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AUSTRALIAN NUCLEAR SCIENCE AND TECHNOLOGY ORGANISATION

LUCAS HEIGHTS RESEARCH LABORATORIES

RADIOACTIVITY DETERMINATION BY COINCIDENCE COUNTING

PART 1 : SETTING UP THE EQUIPMENT AND THE STANDARDISATION OF COBALT-60

by

H.A. WYLLIE

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ABSTRACT

The 4 π β-γ coincidence counting equipment used in the Lucas Heights radioisotope standards laboratory for international comparisons is described in detail. A radioactivity standard is a method rather than a material object; as such it can be specified clearly only by discussing fully the setting-up and operation of one particular set of equipment. This manual is written for persons who are setting up such equipment for the first time.

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BETA PARTICLES; COINCIDENCE CIRCUITS; COINCIDENCE METHODS; COUNTING TECHNIQUES; COBALT 60; CALIBRATION STANDARDS; DEAD TIME; GAMMA DETECTION; GAMMA RADIATION; GAMMA SOURCES; PROPORTIONAL COUNTERS; RADIOACTIVITY; MANUALS;

EDITORIAL NOTE

The Australian Nuclear Science and Technology Organisation (ANSTO) replaced the Australian Atomic Energy Commission (AAEC) on 27 April 1987. Reports issued after April 1987 have the prefix ANSTO with no change of the symbol (E, M, S or C) or numbering sequence.

CONTENTS

1. INTRODUCTION

The coincidence counting equipment in the radioisotope standards laboratory at Lucas Heights has been used for many years for international comparisons of radioactive solutions. For these comparisons, sources are prepared from weighed aliquots of the solution provided, and measured simultaneously by a proportional gas counter and a sodium iodide detector. The equipment has been used recently in the experimental verification of a new formula used in making corrections for dead-time losses and accidental coincidences when calculating radioactivity from coincidence counting results [Wyllie 1987a, 1987b].

Bearing in mind that a radioactivity 'standard' is a method, rather than a material object stable over a long period, it is appropriate for it to be recorded in full detail. Further, it is advisable to illustrate the method by describing the equipment which is actually used. It is left to the reader to amend the method to suit alternative items of equipment. This report is intended to be of use to persons setting up 4 π β-γ counting equipment for the first time.

In describing the method, the standardisation of ^{60}Co has been used as an example. Additional procedures and corrections required for other radionuclides are discussed in a report of the US National Council on Radiation Protection and Measurements [NCRP 1985]. For readers unfamiliar with coincidence counting, a general introduction is provided by Tsoulfanidis [1983].

2. OUTLINE OF METHOD

If a radionuclide emits β particles and γ photons, the radioactivity (disintegrations per second) of a 4π source containing the nuclide can be determined by coincidence counting as follows. The source is placed in a gas-flow proportional counter in which the emission of β particles is measured. The emission of γ photons is measured simultaneously by a sodium iodide detector which is outside and above the gas counter. A disintegration may go undetected by either or both detectors. When, however, a disintegration is detected by both instruments, it is recorded by the coincidence mixer. The experimental results thus consist of the β count-rate, γ count-rate, and the coincidence rate.

The disintegration rate of the source is obtained in a calculation which corrects for background, dead-time in the counters, and accidental coincidences; other corrections may be necessary because of the nature of the decay scheme. An accidental coincidence is recorded by the mixer when the β particle and γ photon detected within the resolving time do not come from the same atom. The advantage of coincidence counting is that it eliminates the problem of no source or detector being 100 per cent efficient. The theory of the proportional counter and coincidence counting is discussed briefly in **appendix A** and in the NCRP report.

3. SUMMARY OF THE EQUIPMENT FOR COINCIDENCE COUNTING

A block diagram of the equipment used in the radioisotope standards laboratory is shown in **figure 1**.

3.1 Warning

Switches for the EHT and power to the AAEC* type 494 β amplifier must be OFF when the β pre-amplifier is disconnected from or connected to the gas counter.

3.2 The Beta Channel

Pulses generated in the β gas counter are amplified first by the β pre-amplifier then by the type 494β amplifier. The AAEC type 291 discriminator rejects less than one per cent of the pulses at the low end of the voltage range and produces fixed amplitude, tailed pulses. The Ortec 551 timing single channel analyser (SCA) produces square pulses of constant amplitude which are suitable for counting. The Sample Electronics (SE) discriminator (paralysis unit) allows for the setting of a fixed β dead-time.

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* Instruments so marked were designed and developed at the Lucas Heights Research Laboratories by staff of the AAEC (now the Australian Nuclear Science and Technology Organisation).

