[Radiocarbon, Vol 25, No. 2, 1983, P 565-575]

DEVELOPMENT AND OPERATION OF THE HARWELL SMALL COUNTER FACILITY FOR THE MEASUREMENT OF ^{14}C IN VERY SMALL SAMPLES

R L OTLET, GEORGE HUXTABLE, G V EVANS, D G HUMPHREYS
T D SHORT*, and S J CONCHIE

Nuclear Physics Division, AERE Harwell, Oxfordshire, OX11 ORA
United Kingdom

ABSTRACT. The Harwell system for measuring milligram size samples using Brookhaven miniature gas counters is fully operative. It comprises 12 counters of different sizes which operate simultaneously within a single NaI crystal (300mm diameter x 300mm long) acting as an anti-coincidence guard counter. Brief details are given of the construction and commissioning of the system, including counter assembly, shield design, electronics, data capture, data analysis, and chemical processing and filling procedures. The performance of the system and an overall view of the fields of application for which the counters have important applications are discussed.

INTRODUCTION

This paper describes the development and first few months of operation of the Harwell small sample ^{14}C measurement facility which uses Brookhaven miniature gas counters (Harbottle, Sayre, and Stoenner, 1979). The collaboration with Brookhaven National Laboratory began in the initial testing of the counters and the design and planning of the proposed facility with its multi-counter array were previously described (Sayre et al, 1981; Otlet and Evans, in press). Up to that time, work had been confined to test, background and calibration samples with no actual samples.

COUNTER ASSEMBLY AND SHIELD

Six 5mℓ 'micro-counters' and six 30mℓ 'mini-counters' which, filled to 4 atmospheres pressure of carbon dioxide, mean equivalent quantities of ca 11mg and 66mg respectively, are employed.

* Computing Centre, North Staffordshire Polytechnic, Blackheath Lane, Stafford, United Kingdom

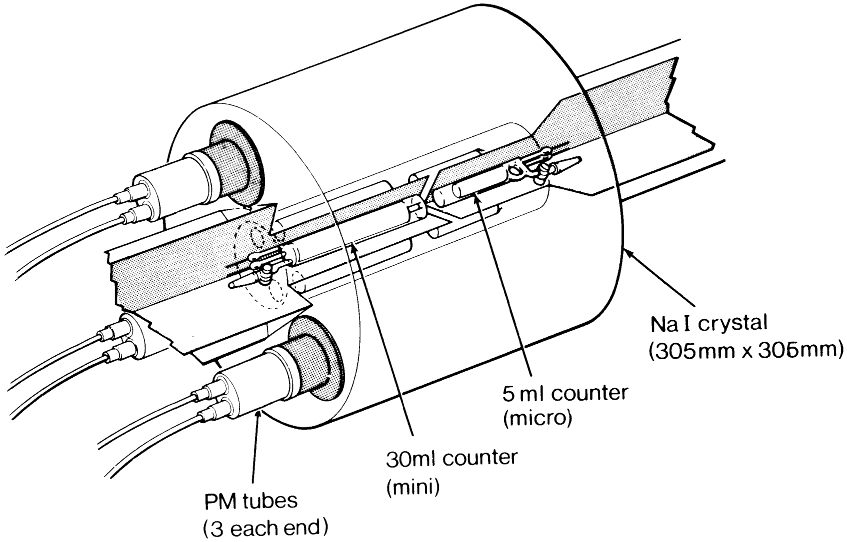


FIG 1. **NESTING OF COUNTERS IN GUARD COUNTER CAVITY**
Showing a 'micro'-counter (5 ml) back to back with a 'mini'-counter (30ml) in one of the segmented compartments

