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HIGH-PERFORMANCE ^{14}C GAS-PROPORTIONAL COUNTING SYSTEM APPLYING PULSE-SHAPE DISCRIMINATION

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ABSTRACT. The new ^{14}C dating system of the Geological Survey of Finland consists of three CO_2 proportional counters of conventional size, a liquid scintillation anticoincidence guard, and a passive shield of about 90-year-old iron. The counting room is an air-conditioned Faraday cage constructed of concrete with aggregate of crystalline Precambrian limestone. We apply pulse-shape discrimination that greatly improves the precision of our ^{14}C analyses – the reduction of background is over 70%, while the modern count rate is reduced only by 15 to 20%. The purity of the counting gas is monitored by recording the cosmic muon pulse-height spectrum. At present, the dating limit for a two-day measurement is 57 ka. We summarize here the most significant results and aspects of pulse-shape discrimination

INTRODUCTION

During the past 40 years of ^{14}C dating, the applications of the method have been increasing at an accelerated rate. New applications, as well as the ever-increasing use of calibrated radiocarbon dates, demand the development of highly sensitive measuring techniques (see Polach 1987).

A few institutions have underground laboratories where very low backgrounds can be achieved. In Finland, due to the granitic bedrock with high U- and Th-content, building a low-level underground ^{14}C laboratory would be extremely expensive. In 1984, we began updating our existing equipment (Heikkinen, Koivisto & Äikää 1974) in collaboration with Wallac Oy (Kaihola *et al.* 1985). We replaced a Geiger anticoincidence guard with a liquid scintillation guard counter, and applied a pulse-shape discrimination technique to reduce further the background. All information including time of arrival for each event is stored for further analysis (Kaihola *et al.* 1984). Software consists of several diagnostic programs (*e.g.*, time series analysis), which play a significant role in data validation.

DISCUSSION AND RESULTS

The details of the new dating system and general improvements have been published elsewhere (Mäntynen *et al.* 1987 and references therein). In the following, we restrict ourselves to our development work since 1987.

Active Shield Performance

Originally, five standard-quality electron-multiplier photo tubes, selected on the basis of dark current and gain, were mounted at each end of the liquid scintillation guard. Measurements carried out in the low-level laboratory of Wallac Oy demonstrated that the tubes contained some gamma radiation, mostly ^{40}K. They were replaced by low gamma activity tubes made especially for this study, in the belief that this would further reduce the background. However, no performance enhancement was achieved with this procedure, for which we can give three explanations: 1) the guard has good counting efficiency for gamma radiation; 2) the counting efficiency of the sample counters is low for gamma radiation; 3) the photo tubes are not very close to the sample counters.

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Counting Characteristics

The dating system includes three counters, originally made of oxygen-free copper. The anode wire insulators were quartz tubes attached to counter ends with epoxy adhesive. The quartz tubes reduced the effective counting volume, and the epoxy glue gradually polluted the counting gas (CO_2). Gas gain was reduced during long counting periods. The same reduction occurred with repetitive measurement of background and modern standard samples without repurification.

We tried to avoid these problems by building counters with quartz ends. This was not successful, because it was not possible to build a working copper-quartz junction. O-rings (viton, kalrez, *etc.*) proved no better than epoxy glue. Finally, we constructed a quartz counter with a copper foil cathode (Fig. 1). We used commercial quartz-metal electrical feed-throughs to assemble the anode wire and the cathode. They were fused directly to the counter body. The counter contains no glue or o-rings. This counter construction seems to be working well. Effective volume is large and counting properties of the gas remain stable. This makes it possible to use larger counter diameter, higher pressure and lower high voltage, all of which increase the resolution of our pulse-shape discrimination. Stable counting properties are particularly important with small counters, due to the longer counting periods needed. In our experience, the only way to make pulse-shape analysis function with small counters is to use a counter with the largest diameter possible.

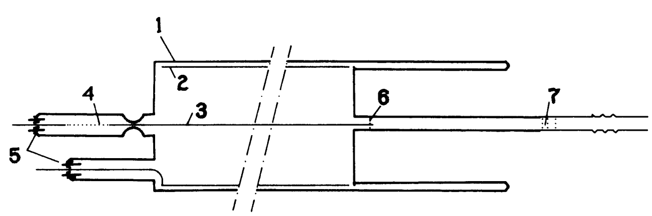

Fig. 1. Quartz counter, inner diameter = 45 mm, active length = 150 mm. 1. Counter body, quartz, 2 mm thick; 2. Copper foil cathode, 0.3-mm thick; 3. Anode wire, kanthal, diameter = 0.025 mm; 4. Tension spring for anode wire (spot welded to the wire and to the feed-through); 5. Quartz-metal feed-throughs; 6. Quartz centering piece for anode wire (holes allow gas flow to the counter); 7. Quartz to metal junctions.