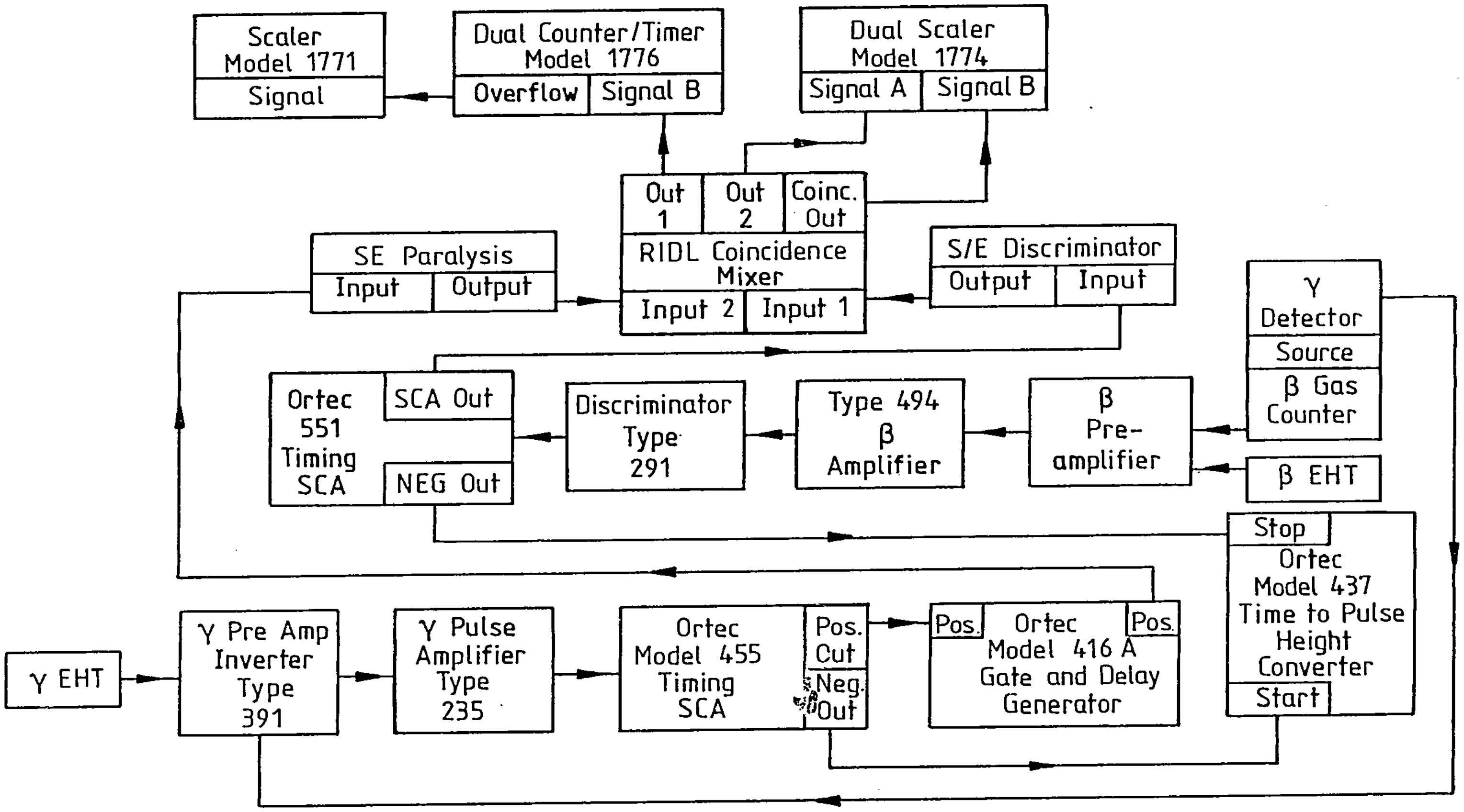

Figure 1 Block diagram of the coincidence counting equipment

3.3 The Gamma Channel

Pulses from the sodium iodide γ detector are amplified in the AAEC type 391 pre-amplifier inverter and the type 235 pulse amplifier. The Ortec 455 timing SCA produces square pulses of a fixed amplitude which are suitable for counting (see **figure 2**). The timing SCA also provides a γ gate, *i.e.*, it rejects incoming pulses below and above a range which admits the pulses from selected photopeaks of the γ spectrum of the radionuclide in the gas counter. The Ortec 416A gate and delay generator delays the passage of the pulses by a variable time interval, as described in **section 3.5**. The SE paralysis unit allows the γ dead-time to be set.

3.4 The Coincidence Mixer and Scalers

When a disintegration gives rise to both a β and a γ pulse, this 'coincidence' is detected in the coincidence mixer. WIDTH controls on the coincidence mixer set the coincidence resolving time. The scalers show the β, γ and coincidence counts.

3.5 Adjustment of the Delay of Gamma Pulses

A β pulse leaves the gas counter after the γ pulse from the same disintegration leaves the γ detector. This delay in the emergence of the β pulses varies over a range of time and is displayed as follows. The Ortec 437 time-to-pulse-height converter (TPHC) produces pulses which are proportional to the time intervals between the arrival at the TPHC of the γ and β pulses. A Canberra series 80 multichannel analyser (MCA) is connected to the TPHC, where it displays a curve showing the number of β pulses versus time interval between the γ and β pulses. The Ortec 416A gate and delay generator is then used to impose a delay on each γ pulse so that it arrives at the coincidence mixer at a time corresponding to the average time of arrival of a β pulse.

4. CHECKING THE EQUIPMENT

The normal β background is not more than two counts per second. A high β background ($\gg 2$ counts per second) may be due to moisture on the high voltage insulators at the rear of the gas counter. The background can be reduced by operating the room air conditioner on the cooling cycle to reduce the humidity of the atmosphere. It may help to dry the insulators with a hair dryer and the gas counter in an oven.

