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A LOW-COST MINIATURE COUNTER SYSTEM FOR RADIOCARBON DATING

KAZIMIERZ JELEN* and MEBUS A GEYH

Niedersächsisches Landesamt für Bodenforschung
D 3000 Hannover, Federal Republic of Germany

ABSTRACT. Despite the great potential of the AMS technique for ^{14}C dating of milligram samples, the use of multiple miniature counter systems is still promising. Investment costs are relatively low and long-term financing of age determinations can be held within bounds. We have developed a ^{14}C dating system containing 10 miniature counters for $30,000, whereas commercial systems usually cost more than $150,000. Counting is not quite as good as that of the technically more sophisticated commercial systems, but the disadvantage for routine work is not significant. CO_2 preparation and purification take less than 30 minutes of active work including measurement and data evaluation. The cost of one age determination on a 22mg sample is about $35.00.

INTRODUCTION

A statistical evaluation of the weights of the submitted ^{14}C samples in 1972 showed (Geyh, 1967) that 20% contained <150mg C. As a result, a ^{14}C counter for 40mg samples was constructed within a plastic scintillation guard counter. However, it had three disadvantages:

1) background counting rate of 0.25 cpm was too high,

2) memory effect was unacceptably large due to absorption of gas in the plastic scintillator, and

3) long-term stability was inadequate.

Harbottle, Sayre and Stoenner (1979) constructed quartz-tube miniature counters for small ^{14}C samples which were introduced in routine dating by Otlet *et al* (1983). Despite the success of the AMS technique (Wölfli, Polach & Andersen, 1984), the development of commercial miniature counter systems was stimulated (Kaihola *et al*, 1984) by vast financial savings. Miniature counter systems costing ca $150,000 are still much less than $1,000,000 or more for AMS accelerators. Moreover, maintainance is inexpensive and staff need not be highly skilled.

Our goal was to construct a low-cost miniature counter system with simple techniques which can also be used in Third World countries, whose interest in archaeologic and ecologic research is growing rapidly.

GENERAL SCHEME

Miniature counter systems consist of 10 to 16 low-level counters for ^{14}C dating of samples containing 10 to 50mg C, a plastic or NaI(Tl) guard detector, a passive shield made of low-level lead, a filling line for the counters, and sophisticated electronics. The general scheme has been described by Otlet *et al* (1983). Storage and statistical evaluation of the counts, including pulse-height (PH) analysis (Otlet *et al*, 1983), analysis of rise-time data (RT) or time series data (TA) (Kaihola, Polach & Kojola, 1984) is done by simple personal computers.

Carbon dioxide (Otlet *et al*, 1983) or methane (Kaihola *et al*, 1984),

* Guest scientist from the Institute of Physics and Nuclear Technology, ul Mickiewicza 30, Pl 30018 Cracow, Poland

which require rather complicated chemical preparation lines, are used as counting gases. Gas samples are stored in small metal or glass bottles. Measuring time is between 2 weeks and 3 months.

COUNTER CONSTRUCTION AND FILLING SYSTEM

The most sensitive part of the system is the low-level miniature counter and the material used in its construction must be of lowest activity. According to our experience, electrolytic copper and quartz seems to be the most promising for construction of small-volume, low-level counters for high-pressure operation (Geyh, 1967). The anode is made of 25μm stainless steel (Leico Industries Inc, NY). Nylon (Polypenco, Cologne, FRG) and quartz are used for insulators. Instead of soldering, a two-component glue (UHU hart 300, FRG) was used. The construction details are shown in Figure 1. On the basis of the range of small samples that have been submitted to our laboratory, we have chosen a counter size of 15ml with an active length of 6cm and a diameter of 1.8cm filled with CO_2 at 3 bar. This corresponds to samples containing 22mg C.

