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RADIATION PHYSICS NOTE 130

Induced ^{11}C Activity Buildup in Uniform Pulsed Beams

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I. Introduction

Early in 1997 it was decided to reopen a previous effort by the Radiation Physics Team to develop a neutron detector based on the production of ^{11}C in a plastic scintillator. In order to quantify the specific activity of ^{11}C during and after exposure in pulsed radiation fields, it is necessary to derive an appropriate expression for its specific activity at any given time. That is the purpose of this note. It should be noted that in 1959-60, J.B. McCaslin set forth the basic idea and principles involved in developing a detector based on the $^{12}C(n,2n)^{11}C$ reaction in ref. (1). Reference (2) provides direction in how to use radiation measurement instruments in pulsed fields. The exponential decay of charge in radiation measuring instruments provides a direct analog for the buildup and decay of radionuclides in material during exposures to pulsed radiation fields. Thus the results of ref. (2) can be used as a check for the derivation presented in this note.

II. Derivation

Assume that some polymer type material composed of carbon and hydrogen or other low Z materials, e.g., plastic scintillator, is exposed to a pulsed neutron radiation field. Define the following symbols:

$N(t)$	= The number of ^{11}C atoms present in detector at time t.
λ_{11}	= Decay constant for ^{11}C. (sec^{-1})
I_0	= Neutron flux density in each pulse. (n/cm^2-sec)
ρ	= Density of detector material. (gm/cm^3)
x_0	= Thickness of detector normal to the beam. (cm)
N_A	= Avogadro's number. (atoms/GMW)
A	= Atomic weight of detector material. (grams/GMW)
σ	= Nuclear reaction cross section for $^{12}C(n,2n)^{11}C$. (cm^2/atom)
τ_{on}	= Beam on period or pulse width. (seconds)
τ_{off}	= Beam off period or time interval between pulses. (seconds)
V	= Volume of material exposed to the neutron radiation field. (cm^3)

The periodic nature of the irradiation of a detector in a pulsed hadron beam can be schematically visualized in Figure 1.

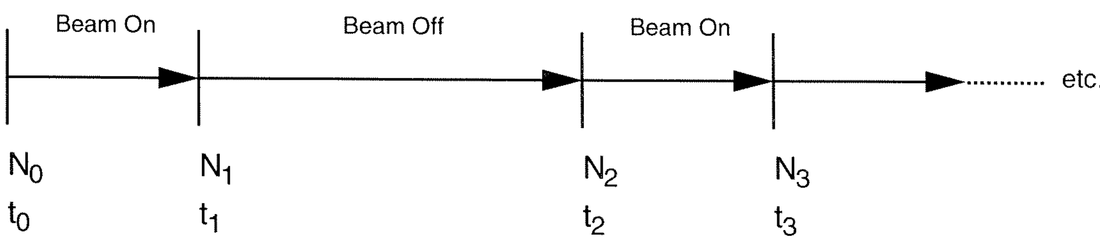

Figure 1

During the beam on time period the number of ^{11}C nuclei present at a time t is governed by the differential equation:

$$\frac{dN(t)}{dt} = -\lambda_{11} N(t) + I_0 (\rho \sigma V)\left(\frac{N_A}{A}\right) \tag{1}$$

The number of ^{11}C nucleii present during sentient periods, i.e., beam off periods, can be represented by the general radioactive decay equation:

$$\frac{dN(t)}{dt} = -\lambda_{11} N(t) \tag{2}$$

Since the second term on the right hand side of the beam on production equation, equation (1), is dependent on a group of material and beam dependent constants a single constant, P_0, can be substituted for it without any loss of generality. A critical caveat here is that uniform beam irradiation conditions must exist.

$$\frac{dN(t)}{dt} = -\lambda_{11} N(t) + P_0$$

A general solution to this differential equation on the interval defined by (N_i, t_i) and (N_f, t_f) is:

$$e^{-\lambda_{11}(t_f - t_i)} = \frac{\left(P_0 - \lambda_{11} N_f\right)}{\left(P_0 - \lambda_{11} N_i\right)} \tag{3}$$

In the special case where N_f=N(t), N_i=0, t_i=0, and t_f=t the above equation reduces to the familiar steady state irradiation (for time t) equation:

$$N(t) = \frac{P_0}{\lambda_{11}}\left(1 - e^{-\lambda_{11} t}\right)$$

Thus for pulsed beam irradiations, the following sequence of equations would describe the number of ^{11}C atoms, with decay constant λ_{11}, present.