A ^{60}Co 4π counting source is placed in the gas counter, and a mixture of dry argon and 10 per cent methane is flowed through the counter and the bubbler. Insufficient flushing by the counting gas results in a drift in the counting rate until all remaining air has been removed. An initial rate of several bubbles per second is reduced to two per second after a few minutes. The flow of dry nitrogen through the pre-amplifier is checked. The β EHT (Ortec 456 high voltage power supply) is switched on, and the start button of the β scaler (Canberra dual counter/timer, model 1776) is pressed. The EHT is raised to plateau volts (the establishment of the plateau is described in **section 5.1**). The Tektronix type 317 cathode-ray oscilloscope (CRO) is switched on and connected in turn to the various outputs.

4.1 Gamma Pulse Amplifier, Type 235

CRO trigger selector : Auto (red knob)
INT + (black knob)
Amplifier : Set switch on NORM.

The required pulse shape is shown in **figure 2**. Shape can be modified by DIFF and INTEG. Amplitude of pulses is changed by

(a) triple-position (multiplier) switch (X1, X3 or X10);
(b) coarse gain; and
(c) fine gain.

4.2 Ortec 455 Timing Single Channel Analyser

Connect POS. OUT to CRO. Use a tee connector with a 100 ohm terminator on the CRO input. **Figure 2** illustrates the pulses from the γ amplifier and the timing SCA. The amplitude of the timing SCA pulses is independent of the amplitude of the pulses from the amplifier.

The back part of the top switch is set to WIN. 100 per cent. The window is set by adjusting the upper and lower level discriminators. With the top switch set to WIN. 100 per cent, the upper level is actually LOWER LEVEL setting plus UPPER LEVEL setting. The timing SCA eliminates 'walk'. **Figure 2** shows how a weak output pulse from the amplifier enters the window later than a strong pulse. 'Walk' is the time difference.

The function of the delay control can be observed as follows. The CRO is externally triggered by connecting the output of the γ amplifier to TRIGGER INPUT on the CRO. The trigger selector knob (black) is moved from INT + to EXT +. The delay potentiometer of the timing SCA can then move the square pulse along the time scale [Leave on 1.0].

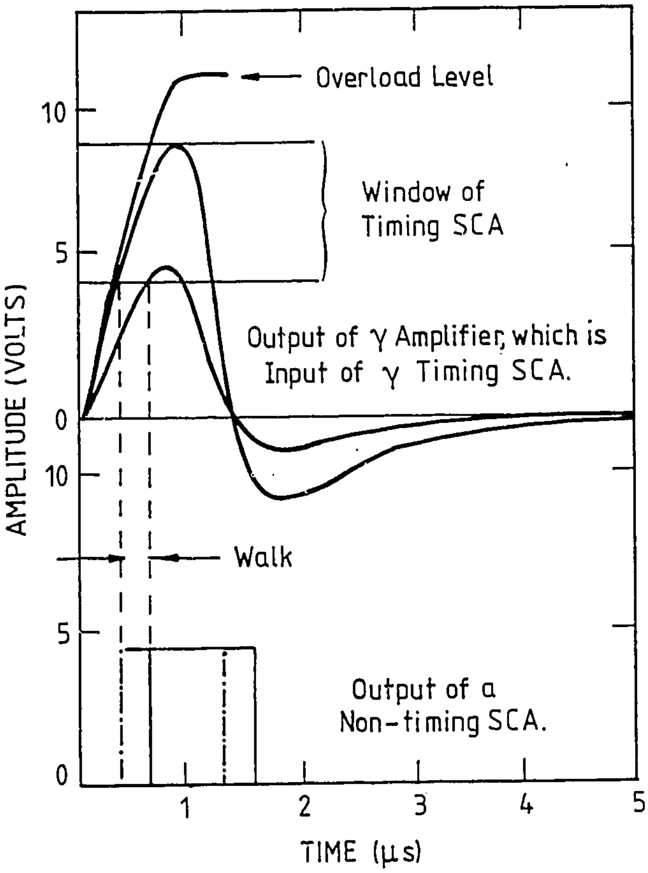

Figure 2 Output of γ amplifier and γ SCA

4.3 Ortec Type 416A Gate and Delay Generator

Set switch to 1.1 μs. The DELAY knob is used to delay the pulse as follows. Externally trigger the CRO by connecting the output of the γ amplifier to TRIGGER INPUT on CRO; move the CRO trigger selector knob (black) to EXT +. Connect CRO INPUT to DELAYED OUT (POS or NEG). Turning the DELAY knob moves the square wave along the time scale.

4.4 AAEC Type 494 Beta Amplifier

The output of the β amplifier is illustrated in **figure 3**. This amplifier is designed for low-noise, overloaded operation with fast recovery from overload.

4.5 AAEC Type 291 Beta Discriminator

The output of the β discriminator is illustrated in **figure 4**. Set switch to 500 ns O'P WIDTH. Set LEVEL dial to 3.50 V. Because the amplifier is run in overload mode, setting at this level causes the rejection of noise and betas of very low energy, as is seen from the low background count-rate.

4.6 Ortec Type 551 Timing SCA

Set the following:

(i) UPPER LEVEL to 10.

(ii) LOWER LEVEL to zero.

(iii) Left-hand switch to '1-11 μs'.

(iv) Right-hand switch to 'NOR'.

