AECL-7253

ATOMIC ENERGY OF CANADA LIMITED

L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

STATUS OF THE COINCIDENCE-COUNTING SYSTEM USED FOR THE METROLOGY OF RADIONUCLIDES, AND THE STANDARDIZATION OF ^{60}Co AND ^{57}Co

L'état du système métrologique des coïncidences et l'étalonnage du ^{60}Co et ^{57}Co

L.V. SMITH, A.R. RUTLEDGE and J.S. MERRITT

Chalk River Nuclear Laboratories

Laboratoires nucléaires de Chalk River

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L'ÉTAT DU SYSTÈME MÉTROLOGIQUE DES COÏNCIDENCES ET L'ÉTALONNAGE DU ^{60}Co ET ^{57}Co

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RÉSUMÉ

On a employé une méthode des coïncidences pour la métrologie et l'étalonnage des solutions de ^{60}Co et de ^{57}Co à l'aide d'un compteur proportionnel de géométrie (4π(CP)) et des détecteurs NaI(Tℓ) pour la voie γ. On décrit l'équipement électronique. Les procédés sont présentés pour l'ajustement et pour la mesure des temps morts, des temps de résolution, et du délai entre les deux voies, 4π(CP) et γ. Des échantillons des étalons résultants sont soumis au Bureau International des Poids et Mesures (BIPM) et à l'Agence Internationale de l'Energie Atomique (AIEA) pour les comparer avec les étalons internationaux. Finalement on a distribué les échantillons de ^{60}Co et de ^{57}Co aux laboratoires locaux.

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ABSTRACT

A coincidence method, employing a 4π proportional counter (4π(PC)) and NaI(Tℓ) crystals as detectors, was used for the accurate standardization of stocks of ^{60}Co and ^{57}Co. The experimental arrangement is described in detail. Procedures are given for the adjustment and determination of dead times, resolving times, and time delay between channels. Samples of the standardized solutions were submitted to the Bureau International des Poids et Mesures (BIPM) and the International Atomic Energy Agency (IAEA) for comparison versus measurements from other countries. Other samples were issued for local use.

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1. INTRODUCTION

For approximately thirty years the radioisotope standardization group at the Chalk River Nuclear Laboratories (CRNL) has chosen a coincidence method for its most accurate measurements of activities of radionuclides. Hawkings and Mann (1) were the first from this laboratory to use the method; they standardized ^{131}I and other β-γ emitters. The detectors for both the beta and gamma channels were Geiger counters and although these detectors and their electronic instrumentation bear little resemblance to those used today, the basic principles were the same. A few years later Campion (2) employed high efficiency detectors and studied the method in detail. The massive lead shielding installed by Campion (2) is still in its original location, but surrounds a different 4π proportional counter (4π(PC)) and other NaI(Tℓ) γ-ray detectors. The works of Campion (2) and others (3,4,5) were effective in establishing the 4π(PC)-γ coincidence method as the primary tool for the metrology of radionuclides in most international laboratories.

A major improvement was introduced at CRNL in 1968. This was an automatic sample changer designed by Taylor and Love (6), which required a specially designed 4π(PC) to accommodate the sample holders employed. A computer-based control system, for operation of the sample changer and for data accumulation, was designed, built, installed and is maintained by members of the Electronic Systems Branch of CRNL (7,8,9). Together, the sample changer and its control system have provided the means of accumulating much more data than previously was possible. This has improved the precision and enabled workers to search for sources of small systematic errors, which, in turn, has led to improvements to the system and more accurate measurements.

Throughout the history of 4π(PC)-γ coincidence counting in this laboratory, the associated electronic instrumentation has undergone almost continual change. From time to time the coincidence equations, miscellaneous test routines, and procedures have also been modified. Some of these developments have been documented, but others have come, and sometimes gone, with little or no notice. The purpose of this report is two-fold: a) to describe the system that was used for two major standardizations, ^{60}Co and ^{57}Co, in sufficient detail so that it can serve as a working document to which future changes and measurements may be referenced, and b) to report the results of these standardizations.

2. COMPONENTS AND LAYOUT

The block diagram of Fig. 1 shows that the computer-control system (7,8,9) now serves three experimental arrangements: 1) a γ-ray spectrometer, 2) an ionization-chamber system, and 3) a 4π(PC)-γ coincidence system. Separate sample changers serve each of the latter two systems. Another small computer (Fig. 2) can be used for γ-ray spectrometry(10), but not for the control of the sample changers.

The capabilities and general layout of the computer-control system (Fig. 1) have been described by various members of the Electronic Systems Branch (7,8,9). The details are given in internal CRNL reports or drawings and Table I gives a list of this documentation. Figure 3 shows the data acquisition system including the peripheral equipment.

Figure 4 shows the arrangement of detectors and electronic instrumentation used for 4π(PC)-γ coincidence counting.

The 4π(PC) is made of stainless steel. The interior has a pill box shape with a 3.8-cm diameter and a 1.8-cm height for each of its 2π sections. The wires are 0.01 mm in diameter.* The counter walls are thick and specially shaped to accommodate the sample holders of the changer and to serve as mounts for the vertical location of two 7.6 cm x 7.6 cm NaI(Tℓ) detectors, above and below the 4π(PC). The ends of the 4π(PC) are of much thinner steel (0.86 mm) to reduce γ-ray attenuation. When required, additional absorbers can be inserted between the 4π(PC) and the NaI(Tℓ) crystals to prevent higher energy β particles or X rays from reaching the γ-ray detectors.

* See CRNL drawing E-4434-3.

