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THE APPLICATION OF ICELS SYSTEMS FOR RADIOCARBON DATING

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ABSTRACT. Liquid scintillation counting (LSC) for radiocarbon dating is a less expensive method than accelerator mass spectrometry (AMS), provides a high degree of accuracy, and is less prone to contamination due to the larger sample sizes. However, to obtain high precision, a long counting time is needed. The Gliwice Radiocarbon Laboratory is seeking to obtain an increased counting capacity with 2–3 mL benzene samples than we presently can achieve with our 2 Quantulus systems. We are therefore investigating the possibility of using a simple, single-phototube LS system (ICELS) for dating samples younger than 5000 yr. We present the first results of this investigation, including the measurement of 3 VIRI and 3 FIRI intercomparison samples.

INTRODUCTION

Quantulus 1220™ is a sophisticated liquid scintillation counting (LSC) system designed in the 1980s for the measurement of a variety of low-radioactivity radiocarbon samples. The ICELS system was designed a few years ago specifically for ^{14}C dating with an emphasis on simplicity. It has been thoroughly tested using elevated ^{14}C-activity samples, and its high stability has been verified (Theodórsson 2005). Here, results of measurements using the Quantulus 1220 and the ICELS systems are compared in order to test the ICELS as an alternative counting system that could be used by an established ^{14}C dating laboratory to increase its dating capacity.

EXPERIMENTAL

Liquid Scintillation Systems

Both the Quantulus 1220 and the ICELS (Figure 1) liquid scintillation (LS) systems used in the present study have been described thoroughly in previous publications (e.g. Kojola et al. 1984; Theodórsson 2005). Thus, only a brief description is needed here, with an emphasis on the systems' differences. The detector unit of Quantulus 1220 has, as most other LS systems do, two 180-degree exposed 50-mm-diameter photomultiplier tubes (PMT), facing the counting vial, for detecting the weak scintillation pulses caused by the decay of ^{14}C nuclides. To eliminate the background cathode thermionic pulses of the phototubes, only events giving simultaneous pulses in the PMTs are registered. This core unit is housed inside a long annular LS anticosmic counter that significantly reduces the cosmic radiation background component. To further enhance background reduction, the Quantulus 1220 has an external lead shield (10–20 cm) weighing 1000 kg.

The Quantulus 1220 has a fixed factory setting for high voltage and amplification (semi-logarithmic). In high-precision LS dating (±20 ^{14}C yr or better), the systems are generally operated in balance-counting mode, i.e. the high voltage is set at the value where the count rate reaches a maximum (Figures 2–3) for the ^{14}C reference sample with rising voltage (Theodórsson et al. 2003). This setting minimizes the influence of fluctuating parameters that can affect the ^{14}C spectrum (high voltage, amplification, and temperature), and it reduces the correction due to sample light quenching.

Modern PMTs (such as the R6094 Hamamatsu Photonics) have a greatly reduced thermionic background, and particularly when the lowest-energy nuclides (e.g. tritium) are not being measured, the 2-PMT coincidence arrangement is not needed for background reduction. For this reason, ICELS

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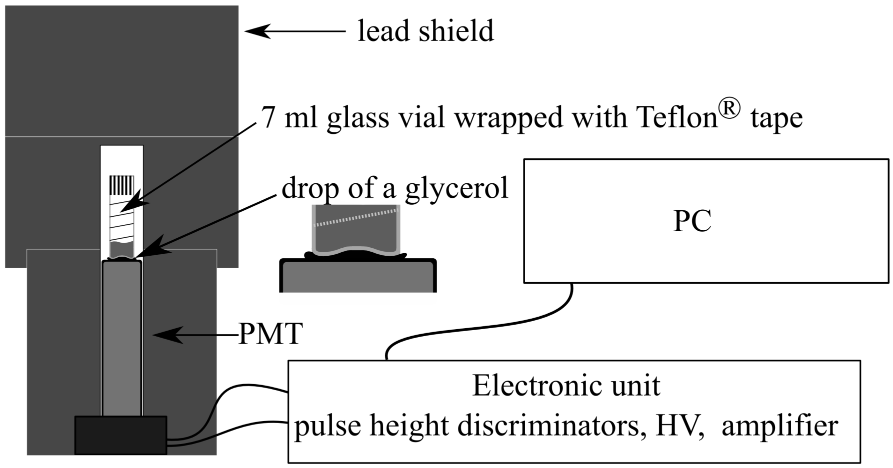