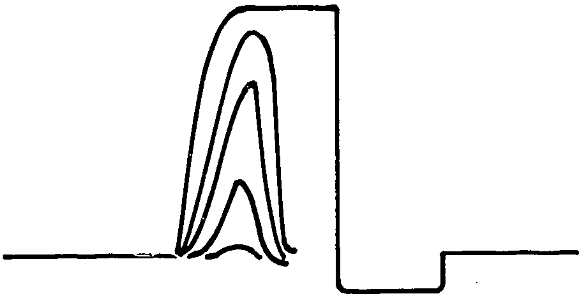

Figure 3 Output from β amplifier

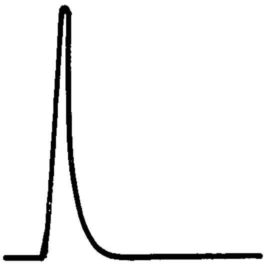

Figure 4 Output from β discriminator

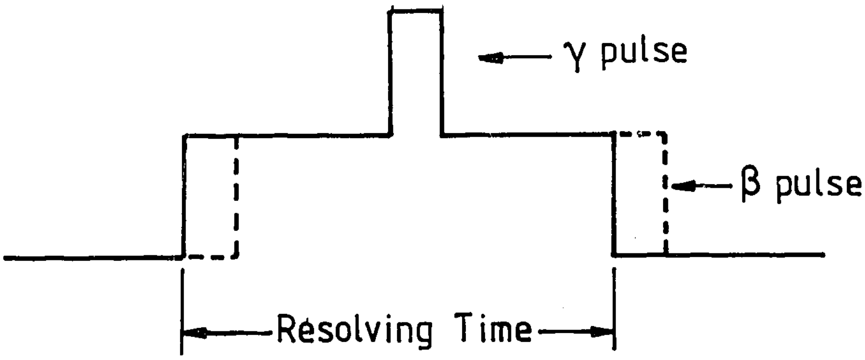

Figure 5 Production of a coincidence pulse by the summing of a β pulse and a γ pulse

4.7 Gamma Paralysis Unit

Set BIAS to 1.50 V.

4.8 Beta Paralysis Unit

Set DISCRIMINATOR to 1.00 V.

4.9 Coincidence Mixer

Set 'WIDTH' switches to X 10. Connect MONITOR to CRO INPUT. Connect OUT of the type 235 pulse amplifier to CRO TRIGGER INPUT. Turn CRO TRIGGER SELECTOR to EXT +. A coincidence pulse is produced by summing a β pulse and a γ pulse (see **figure 5**). The knob marked WIDTH, MIN MAX allows the resolving time to be adjusted. Normally it is set on the central reading. The resolving time (about 1.2 µs) can be estimated approximately on the CRO by observing the width of the pulse.

5. SETTING UP THE EQUIPMENT

The procedures described in this section are carried out before a radionuclide standardisation is commenced. In addition, the β plateau (**section 5.1**) is checked at the start of each day.

5.1 Establishing the Plateau for Beta Count-rate *v.* EHT

(a) A β count-rate versus EHT curve is obtained for each radionuclide. This is normally done with the potenticmeter of the beta discriminator set at 3.5.

(b) It is advisable to store the source overnight in a dry atmosphere before counting.

(c) Flush the counter with counting gas long enough to achieve stable counting conditions.

(d) With the argon-methane mixture, the EHT plateau lies between 1800 and 2500 V; with methane it lies between 1800 and 3800 V.

(e) Select on the Ortec 456 high voltage power supply an EHT value 100 V below the expected plateau region and count long enough to accumulate more than 100 000 counts. Record the counts.

(f) Increase the EHT in 50 V steps, recording the counts at each voltage. Stop when the count-rate rises steeply.

(g) Record the voltage at which the change in count-rate with respect to EHT is at a minimum. Count the source at this voltage.

(h) If the EHT is changed by REDUCING the setting, wait a few minutes for it to stabilise then recommence counting.

5.2 Establishing the Plateau for Gamma Count-rate *v.* EHT

(a) Set the window of the timing SCA wide open by turning the lower level potentiometer to its minimum setting and the upper level potentiometer to its maximum setting.

(b) The usual setting of the gamma EHT is 1200 V. The gain of the gamma amplifier has been set so that at 1200 volts EHT, the output pulses do not exceed 8 volts on the CRO.

(c) Reduce the EHT to 800 V. It will be noticed that the pulse height has dropped. Measure the γ count-rate.

(d) Increase the EHT in 50 V steps, measuring the γ count-rate at each setting. The count-rate rises at first, levelling off at about 950 V. Beyond 1200 V, it is necessary to reduce the γ amplifier gain to keep the output pulses less than 8 V.

(e) At 1400 volts, increase the EHT in 20 V steps. It will be noticed that the count-rate increases slightly, but that the pulse height no longer rises with increasing EHT. At 1520 V, the count-rate rises sharply. The EHT should not be increased beyond this point.

(f) Reduce the EHT to 1200 V, which is the mid-point of the plateau, and adjust the gain of the γ amplifier so that the maximum pulse height is 8 V.

5.3 Setting the Gamma Gate

The procedure described in this section is for cobalt-60:

(a) Bias on photomultiplier: the EHT is set at 1200 V.

(b) Connect the type 235 γ pulse amplifier OUT to the analogue- to-digital converter (ADC) of the Canberra series 80 MCA, and to the Tektronix CRO CH1 or X, using a tee-connector.

