ABSOLUTE DISINTEGRATION RATE DETERMINATION OF BETA-EMITTING RADIONUCLIDES BY THE PULSE HEIGHT SHIFT-EXTRAPOLATION METHOD

by

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It has long been the hope of investigators using liquid scintillation counters to be able to determine the disintegration rate of a radionuclide in a sample directly, without having to use a standard solution of the radionuclide to calibrate the counting system. In this paper, a method will be described which enables the investigator to determine the disintegration rate of a beta emitting radionuclide in a sample by measuring the pulse height shift produced by quenching (either real or simulated). This paper also describes two new liquid scintillation counters, Beckman LS-8000 and Beckman LS-9000, which provide the capability for determination of disintegration rates by this method.

Three previous reports(1-3) have dealt with the application of a double extrapolation method based upon the measurement of the amount of successive increases in quench by the Compton edge-half-height method. The first two reports dealt with quench produced by the addition of a quenching agent directly into the sample-liquid scintillation solution. The last report dealt with a method of simulated quench by introduction of an optical filter between the sample and the detector (multiplier phototube). All three of the reports utilized a single multiplier phototube (MPT) detection system; i.e., a non-coincidence system. Because of the high background which results from use of the single MPT, it was necessary to count the sample at various threshold levels and extrapolate to a zero threshold in order to obtain the sample count rate at each quench level. The logarithm of the extrapolated count rate is then plotted as a function of the relative pulse height for the half-height of the Compton edge. Extrapolation of these plots to give a zero relative pulse height ratio would provide the sample disintegration rate. The disintegration rates were determined by this method for samples of the following radionuclides: reference 1 - ^{14}C; reference 2 - ^{14}C and ^{63}Ni; reference 3 - ^{3}H, ^{106}Ru, ^{14}C,

^{95}Nb, and ^{60}Co. The accuracy of the method was ± 2%.

NEW METHOD

This method differs from the previous in that it is accomplished in a coincidence type liquid scintillation counter and does not require the extrapolation of measured count rate to zero threshold at each quench level. The method also requires an instrument with pulse summation which gives a truer representation of the pulse height spectrum. The coincidence technique allows for the threshold to be set at essentially the threshold of detection without including excessive backgrounds. The count rates measured, with the threshold set at zero, will be the same as that obtained by the count rate extrapolation method used in previous methods.

The other requirement for this method is the measurement of a parameter which is indicative of the scintillation efficiency. In previous methods the pulse height equivalent to the half-height of the Compton edge for a gamma ray source was used to monitor the scintillation efficiency of the solution as a function of the quench level (real or simulated). In the present method, a new parameter is measured which is a direct measurement of the scintillation efficiency. This new parameter is called the H-number (H#).(4) The H# measures the pulse height of the inflection point $\frac{d^2\ (CPM)}{d\ (PH)^2} = 0$ of the Compton edge. This inflection point is unique, i.e., there is only one point on the Compton edge for which the second derivative is zero. The use of the H# technique requires the use of a gamma ray source which will produce a Compton distribution that is the result of a single gamma ray energy. One ideal source for producing this type of distribution is ^{137}Cs - ^{137m}Ba which produces the 662 keV gamma rays with no other interfering gamma rays.

A plot of the ratios of the relative scintillation efficiency for a ^{3}H sample with successive additions of a quenching agent vs. the logarithm of the zero threshold count rate is shown in Figure 1. Table 1 lists the actual data obtained from the liquid scintillation counter. A least squares fit of the data to the equation:

