

BALANCED WINDOW METHOD IN ^{14}C LIQUID SCINTILLATION COUNTING

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ABSTRACT. The authors present a detailed theoretical and experimental study of the liquid scintillation balanced counting method, widely used in radiocarbon dating, using a simple, laboratory-made system. A fixed counting window becomes a balanced window when the high voltage is set where the ^{14}C count rate rises to a maximum. Using a measured ^{14}C pulse height spectrum, we have calculated the lower and upper limits for 11 balanced windows of varying width and their respective counting efficiencies. Furthermore, we have studied: (1) theoretically and experimentally, the counting efficiency for up to a $\pm 15\%$ shift in pulse height from the balanced setting, (2) the change in pulse height due to temperature variations, (3) the long-time stability of the system, and (4) a method that allows a quick determination of the balance voltage for individual samples, using the Compton spectrum of ^{133}Ba . The standard deviation for thirty 24-hr measuring periods for a ^{14}C standard (190 Bq) was within the expected statistical standard error (0.03%).

INTRODUCTION

In high-precision radiocarbon dating, the ^{14}C counting efficiency must be highly stable over the weeks or months it takes to count standards and to date samples. When using a liquid scintillation (LS) counter, the most effective way to attain this is to apply the balanced window method, where the high voltage is set at the maximum ^{14}C count rate in the system's fixed counting window. Pearson (1979, 1983) used the balanced window method in his pioneering high-precision LS dating, McCormac has studied it in some detail with a 2-photomultiplier tube (PMT) system (1992) and Arslanov (1991) discusses the method in some detail in his book on ^{14}C dating. The present study is an extension of their work, in which we strengthen the basis of this technique.

METHODS

We use a laboratory-built, single PMT system (ICEL-C14) specially designed for ^{14}C dating with an emphasis on simplicity. It consists of a detector unit and a compact, dedicated electronic unit connected to a laptop computer, which controls the counting operations and processes the data.

A semi-spherical, 3-ml, quartz vial sits on top of a vertical, 30-mm diameter phototube (Theodórssón 2000). The amplifier of the electronic unit is followed by 4 pulse height discriminators (D1–D4) with inputs connected in parallel order. Their state is read by the computer and when activated, 1 count is added in the respective internal pulse summing registers (N1–N4) of the computer.

D1 and D3 define the lower and upper limits of the ^{14}C counting window, denoted by $D(L)$ and $D(U)$. The number of pulses in the ^{14}C window is $N(L) - N(U)$. In the following, L (lower) and U (upper) will refer to discriminator voltages, the corresponding counting registers, or to channel numbers of the ^{14}C spectrum. D2 and D4 are set at $0.60D(U)$ and $1.25D(U)$, respectively, and are denoted by $D(M)$ and $D(H)$. Their use will be discussed in the following. We have set the $D(L)$ at 0.9492 V and the $D(U)$ at 5.088 V, giving $h = 5.36$.

RELATIVE GAIN VERSUS HIGH VOLTAGE AND COUNTING EFFICIENCY

In our studies, we use a laboratory-prepared ^{14}C standard (190 Bq in 3 ml of benzene, to which we added 16 g/L of butyl-PBD). We first measured, at varying high voltage (HV), the ^{14}C activity with the ICEL-C14 for the determination of the counting efficiency (ε) given by:

$$\varepsilon = [N(L) - N(U)] / T / 190 \quad (1)$$

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where T is the counting time in seconds and 190 is the activity of our ^{14}C standard (Bq). ϵ rises to a maximum at 533.8 V, becoming the balance voltage, denoted as HV_{bal} (Figure 1). At this voltage, the fixed window is called the *balanced widow*. A shift in the pulse height spectrum will then have a minimal effect on the ^{14}C count rate.

