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Distinguishing Pu metal from Pu oxide using fast neutron counting

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Generalization of Hage-Cifarelli Eqs for Pu oxides

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In an object with (α,n) neutron generation the Feynman 2-neutron correlation function Y_{2F} (t) is given by:

$$Y_{2F}(t) = \frac{(\varepsilon q)_f^2 M_f^2}{(\varepsilon q)_f [M_f + \alpha(M_\alpha - 1)] + \alpha(\varepsilon q)_\alpha} \left\{ D_{2s} + [(M_f - 1) + \alpha(M_\alpha - 1)] D_2 \right\} \left(1 - \frac{1 - e^{-\lambda t}}{\lambda t} \right), \quad (1)$$

while Feynman 3-neutron correlations Y_{2F} and Y_{3F} as a function of time have the form

$$Y_{3F1}(t) = \frac{(\varepsilon q)_f^3 M_f^3}{(\varepsilon q)_f [M_f + \alpha(M_f - 1)] + \alpha(\varepsilon q)_\alpha} \left\{ D_{3s} + [(M_f - 1) + \alpha(M_\alpha - 1)] D_3 \right\} \left(1 - \frac{3 - 4e^{-\lambda t} + e^{-2\lambda t}}{2\lambda t} \right)$$

(2)

$$Y_{3F2}(t) = \frac{2(\varepsilon q)_f^3 M_f^3}{(\varepsilon q)_f [M_f + \alpha(M_\alpha - 1)] + \alpha(\varepsilon q)_\alpha} (M_f - 1) \left\{ D_{2s} D_2 + [(M_f - 1) + \alpha(M_\alpha - 1)] D_2^2 \right\} \left(1 - \frac{2 - (2 + \lambda t) e^{-\lambda t}}{\lambda t} \right),$$

where $(\varepsilon q)_f$, $(\varepsilon q)_\alpha$, and M_f , M_α are detection efficiencies and neutron multiplications for fission and (α,n) neutrons. These quantities are in general different for fission and (α,n) neutrons because the fission and (α,n) neutrons have different energy spectra. Indeed for the case of PuO_2 the efficiency for detecting the oxygen (α, n) neutrons in our liquid scintillators will be significantly higher than for fission neutrons, because the energy spectrum for the oxygen (α,n) neutrons is peaked around 2 MeV where the intrinsic efficiency of the detector is relatively large, whereas almost half the fission neutrons have energies below 1 MeV where the intrinsic efficiency of our liquid scintillators is very small. However, because the fission cross-section is fairly flat across all the energies of interest, the neutron multiplications for fission and (α,n) neutrons are nearly the same. Therefore in the following we will set $M_\alpha = M_f \equiv M$. In Eq.s 1-2 α is the usual parameter specifying the strength of (α,n) neutron emission relative to the rate of neutron emission from spontaneous fission (mainly due to Pu240). The denominator of the first factor in Eq's. 1-2 is proportional to the observed count rate

$$R_1 = (\varepsilon q)_f [M_f + \alpha(M_\alpha - 1)] \bar{\nu}_s F_s + (\varepsilon q)_\alpha \alpha \bar{\nu}_s F_s \quad (3)$$

Following Hage-Cifarelli [1] one might contemplate using the ratio of R_{3F} /R_{2F}^2 to determine the multiplication *M*. In our case we can write this ratio as:

$$\frac{R_{3F}}{R_{2F}^2} = \frac{2(M-1)D_2 + D_{3s} + (1+\alpha)(M-1)D_3}{[D_{2s} + (1+\alpha)(M-1)D_2]^2}\left(\frac{R_1}{(\varepsilon q)_f M \bar{\nu}_s F_s}\right) \tag{4}$$

The factor in parenthesis represents the ratio of the count rate including (α,n) emission to the count rate minus the terms proportional to α (note: that this is different from what the count rate would be without (α,n) emission because the multiplication would be different). Unfortunately, in contrast with the situation considered by Hage-Cifarelli where there were no (α,n) neutrons, we cannot use expression (4) to determine the multiplication because of the presence of the unkowns α, ε_f, and ε_α. We can however eliminate the efficiencies be making use of the theoretical fission and (α,n) spectra to calculate the ratio $\varepsilon_\alpha/\varepsilon_f$ for the particular kind of fast neutron detector (e.g. a liquid scintillaor) one is using. Calling this ratio r we have

$$\frac{R_{3F}}{R_{2F}^2} = \frac{2(M-1)D_2\left[D_{2s} + (1+\alpha)(M-1)D_2\right] + D_{3s} + (1+\alpha)(M-1)D_3}{[D_{2s} + (1+\alpha)(M-1)D_2]^2}\left[1+\alpha\left(1+\frac{r-1}{M}\right)\right] \tag{5}$$

Given a value for α we could in principle use Eq 5 to solve for the multiplication. Unfortunately this equation is now a cubic equation rather than a quadratic equation as in the case of pure metal. As an alternative we can use the observed spectrum to directly evaluate the quantity in brackets on the r.h.s. of Eq. 5. In particular, fitting the observed fast neutron spectrum to a sum of fission and (α,n) spectra yields two coefficients whose ratio ρ allows us express the quantity in brackets on the r.h.s. of Eq. 5 in terms of experimental quantities . The ratio ρ is related to α and the multiplication *M* by

$$\rho = \frac{\alpha r}{(1+\alpha)M - \alpha} \tag{6}$$

Given this additional experimental input, one can express the quantity in brackets on the r.h.s. of Eq. 5 in the form

$$!+\alpha\left(1+\frac{r-1}{M}\right)=\frac{(1+\alpha)(1+\rho)}{(1+\rho/r)} \tag{7}$$

Given a values for r, ρ, and α, one may simply use Eq. (6) to solve for *M*. If α is unknown, one can substitute Eq. (7) into Eq. (5) to obtain a second independent equation connecting *M* and α:

$$2(1+\alpha)(M-1)D_2\left[D_{2s}+(1+\alpha)(M-1)D_2\right]+(1+\alpha)D_{3s}+(1+\alpha)^2(M-1)D_3$$
$$=\frac{1+\rho/r}{1+\rho}\frac{R_{3F}}{R_{2F}^2}\left[D_{2s}+(1+\alpha)(M-1)D_2\right]^2 \tag{8}$$

This equation may be regarded as a generalization of the Hage-Cifarelli quadratic equation for *M* that includes (α,n) processes, and can be used together with Eq. (6) to determine both *M* and α. Of course, simply determining that ρ >1 insures that one is dealing with oxide rather than metal. However substituting the relation

$$(1+\alpha)(M-1)=\frac{\alpha r}{\rho}-1 \tag{9}$$

into Eq. (8) leads to a quadratic equation for α that is readily solved if r and ρ are known. We have applied this method to Monte Carlo simulations of the signal recorded in an array of liquid scintillators due to fast neutrons emitted by a Pu oxide sample with a known α-ratio, and the value for α obtained from Eq.s (8) and (9) agreed very well with the known α-ratio.

In conclusion, by fitting the observed distribution of energies deposited in a liquid scintillator detector to the distribution one would expect from a fast neutron spectrum that was a sum of theoretical fission and (α, n) spectra, we were able to obtain quite accurate values for both *M* the α.

[1] W. Hage and D. Cifarelli, *Nucl. Inst. and Methods* A251, 550 (1986).

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