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LUMP CORRECTION AND IDENTIFICATION IN THE COMBINED THERMAL/EPITHERMAL NEUTRON (CTEN) METHOD

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Abstract
We present a model for self shielding in lumps of fissile material in active-neutron assays. The model combines the formula for self-attenuation of gamma-ray in lumpy sources with the multi-group analysis techniques used in neutron transport calculations. Models for thin foils and for spheres are examined in terms of error multiplication in determining lump corrections and the basic accuracy of the model.

INTRODUCTION

A problem we face in developing a lump correction algorithm for combined thermal/epithermal neutron (CTEN) systems is that we are limited in the laboratory to studying self shielding in large metallic sources, in foils, or in diffuse sources such as suspensions of SNM in diatomaceous earth. In large metallic sources, only the outer skin of the source is interrogated and the self shielding is given by the mean free path of the neutrons times the ratio of the source's surface area to volume. Real lumpy waste is more likely to consist of a multitude of microscopic particles that are significantly smaller than the mean free path. Study of self shielding in foils that are thinner than the mean free path gives us a picture of the self shielding in the low-thickness region, but foils are not a direct model of the fine particulates in real waste, which are usually assumed to be nearly spherical. The description of self shielding in diffuse sources is complicated by the moderating and absorbing properties of the suspension medium and by questions of uniformity of mixing.

The approach we have taken to study self shielding experimentally is to develop parallel mathematical models for self shielding in spheres and in foils. We then experimentally demonstrate that our model for foils is accurate in the low-thickness region, and that both models are accurate in the high-thickness region. Because the likely causes of inaccuracy are common to both the foil and sphere models, we assume that the spherical-source model is just as accurate (or inaccurate) as the foil model. Neither the sphere nor foil model can be rearranged to give, for example, the self shielding as a function of the epithermal-to-thermal signal ratio, which is what we require of our lump-correction algorithm. However, we can use our mathematical model for spheres to generate a set of self shielding values for cases that are not accessible in our laboratory measurements. These values can be used to create look-up tables for correction factors - the approach we are currently using - or can be fitted to find a simpler closed-form approximation function.

MATHEMATICAL MODEL FOR SELF SHIELDING

Our model for calculating the self shielding in fissile materials is the essentially the same as the model described in 1985 by Cogbill and Swinhoe[1] for estimating self shielding in DDT assays, and combines features of the differential absorption method[2] used for correcting gamma-ray self-attenuation* in SGS and TGS assays with the multi-group analysis (MGA) used in neutron-transport calculations. If the interrogating neutrons were monoenergetic, we could accurately describe neutron self shielding using the same equations that describe gamma-ray self-attenuation. Since the neutrons are not monoenergetic, we use multi-group analysis to numerically combine the neutron energy spectrum with the neutron absorption cross section.

One difference between our approach and that of Cogbill and Swinhoe is that their expression for self shielding in spheres involves numerical evaluation of a double integral. They were apparently unaware that the same expression, when applied to gamma-ray self-attenuation, had been evaluated to an analytical function by Francois in 1974. This is the expression we use, and is the expression used by Parker, et al., in their 1981 paper[2] on the differential absorption technique for lump corrections in SGS. The energy groups used in our multi-group analysis are different from those of Cogbill and Swinhoe; we used 25 energy groups to their 30, but our groups sample the low-energy resonances in ^{235}U and ^{239}Pu more finely. We use cross sections from published tables for some cases, but to achieve the best accuracy we have used our own group-averaged cross sections extracted from the most recent version of the MCNP neutron transport code. We are skeptical of the unproved assertion in ref. 1 that any expression for self shielding in ^{239}Pu that does not account for down-scattering of neutrons to lower energies will necessarily be inaccurate. We will attempt to show that downscattering is not a significant effect either in thin foils or in small spheres of ^{239}Pu. In any case, downscattering and upscattering cross sections are available in tables and with some extra effort could be included in the multi-group analysis.