Figure 1 shows nesting of the full array of counters inside the guard counter, which is a large, 300mm diameter by 300mm long, NaI crystal with three photomultiplier tubes on opposite parallel faces (six in all), providing annular shielded volume for the counters ca 90mm internal diameter. The complete assembly, guard counter, and gas counters are further shielded within a large passive shielding castle. The shield comprises old steel, total thickness 230mm, with an inner layer of boron-loaded paraffin wax (neutron shield) arranged in the order, 204mm steel, 150mm paraffin wax plus 26mm steel as the inside layer. During commissioning we found it necessary to contain each counter in its own individual metal (copper) box. This design feature greatly assists removal for counter filling and minimizes spurious cross-talk-type interference among neighboring counters in operation. The individual, segment-shaped containers are illustrated in plate 1, designed to pack numerous counters (six at each end) inside the concentric cavity of the guard counter, with the EHT filter components and pre-amplifiers as close to the counter as possible, as well as to leave space for the photomultiplier tube terminations.

Table 1 lists characteristics of the counters operated within the shield and, in the case of the micro-counters, can be compared with the Brookhaven values. The background so far obtained, although very low and acceptable from an operational point of view, is not so low as the best Brookhaven result, the achievement of which will be further considered. On the other hand, the modern carbon to background ratio achieved with the larger mini-counters (∿ 8) is close to the Brookhaven value and compares favourably with large sample scintillation counting.

TABLE 1. Comparison of some miniature counter characteristics in different shields

Guard shield	Place	Counter Volume (mℓ)	Background (counts/day)	Modern carbon to background ratio
Conventional multi-gas counters	Brookhaven	5	56	3.3
Conventional multi-gas counters	Harwell	30	180*	5.3
Conventional multi-wire gas counter	Harwell	5	124	1.5
Conventional multi-wire gas counter	Harwell	30	400	2.4
254mm x 254mm NaI crystal	Brookhaven	5	21	8.9
300mm x 300mm NaI crystal	Harwell	5	30	6.2
300mm x 300mm NaI crystal	Harwell	30	130	7.5
300mm x 300mm NaI crystal	Harwell	10	80**	∿4.7
Gas multi-wire counter with inner plastic phosphor shield (10mm thick)	Harwell	5	72	2.6

*Approximate result with a 4-atmosphere filling of P10 gas
** Counter tested for Wallac, Finland - filled to 4 atmospheres CH_4
Other counters all of Brookhaven origin - filled to 4 atmospheres CO_2

https://doi.org/10.1017/S0033822200005877 Published online by Cambridge University Press

The validity of the decision to use the large NaI crystal as the guard counter instead of a less expensive gas proportional or plastic phosphor type is still under investigation. The comparative tests with a multi-wire gas proportional guard counter operated in the same passive shield (table 1) support the Brookhaven results and our decision. Further tests will include a direct comparison with a plastic phosphor guard shield.

ELECTRONICS AND DATA HANDLING

Recent improvements in technology, eg, monolithic integrated circuit components and micro-processor data capture have been fully exploited, facilitating multi-counter operations and providing stability throughout the counting times. The electronics hardware is made at Harwell using standard, commercially available components. The full system, including micro-processor and computer display, acts as 14 multi-channel analysers, each of 512 channels and split-store operational capability. Additional features of the Harwell circuitry enable 1) full spectrum display of both coincident (cosmic) and anticoincidence (sample and background) pulses, 2) pulse height analysis of events recorded simultaneously in any counter (without which it would be impossible to record the coincident spectra of every counter separately), 3) variable dead time (blanking time) following guard counter (veto) pulses. The relatively high count-rate of the large NaI crystal detector and the excessively large amplitude range of pulses ca 1000:1 made this necessary. Figure 2 is a simplified schematic diagram of the electronics and data handling circuitry EHT for the 12 counters which are currently in operation is individually provided from a single unit (Le Croy 4032A) which uses plug-in EHT pods. Positive high voltage is used, applied (after filtering) directly to the anodes, the signal being taken from the cathode terminations at ground level. This particular connection arrangement means that any spurious break down pulses in the EHT filter circuit have an opposite polarity to the genuine counter pulses and is more convenient from the point of view of the physical layout in the confined space of the counter box.