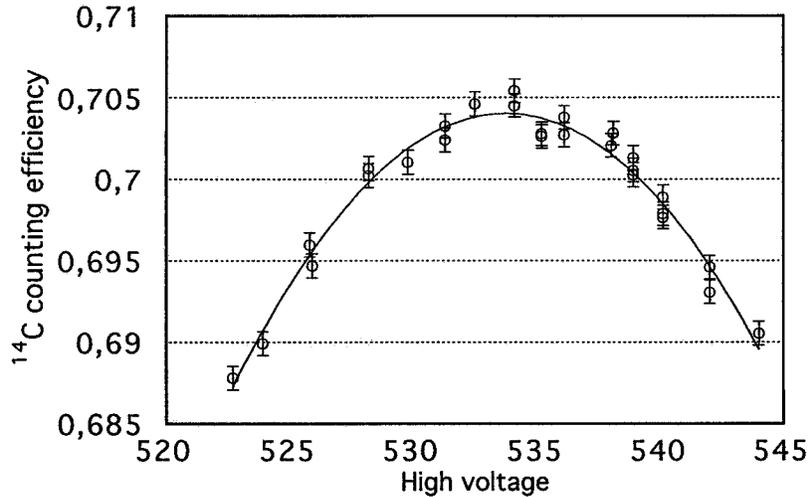


Figure 1 Measured ^{14}C counting efficiency versus high voltage. The solid line is a parabolic fit.

Simultaneously with these measurements, we measured the ^{14}C pulse height spectrum with a multi-channel analyzer. From each spectrum, we determined (with interpolation) the channel where the number of counts has fallen to 10% of that in the peak channel (Figure 2). There is a linear relationship between this channel number, $Ch(10\%)$, and the HV :

$$Ch(10\%) = -1125.1 + 2.402HV \tag{2}$$

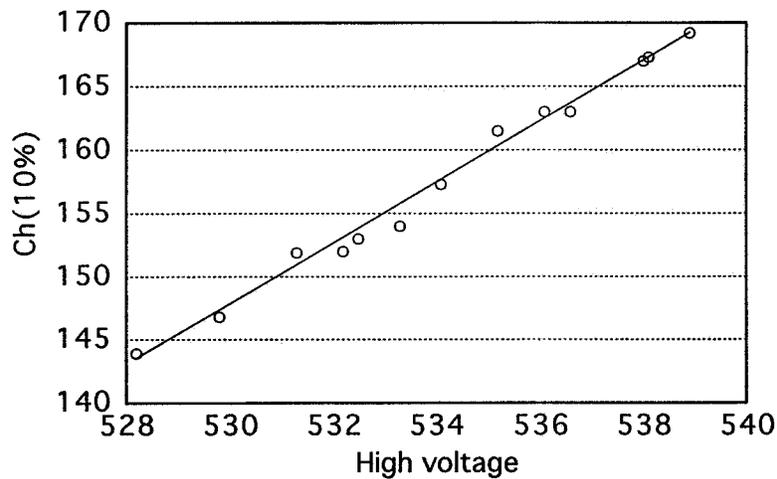


Figure 2 The channel number where the count rate has fallen to 10% of the maximal count rate versus the high voltage

where $Ch(10\%)$ is a measure of the gain and depends on the system used. In the following, we use another gain parameter, the relative gain (RG), which is linearly dependent on $Ch(10\%)$ and is instrument independent. We set the $RG = 1.00$ at HV_{bal} , where $Ch(10\%)$ has a value of 157.1. Using Equation 2, we get:

$$RG = -7.162 + 0.01529HV \quad (3)$$

This shows that when the high voltage changes by 1.0 V, the RG changes by 0.015.

Using a parabolic fit for the measured counting efficiency (Figure 1), we have calculated the relative counting efficiency (ε_{rel}) defined by:

$$\varepsilon_{\text{rel}}(HV) = \varepsilon(HV) / \varepsilon(HV_{\text{bal}}) \quad (4)$$

Figure 3 displays the deviation of this parameter from its maximum value (1.00) as a function of RG . Since the curve is symmetric at about $RG = 1.0$, only the part above 1.0 is shown. The figure shows that if the RG shifts 3.5% (corresponding to a change of 2.3 V in the high voltage) from the balance point (RG from 1.00–1.035 or 0.966–1.00), the decrease in the count rate will be 0.1%, corresponding to 8.0 ^{14}C yr, a shift that can be tolerated in most cases.

