

BALANCED-ENERGY COUNTING WINDOW FOR STABLE LIQUID SCINTILLATION RADIOCARBON DATING

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ABSTRACT. This paper describes an optimal radiocarbon counting window for liquid scintillation (LS) ^{14}C dating that secures for unquenched as well as for heavily quenched dating samples maximal stability of ^{14}C counting efficiency and theoretically minimal quench correction. In high-precision dating, a balanced counting window with fixed channel limits is frequently used, where about 3% of the highest part of the ^{14}C spectrum is sacrificed for high ^{14}C counting stability. The stability is, however, diminished for quenched samples. Therefore, this window is here replaced by a balanced fixed-energy ^{14}C counting window where the channel limits depend on the quench level. The LS system used must have a linear amplifier and a multichannel analyzer. All samples are measured at a fixed high voltage. For energy calibration and determination of the quench level, the channel number of the middle of the 59.5-keV peak from an external ^{241}Am gamma source is determined before and after measuring each sample. This counting mode is valuable in high-precision dating. It could be widely applied if adapted to systems with a logarithmic amplifier, generally used in LS dating.

INTRODUCTION

In his pioneering radiocarbon dendrochronological studies, G W Pearson measured benzene (8 mL) samples to high precision with his vintage 1960s liquid scintillation (LS) system that only had 4 discriminators for pulse analysis. To secure high ^{14}C counting stability, he operated the system in a balanced mode with a fixed pulse-height counting window where highest part of the ^{14}C spectrum was sacrificed to maximize the ^{14}C counting stability (Pearson 1979). As his LS system operated with automatic sample changing, all samples were counted at the balance setting of his ^{14}C reference sample. His work was used as a guiding line in the development at the University of Iceland of a single-phototube LS system, ICELS.

The ^{14}C spectrum of quenched samples is shifted to lower pulse height compared to unquenched samples. This diminishes the ^{14}C counting stability in the fixed-channel balanced window. The first version of the electronic unit of ICELS had, like Pearson's LS counter, only 4 pulse-height discriminators. As sample changing is manual in ICELS, it was now possible to modify the high voltage for individual samples according to their quench level, to shift the ^{14}C spectrum to the position it would have in absence of quenching, so that also quenched samples were counted at their balance point (Theodórsson 2005). The ^{14}C counting stability is then secured for all samples. This is the earlier version of the ^{14}C balanced-energy window mode, called the spectrum restoration method.

The new electronic unit of ICELS has a multichannel analyzer and all samples are now counted at fixed high voltage. The balanced-energy counting mode then comes to its full merits as the position of the Am peak gives the quench level of the sample and no high voltage modification is needed. This counting mode can be used in all LS systems with linear amplification and a multichannel analyzer and probably also for systems with a logarithmic amplifier.

METHODS

The LS System

A single photomultiplier tube (PMT; 28 mm diameter) LS system, ICELS, specifically developed for ^{14}C dating, is used (Theodórsson 2008). Its new compact electronic unit has a high-voltage power supply, linear amplifier, and a pulse-height analyzer with 256 channels, which match well the resolution of the LS detector when a single radionuclide is measured. A 7-mL vial, usually with 3 mL of benzene, sits on the top of the vertical phototube. Some 10 mg of butyl-PBD scintillator is

added per mL benzene. The vial is wrapped (except for the bottom facing the PMT) with 2 layers of highly reflective polytetrafluoroethylene (PTFE) tape. This arrangement gives good scintillation photon collection at the cathode of the PMT and it is enhanced by a factor of nearly 2 by 60 mg of glycerol between the concave bottom of the vial and the PMT as it forms optical coupling. The detector unit is shielded by 5 cm of lead. Sample changing is manual.

Energy Calibration and ^{14}C Benzene Standard

All dating samples are measured at a fixed high voltage. For energy calibration, an external ^{241}Am gamma source (59.5 keV) inside the end of a 3-mm-diameter steel tube is used, which is manually inserted into the lead shield. The middle of the ^{241}Am gamma peak (59.5 keV) of each sample gives both energy calibration and the quench level of the sample. In stability and quench-correction studies, a laboratory mixed ^{14}C benzene standard is used, which gives ~ 250 cpm in the chosen ^{14}C window. Two days of counting time gives a precision of 0.1%.

