A COUNTER SYSTEM FOR HIGH-PRECISION ^{14}C DATING

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ABSTRACT. A multicounter radiocarbon dating system was developed applying the experiences of the previous one-channel low-level counting facility. The counter system consists of nine electrolytic copper proportional counters of identical diameters with sensitive volumes of 0.35–0.7dm^3 and filled with either methane at high pressure (6 bar) or CO_2 at 1 bar. The inner counters are surrounded by an anticoincidence shield consisting of five multiwire proportional flat counters filled with propane. The pulses of the detectors are handled by integrated amplifiers, discriminators and anticoincidence units interfaced to a microprocessor-controlled data evaluation unit. Software is written in BASIC using ASSEMBLER sub-routines. The overall precision of the system for modern carbon samples using high-pressure methane-filled counters (B ≈ 0.7 cpm, S ≈ 14 cpm) is better than 4‰ after a counting period of seven days.

INTRODUCTION

For the past 30 years, gas proportional counters have become increasingly important devices for low-level counting of several cosmogenic radionuclides. Gas counters for ^{14}C dating were first built by Libby (1955). However, a special proportional counter (DeVries, Stuiver & Olsson, 1959) has proved to be more suitable for high-precision ^{14}C measurements. The successful features of these and similar counters are high counting efficiency, allowing increased precision, as well as increased age range and comparatively easy sample preparation and filling, eliminating the memory effect, which was a serious problem with Libby's screen-wall counter.

Today, ^{14}C measurements needed for oceanographic applications or for dendrochronologic calibration of the ^{14}C time scale require increased precision (standard error better than 4‰) and high measuring capacity. To reach high counting precision and high capacity, various multicounter arrangements were developed (Tans & Mook, 1979; Povinec, Szarka & Usacev, 1979; Gulliksen & Nydal, 1979; Stuiver, Robinson & Yang, 1979; Schoch et al, 1980; Schoch & Münnich, 1981).

Using our combined experiences, we developed a multicounter dating system in our laboratory. The new detector system provides sufficient measuring capacity with a precision <4‰ requiring a counting time of one week per sample per counter.

PRACTICAL CONSIDERATIONS

We achieve a statistical precision of 4‰ (1σ) with 1.42×10^5 counts for modern sample in a one-week period including the error contribution from standard and background of 1 cpm. The counter must contain 1.03g of carbon, as the specific disintegration rate of ^{14}C in modern material is 13.6

dpm/gC; 1.03g of carbon is equivalent to 2.06 bar-L CO_2. We choose high pressures to avoid high background count rates due to large counter volumes. However, increasing the gas pressure is limited, because of the difficulties with high voltage and an attachment of primary electrons to the electronegative contaminants. Transit time of electrons through a counter is seven times longer for CO_2 than for CH_4 (Povinec, Szarka & Usacev, 1979). Thus, a counter filled with CO_2 is much more sensitive to electronegative impurities than a counter with methane filling. Transit time of electrons can be lowered with an anode of larger diameter and higher tension on the wire, but it increases the danger of spurious pulses due to electrical leakage across the insulators. Eliminating spurious signals is of paramount importance in measuring low activities. Preliminary experiments on anode wires showed that our counters can operate satisfactorily with wires of very small ($5\mu m$) diameter.

From these considerations and technical experience gathered during preliminary experiments, we decided to use high pressure (6 bar), small wire diameter ($5\mu m$) and, at the same time, we adhered to our CH_4 traditions.

PASSIVE/ACTIVE SHIELDING

We minimized cosmic-ray muon events with an anticoincidence arrangement. For economic reasons, only 1 anticoincidence shield was used for all 9 measuring counters, which differs from the previous cylindrical geometry (Csongor & Hertelendi, 1986) with 5 stainless steel flat counters ($844\times396\times50$mm, $844\times276\times50$mm, $496\times456\times50$mm).

To maximize shield efficiency for cosmic-ray muons that occur at large angles to the vertical, the anticoincidence shield was opened only at one side where the pumping pipe lines leave the system. Large-diameter ($100\mu m$) silver-plated BeCu wires (Little Falls Alloys, Inc, Patterson, New Jersey) and a teflon insulator were applied, ensuring long-term stability of the guard counters. The filling of the anticoincidence system proceeds by flushing it with clean propane for two days. Then the counter system is closed and it works for about one year with the same filling.

The flat counter system is in a lead shield ($1250\times95\times80$mm, weight: $\approx 7\times10^3$kg); the wall is 150mm thick and 250mm on the top. We use a lead layer, 50mm thick, between the guard and proportional counters. We placed a 50mm boron-loaded paraffin layer inside the passive shield for thermalization and absorption of neutrons produced by cosmic-ray particles in the lead. The low-level counting equipment is in the basement of a two-story building, 2m underground.