(c) Put the cobalt-60 source in the gas counter. Adjust amplifier gain with the CRO so that the amplitude of the pulses is below the overload level of the amplifier (see **figure 2**).

(d) Obtain a spectrum of counts *v.* γ-photon energy on the Canberra MCA:

   (i) Function– Status, select. Type– 1–2. Press– Store, Yes.

   (ii) Function– Preset, select. Press– No. Type– 10000. Press– Store, Yes, Yes. (This sets the counting time in the MCA.)

   (iii) Function– ADC, select. Press– No, Yes, Yes. Type– 1–2. Press– Store, Yes, Yes.

   (iv) Press– Collect (this starts the counting).

   (v) To stop counting, again press– Collect. To remove spectrum, press– Clear, Data (simultaneously), Yes.

   (vi) Move cursors onto the two photopeaks (after collecting sufficient counts for well-defined peaks). Stop counting. Remove spectrum.

(e) Disconnect the γ pre-amplifier from the γ main amplifier and connect the mercury relay pulser (MRP) of a Franklin Electronics 370 precision pulse generator and a 100 ohm terminator to the input of the γ main amplifier.

(f) Set the pulse height dial on the MRP to a multiple of the energy of one of the photopeaks, *e.g.* set 586.6 on the dial, corresponding to 1173.2 keV. Set the potentiometer labelled NORMALISE at its maximum reading.

(g) Turn on MRP switch labelled RELAY. Look at the pulse height on the CRO. Adjust MRP attenuators and potentiometer labelled NORMALISE to get an ~ 7 V pulse on the CRO. Press the MCA switch labelled COLLECT. Counts will accumulate on the MCA in a channel near the left-hand cursor. Adjust NORMALISE so that left-hand cursor rises. Pulser volts then equal photopeak volts. Stop counting and remove the spectrum.

(h) Check the NORMALISE adjustment as follows. Set MRP pulse height dial to 666.3, corresponding to the second photopeak energy of 1332.5 keV. Press the COLLECT switch. The second cursor should start to rise. Stop counting, and remove the spectrum.

(i) Disconnect the MRP from the γ amplifier and connect the γ pre-amplifier to the γ amplifier. Press the COLLECT switch. Move the MCA cursors to the required energy levels of the γ gate, *i.e.*, to the left of the left-hand photopeak, and to the right of the right-hand photopeak. Stop counting and remove the spectrum. Reconnect the MRP to the γ amplifier. Press the COLLECT switch. Adjust the MRP pulse height dial so that the MCA left-hand cursor rises. Record the energy (keV). Stop counting and remove the spectrum.

(j) Start the β and γ scalers. The γ scaler shows the MRP pulses being counted. Adjust the lower level discriminator of the timing SCA until the γ scaler flicks from 'counting' to 'not counting'.

(k) Press the COLLECT switch . Adjust the MRP pulse height dial so that the MCA right-hand cursor rises. Record the energy (keV).

(l) Adjust the upper level discriminator of the timing SCA until the γ scaler flicks from 'counting' to 'not counting'.

(m) Remove the MRP and reconnect the pre-amplifier.

5.4 Beta Dead Time

(a) On the β paralysis unit, turn the knob marked 'PARALYSIS µs' to the required dead-time, *e.g.* 5 or 10 µs. This value is nominal only. The actual dead-time is measured as follows.

(b) Connect OUTPUT of the AAEC type 144 double pulse generator (DPG) to INPUT of CRO. On the DPG, set SHAPE to 'Rectangular', and POLARITY to +. On the DPG, set Frequency dial to a value between 4 and 5 kHz. On the DPG, set left-hand knobs of attenuators A and B to 100 mV (avoiding the vacant settings between the figures on the dial), and set the right-hand knobs to about '2 o'clock' to produce pulse heights of 15 mV, as shown on the vertical scale of the CRO.

(c) Connect OUTPUT of the DPG to the INPUT of the type 494 β amplifier. To check that the DPG pulses will not overload the β amplifier, connect the β amplifier OUTPUT to CRO INPUT. Spread out the wave form by setting the CRO TIME/DIV. at 2 µs. Check that the amplifier is not overloaded, *i.e.*, that the output wave is not flattened. Reconnect the β amplifier OUTPUT to the AAEC type 291 discriminator.

(d) On the DPG, delay time (µs) = {(Reading on DELAY dial) x 2.5} + 2.7. Set the DELAY potentiometer to give a delay time greater than the dead-time which has been set on the β paralysis unit. (Note: the window on the potentiometer shows units, and the dial shows tenths and hundredths.)

(e) Connect the CRO INPUT to the coincidence mixer SINGLES OUT 1 (β output). Set CRO TIME/DIV. to 2 µs and observe the double pulses.

(f) Reduce the DPG DELAY until the second pulse is extinguished.

(g) The β dead-time is obtained by one of the following methods:

  (i) The time between pulses is measured on the horizontal (time) scale of the CRO.

  (ii) The dead-time equals the delay time which is calculated by the equation in paragraph (d).

  (iii) Method (i) using the more advanced Tektronix digital storage CRO (if available) gives the most accurate result.

5.5 γ Dead-time

(a) The γ dead-time is set to equal the β dead-time. On the γ paralysis unit, turn the knob marked 'PARALYSIS µs' to the required dead-time, *e.g.* 5 or 10 µs. This value is nominal only; the actual dead-time is measured as follows.