The first counters were made of technical-grade quartz coated with gold. High background counting rates were obtained (Table 1). Low-level gamma-spectrometric analysis showed that the technical-grade quartz was contaminated with daughter products of the uranium decay series. Quality was improved by using synthetic quartz (Suprasil from Hereaus, Hanau, FRG). The best results have been obtained with OFHC copper counters, which can be built in a day. Stuiver, Robinson and Yang (1979) found, on

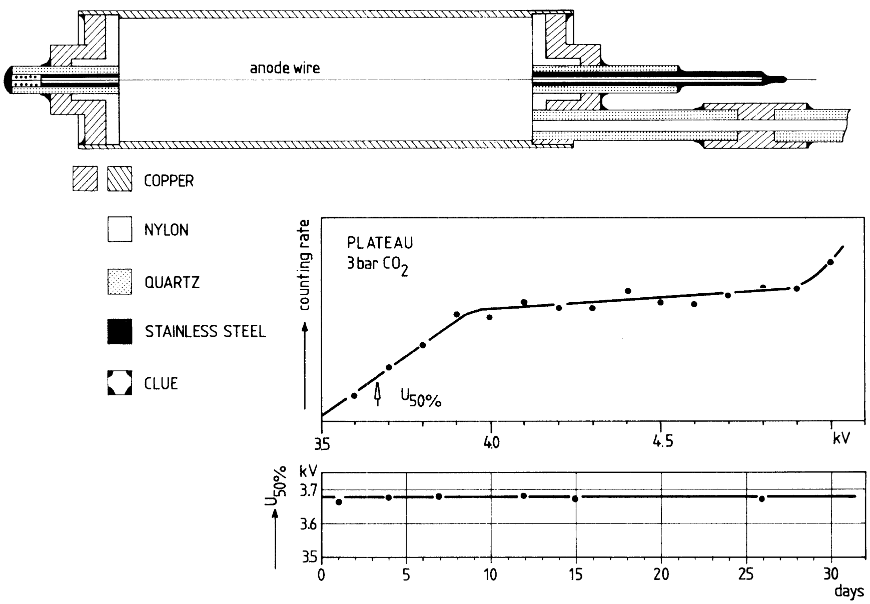

Fig 1. Scheme of the Hannover miniature counter, its plateau, and long-term constancy of the "half voltage" $U_{50\%}$

TABLE 1
Specifications of various types of counters

Construction material	Volume (ml)	Pressure (bar)	Quantity (mg C)	Background (cpm)	Standard (cpm)
Techn-grade quartz	10	3	15	0.18	0.20
Suprasil quartz	10	3	15	0.14	0.20
OFHC copper	15	3	22	0.04	0.28
Otlet system	5	4	10	0.02	0.15
Kaihola system	10	7.5	37	0.0625	0.50

the contrary, that synthetic quartz has the lowest specific activity. High-purity aluminum is being tested.

Otlet *et al* (1983) use separate high-voltage power supplies for each counter. Our studies show that two counters can be easily operated with one power supply if costs (Table 2) must be cut.

Results of a representative long-term test are shown in Figure 1. Using high-purity CO_2 as counting gas, measurements of at least 4 weeks show no change in plateau. The anticoincidence counting rates fit Poisson statistics during this time.

Memory effect was checked with a measurement of 3 to 5 days using the Heidelberg standard (Kromer, 1984) with an activity of 10.3 times the NBS oxalic acid standard, pumping for only 2 to 3 hr, followed by four weeks of background counting. The memory effect was <0.4%. Improvement is possible by lengthening pumping time.

We use a stationary counter filling line which reduces the effective volume of the counter by 10 to 12%. A counter with an effective volume of ca 92% is being constructed.

SHIELDING AND GUARD COUNTER

An inexpensive 10cm shield constructed with low-level lead (Gruvaktibolag, Gotenborg, Sweden) is used. Taking into account the experience by Otlet *et al* (1983), common well-type NaI(Tl) scintillation detectors (HARSHAW Chemie GmbH, Wermelskirchen, FRG) are used as guard counters. But instead of large crystals, two small ones were chosen to save 80% of the costs and to be more flexible in case of defects. The most eco-

TABLE 2
Costs of a dating system with 10 miniature counters ($1.00 = ca DM 3.00)

2 NaI(T1) well-type crystals with photomultipliers	DM 33,000
1 lead shielding (10 cm thick) for two guard counters	DM 15,000
10 low-level miniature counters (construction materials)	DM 2,000
1 chemical preparation line made of glass, pumps, etc	DM 7,000
1 filling line with 10 storage bulbs and pressure gauge	DM 6,000
10 preamplifiers, amplifiers and discriminators	DM 6,000
1 Commodore personal computer, disk drive, and printer	DM 5,000
6 high-voltage power supplies	DM 13,500
Total	DM 87,000

nomic choice was a 6 in^2 crystal with a 6.1 × 12cm hole in an aluminum casing and a 5″ photomultiplier (model EMI 97918) with magnetic shield. The crystal surrounding 4 or 6 counters has an active thickness of 4.5cm.