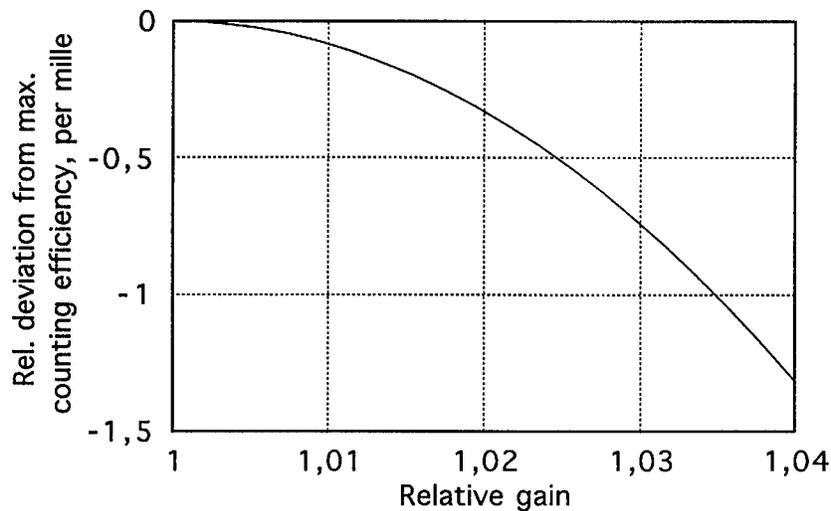


Figure 3 Calculated relative deviation from maximal counting efficiency versus relative gain

^{14}C BALANCED WINDOW

In order to calculate the lower and upper limits of a balanced window, an experimentally-determined ^{14}C pulse height spectrum is needed, which we measured with our ^{14}C standard for 5.0 days (Figure 4). To explain the principle of a balanced window let us assume that in Figure 4, $D(L)$ is at channel 20 and $D(U)$ is at channel 120. If a pulse height shift occurs where each pulse is attenuated by 5%, the pulses in channel 20 will not be registered by $D(L)$, nor the pulses in channels 121–126 by $D(U)$. If these 2 changes are equal, the number of pulses in the ^{14}C counting window, $N(L) - N(U)$, will not be affected by this shift. A ^{14}C counting window of this type is called a balanced window.

We denote the ratio of channel numbers of the upper- and lower-window limit [$Ch(U)$ and $Ch(L)$] by h :

$$h = Ch(U) / Ch(L) \quad (5)$$

For a selected value of h , the general condition of a balanced window can be described by the following equation:

$$F(L) = hF(U) = hF(hL) \tag{6}$$

where $F(\text{Channel number})$ is a continuous function describing the measured ^{14}C spectrum versus the channel number and L and U are the channel numbers at the lower and upper limits of the balanced window. For a given value of h , Equation 6 unambiguously determines the window limits and the counting efficiency.

When we calculate the limits of a balanced window, we use a 6th degree approximation for the measured ^{14}C spectrum (Figure 4). Using Equation 6 for 11 values of h , we calculated the corresponding values of L and U (channel numbers, Table 1). The detection efficiency (ϵ) was then calculated for the balanced windows using the following equation:

$$\epsilon = \int_L^U F(\text{Ch})d\text{Ch} / 190 \tag{7}$$

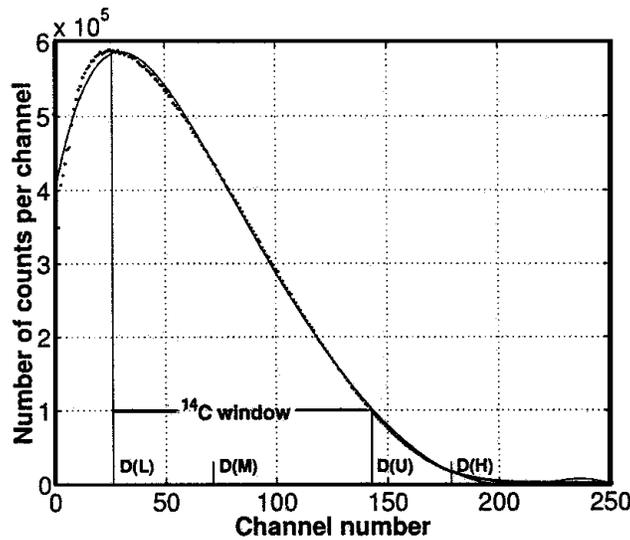


Figure 4 The pulse height spectrum of a 190-Bq ^{14}C standard measured for 5.0 days. The solid line is a 6th degree fit to the measured spectrum. The discriminator settings for $h = 5.36$ are shown on the x-axis.

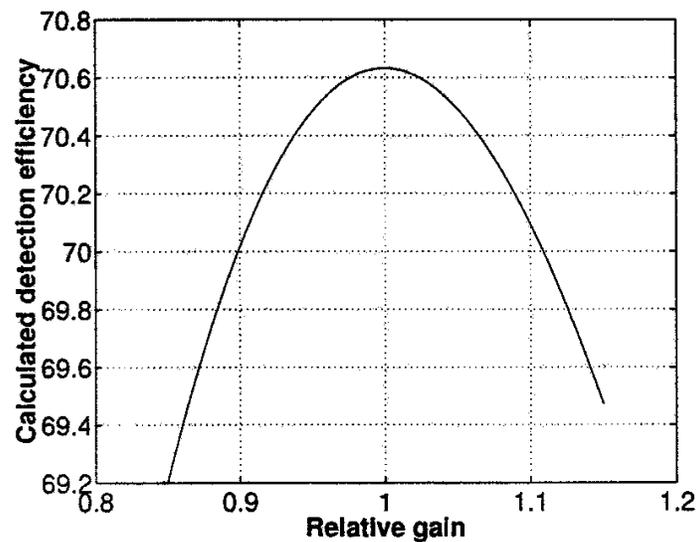
The results of these calculations are shown in Table 1. Furthermore, we have calculated the counting efficiency for a varying shift in gain for $h = 5.36$ (Figure 5).

Table 1 Limits of balanced windows and counting efficiencies

| h | D(L) Channel number | D(U) Channel number | Counting efficiency | Ch(L) keV | Ch(U) keV |
|-----|---------------------|---------------------|---------------------|-----------|-----------|
| 4.0 | 32.7 | 130.9 | 0.622 | 30.7 | 105.7 |
| 4.5 | 30.0 | 135.1 | 0.658 | 28.6 | 108.8 |
| 5.0 | 27.7 | 138.7 | 0.688 | 26.7 | 111.4 |
| 5.5 | 26.0 | 142.9 | 0.713 | 25.4 | 114.6 |
| 6.0 | 24.1 | 144.6 | 0.734 | 23.9 | 115.8 |
| 6.5 | 22.8 | 148.2 | 0.752 | 22.9 | 118.5 |

Table 1 Limits of balanced windows and counting efficiencies *Continued*

| h | D(L) Channel number | D(U) Channel number | Counting efficiency | Ch(L) keV | Ch(U) keV |
|-----|---------------------|---------------------|---------------------|-----------|-----------|
| 7.0 | 21.3 | 149.3 | 0.768 | 21.7 | 119.3 |
| 7.5 | 20.3 | 152.5 | 0.781 | 20.9 | 121.6 |
| 8.0 | 19.1 | 153.1 | 0.793 | 20.0 | 122.1 |
| 8.5 | 18.4 | 156.0 | 0.804 | 19.4 | 124.1 |
| 9.0 | 17.4 | 156.3 | 0.813 | 18.6 | 124.4 |

Figure 5 Calculated counting efficiency for a balanced window corresponding to $h = 5.36$ for the varying drift in relative gain

ENERGY CALIBRATION

In order to transform the calculated upper and lower channel number limits of the balanced windows (Table 1) to the corresponding energies of the ^{14}C beta particles, we have energy calibrated the system. Due to ionization quenching, the size of the amplified pulses is not strictly proportional to the energy deposited by the ^{14}C beta particles in the scintillator. This quenching increases with decreasing beta energy (Horrocks 1979). At HV_{bal} , we have determined the channel number at 8 different beta particle and electron energies. The result is shown in Table 2 and Figure 6, where we also show points based on similar measurements made by Horrocks (1979). This calibration curve is used to determine the energy limits (keV) of the 11 balanced windows (see Table 1).