$0 \le t \le t_1$ $$N(t) = \frac{P_0}{\lambda_{11}}\left(1 - e^{-\lambda_{11} t}\right)$$

$t = t_1$ $$N(t_1) = \frac{P_0}{\lambda_{11}}\left(1 - e^{-\lambda_{11} t_1}\right) \equiv N_1$$

$t_1 \le t \le t_2$ $$N(t) = N_1 e^{-\lambda_{11}(t - t_1)} = \frac{P_0}{\lambda_{11}}\left(1 - e^{-\lambda_{11} t_1}\right)\left[e^{-\lambda_{11}(t - t_1)}\right]$$

$$N(t_2) \equiv N_2$$

Using the general formula from above for buildup and decay during an irradiation period, i.e., equation (3), and substituting t for t_f:

$t_2 \le t \le t_3$

$$N(t) = \frac{1}{\lambda_{11}}\left\{P_0 - \left(P_0 - \lambda_{11}N_i\right)e^{-\lambda_{11}(t-t_i)}\right\}$$

$$N_i \equiv N_2$$

$$N(t) = \frac{1}{\lambda_{11}}\left\{P_0 - \left(P_0 - \lambda_{11}N_2\right)e^{-\lambda_{11}(t-t_2)}\right\}$$

$$N(t) = \frac{1}{\lambda_{11}}\left\{P_0 - \left[P_0 - \lambda_{11}\left(\frac{P_0}{\lambda_{11}}\left(1-e^{-\lambda_{11}t_1}\right)\left(e^{-\lambda_{11}(t_2-t_1)}\right)\right)\right]e^{-\lambda_{11}(t-t_2)}\right\}$$

$$N(t) = \frac{1}{\lambda_{11}}\left\{P_0 - \left[P_0 - P_0\left(1-e^{-\lambda_{11}t_1}\right)\left(e^{-\lambda_{11}(t_2-t_1)}\right)\right]e^{-\lambda_{11}(t-t_2)}\right\}$$

The time period t_1 corresponds to τ_{on} and the interval (t_2-t_1) corresponds to τ_{off}. So:

$$N(t) = \frac{1}{\lambda_{11}}\left\{P_0 - \left[P_0 - P_0\left(1-e^{-\lambda_{11}\tau_{on}}\right)\left(e^{-\lambda_{11}\tau_{off}}\right)\right]e^{-\lambda_{11}(t-t_2)}\right\}$$

$$N(t) = \frac{P_0}{\lambda_{11}}\left\{1 - \left[1 - \left(1-e^{-\lambda_{11}\tau_{on}}\right)\left(e^{-\lambda_{11}\tau_{off}}\right)\right]e^{-\lambda_{11}(t-t_2)}\right\}$$

$$N(t_3) = \frac{P_0}{\lambda_{11}}\left\{1 - \left[1 - \left(1-e^{-\lambda_{11}\tau_{on}}\right)\left(e^{-\lambda_{11}\tau_{off}}\right)\right]e^{-\lambda_{11}(t_3-t_2)}\right\} \equiv N_3$$

$$N_3 = \frac{P_0}{\lambda_{11}}\left\{1 - \left[1 - \left(1-e^{-\lambda_{11}\tau_{on}}\right)\left(e^{-\lambda_{11}\tau_{off}}\right)\right]e^{-\lambda_{11}\tau_{on}}\right\}$$

