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Low Level Proportional Counter for Tritium

by

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G E N E V A

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A B S T R A C T

A proportional counter for the measurement of low Tritium activities is described. The cylindrical proportional counter (principal counter) with a volume of about 600 cm^3 is the inner part of a low level counting system in which the guard counter (anti-coincidence counter) surrounds the principal counter which is only separated from the annular counter by a thin foil. The ratio of the "transmit" to the "out" counting rate is high compared to other low level counting systems (of the order of 300). The specific feature of the design is the separation of the volume of the internal counter and the anti-coincidence counting ring. The special properties of the counter and some of its operations are discussed.

A. Introduction

The very short range of the β particles from the Tritium, 0.1 mg/cm^2 ¹⁾ for the average energy $\bar{E}$ of 5.6 KeV, require special counting techniques for their detection, because, for example, the Tritium is always brought into the sensitive volume of the detector and mixed with the counting gas or in liquid form with scintillating liquids. In our particular case the Tritium was produced as a spallation product in meteoric matter under the influence of cosmic radiation, and it was mixed with large quantities of hydrogen. The counter has therefore to contain fairly large quantities of hydrogen and at the same time be able to measure accurately net count rates of the order of 0.2 to 5 cpm. In order to make measurements in a reasonable time special low level counting techniques are necessary to ensure a suitable low background.

At low count rate the statistical error σ is proportional to $\frac{\sqrt{B^*}}{S^*}$, (App. A), where B^* is the background rate and S^* the net counting rate of the sample. A counter minimising this value is required for these particular Tritium measurements. The background of a counting system arises from the following causes:

- a. The charged component of the cosmic radiation;
- b. the radioactivity of counter material and counting gas;
- c. the effects of the neutral components of the cosmic radiation and neighbouring radioactive materials in the counter which give rise to a count.

To reduce the charged component of the cosmic radiation as well as the surrounding activity, the counter is placed in heavy shielding. The penetrating particles trigger a guard counting ring which vetoes a simultaneous event in the principal counter.

A high guard counter efficiency is indispensable. The radioactivity of the counter itself can be reduced by selecting

suitable material.²⁾ The effect of the uncharged component of the cosmic radiation and the γ radiation from the surroundings can be reduced by selecting material for the counter wall of low Z and by keeping the wall of the principal counter as thin as possible.

Working in the proportional region has the advantage of being able to limit the counting to the energy regions of Tritium. In this way the counter background is reduced without reducing to any great extent its efficiency to Tritium radiation.

Houtermans and co-workers³⁾ have already developed a quite suitable annulus counter in which the guard counter ring is incorporated in the same counter tube and only separated by a thin foil from the principal centre counter. In this and many other counting devices⁴⁾ the counting gas and the Tritium to be measured circulate in the whole counting tube as well as in the centre counter volume and in the guard counter volume. This forces one to keep the guard counter volume small compared to the total volume and consequently to make the guard counter ring as thin as possible. This creates problems in causing a high guard counter efficiency and in avoiding sparking of the guard counter.

We studied, therefore, the possibility of having the two volumes separated and in this paper we describe a Tritium low level proportional counter with this special feature. The separated counting volumes create special problems for the filling system but the advantages of doing this are many. In separating the two counters it becomes possible to make the guard counter much longer than the principal counter and conveniently thick, to obtain a high counting efficiency in the coincidence ring and to protect as much as possible the principal counter from low angle radiation coming towards the axis. The counting efficiency of the principal counter depends on the threshold energy for which the lower discriminator has been set up. To accept the lowest possible energy of the Tritium β particles a very small noise level of the electronic circuits is required. The

sensitive circuit has to be protected against parasiting electronic events.

In the following sections the counter, the filling system and the electronic equipment will be discussed in detail.

B. The Proportional Counter

1. Counter Design

The counter which will be described in this section consists mainly of two parts:

- a) The Inner or Principal Counter and
- b) a Counting Ring surrounding the inner counter which will be called Anti-coincidence counter or Reject counter.

Both counting systems are housed in a steel cylinder which itself is placed in a heavy shield of 25 cm iron, 5 cm lead and 2 cm mercury. The inner and the reject counter are separated by a thin Mylar foil which is coated on both sides with Al. To improve the counting efficiency the counter described here is different from other similar counting systems,³⁾ in that the thin foil between the two counters separates also the gas volumes. This has some consequences for the filling system and for the counter construction, as, on the one hand, the pressure in both the reject and inner counter has to be about the same, and, on the other hand, the two parts have to be gas tight.

