison of calculated with experimental activities for Sample 10.

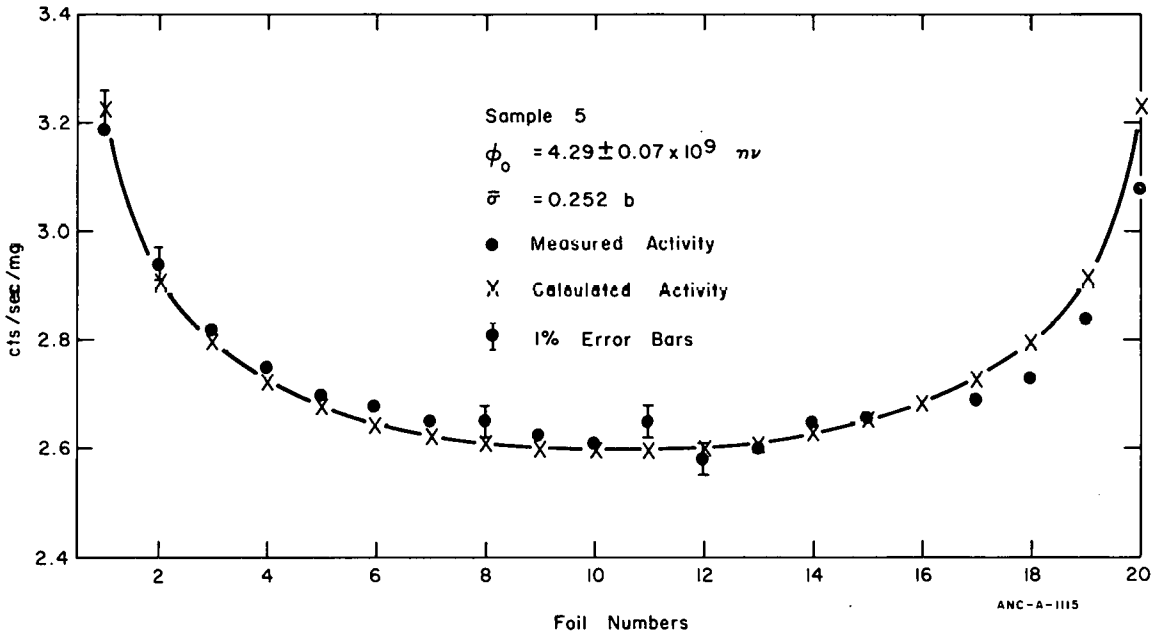

Figure 6. Comparison of calculated with experimental activities for Sample 13.