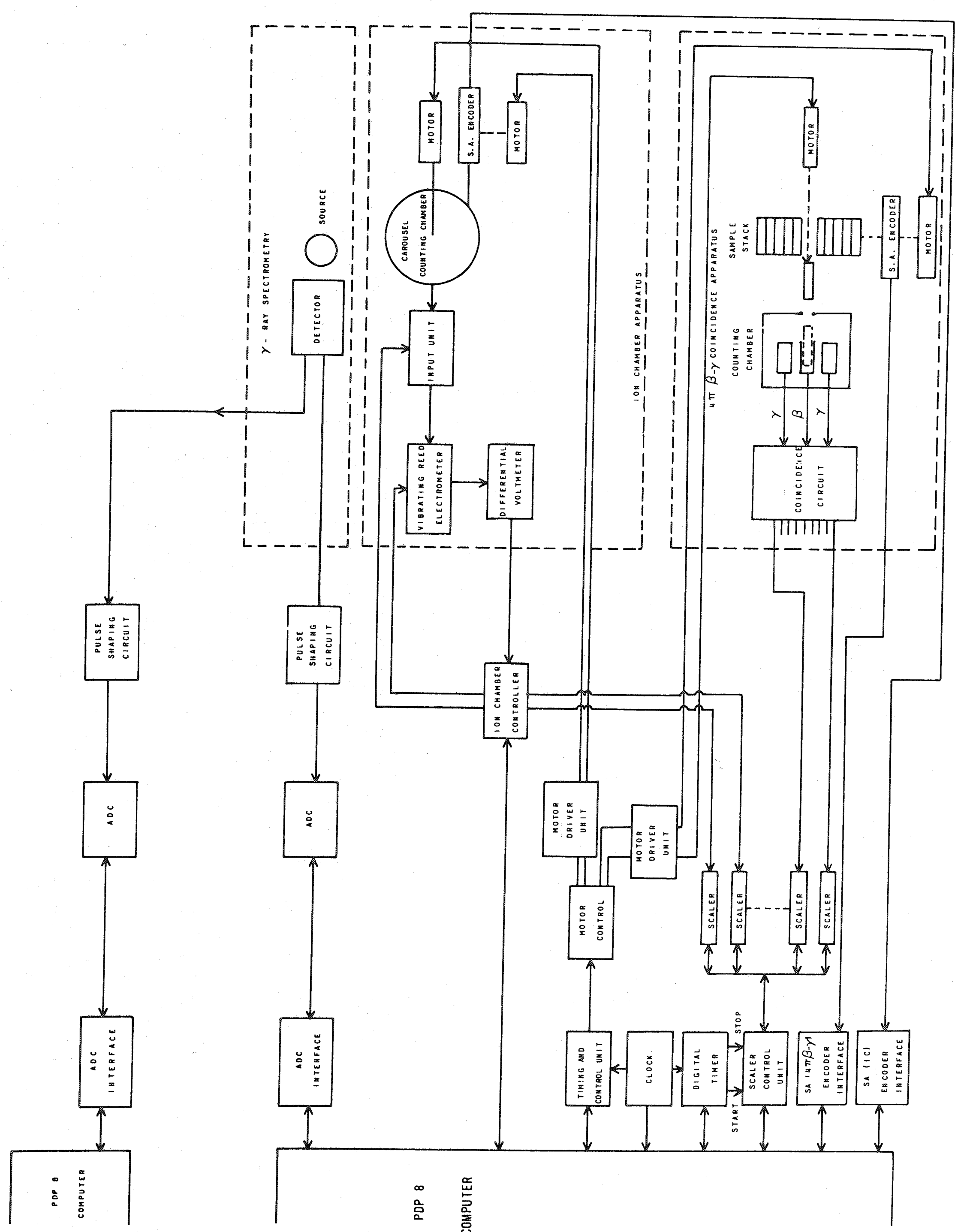


Fig. 1: Computer Control Systems

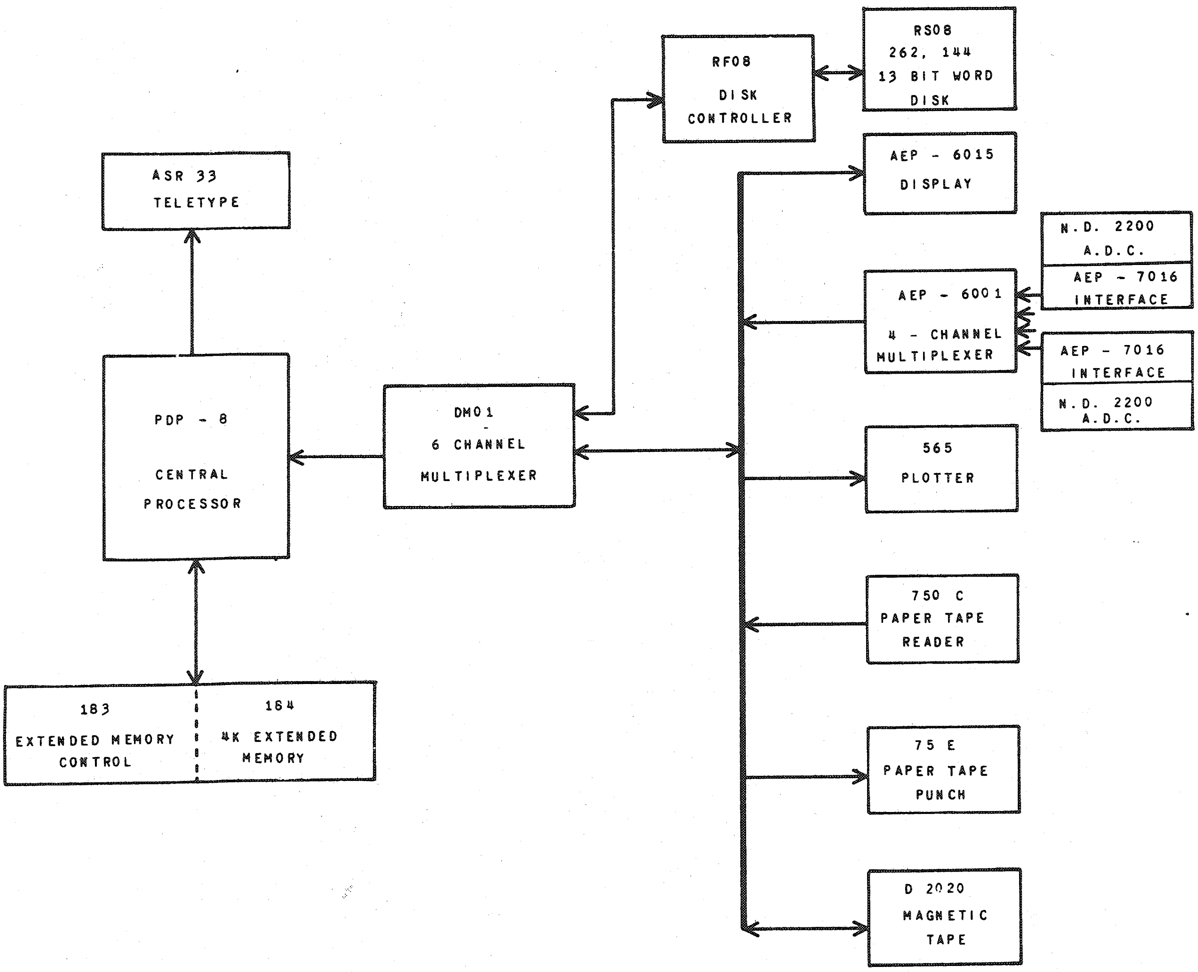


Fig. 2: Succeed Data Acquisition System

Table I

List of Computer Control Equipment

Used in the CRNL $4\pi(PC)-\gamma$ Coincidence System

Man/Comp Program Units	AEP - 7022
Digital Timer	EB - 5882
Motor Control Unit	AEP - 7053
Manual Motor Burst Generator	EB - 5841
Shaft Angle Indicator	EB - 5884
Position Motor Control	EB - 5883
Shaft Angle Encoder Input Module	EB - 5928
Motor Drive Unit	EB - 5894
Dual 2^{36} Bit Scalers	AEP - 7030
Dual Power Entry	EB - 5887
Real Time Clock (Conuclear)	C - 7029
Timing and Control Unit	AEP - 7017
3.6 V Power Entry	AEP - 7028
AC Power Entry	EB - 5656
Gated Oscillator	AEP - 7013
Data Break Multiplexer	AEP - 6001
Level Converter	EB - 5931
General Purpose Interface	EB - 5558
Junction Box A	EB - 5924
Junction Box B	EB - 5925
Junction Box C	EB - 5926

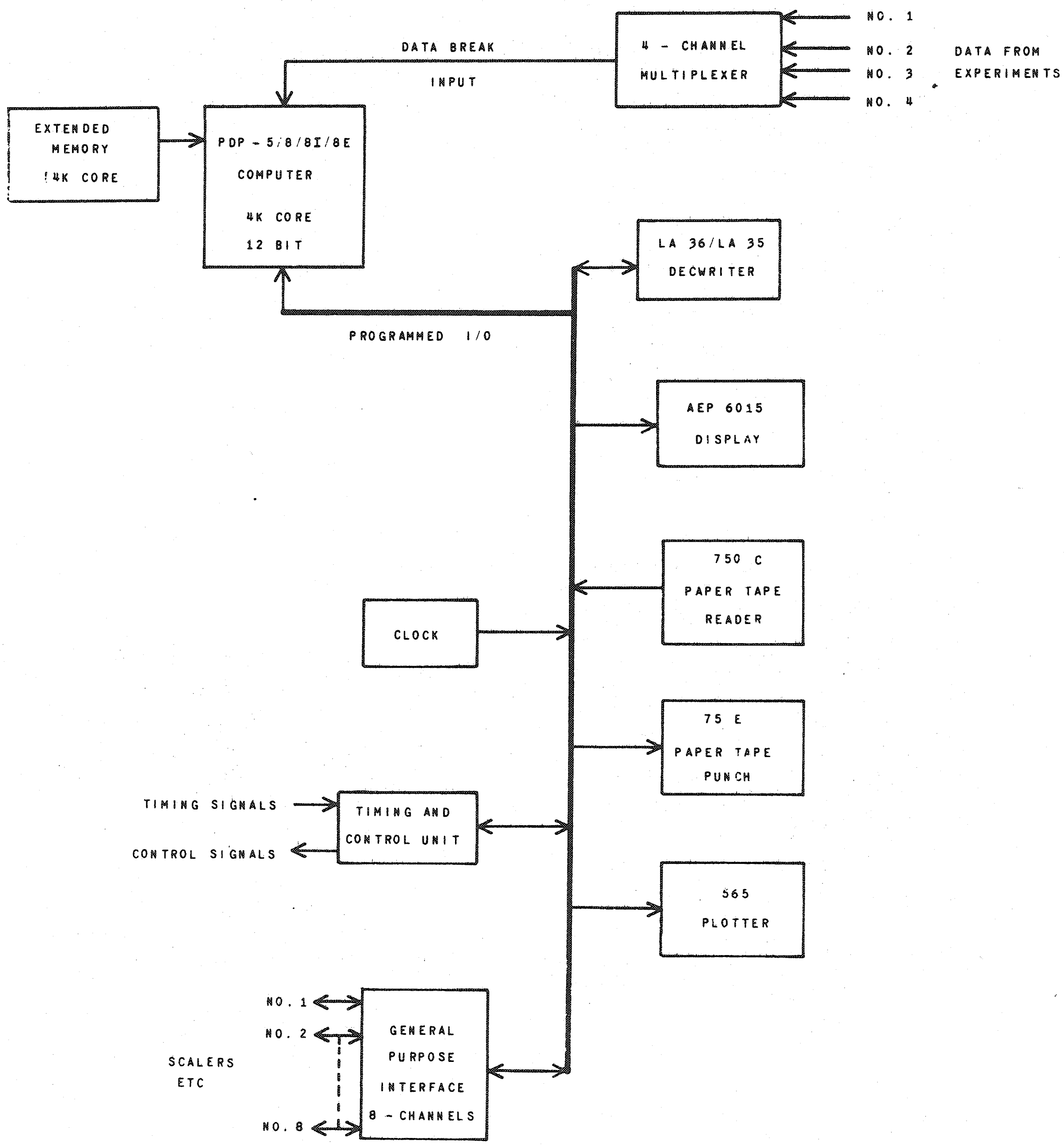


Fig. 3: Success 1 Data Acquisition System

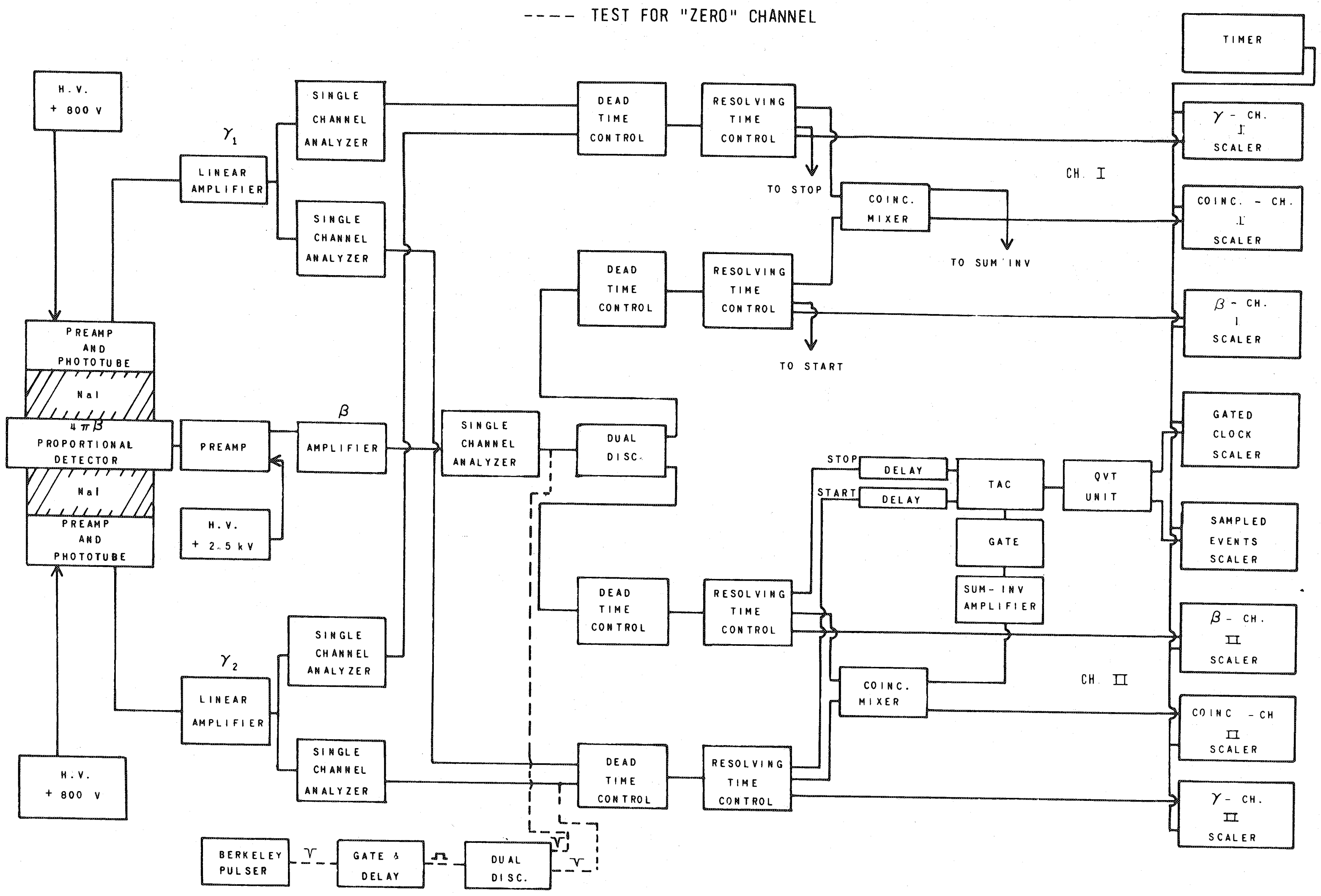


Fig. 4: Detectors and Electronic Instrumentation Used for 4π(PC)-γ Coincidence Counting