$$\log CPM = c + d\ R$$

gives

$$\log CPM = 5.22766 - 0.23595\ R$$

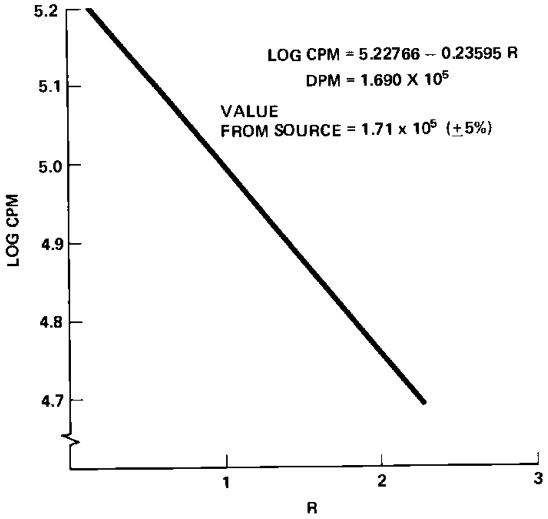

Figure 1. Extrapolation of plot of logarithm of measured CPM in wide open counting channel (zero threshold) vs. relative scintillation yield ratio (R) to obtain DPM of sample. Successive additions of quenching agent produced increased values of R.

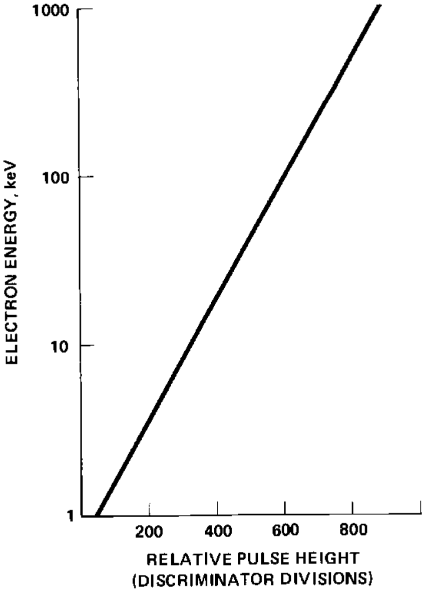

Figure 2. Pulse Height - Energy relationship for the Beckman LS-8000 and LS-9000 Liquid Scintillation Counters.

where c = 5.22766 is the logarithm of the DPM. Thus the DPM of the sample is 1.69×10^5. The actual DPM value calculated from the stock calibration is 1.71×10^5 (± 5%). The value of c is the logarithm of the sample count rate when R is zero. When R is zero, every beta decay will produce a measurable count; i.e., the count rate will be equal to the disintegration rate.

USE OF H-NUMBER

The pulse height-energy relationship has been previously investigated.(5,6) The Beckman LS-8000 and Beckman LS-9000 liquid scintillation counters utilize a logarithmic pulse height conversion leading to a pulse height-energy relationship:

$$\text{PH (pulse height)} = a + b \log E$$

where E is the energy of an electron producing the measured pulse height. For these systems, the pulse height response is shown in Figure 2. If E is expressed in keV, the value of a is the pulse height corresponding to a one keV electron. The equation for the response of this particular instrument used in this investigation (Note 1) was:

$$\text{PH} = 121 + 250 \log E.$$

NOTE 1. Due to use of a prototype LS-8000 for this work, the relationship
for commercially available instruments may be different.

The H# is a measure of the difference in pulse height units of the inflection point of the Compton edge of any sample (PH_q) relating to the inflection point of the Compton edge of an unquenched sample (PH_o):

$$H\# = PH_o - PH_q$$

The response of the two samples is:

$$PH_o = a + b \log E_o$$

$$PH_q = a + b \log E_q = PH_o - H\#$$

Subtracting the second equation from the first gives:

$$H\# = b \log (E_o/E_q)$$

TABLE 1

SAMPLE	ZERO THRESHOLD CPM	RELATIVE SCINTILLATION EFF.	R(a) VALUE
1(b)	98,111	1.000	1.000
2(c)	86,582	0.808	1.237
3	75,341	0.673	1.486
4	60,666	0.534	1.871
5	50,626	0.449	2.229

(a) $R = \frac{\text{Relative Scintillation Eff. of Sample 1}}{\text{Relative Scintillation Eff. of measured Sample}}$

(b) Unquenched sample

(c) Samples 2-5 contain increasing amounts of quench.

where R is defined as:

$$R = E_o/E_q$$

Thus knowledge of b (an instrument parameter) and a measure of H# is all that is necessary to calculate the value of R:

$$R = \text{antilog } (H\#/b)$$

Table 2 lists the values of the DPM of a sample obtained by this method utilizing different values of the slope b. The real DPM value of this sample was 97,370. The value of b is very critical to the accuracy of the method. A known DPM sample can be used to check on the accuracy of the value of b.