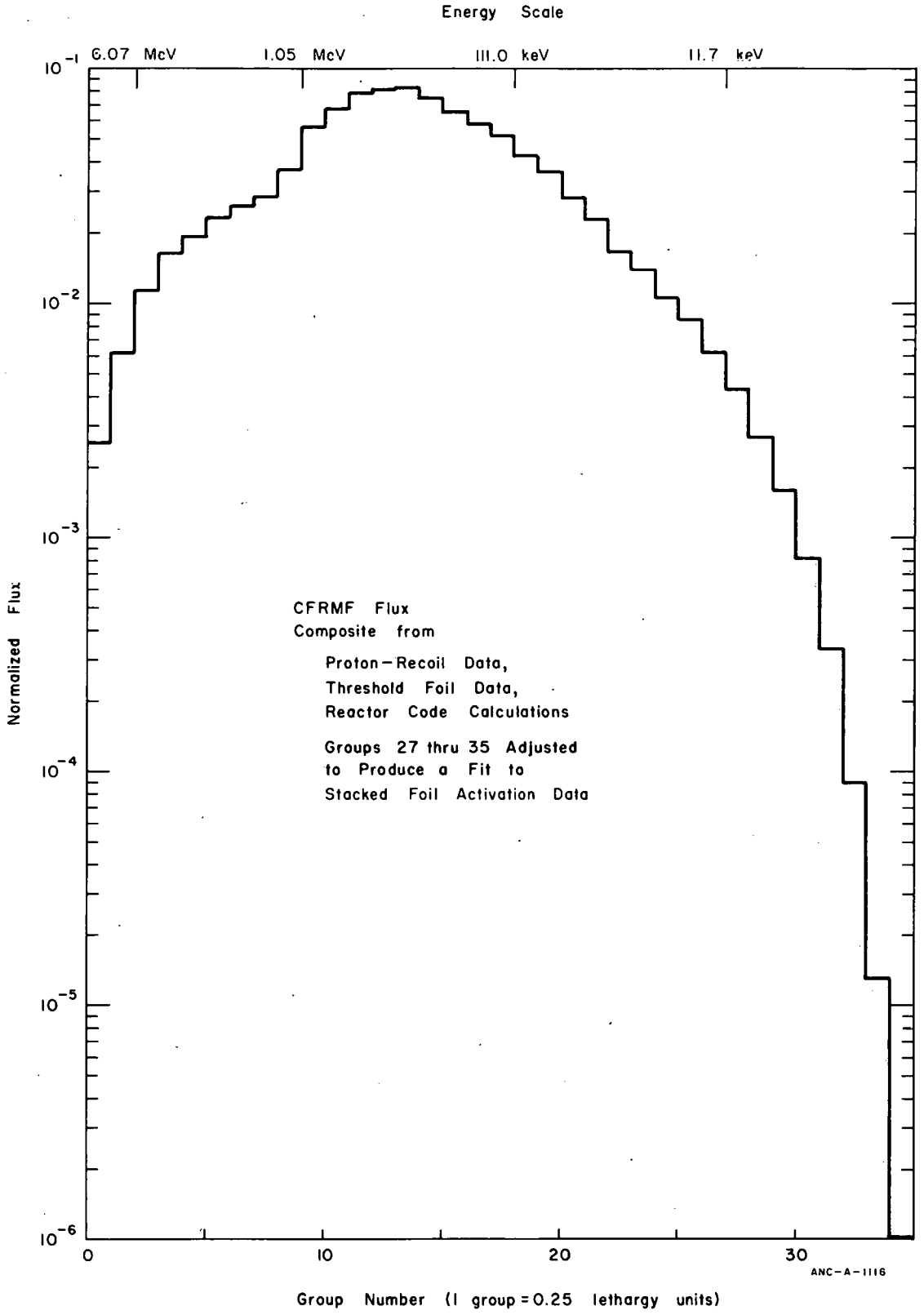

Figure 7. Flux spectrum derived from proton-recoil data, threshold foil data, reactor code calculations, and stacked foil data.

IV. CONCLUSION

The results indicate that self-shielding in a stack of absorber foils can be used to derive flux spectrum information from activation data. The computer program used in this work to calculate the fits to the measured activities did not include a systematic convergence routine, since the PDP-15 has an inadequate memory for this purpose. The bracketed term in Eq. 6 can be used, however, to calculate a set of psuedo-cross sections as for $\hat{\sigma}$. These latter can then be used in a more powerful program such as INSPECT or SAND II to derive a spectrum.

A cursory examination of the measured activities might lead one to conclude, intuitively, that a significant portion of the activity was due to flux in the energy range below one keV. Such a conclusion might be based on the fact that cross sections in this energy range are so much larger than those above this energy. The results indicate that quite the contrary is the case; namely, that significant self-shielding exists at all energies in stacks of two or more foils of thicknesses of 0.005 inch.

That is, a large number of foils of a single, or a few absorbers in an alloy foil, replaces sandwich sets of a limited number of suitable absorbers. For example, if ten suitable absorbers are available, ten data points result. Whereas, in the case of gold, a stack of twenty foils yields ten data points (assuming symmetrical results in the stack) with a single absorber, or twenty points in a binary alloy such as Au - Cu. Thus, the number of data points is not limited by the availability of suitable absorbers.

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