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Self Shielding in Cylindrical Fissile Sources in the *APNea*[a] System

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Abstract

In order for a source of fissile material to be useful as a calibration instrument, it is necessary to know not only how much fissile material is in the source but also what the effective fissile content is. Because uranium and plutonium absorb thermal neutrons so efficiently, material in the center of a sample is shielded from the external thermal flux by the surface layers of the material. Differential dieaway measurements in the *APNea* System of 5 different sets of cylindrical fissile sources show the various self shielding effects that are routinely encountered. A method for calculating the self shielding effect is presented and its predictions are compared with the experimental results.

Thermal neutron flux will be attenuated as it traverses a matrix because it is absorbed by the matrix; alternamtely, the flux can escape from the matrix. Both of these effects lead to a diffusion dieaway time of the thermal neutron flux. The *APNea* cavity is large enough that the diffusion time for thermal flux out of the cavity is of the order of 800μs. When a matrix material (in a drum) is introduced, the dieaway time of flux in the drum matrix drops to 600μs for ethafoam, to 400μs for concrete, and to as little as 100μs for raschig rings.

But, there is a special concern for the measurement of fissile materials, such as ^{235}U and ^{239}Pu, which have large thermal-neutron-capture cross sections. The attenuation of the flux as it penetrates clumps of these fissile materials can be so severe that the fissile signal will be significantly reduced over that which would normally be hoped for. The purpose of this paper is to study the effect for a certain class of sources encountered by the *APNea* System and to assess the overall accomodation that must be made in measuring these materials. In particular the focus of this paper will be on fissile sources in the form of right circular cylinders, as all of the calibration sources for the *APNea* System have this form factor (or can be put into this shape). The strengths of the various sources are listed in the first three

[a] *APNea* — It takes your breath away!

tables.

Fissile Sources

Uranium Pins

# Pins	# Pellets	^{235}U(mg)	Factor
2	0	0	1.0
2	1	61	0.917
2	2	123	0.887
2	4	244	0.851
30	6	368	0.832
6	13	797	0.809
Enriched Uranium Pins			
2	3	803	0.634
2	5	1350	0.601
30	8	2150	0.580
20	17	4560	0.562

Table 1:

Fig. 1 shows the results of assaying two different sets of fissile sources in the *APNea* System. These sources were specially made for the *APNea* in order to form the basis of an absolute calibration for the active mode of the *APNea* System. The 98 pins which make up the calibration set are loaded with various numbers of uranium fuel pellets. One set of pins use natural uranium fuel pellets with a ^{235}U fraction of 0.76%; the other set uses fuel pellets with a ^{235}U fraction of 4.45%. The strengths of the sources vary as the number of fuel pellets in each pin assembly. Fig. 1a shows the *APNea* ^{235}U mass values versus the reported mass values for the various stacks of natural uranium fuel pellets. In this figure and in several of the following figures is included the contribution from a one inch cylinder of uranium metal (labeled **dU**) depleted to the 0.2% level. The actual amount of ^{235}U in this slug is somewhat in question, but it is used as the (relative) reference point for all of these active measurements of fissile sources. That is, the slug is measured along with whatever reference source so that the results from all reference sources can be correlated in the final analysis.

Fig. 1b is the equivalent plot for fuel pellets enriched in ^{235}U to 4.45%. It is seen in

these figures that the assay values deviate more significantly from the reported mass values for both sets of pins as the mass increases. 800mg of ^{235}U in natural uranium pins looks like 650mg but looks like only 500mg when it is in the 4.45% enriched uranium pins. This difference is essentially the self shielding distortion. Fig. 2a shows results for a set of small cylinders of pure ^{235}U dust. Here the self shielding is greater than 50% as 500mg of ^{235}U is barely 200mg in the assay.