A photograph of the automatic sample changer designed by Taylor and Love (6) has been published by Taylor and Baerg (11), and is reproduced here in Fig. 5. With most of the lead shielding removed, as well as the upper NaI(Tℓ) detector, which is lying on some of the shielding at the right, the 4π(PC) can be seen at table level toward the right of the photograph. The system accommodates thirty-six samples, on thin source mounts (see section 7 of this report). They are loaded into trays that can be seen stacked vertically on shelves in the magazine at the left. A stepping motor, located underneath near the floor, is used to move the selected sample vertically. Then the sample is moved along the table into the 4π(PC) via a carrier, which can be seen in this picture midway between the magazine and the counter.

To obtain the most accurate results with the 4π(PC)-γ coincidence method, good counting statistics and favourable signal-to-background ratios are obvious requirements. This implies high counting rates (10^4 to 10^5/s) which can, in turn, introduce other sources of uncertainty. Thus the choice of electronic circuitry relates to the ability to use high rates while maintaining good pulse shaping which in turn provides precise timing with minimal losses. Insofar as is practicable, commercially available modular electronic circuitry has been selected. Table II lists the particular modules used during 1980 for the functions outlined in Fig. 4, and differs somewhat from previous tabulations for other standardizations (12,13). The rationale for the selection of these units and some information about their performance characteristics are discussed in section 2.1 and 2.2 of this report.

Figure 4 shows that, following the amplifiers in the γ channel and the single channel analyzer (SCA) in the 4π(PC) channel, the system is essentially twinned. The resulting dual set of electronics offers the opportunity to test one system versus the other for the same, or for different, experimental conditions. When comparing the two systems under the same conditions, the counting statistics effectively are cancelled. Thus small differences can be detected and this can be useful in trouble shooting. Furthermore, this twinned arrangement has proved to be a useful tool for the development and

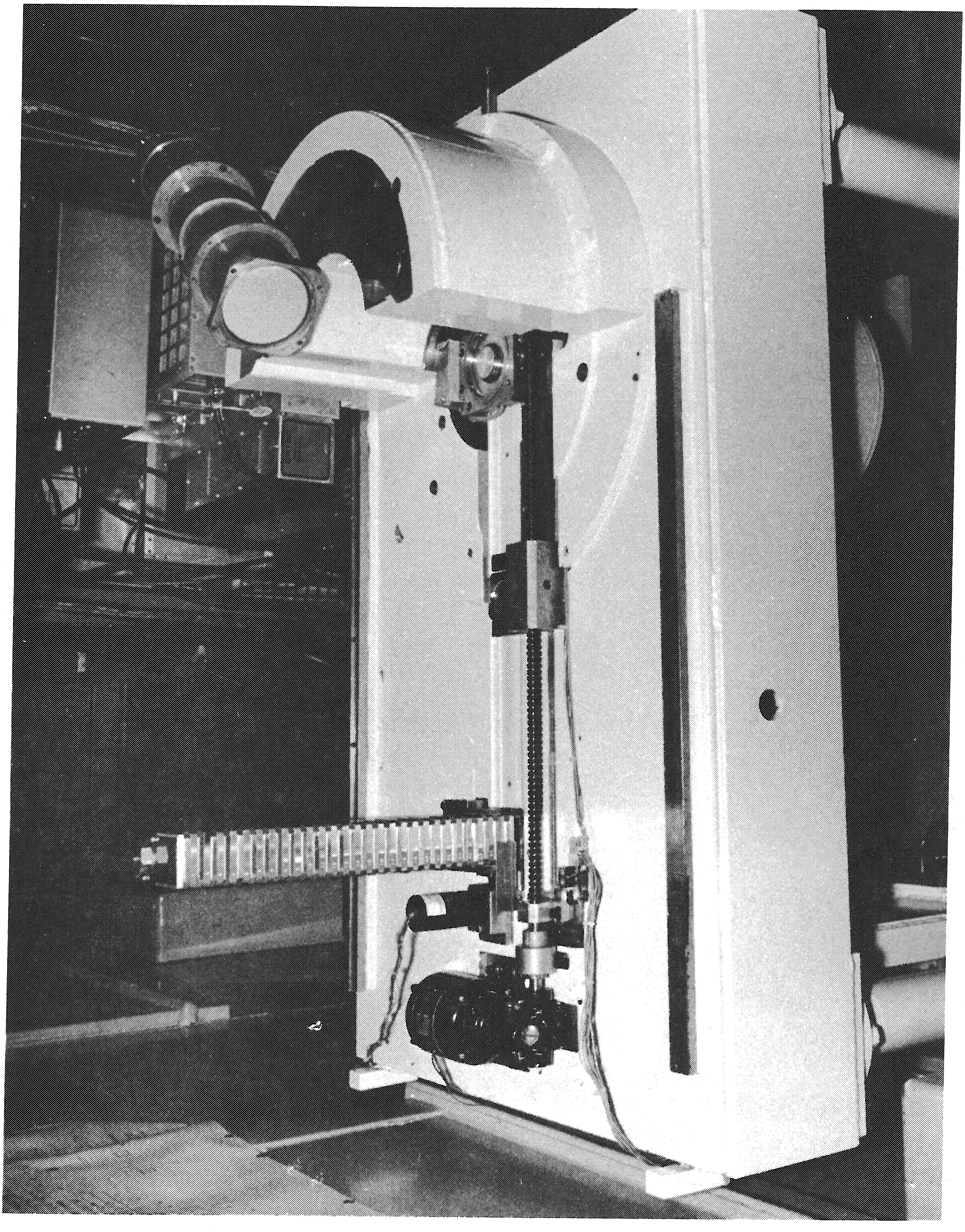


Fig. 5: The 4π(PC) Automatic Sample Changer

Table II

List of Model Numbers/Suppliers of Electronic Instrumentation

Used in the CRNL 4π(PC)-γ Coincidence System

Module	Quantity	Model Number	Supplier
β Preamplifier	1	CRNL designed preamplifier	
γ Preamplifier	2	CI 802-9	Canberra Industries
Linear Amplifier	4	CI 1411 DDL AMP	Canberra Industries
Single Channel Analyzer	4	420 SCA	Ortec
Single Channel Analyzer	1	551 SCA	Ortec
Sum/Invert Amplifier	2	433 SIA	Ortec
Dead-Time Control	4	LRS 222N (2) & 125 (2)	LeCroy Research System
Resolving-Time Control	4	LRS 123, 125 & 222N (2)	LeCroy Research System
Coincidence Mixer	1	LRS 162 (2)	LeCroy Research System
Dual Discriminator	1	LRS 108D	LeCroy Research System
Dual Discriminator	1	LRS 123	LeCroy Research System
Clock	1	TRI 1802 dual clock	Tomlinson Research Institutes
Timer	1	AECL EB 5882	CRNL Electronics Br.
Scaler	8 1	C7030 Multiscaler Level Translator (4)	Conuclear CRNL Standards Group
H.T. Supply	3	(2) HP6110A (0-3000V) (1) 413C (0-3000V)	Hewlett Packard Fluke
Delay	2	CI 1445	Canberra Industries
Delay	1	AD-YU 20AIC	AD-YU Electronics
Gate	1	Ortec 409	Ortec
Delay & Gate Generator	1	Ortec 416	Ortec
Time to Amplitude Converter	1	Ortec 437A TAC	Ortec
Pulser	1	BH-1	Berkeley Nucleonics
QVT Multichannel Analyzer	1	LRS 3000-1	LeCroy Research System
Analog to Digital Converter	1	ND 2200 ADC	Nuclear Data Inc.
Interface	1	AECL AEP-7016	CRNL Electronics Br.
Oscilloscope Display	1	TEK RM561A	Tektronix Inc.

assessment of appropriate experimental conditions for the metrology of various nuclides. Recent examples were the simultaneous use of different γ-channel windows in the assay of ^{134}Cs (13), and of different dead times and resolving times in the assay of ^{233}Pa (14).

2.1 ELECTRONIC INSTRUMENTATION FOLLOWING THE 4π(PC) DETECTOR

Requirements of the 4π(PC) channel are fast recovery time, good overload characteristics, good timing that is constant for a large range in pulse amplitude, a variable gain, and low noise.

The preamplifier is situated as close as possible to the 4π(PC) to minimize noise and stray capacitance. Because commercially available preamplifiers lack the ability to handle a large dynamic range without severe overloading, Frketich and Taylor (15) assembled a logarithmic preamplifier in this laboratory some years ago, and an adequate replacement has not yet been obtained from a commercial supplier. The first stage is a commercially produced non-blocking preamplifier, model Tennelec TC-132. This is followed by a logarithmic stage which consists of a Philbrick/Nexus PP45U operational amplifier with diode-connected transistors as the logarithmic feedback elements. This preamplifier cannot be driven into overload and it recovers from any input pulse in <2 μs.

The rest of the electronic equipment is located one or more metres away. The amplifier, a Canberra model CI 1411 Double Delay Line (DDL) amplifier, has a clipping time of 0.5 μs for a single lobe, which provides a recovery time that is comparable with that of the 4π(PC) detector. The gain is selected to avoid overload. The linear bipolar output pulses are suitable for feeding a timing single channel analyzer (SCA). The fast negative output from the SCA is used as a trigger circuit. The same model, Ortec-420, is used in both the 4π(PC) and γ channels to provide precise crossover time pickoff. In the 4π(PC) channel it is operated in the integral mode with the discrimination level suitably set at ≈ 1 V to eliminate noise (16).