Output from the preamplifiers is passed to the amplification, pulse shaping, discrimination and peak stretching (sample and hold) circuits. All are internally connected to a common logic board which includes the analogue to digital converter and logic circuitry which encodes all signals ready for acceptance and storage by the dedicated micro-processor (NASCOM). Data transfer is effected on a priority interrupt basis and in the case of multi-events (simultaneous triggering of more than one counter at a time) a hierarchical system reads

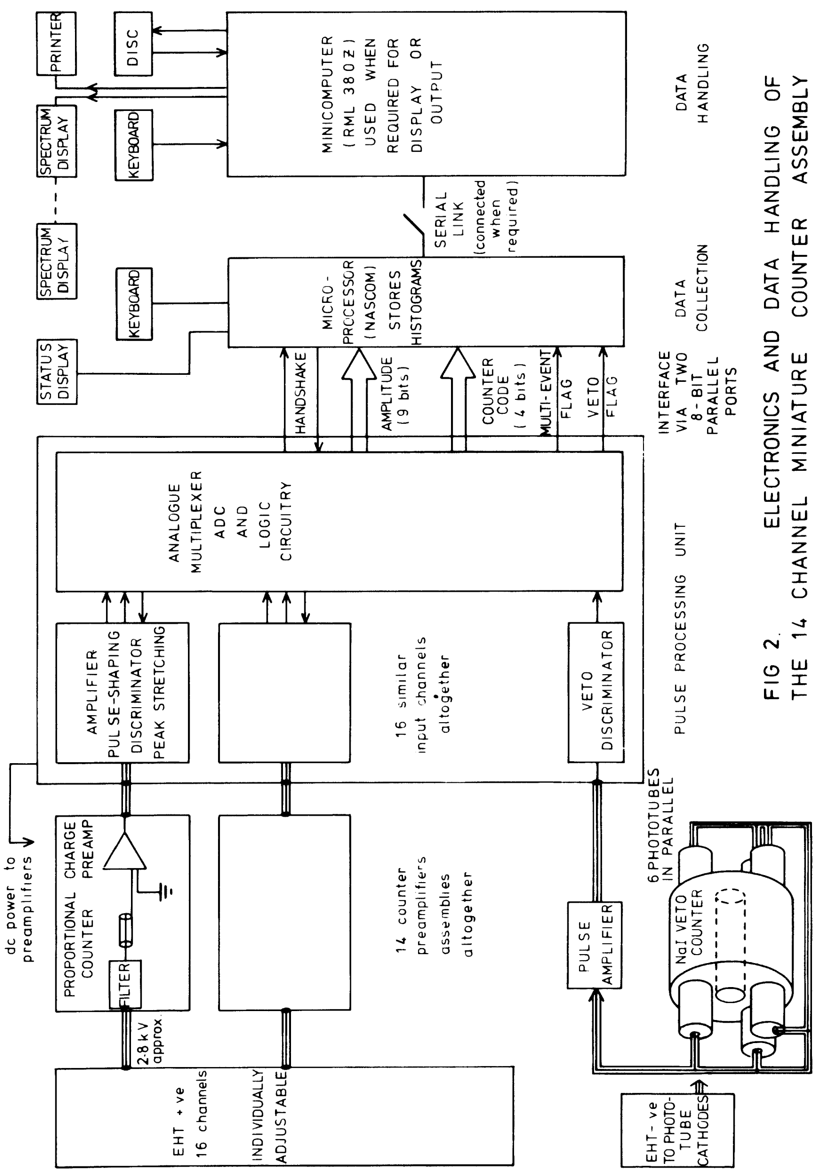


FIG 2. ELECTRONICS AND DATA HANDLING OF THE 14 CHANNEL MINIATURE COUNTER ASSEMBLY