Pure ^{235}U Powder

Pin ID	^{235}U(mg)	Factor
EU6	9.97	0.663
EU1	25.01	0.642
EU3	49.94	0.614
EU4	74.91	0.589
EU2	99.98	0.566
EU5	149.78	0.529
EU8	424.38	0.428
EU7	498.75	0.416

Depleted Uranium Slug

Height(in)	^{235}U(mg)	Factor
1.02	~500	0.794

Table 2:

Fig. 3 features the Quinby sources, a set of weapons grade plutonium cylinders. The roughly 20% deviation of the 2.96g source prompted a special study of the Quinby Sources plutonium distribution. These sources are cylinders of alumina, 7 inches in diameter and roughly 8 inches high — they are supposed to be uniformly doped with increasing amounts of weapons grade plutonium. In order for self shielding to account for the apparent 20% depletion of the 2.96g source, the plutonium would have to be crammed into a sphere with a radius of 1 inch. In fact, the γ-ray imaging of the Quinby Sources reported in a separate paper at this meeting indicates that the sources are reasonably close to being uniform, close enough that the factors indicated in Tab. 3 don't come anywhere close to explaining the active descrepancy.

Quinby Pu Sources

#	^{239}Pu(mg)	^{240}Pu(mg)	Factor
12	1	0.07	1.0
10	10	0.73	1.0
9	100	7.3	0.9993
12	1000	73	0.994
1	2940	213	0.973

Table 3:

Self Shielding in a Cylinder with Dimensions R,H

First Approximation

It is assumed that the fissile material does not appreciably attenuate the overall flux. This means that only effects within the calibration sample will be considered. The absorption within the sample will depend on the cross section for absorption of the materials in the sample. If several materials are present then the sum of the absorption probabilties should be used. Tab. 4 lists the relevevant parameters for both uranium and plutonium. The derivation assumes that the flux impinging on the sides of the slug is the same as that impinging on the top surface. The final form contains an integral which I could not put into closed form, necessitating a numerical integration.

$$\begin{aligned} Fission &= Flux * \rho * Avagadro/A * \sigma_f \\ Absorption &= Flux * \rho * Avagadro/A * \sigma_a \\ Probability &= Avagadro/A * \sigma_a \\ &= 2.574\ (cm^2/g) \qquad ^{239}Pu \\ &= 1.802 \qquad ^{235}U \\ Flux(x) &= Flux(0) * e^{-x\lambda_a} \qquad \lambda = Probability * \rho \end{aligned} \tag{1}$$

$$Fraction * volume = \frac{2H}{3}\int_0^{\pi/2} 2Rcos(\theta)d\theta \int_0^{Dcos(\theta)} e^{-\lambda x}\lambda dx + \frac{\pi R^2}{3}\int_0^H e^{-\lambda h}\lambda dh \tag{2}$$

$$Fraction = \frac{8}{3\pi\lambda D}\left[1 - \int_0^{\pi/2} cos(\theta)e^{-\lambda\, Dcos(\theta)}d\theta\right] + \frac{1-e^{-\lambda H}}{3\lambda H} \tag{3}$$

Cross Sections(barns)

Isotope	Fission	Absorb	Elastic
^{235}U	584.4	683.21	15.04
^{238}U	0.0	2.717	9.360
^{238}Pu	17.89	558.19	28.54
^{239}Pu	747.4	1017.7	7.968
^{240}Pu	0.06	289.5	1.642
^{241}Pu	1012.0	1373.5	11.35
^{242}Pu	0.0	18.79	8.318
^{241}Am	3.02	603.4	11.14
^{16}O	0.0	0.0	3.87

Table 4:

Self Shielding Calculations

Internal Flux

The challenge at this point is to generate self shielding corrections sufficiently accurate that the various calibration sets can be used to form the absolute calibration for the *APNea* active mode. The first approximation formula listed above does fairly well for a squat cylinder but increasingly misses the mark as the cylinder becomes taller. The first term of Eq. 3 is independent of H, so all of the effect of having a taller cylinder is contained in the second term, which goes to zero as H becomes large. The measurements of the various pellet stacks in Fig. 1 indicate a much more gradual increase in the self shielding effect with increasing height. The focus of the next order calculation is to try to include and consider other aspects of the thermal flux.

The first insight was a simple one and was quickly included in the first order formula. Simply considering the fission capture cross section to calculate the flux attenuation neglects the fact that both 235U and ^{239}Pu have additional capture cross section which does not lead to fission. Therefore, in calculating the attenuation of the flux in the source material, the total absorption must be used. Surprisingly, it is the case for depleted uranium, a favorite calibration source material, that the ^{238}U contributes significantly to the absorption though

it contributes essentially nothing to the fission output. In natural uranium the absorption due to ^{235}U is less than twice that due to the ^{238}U — whereas this drops to less than 1 for depleted uranium. Thus, for the depleted uranium slug used with the *APNea* system, greater than half of the observed ($\sim$20%) self shielding is associated with the ^{238}U.