SELF-SHIELDING FORMULAE

The formula for estimating self-attenuation of monoenergetic gamma rays in spheres is

$$(1) \qquad f_{\text{sphere}}(D,\mu) = 3/(2D\mu) - 3/(D\mu)^3 + \exp(-D\mu)\cdot\{3/(D\mu)^2 + 3/(D\mu)^3\} \ ,$$

where D is the sphere diameter and μ is the linear attenuation coefficient for the gamma rays. The attenuation of monoenergetic neutrons interrogating a lump of fissile material is also described by (1). In that case the formula is describing the attenuation of the interrogating neutrons entering the spherical source, rather than that of gamma rays leaving the source along the same trajectories. The relationship between the neutron absorption cross section $\rho_{Abs}(E)$ (in barns) at neutron energy E and its corresponding attenuation coefficient $\mu(E)$ (in 1/cm) is

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* For gamma rays we use the term "self-attenuation" while for interrogating neutrons we use the term "self-shielding". Both terms are describing essentially the same phenomenon.

(2) $\mu(E)\ (1/\text{cm}) = 10^{-24} \cdot N_0 \cdot \rho \cdot \sigma_{Abs}(E)(\text{barns})/A$,

where

A = atomic weight (g/mol)
N_0 = Avogadro's number = $6.022 \cdot 10^{23}$
ρ = source density in g/cm^3 .

For fissile radionuclides, the total absorption cross section can be replaced by the sum of the (n,fission) and (n,gamma) neutron-capture cross sections.

The fractional self shielding in foils, f_{foil}, is given by the integral

(3) $f_{\text{foil}}(D,\mu) = \int_0^{\pi/2} 2\sin(\theta)\cos(\theta)\exp\{-D\mu/\cos(\theta)\}\,d\theta$

where D is now the foil's thickness. We used the MathCad 6.0 computer software to numerically evaluate eqn. (3) for the results presented here. Notice that because of spherical symmetry, eqn. (1) is valid even with an anisotropic interrogating flux. Equation (3), however, is valid only for an isotropic interrogating flux. Another difference is that with spheres we can associate each self-shielding value with a unique particle mass. We cannot do this with foils, since by assumption they are infinite in areal extent.

A multi-group analysis is used to combine equations (1) and (3) with the time-dependent neutron energy spectra seen in CTEN assays. For a given matrix type and position within the drum, we perform Monte Carlo simulations with the program MCNP to tabulate the neutron flux $\phi(e,t)$ in each of N_e energy bins and N_t time bins[3]. Simulating just the neutron energy spectrum instead of the entire self-shielding problem makes the calculation feasible. These spectrum simulations are relatively fast, even with the detailed, time-dependent, as-built CTEN model that we are using. In contrast, an attempt to fully simulate self shielding in microscopically small particles using the detailed CTEN model would be too time-consuming to be generally useful.

The same energy bins used for calculating the flux are used to define the lethargy-weighted group-average attenuation coefficients for the SNM radionuclide of interest, i.e.,

$$\mu(e) = \Sigma_i\ \mu(E_i, E_{i+1}) \cdot \{\ln(E_{i+1}) - \ln(E_i)\} / \Sigma_i\ \{\ln(E_{i+1}) - \ln(E_i)\} \quad ,$$

where the sum is over discrete attenuation coefficients $\mu(E_i, E_{i+1})$ within the e'th energy group. The group $\mu(e)$ values are used in equation (1) to compute the e'th self-shielding fraction for a sphere of diameter D, $f_{\text{sphere}}(D,\mu(e))$, or in equation (3) to compute the e'th self-shielding fraction for a foil of thickness D, $f_{foil}(D,\mu(e))$. The weighted sum over the individual group self-shielding fractions $f_X(D,\mu(e))$ (X=sphere or foil) multiplied by the fluxes $\phi(e,t)$ gives the overall self-shielding fractions $F_X(D,t)$ for the t'th time bin and for the matrix type and position for which the $\phi(e,t)$ were calculated. That is,

$$(4) \qquad F_X(D,t) = \Sigma_e f_X(D, \mu(e)) \cdot \phi(e,t) \cdot w(e) \,/\, \Sigma_e \ \phi(e,t) \cdot w(e) \ .$$