Figure 1 ICELS block diagram with detector cross-section

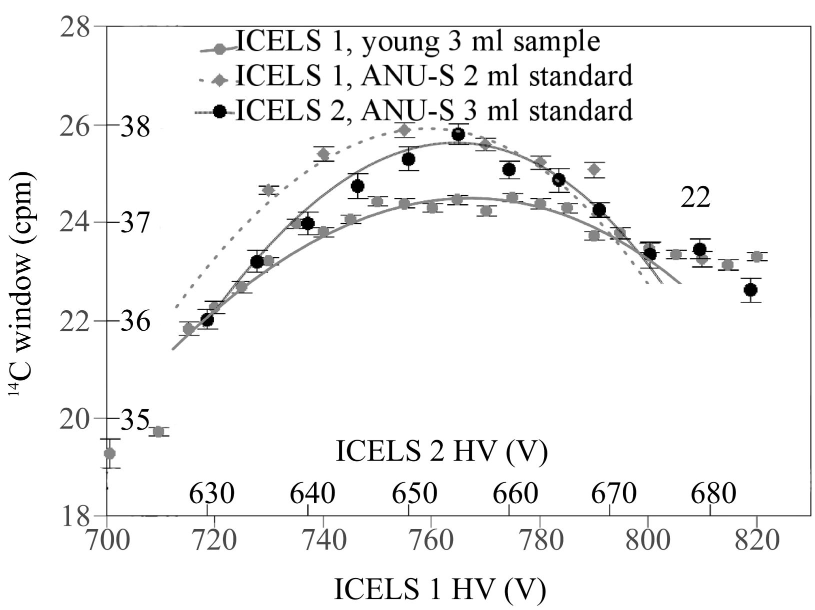

Figure 2 Setting the balance point in ICELS systems

has a single 28.5-mm-diameter PMT (Figure 1), which eliminates the coincidence electronics, simplifies the system, and makes it more compact (Figure 1). The benzene samples are in 7-mL glass vials sitting on the top of the PMT. The vials are wrapped, except for the bottom, with polytetrafluoroethylene (PTFE) tape. A drop of glycerol is added between the PMT and the vial for high-photon transmission (Theodórsson and Gudjónsson 2009).

Using ICELS, the quench correction is reduced to a theoretical minimum by adjusting the high voltage of each sample to its balance point (Pearson 1983; Theodórsson et al. 2003). The ^{14}C pulse-height spectrum is then adjusted to the same position as that of the reference sample (spectrum restoration balance mode).

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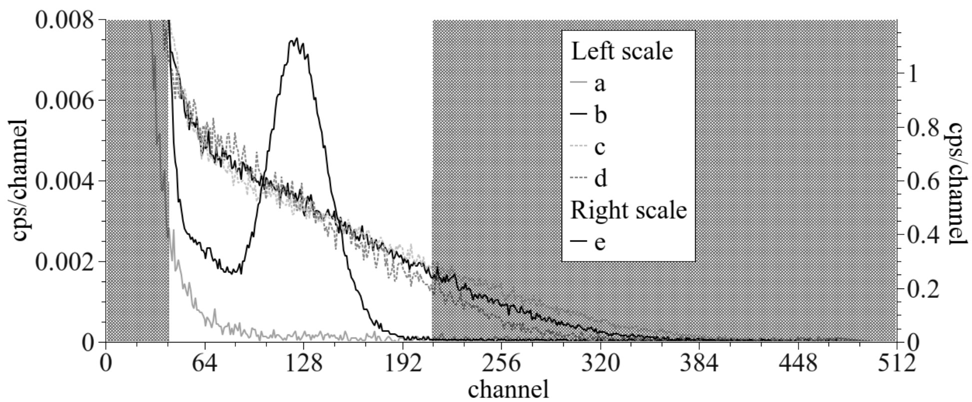

Figure 3 Typical spectra of ANU-S standard (b) and background (a) in 3-mL geometry. ^{241}Am spectrum (e) shows 59.5-keV γ line that is used to find balance point in each measurement. Lines (c) and (d) show the influence of different PMT voltages (665 and 645 V, respectively) on the ANU-S standard shifted from the balance point.