PLATE 1. Individual counter assembly

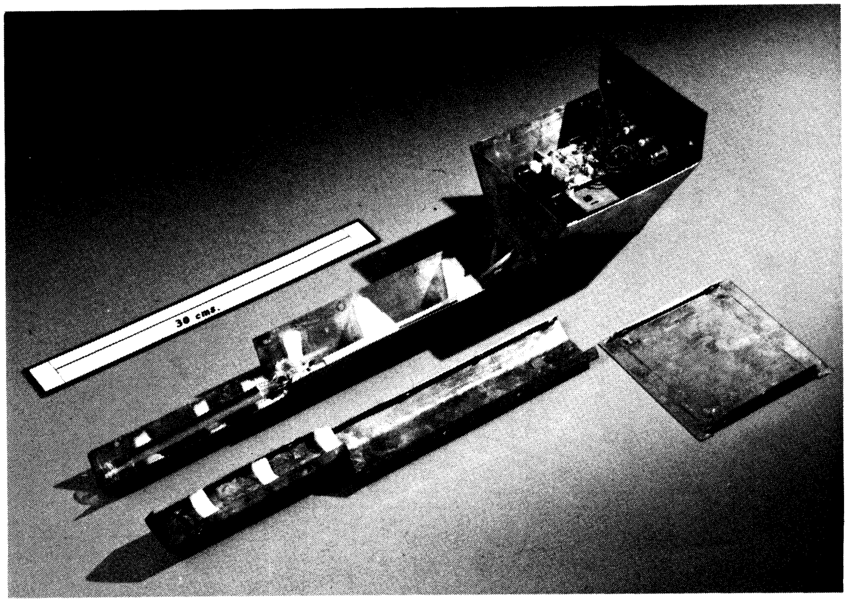
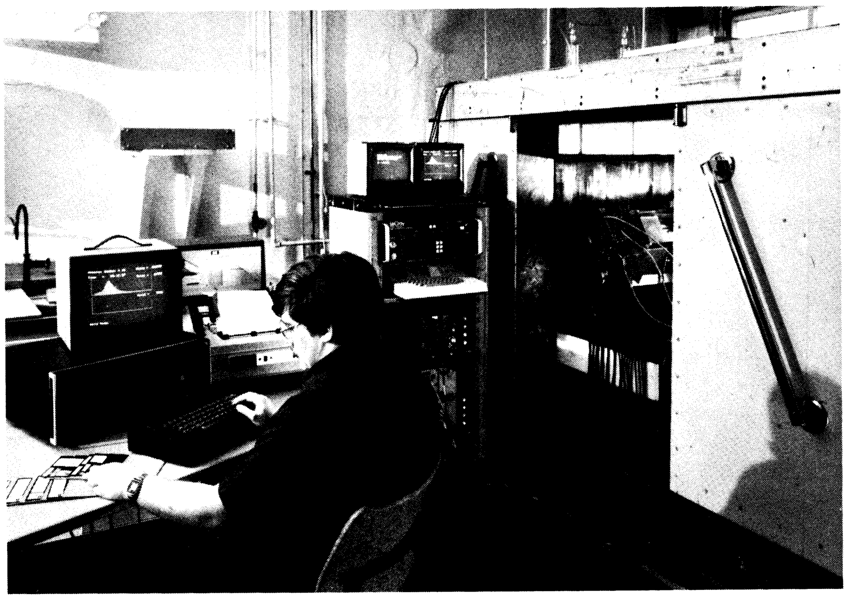


PLATE 2. Complete set-up - shield, counter assembly, electronics, and micro-computers



https://doi.org/10.1017/S0033822200005877 Published online by Cambridge University Press

from each counter in turn whilst the peak stretching facility holds the sampled pulse waiting for its turn to transfer. Priority interrupt also ensures that data transfer from the counter electronics is not impeded by read-out of the Nascom stores by the data handling general purpose computer.