A LeCroy model 108D discriminator provides the larger pulse height which is required to drive the units used for setting channel dead times. This unit provides two independent outputs. The dead time unit is a LeCroy model 222N dual gate and delay generator, and has been chosen primarily because it gives a dead time that is nonextendable. Without this feature accurate dead-time corrections cannot be made (17). It has been found that the dead time of this unit remains constant as set, even at rates that approximate the reciprocal of the dead time. Furthermore, the duration of the output pulse, or dead time, can be selected over a wide range. This is convenient for experimental work, and for activity measurements of nuclides that decay through delayed transitions where longer dead times and coincidence resolving times are required (2). For routine work 2 μs is a typical dead time that satisfies the requirement for it to be 1) longer than the dead time of any other electronic component in the channel (17), and 2) greater than twice the coincidence resolving time (2). Other good features of the 222N are the sharpness of the pulse shape and its dual nature, i.e. it is a set of two gate and delay generators with essentially no delay between its halves. Thus it is convenient to use the other half of it to set and maintain the coincidence resolving time of the 4π(PC) channel.

A LeCroy model 162 dual three-fold logic unit is used to detect coincident pairs of pulses from the 4π(PC) and γ channels. Its special feature, called "deadtimeless operation," is advantageous in allowing the circuit to be retriggered immediately after the output pulse, for which a width of 150 ns normally is selected. For nuclides that decay promptly, the resolving time is set at 0.7 μs. This is sufficiently long to prevent loss of true coincidences from time jitter that arises largely from the 4π(PC) (2).

Finally the pulses are registered in 10-MHz scalers which are used in the decimal mode. Up to nine digits are accommodated in each scaler.

2.2 ELECTRONIC INSTRUMENTATION FOLLOWING THE GAMMA DETECTORS

New preamplifiers and phototubes were obtained in 1978, assembled as integral units with NaI(Tℓ) detectors by the supplier. The phototubes were selected to give gain stability within 0.1% for input rates up to $2 \times 10^4\ s^{-1}$. Previously gain stability had proved to be a problem for nuclides with complex decay schemes, as changes in the γ-channel gate that resulted from source-rate variations gave irregularities in the slope of the 4π(PC)-efficiency function.

The amplifier and its following trigger circuit (Fig. 4) are selected to satisfy the need for precise timing in the determination of coincident pairs of pulses from the 4π(PC) and γ-ray detectors. Optimum timing is realized through crossover pickoff. For this purpose bipolar output pulses are required. Optimum γ-ray resolution is sacrificed in favour of fast recovery time and reduced time jitter of the output signal. The Canberra 1411 DDL amplifier has served well in this regard, with a clipping time of 0.35 μs for a single lobe. The clipping times of the two amplifiers following the two γ-ray detectors are equal within 20 ns. Even with these short clipping times a γ-ray resolution of 6.7% is obtained at 660 keV. Because individual γ-channel windows are set via the SCA's, some system non-linearity can be tolerated. The potentiometer-controlled fine gain adjustment of the amplifier is a convenience for matching pulse heights from the two NaI(Tℓ) assemblies.

Ortec Model 420 timing SCA's are used because they offer selection of γ-channel windows to accept the desired part of the spectrum, and individual adjustment of time delays for balance versus the arrival time of coincident pulses from the 4π(PC). In addition the fast negative output is required for timing precision. The slow positive output is useful during experimental setup for setting the γ-channel windows via a multichannel analyzer (see Section 2.3). The stability is adequate provided the operating conditions are chosen so that the potentiometers are not set in the lowest part of their range. As the thermal stability for the delay is specified as 0.4 ns/K and the precision with which the relative delays are adjusted is ±2 ns, small temperature fluctuations in the laboratory can be tolerated. For each SCA the walk adjustment is set for minimum time walk according to the manufacturers recommendations. Once set, the walk adjustment should remain stable; however, experience indicates the adjustment should be examined occasionally.

Currently various modules (see Table II) are used in the γ channels for setting and controlling dead time. They are as satisfactory as the LeCroy 222N, insofar as nonextendable dead time is concerned. However, they are less convenient to use, having a limited range for the output pulse duration, and the pulse rise time is less sharp. Furthermore, some time delay is introduced by cable between one module and the next. Therefore it is planned to install twin units of the 222N in the γ channels in the near future.

3. INSTRUMENTATION AND PROCEDURE FOR SETTING GAMMA-CHANNEL WINDOWS

The instrumental requirements for accuracy at some of the various stages in the standardization of a radionuclide are quite simple. The setting of γ-channel windows is one of the more straightforward steps, but careful judgement during set-up is needed.

Figure 6 shows the instrumentation used to set γ-channel windows conveniently and precisely. The SCA's are used in the differential mode. A sufficiently high amplifier gain is selected so that the discrimination levels of the SCA's can be employed in the upper range of the potentiometers. It is preferable to make gain adjustments via the DDL amplifiers, instead of via the high voltage supplies. There are two reasons for this, 1) the gain of the system has been found less stable immediately after a change in applied voltage, and 2) there is increased risk of spurious pulses from excessive applied voltage (16). After examining the full γ-ray spectrum for each NaI(Tℓ) detector, the gain of one DDL amplifier is adjusted so that the location of the peak of a prominent γ ray appears at the same channel number of the ADC for both detectors.

Next, the positive output pulse from one of the SCA's is suitably delayed so that an appropriate gating pulse can be fed to the ADC. The full and gated spectra are examined alternately while the discrimination levels are adjusted to include the desired part of the γ-ray spectrum in the window. With care this normally can be judged visually from the oscilloscope display, without consuming time to plot or list data. Then this procedure is repeated for each of the other three SCA's.