Figure 3 shows the application of this method for a series of samples with the same amount of ^3H but different amounts of quench. The value of R for the least quenched sample is not 1.00 because it was not a totally quench free sample. However, this method does not require that the least quenched sample be totally quench free.

SIMULATED QUENCH

This method works equally well when the quench is artifically created by introduction of some optical absorber between the sample and the detectors (MPTs). Flynn, et al.(3) first demonstrated use of simulated quench by use of calibrated filters. However, in the present method any filter material can be used because it is calibrated at the time the zero threshold CPM is measured. This method has great desirability because the sample-liquid scintillator solution remains unaltered. Figure 4 shows the determination of DPM of a ^3H sample using the simulated quench monitored by the H#. The vial was merely wrapped with paper of different color. The paper was typewriter paper of white, pink, blue, and yellow color. The paper was wrapped carefully around the vial and taped in place so as not to come lose or jam the liquid scintillation counter elevator mechanism.

Figure 5 and 6 show the determination of the DPM of ^{14}C samples by this method using the simulated optical and added chemical quench. Figures 7 and 8 show similar plots for determination of the DPM of ^3H-water samples in an emulsion liquid scintillation solution using the simulated optical and added chemical quench techniques.

TABLE 2

THE INFLUENCE OF THE VALUE OF THE SLOPE b ON THE DETERMINATION OF THE DPM OF A ^{3}H SAMPLE. ACTUAL DPM = 97,370

SLOPE b	DPM
267	102,605
258	99,867
250	97,481
240	94,410

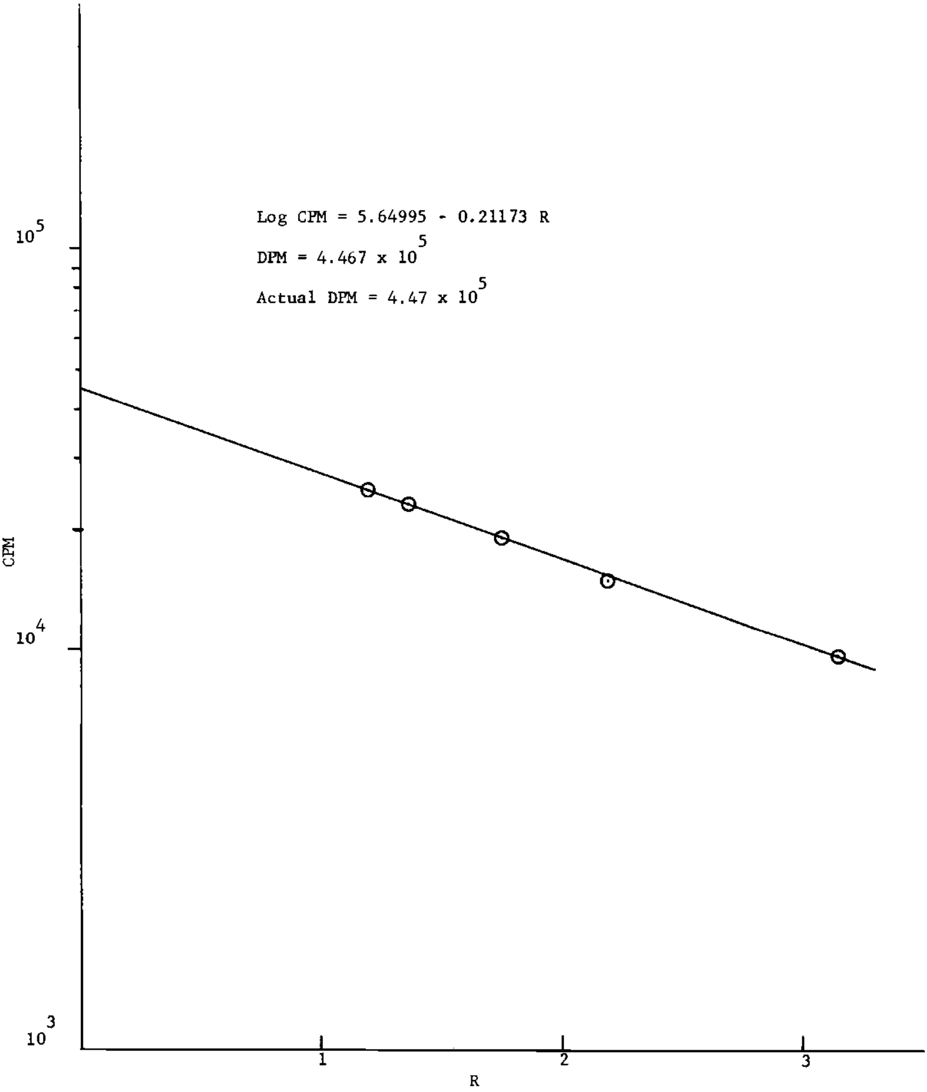

Figure 3. Extrapolation method plot applied to a set of samples with the same amount of ^3H but different amounts of quench. Each R value corresponds to a different counting sample.

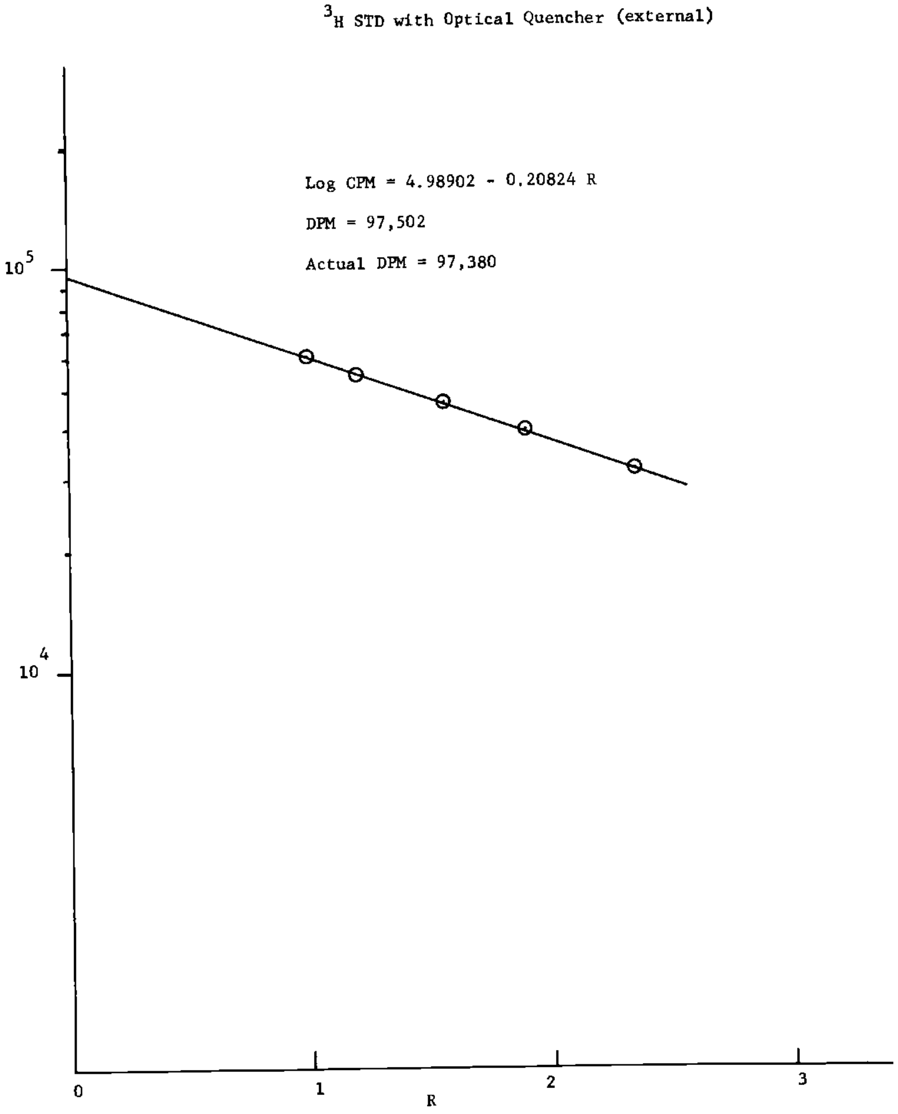

Figure 4. Extrapolation method plot applied to a single sample containing ^{3}H but producing simulated quench by the use of optical filters around the counting sample vial.