Table 2 Energy calibration of the LS counting system

| | keV | Channel number |
|----------------------------------|-------|----------------|
| X-rays of ^{133}Ba | 31.0 | 31.3 |
| Gamma from ^{119}Sn | 31.0 | 32.0 |
| Gamma from ^{241}Am | 59.5 | 65.3 |
| 1% of ^{14}C spectrum | 127.9 | 165.5 |
| 3% of ^{14}C spectrum | 116.6 | 146.5 |
| 10% of ^{14}C spectrum | 95.7 | 118.2 |
| Max. of ^{14}C spectrum | 22.8 | 25.0 |
| End of ^{14}C spectrum | 156.0 | 182.0 |

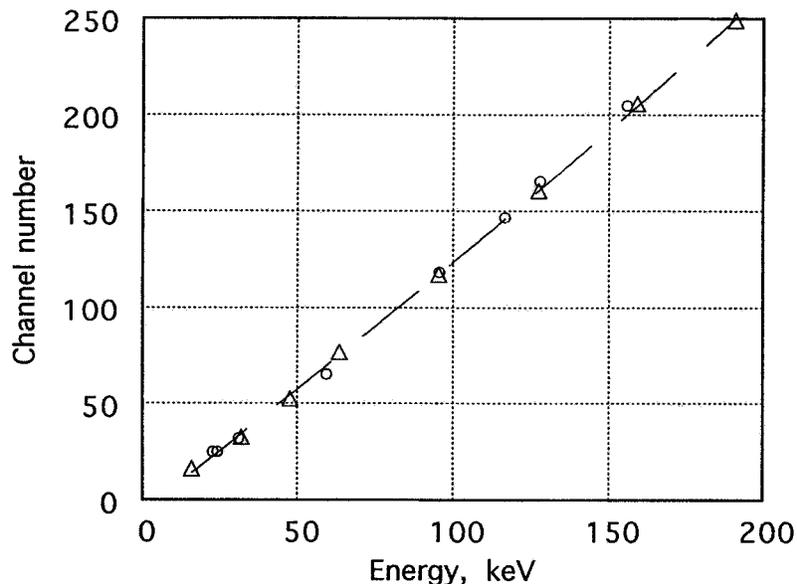


Figure 6 Energy calibration of the LS system at the balance high voltage (HV_{bal}). Circles are our measured points, triangles are measurements made by Horrocks (1979) fitted to our y-scale. The line is a 2nd order fit to the measured points.

SETTING AND CHECKING THE BALANCE POINT

It is very useful to have a quick and reliable method for determining the HV_{bal} for each sample to be counted, so that the eventual light quenching, vial variation, or vial eccentricity on the photomultiplier tube is corrected for, and also for checking an eventual drift from initial pulse gain at the end of a counting period. For this purpose, we use the Compton spectrum of an external ^{133}Ba gamma source (about 2 μCi), which has a dominating gamma line at 356 keV (Compton edge 207 keV). The ratio b , where:

$$b = N(H) / [N(M) - N(H)] \quad (8)$$

is a good measure of the relative number of pulses in the high part of the Compton spectrum and, therefore, a good indicator of a shift in relative gain (Figure 7). $N(H)$ and $N(M)$ are the number of pulses that have activated discriminators D2 and D4.

This is, in principle, the same method that Pearson (1983) used for checking the gain for each counting sample, except that he did not use it to correct the high voltage, but only to check that the shift in gain for individual samples was within a given tolerance limit.

At the balance point (533.8 V) and with our discriminator voltage setting, b has a value of 0.86. For fixed ratios of the discriminator voltage, b is presumably independent of the system used. Measuring the ^{133}Ba standard for 2 min gives a relative gain accuracy of 0.01 through the value of b , which corresponds to less than a 0.1% shift in the count rate at the balance setting. If b deviates from 0.86 for a sample to be measured, the high voltage is corrected.