A test showed that the background counting rate n_0 is not lowered using a flat guard counter in the roof shielding (Gulliksen & Nydal, 1979). Figure 5 shows that even lowering n_0 by 60% would not improve dating precision significantly.

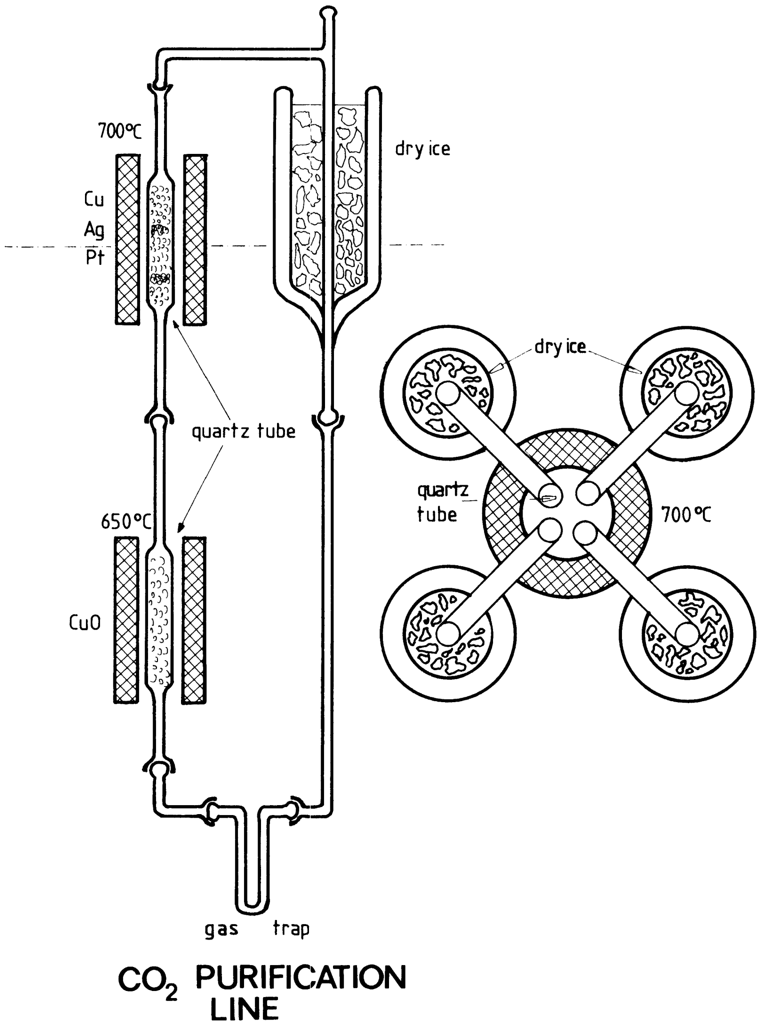

Fig 2. Scheme of the semi-automatic purification line for CO_2, which is circulated through hot CuO and Cu, Ag and Pt for two hours; cross-section of the planned fourfold system

ELECTRONICS

Otlet *et al* (1983) use separate charge pre-amplifiers, amplifiers with pulse-shaping, discriminators and peak stretching for each counter, which are connected to an analog multiplexer, ADC, and logic circuitry. The data are stored with a micro-processor. High-resolution pulse-height analysis (Otlet *et al*, 1983) and time-series and rise-time analysis (Kaihola, Polach & Kojola, 1984) are recommended.

Our electronics are similar, although we use only 18 channels for each counter and 6 for routine work. We have found in routine measurements that more detailed information is needed only during test phases or the exceptional case of a counter malfunction.

COUNTING GAS CHEMISTRY

Carbon dioxide was chosen as the counting gas because the chemical preparation is simple and fast with minor contamination problems since chemicals or catalysts are not needed. A semi-automatic preparation line has been constructed (Fig 2).