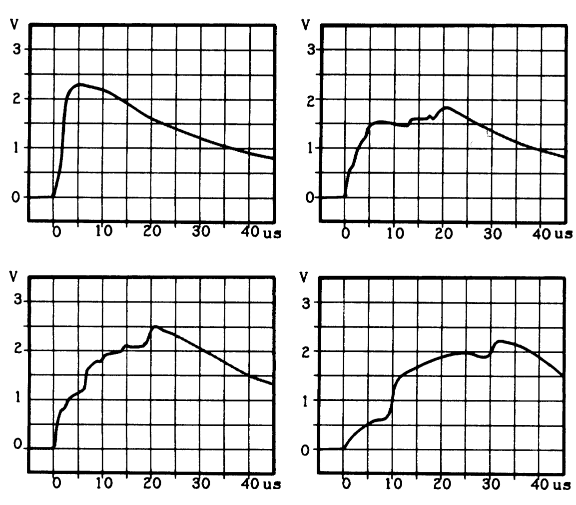

Fig. 2. Examples of different pulse shapes. Top left is a β-particle. The others are typical background pulses generated by cosmic radiation.

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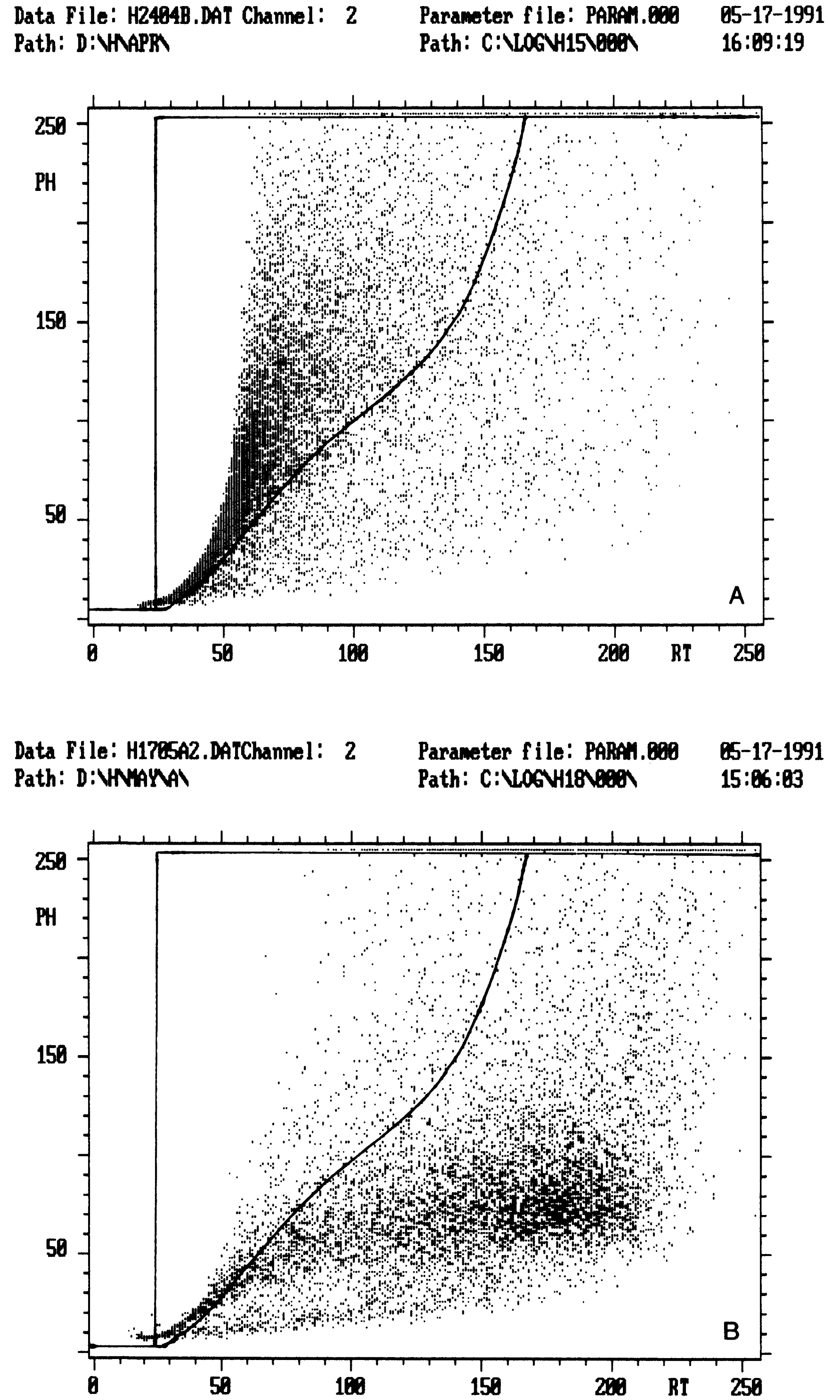