The next insight was a less obvious one for the author; essentially that the elastic scattering of thermal neutrons in the source medium must be included. This arises because, even though the elastic scattering does not deplete the flux, it does retard its penetrating a volume and it tends to hold some of the flux within the source volume where it has a further chance to interact with the fissile material. What this entailed was to do an integration through the source volume of the capture fission response, of the flux attentuation, and of the flux elastic scatter. A second integration integrated the fission response to the now internal flux and the attenuation and scatter of this internal flux. This procedure is continued until the internal flux dwindles to an uninteresting level. Again, one must include all of the materials in the source matrix which contribute to elastic scattering. For the fuel pellets which are in the molecular form of UO_2, the oxygen in the molecule ups the elastic cross section from 9 barns to over 16 barns. For the 100% enriched ^{235}U dust, the ^{235}U contribution is so large that the oxygen plays essentially no role.

The results of these more detailed calculation have been included in the *Factor* columns of Tabs. 1,2 and have been incorporated in several of the figures. It must be remember, however, that the point of the self shielding calculations is to provide the basis for an absolute calibration of the *APNea* active mode. Because this work is still preliminary, some of the observed deviations have to do with the absolute calibration and only indirectly with the self shielding calculation. In particular, some of the discrepancy of the enriched pin assays at 2.1g and 4.5g is surely related to the uncertainty in the absolute calibration.

It can be seen in the pin assay data of Figs. 1a,b that the self shielding calculation has the self shielding factor (SSF) falling too rapidly with increasing height of the pellet stack. It is likely that the calculation of the internal flux is still not detailed enough and more

experimental and calculational work is planned. The results for the enriched dust are hidden in Fig. 2b. Here the assay results are plotted as a function of the shielded mass calculations. Of interest is the result of the vertical/horizontal measurements. Since the source is in the form of dust, it takes on a truly cylindrical shape only when the tiny container is the the **vertical** position. Then the dust settles to the bottom and takes on the cylindrical shape of the container. When the container is in a horizontal position, the dust spreads out and increases its surface area. It turns out that the *APNea* measurements are sufficiently precise as to see the difference, as can be seen in the figure. There should be less self shielding in the horizontal position, and that is the case. The vertical measurements agree nicely with the self shielding calculations — there is no obvious effect of too much self shielding with increasing height (mass). Part of this may be the fact that the H/R never becomes too extreme, but it is also explained by the fact that there is little internal flux within the pure dust.

The Quinby sources are of special interest because they feature plutonium rather than uranium, and they also feature a fairly uniform distribution of a low density of the material. The self shielding factors from calculations based on the γ-ray imaging are listed in Tab. reft:quin. Here it can be seen that the factors are simply not large, in fact, small enough to be lost in the measurement uncertainties. The measurements of ^{240}Pu shown in Fig. 3b demonstate the general integrity of the sources. The discrepancy in the assay values over the recorded mass values is less than 2%. On the other hand, the experimental uncertainty in the ^{240}Pu vlues can be seen to be dominated by counting statistics, even for the hottest source.

An interesting aspect for the plutonium sources is that there are 3 independent measures. The active measurement of ^{239}Pu is one, the auto-correlation measurement of ^{240}Pu is another, and the third is the direct measurement of the neutron output from the source (**Singles**). Since all of the Quinby sources were made from the same batch of weapons grade plutonium, one would expect that the singles rate should correlate closely with the actual

plutonium loading. Fig. 4a shows both the assay and the report values for ^{239}Pu plotted against the singles rate. The separation of the two sets of values at 1 gram is largely due to the uncertainty in the absolute calibration, but the Quinby value at 2.96g clearly does not follow the singles as well as do the *APNea* active results. This is unfortunate, since the singles rate should not be affected by self shielding or source distribution, but it is pleasing that the active results do seem to follow the singles rate fairly closely. As a further insight into the problem, Fig. 4b shows the region near 1 gram in more detail. The vertical separation of the two data sets depends on the absolute calibration, but the overall agreement of the various results with the singles measurements clearly indicates that the Quinby results have a degree of error which is noticeable.