$t_3 \le t \le t_4$

$$N(t) = N_3 e^{-\lambda_{11}(t-t_3)}$$

Substituting for N_3:

$$N(t)=\frac{P_0}{\lambda_{11}}\left\{1-\left[1-\left(1-e^{-\lambda_{11}\tau_{on}}\right)e^{-\lambda_{11}\tau_{off}}\right]e^{-\lambda_{11}\tau_{on}}\right\}e^{-\lambda_{11}(t-t_3)}$$

$$N(t)=\frac{P_0}{\lambda_{11}}\left\{1-\left[e^{-\lambda_{11}\tau_{on}}-\left(1-e^{-\lambda_{11}\tau_{on}}\right)e^{-\lambda_{11}(\tau_{off}+\tau_{on})}\right]\right\}e^{-\lambda_{11}(t-t_3)}$$

Now define the pulse repetition time, i.e., the period as:

$$\tau=\tau_{on}+\tau_{off} \tag{4}$$

Then

$$N(t)=\frac{P_0}{\lambda_{11}}\left\{1-e^{-\lambda_{11}\tau_{on}}+e^{-\lambda_{11}\tau}-e^{-\lambda_{11}(2\tau_{on}+\tau_{off})}\right\}e^{-\lambda_{11}(t-t_3)}$$

At the end of the time interval, i.e., $t = t_4$;

$$N(t_4)=\frac{P_0}{\lambda_{11}}\left\{1-e^{-\lambda_{11}\tau_{on}}\left(1-e^{-\lambda_{11}\tau_{off}}+e^{-\lambda_{11}\tau}\right)\right\}e^{-\lambda_{11}(t_4-t_3)}\equiv N_4$$

$t_4 \le t \le t_5$

$$N(t)=\frac{1}{\lambda_{11}}\left\{P_0-\left(P_0-\lambda_{11}N_4\right)e^{-\lambda_{11}(t-t_4)}\right\}$$

$$N(t)=\frac{P_0}{\lambda_{11}}\left\langle 1-\left\{1-\left[1-e^{-\lambda_{11}\tau_{on}}\left(1-e^{-\lambda_{11}\tau_{off}}+e^{-\lambda_{11}\tau}\right)\right]e^{-\lambda_{11}\tau_{off}}\right\}e^{-\lambda_{11}(t-t_4)}\right\rangle$$

$$N(t)=\frac{P_0}{\lambda_{11}}\left\langle 1-\left\{1-e^{-\lambda_{11}\tau_{off}}+e^{-\lambda_{11}(\tau_{on}+\tau_{off})}-e^{-\lambda_{11}(\tau_{on}+2\tau_{off})}+e^{-2\lambda_{11}(\tau_{on}+\tau_{off})}\right\}e^{-\lambda_{11}(t-t_4)}\right\rangle$$

At the end of the time interval, $t = t_5$, $N(t_5) \equiv N_5$, i.e.;

$$N(t_5)=\frac{P_0}{\lambda_{11}}\left\langle 1-\left\{1-e^{-\lambda_{11}\tau_{off}}+e^{-\lambda_{11}(\tau_{on}+\tau_{off})}-e^{-\lambda_{11}(\tau_{on}+2\tau_{off})}+e^{-2\lambda_{11}(\tau_{on}+\tau_{off})}\right\}e^{-\lambda_{11}(t_5-t_4)}\right\rangle\equiv N_5$$

Since $(t_5-t_4) \equiv \tau_{on}$;

$$N_5=\frac{P_0}{\lambda_{11}}\left\langle 1-\left\{e^{-\lambda_{11}\tau_{on}}-e^{-\lambda_{11}\tau}+e^{-\lambda_{11}(2\tau_{on}+\tau_{off})}-e^{-2\lambda_{11}(\tau_{on}+\tau_{off})}+e^{-\lambda_{11}(3\tau_{on}+2\tau_{off})}\right\}\right\rangle$$