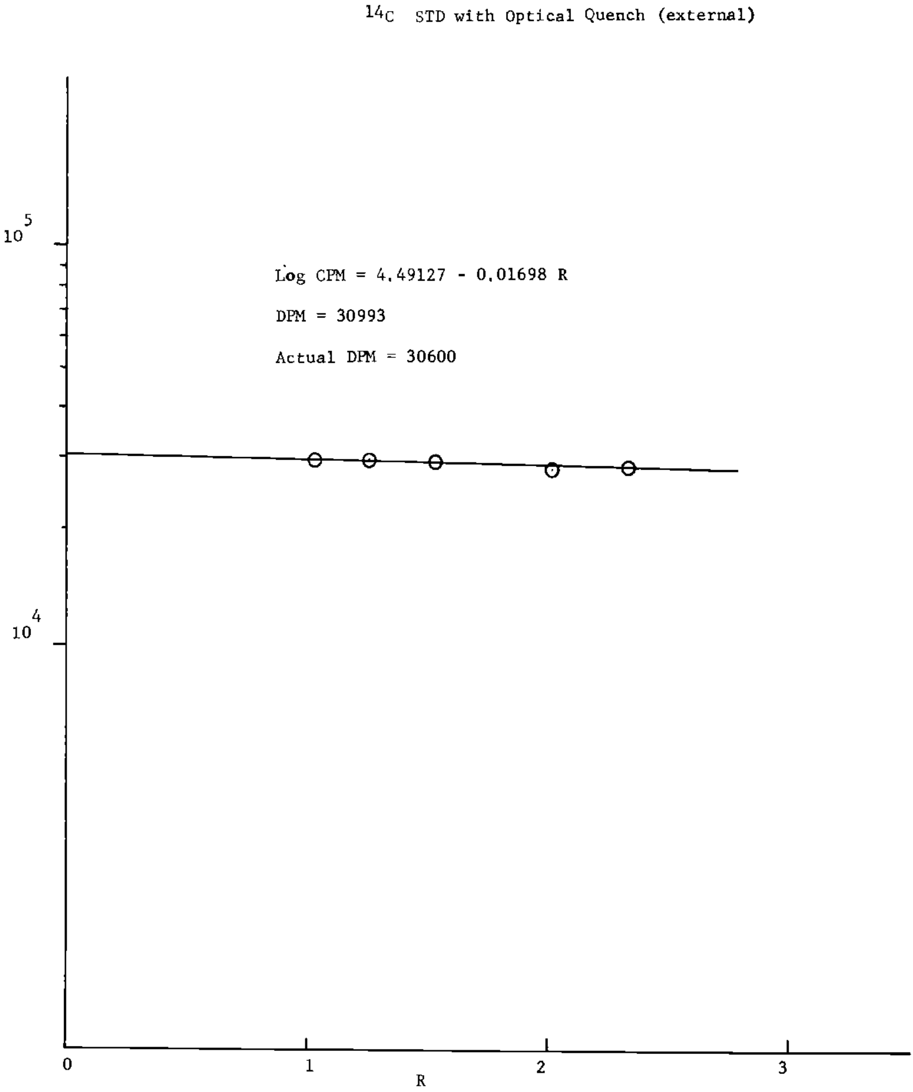

Figure 5. Extrapolation method plot applied to a single sample containing ^{14}C but producing simulated quench by the use of optical filters around the counting sample vial.