Fig. 3. Rise-time–pulse-height distribution of modern standard (A) and of coincidence events with the guard (B). RT-channel 256 roughly equals 25 μs.

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Electronic Background Reduction

The present electronics of the pulse-shape discrimination were described by Mäntynen *et al.* (1987). We have begun a new cooperative project with Coronel Oy in order to produce commercially available counting electronics with pulse-shape discrimination.

Development of the charge-sensitive preamplifier was completed in the summer of 1988. The old preamplifier was based on an operational amplifier; the new one is based on discrete semiconductors. The slew rate is *ca.* 170 V μs^{-1} and input capacitance 2 pF, and the noise is lower than the old one. At present, the coupling capacitor and the anode resistor are soldered directly to the anode. The preamplifier is at the end of the counter in its own housing. All high-voltage components are covered with electronic-quality silicone rubber.

Figure 2 illustrates the difference between β- and background pulses using the new preamplifier. In our pulse-shape analyzing method, the total length of the rising edge of the pulse is measured; in this way, the accumulated rise time of background pulses is longer than that of β-pulses.

Figure 3 illustrates rise-time–pulse-height distributions for the modern standard and for pulses generated by cosmic events. The average rise time of coincidence events is so much longer than that of β-events, that by applying pulse-shape discrimination, only 15–20% of modern standard counts are lost, whereas the background is reduced by more than 70% (see Table 1). Comparison

TABLE 1. Description and performance of the counters. The guard counter has three enclosures for sample counters. Performance of the four counters, P, K, S and Q, is included.

	Counters			
Performance data	P	K	S	Q
Counter Parameters				
– body material	Copper	Copper	Copper	Quartz
– length (mm)	360	600	600	150
– diameter (mm)	50	50	50	45
– total volume (liter)	0.707	1.178	1.178	0.287
– eff. volume (liter)	0.511	0.982	0.982	0.244
– working press. (kPa)	345.1	345.1	400.0	345.1
– working voltage (V)	5130	5150	5550	4900
– anode wire d. (mm)	0.025	0.025	0.025	0.025
Modern standard (S_o):				
– total (cpm)	12.53	24.82	27.93	5.28
– after discr. (cpm)	10.71	19.86	23.17	4.29
– after discr. % tot	85	80	83	81
Background (B):				
– total (cpm)	0.48	0.90	0.85	0.314
– after discr. (cpm)	0.14	0.25	0.25	0.071
– after discr. % tot	29	28	29	23
Figure of merit ($S_o/\sqrt{B}$):				
– total	18.2	26.1	30.3	9.4
– after discr.	28.7	39.5	46.4	16.1
Max. measurable age,				
– 44-h counting time:				
– total	49,400	52,300	53,500	44,100
– after discr.	53,000	55,600	56,900	48,400

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of the pulse-height spectrum of coincidence events with that of background events before and after discrimination (Fig. 4) demonstrates that our pulse-shape discrimination method removes mostly background pulses that are of cosmic origin. The cosmic muon peak in Figure 4A, also visible in the upper curve of Figure 4B, cannot be seen after discrimination (lower curve).