PMT GAIN VERSUS TEMPERATURE CHANGES

Figure 3 shows that the ^{14}C counting efficiency in a balanced window changes less than 0.1% (corresponding to 8.0 ^{14}C yr) for a $\pm 3.5\%$ shift in the pulse height spectrum from the balance point.

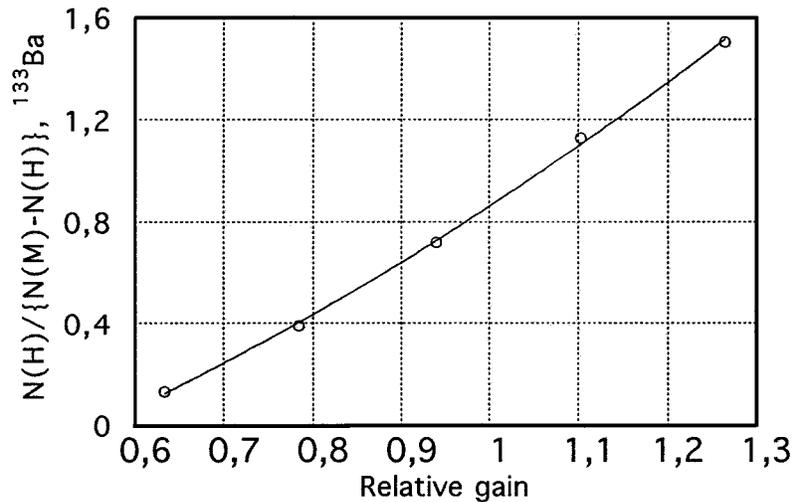


Figure 7 Parameter $b = N(H) / [N(M) - N(H)]$ versus relative gain for an external ^{133}Ba gamma source. The solid line is a 2nd order fit.

Using modern LS counting systems, the change in the ^{14}C count rate in a balanced window due to any likely drift in high voltage, electronic amplification, or discriminator voltages is negligible. In fact, a significant shift in the pulse height is only to be expected from a drift in internal PMT amplification due to a change in temperature.

We have, therefore, studied this effect. The PMT was cooled and heated a few $^{\circ}\text{C}$ below and above the mean ambient temperature. The influence of these variations on the internal gain was determined by the shift of the Compton edge of an external ^{137}Cs gamma source at HV_{bal} using a multichannel analyzer. From the results (Figure 8), we find that the relative internal gain of the PMT decreases linearly by -0.0046 per $^{\circ}\text{C}$. If we want to keep the decrease in counting efficiency within 0.1% from its balance point value, the temperature should be kept within ± 8 $^{\circ}\text{C}$ from the initial temperature, a condition not difficult to fulfill.

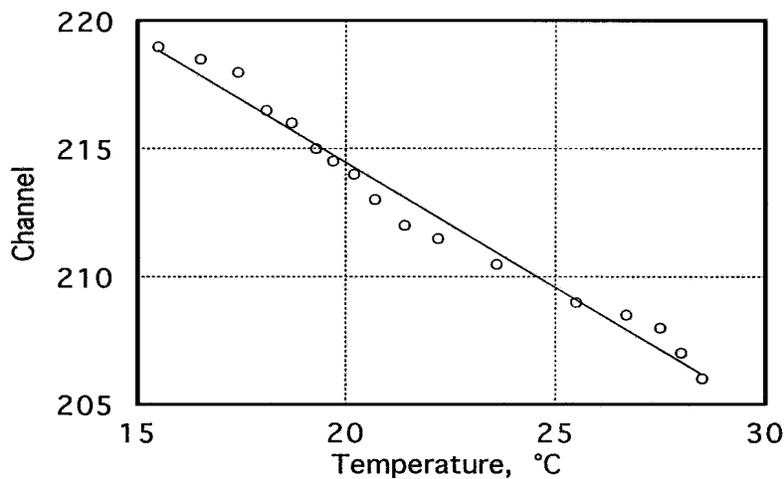


Figure 8 Shift in channel number at ^{137}Cs Compton edge versus temperature of the PMT