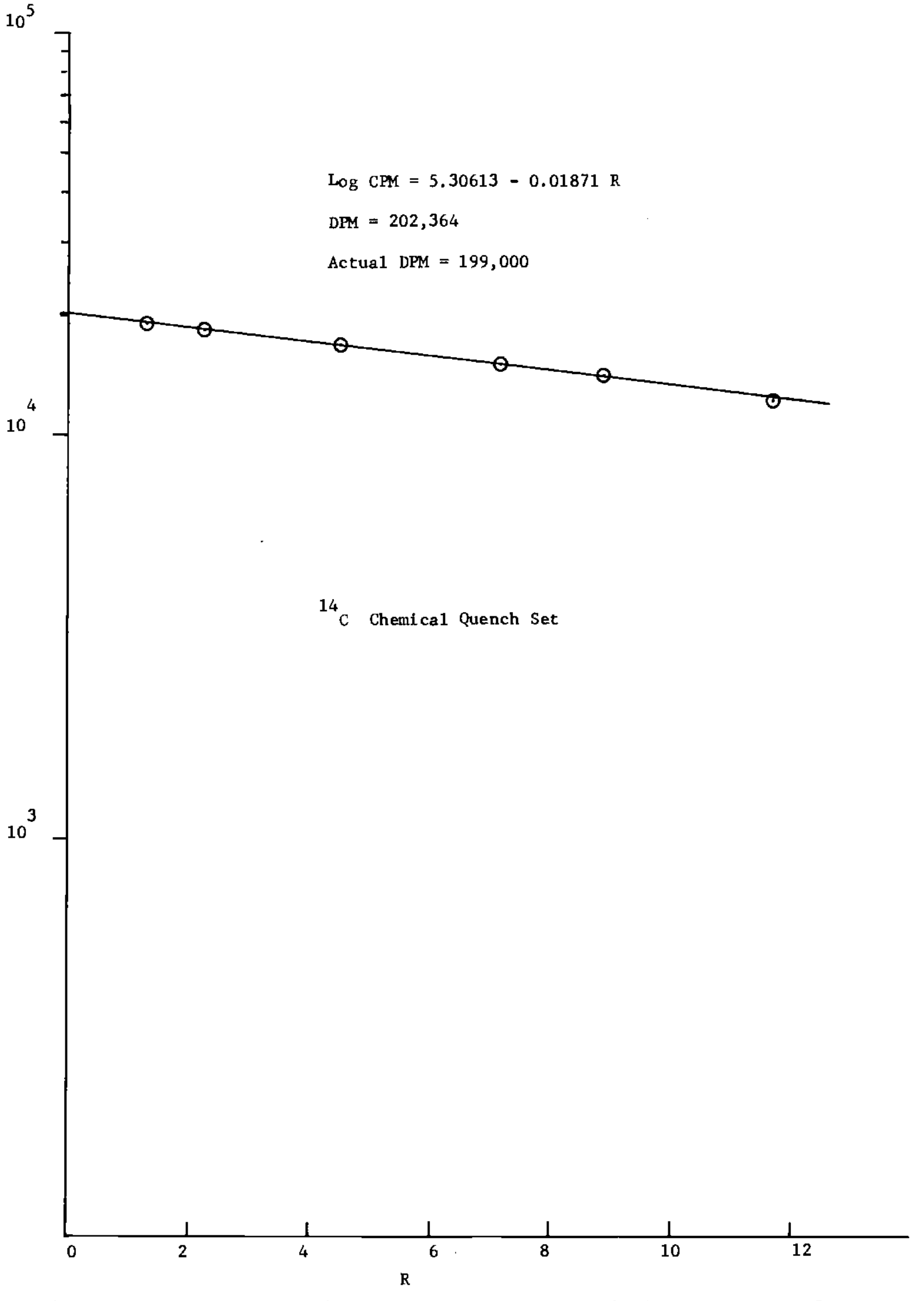

Figure 6. Extrapolation method plot applied to a set of samples with the same amount of ^{14}C but different amounts of quench. Each R value corresponds to a different counting sample.