Figure 1 shows the net pulse-height spectrum of the unquenched ^{14}C standard, and the spectrum of the external ^{241}Am gamma source as a function of channel number. The channel number of the middle of the ^{241}Am peak, $ChAm$, is used to transform the x axis from channel number to keV:

$$\text{keV} = (\text{channel number}) / ChAm \times 59.5 \quad (1)$$

For quenched samples, the pulse size of each ^{14}C beta particle is attenuated by the same constant factor, Att , compared to that it would have in absence of quenching. This ratio is measured by the shift of the channel number of the middle of the 59.5-keV peak:

$$Att = ChAm_q / ChAm_{unq} \quad (2)$$

where $ChAm_q$ and $ChAm_{unq}$ are the mid- ^{241}Am peak channel numbers for a quenched sample and the unquenched ^{14}C reference standard, respectively.

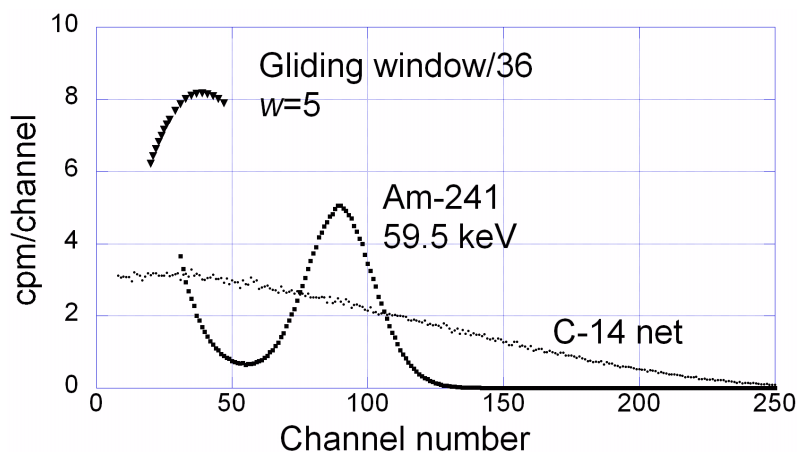


Figure 1 The net spectrum of a ^{14}C standard, the ^{241}Am spectrum with a gamma peak at 59.5 keV, and count rate in a gliding window for $w = 5$.

Balanced-Energy ^{14}C Counting Window

The channel numbers that determine the lower and upper limits of the ^{14}C counting window for a given net ^{14}C pulse-height spectrum are denoted by L and H . The ratio w between H and L ($w = H/L$)

L) is called the window width. For a chosen value w , only 1 value of L gives a balanced window. The lower the chosen value for L , the wider will the balanced window be and higher the ^{14}C counting efficiency. The lowest part of the spectrum is always rejected, to exclude either a contribution from eventual ^3H contamination in the benzene or because the background spectrum rises steeply in its lowest part.

The balanced window is determined in the following way. A value for w is first chosen, based on these considerations, and then the values of L and $H (= w \times L)$ are determined that give a balanced window. The lower limit L of the balanced window for a selected w is then determined from the net ^{14}C pulse-height spectrum. The ^{14}C count rate is calculated for this w in a series of gliding windows for varying values of $L(Ch)$. This is demonstrated here for the spectrum shown in Figure 1 for $w = 5$. The number of pulses in channels from 20 to 100 is first calculated, next in channels from 21 to 105, and so on. The result is shown in Figure 1. The count rate in the gliding windows rises to a maximum in channel 39.0, so the balanced window is from channel 39.0 to 195.0. This corresponds to energies from 24.0 to 120.0 keV according to the ^{241}Am energy calibration. This operation has been carried out to find L and H for integer values of w from 3 to 7 for the same spectrum (Figure 2). The absolute counting efficiency was calculated in the same way as shown above from the net spectrum of the Russian ^{14}C benzene standard with 501% activity of recent carbon standard.