Again on the time interval t_5 to t_6 there is only decay of the built up ^{11}C and the representative equation is:

$$t_5 \le t \le t_6 \qquad\qquad N(t) = N_5 e^{-\lambda_{11}(t-t_5)}$$

$$N_6 = \frac{P_0}{\lambda_{11}} \left\langle 1 - \left\{ e^{-\lambda_{11}\tau_{on}} - e^{-\lambda_{11}\tau} + e^{-\lambda_{11}\tau_{on}} e^{-\lambda_{11}\tau} - e^{-2\lambda_{11}\tau} + e^{-\lambda_{11}\tau_{on}} e^{-2\lambda_{11}\tau} \right\} \right\rangle e^{-\lambda_{11}(t_6-t_5)}$$

$$N_6 = \frac{P_0}{\lambda_{11}} \left\{ e^{-\lambda_{11}\tau_{off}} - e^{-\lambda_{11}\tau} + e^{-\lambda_{11}\tau_{off}} e^{-\lambda_{11}\tau} - e^{-2\lambda_{11}\tau} + e^{-\lambda_{11}\tau_{off}} e^{-2\lambda_{11}\tau} - e^{-3\lambda_{11}\tau} \right\}$$

If the irradiation is now terminated at a time, t_n, then the final expression for N as a function of time would be:

$$N(t) = \frac{P_0 e^{-\lambda_{11}(t-t_n)}}{\lambda_{11}} \left[1 - \left(e^{-\lambda_{11}\tau_{on}} + e^{-\lambda_{11}\tau_{on}} e^{-\lambda_{11}\tau} + e^{-\lambda_{11}\tau_{on}} e^{-2\lambda_{11}\tau} + L \right) + \left(e^{-\lambda_{11}\tau} + e^{-2\lambda_{11}\tau} + e^{-3\lambda_{11}\tau} + L \right) \right]$$

The time dependent piece of this equation can be compressed by letting

$$N_0(t) \equiv \frac{P_0}{\lambda_{11}} e^{-\lambda_{11}(t-t_n)}$$

where t is the start time for the sample count and t_n is the irradiation time (assuming the irradiation is started at t=0). If we now define m to be the exponential series index during time t_n then the above equation for N(t) can be generally expressed as:

$$N(t) = N_0(t) \left[1 - e^{-\lambda_{11}\tau_{on}} \left(1 + e^{-\lambda_{11}\tau} + e^{-2\lambda_{11}\tau} + L + e^{-m\lambda_{11}\tau} \right) + \left(e^{-\lambda_{11}\tau} + e^{-2\lambda_{11}\tau} + e^{-3\lambda_{11}\tau} + L + e^{-m\lambda_{11}\tau} \right) \right]$$

So far two indices have been introduced; (1) the time interval counting index, n, and (2) the exponential series index, m. However, the number of beam pulses, p, is the index of primary interest to this derivation. By inspection, it can be deduced that the relationships between these three indices are;

$$n = 2m + 1 \qquad (5)$$

and

$$n = 2p - 1 \qquad (6)$$

From these two relationships it is straightforward to derive the following relationship between m and p;

$$m = p - 1 \tag{7}$$

Collecting terms;

$$N(t) = N_0(t)\left[\left(1 - e^{-\lambda_{11}\tau_{on}}\right)\left(1 + e^{-\lambda_{11}\tau} + e^{-2\lambda_{11}\tau} + \ldots + e^{-m\lambda_{11}\tau}\right)\right] \tag{8}$$

The second exponential series meets the definition of a geometric series with;

$$r = e^{-\lambda_{11}\tau}, \quad a = 1, \quad n_s - 1 = m, \quad \text{and a finite sum of,} \quad S = \frac{\left(1 - e^{-(m+1)\lambda_{11}\tau}\right)}{\left(1 - e^{-\lambda_{11}\tau}\right)}$$

Substituting the expression for the finite sum of a geometric series into the last equation for N(t);

$$\boxed{N(t) = N_0(t)\left[\left(1 - e^{-\lambda_{11}\tau_{on}}\right)\frac{\left(1 - e^{-(m+1)\lambda_{11}\tau}\right)}{\left(1 - e^{-\lambda_{11}\tau}\right)}\right]}$$

Or in terms of the number of beam pulses:

$$\boxed{N(t) = N_0(t)\left[\left(1 - e^{-\lambda_{11}\tau_{on}}\right)\frac{\left(1 - e^{-p\lambda_{11}\tau}\right)}{\left(1 - e^{-\lambda_{11}\tau}\right)}\right]} \tag{9}$$

This now is a useable expression for the number of ^{11}C atoms present in a material at the beginning of a count period under uniform beam conditions, i.e., I_0 and the pulse repetition rate are reasonably constant during the irradiation time.

