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EFFICIENCY AND BACKGROUND VARIATIONS OF A PROPORTIONAL COUNTER WITH CARBON DIOXIDE FILLING

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Sources of detection efficiency variations, namely concentration of electronegative impurities, gas pressure and temperature variations, instability of electronics and barometric effect are studied and a method of their elimination is presented.

I. INTRODUCTION

We have developed a method of low level counting of ^{14}C with an internal proportional counter using a carbon dioxide as a gas filling [1, 2] for radiocarbon dating and ^{14}C concentration measurements in atmosphere and biosphere. The carbon dioxide is prepared from samples of organic or inorganic origin by a combustion in a stream of oxygen or by an acid evulotion. Before filling the counter the carbon dioxide is purified by a vacuum condensation and by passing through furnaces and traps [1, 2].

It is impossible to ensure the stability of all parameters which have an influence on the accuracy of measurements [3]. One of the most important parameters is the amount of electronegative impurities present in the counter filling. Out of the other parameters variations of the working voltage the input sensitivity variations of electronics, pressure and temperature variations of the gas filling, and the barometric effect are also important. These variations cause a change in the gas amplification factor of a proportional counter.

The contributions of the above mentioned variations to total errors of measurements for the 2.3 l proportional counter filled with carbon dioxide are studied in the present paper.

II. VARIATIONS OF DETECTION EFFICIENCY

It is well known that electronegative impurities present in the carbon dioxide filling of proportional counters cause a drop in detection efficiency

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[4—7]. This phenomenon is connected with the attachment of secondary electrons on atoms of electronegative impurities. The gas amplification factor of the proportional counter decreases therefore with increasing concentration of electronegative impurities. At the stable discrimination level of an electronic apparatus and the stable working voltage of a counter, a change in the concentration of electronegative impurities causes a change in the counting rate of the proportional counter.

The amount of impurities (oxygen, water vapours and in samples of organic origin also nitrogen and sulphur oxides and halogens) depends on the CO_2 preparation, i. e. on the absorption media that are partially inactivated after purification of CO_2, on vacuum leakage, on the sample contamination by impurities and on the number of purification cycles. Even if the differences in the concentration of impurities are not large (e. g. in our counter the oxygen concentration is 0.06—0.08 torr), their influence on the counting rate is very important. Fig. 1 shows the counter characteristics for these concentrations of oxygen in the CO_2 filling. The slope of the characteristic for 0.06 and 0.08 torr of O_2 in 800 torr of CO_2 filling is 2.0 and 3.6 %, respectively. At 5.3 kV working voltage and 50 mV input sensitivity the change in the counter background is 10 %. The change in the counting rate of modern ^{14}C standard is even higher, about 12 %.

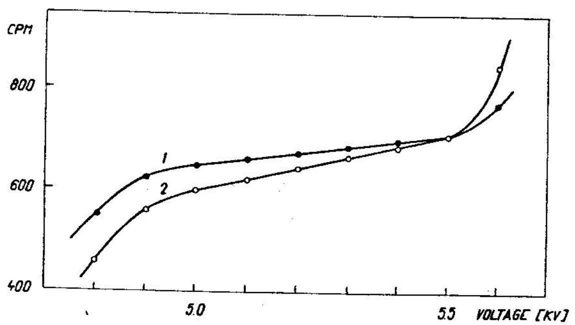

Fig. 1. Working characteristics of the proportional counter. 1— 0.06 torr of O_2; 2 — 0.08 torr of O_2 in 800 torr of CO_2 filling. Background for (1) and (2) curves is 17.6 and 16.0 cpm respectively.

We did not observe a change of the detection efficiency during one measurement with the same CO_2 filling during 48 hours counting time. During longer counting times a decrease of the detection efficiency in copper wall proportional counters is possible [8].

III. PRESSURE AND TEMPERATURE VARIATIONS

The pressure of CO_2 filling in the counter is measured by a mercury manometer. The relative change of the counting rate on the pressure of gas fillig depends also on the ^{14}C concentration in a sample. While the background depends only slightly on a change of gas pressure, the counting rate of the modern ^{14}C concentration in carbon increases considerably with increasing CO_2 pressure. In this case not only the gas amplification factor is changed but the ^{14}C activity increases with an increasing gas pressure. As can be seen from Fig. 2, the error of ± 2 torr of the working pressure represents a 0.05 % change in the counter background, 0.12 % in the counting rate of modern carbon, and 0.15 % for the sample with 100 % excess of ^{14}C content.