Conclusions

The method for calculating the self shielding correction for cylindrical sources clearly predicts too much shielding as the height and fissile mass of the source increases, but the deviation is relatively small. It does mean, however, that the quality of a fissile source is compromised if the actual fissile contribution in a particular differential dieaway device cannot be precisely predicted. In general, the quality of the current predictions is adequate to provide calibration points for the active mode the *APNea* System. The experience is sufficient, however, to indicate that establishing the basis for the active mode calibration is not trivial and that correcting for the actual clumping of fissile material in waste will be difficult whenever significant clumping is suspected.

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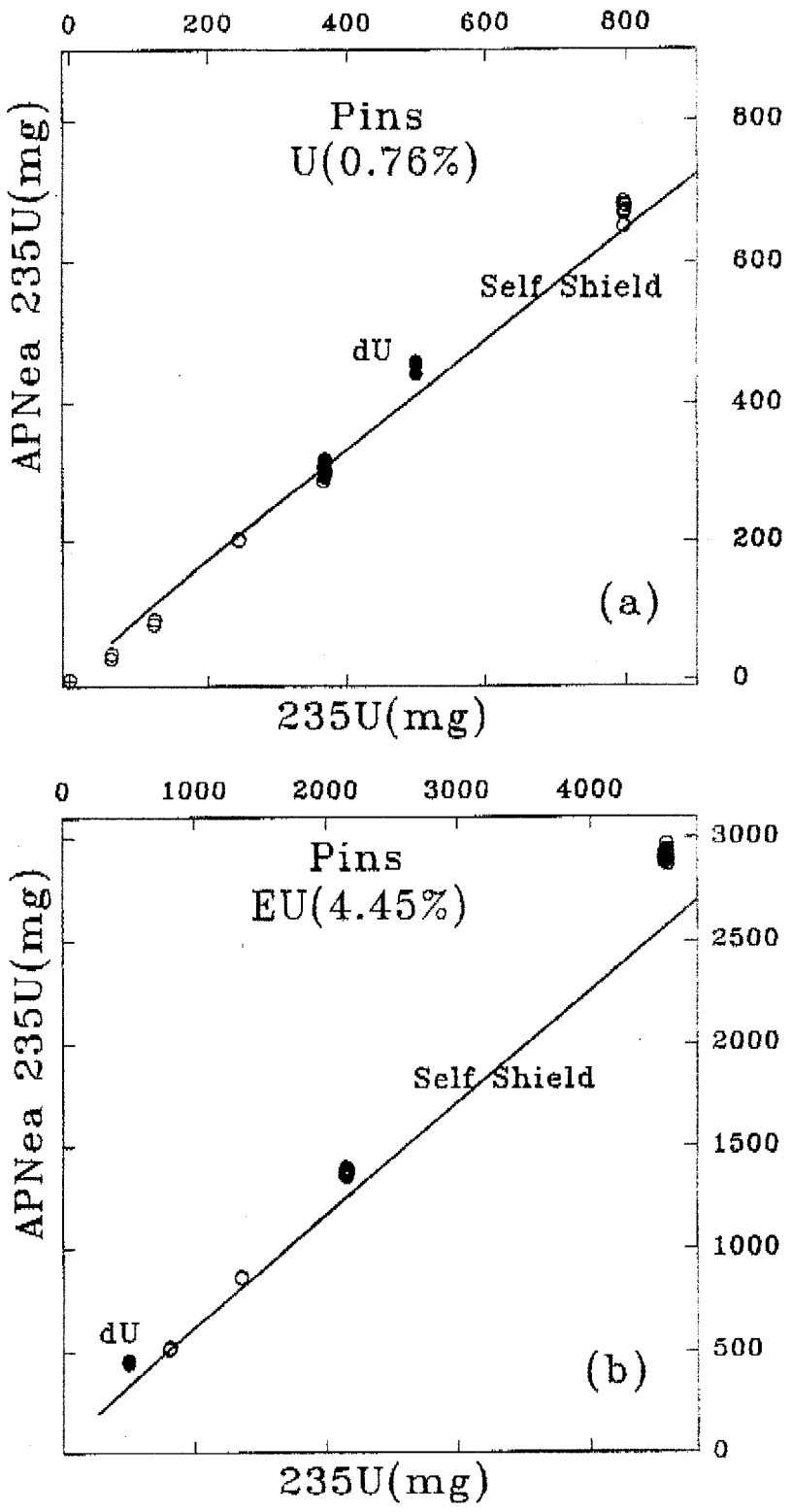