High-yield oxidation of various substances such as charcoal, peat, humic acids, collagen, and wood showed that CuO does not work satisfactorily as an oxidant. Burning in a 6L glass bulb filled with 0.9 bar oxygen is successful. The sample is ignited electrically in a stainless steel boat and the

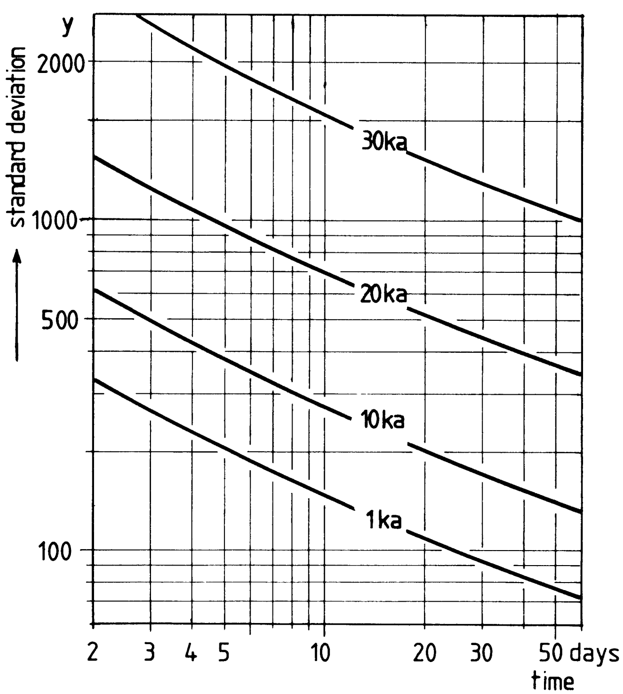

Fig 3. Increase in dating precision as a function of measuring time and the age of the sample

CO_2 is separated from the resulting gas mixture by condensation with liquid air.

For purification, CO_2 (at a pressure of 0.5 to 1 bar in a 0.08L convection line (Fig 2) containing a water trap) is first circulated in a tube filled with CuO and Pt-asbestos at 650 to 700°C for two hours to oxidize any remaining hydrocarbons. CuO is used instead of adding oxygen to the CO_2 during purification (Srdoc & Sliepcevic, 1963) and makes it possible to semi-automatize the process. After 2 hours, the gas is circulated for 2 hours through a tube filled with Cu and Ag heated to 700°C. In this step, nitrogen oxides are reduced to nitrogen, halogens and sulfur oxides are absorbed. No loss of CO_2 occurs. A maximum of 20 min of active work is needed for the preparation of one sample.

Four purification systems will operate simultaneously and will not need any maintenance. The purity of the gas is tested by plateau measurements with a counter installed in the line. The purified CO_2 is stored for one month in glass bulbs before measurement.

After more than 30 purifications, the Cu is regenerated with hydrogen at 600°C for 15 min.

Preliminary attempts with the $CaO/CaCO_3$ purification technique were not successful. Apparently, gaseous impurities were enclosed in the

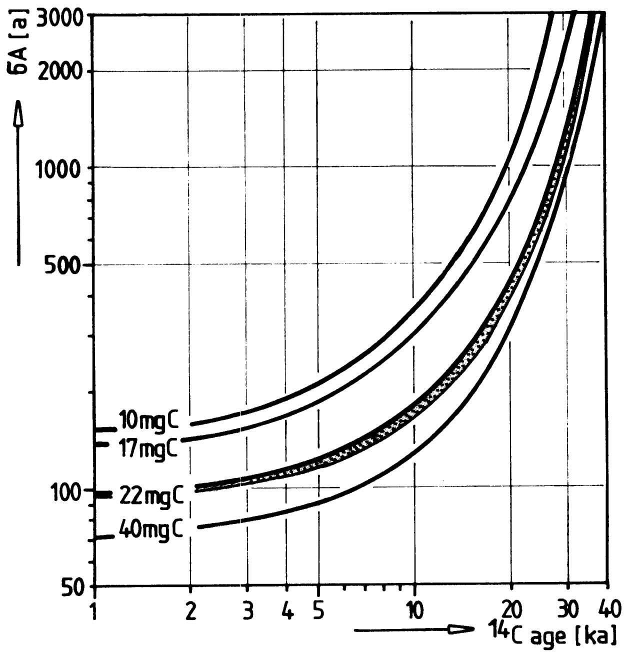

Fig 4. Dating precision of different miniature counting systems for various sample sizes. Ten and 40mg are used by Otlet *et al* (1983) and Kaihola *et al* (1984), respectively. Our quartz counter needs 17mg C. The copper counter is filled with CO_2 of 22mg C. The dotted area shows the improvement if the background of this counter is reduced by 60%.

pores of the encrustations and therefore could not be removed by pumping.