Comparison of ^{14}C LSC Measurements

Table 1 gives typical values of basic parameters used in ^{14}C dating (Pazdur et al. 2000). The factor of merit (FOM; Theodórsson 1991) in this work is calculated as:

$$FOM = \frac{S_0}{\sqrt{B}} \tag{1}$$

where S_0 is the modern biosphere standard and B is the background counting rate. Figure 4 shows the FOM of ICELS at 655 V (HV_{bal}) as a function of the lower discriminator (LD) and the upper discriminator (HD) in the ^{14}C counting window. The ^{14}C window spans from channel 40 to 214 in the 512-channel MCA correction mode. The position of the ^{14}C window, in the case of ICELS, responds to almost the maximum FOM. However, this FOM is only relevant when very old samples are dated. In our laboratory, ICELS will only be used for relatively young samples (<5000 yr). Samples older than ~5000 yr are measured in the Quantulus due to its much lower background..

Table 1 Comparison of 2-mL geometry LSC systems in the Gliwice Radiocarbon Laboratory.

Lab name	ICELS 1		ICELS 2	Quantulus 1		Quantulus 2
Geometry	2 mL	3 mL	2 mL	3 mL	2 mL	2 mL
S_0 (cpm)	14.661 ± 0.068	21.790 ± 0.067	14.304 ± 0.016	21.377 ± 0.029	13.818 ± 0.036	14.252 ± 0.066
B (cpm)	4.052 ± 0.032	4.471 ± 0.042	3.980 ± 0.025	0.4519 ± 0.0047	0.3072 ± 0.0048	0.4057 ± 0.0063
FOM (cpm^{½})	8.57	11.7	7.17	34.8	24.9	23.4
T_{max}[a] (BP)	35,300	39,100	35,100	47,200	45,300	44,400
Efficiency	62%	62%	60%	60%	58%	60%

[a]Value is calculated for 1000-min counting time.

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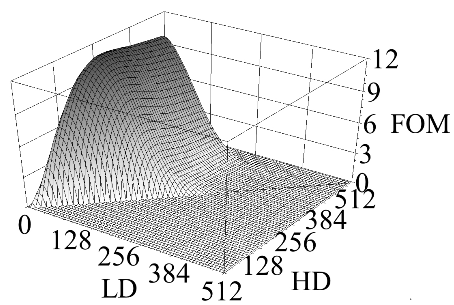

Figure 4 ICELS 2, 3-mL geometry FOM as a function of lower and higher band of ^{14}C window. The FOM of ICELS 1 looks very similar.

To compare our LSC systems and check their reliability, we used samples from the Fifth International Radiocarbon Intercomparison (VIRI). Consensus values were taken from Scott et al. (2007). Three of 4 VIRI samples (B, C, and D) were converted to 2-mL benzene samples. $\delta^{13}C$ values were measured as CO_2 before benzene preparation. We also used 3 samples from the Fourth International Radiocarbon Intercomparison (FIRI) program (Pazdur et al. 2003; Scott 2003). Figure 5 shows the plotted results, and Figure 6 presents a scaled deviation plot of the obtained results. The scaled deviation is calculated as:

$$\text{scaled deviation} = \frac{T_G - T_C}{\sqrt{\sigma^2{}_G + \sigma^2{}_C}} \qquad (2)$$

where T_G and T_C correspond to the Gliwice Radiocarbon Laboratory dating result and consensus value, respectively, and $_G$ and $_C$ are uncertainties of the values. The larger uncertainty in ICELS systems is caused by the shorter counting time. The Quantulus 1 and 2 VIRI C values are significantly different than consensus values. This is caused by an unknown gross error.

ICELS Counting Stability and Reproducibility

The stability of ICELS systems was presented previously (Theodórsson 2005) but not directly for ^{14}C dating. Some tests of ^{14}C dating stability and reproducibility were performed in our laboratory using ICELS 2. Table 2 and Figure 5 (with data points labeled as "1st value" and "2nd value") show the dating results obtained from sample measurements that were repeated. The results are in a very good agreement, and statistically they are the same. Moreover, there was a lapse of time between measurements of approximately 1 month and a different standard of activity was used.

Table 2 ICELS 2 reproducibility. Results of 2 different sets of measurements.