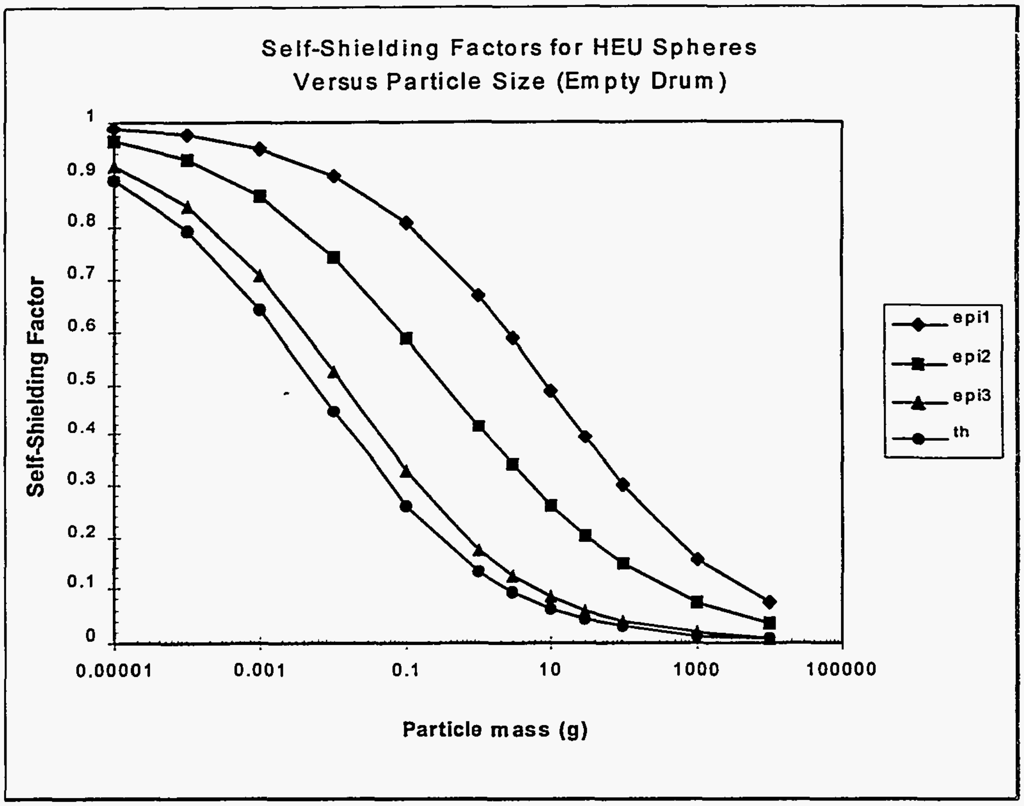

Figure 1. Calculated self-shielding factors for HEU spheres (93.15% ^{235}U) in an empty drum for the CTEN time windows. The three epithermal interrogation time windows are: epi1 = 60-100 μs, epi2 = 100-240 μs, epi3 = 240-800 μs. The epi2 window is currently used in obtaining CTEN lump corrections. The thermal time window (th) is from 800-2800 μs.

The additional weights $w(e)$ are needed to convert the actual self shielding to the observed self shielding, which is complicated by the variation with energy of the neutron multiplicity and the ratio of the fission-to-absorption cross section. The weights are given by

$$(5) \qquad w(e) = \nu(e) \cdot \mu(e,fission) \,/\, \mu(e,absorption) \quad .$$

The neutron multiplicity, ν(e), is constant for an interrogating neutron energy below 1 MeV and so can be safely omitted for descriptions of our prototype CTEN system, since the neutron flux is essentially zero above 1 MeV by the time the detectors have recovered from the zetatron burst.

Figure 1 shows computed values of the self shielding ($F_{sphere}(D,t)$) in enriched U as a function of sphere size for the time bins currently used in active CTEN assays.