COUNTER MATERIALS AND CONSTRUCTION

Quartz and copper have very little radioactive contamination and are preferred construction materials for low-level counters. Walls at least 2.5 and 3mm thick were required for the quartz end tubes. Our insulators exhibited a "moderate shock" effect over 5 kV when voltage was suddenly changed. Although the number of spurious pulses diminished exponen-

tially in time after stabilization of high voltage, the effect disappeared only when <5kV were connected to the wire. We used small gold-plated tungsten wires (LUMA, Sweden) for this reason.

The metal parts of the counters were made of electrolytic copper. All joint between metal and quartz were sealed with two-component epoxy resin (Araldit AV-134B, HY 996, Ciba AG, Basel). The gas inlet tube was brazed to the mantle of the counter (Fig 1).
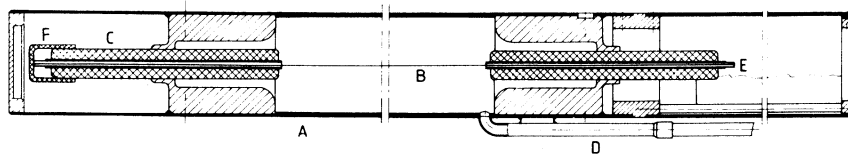

Fig 1. Schematic cross-section of the proportional counter: A) electrolytic copper cathode; B) gold-plated tungsten wire; C) quartz end tube; D) gas inlet; E) copper capillary; F) teflon cup

The vacuum/pressure valves are soft-sealed metal valves. Owing to the clean system, sample quality did not deteriorate, even after an extremely long measuring time. All counters run with a common high voltage due to their identical anode and cathode diameters and filling pressures. However, the length of the counters are different, ensuring optimum measuring conditions for varying sample amounts.

Wire supports and counter endplate shapes were designed in such a way that the active volumes of the counters were nearly equal to their total volume (Stuiver, Robinson & Yang, 1979). The counting efficiency was >90% for the geometric volume of the counter body (excluding the filling line).
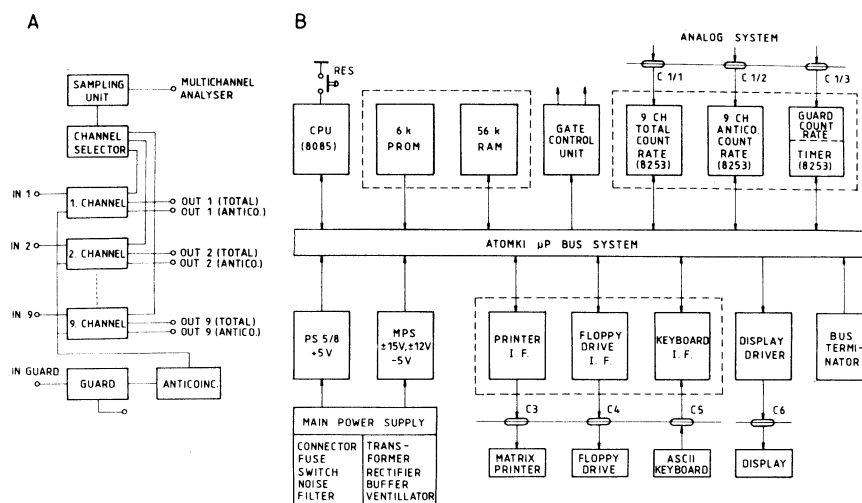

Fig 2. Block diagram of the analog A) and digital B) data processing systems

SIGNAL PROCESSING

Figure 2 shows the electronics and data acquisition system. The charge-sensitive preamplifiers, high-voltage capacitors (C=2nF), and high-voltage filter units are mounted in the copper endcups of the proportional counters. Other electronic parts work outside the lead shield. The linear amplifiers, discriminators and coincidence circuits of the nine channels are in one unit separate from the unit for signal handling of the anticoincidence counter. This part of the system also contains an analog circuit, for obtaining spectral information of the anticoincidence counts with a multichannel analyzer. Bipolar pulse shaping is performed in the linear amplifier with active filters (Table 1).