(b) Connect the OUTPUT of the DPG to IN of the type 235 γ pulse amplifier. Connect the CRO INPUT to the coincidence mixer SINGLES OUT 2 (γ output).

(c) Set CRO TIME/DIV. to 2 µs. On the DPG, set the FREQUENCY dial to a value between 4 and 5 kHz.

(d) On the DPG, delay time (µs) = [(reading on DELAY dial) x 2.5] + 2.7. Set DELAY dial to give a delay time greater than the dead-time which has been set on the γ paralysis unit.

(e) On the DPG, the voltage of the double pulses has to be set so that it is between the upper and lower levels of the γ gate. Set the CRO VOLTS/DIV. to 1. On the DPG, adjust attenuators A and B as follows (this is the procedure for cobalt-60). Turn the two COARSE (left-hand) knobs to '1 volt'. Then turn the FINE (right-hand) knobs so that both pulses appear on the CRO. The MCA can also be used.

(f) On the DPG, reduce DELAY until the second pulse is just extinguished. To obtain the γ dead-time, see **section 5.4, paragraph (g)**.

(g) The dead-time can be set exactly to a required value by adjusting the appropriate screw on the paralysis unit.

5.6 Resolving Time

(a) Convert the DPG to a single pulse generator by turning the COARSE knob of one of the attenuators to one of the vacant positions between the dial voltage figures.

(b) The coincidence resolving time can be set to a maximum value of about 2.3 µs. Set a nominal value of 1.1 µs on the coincidence mixer by turning the left-hand and central WIDTH knobs to their central positions.

(c) The actual value is measured as follows. Place a cobalt-60 source in the proportional counter. On the DPG, turn the FREQUENCY knob to 5 kHz. Connect the DPG OUTPUT to IN of the type 235 γ pulse amplifier (use a terminator). Set the DPG Shape to Rectangular.

(d) On the DPG, turn the Fine knob of the other attenuator to get counts accumulating on the γ scaler (Canberra 1774 dual scaler, channel A; channel B shows the coincidence count). The voltage of the pulses will then allow the pulses to pass through the γ gate.

(e) Record β, 'γ' and coincidence counts after sufficient counts have been accumulated to give good accuracy.

$$\text{Resolving time } (\mu\text{s}) = \frac{\text{Coincidence count rate} \times 10^6}{2 \times \beta \text{ count rate} \times \text{'}\gamma\text{' count rate}} .$$

This equation is discussed by Baerg [1965].

5.7 Delay Time

(a) On the Ortec TPHC, connect

   (i) START to NEG. OUT of the Ortec 455 timing SCA;

   (ii) STOP to NEG. OUT of the Ortec 551 timing SCA;

   (iii) OUTPUT, $Z_0 = 1\ \Omega$ to ADC IN of the Canberra series 80 MCA.

   The time intervals between START and STOP give rise to pulses whose heights are indicated by the horizontal coordinates of the points on the MCA screen. The vertical coordinates measure the number of disintegrations corresponding to each time interval.

   On the Ortec TPHC, set

      (i) AMPLITUDE to 10 V,

      (ii) GATING MODE to 'Anti-Coinc.'

(b) On the Canberra MCA, start counting:

   (i) Function– Pre–set, select. Press– No. Type counting time in seconds, *e.g.* 10 000. Press– Store, Yes, Yes, Collect.

   (ii) Vertical range: AUTO.

(c) On the Ortec TPHC, set RANGE to obtain a satisfactory time delay spectrum on the MCA screen:

e.g. RANGE: 0.1 µs.
MULTIPLIER: X 10.
Stop counting. Remove spectrum.

(d) Press COLLECT. On the Ortec 551 timing SCA, adjust the β DELAY so that the MCA spectrum is well clear of the left-hand end of the base line; *e.g.* DELAY = 1.30. Stop counting. Remove spectrum.

(e) COLLECT spectrum on the MCA. Press COLLECT to stop counting when sufficient counts have accumulated.

(f) Set cursors either side of peak. INT indicates total counts between cursors, *i.e.* peak counts. Divide INT by 2. Move one cursor until INT indicates half the original value. Remove spectrum.

(g) On the Ortec 416A gate and delay generator, connect DLYD MARKER to STOP on the Ortec TPHC. Press COLLECT. Adjust the DELAY on the Ortec 416A until the cursor at the mid-point of the peak starts to rise. This indicates that the γ pulses to the coincidence mixer will be delayed so that half the β pulses reach the mixer ahead of the corresponding γ pulses, and half after.

(h) Reconnect NEG OUT of the Ortec 551 timing SCA to STOP on the Ortec TPHC. The system now operates with optimum delay matching.

6. COUNTING A 4π SOURCE

6.1 Choice of Source Strength

Uncertainty in the correction for dead-time and accidental coincidences can be kept low by using a 4π source of not more than 7 kBq. However, counting accuracy is reduced as the chosen radioactivity decreases.

6.2 Flushing the Gas Counter

The counter should be flushed with counting gas for 20 minutes before counting is commenced.

6.3 Setting the Beta EHT

The β EHT is set on the Ortec 456 high voltage power supply at a value which allows a minimum change in count-rate with respect to EHT. This reduces to a minimum any change in count-rate during counting due to drift in EHT.