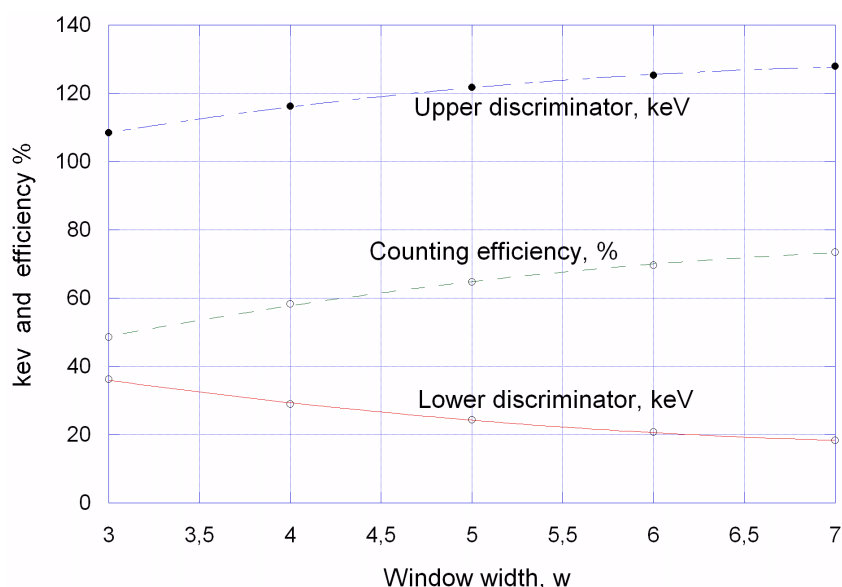


Figure 2 The upper and low energy limits of a balanced-energy window and the ^{14}C counting efficiency versus window width.

Balanced-Energy Window of Same Width for Quenched Samples

^{14}C standard samples at varying quench levels, i.e. for varying values of Att , are obtained by adding in steps up to 20 μL of acetone to the 3-mL ^{14}C benzene standard, taking care that the benzene loss during acetone addition is kept below 1 mg to secure that the ^{14}C activity in the vial remains constant. The pulse-height spectrum of the quenched samples is shifted to a smaller pulse height. The fixed-channel window then includes ^{14}C pulses of higher beta energy. Figure 3 shows the spectrum of the same standard sample, unquenched ($Att = 1.00$) and quenched ($Att = 0.76$).

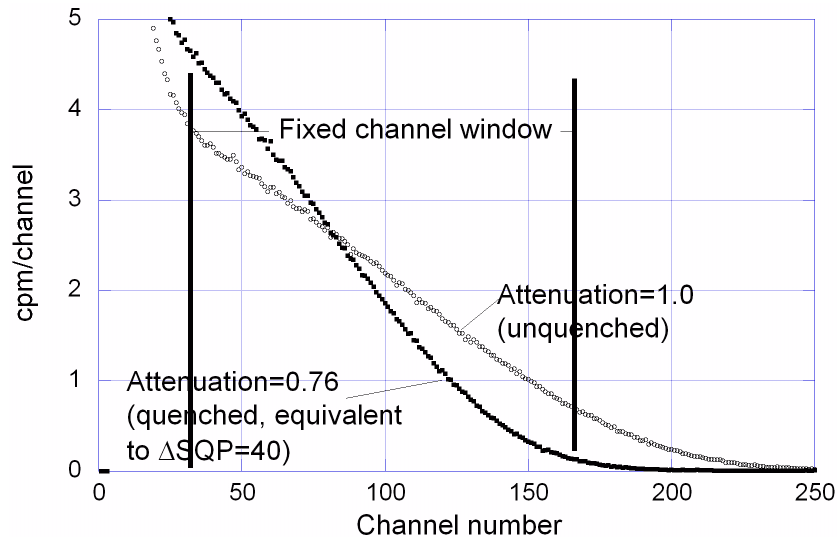


Figure 3 The spectra (total) of an unquenched and quenched ($Att = 0.76$) ^{14}C standard. A balanced ^{14}C counting window for $w = 5$ is shown.