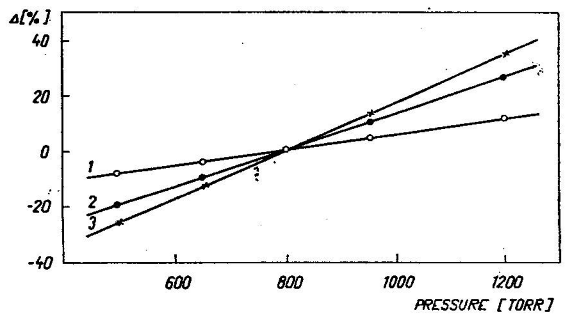

Fig. 2. Relative change of the counting rate as a function of CO_2 pressure. 1 — background; 2 — the modern ^{14}C standard; 3 — sample with 100 % excess above the modern ^{14}C standard.

A change in the counter temperature, Δt, represents a change in the gas pressure

$$p/p_0 = -\alpha \Delta t, \quad (1)$$

where p_0 is the working pressure and α is the coefficient of the temperature expansivness of CO_2.

As the relative change of the detection efficiency is 0.025 %/torr, 3 °C change of temperature of the gas filling will not cause a higher change of the counting rate than 0.2 %.

Trost [9] found a relation between a change of the working voltage and the pressure of the gas filling

$$V = \varepsilon(\Delta p + nk) \ln (r_k/r_a), \quad (2)$$

where ε, k are constants characterizing the gas filling, n is the number of ions produced by an electron on its way to the anode, r_k and r_a are the radii of the cathode and the anode, respectively. Using relation (2) one can determine from the plateau rise the change of the counting rate for the given change of the pressure. From this point of view a 3 °C change in temperature of the gas filling represents a 10 V change of the working voltage.

IV. INSTABILITY OF THE DISCRIMINATION LEVEL AND THE WORKING VOLTAGE

A change of the amplification factor of the linear amplifier or a change of the discrimination level can be characterized by a change of the input sensitivity of electronics. We have found that 1 mV change of 50 mV input sensitivity represents 0.36 % change of the counting rate.

The high voltage power supply used for the proportional counter is refilled with batteries. The instability of high voltage during one measurement does not exces ±5 V, which represents 0.12 % error in the counting rate.

V. BAROMETRIC EFFECT

The intensity of cosmic rays and the counter background depend on the barometric pressure. The barometric effect of gas couters can be essentially lowered uslowered using an anticoincidence shielding. The contribution of the hard component of cosmic rays to counter background is in this case much lower. The number of μ-mesons M, registered in the counter is a function of barometric pressure [10].

$$M = M_0 \exp(-B/B_0), \tag{3}$$

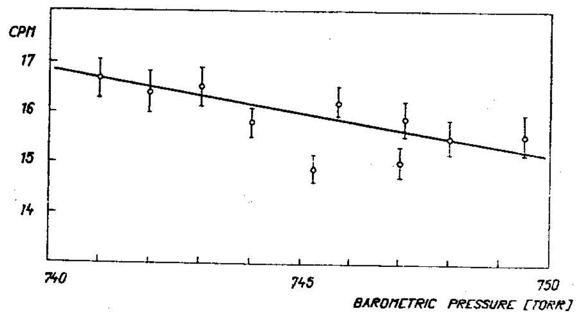

Fig. 3. Dependence of the background on the barometric pressure. The errors represent the maximum standard deviations.

where B_0 is the normal barometric pressure and M_0 is the corresponding number of μ-mesons.

After derivation we obtain

$$\frac{dM}{dB}\frac{1}{M} = -\frac{1}{B_0}. \tag{4}$$

The left-hand side of equation (4) represents the relative change of the registered number of μ-mesons. It is clear that this does not depend on the barometric pressure. Fig. 3 shows the dependence of the counter background on the barometric pressure. Using the least square method the relative change of the counter background due to barometric pressure was determined to be 0.86 %/torr. The motion of air masses and the changes in the atmospheric temperature and their influence on the cosmic ray intensity were not taken into account.

