INSTRUMENTATION FOR LIQUID SCINTILLATION COUNTING AT LOS ALAMOS

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INTRODUCTION

The instrumentation problems involved in liquid scintillation counting are not particularly difficult and a good coincidence system can be designed by employing basic electronic pulse handling techniques. This paper will discuss a little of the history of the Los Alamos counting systems, with emphasis on the reasoning behind transitional developments since the 'early days'. Some of the operational features and performance characteristics of the Los Alamos counters will be given and, finally, a few ideas for the future counting refinements will be proposed.

BASIC SYSTEM

Figure 1 is a block diagram of the Los Alamos counting apparatus. These systems can be set up to count only one isotope energy interval at a time.

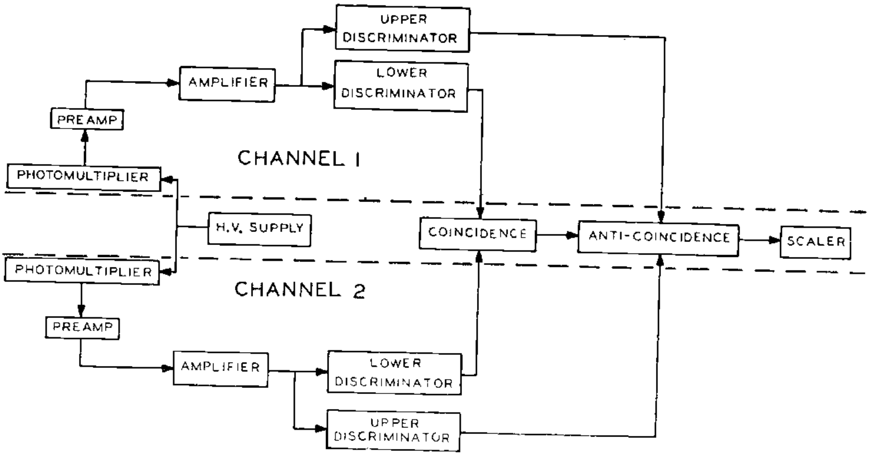

Fig. 1. Block diagram of Los Alamos counting apparatus.

Photomultiplier pulses are amplified and passed to pulse height discriminators for amplitude bounds selection. The normalized duration of the pulses generated in the lower discriminators fixes the system resolving time, since these pulses pass directly to the coincidence circuit. The output of the coincidence circuit goes to anticoincidence and, if there is no inhibiting pulse from either of the upper discriminators, passes out to be counted by the scaler.

The adequacy of such a system for liquid scintillation counting depends to a large extent upon the time resolution which can be obtained, since the time element determines the number of undesired 'chance' counts which will be recorded due to uncorrelated noise events from the photomultipliers and electronic system. The Los Alamos counters have resolving times of 0.08–0.1 μsec which is a speed compatible with all the system elements, and sufficiently fast to give a low background from 'chance' count sources.

Another workable configuration of the blocks in Fig. 1 would have the upper and lower discriminators in each channel feeding anticoincidence in each channel, thus giving pulse height selection prior to coincidence for time selection. This configuration was decided against, since it required an extra anticoincidence block, and required that the critical, fast, time determining pulses pass through the added circuitry before reaching coincidence.

CIRCUIT DETAILS

Photomultipliers and high voltage supply

The first Los Alamos counters used RCA type 5819 photomultiplier tubes and these were later replaced by DuMont type 6292 tubes because of their slightly lower noise emission. The most recent system uses DuMont type 6467 tubes, since they have a cathode efficiency equal to that of the 6292, with only 40% of the cathode area. This system operates at room temperature and is adequate for counting tritium at low efficiencies.

The access port to the counting sample well between photomultipliers has a shutter system which allows high voltage to remain on the photomultipliers at all times. The sample well is large enough to accommodate the Kimble Opticlear 10 dram vial and the reflecting surfaces are coated with white Tygon paint. This easily applicable TiO_2-containing reflector requires the use of a long wavelength emitting scintillation solute such as POPOP.

The high voltage supply for the photomultipliers must be stable and free from noise. Positive polarity is used to allow grounding of the photocathodes. Several Los Alamos models and commercially available high voltage supplies have been used with the Los Alamos counters.