Figure 4 shows the same 2 spectra where the x axis is now given in energy (keV, using Equation 1) but not in channel number, and the y axis in cpm/keV. For clarity, only every 6th point in the 2 spectra is shown. The figure shows that in this normalized form, the 2 spectra are nearly identical. When we therefore use a ^{14}C window with fixed energy limits (determined by ^{241}Am calibration) from 24.0 to 120.0 keV (for $w = 5$), the change in the ^{14}C count rate for quenched samples is theoretically minimal. This is to be expected as we for each sample are counting pulses from the same beta particle energy interval. A 3.3% spectrum shift to higher or lower pulse height will decrease the ^{14}C counting rate by only 0.1%, irrespective of quench level, when the balanced-energy window is used.

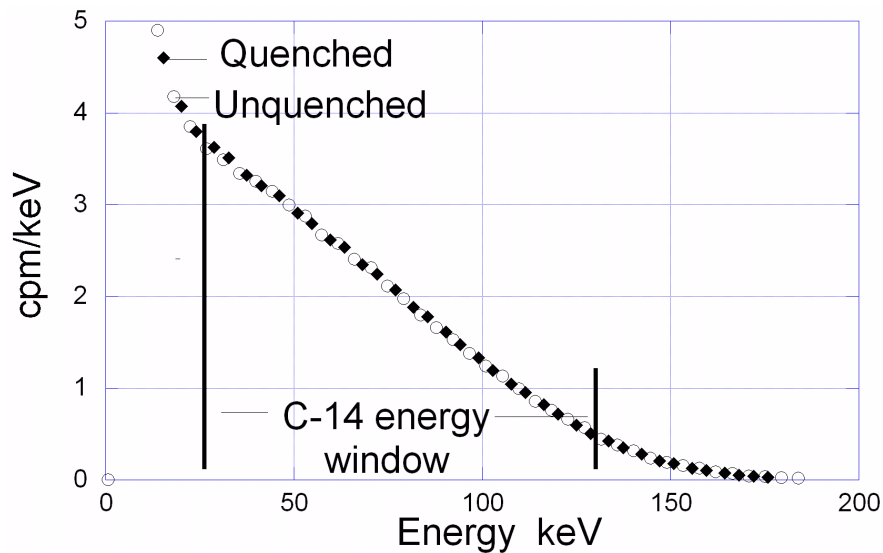


Figure 4 The same spectra as in Figure 3, but x and y axis are normalized to keV and cpm/keV, respectively. For clarity, only every 6th point is shown in both spectra.

It should, however, be noted that the background pulse-height spectrum is widely different from that of ^{14}C . The background count rate in the fixed-energy counting window increases significantly with quenching (Figure 5).