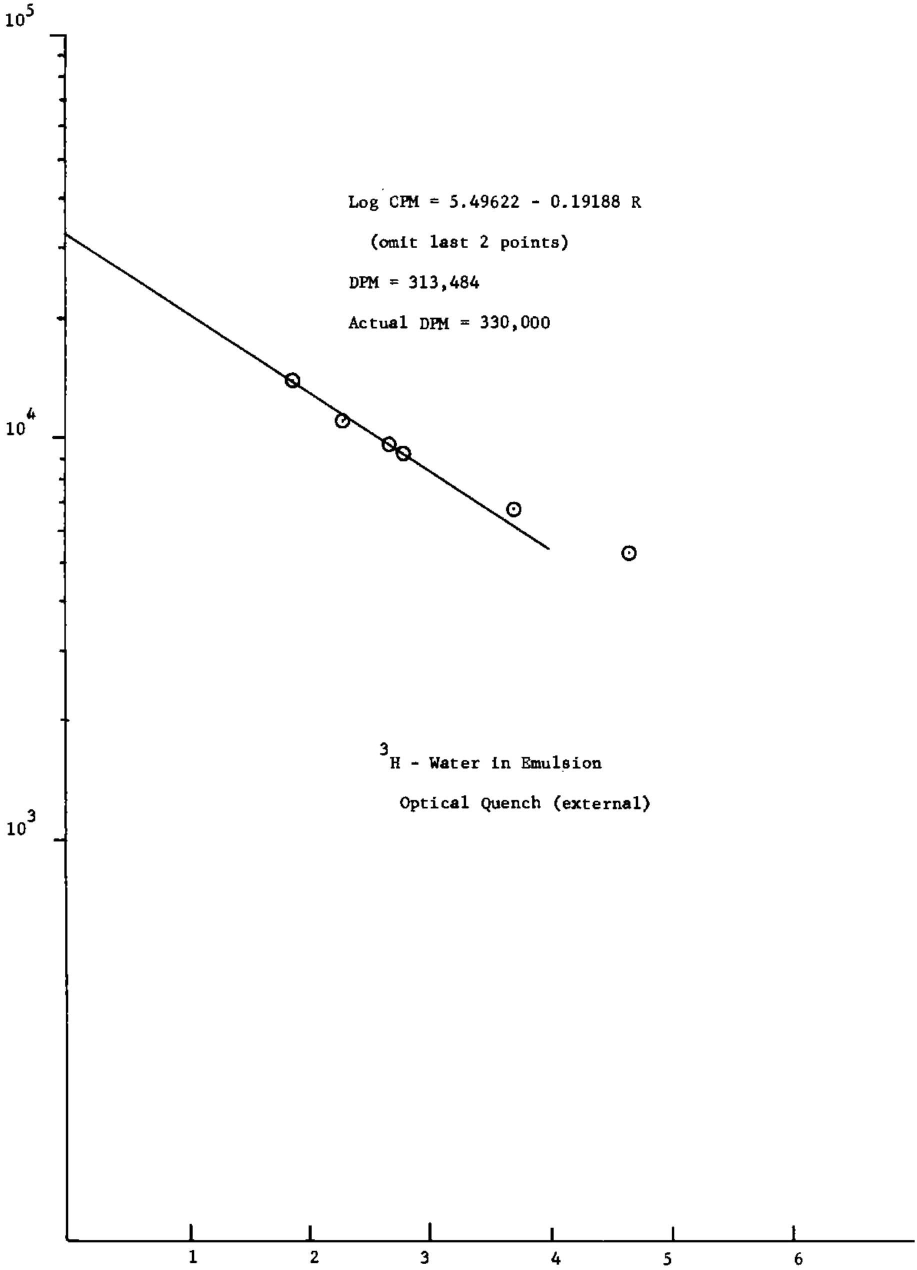

Figure 7. Extrapolation method plot applied to a single sample containing ^{3}H-water but producing simulated quench by the use of optical filters around the counting sample vial.