Amplifiers

A complete description of the first Los Alamos liquid scintillation counter can be found in the literature.[1] At the time that the system was designed it was considered necessary that amplifier rise time be faster than the coincidence resolving time so that coincident pulses of large amplitude difference in the two channels would still be counted. The amplifier used in that system was of a conventional three-tube loop design, with wideband response giving a rise time of about 0.08 μsec. Its resulting characteristics were not ideal with regard to background stability since the optimum system operating point called for a counting threshold very near the level of the amplifier noise pulses. Further, the amplifier had very poor overload properties for the occasional large scintillation pulses, and it consumed a great deal of power.

In order to correct these difficulties it was decided that a different design would be used, with a sacrifice in bandwidth and rise time in order to achieve a lower amplifier noise level. Also, this slower amplifier would have more graceful overload properties and be more efficient with regard to power. The amplifier design was patterned after that of HIGINBOTHAM,[2] using cathode coupled amplifier loops and double differentiation.

Since it was desired that both amplifiers for a system be built on one chassis and fed from one standard power supply, power consumption had to be considered. On the basis of heater power requirements, and transconductance at low plate currents, type 6BQ7A tubes are used in the cathode coupled amplifier loops. The output stage produces 50 V pulses linearly. Preamplifiers used are modified versions of the stock Los Alamos model used with other overloadable type amplifiers. They have gain selection of unity or ten, and the main amplifiers have a maximum gain of about 4000, with overlapping gain controls to give a minimum gain of about 250. Amplifier rise time is 0.15–0.2 μsec,which is compatible with the coincidence circuit and method of operation to be described later. Line voltage stability of the amplifiers shows about 0.15% gain change per volt change in the line. The amplifiers have no detectable base line shift at rates of 10,000 pulses/sec of ten times overload amplitude.

Discriminators

The discriminators used in the original Los Alamos equipment[1] were of the Harwell type and they performed very well. However, they were inefficiently designed and used much power. A simpler, two-tube version of the lower discriminators is shown in Fig. 2. A 6BN6 gated-beam tube is used at the input, primarily for its higher limiter grid impedance when that grid is driven positive with respect to the cathode. Loading can not be tolerated at this grid, since the input signal must simultaneously be fed to the upper discriminator for possible action there. The negative anode output of the 6BN6 feeds the zero-biased pentode section of a 6U8. The plate of the pentode feeds the grid of the 6U8 triode section, the plate of which ties back to the grid of the pentode. Thus there is rapid regenerative action upon triggering, and the shorted delay line stub in the pentode plate fixes the duration and amplitude of the circuit output pulse. Since the 6U8 triode section is normally biased off, no pulse appears when the 6BN6 input pulse recedes and the delay line recharges. The amount of this bias on the triode determines how 'trigger happy' the discriminator will be. This bias should be large enough to keep the positive feedback loop gain less than unity in the absence of triggering, but small enough to let the triode close the regenerative path at the earliest point on the signal from the pentode.

This discriminator produces a nominal 0.1 μsec positive pulse to be fed to coincidence. The actual pulse shape is 0.03 μsec rise, quite flat for 0.06 μsec, and 0.04 μsec fall. Amplitude is 5–6 V. The circuit requires about 18 mA of current at 300 V, and 1 mA from a negative voltage source for input and loop biasing. The threshold is accurate at levels as low as 1 V. Recovery time of the discriminator is about 0.4 μsec.

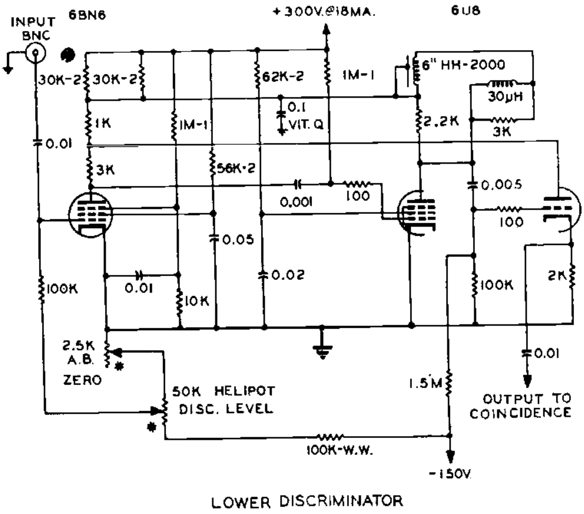

Fig. 2. Lower discriminator.

