EXPERIMENT 5.

SCINTILLATION COUNTING AND QUENCH CORRECTION.

(The report for this experiment is due 1 week after the completion of the experiment)

5.1 Introduction

Liquid scintillation is the method of choice to measure beta activity in particular if detection efficiency is important. Actually, for weak beta-emitters ($E_{\beta max} < 0.1$ MeV) it is the only practical way of detection. With other type of detectors, even before reaching the detecting medium, a weakly penetrating (low energy) radiation may be completely stopped by the window of a gas-based detector, or by the envelope of a solid state detector. Whereas in liquid scintillation, the radioactive substance is intimately mixed with the scintillating "cocktail" such that the radiation is emitted inside the detecting material itself.

The principle of scintillation counting is to measure the amount of light emitted from the scintillator as the result of ionizing radiation interacting with the medium. Many factors affect the actual amount of light detected as it relates to the amount of energy deposited in the scintillator. More precisely, light emission can be *quenched* (*i.e.*, the excited scintillator relaxes by transferring it excess energy to some other molecules), it can be absorbed before reaching the outside world (as would be the case for a deeply colored sample) or in some case, extra light is emitted due to *chemiluminescence* (ambient light may be absorbed then re-emitted as phosphorescence under certain conditions). One must be aware of these factors in order to obtain quantitative information from Liquid Scintillation Counting (LSC).

In the present experiment the effect of chemical quenching will be studied. The purpose of the experiment is to construct a *quench correction curve* using standard quenched samples in order to determine the true decay rate of unknown samples. The quench correction curves will be based on instrumental factors known as Single Channel Ratio (CHR) and External Standard Ratio (ESR).

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5.2 Instrumentation

Figure 1. shows schematically the basic components of a modern liquid scintillation counter. A beta particle emitted during the decay of a radionuclide in the scintillating solution will transfer some of its energy to the medium resulting in the production of a number of photons which are allowed to strike the sensitive photocathode of a photomultiplier tube (PMT). The net result is a detectable voltage pulse at the output of the PMT; the height of the pulse is proportional to the number of photons detected which is itself proportional to the amount of energy deposited in the stopping medium. A preamplifier and amplifier turn this small voltage into a workable signal. The diagram shows two PMT's operating in coincidence to ensure that mostly *true* signals are recorded while random background signals are rejected. A pulse height analyzer allows selection of specific voltage range (*i.e.*, energy range) and eventually the numbers of events of interest are recorded on a scaler or other convenient output device.
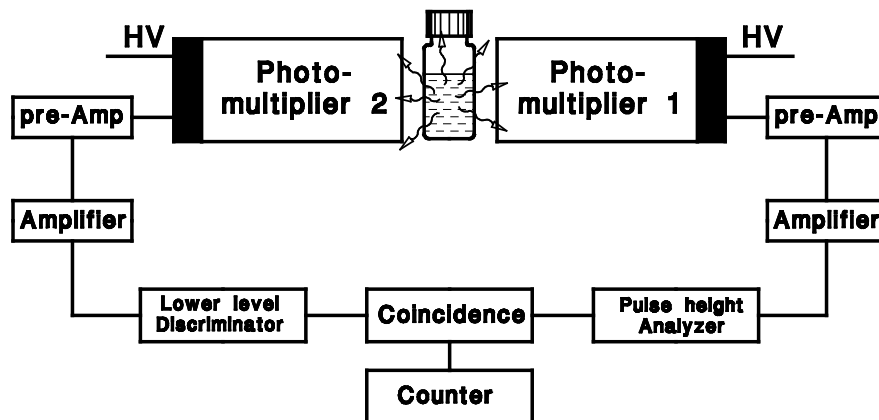

Figure 1. Block diagram of a typical liquid scintillation counter.

Beta particles have a continuous energy spectrum ranging from 0 to $E_{\beta max}$. The voltage pulses resulting from each beta event would show a range of heights depending on the energy deposited during a particular event. Figure 2 pictures such a series of voltage pulses reaching the analyzer as a function of time. This string of pulses could be sorted out with a multichannel analyzer according to their height, and then the result plotted as number of

occurrences of a certain pulse height versus pulse height. One would obtain a histogram of pulse height occurrences which is equivalent to the energy spectrum shown in Fig. 3.