NOTE: In coincidence counting, any value of the EHT in the plateau region could be chosen if there were no drift in the EHT. In that case, the selection of a new EHT between two separate counting periods would lead to a different calculated efficiency of the β counter for the second period, but there would be no change in the calculated radioactivity.

6.4 Counting Periods

For accurate results, before counting, the source should be left in the counter overnight with the counting gas flowing. The source is counted the following day. The number of counting periods, and their duration, depend on the accuracy required [NCRP 1985]. The date and the time at the start of each counting period is noted. The source is removed and the background is counted. Lack of precision in the beta count-rate may be due to dirty filaments in the gas counter. The installation of new filaments is described in **appendix B**.

6.5 Calculations

The calculation of the disintegration rate of cobalt-60 is given below. Additional corrections are required for nuclides with more complex decay schemes.

After each counting period, the count-rates (counts per second) are calculated from the β, γ and coincidence scaler readings. The corresponding background count-rates are then subtracted:

let A' = observed count-rate in the β counter with the background rate subtracted;

B' = observed count-rate in the γ counter with the background rate subtracted;

C' = observed count-rate in the coincidence counter with the background rate subtracted;

T = dead-time in seconds;

T_R = resolving time in seconds; and

N = disintegration rate in becquerel.

If the disintegration rate of the source is less than 10 kBq, it can be calculated by an equation derived by Bryant [1963]:

$$N = \frac{A'B'}{C'-2A'B'T_R}\left[1 + \frac{2C'T-2(A'+B')T_R}{2-(A'+B')T}\right] .$$

For higher disintegration rates, use is made of a set of equations derived by the author [Wyllie 1987a, 1987b]. The computer program calculates N when the following data are entered:

(a) the dead-time in microseconds,

(b) the resolving time in microseconds,

(c) the half-life of the source in days,

(d) the reference time, in days and fraction of a day, measured from the beginning of the year,

(e) the counting period of the source in seconds,

(f) the time at the start of the counting of the source as shown on the digital clock, in days and fraction of a day,

(g) the source counts from the β, γ and coincidence scalers,

(h) the background counting period in seconds,

(i) the background counts from the β, γ and coincidence scalers,

(j) the mass of solution deposited on the source mount, and

(k) the dilution factor of the solution used for preparing the source.

The computer prints out the following information:

(a) The radioactivity of the source at the reference time. For this calculation, the time half way through the counting period is taken as the time of measurement.

(b) The radioactivity concentration of the original solution (before dilution for source preparation) at the reference time.

(c) The β and γ counting efficiencies.

(d) The values for (a) and (b) calculated using Bryant's formula.

The counting data from ten runs can be entered, in which case the **average** radioactivity concentration of the original solution at the reference time is printed. As indicated above, the program calculates the radioactivity of the source by using Bryant's formula. This latter value of the radioactivity is the starting value for an iteration which uses the author's formula.

7. ACKNOWLEDGEMENTS

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APPENDIX A

COUNTING THEORY

A1. GAS-IONISATION DETECTORS

Consider a volume of gas within an enclosure containing an insulated anode and cathode by means of which an electric field can be applied across the gas. When charged particles and electromagnetic radiation emitted by radionuclides traverse the gas, they undergo inelastic collisions with the gas atoms or molecules, which produce positive ions (atomic or molecular) and electrons (in an electropositive gas) or negative ions (in an electronegative gas). An alpha particle with energy 5 MeV will produce about 150 000 ion pairs while expending all its energy in a gas. The positive ions and electrons (or negative ions) will move towards the cathode and anode, respectively. The resultant ionisation current is utilised in two types of radiation detector — ionisation chambers and proportional counters.

If the voltage across the electrodes is increased from zero, the ion current increases and then levels off. In the voltage range where the current is increasing, some recombination of the ions takes place; in the range where the current remains constant, there is no recombination and the ions are all collected. This current is called the saturation current. The probability of recombination is lower in a gas in which only free electrons are formed (such as argon) rather than negative ions (as in oxygen); saturation in the former type of gas can be reached with a lower field intensity.

A2. GAS MULTIPLICATION

When the voltage applied across an electropositive gas is increased beyond the region of the saturation current, the electrons formed by the primary ionising event acquire sufficient energy to cause secondary ionisation. The ionisation current will start to rise again; this phenomenon is known as gas multiplication or gas amplification. If a fine wire is used as the anode, as in the proportional counter, the electrons pass through an intense electric field before collection on the anode. The secondary ionisation occurs in a very small volume of gas close to the anode wire. The gas multiplication is in the form of an avalanche which, in a gas ionisation detector with a wire anode of 0.02 mm diameter, occurs within 0.05 mm of the wire surface.

Ionisation detectors which employ gas multiplication are operated in the pulse mode to count individual disintegrations. In the lower part of the gas amplification range of voltage, the charge collected on the anode is proportional to the energy deposited by the incident radiation, *i.e.* to the number of ion pairs formed in the primary ionising event. This voltage range is known as the proportional region.

A proportional counter usually consists of two detectors, each subtending a solid angle of 2π to a source mounted on a thin plastic support; it is known as a 4π counter.