LONG-TERM STABILITY AND REPRODUCIBILITY

The long-term stability of the ICEL-C14 was studied by first setting the high voltage at HV_{bal} . Our ^{14}C benzene standard was then measured continuously at 24-hr counting periods for a month (Figure 9). The small, gradual decline in the count rate is due to the loss of benzene through evaporation, about 0.13% over 30 days (0.9 mg/week). The standard deviation from the line of best fit is -0.023% . As we count $11.2 \cdot 10^6$ pulses in the ^{14}C window in each 24-hr counting period, the relative statistical standard deviation is 0.030%. Even at this high level of precision, we can see no drift from a constant value. These measurements prove convincingly the extremely high stability of the ICEL-C14 at the balance setting, and is well below what is needed in high-precision dating.

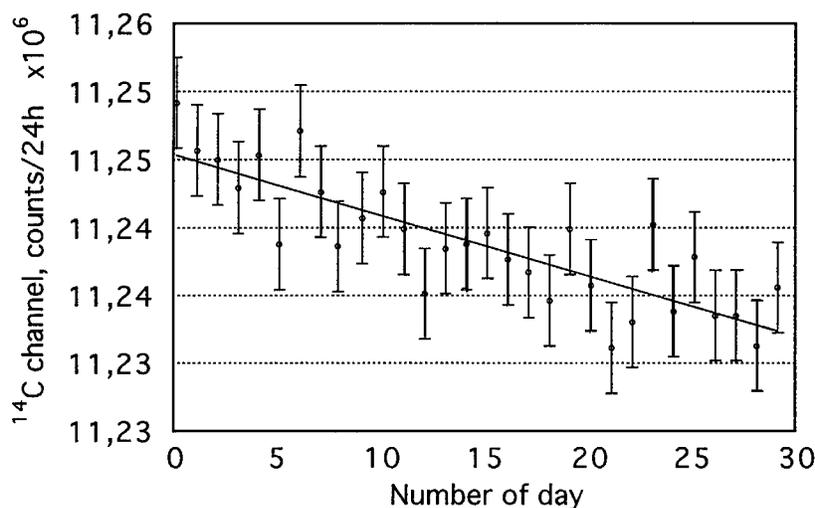


Figure 9 Stability test using consecutive 24-hr measurements of a ^{14}C standard (190 Bq)

The counting data contains information that can be used for checking the drift in relative gain, initially set close to 1.00, during the 30-day testing period. We use a parameter t , defined by:

$$t = [N(U) - N(H)] / [N(L) - N(U)] \quad (9)$$

which measures the relative number of pulses in the tail of the ^{14}C spectrum. We determine the value of t versus the relative gain by measuring the ^{14}C standard at varying high voltages, i.e., by using the same data as for determining the counting efficiency, as described above. There is a linear relationship between t and the relative gain in the interval of 0.85 to 1.15:

$$\text{RG} = 0.822 + 3.27t \quad (10)$$

At HV_{bal} ($\text{RG} = 1.00$), t has a value of 0.056.

During the 30-day stability test period, the number of pulses in the 4 counting channels of ICEL-C14 was recorded in 6-hr intervals, from which the value of t was determined (Figure 10). There was a small, gradual increase in the value of t , corresponding to a drift from an initial relative gain of 1.00 to 0.987 at the end of the 30-day period (September), which could have been caused by a 2.8 °C decrease in temperature. This shift in relative gain should, according to Figure 3, cause a decrease of 0.012% in counting efficiency at the end of the period.