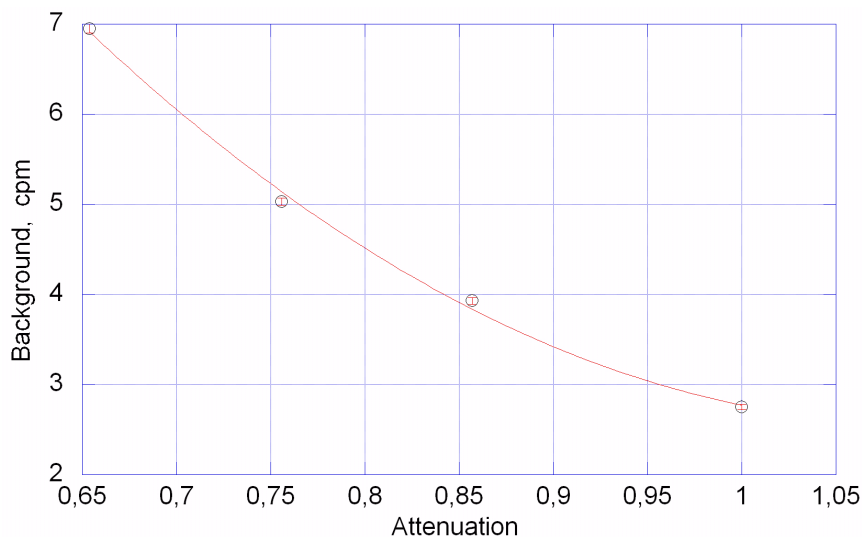


Figure 5 The background count rate for 3 mL of benzene versus attenuation

The net ^{14}C count rate of the ^{14}C standard was measured for 4 different values of *Att*. Figure 6 shows the relative net count rate for balanced windows ($w = 5$), (a) fixed-channel (channel 31 to 167) and (b) fixed-energy limits from 24.0 to 120 keV, as well as for a (c) wide fixed-channel window, from channel 39 to the end of the ^{14}C spectrum. The figure shows clearly the advantage of the balanced-energy window compared to fixed-channel limits. However, it should be noted that in the *Att* range from 1.0 to 0.9, there is only a small difference between the fixed-channel and fixed-energy windows.

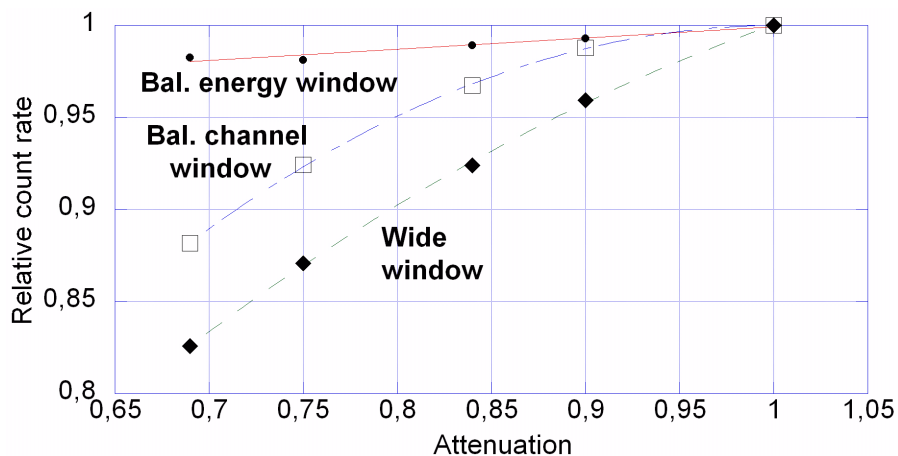


Figure 6 The relative net count rate of the ^{14}C standard for balanced windows with fixed channel and fixed energy and wide fixed-channel limits as a function of attenuation.

The standard deviation from the line for the measured balanced-energy window points in Figure 6 is 0.05%, actually lower than the theoretical (statistical) counting error of 0.10%. This difference must be due to the small number of measuring points. These measurements demonstrate the very high precision in these measurements and the stability of ICELS.

An important additional advantage of the presented counting mode is that the uncertainty of the quench correction depends much less on the accuracy of the quench indication parameter, *Att*, compared to other counting windows. The same relationship for the ^{14}C counting efficiency versus *Att* is found in all our 4 ICELS systems, as it is independent of the system used, so long as the response of the phototube is nearly the same. Individual calibration of each system is therefore not necessary. Frequent checking of the ^{14}C reference standard is not necessary as the ^{241}Am calibration secures constant ^{14}C counting efficiency.

Finally, it should be noted that it should be possible to adapt the balanced-energy ^{14}C counting window to all LS systems used in ^{14}C dating. However, for systems with logarithmic amplification this calls for a special study.

CONCLUSION

When the balanced-energy ^{14}C counting window is used, maximal counting stability is secured for samples at all quench levels. A 3.3% spectrum shift due to drift in system parameters will cause only a 0.1% decrease in the ^{14}C count rate. Furthermore, the quench correction as well as its dependence on the quench indication parameter (*Att*) is decreased to a theoretical minimum.

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