The counter volume of the inner counter is about 600 cm^3 . This has a diameter of 50 mm and a length of 300 mm. The reject counter ring considerably overlaps the inner counter on both sides and has a total sensitive length of about 450 mm. Against other designs this geometry protects the inner counter much better against cosmic ray mesons coming from the side. The details can be seen from fig. 1. For calibration purposes a window is provided in the middle of the counter, which allows soft x-ray radiation to penetrate the inner counter. There are eight anode wires in the anti-coincidence counter ring which are connected in parallel. The distance between two wires is about the same as the thickness of the anti-coincidence counter. The gas tight seal between the two counters is achieved by squeezing the

Mylar foil between two cones and by additional glueing with Araldite.

The thickness of the separating foil must be chosen according to the following three conditions.

1. The foil should be as thin as possible to give the lowest background (see Appendix B and fig. 9) ⁵⁾.
2. On the other hand the foil must be thick enough to stop all β particles from the inner counter penetrating into the anti-coincidence counter and triggering it.
3. The foil should be able to stand a difference of pressure of some ten torr between the two counters.

For the Tritium measurements we used a foil of 3.4 mg/cm^2 . This thickness is sufficient for a low background a cuts off electrons below 34 KeV, so no "inner event" triggers the anticoincidence ring. Before putting the counter together the inner part was tested for vacuum tightness and for over-pressure resistance. It was found that the foil stands a 50 torr over-pressure.

All insulators are made of teflon, all metal parts of stainless steel. The anode wires are of 0.09 mm stainless steel in order to avoid too big end effects of the counter. Some field correction tubes ⁶⁾ have been applied on both sides of the principal counter.

2. The Counter Filling System

In fig. 2 the counter filling system is shown. It consists mainly of a filling line connecting both parts of the counter with a steel cylinder containing the counting gas (90% Argon, 10% Methane). By means of a Töpler pump the sample containing the Tritium can be pumped into the inner counter. Two CO_2 traps retain the mercury vapour. Between the two parts of the counter a safety valve ensures that the filling pressure in both parts does not differ more than about 10 - 20 torr.

The mercury valve (b in fig. 2) is used to fill the principal counter, and the tube plunging into the mercury fixes the difference of the pressure of the two counters. By this valve only a uni-directional connection exists between inner counter and gas container. Normally, we applied the following filling procedure:

After the counter was pumped to a vacuum of about 10^{-4} torr, counting gas is added in both parts up to about 300 torr. Then the Töpler pump is put into operation transferring the gas sample containing the Tritium into the inner counter. To adjust the filling pressure during this procedure, some counting gas is added from time to time into the reject counter. The Tritium sample is prepared in such a way that always 30 torr of hydrogen will be added in the inner counter. Finally for normal operations we have therefore 320 torr counting gas in the reject counter, 30 torr hydrogen plus Tritium mixture in the inner counter and 290 torr counting gas in the inner counter. The pumping and filling procedure needs about 3 to 4 hours. During the whole counting time the safety valve is connected with the counter. After a measurement the counting gas of the inner counter could be stored for further use, by extracting it from the counter using a second Töpler pump*.

The counting and filling system can also be connected directly with the Tritium extracting apparatus described elsewhere.⁷⁾ During more than one year's operation the filling system worked satisfactorily and the thin foil of the counter remained intact during the whole period.

* not shown in the figure

3. The Electronic Counting System

Both counters, the reject and the principal counter, were used in the proportional region. The output of both counters was connected directly to two pre-amplifiers fixed on the counter. After a linear amplification by a factor of 25 the signals were passed through a cable of 5 m length, and applied to two non-overloading linear amplifiers (Hamner type N301). The signal of the inner counter passed two discriminators which were adjusted in the following way:

The lower discriminator was set to a signal height corresponding to a particle of 2 KeV, the upper discriminator to about 18 KeV. Both discriminator outputs are gated separately by the anti-coincidence signals. In this way the "transmit" and "out" counting rates above the two discriminator levels could be recorded separately ("out" = gated signal). By subtracting the two "out" rates the inner counter count rate over the energy region determined by the two discriminators would be found.

All electronic parts were checked for spurious counts and for parasitic events from external influences. The whole system was found very stable and no signals other than real events were recorded. As indicated in the block diagram (fig. 3) an x-ray tube was installed underneath the counter (outside the iron shield) to produce characteristic fluorescent x-ray lines from different elements. These lines were used to set up the discriminators. This method of energy calibrations was found to be both quick and convenient as an x-ray spectrum could be taken in some few seconds using the Hutchison Skarott kicksorter. The line half-width for the 5.6 KeV iron x-rays was found to be 12%.

C. General Properties and Calibration of the Counter

The counter described in the foregoing section was studied extensively for its general properties such as plateau, amplification, linearity, background, efficiency and stability.

The dependence of the plateau on filling pressure and gas mixture was found to be sufficiently low near the values generally used for the measurements. The dependence of the count rate on the high tension applied to the centre wire was studied with the discriminator level and the electronic gain constant. An external source was used (Co^{60}) and filling pressures between 300 and 600 torr. The slope of the plateau was less than 2% per 100 V and the plateau length more than 300 V. The fig. 4 shows the plateau dependence on filling pressure. As standard values a total filling pressure of 320 torr was chosen and the working E.H.T. was about 1600 V.