Coincidence and anticoincidence

The coincidence detector shown in Fig. 3 is very similar to the lower level discriminators. Here the shaped outputs from the lower discriminators are fed to the negatively biased limiter and quadrature grids of the 6BN6. When both grids are pulsed coincidentally plate current flows and the 6U8 circuit is triggered. The time of regeneration is fixed by the grid *RC* time constant of the 6U8 pentode and it is adjusted to deliver a 1 μsec pulse to anticoincidence.

The two control grids of the 6BN6 are switched to positively bias one or the other for single channel counting. Also they have independent negative bias controls which allow some freedom in coincidence resolving time adjustment. Since the input pulses are somewhat trapezoidal, the triggering threshold can be set to give resolving times from 0.05 μsec to about 0.12 μsec. Very little time overlap is required on the grids before triggering will occur, since the response of the coincidence circuit is excellent for short pulses.

The upper level discriminators are identical to the coincidence detector except that the quadrature grid of the 6BN6 is positively biased, and the plate load of the 6U8 pentode is a little larger to give larger output pulses for the anticoincidence action. The upper level discriminator outputs feed through a mixer circuit into the anticoincidence circuit. This circuit is similar to that described by VAN RENNES,[3] has been used by numerous people elsewhere, and need not be described here. It works very well with pulses of sharp trailing edges such as those generated in these circuits. Pulses which survive anticoincidence are positive, 20–25 V amplitude for use by standard Los Alamos scalers. Recovery time of the circuit configuration of Fig. 3 is about 0.5 μsec after the trailing edge of the output pulse.

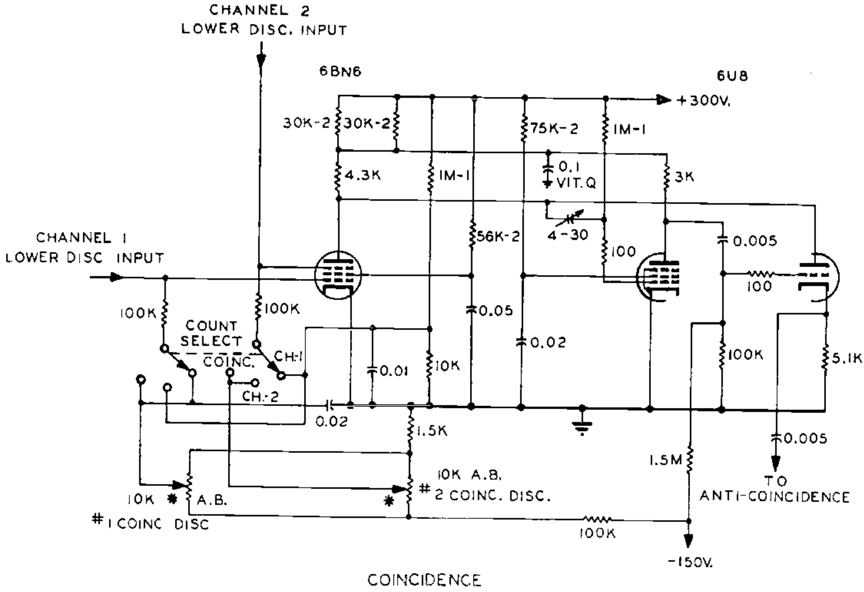

Fig. 3. Coincidence detector.

Scaler

The output pulses generated by the pulse selecting system described above can be counted by any of the standard Los Alamos scalers and the early models were used with the Model 750A scaler with 1 μsec resolving time and hexadecimal light interpolation. When the desirability of a scaler with preset count and preset time was expressed, the Model 770 counter was developed. This scaler uses two Burroughs beam switching tubes in the first fast decades, followed by three decades of Dekatron counters and a four-digit Sodeco register. Preset count is set up from the Dekatron outputs to give a selected total of from 100 counts to 90,000 counts. Preset time is furnished by a panel mounted mechanical Cramer timer. Two pulse resolving time of these scalers is about 0.5 μsec.