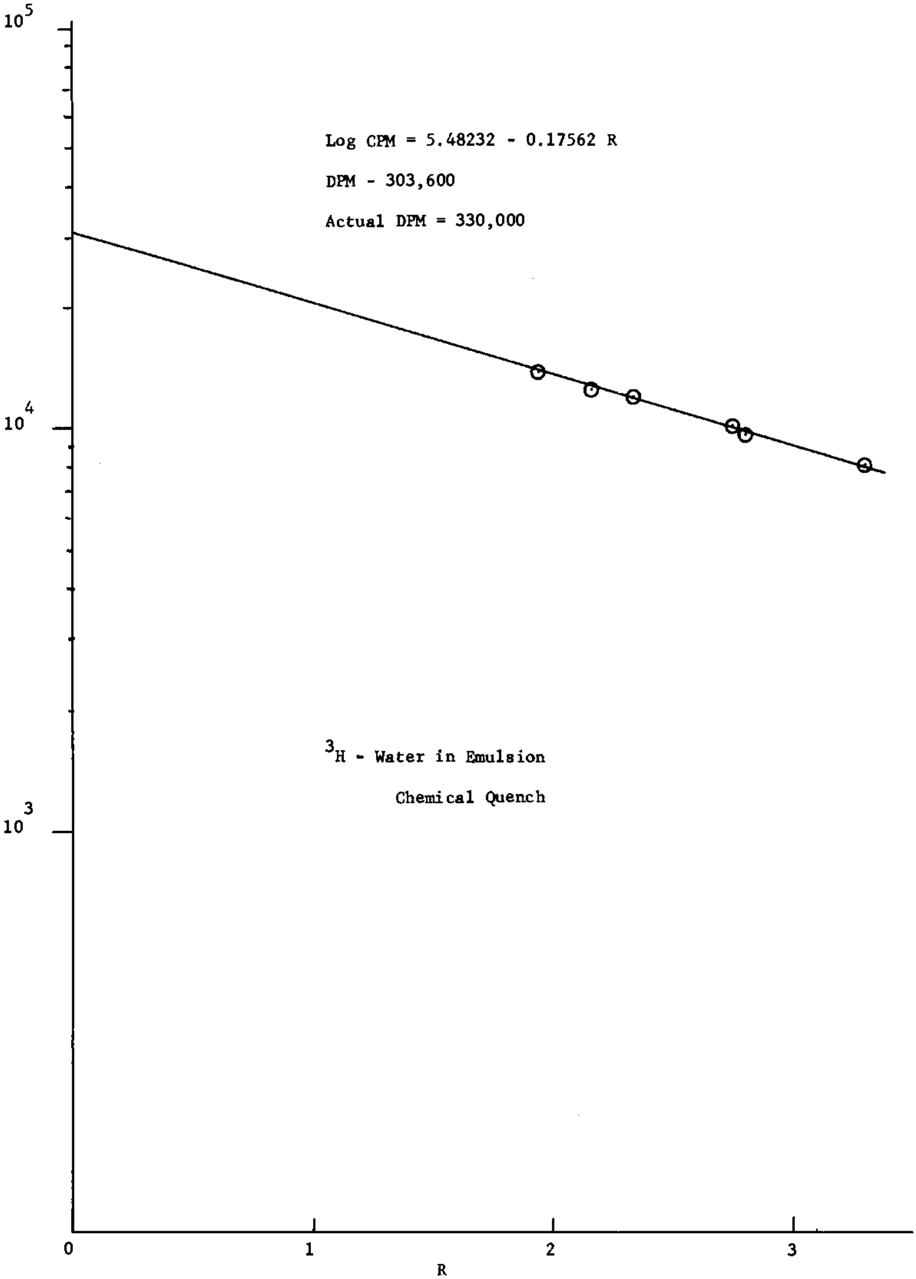

Figure 8. Extrapolation method plot applied to a set of samples with the same amount of ^{3}H-water but different amounts of quench. Each R value corresponds to a different counting sample.

CONCLUSIONS

The combinations of the H# and a coincidence type counter make possible the determination of the DPM of beta emitters by employing a single extrapolation. The method does have some limitations. It is not possible to start with highly quenched ^3H containing samples, i.e., R > 3.0. The method is probably accurate to within ± 3%. Further work will be performed to extend this method to a series of beta emitting nuclides and to further define the accuracy.

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