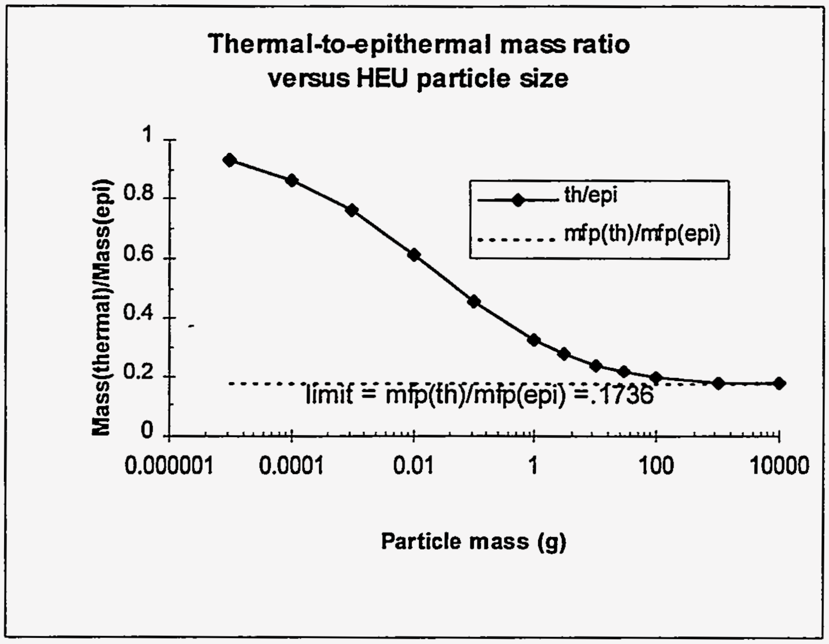

Figure 2. Computed thermal-to-epithermal apparent mass ratio as a function of the individual HEU particle size (93.15% ^{235}U), based on MCNP fluxes computed for the center of an empty drum. This ratio, which is equal to the ratio of the thermal-to-epithermal self-shielding factors, asymptotes to the ratio of the thermal-to-epithermal (neutron absorption) mean free paths (the dashed line).

LIMITING THERMAL/EPITHERMAL RATIOS

The mean-free-path estimate for self shielding of monoenergetic neutrons ($f_{mfp,X}$), which is accurate only when the sphere diameter or foil thickness is large compared to the mean free path of the neutrons, is given by

(6) $f_{mfp,sphere}(D,\mu) = 3 / (2D\mu) = 3\lambda / (2D)$ (for spheres)

or

(7) $f_{mfp,foil}(D,\mu) = 2 / (D\mu) = 2\lambda / D$ (for foils) ,

where λ is the mean free path of the neutrons and D and μ have the same meanings as in equations (1) and (3). We compute the overall mean-free-path self-attenuation fraction, $F_{mfp}(D,t)$, in the same way as for the more accurate estimate in eqn. (4), i.e.,

$$F_{mfp,X}(D,t) = \Sigma_e f_{mfp,X}(D,\mu(e)) \cdot \phi(e,t) \cdot w(e) / \Sigma_e \ \phi(e,t) \cdot w(e) \ ,$$

which simplifies to

(8) $F_{mfp,X}(D,t) = k_X \langle\lambda\rangle / D$,

$$\langle\lambda\rangle = \Sigma_e \lambda(e) \cdot \phi(e,t) \cdot w(e) / \Sigma_e \ \phi(e,t) \cdot w(e) \ ,$$

$$k_{sphere} = 3/2 \text{ and } k_{foil} = 2 \ .$$

When the weights $w(e)$ are given by eqn. (5), $\langle\lambda\rangle$ is the apparent average mean free path; with all the $w(e)$ set to one, $\langle\lambda\rangle$ becomes the true average mean free path. Note that this average is only meaningful at large particle sizes and cannot be used to circumvent the summation over energy groups in eqn. (4) for the true self-shielding estimate.