By definition, the ^{11}C activity, A_{11}, at time t after the irradiation can be expressed as:

$$\boxed{A_{11} = \lambda_{11} N(t)}$$

III. <u>Determination of Beam Flux from ^{11}C Induced Activity</u>.

If the sample material is counted at a time t_c, then the number of ^{11}C atoms present at the beginning of the count time is given by;

$$N(t_c) = \frac{P_0}{\lambda_{11}} \left[\frac{\left(1 - e^{-\lambda_{11}\tau_{on}}\right)}{\left(1 - e^{-\lambda_{11}\tau}\right)} \right] \left(1 - e^{-p\lambda_{11}\tau}\right)\left(e^{-\lambda_{11}(t_c - t_n)}\right) \equiv N_c \qquad (10)$$

and the number of ^{11}C atoms present in the material at the conclusion of the sample count, N_f, is given by;

$$N_f = N_c e^{-\lambda_{11}(t_f - t_c)}$$

To simplify nomenclature the time intervals $(t_c - t_n)$ and $(t_f - t_c)$ can be defined as the decay time, T_d, and the count time, T_c, respectively. Note that T_c is the counting system real time and **not** the live time.

When one counts a radioactive sample, it is a fraction of the number of decays during the counting time interval, T_c, which is actually recorded. For a given detector, the number of decays counted, ΔN_D is given by the recorded counts, and the actual number of decays, ΔN_A, is given by;

$$\Delta N_A = \frac{(\Delta N_D)(DTC)}{\varepsilon(\Omega_f)(F_a)(R_B)} \qquad (11)$$

where:

- ε $\equiv$ the ratio of ^{11}C radioactive emissions incident on the detector which are converted by the detector into counts, i.e., the intrinsic efficiency of the detector.
- Ω_f $\equiv$ the fractional solid angle (of 4π) subtended by the detector at the sample being counted.
- F_a $\equiv$ the ratio of ^{11}C radioactive emissions transmitted from their point of origin to the detector without any modification in properties by intervening material which would prevent them from being counted, i.e., self absorption of the material.
- R_B $\equiv$ the ratio of ^{11}C radioactive decays which produce the specific radiation being counted by the detector, i.e., the transition branching ratio.
- DTC $\equiv$ the detector system dead time correction factor.

The actual number of decays can also be expressed as the difference between the final and initial number of ^{11}C atoms present, i.e.

$$\Delta N_A = -(N_f - N_c) = -\left(N_c e^{-\lambda_{11}T_c} - N_c\right) = N_c\left(1 - e^{-\lambda_{11}T_c}\right)$$

Setting the expressions for ΔN_A equal we obtain the equality:

$$N_c\left(1-e^{-\lambda_{11}T_c}\right)=\frac{(\Delta N_D)(DTC)}{(\varepsilon)(\Omega_f)(F_a)(R_B)}$$

In order to simplify manipulation of the above equation, define a parameter, B_0, which is dependent only on the beam pulse width, τ_{on}, and the pulse repetition time, τ, as;

$$B_0 \equiv \frac{\left(1-e^{-\lambda_{11}\tau_{on}}\right)}{\left(1-e^{-\lambda_{11}\tau}\right)}$$

Substituting for N_c, the following equality results;

$$\frac{P_0}{\lambda_{11}}B_0\left(1-e^{-p\lambda_{11}\tau}\right)\left(e^{-\lambda_{11}T_d}\right)=\frac{(\Delta N_D)(DTC)}{(\varepsilon)(\Omega_f)(F_a)(R_B)\left(1-e^{-\lambda_{11}T_c}\right)}$$