TABLE 1

Specifications of the analog system

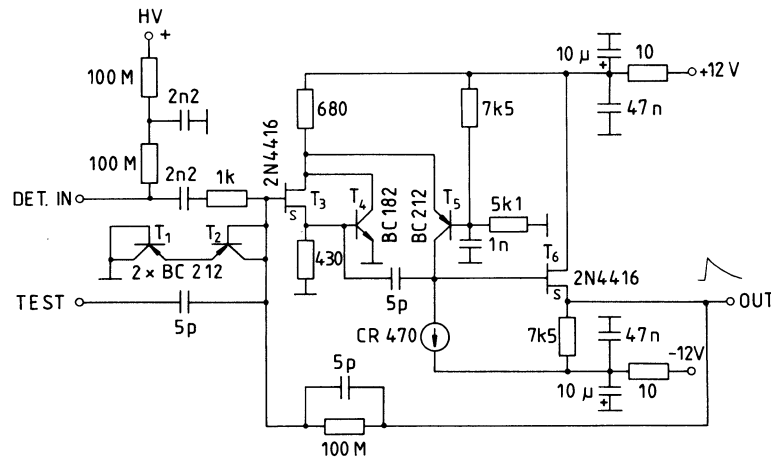| Specified parameters	Numerical values
Charge to voltage conversion coefficient	2×10^{11}V/C
Gain in the preamplifier	2
Gain in the linear amplifier	12 (proportional counters)
	18 (flat counters)
Time constant of shaping	$1.6\mu s$
Discriminator window	50–5000mV (2–160keV)
Coincidence resolving time	$500\mu s$
Operating voltage	3300V (proportional counters)
	5100V (flat counters)

Fig 3. Circuit diagram of the charge-sensitive preamplifier

For the charge-sensitive preamplifier, we use the circuit shown in Figure 3. Its design is, in most respects, similar to a conventional charge-sensitive unit aplying a cascade stage. The main difference is that instead of a single FET at the input, a FET-bipolar transistor arrangement is used as it is shown in Figure 3. Using this solution, the transconductance of the FET-bipolar transistor arrangement is ca 20 times larger (depending on the resistor in the FET source and the current gain of the bipolar transistor) than that of a single FET, *ie,* the open loop gain (dc gain) of the overall circuit is increased by this factor. Besides this, the unity gain band width is also increased without sacrificing the frequency stability, which is an inherent feature of the conventional circuit. However, the noise at zero input capacity is somewhat larger than in the single FET arrangement. From these it follows that the use of this kind of circuit may be advantageous with high – capacity detectors, such as large volume proportional counters.

In the linear amplifier, we use a shaping circuit, which is a simplified variant of the widely used semi-Gaussian shaper. Its functional diagram is shown in Figure 4. Basically, it is a unipolar shaper and the time function of
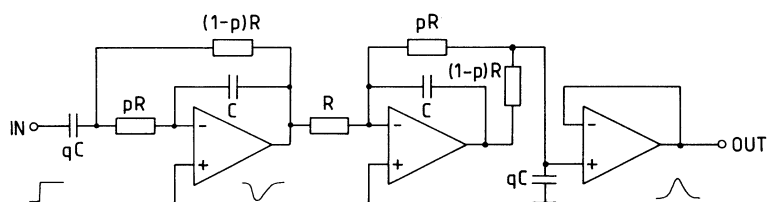

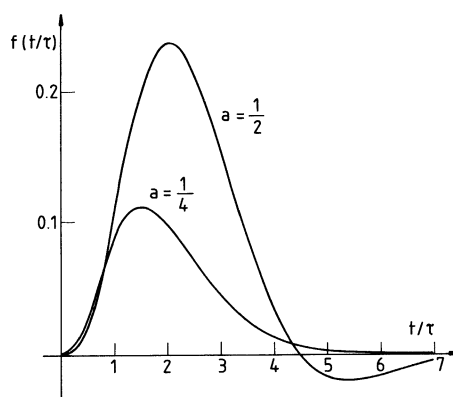Fig 4. Simplified circuitry of the semi-Gaussian shaper

Fig 5. Step response ($f(t/\tau)$, $r=RC$) of the semi-Gaussian shaper

the step response depends on the $a = (p-p^2)\ q$ parameter. The calculated signal shapes for two values of a are shown in Figure 5.

From the point of view of noise filtering, an important parameter of the circuit is the "cusp factor", F, which is the relative noise-to-signal ratio relating to the infinite cusp shaping. For the given circuit, F=1.17 at $a = 1/2$. This F value is significantly smaller than that of the simple CR-RC shaper (F = 1.36) and it is not significantly larger than that of the ideal Gaussian filter (F = 1.12). For bipolar shaping, a simple CR filter is used in the next stage.

Digital signals were fed into 19 scaler registers built from INTEL 8253 programmable counters. The scalers are controlled by a microcomputer based on an INTEL 8085 CPU and equipped with 56 kbyte RAM, 6 kbyte PROM, alphanumeric display driver, multivoltage power supply and interfaces to matrix printer, floppy drive unit and keyboard. Software is written in BASIC, using ASSEMBLER sub-routines.