Currently, our lump correction for the prototype CTEN system is based on two time windows: an epithermal window from 100 μs to 240 μs (called epi2 in CTEN nomenclature) and a thermal window from 800 μs to 2800 μs. Assuming that all other factors (matrix corrections, efficiency, etc.) have been corrected for, the ratio of the apparent masses for the epithermal and thermal time windows is given by the ratio of their self-shielding factors, i.e.,

(9) $ratio(D) = mass(D, t_{th})/mass(D, t_{epi}) = F_X(D, t_{th}) / F_X(D, t_{epi})$.

This ratio starts at 1.0 in the zero-mass limit ($D = 0$) and assymptotes at large particle sizes to the ratio of the mean-free-path estimates, i.e.,

(10) $\lim(D \to 0)\ F_X(D, t_{th}) / F_X(D, t_{epi}) = 1$,

(11) $\lim(D \to \infty)\ F_X(D, t_{th}) / F_X(D, t_{epi}) = F_{mfp,X}(D, t_{th}) / F_{mfp,X}(D, t_{epi}) = \langle\lambda_{th}\rangle / \langle\lambda_{epi}\rangle$.

Figure 2 shows the the thermal-to-epithermal ratio as a funtion of particle size calculated for enriched U (93.15% ^{235}U) spheres in an empty drum, illustrating these limits. Figure 3 shows the spherical-lump correction factor $C_{sphere}(D,t_{th}) = 1/F_{sphere}(D,t_{th})$ for the thermal time window as a function of the thermal-to-epithermal mass ratio, computed for an empty drum and for a drum matrices with hydrogen densities of .0055 and .0118 g/cm^3. These correspond to 4.95% and 10.6% water by volume in an otherwise benign matrix.