Rearranging terms;

$$P_0=\frac{(\Delta N_D)(DTC)\lambda_{11}\left(e^{\lambda_{11}T_d}\right)}{(\varepsilon)(\Omega_f)(F_a)(R_B)(B_0)\left(1-e^{-p\lambda_{11}\tau}\right)\left(1-e^{-\lambda_{11}T_c}\right)} \tag{12}$$

Substituting for P_0;

$$\boxed{I_0=\frac{\lambda_{11}(\Delta N_D)(DTC)(A)\left(e^{\lambda_{11}T_d}\right)}{(\rho\sigma V)(N_A)(\varepsilon)(\Omega_f)(F_a)(R_B)(B_0)\left(1-e^{-p\lambda_{11}\tau}\right)\left(1-e^{-\lambda_{11}T_c}\right)}} \tag{13}$$

an expression for the instantaneous neutron flux density, I_0, is the result. The average neutron flux density during irradiation is given by:

$$\boxed{I_A=\frac{I_0\times\tau_{on}}{\tau}} \tag{14}$$

The total neutron flux density or fluence, N_N, can now be obtained by mulitplying I_0 by the number of pulses, p, and the pulse width in time, τ_{on}, i.e.;

$$\boxed{N_N=I_0(p\tau_{on})} \tag{15}$$

This expression provides information about the number of neutrons incident on the original detector material with sufficient energy to initiate the $^{12}C(n,2n)^{11}C$ nuclear reaction.

IV. Example: Supplement to Multisphere Measurements.

A neutron detector based on ^{11}C activation serves to supplement measurements with the usual multisphere system because such a system is rather insensitive to neutrons with energy much greater than 20 MeV, even when the largest moderating sphere is used. In order to utilize a plastic scintillator along with a set of multispheres the so-called response function of the scintillator to neutrons of energy above 20 MeV must be known. An example for a 2" diameter by 2" high cylindrical plastic scintillator is discussed below.

For a 2" diameter by 2" high plastic scintillator, the volume is 103 cm^3. With a density of 1.032 g/cm^2 the mass is 106.3 g, and for a ratio of Hydrogen to Carbon atoms of 1.1, A, the molecular weight is 13.1, and the total number of atoms in the material is 4.89×10^{24}. The energy dependence of the crosssection for the $^{12}C(n,2n)^{11}C$ reaction is not well defined, but is usually taken to be 0 at energies below 20 MeV, to rise rapidly to a value of 22×10^{-27} cm^2, and to remain constant at that value up to about 1 GeV.

Based on the above numbers the material dependent part of the constant P_0, defined in Sect. II, equals 0.108 cm^2. For irradiation to saturation, the response function (or,as it is sometimes called, the Figure of Merit) for a 2" diameter by 2" high plastic scintillator is 0.108 counts/sec (or 6.5 counts/min) per unit flux density (n/cm^2-sec), and is constant with neutron energy.

The neutron fluence spectrum obtained from measurements with a set of multispheres is determined from the appropriately corrected and normalized counts in each detector by unfolding the fluence from its product with the sphere response functions in each energy bin. Therefore, when ^{11}C activation is included to supplement the sphere measurements a similiar quantity is required. In fact, it is the actual ^{11}C counts, averaged over the irradiation, and appropriately corrected for decay and saturation, that is needed. Thus, to include ^{11}C activation along with a multisphere measurement of neutron fluence spectra one should use the expression for total ^{11}C counts, P_0/λ_{11}, Eq. 12, multiplied by τ_{on}/τ to average over the irradiation, as input to the unfolding codes.

V. Summary.

While the expressions for induced activity and neutron flux discussed in this report were derived explicitly for the buildup of ^{11}C in plastic scintillator, they can be straightforwardly applied to any induced isotope in a material matrix under pulsed beam irradiation.

VI. References

1. J.B. McCaslin, A High Energy Neutron-Flux Detector, **Health Physics**, Vol. 2, p399-407, 1960.

2. J.D. Cossairt, Usage of Chipmunks and Scarecrows in Tevatron Radiation Fields, Fermilab Radiation Physics Note No. 44, August, 1984.