Data collection and handling is enacted by two Z80 based devices. The first is a simple, inexpensive extended memory (16K) microprocessor (NASCOM), the basic function of which is to accept pulses from the counters and to store them as histograms according to the encoded label. For each counter 256 locations are for multi- and veto-flagged events and 256 for non-flagged pulses. The NASCOM is permanently connected to the counter electronics circuitry and buffers the system breakdowns or malfunction caused by other activities of the second, more sophisticated, general purpose micro-computer (RML 380Z). This second computer, which includes full facilities for high resolution graphics provides display of the counter pulse-height spectra, analysis and long term (back-up) floppy disk storage. In these first few months of operation, display of spectra has been made frequently but the dumping to disk should only need to be carried out twice a week, each time resetting the NASCOM stores to zero. This procedure enables possible problems to be spotted rapidly enough and at no time is there any risk of losing a set of data which may have taken up to 7 days to collect. A photograph of the complete shield, electronics and data-handling set-up is given in plate 2.

INTERPRETATION OF SPECTRA

Figure 3 A-D shows a set of print-outs derived from displays of spectra obtained with the micro and mini-counters under different conditions. Each display is a composite of the coincident (cosmic) spectrum, above, with the anticoincident spectrum below. Counts per channel are plotted against channel number which, in terms of energy, is linear up to channel 255. Counts below channel 10 are cut off by the preset discriminator contained in the pulse processing unit (see fig 2). The displayed range represents ca 2 KeV to 59 KeV for both sizes of counter. All pulses above the equivalent energy of channel 255 (> 59 KeV) are dumped into channel 256 but are included in the total count calculations. A new counter filling is set up, using an external ^{241}Am or ^{238}Pu gamma-ray source to excite the Kα X-ray spectrum off the iron cathode, before the counter is put into the shield. The peak is moved to channel 31 (both sizes of counter) by adjusting the EHT and forming the primary energy calibration of the channel's scale (6.4 KeV at channel 31). The sharpness of the peak is important for assessing the quality of the filling gas (Sayre et al,

https://doi.org/10.1017/S0033822200005877 Published online by Cambridge University Press