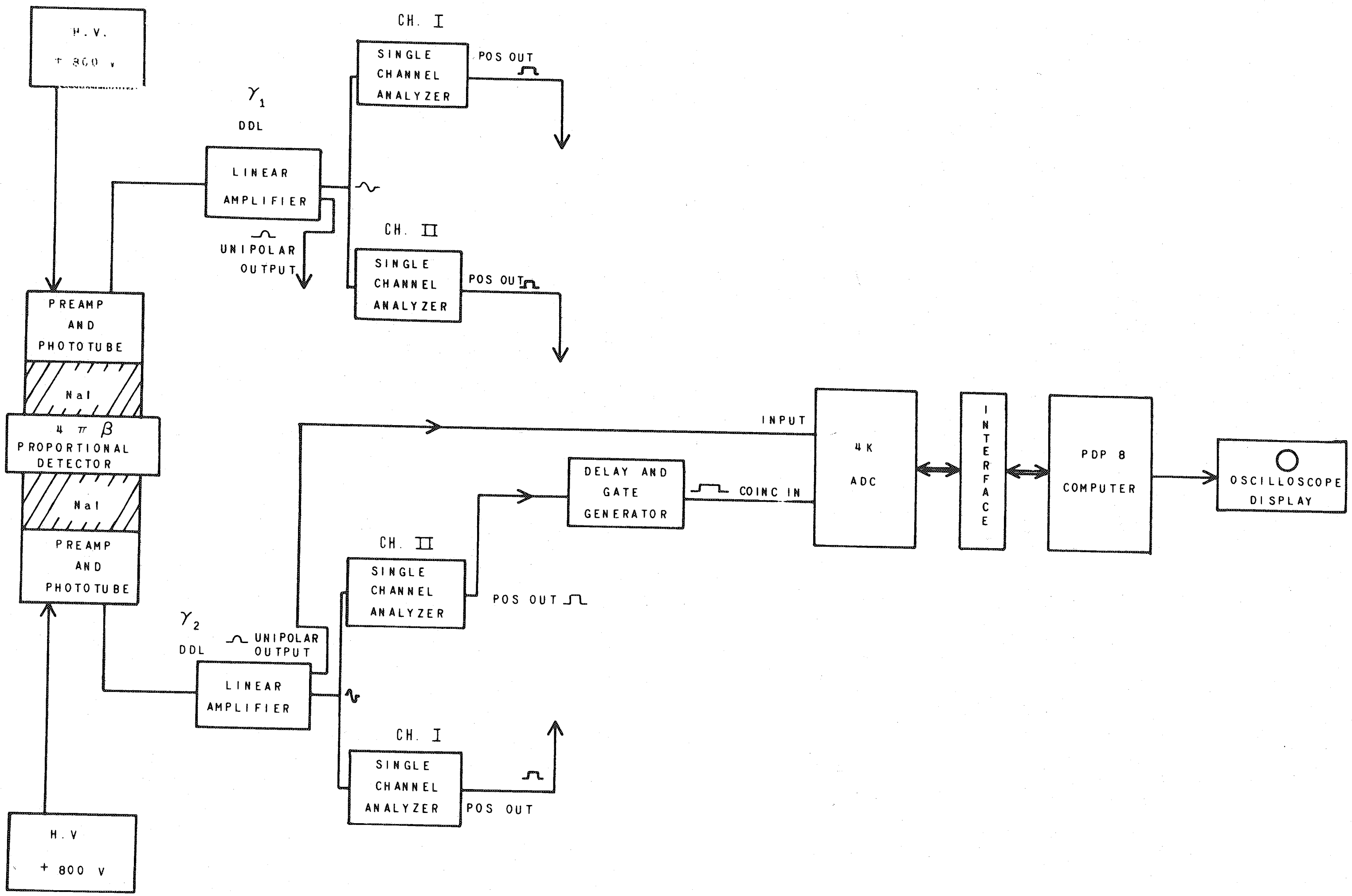


Fig. 6: Instrumentation Used to Set γ-channel Windows

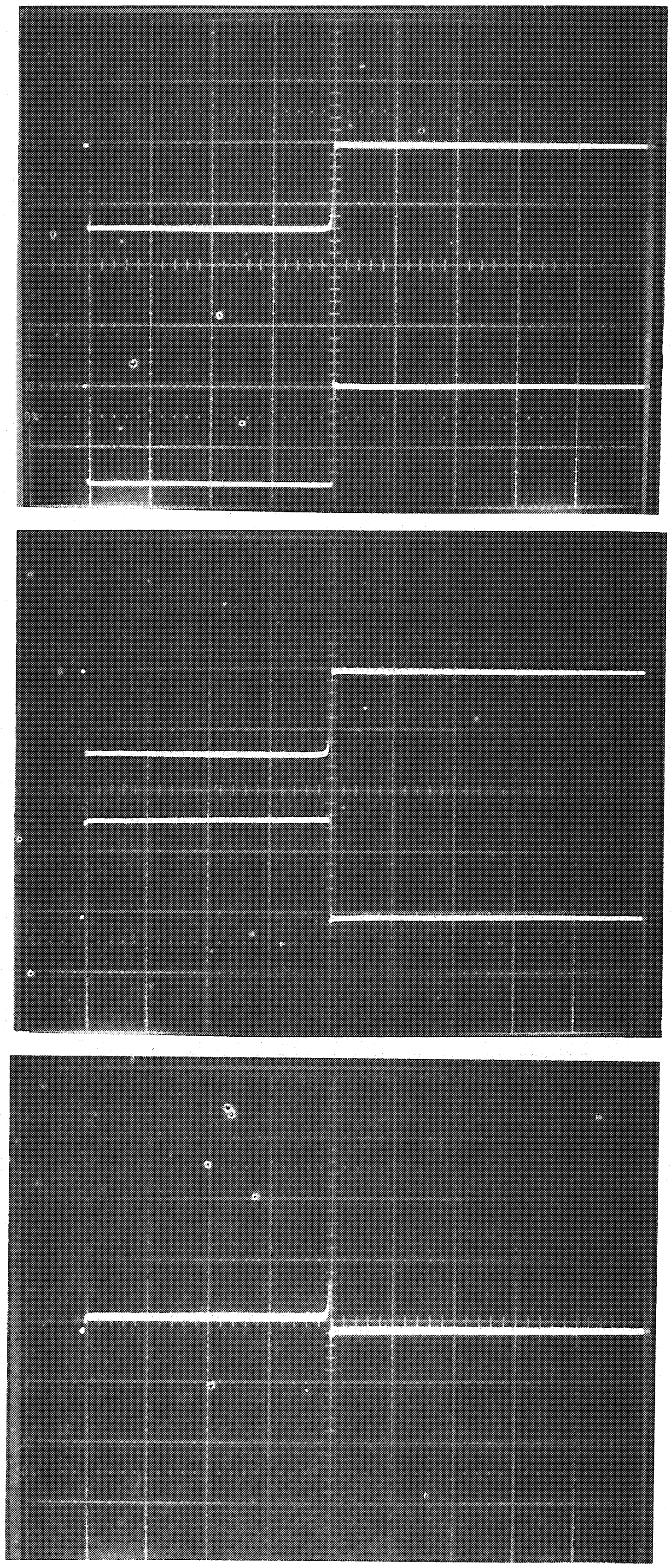
4. ADJUSTMENT AND MEASUREMENT OF DEAD TIMES AND RESOLVING TIMES

Two requirements of the coincidence equations developed by Bryant (18) are (1) equal non-extending dead times for the 4π(PC) and γ channels, and (2) dead-time values greater than twice the coincidence resolving times (2).

Our procedure for setting the duration of dead times is as follows. Suitable pulses are fed to the dead-time control units. The level outputs from both the 4π(PC) and γ channels are examined with an oscilloscope as shown in Fig. 7a, and set approximately for the desired dead time, which typically is 2 μs. By inverting one of the outputs displayed on the oscilloscope, as shown in Fig. 7b, and then summing them, as shown in Fig. 7c, a small difference between the two dead times is easily noticed. While observing these summed outputs, one of the pulse widths is adjusted with respect to the other so that overshoots are minimized. This final adjustment gives dead times that are equal within ≈10 ns.

The same technique is used to set the coincidence resolving times of the 4π(PC) and the 4π(PC) and γ channels to be equal (Figs. 8a,8b,8c). These are set at less than half the duration of the dead times to prevent loss of true coincidences from excess time overlap (2, 18). Typical resolving times are 0.7 μs. However, for nuclides that decay via delayed transitions, the resolving times are set to be about twelve times the half-life of the delayed state to reduce the probability of non-detection of true coincident pairs of pulses to a negligible level (1 in 2^{12}). ^{57}Co with its 98-ns delayed γ transition is an example for which somewhat longer resolving times and dead times are used.

The dead times are measured with the source-pulser method as described in a recent handbook (17). Figure 9 shows the electronic circuit used for these measurements. Pulses from the 4π(PC) are fed via the preamplifier and main amplifier to a sum invert amplifier (SIA); the output then feeds an SCA for each of the 4π(PC) and γ channels (only one of the γ-channel SCA's is employed in each system, and in the integral mode). A radioactive source is counted in the 4π(PC) at a voltage which is near the starting point of the plateau, in

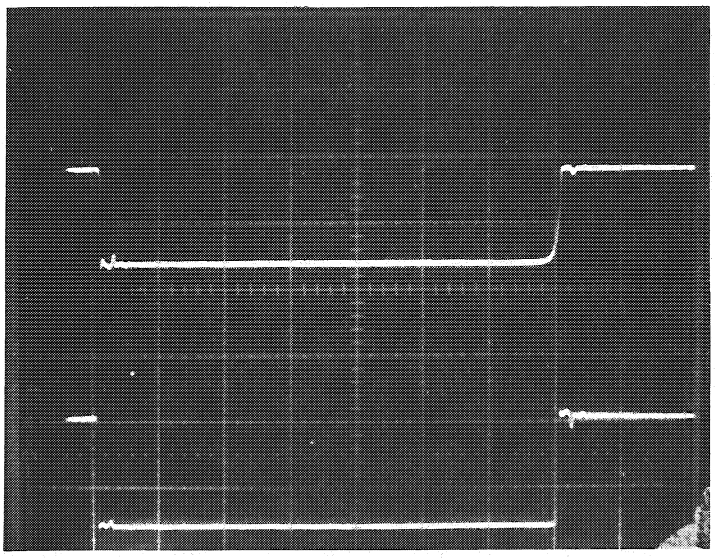


(a)

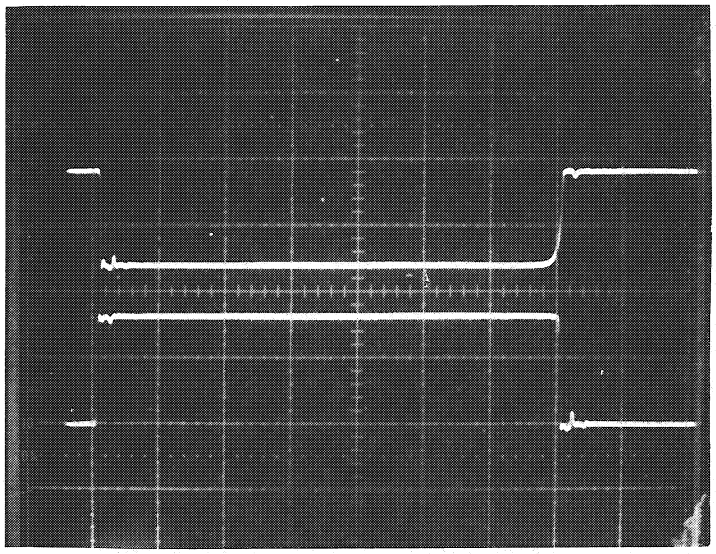
(b)

(c)

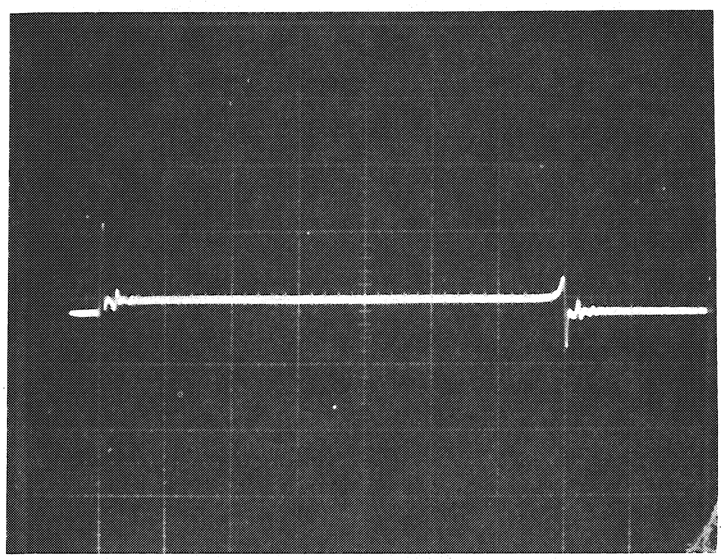
Fig. 7: The Procedure for Setting and Matching the Dead Times of the Beta and γ Channels. Scale of the Oscilloscope Trace is 0.5 μs/div. and the Dead Time is 2.0 μs.



(a)