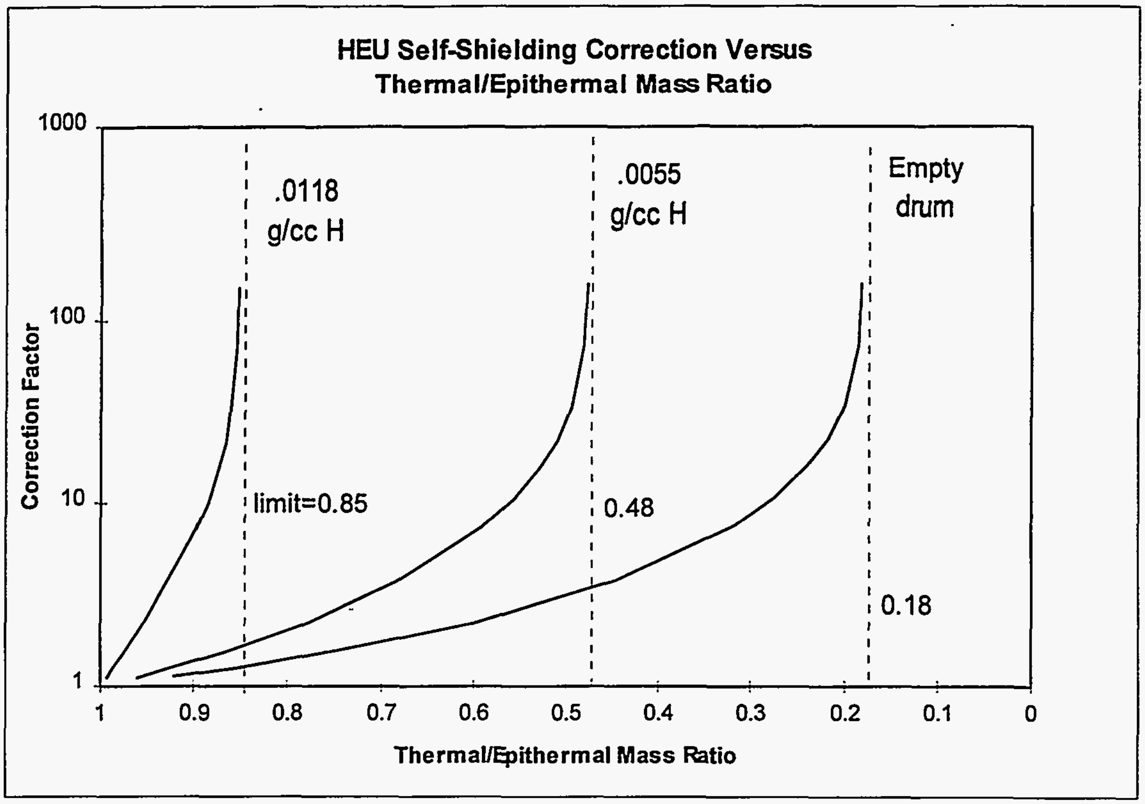

Figure 3. Calculated HEU (93.15% ^{235}U) self-shielding corrections (for the thermal time window) as a function of the thermal/epithermal mass ratio, for an empty drum and for two hydrogen densities. The hydrogen density of .0055 g/cc corresponds to a 4.95% water content (by volume), the 0.0118 g/cc case to 10.6% water. The calculations are for the drum center, where the degree of moderation is the highest. The corrections assymptote to a limiting *ratio* given by $\langle\lambda_{th}\rangle/\langle\lambda_{epi}\rangle$, the ratio of the group-averaged absorption mean free paths.

We can see from figs. 2 and 3 that estimates of the thermal lump correction factor C_X based on noisy measurements of the thermal/epithermal mass ratio (*ratio*) will become unstable as the ratio approaches the $\langle\lambda_{th}\rangle/\langle\lambda_{epi}\rangle$ lower limit. We can quantify this instability as an amplification $\alpha = (dC_X/d\,ratio)$ in the error or noise level when estimating the corrections. That is,

$$(12) \qquad \sigma_{Cx} = \alpha \cdot \sigma_{ratio} = \left| dC_X/d\,ratio \right| \cdot \sigma_{ratio} \approx \left| \Delta C_X / \Delta ratio \right| \cdot \sigma_{ratio} \; ,$$

where σ_{ratio} is the error in the measured *ratio* and σ_{Cx} is the corresponding error in the estimate of C_X (based on the measured *ratio*). Division of α by C_X gives the relative error amplification in the correction. If there were no other source of error, the final estimated mass would have same relative error as C_X. To compute α, an estimate of $\Delta C_X(ratio)$ can be made from a pair of neighboring computed C_X values with separation of $\Delta ratio$, i.e.,

$$(13) \quad \Delta C_X = C_X(ratio + \Delta ratio) - C_X(ratio) \; .$$

The value of α becomes infinite as *ratio* approaches $<\lambda_{th}>/<\lambda_{epi}>$ or as $<\lambda_{th}>/<\lambda_{epi}>$ approaches 1.

Figure (4) shows the relative error amplification α/C_X as a function of *ratio* for the (X=sphere) empty-drum HEU thermal-to-epithermal mass ratios in figure (3).