Note that the beta spectrum thus obtained is different for different radionuclides; also one should appreciate that the conversion efficiency of beta energy to photons is dependent on the solvent and scintillating material. Consequently, the pulse height spectra will somewhat vary from one type of scintillation cocktail to another.
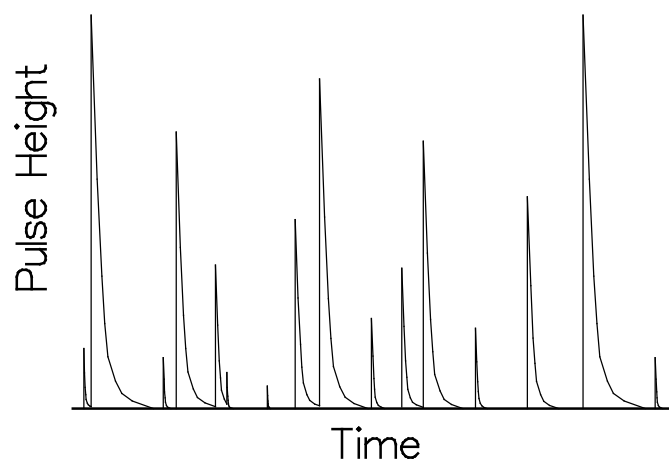

Figure 2. Schematic diagram of the time profile of a string of signals out of the amplifier; each voltage spike (pulse) corresponds to a decay event exciting the scintillating liquid.

Commercial LS counters can be set to merge into "windows" selected ranges of pulse heights to be counted. For example, in Fig. 3, window A and window B represent possible range settings if one were to measure ^{3}H or ^{14}C activities respectively. It is clear that if the lower level is set to a high value, or the high level to a low value, some *good* pulses will not be counted. Thus the counting efficiency, defined as

$$\text{Efficiency} = \frac{\text{cpm}}{\text{dpm}} \tag{1}$$

where cpm is the Counts Per Minute detected and dpm is the true number of Disintegration Per Minute occurring in the sample, is dependent on the window settings.

In general, the settings of the detecting windows are not chosen to include the entire pulse height spectrum, but rather to achieve maximum sensitivity for a particular radionuclide. To determine the best energy window setting, one needs to calculate for each setting some kind of performance parameter – the figure of merit. The E^2/B is one parameter commonly used for this purpose and is defined as:

$$\text{figure of merit} = \frac{E^2}{B} = \frac{(\text{percent efficiency})^2}{\text{background (cpm)}} \tag{2}$$

As it turns out, the optimum E^2/B is obtained for the energy window upper level set just below the cut-off point of the pulse height spectrum; above this point background counts make the major contribution to the spectrum. The lower level setting is found by increasing its value from zero to a point where the background counts are insignificant compared to the loss of sample counts. To be rigorous, the optimum E^2/B should be determined for different experimental conditions; it depends in particular on the overall count rate, type of fluor and degree of quenching.
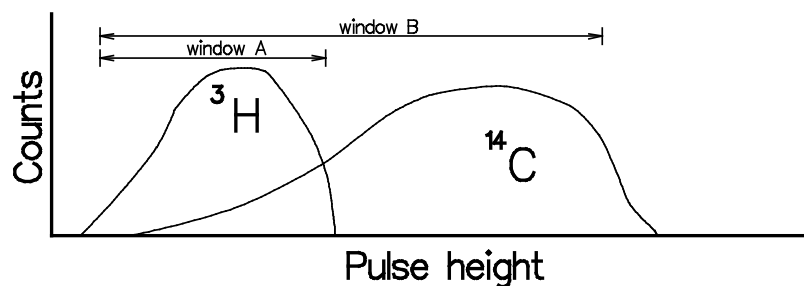

Figure 3. Pulse height spectra for ^{3}H and ^{14}C showing possible energy window settings suitable for activity measurement of unquenched samples.

Practically, for samples with an appreciable count rate a simpler method of setting the energy window can be adopted. For a given radioisotope, the upper level is set to some arbitrary value just below the cut–off point. The lower level is then set to a value ($\approx 2\%$ full scale) which just discriminates against the small electronic noise signals inherent to the system.

Modern scintillation counters offer in general a choice of preset energy windows optimized for common type of radionuclides.

5.3 Channel Ratio Method of quench correction (CHR)[1]

In the presence of quenching agents, for each decay in the scintillating solution either the number of photons produced is reduced (chemical quench) or the number of photons actually reaching the PMT is reduced (color quench).   Both processes reduce the pulse height corresponding to a particular decay event; the net result is a shift of the energy spectrum towards lower energy as is illustrated in Fig. 4. It is apparent that for a fixed window setting, the number of counts recorded (*ie* the counting efficiency) will diminish with increasing quench. The relationship between the pulse height spectrum shift and the counting efficiency reduction are the basis for the quench correction method known as the channel ratio (CHR).