MEASUREMENTS AND STATISTICAL EVALUATION

Ten counters are used simultaneously but independently of each other. A counting time of ca 3 weeks is usually sufficient to provide dating precision of 1 to 2% for Holocene samples (Fig 3).

Further lowering the background counting rate does not improve this figure significantly (Fig 4).

A statistical evaluation of the raw data is made every day for each counter to determine whether the sample should continue to be measured (Fig 5). This simple statistical treatment seems to be sufficient for routine work. Calculation shows how an extension of the counting time may be expected to increase dating precision. This information is useful as sample submitters usually request maximum dating accuracy without considering eco-

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--------------------------------------------------------------------------------
Hv 12986                                           11.06.85  -  12.06.85
--------------------------------------------------------------------------------
Zählrohr          : 80                 Anlage                 : 1
Nulleffekt (cpm): .0406+-0 0008        Standard       (cpm): .2824+-0 0030

Füldruck (torr): 2280                  U-50%               (V): 4410
Messzeit   (min):  900                 Messintervall (min):  100
--------------------------------------------------------------------------------
                           5mV <-              Anti (ADM)              -> 100 mV
 Nr Anti  Coin Guard
--------------------------------------------------------------------------------
 1 -  17 + 9.17  1375   1  0  1  1  0  0  1  1  0  3  1  1  0  0  1  1  0   5
 2 -   7 + 8.87  1381   1  0  0  1  1  0  1  0  0  1  1  0  0  0  0  0  0   1
 3 +  13 + 9.07  1376   1  0  0  0  0  0  2  0  1  1  2  1  2  0  0  0  0   3
 4 +  10 + 9.13  1364   0  1  0  0  0  1  1  2  1  0  0  0  0  0  0  0  0   4
 5 +   9 + 9.14  1377   1  0  3  0  1  0  0  1  1  1  1  0  0  0  0  0  0   0
 6 +   9 - 8.67  1373   2  0  1  0  1  0  0  0  0  0  1  0  0  0  0  0  0   4
 7 -  18 + 9.09  1378   2  1  3  2  1  1  1  0  0  0  2  1  0  0  1  1  0   2
 8 +  13 + 8.89  1379   2  1  0  0  0  0  1  1  2  0  0  1  1  0  0  0  0   4
 9 +  15 + 8.90  1374   0  0  0  1  0  0  0  1  0  6  0  1  0  2  0  0  0   4
--------------------------------------------------------------------------------
sm                     10     8     4     7     5     8     3     2     0
                          3     5     2     6    12     5     2     2     27
--------------------------------------------------------------------------------
           cpm    1s      2s     ex
--------------------------------------------------------------------------------
Antis  :          6       3      0
Koinz  :  8.9     8       1      0

Kanal 1:   10
Kanal 2:   40     6       2      1
Kanal 3:   34     4       4      1
Kanal 4:   27

Kanal 2/Kanal 3          = 1.176 +-  0.274
--------------------------------------------------------------------------------
Zählrate (cpm):  0.1233 +- 0 0117

pcm     (%mod):   29.29 +-    4.17

Alter  (abp)  9862 +-   1140 nach  0.6 Tagen            (- 300 a/d)
                   +-    576 nach  2.5 Tagen            (-  65 a/d)
                   +-    413 nach  5.0 Tagen            (-  30 a/d)
                   +-    353 nach  7.0 Tagen            (-  13 a/d)
                   +-    259 nach 14.0 Tagen            (-   5 a/d)
                   +-    219 nach 21.0 Tagen            (-   3 a/d)
                   +-    196 nach 28.0 Tagen
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Fig 5. Printout of raw data and results of the statistical treatment for a sample nomic aspects. Counting time is extended only if it is shown that the standard deviation can be expected to decrease significantly.

COSTS

The costs of the system are specified in Table 2. This system allows dating of 160–180 samples per year, including background and standard measurements. Chemical preparation takes 20 days (DM 4000). With DM 1000 for chemicals as well as DM 8000 for amortization and maintenance, dating costs are ca DM 100 per sample.

SUMMARY

A low-cost miniature counter system for ^{14}C dating of 22mg samples with CO_2 is presented which, comparing both cost and specifications, is quite capable of competing with commercial systems (Fig 5). Costs do not exceed $30,000 and the dating expenses are ca $35 per sample.

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