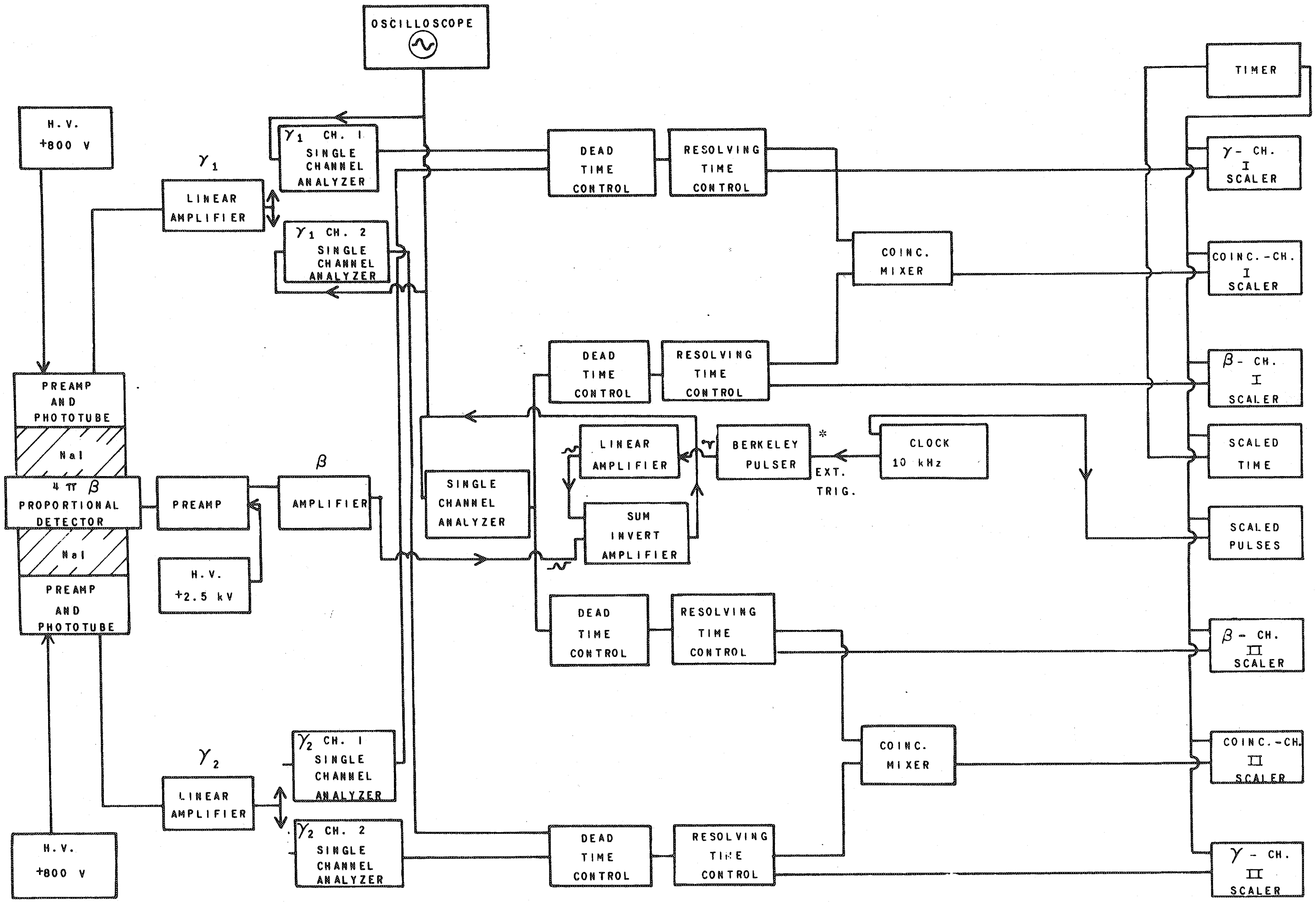


(b)



(c)

Fig. 8: The Procedure for Setting and Matching the Resolving Times of the Beta and γ channels. Scale of the Oscilloscope Trace is 0.1 μs/div. and the Resolving Time is 0.7 μs.



*EXT. TRIGGERING TO BERKELEY PULSER IS CONNECTED FOR COUNTING SOURCE & PULSER THEN IS DISCONNECTED FOR COUNTING SOURCE ONLY

Fig. 9: Instrumentation used to Measure Dead Times

order to inhibit spurious pulses (16). Figure 10a shows an oscilloscope display of the output pulses from the DDL amplifier for a source of ^{90}Sr + ^{90}Y. The pulse shape is selected to minimize overshoot of the baseline, which could trigger a false count.

The same source is counted again, but with a train of similarly shaped pulses superimposed (Fig. 10b). These are generated by a tail pulser, which is externally triggered by a precision clock, and then fed through a linear amplifier and SIA to the same SCA's. Typical rise and fall times from the pulser are 0.1 and 0.5 μs , respectively.

From the counting results, the dead time, θ, is calculated as follows (17), where N_s is the source rate, N_p is the pulser rate, N_{sp} is the rate for source counted together with the pulser, and $\Delta = N_s + N_p - N_{sp}$

$$\theta = [1 - (1 - \Delta/N_p)^{\frac{1}{2}}]/N_s$$

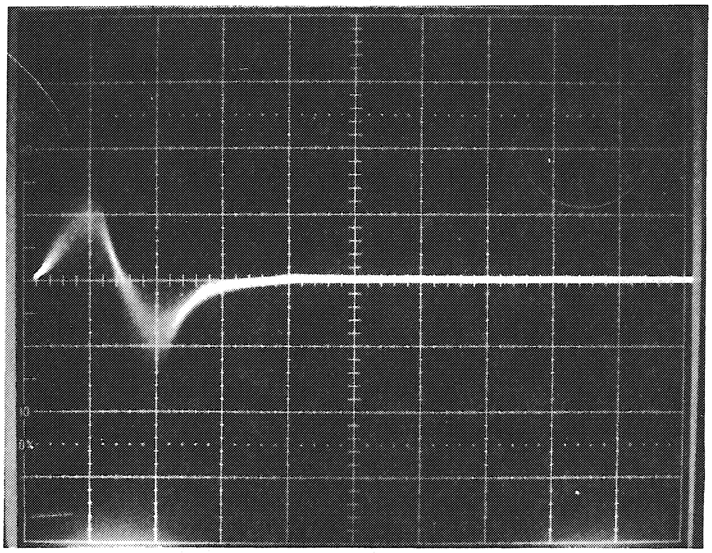
The resolving times are measured by counting the pulses from a source in the 4π(PC) while feeding pulses from a pulser to the gamma channel, and observing the coincident rate, which is entirely from accidental coincidences. The circuit is shown in Fig. 11. The resolving time, τ_R, is given by

$$\tau_R = N_c / 2N_sN_p$$

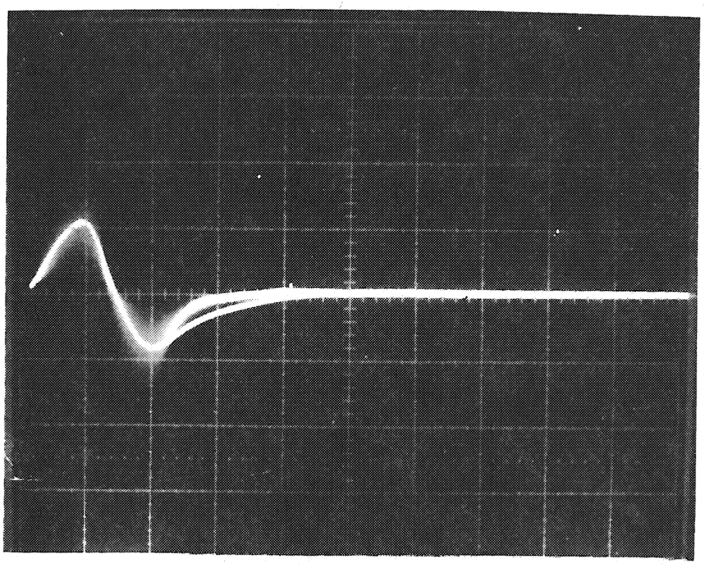
where N_c is the rate in the coincidence channel. Normally several consecutive measurements are made for these parameters. The standard deviation in the mean value is taken as the uncertainty.

5. TREATMENT OF THE TIME DELAY BETWEEN CHANNELS

Gandy (19) showed that inaccuracy arises in the 4π(PC)-γ coincidence method from mismatch in the relative delay between coincident pairs of pulses from the 4π(PC) and γ detectors. He developed the coincidence



(a)



(b)

Fig. 10: (a) Output Pulses from the DDL Amplifier of the 4π(PC) for a source of ^{90}Sr + ^{90}Y (b) Output of Both the Source and 10 kHz Tail Pulser. Scale of the Oscilloscope Trace is 5.0 V/div. Vertical and 0.5 μs/div. Horizontal.

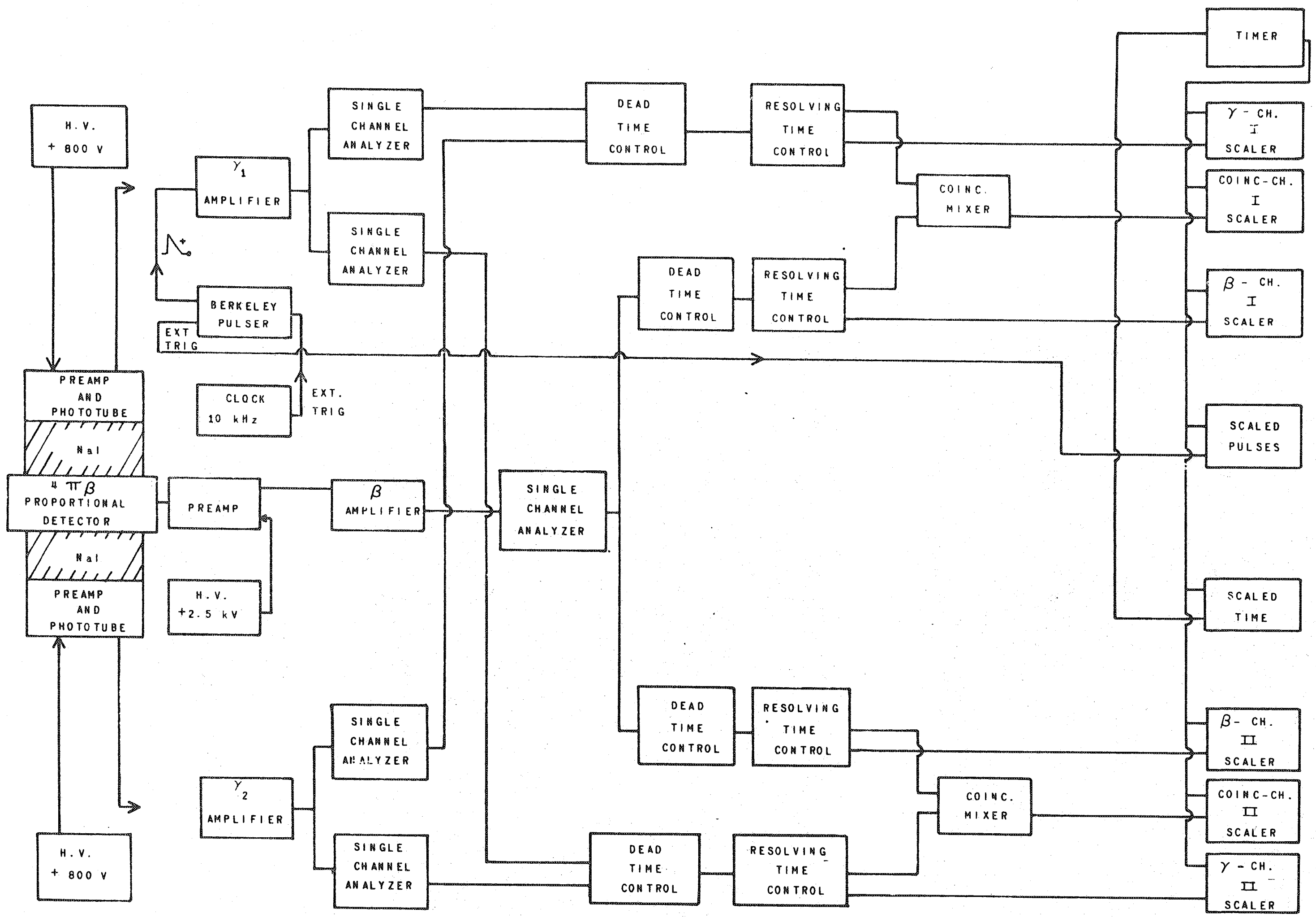


Fig. 11: Instrumentation Used to Measure Resolving Times

equations to correct for mismatch, which invariably exists in practical counting systems. This correction is rate-dependent:

$$\Delta N_o \propto (N_{4\pi} - N_{\gamma})\delta$$

where ΔN_o is the correction to the activity, $N_{4\pi}$ and N_{γ} are the 4π(PC) and γ-channel rates, respectively, and δ is the difference in arrival times at the coincidence mixer between pairs of pulses from the 4π(PC) and γ channels. In practice the instrumentation can be set up so that δ is very small. Williams and Campion (20) proposed such an arrangement of electronics. With it the system can be set up to compensate the delays so that mismatch is negligible for the particular source and conditions that prevail during set-up. However, the resulting compensation does not give $\delta = 0$ for other sources (of even the same radionuclide) with different counting rates and efficiencies; it may be even less appropriate for other γ-channel windows, or other gain conditions in the 4π(PC) system (amplifier settings, HT voltage, or discrimination level). As a consequence, the delay settings invariably are inappropriate ($\delta \neq 0$) during a run in which several different sources are counted with an automatic sample changer.

Figure 4 outlines the electronics used here. It serves two purposes: 1) during setting up prior to a run, to match the delay between the 4π(PC) and γ channels, with the γ-channel windows and other conditions that have been selected for the run, in a manner similar to that of Williams and Campion (20), and 2) during the run, to collect information from which corrections for the prevailing delay mismatch can be computed for each counting interval, in a manner patterned after one used here by Taylor and Gibson (21). Instead of the latter, some workers have proposed continuous instrumental compensation of the delays during a run (22). There are two reasons why we prefer to obtain data from which appropriate corrections can be deduced: 1) the output data alerts us to instrumental faults that might otherwise go unnoticed; and 2) this data is useful in the assessment of that part of our overall uncertainty that comes from delay mismatch. However, we note that automatic compensation is advantageous in a system that employs a pressurized 4π(PC) with variable threshold discrimination, where the required delay compensations between successive counts normally are very much larger.

Thus our operating practice is as follows. In setting up for a standardization, first the γ-channel windows and gain conditions for the 4π(PC) channel (high voltage, amplifier gain and SCA discrimination level) are selected, and then the arrival times of pulses from the 4π(PC) and γ channels are adjusted so that $\delta = 0$. This is done for each of the twin systems in turn. The same train of logic pulses from an externally triggered precision pulser is shaped and fed to the dead-time control units for the 4π(PC) and γ channels (see Fig. 4), but with all amplifier inputs disconnected. The start or stop times are adjusted via either an active or a passive delay circuit, so that they are displayed conveniently at about mid-range of the multichannel analyzer (MCA) (Table II). For reasons of stability most of the required time delay is inserted via a passive circuit. Scaled outputs from the MCA are recorded: 1) n pulses for each count recorded in channel n, and 2) a busy signal which indicates the number of counts analyzed. Then the ratio of these scaled outputs gives the channel number, A, that corresponds to zero relative delay between channels, which is called "zero channel." Then the time delay of the passive delay circuit is altered by a known amount, x (200 ns is typical), and the channel number, B, that now corresponds to zero relative delay is observed again. The time calibration of the system (ns per channel) is given by $x/(A - B)$. Next, the delay, x ns, is removed, the amplifier inputs are reconnected and the pulser outputs are disconnected (i.e. normal counting conditions). A radioactive source (representative of the run) is counted and the ratio of the scaled outputs from the MCA, A/B, is observed. Adjustments are made to the γ-channel delay at each SCA, in turn, so that this ratio matches "zero channel."

During the run the scaled outputs from the MCA are recorded for each counting interval, which then allows individual corrections for the delay mismatch to be computed.

Figure 12a shows the relative time spectrum for a ^{60}Co source with methane as the counting gas; the same is shown for ^{57}Co with methane and with a gas mixture of 10% methane in argon (P-10) in Figs. 12b and 12c, respectively.

6. *CALCULATION OF THE RESULTS*

The coincidence formulae used follow those developed by Bryant (18) with modifications to accommodate small differences between the dead times of the 4π(PC) and γ channels, and to correct for delay mismatch (19).

Dead-time and background corrections to the observed rates in the various channels are computed as follows, where

$N_{4\pi}$, N_γ and N_c are the corrected rates of the 4π(PC), γ, and coincidence channels, respectively,

$N'_{4\pi}$, N'_γ and N'_c are the observed rates,

θ's are the dead times,

B's are the background rates,

τ's are the resolving times, and

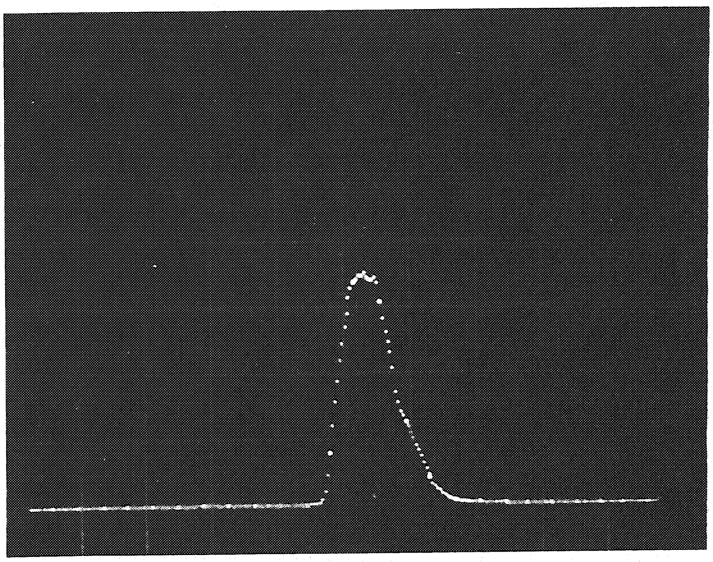
δ is the mismatch in delay; it is positive when a pulse from the γ channel arrives before its coincident counterpart from the 4π(PC):

$$N_{4\pi} = [N'_{4\pi} / (1 - N'_{4\pi}\theta_{4\pi})] - B_{4\pi}$$

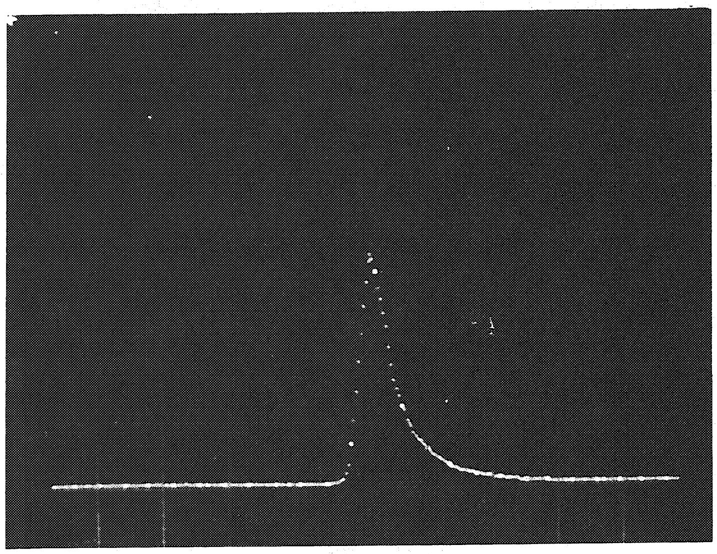
$$N_\gamma = [N'_\gamma / (1 - N'_\gamma\theta_\gamma)] - B_\gamma$$

$$N_c = \frac{[N'_c - N'_{4\pi}N'_\gamma (\tau_{4\pi} + \tau_\gamma)]\ [(1 - N'_{4\pi}\theta_{4\pi}) + (1 - N'_\gamma\theta_\gamma)]}{K[(1 - N'_{4\pi}\theta_{4\pi}) + (1 - N'_\gamma\theta_\gamma) + (2N'_c\theta_c) - 2(N'_{4\pi}\tau_\gamma + N'_\gamma\tau_{4\pi}) + 2(N'_{4\pi} - N'_\gamma)\delta]} - B_c$$

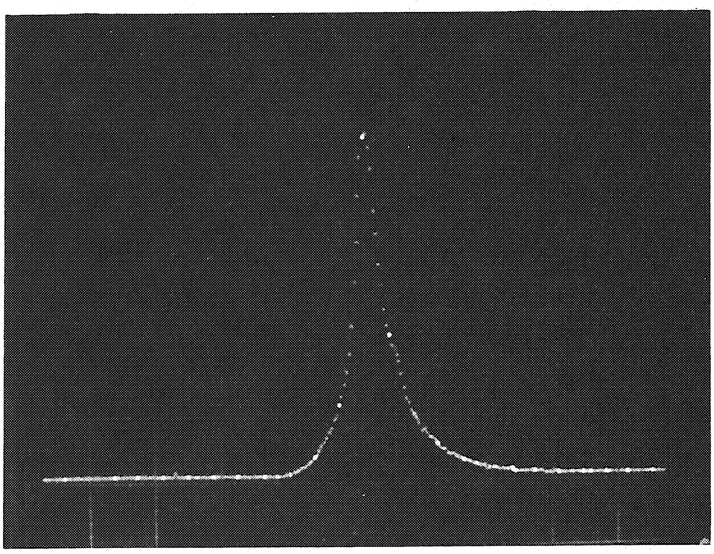
where $K = (1 - N'_{4\pi}\theta_{4\pi})\ (1 - N'_\gamma\theta_\gamma)$



(a) ^{60}Co with methane gas



(b) ^{57}Co with methane gas



(c) ^{57}Co with P-10 gas

Fig. 12: Oscilloscope Trace Showing the Time Spectrum: the TAC Output versus Time (10 ns/ch).