WORKING CHARACTERISTICS

The working characteristics of the proportional counters filled with methane at 6 bar and the anticoincidence counters filled with propane at 1 bar are shown in Figure 6. The plateaus of the proportional counters are 600V long with a slope of <2%/100V; the flat counters have a 700V plateau length with a similar slope. We observed no shift of characteristic normally in three years of operation.
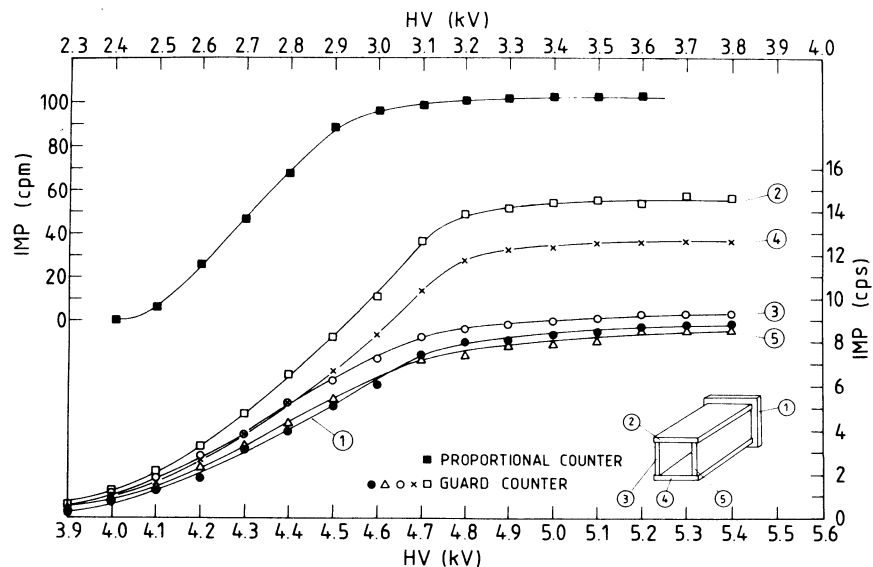

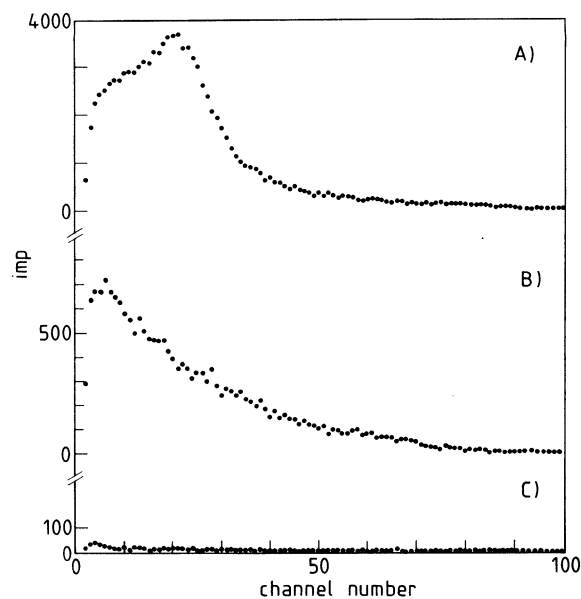Fig 6. Working characteristics of the counters

Fig 7. Energy spectra of the shielded proportional counter filled with methane: A) total energy spectrum; B) ^{14}C beta energy-loss spectrum; C) background spectrum (accumulation time 6×10^5 s). The broad peak in the total spectrum (A) is caused by cosmic-ray μ-mesons traversing the counter.

BACKGROUND

High-precision ^{14}C measurements depend primarily on stability of counting efficiency, good reproducibility of actual values during measurement and not so much on low background. We estimated the actual value of background during measurement from backgrounds before and after the sample measurement.

The energy spectrum of the background proves that the direct contribution of mesons is negligible (Fig 7). However, the dependence of background on barometric pressure supports the assumption that muon-initiated radiation cascades are responsible for most of the background.

MEMORY EFFECT

Small variations of background with time indicates that, generally, sample-to-background memory effect is immeasurably small. The use of more active gases obtained from stack samples of a nuclear power plant is safe as long as the activity is not higher than 100 times the NBS standard activity. We observed 0.7 cpm (100%) increase of background after measurement of a stack sample which was 1000 times more active than the NBS standard. However, this type of memory disappears when we use longer evacuation time (*ie*, 12 hr).

SYSTEM PERFORMANCE

Table 2 summarizes the most important operating parameters of the multicounter measuring system. The systematic error of the multicounter arrangement was checked by intercalibration (Scott *et al,* in press)

TABLE 2

System characteristics

System parameters	Counters		
	Type 1	Type 2	Type 3
Total active volume (dm³)	0.35	0.5	0.7
Standard counting rate (S) (cpm)	14	19	26.5
Background (B) (cpm)	0.7	1	1.4
Figure of merit (S/$\sqrt{B}$)	16.7	19	22.4
Max measurable age (2 days) (yr)	40,700	41,700	43,100
Precision for modern sample (7 days) (‰)	4	3.4	2.9

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