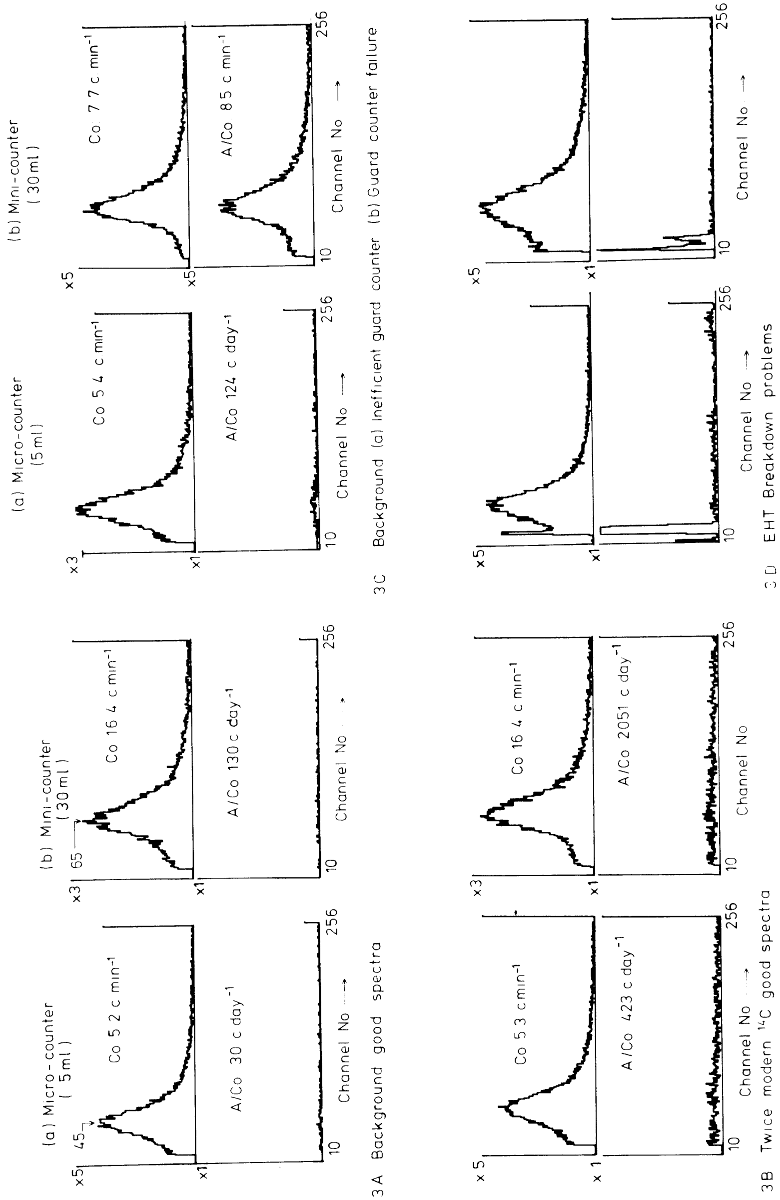


FIG 3 EXAMPLE COINCIDENT / ANTICOINCIDENT SPECTRA FROM "MICRO" AND "MINI" GAS COUNTERS

https://doi.org/10.1017/S0033822200005877 Published online by Cambridge University Press

1981). A resolution of the order of 25% should be obtained or repurification will be necessary.

The counter is then put into the shield and the position of its cosmic peak and its resolution determined. The occurrence of these peaks, shown in the upper half, coincident, spectra, in fig 3 at ca 9.5KeV and 13.6KeV for the micro- and mini-counters, respectively, are characteristic of gas counters generally (Wolfendale, 1963). They are produced from charged particles (muons) causing ionization during passage through the counter, the total energy deposited being a function of the product of the track length possible in the dimensions of the counter and of the gas density for the particular gas filling, thus it reflects the pulse produced by the passage of such a particle across the diameter of the counter and for larger counters filled to the same pressure, the equivalent energy it represents appears at higher levels. The peak is used to provide a continuous monitor of the gas quality and electronics (EHT and gain) stability throughout the long counting periods. This technique has been used for many years in our laboratory for ^{14}C gas counting, having been developed from a similar procedure we used in ^{3}H counting (Otlet, 1968) but has also been reported independently by Oona and Fan (1977). No change in either its position or resolution is expected even after 70 days of counting.

Examination of the anticoincidence (sample) spectra also provides valuable confirmation of valid counting. Figure 3A and B shows good background and ^{14}C (2 x modern) plus background spectra for both sizes of counter. The difference between these good spectra and the others, which demonstrates problems, is clearly seen. So far, sporadic EHT breakdown has been the only problem which has had to be contended with and the ability to identify it and to cure it quickly on the occasions when it has arisen has been invaluable.

GAS PURIFICATION AND COUNTER FILLING

Sample preparation basically follows the well established procedures for gas counting with CO_2 (Srdoč and Sliepčevic, 1963; Sayre et al, 1981) but using rigs suitably scaled down for the smaller size of samples being handled. Dry reagents are used throughout, the essential stages of the final purification being thermal cycling for 2 to 3 hours, first over copper at 700°C and afterwards over silver and platinum at the same temperature in the presence of a small quantity of added oxygen. The last step is the passage of the CO_2 through activated charcoal at 0°C as Bruns (ms) followed by freezing and pumping.

The filling procedure is similar to that which Brookhaven uses (Harbottle, Sayre and Stoenner, 1979). Mercury under

https://doi.org/10.1017/S0033822200005877 Published online by Cambridge University Press

pressure is used to compress the purified CO_2 into the counter. The mercury passes through the pre-filling measurement vessel and connecting lines right into the counter filling leg stopping only 2-3mm short of the counter sensitive volume. In this way, dead volume is absolutely minimized and no loss of sample CO_2 occurs even for samples which yield amounts very close to the minimum required to fill the counter. When the counter stopcock is closed the mercury above it helps the seal and prevents the gas making contact with the stopcock grease.