The variation of the pulse height spectrum with increasing quench is determined by setting two counting windows. In Fig. 4., window B is set to record the total unquenched spectrum. Window A is set to accept only the lower end of the spectrum ($\approx$ 30% of the full unquenched spectrum). As the energy spectrum shifts to lower energy with increasing quench, the ratio of counts in A over counts in B (the CHR value) will increase from some fractional value to a maximum value of one.   An empirical quench correction curve can be constructed by measuring the count rate of a set of samples containing the same known activity but varying amount of quench. The results are plotted as counting efficiency versus CHR value. The true decay rate of an unknown sample can be obtained by reading off the plot the counting efficiency corresponding to its measured CHR value. This method of quench correction can be used for a wide variety of quenched samples. However the method looses accuracy with low activity or highly quenched samples.

5.4 External Standard Methods of quench correction (ESR)[2]

In this type of methods, first the count rate of a sample is obtained in the usual manner, and then the sample is again measured in the presence of an external gamma source (^{137}Cs, or

[1] Depending on instrument, this parameter may be labeled as IC#

[133]Ba, or [226]Ra). The amount of quenching can be obtained from the shape of the energy spectrum of this second measurement.

   One of the possible interaction processes of gamma rays with matter is the production of Compton electrons; they arise from a partial transfer of energy from the gamma photon to an orbital electron. For the present purpose, Compton electrons produced in the scintillator are indistinguishable from beta particles of the same energy and accordingly will give rise to scintillations and a corresponding energy spectrum (Fig. 5.).
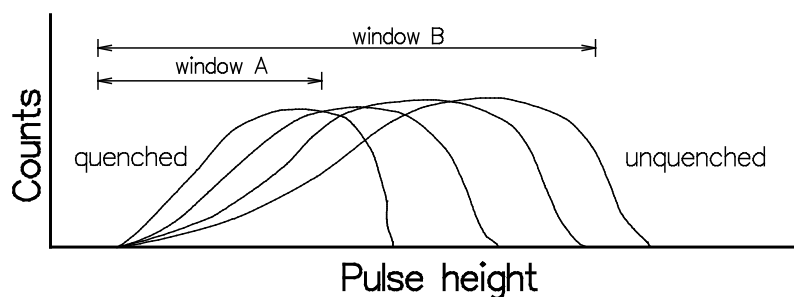

Figure 4. Shift to low energy of the pulse height spectrum with increasing quench; possible window settings are shown for Channel Ratio method of quench correction.

   The net photon output from the Compton electrons will be the result of the same quenching effects as that from the beta particles due to sample decay. Therefore the Compton spectrum will be shifted to lower energy as quenching increases. One way to quantify the amount of quenching is to take the ratio of counts recorded in two predefined energy windows (window C and D in Fig. 5). As in the CHR method, a correction quench curve is constructed by plotting the observed counting efficiency of a set of samples of same known activity but varying quench, versus the observed C/D ratios (ESR values). For an unknown sample the ESR value can be measured then used to obtain the corresponding counting efficiency from the standard quench curve.

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[2] Depending on instrument, this parameter may be labeled H#

This technique has the advantage of producing very high scintillation rates and therefore precise numbers may be obtained in short counting times. It is a non-invasive technique and no special preparation of the sample is required. Also by taking the ratio of counts in two channels, volume changes or variation in sample or vial position are not critical because these factors are expected to affect both channels proportionately and cancel each other when the ratio is taken.
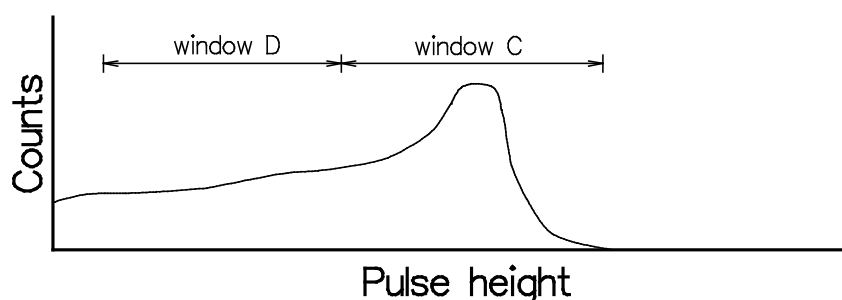

Figure 5. Typical pulse height spectrum of Compton electrons resulting from gamma irradiation of a liquid scintillation sample.

5.5 Other External Standard quench correction methods

Two other features of the Compton energy spectrum have been used to characterize the degree of quench of a sample.

5.5.1 H#.

The Compton spectrum shows an inflection point near its high energy cut-off whose position in energy (the so-called H#) varies in a systematic way with amount of quenching. Modern electronic allows to locate precisely this point from an on-line calculation of the second derivative of the spectrum; this quantity vanishes to zero at the inflection point. This method is a proprietary technique of BECKMAN instrumentation.