Figure 1: Pins of Fuel Pellets

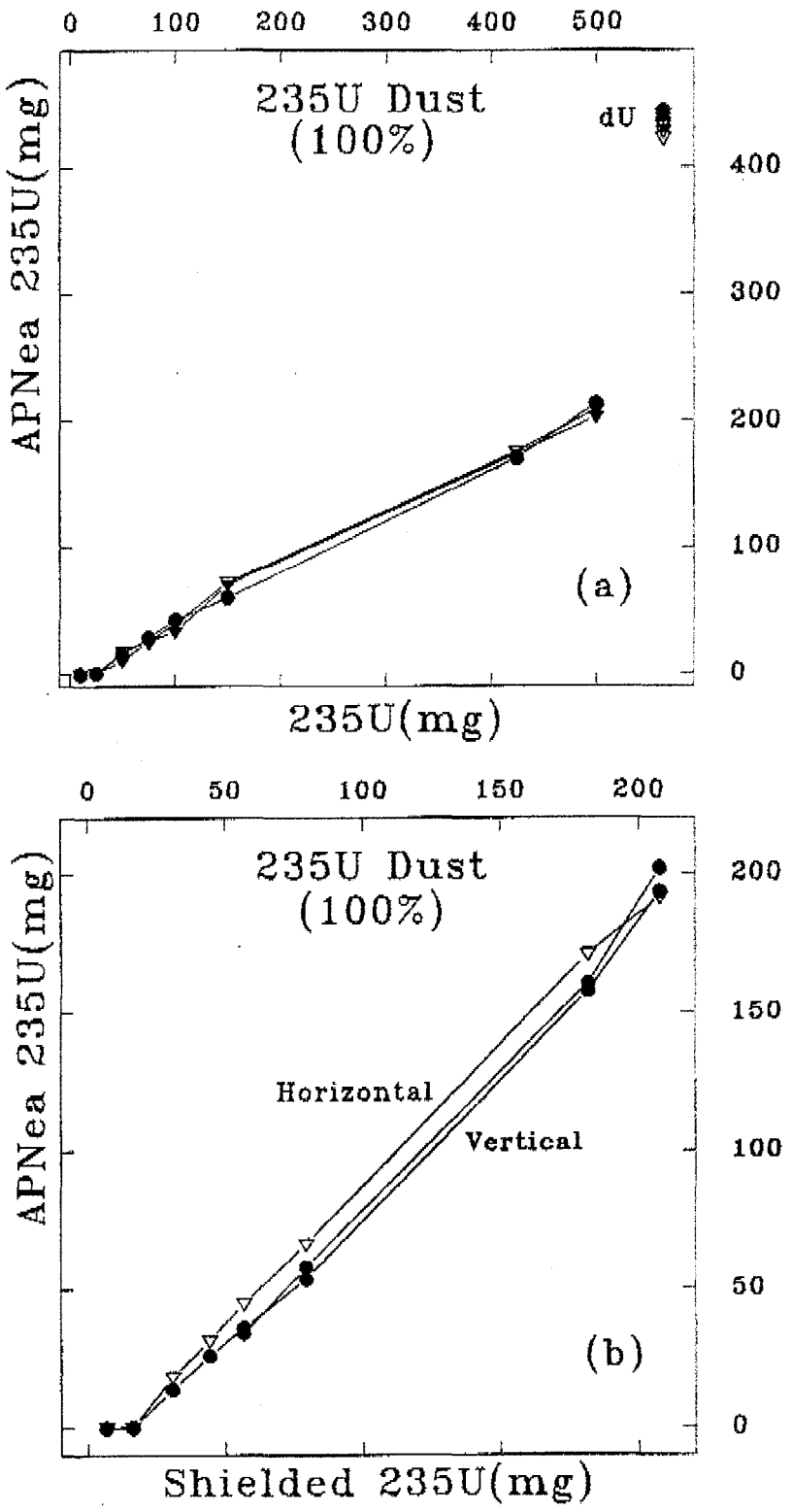