A3. THE RANDOM NATURE OF RADIOACTIVITY

Radioactivity is a random process; the time of decay of an individual atom cannot be predicted. The equation $N_t = N_o e^{-\lambda t}$ (where N_o is the initial number of atoms and N_t is the number after time t) describes the behaviour of an infinitely large number of atoms. The number of atoms which will decay in time t is $N_o(1 - e^{-\lambda t})$. This expression is called the expectation value. If a series of radioactivity measurements is carried out, as the number of measurements increases their mean value approaches the expectation value. The values found in the individual measurements are scattered about the mean value owing to the random nature of radioactive decay.

An expression which gives a measure of the random scatter of results in an experiment is the standard deviation (σ) which is defined by the equation

$$\sigma^2 = \left\{ \sum_{i=1}^{k} (n_i - \mu)^2 \right\} / k \quad ,$$

where n_i is the result of the i^{th} measurement of a very large number (k) of measurements and μ is the mean of the results. For radioactive decay, μ is equal to the expectation value, *i.e.* $N_o(1\text{-}e^{-\lambda t})$.

If a very large number of radioactivity measurements is made, the fraction of the measurements in which n atoms are found to decay in time t is called the probability (P_n) of the result being n. Let $\mu - n = x$. Now, if μ is much less than N_0 (*i.e.* t is small compared to the half-life) and $|x|$ is much less than μ (*i.e.* only values of n close to the expectation value are to be considered), then it can be shown from probability theory that the probability (P_x) of obtaining the difference x in any particular measurement is given by

$$P_x = \frac{1}{\sqrt{2\pi\mu}} e^{-x^2/2\mu} \quad .$$

This means that if a very large number of measurements is carried out, P_x is the fraction of measurements which gives a result which differs from the mean value (μ) by a particular value of x. It can also be shown that

$$\sigma^2 = \mu \quad .$$

The above expression for P_x is known as the Normal or Gaussian distribution, and from it the scatter of results about the mean value (*i.e.* the expectation value) can be calculated. It can be shown that if a very large number of measurements were to be carried out, 68.3 per cent would give values of n in the range $\mu \pm \sqrt{\mu}$, 95.4 per cent would lie within $\mu \pm 2\sqrt{\mu}$, and 99.7 per cent would lie within $\mu \pm 3\sqrt{\mu}$.

Thus, if a radioactive source is counted many times and the mean value is 10 000 disintegrations, 99.7 per cent of the results will lie within 3.0 per cent of the mean value. If the counting period is long enough to obtain a mean value of 1 000 000, 99.7 per cent of the results will lie within 0.3 per cent of the mean value. Therefore, from considerations of the random nature of radioactive decay, the scatter of the results is reduced by increasing the time of counting.

A4. COINCIDENCE COUNTING

Some of the particles and photons emitted by atoms in a radioactive source are absorbed within the source. Others are not seen by the detectors. The count rate is thus always less than the disintegration rate. However, coincidence counting can be used to obtain the disintegration rate for those nuclides which emit both a particle and a gamma photon. In the case of cobalt-60, if A′ is the count-rate in a 4πβ counter, and N is the disintegration rate of the source, the efficiency of the counter (ε_β) is given by the equation

$$\varepsilon_\beta = A' \div N \quad .$$

Similarly, for the gamma counter, which is mounted above the 4πβ counter,

$$\varepsilon_\gamma = B' \div N \quad .$$

The equipment also includes a coincidence counter which receives pulses from the β and γ counters. These pulses do not arrive at the coincidence counter simultaneously. When the γ pulse from a disintegration arrives, the coincidence counter waits for about 1 µs (this is known as the resolving time) and, if a β pulse arrives during this interval, a coincidence is counted. The count-rate in the coincidence counter (C′) is given by

$$C' = N\varepsilon_\beta\varepsilon_\gamma \quad .$$

Combining these last three equations, we get the disintegration rate in the source:

$$N = \frac{A'B'}{C'} \quad .$$

In practice, various corrections have to be made. For example, during the resolving time, the coincidence counter may receive a pulse arising from a β particle emitted by an atom other than that which gave rise to the γ pulse; this is known as an accidental coincidence.

For every disintegration detected, a dead-time is imposed in the β or γ detector; this is the minimum interval of time (*e.g.* 5 µs) which the system requires to process a detected event. Events occurring during this time cannot be processed, and are lost.

Equations have been developed which give corrections for both accidental coincidences and dead-time losses. The two equations which are used in this laboratory are given in **section 6.5**.

APPENDIX B

INSTALLATION OF FILAMENTS IN GAS COUNTER

About 300 mm of tungsten wire of diameter 0.025 mm is washed as follows. A detergent solution is prepared in a beaker by mixing one volume of RBS 25 Concentrate (Chemical Products, R. Borghgraef, Belgium) with three volumes of distilled water. The wire is dropped into the solution and left for 20 minutes. The solution is poured off, and the wire is washed ten times with distilled water by decantation. It is easy to pour out nearly all of the water after each washing; the tangled wire remains at the bottom of the beaker with a small amount of water trapped by surface tension. After the last washing, the beaker is placed in an oven to evaporate the remaining water.

The filament posts in the counter are prepared by removing old solder by means of a soldering iron and copper wick. One end of the wire is laid within the groove in a post and solder is applied. The wire is stretched through the groove of the other post and solder is applied. The wire must be kept taut while producing this second 'solder swage'. The ends of the tungsten wire are removed by shaving off with a knife a layer of solder from which the wire protrudes.