CONCLUSIONS

Operation of the 12-counter assembly began in November, 1981. Since then a simultaneous programme of sample measurements and calibration of the counters has been operated. Preliminary experience with the multi-counter set-up suggests that miniature counters have a promising future for low-level ^{14}C measurements in a variety of applications including Art, Archaeology, Hydrology and the Environment. The obvious limitation of counting time required for acceptable statistics is minimized when several counters are operated simultaneously. For many applications, the sample quantities available enable use of the larger sized counters and a corresponding reduction in counting time for a given measurements precision to be realized (table 2).

TABLE 2. Poisson precision from micro- and mini-counters

Age of sample (years)	± 1σ precision*(%)					
	Micro-counters (5mℓ)			Mini-counters (30mℓ)		
Count time (days)	30	60	90	15	30	60
Modern	1.5	1.1	0.9	1.0	0.7	0.5
1000	1.7	1.2	1.0	1.0	0.7	0.5
2000	1.8	1.3	1.0	1.1	0.8	0.5
5000	2.3	1.6	1.3	1.4	0.9	0.7
10 000	3.6	2.5	2.1	2.1	1.5	1.1
20 000	9.6	6.8	5.5	5.7	4.0	2.8
30 000	18.4	13.0	10.6	17.0	12.0	8.5

* Represents minimum errors in the full procedure, based on the statistics of the estimated sample and background counts only

Modern electronics simplify the operation of many counters simultaneously and their small size allows for containment within an efficient guard counter shield. Experience with a large NaI crystal guard counter reinforces Harbottle, Sayre, and Stoenner's (1979) conclusions that a substantial reduction in background, over and above conventional gas proportional guard systems, is obtained by its use.

ACKNOWLEDGMENTS

The continued collaborative support of the Brookhaven National Laboratory (G. Harbottle and staff) in this project is gratefully acknowledged. Also the assistance of our colleagues L L Wassell and D B Punter, in the design and construction of the counter assembly, and D Sanderson (Research Assistant, Bradford University), who has recently joined the Group, for the provision of some of the latest data, should be mentioned.

REFERENCES

Bruns, M, (ms), 1976, Gas chromatography with Radon-222, Doctoral thesis: Inst Environmental Physics, Univ Heidelberg.

Callow, WJ, Baker, MJ, and Pritchard, DH, 1963, National Physical Laboratory radiocarbon measurements I: Radiocarbon, v 5, p34.

Harbottle, G, Sayre, EV, and Stoenner, RW, 1979, Carbon-14 dating of small samples by proportional counting: Science v 206, p683-685.

Oona, H, and Fan, CY, 1977, A counter system for natural ^{14}C measurements: Nuclear Instruments and Methods, v 143, p391-397.

Otlet, RL, 1968, Low-level tritium measurements for hydrological applications, in Nucleonic Instrumentation Conf, Proc: Conf Publication No. 47, Institution of Electrical Engineers.

Otlet, RL, and Evans, GV, in press, Progress in the application of miniature gas counters to radiocarbon dating of small samples: <u>in</u> ^{14}C and Archaeology Symposium, 1st, Proc: Groningen, in press.

Sayre, EV, Harbottle, G, Stoenner, RW, Otlet, RL, Evans, GV, 1981, Small gas proportional counters for the carbon-14 measurement of very small samples, <u>in</u> Methods of Low Level Counting and Spectrometry: IAEA Vienna, p393-407.

Srdoc, D, and Sliepcevic, A, 1963, Carbon dioxide proportional counters; effects of gaseous impurities and gas purification methods: Internatl Jour Appl Radiation Isotopes, v 14, p68.

Wolfendale, AW, 1963, Cosmic Rays: London, Newnes, p46.

https://doi.org/10.1017/S0033822200005877 Published online by Cambridge University Press