As in normal measuring procedure hydrogen is added to the proportional counting gas, the counter properties were studied for different gas compositions. 90% Argon with 10% Methane was used as proportional counting gas. Hydrogen was added in different quantities. Good plateau characteristics could be obtained up to 300 torr hydrogen mixture. In most of our applications the hydrogen quantity is so small that the standard mixture of 30 torr was chosen in general.

Also, with a certain higher Tritium activity in the counter, the plateau was measured with Tritium as an internal source. Here, however, a certain discriminator level means that the spectrum of Tritium is cut off at a certain energy. By raising the tension the gas amplification increases and the Tritium counting rate accordingly, as the same discriminator level now corresponds with another cut-off point in the Tritium spectrum. From the known Tritium spectrum and the measured gas amplification (see below) the expected increase of the

Tritium counting rate could be compared with the measured values.

The gas amplification at different filling pressures and gas mixtures was measured in the following way:

The electronic gain of the system was measured with a precision pulse generator. Knowing the input capacity of the pre-amplifier the charge collected by the centre wire (anode) could be estimated from the pulse-height spectrum. The K x-ray energies of different elements in the region between 3 and 17 KeV were used as defined energies. Under our standard conditions (30 torr hydrogen, 290 torr Methane-Argon mixture) the gas amplification was estimated to be of the order of several 1000. Independent of this absolute determination of the gas amplification the relative increase of the pulse height at the pre-amplifier input was checked (see fig. 5 and 6) for different pressure and different anode voltages.

The overall linearity of the counter was checked in the following way:

As described in the section "counter design" the proportional counter has special windows where the K x-rays of different elements could be applied from the exterior. The K x-rays emitted from radio-isotopes as Fe^{55} , Zn^{65} , Se^{75} traverse the window and the counter foil (see fig. 2) and are absorbed mostly by the Argon in the centre counter. This absorption gives a fairly sharp line, (photo K x-ray line) and a second line of lower intensity, (2.8 KeV lower), the escape line of K x-rays from the Argon ionisation in the K level. In this way each isotope gives two lines and consequently two checking points for the linearity. Fig. 7 shows the position of the photo peaks on a multi-channel analyser for the K x-ray of the different isotopes.

A second check of the linearity has been made using the fluorescent characteristic x-ray emission. For this purpose foils of

and a thin wall (0.125 mm iron) was exposed to a collimated beam which was slid along the whole counter length (see fig. 9). The well-defined Tritium activity in such a counter with defined end efficiency was put into the proportional counter under our normal working conditions. The same was done with the gaseous source made of a sample of tritiated standard water, supplied by the Bureau of Standards in Washington. Within the limits of the errors of both calibrations the two standards gave the same overall efficiency for the proportional counter. The average value is 0.45.

As explained in the last section, the counter was tested for gas tightness, especially for leaking from the principal counter through the Mylar foil to the reject counter ring. For periods of the order of weeks no leaking was observed. (Less than 1% of the filling pressure.) With Tritium samples the leaking of this isotope was tested directly and no measurable effect could be found. For a long period special care had to be given to the maintaining of the overall gain. During the measurements at least once every day the position of a particular K x-ray line on a multi-channel analyser was taken. The overall gain changed by less than 2% in one day. We explain these changes by a slight change in the gas amplification, probably by establishing a final gas mixture. The rate of decrease was flattened out to zero in about four days. In the beginning of the measurements a slight adjustment for this gain variation was applied by adjusting the high voltage on the centre wire so that the test peak always appeared exactly in the same channel of the kicksorter. Under these conditions all the properties of the counter appeared to be very stable. During a continual testing period of more than one year no failure in the detector and in the transistorized circuit occurred.

D. Summary

From other similar types of counters the Tritium low level counter described in this paper differs mainly insofar as its inner volume is completely separated from the anti-coincidence counter ring by a thin foil. In this way it is possible to use a comparatively large guard counter, effectively shielding the principal counter against charged particles.

The result is a low background of 0.2 cpm which is unusual for a counter volume of 0.6 litres. The efficiency of the guard counting ring is shown by the high "transmit"/"out" ratio which is of the order of 300.

In the appendix B the formula of Oeschger and co-workers for the background dependence on the thickness of the separating foil and other counter parameters is given. Some of our background measurements confirm this and a factor for our low background is the thinness of the separating foil (see fig. 10).

Another advantage of the separation of the two counter parts is that no loss of the sample occurs because it is not circulated in the anti-coincidence counter.

The technical difficulties connected with the complete separation of the two counter parts have been described above. From practical working with the filling system it can be said, however, that operation and maintenance are relatively simple and need little time as long as the counter always remains connected with the filling system.