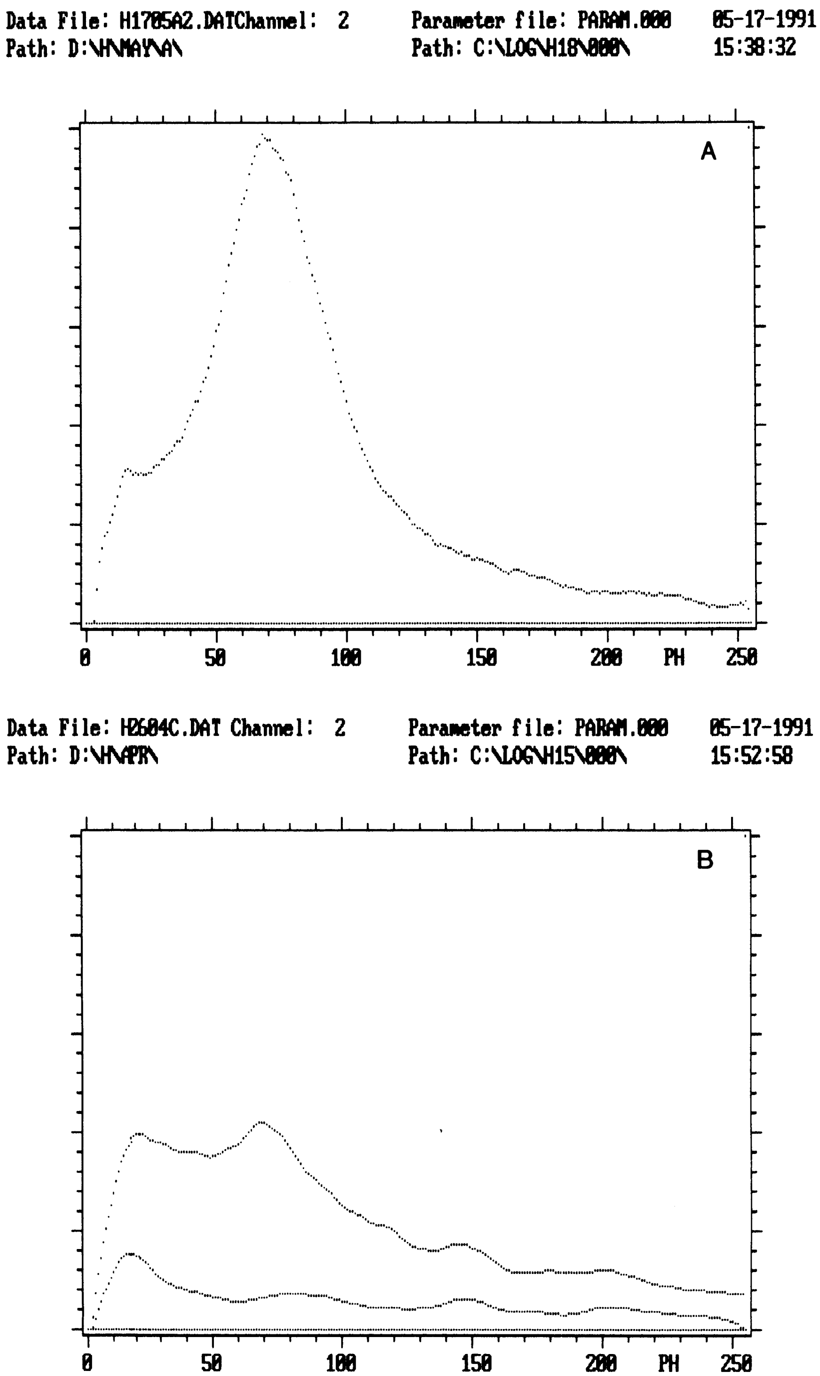

Fig. 4A. Pulse-height spectrum of events generated by cosmic radiation; B. Pulse-height spectrum of total background (upper curve) and background after discrimination (lower curve).

To ensure the purity of the counting gas, we check the peak and the median of the coincidence counts. If the counting gas is not pure, quenching can be seen from the change of the location and shape of the muon peak (Fig. 4A). We can compensate for this by increasing the high voltage by a few tens of volts. If this HV change is not enough, the sample is repurified.

Table 1 presents the parameters and performance of different counters.

Conclusion

According to Polach (1987), we can achieve the performance level of an underground laboratory with pulse-shape discrimination. We believe the most important aspects for the successful use of pulse-shape discrimination are pulse analyses, the fast charge-sensitive low-noise preamplifier and counter construction. The sensitivity of our gas purity test, based on the behavior of the cosmic muon peak, together with versatile diagnostic software, are also important in controlling the measurements.

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