5.5.2 EXTERNAL STANDARD PULSE (ESP)

Here the parameter used to quantify the degree of quenching is the weighted average pulse height of the Compton spectrum; again a fast processor allows on-line calculation of this parameter.

Modern scintillation counters have automated many of the procedures necessary for sample handling and quench correction. In particular, the quench correction curve can be stored in the machine to obtain the appropriate interpolation and calculate the true dpm of an unknown sample. However, these correction curves need to be checked regularly to account for systematic drift in the machine performance over time.

5.6 Procedure

WARNING !! ³H *and* ¹⁴C *are beta emitters the energies of which are so low that the corresponding radiation will not (hardly) penetrate the thin window of the GM detectors and will not (hardly) be detected. Only swipe tests can show the level of contamination of your working space.*

Review the technique for using automatic pipette properly (see Expt. 1) as the quality of results is entirely dependent on the accuracy and reproducibility of your pipetting.

Quench curves will be obtained for both ³H and ¹⁴C. Except for the isotope used and the programs used on the LKB scintillation counter, the procedures for each determination are the same.

- Prepare and mark for identification six scintillation vials.

- Add 4 ml of scintillation cocktail to each vial,

- Pipette accurately into each vial the *same* aliquot of standard tracer (your instructor will specify the exact amount to use), *cap tightly then shake thoroughly* to mix the radioactive tracer with the cocktail.

NOTE: the sources provided are calibrated standard solutions and are expensive. Also other groups are relying on the accuracy of the information provided with the source to calibrate their instrumentation; handle these sources with respect. Do not forget to record in your note book all the information relevant to these standard solutions, in particular the <u>chemical identity</u>, the <u>density</u>, the <u>specific activity</u> and the <u>reference date</u>.

– If time permits, dark adapt the samples for ≈ 5 min, then measure the activity on the LKB counter; the instructor will tell you which program to use.

- Verify that the *cpm* of the six samples agree within 4 standard deviations; re-prepare and recount samples which deviate too much.

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- Once you are satisfied that you have six samples containing the same amount of radioactive tracer, add to each sample different amount of quenching agent (CCl_4 in the present case), cap *tightly* then *shake well*. For ^3H, use the following volumes: 0, 10, 20, 50, 100, 200 μl; for ^{14}C use 0, 10, 20, 50, 100, 150 μl.

– Measure the *apparent* activity of the quenched samples on the LKB counter using first a program which outputs the CHR values, then one which gives the ESR values. Measure also the activity of the unknown samples provided[3]. In addition a background sample containing only scintillation liquid with no added activity should be included.

- Finally measure the activity your unknowns and background sample with programs with a built-in quench curve already loaded in the LKB counter.

- Perform contamination tests of your working area and record the results.

5.7 Data analysis

First tabulate clearly all the data collected during the experiment along with the corresponding error estimates and relevant quench parameters according to the Excel spreadsheet template available from the course web site (LSC_results.XLS from *www.sfu.ca/~brodovit/files/nusc346/templates/* ; hand in a copy of this file with your report).

From your data, construct quench correction curves as follows:

1. Determine the %counting efficiency for each of the sample by taking the ratio of the measured count rate to the true decay rate (in the case of ^3H, the actual dpm of the standard may need to be corrected for radioactive decay since the reference date). Plot %counting efficiency versus CHR for ^3H and ^{14}C,

2. Same thing for the ESR set then plot %counting efficiency versus ESR for ^3H and ^{14}C,

3. If possible, parameterize the curves with some curve fitting procedure or just hand draw a smooth curve through the experimental points.

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[3] To sample the unknown, inject 100 μl into 4 ml of LSC cocktail; do not forget to record the unknown ID.

4. Use these curves to obtain first the true activity of your unknowns in the scintillation vial, and then report the true activity of your unknowns in Bq l^{-1}.

5. Compare your results with the ones obtained using pre-loaded program. Comment on the accuracy (or lack of) of the different methods, in particular when dealing with highly quenched samples or lightly quenched samples. The suggested volumes of CCl_4 were chosen to obtain a convenient range of quenching. What adjustment would have to be made to the procedure if acetone was used as a quenching agent instead of CCl_4? Could the same quench curves be used if one were to switch to larger (10 ml) size vials (hint: consider how the quenching parameters CHR, SCR, IC# or H# are defined)? How could the window setting be adjusted to improve the accuracy of determination of highly quenched samples? Estimate the 50% MDA (Minimum Detectable Activity within a 50% confidence level) with the present detection conditions; justify your answer (you may want to check the SFU radiation Safety manual - *http://www.sfu.ca/radiation-safety/toc.htm* - which contains some practical information on counting statistic).

Hand in a copy of your data entered in the spreadsheet LSC_results.XLS.

References

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