Sample	1st value	2nd value
Standard S_0 (cpm)	14.25 ± 0.12	14.319 ± 0.063
VIRI B (BP)	2740 ± 115	2790 ± 120
VIRI D (BP)	2810 ± 75	2905 ± 60
VIRI C (pMC)	110.74 ± 1.2	111.54 ± 0.64

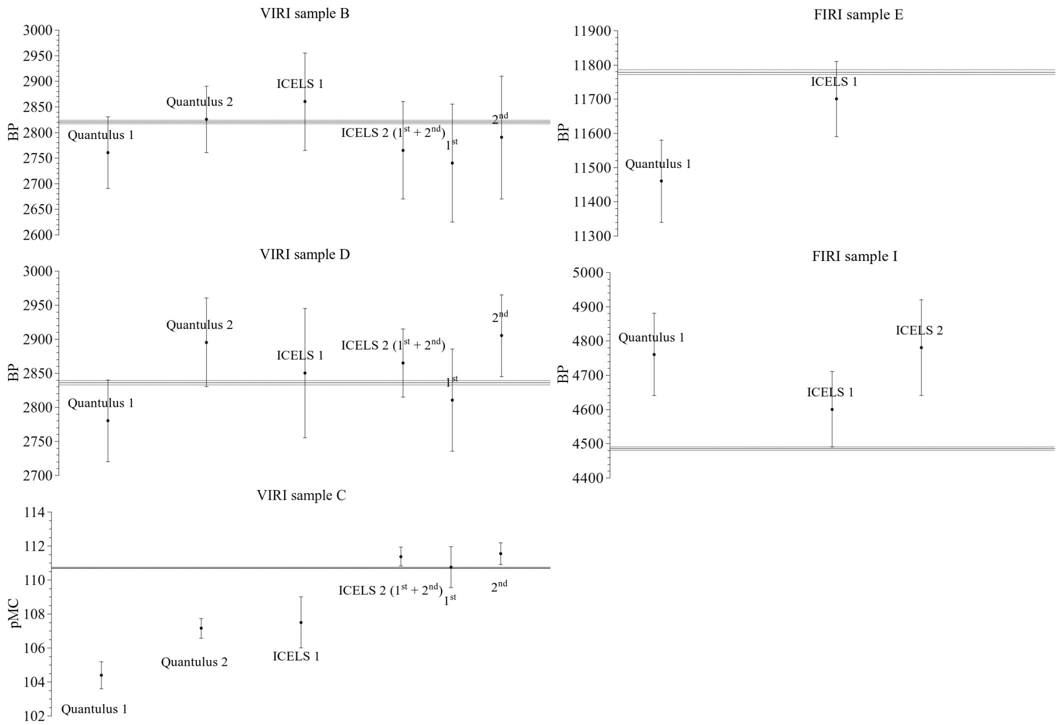

Figure 5 Comparison of consensus values with measurement results

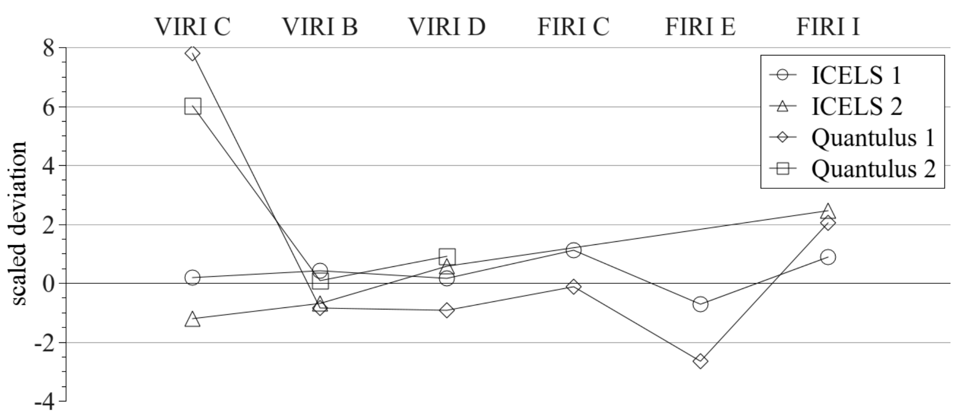

Figure 6 Scaled deviation of LSC systems for 3 VIRI samples

CONCLUSIONS

The measurement results of VIRI and FIRI samples for the Quantulus 1220 and the ICELS LSC systems are in acceptable agreement with consensus values. The deviation from the consensus value for VIRI C is higher than expected in both our LSC systems. Values obtained from ICELS 2 and ICELS 1 are satisfying, although higher precision can be reached. In this initial testing of ICELS, the factor limiting the precision was counting statistics. We expect that our ICELS results can be greatly improved in further work.

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