The ^{14}C count rate reproducibility of sample changing was studied by setting the system initially at the balance point, and then counting the ^{14}C benzene standard 10 times in 23-hr periods, removing

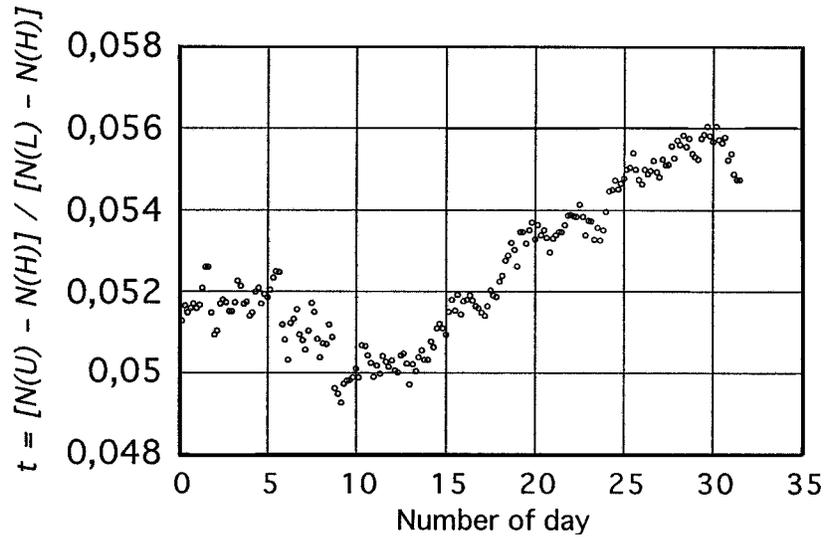


Figure 10 The measured value of parameter $t = [N(U) - N(H)] / [N(L) - N(H)]$ during the 30-day stability test period

and reinstalling the sample between measurements (Figure 11). After correction for evaporation, we get:

- Mean ¹⁴C count rate 130.17 cps
- Measured standard deviation 0.040%
- Calculated relative statistical deviation 0.030%
- Value of t 0.052–0.054

Finally, we measured the sample twice at 23-hr periods, where the vial had an obvious eccentricity of 2–3 mm relative to the axis of the PMT. This displacement reduced the count rate by 0.07% (Figure 11), while the t value was 0.049. These measurements show that no special care is needed in placing the vial on the top of the PMT, even in work at the highest practical precision.

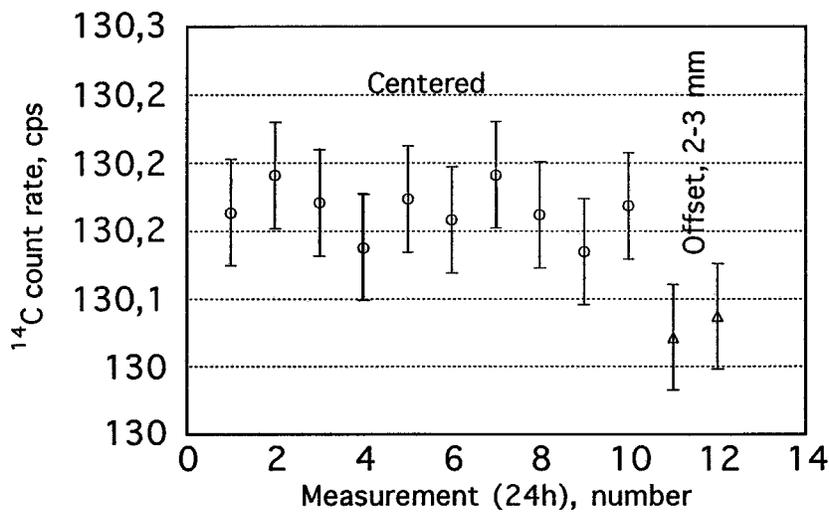


Figure 11 Repeated 23-hr measurements for ¹⁴C standard (190 Bq). The vial was removed and reinserted between measurements. Circles: vial centered; triangles: vial displaced 2–3 mm from center

CONCLUSION

When an LS counting system is used in ^{14}C dating, a high counting stability is best secured by applying the balanced window method. The balance point can be determined (or checked) quickly for each sample by using an external gamma source. For our system, a change of $\pm 8^\circ\text{C}$ from the temperature when the balance voltage is set will decrease the count rate in the ^{14}C window by 0.1%. The count rate drift in the ^{14}C window was less than 0.03% during the 30-day stability test period and the reproducibility in sample changing was better than 0.015%.

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