Figure 2: Pure 235U Dust

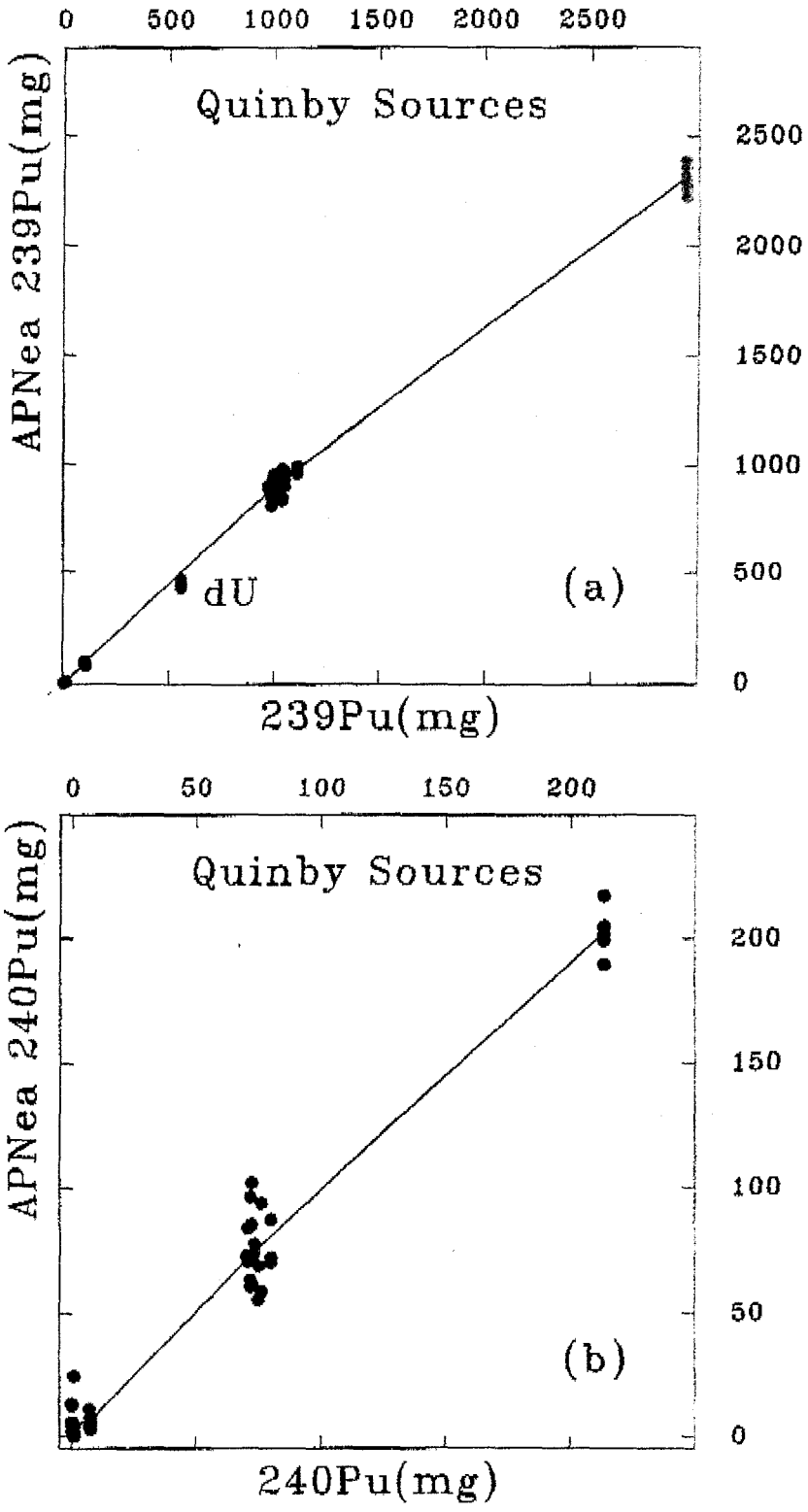

Figure 3: Quinby