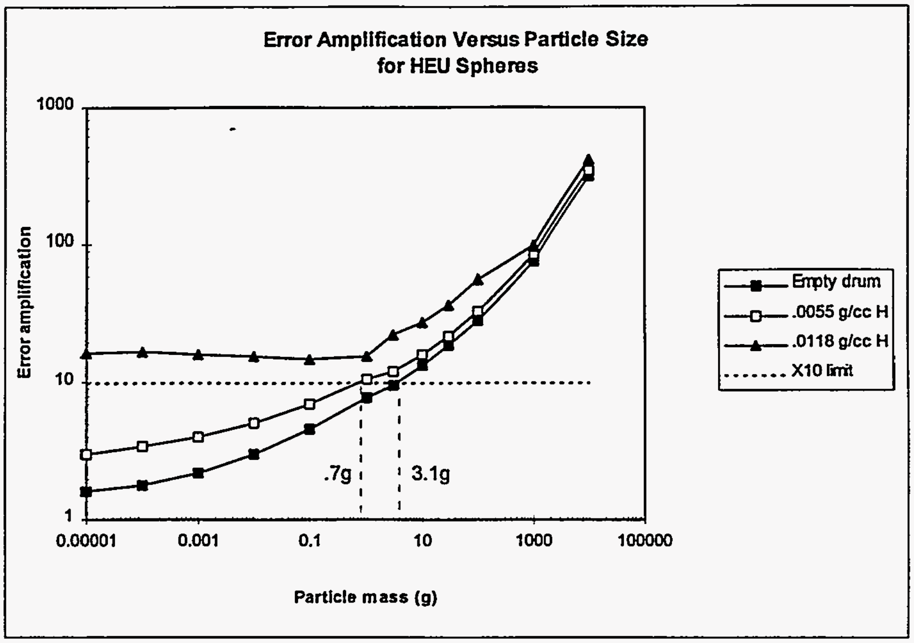

Figure 4. Relative error amplification factor (α) for estimation of the self-shielding correction from the thermal-to-epithermal mass ratio, calculated for enriched U (93.15% ^{235}U) spheres in an empty drum. A relative error amplification of 10 means that with a 3% error in the thermal/epithermal ratio, there will be a 30% relative error in the estimated correction factor. For a maximum of 10X amplification in the relative error, corrections can be obtained in an empty drum up to particle sizes of about 3.1g, and in the .0055 g/cc hydrogen drum up to about .7 g.

TEST OF THE MODEL

To test the model for self-attenuation in foils, we compared calculated and measured self-shielding factors for a series of stacked HEU (93.15% ^{235}U) foils with nominal masses of 7 g each and uniform thicknesses of 5 mil. Using stacks of 1, 2, 3, and 4 foils gave thicknesses of 3, 6, 9, and 12 mils, respectively. These thicknesses are in the region where mean-free-path estimates of the self shielding are invalid. Our results are shown in figure five, which compares calculations from the foil model with measurements in two epithermal time windows and the thermal time window of our CTEN instrument. The agreement, as can be seen, is excellent.

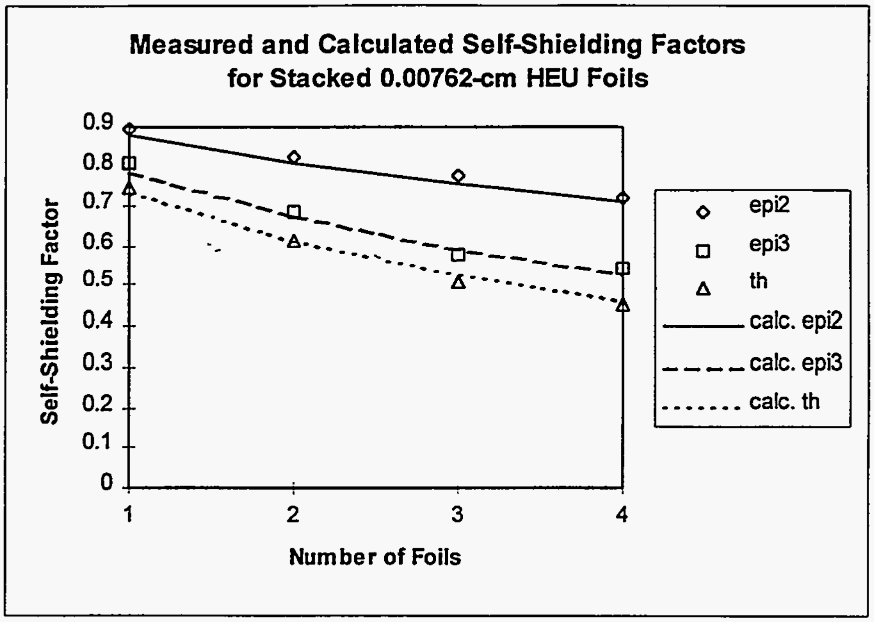

Figure 5. Measured and calculated self-shielding factors for stacked 3-mil HEU (93.15% ^{235}U) foils.

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