From the results of the counter tests and from the measurements of Tritium⁸⁾ produced as a spallation product by cosmic ray particles in meteorites, it can be seen that the described counter is a reliable instrument for counting small quantities of Tritium.

APPENDIX A :

If S and B are the counts from the sample and from the background respectively measured in the time intervals t_s and t_B , the counting rates will be $S^* = \frac{S}{t_s}$ and $B^* = \frac{B}{t_B}$. If the only factor for the variation of the counting rates of source and background is the hazard of disintegration, the Poisson statistical error (deviation) will be:

$$\delta_{S^* + B^*} = \pm \sqrt{\frac{S^* + B^*}{t_s}} \quad (1)$$

and $\delta_{B^*} = \pm \sqrt{\frac{B^*}{t_B}}$

For the net error δS^* we have:

$$\delta_{S^*}^2 = \delta_{S^* + B^*}^2 + \delta_{B^*}^2 \quad (2)$$

From (1) and (2)

$$\delta_{S^*} = \left[\frac{S^* + B^*}{t_s} + \frac{B^*}{t_B} \right]^{\frac{1}{2}} \quad (3)$$

and for the relative deviation

$$\sigma = \frac{\delta S^*}{S^*} = \frac{1}{S^*} \left[\frac{S^* + B^*}{t_s} + \frac{B^*}{t_B} \right]^{\frac{1}{2}} \sim \frac{\sqrt{B^*}}{S^*} \quad (4)$$

According to this formula to keep σ small, which means better results, a combination of B^* small and S^* large must be made. For the second condition, S^* large, because in most cases the radioactive sample is mixed with other inactive materials, a large detector must be used. But on the other hand a large detector means high background. So in the low level technique, firstly one must chose a reasonable counting volume, and secondly the background of this volume must be made as small as possible and the counting efficiency as high as possible.

A P P E N D I X B:

The total background N of a counter is given by the following formula:

$$N = C + S_p V \rho^1 p \left(1 - e^{-\left(\frac{\delta}{2} + \gamma d\right)} \right) + S_p \frac{0}{2\gamma} \left(1 - e^{-\gamma d} \right) \left(1 - e^{-(\delta_p + \gamma d)} \right) + S_{\gamma\epsilon} V \rho^1 p \left(1 - e^{-\left(\frac{\alpha}{2} p + \beta d\right)} \right) + S_{\gamma\epsilon} \frac{0}{2\beta} \left(1 - e^{-\beta d} \right) \left(1 - e^{-(\alpha_p + \gamma d)} \right)$$

- N : total background (cpm)
- C : constant (cpm)
- S_p : source strength of recoil protons (cpm/g)
- $S_{\gamma\epsilon}$: source strength of gamma induced electrons (cpm/g)
- V : Volume of counter (cm^3)
- ρ^1 : specific weight of gas per unit pressure (g/cm^3 , 10cmHg)
- p : gas pressure (10 cm Hg)
- d : thickness of wall (g/cm^2)
- α, δ : absorption coefficients (10 cm Hg^{-1})
- β, γ : absorption coefficients (cm^2/g)
- 0 : surface of counter.

$$\begin{aligned}\beta &= Z \left(\frac{\mu}{\rho} \right) \gamma, \epsilon & \alpha &= \frac{4V}{0} \rho^1 \left(\frac{\mu}{\rho} \right) \gamma, \epsilon \\ \gamma &= Z \left(\frac{\mu}{\rho} \right) p & \delta &= \frac{4V}{0} \rho^1 \left(\frac{\mu}{\rho} \right) p\end{aligned}$$

The total background as functions of wall thickness of the counter is shown in fig. 10.

Figure captions

1. Section of the low level proportional counter.
2. The filling system of the counter.
3. Block diagram of the electronic instrumentation.
4. Plateaux of the principle counter for different counting gas pressures.
5. The impulse height as function of H. Voltage and as parameter of the gas pressure.
6. The pressure - H. Voltage relation for the same pulse height. Counting gases 90% A + 10% CH₄ and 55% A + 45% CH₂CH₂
7. Linearity of the whole system. In the figure the position of the peak in the multi channel analyser is given as a function of the produced x-rays.
8. Dependence of counter efficiency on the hydrogen quantity.
9. Estimation of the effective length of the calibration counter.
10. Dependence of the background of a counter on the thickness of its wall.

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LOW LEVEL PROPORTIONAL COUNTER FOR TRITIUM

Fig. 1

COUNTER FILLING EQUIPMENT

Fig. 2

Block Diagram of Low Level Apparatus and X-ray Equipment

Plateaux of the Principle Counter

H. V.

Fig. 4

Fig. 5

Fig. 6

→ Energy of Electrons

Fig. 7

Fig. 9

Fig. 10