Sources

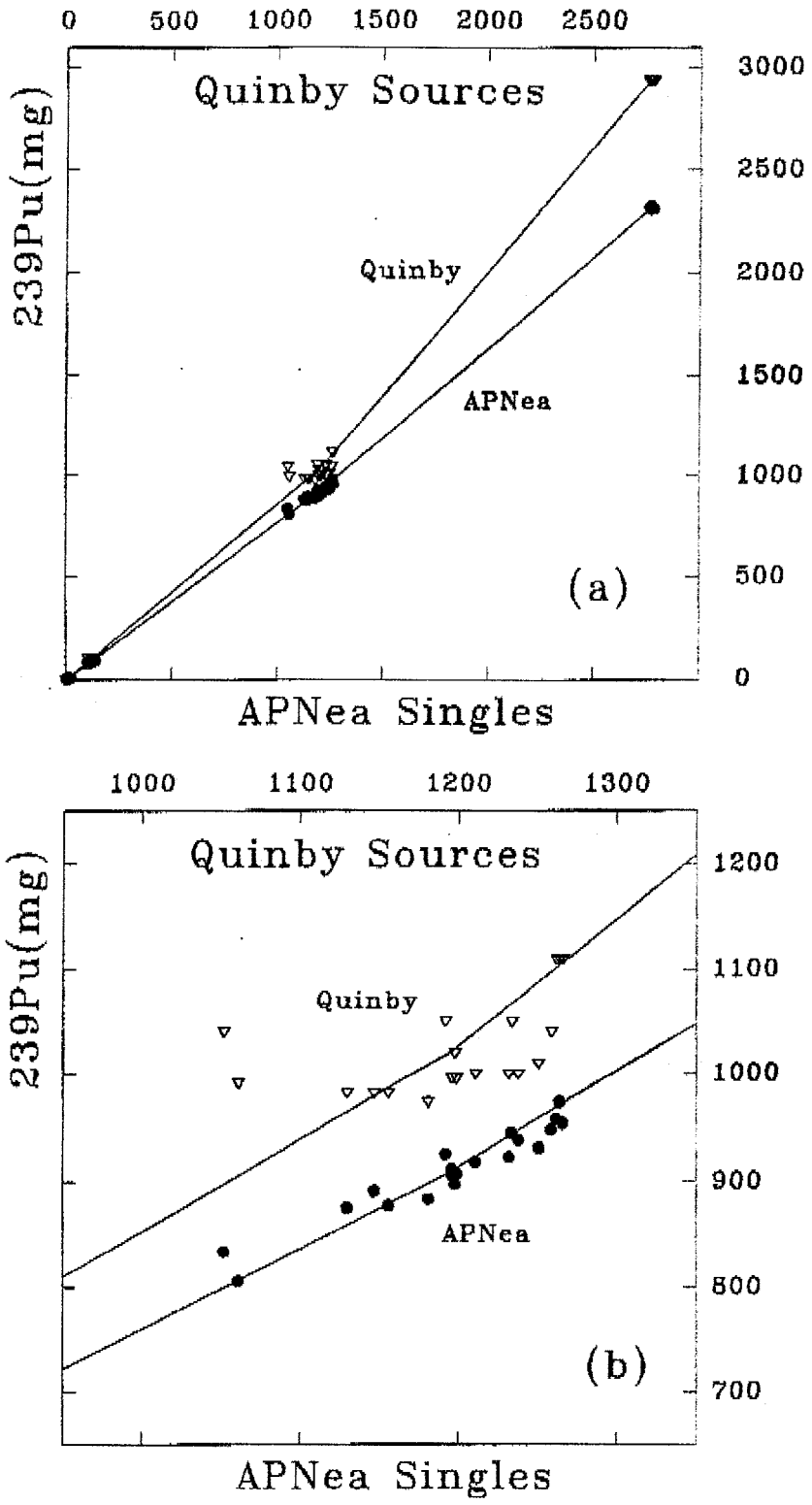

Figure 4: Quinby Singles