Then the efficiency for the 4π(PC), $\mathcal{E}_{4\pi}$, and the activity, N_o, are given by:

$$\mathcal{E}_{4\pi} = N_c / N_\gamma$$

$$N_o = N_{4\pi} N_\gamma / N_c$$

These equations assume that in β or electron capture decay processes, the 4π (PC) detects β particles, X rays or Auger electrons, and that prompt γ rays are detected by the γ detectors only. Campion (2), Taylor (23) and others have discussed the complications that arise when these conditions are not strictly observed. The nature of the problem is as follows. For decay events in which the primary decay process is not detected in the 4π(PC)(i.e. $\mathcal{E}_{4\pi} < 1$) there is a finite probability that a γ transition is detected instead; this could be by detection of either a γ ray or a conversion electron. A similar complication arises for a decay scheme with a branch that decays directly to the ground state. In all such cases the 4π(PC) rate and N_o are inconsistently high with respect to the ratio, N_c / N_γ, or $\mathcal{E}_{4\pi}$. The accepted method for deducing the activity is by 1) the accumulation of data in which $\mathcal{E}_{4\pi}$ is varied over a convenient range (20 to 30%), and then 2) extrapolation of the data to $\mathcal{E}_{4\pi} = 1.0$ (5, 23). For this purpose a least-squares fitting program based upon one developed by Schmidt (24) is used in this laboratory.

The coincidence formulae of Bryant (18) have been found to give deviations at high rates, e.g. $\approx 0.3\%$ for source activities of 10^5 Bq (3 μCi) (25), depending upon counting conditions. The more recent formulae proposed by Cox and Isham (26) and developed by Smith (25) are being introduced, but were not used during 1980 for the standardizations of ^{60}Co and ^{57}Co discussed in this report.

7. *STANDARDIZATION OF ^{60}Co*

A stock of ^{60}Co was obtained from a commercial supplier in 1978. Thus, isotopic impurities should have decayed to negligible levels by 1980, when the stock was purified by elution from an anion exchange column with a solution of HCl (4 mol/L). No impurities were found, either before or after chemical purification, by examination of the γ-ray spectrum with a Ge(Li) detector. We conclude that impurities in this stock are < 0.1%. The stock was evaporated to dryness and dissolved in a solution with HCl and CO^{+2} concentrations of 0.3 mol/L and 10 mg/L, respectively. Two dilutions were prepared.

The methods used for source preparation, dilution, and sampling have been described in earlier reports (27,28).

Eleven sources mounted on thin metallized plastic VYNS films (29) were prepared from the two dilutions and were used for the coincidence measurements. The activity range was from 6 to 40 kBq (0.2 to 1 μCi). Other samples of the master solution and the dilutions were sealed in ampoules. Some of these were submitted to the Bureau International des Poids et Mesures (BIPM) and the International Atomic Energy Agency (IAEA) for registration in the international reference systems for measuring activity of γ-ray-emitting nuclides. Other ampoules were used for calibration of our 4πγ ionization chamber (IC). In addition, thirty-six sealed sources were prepared to serve as standards for calibration of miscellaneous counting systems in other laboratories, mostly within AECL, as well as some samples of the dilutions. The distribution of these samples of ^{60}Co is given in Table III.

For 4π(PC)-γ coincidence counting, the γ-channel windows for both systems of electronics were set to include only the two photo peaks of 1173 and 1333 keV, as shown in Fig. 13.

The standard deviation in the final result from the eleven thin sources, each counted four times with both sets of electronics, was ±0.016%. The results showed that the two dilutions and the two systems of electronics were consistent within this statistical uncertainty. Systematic uncertainty was assessed as described in an earlier report (13) and our estimates of known causes are given in Table IV. Where practicable, a confidence level of 67% was used in arriving at these estimates. Consider, for example, the contribution

Table III

Summary of the Distribution of Samples of ^{60}Co

Type of Sample	Relevant Solution	Use or User
Ampoules	dilution no.1	BIPM
Ampoules	dilution no.2	IAEA
Ampoules	master dilution no.1	4πγ IC calibration
Ampoules	dilutions nos.1 & 2	CP*, Environmental Research Branch of CRNL, WNRE**
Sources	master	Ge(Li) calibration, CP, 6 branches at CRNL
Sources	dilutions nos.1 & 2	4π(PC)-γ coincidence, Ge(Li) calibration, 8 branches at CRNL, WNRE, U. of Guelph

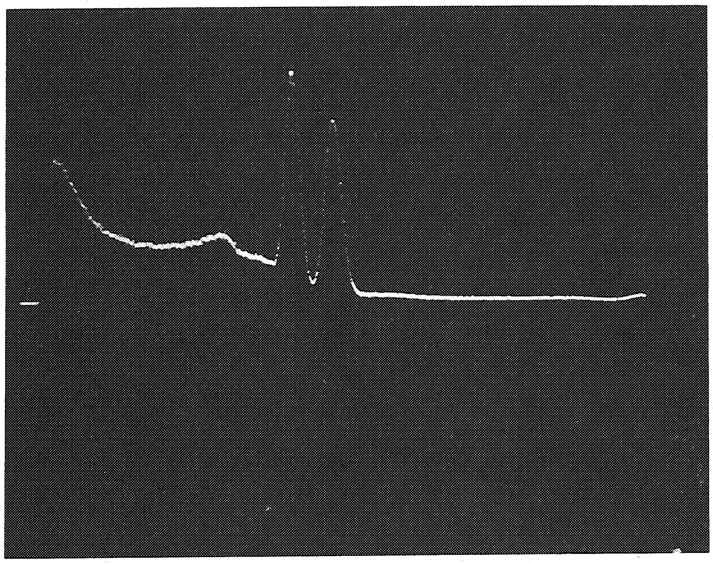
Table IV

Estimates of Systematic Uncertainty in the Final Results from Various Causes

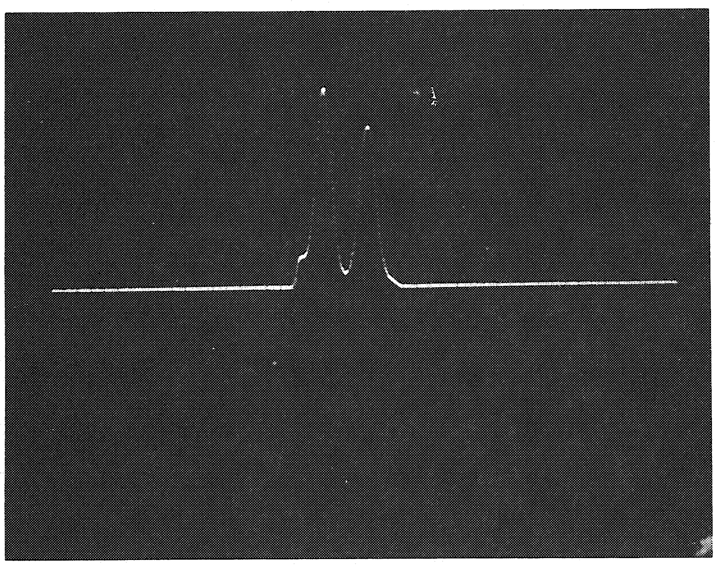
Cause	Uncertainty (%)	
	^{60}Co	^{57}Co
Uncertainty in dead time (20 ns)	0.01	0.01
Uncertainty in resolving time (2 ns)	0.01	0.01
Uncertainty in delay mismatch (5 ns)	0.01	0.01
Sampling error	0.01	0.01
Impurities	<0.1	0.05
Uncertainty in half-life	negligible	<0.05

* CP - Commercial Products, The Radiochemical Company, AECL

** WNRE - Whiteshell Nuclear Research Establishment, AECL



(a)



(b)

Fig. 13: Oscilloscope Trace Showing a 1024-channel Display and the Selection of the γ-channel Window for ^{60}Co

from dead time: the estimated systematic uncertainty is the difference between results for the measured dead time and for a dead time that is longer by one standard deviation in the mean of several measurements of the actual dead time. The statistical and systematic uncertainties were quoted separately for the activity concentration of the solutions submitted to the international reference systems. For other users (Table IV) an overall uncertainty of 0.5% was given.

8. STANDARDIZATION OF ^{57}Co

A stock of ^{57}Co was obtained from a commercial supplier in 1979 and purified by the same method used for ^{60}Co. Impurities of 0.11% ^{56}Co and 0.03% ^{58}Co were found by γ-ray spectrometry on the reference date for the ^{57}Co coincidence measurements. Similarly to ^{60}Co, a master solution, two dilutions, and sources were prepared and distributed (Table V).

The γ-channel windows were set from approximately 60 to 160 keV, and included the 122- and 136- keV photo peaks. Methane and a mixture of 10% methane in argon (P-10) were used in separate runs as counting gases for the 4π(PC). The 4π(PC) efficiency was enhanced by approximately 15% with P-10 gas, which stops a larger fraction of the 6-keV X rays.

Altogether data were recorded from ten runs in which each of seven sources was counted. The range of $\mathcal{E}_{4\pi}$ was from 0.39 to 0.79, and of activity from 15 to 30 kBq (0.4 to 0.8 μCi). The standard deviation in the final result was ±0.03%. Again, the results from the two dilutions and the two independent sets of electronics were consistent, which suggests that instrumental errors were small. Our estimates of sources of systematic uncertainty are given in Table IV, and again an overall uncertainty of 0.5% was stated for locally issued standards.

Table V

Summary of the Distribution of Samples of ^{57}Co

Type of Sample	Relevant Solution	Use or User
Ampoules	master	BIPM
Ampoules	master, dilution no.2	$4\pi\gamma$ IC calibration
Ampoules	dilution no.2	CP, Environmental Research Branch of CRNL, WNRE
Sources	master	Ge(Li) calibration, CP, 2 branches at CRNL
Sources	dilutions nos. 1 & 2	4π(PC)-γ coincidence, Ge(Li) calibration, 9 branches at CRNL, WNRE

9. *CONCLUSION*

The registration of these standards of ^{60}Co and ^{57}Co in the international reference system indicates that they compare favourably with others (30). Similarly the results of an international intercomparison for measurement of the activity of ^{134}Cs (13, 31) demonstrate that a satisfactory level of competence has been obtained with the 4π(PC)-γ coincidence method. We find that an improvement of a factor of $\approx$5 in accuracy has been obtained during the last decade. However, further improvements will be necessary in order to achieve high accuracy in the metrology of nuclides with more complex decay schemes.

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