VI. EFFICIENCY AND BACKGROUND CORRECTIONS

The standard deviation of a sample count rate with a modern ^{14}C concentration after 20 hours of counting is 1.4 %. Variations of the detection efficiency and the background increase the total error of the measurement. In Tab. 1 the contributions from different sources of errors are given a sample with a modern ^{14}C concentration.

It is clear from Tab. 1 that the main contribution to the total error of the measurement is the variation of electronegative impurity concentration in the carbon dioxide filling.

A drop or an increase in the detection efficiency can be compensated by a change in the working voltage of the proportional counter. Besides changes in impurity concentrations one can compensate in this way also a change in the input sensitivity or the gas temperature for different fillings. A 20 % change in the input sensitivity can be compensated by a 120 V change of the working voltage.

Table 1

Contribution to the total error of the measurement

Origin of error	Maximum error %
Statistics	4.2
Instability of the discrimination level	0.5
Instability of the high voltage	0.2
Change in the concentration of electronegative impurities	8.0
Change in the gas pressure	0.2
Change in the gas temperature	0.5

It is clear from Fig. 1 that the counting rate will differ markedly if the working point is chosen in the middle of the plateau curve. This procedure would necessitate plotting the whole plateau characteristics, which is always inaccurate and tedious or even impossible in the case of low level counting of soft beta emitters.

The method of external calibration is more accurate and quicker. In this case it is not necessary to measure the counter plateau for every new filling of the counter. The plateau curve is taken only once when the nominal working voltage and the reference counting rate with a gamma source at this voltage are determined. When a new gas filling is used, it is enough to find the gamma counting rate at the nominal voltage. If this counting rate differs from the reference one, the voltage is adjusted so that the reference counting rate is obtained. We have used for the external calibration ^{137}Cs standard. The activity of the standard was 4 μCi. The standard is during the calibration

Table 2
Reproducibility of the counter background measurements

Counting rate [cpm]	Voltage correction [V]	Background [cpm]	Standard error [cpm]
15.51	0	15.25	0.13
15.56	70	15.44	0.12
14.85	0	14.29	0.11
15.06	0	14.52	0.07
15.77	100	15.07	0.12
16.66	200	16.06	0.15
16.52	110	15.52	0.14
16.40	120	15.32	0.09
15.52	160	15.12	0.13
15.96	−20	15.58	0.14
16.03	−10	15.55	0.15
15.82	90	15.14	0.12
15.85	30	15.44	0.15

Table 3
Reproducibility of modern ^{14}C standard measurements

Voltage correction [V]	Counting rate [cpm]	Standard error [cpm]
200	13.97	0.40
0	13.68	0.34
260	14.10	0.45
210	13.67	0.34
140	13.91	0.45
70	13.74	0.42
−10	13.75	0.30
40	13.92	0.36

reproducible placed on the counter body. The standard counting rate is determined with the maximum deviation of 1 %.

In Tab. 2 and 3 the corrected results of the background and the modern ^{14}C standard (National Bureau of Standards, Washington D. C., USA) measurements are listed. The concentration of oxygen in the samples was 0.05—0.1 torr. The nominal working voltage was 5.300 V. The results of background measurements were corrected in relation to the barometric effect and the results of modern ^{14}C standard measurements were corrected in relation to background.

VII. CONCLUSIONS

Variation of electronegative impurity concentrations present in a carbon dioxide filling of a proportional counter cause changes of the detection efficiency. The contribution of this effect to the total error of the measurement is two times higher than the statistical error. Contributions from other sources of errors are negligible in comparison with the statistical error (Tab. 1). For elimination of detection efficiency variations we have developed the method of the external calibration using the ^{137}Cs standard.

The results of modern ^{14}C standard measurements obtained by this method show that the main errors of measurements are due to poor statistics. The background after a correction on the barometric effect (0.86 %/torr) has an error below the maximum standard deviation.

Using the above mentioned corrections the total error of ^{14}C measurements at natural level is about 2/3 lower than without these corrections.

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