The most recent type scaler to be used with the liquid scintillation counting apparatus retreats from the option of preset count control, but uses a quality Liebel-Flarsheim X-ray timer for preset time counting. It is somewhat novel in its simplicity ; only nine tubes are used in the active counting circuit, which includes an input pulse height discriminator, three scaling decades, and an electrically reset five-digit Sodeco register. Figure 4 shows the circuit for this scaler. A standard Schmitt trigger discriminator is used, and all three beam switching tubes use identical circuitry. This means the last two decades are much faster than necessary, but at best any circuitry compatible with required speed would be very little simpler, and it seemed advantageous to keep all decades alike for construction, checking, and maintenance reasons. Two pulse resolving time of the counter is 1 μsec, maximum counting rate is 35,000 pulses/sec, and total line power consumption is a rather modest 75W. The somewhat elaborate power supply regulation furnished is necessary only

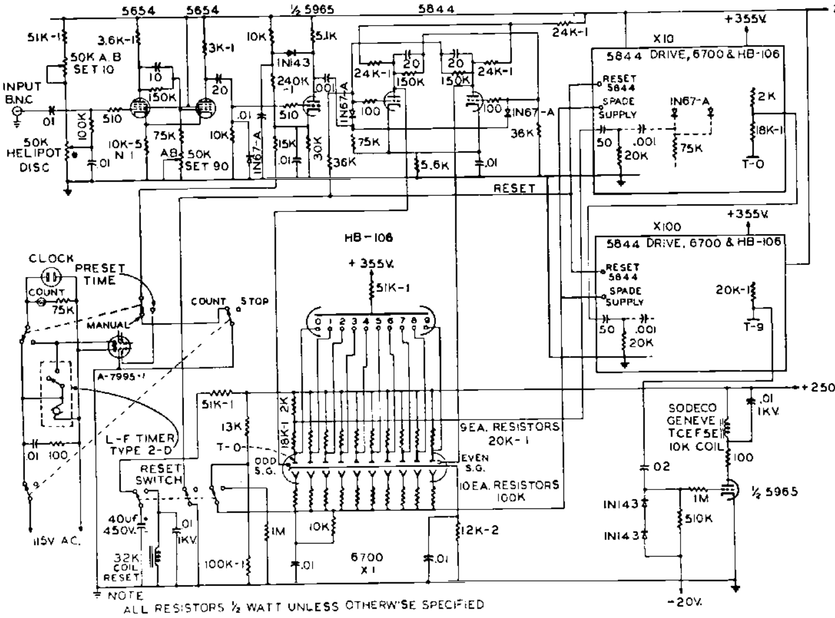

Fig. 4. Circuit of scaler using a quality Liebel-Flarsheim X-ray timer for preset time counting.

for discriminator voltage stability reasons when it is desired to use the unit for integral bias curves. The *B* plus for the remainder of the circuit can change by 20% without affecting operating characteristics. Count readout is on Nixie numerical indicator tubes for a clear, error-free display. The life expectancy of these indicators is still doubtful, and they are probably the weak point of the counter. All other parts of the circuit are very conservatively designed, and computer type tubes are used throughout.

PERFORMANCE CHARACTERISTICS

The question of absolute compatibility between amplifier rise time and coincidence resolving time was checked, and the system which has been described here meets the requirements of this Laboratory. Much of the good performance of such a system can be attributed to the user's operating techniques and to the optimum optics at the photomultipliers. An effort is made to keep the lower discriminator threshold settings about equal in both channels, with near-balanced operating points on both sides. Good optics assures that gross differences in coincident pulse amplitudes will be minimized in the two channels. If the coincidence detector thresholds are set to trigger at the very top of the trapezoidal pulses from the discriminators, i.e. a resolving time of about 0.05 μsec, the counting efficiency for tritium is only 20% poorer than that achieved with a long resolving time, about equal to the rise time of the amplifiers. In actual practice the coincidence detector thresholds are set to trigger about half way down on the discriminator pulses, giving a resolving time of 0.08–0.10 μsec.

Six coincidence counters have been built in the electronics group for use in the biomedical research group. Four of these are now in operation and the other two have been retired. A variety of dual isotope counting has been carried out, some samples of which are : C^{14} and H^3 in lipids, H^3 and Na^{22} in intestinal water, and Ca^{45} and Sr^{90} in bone. Counting time has never been at so much of a premium as to suggest construction of an instrument to indicate coincidence rates in two energy channels at the same time. Thought which has been put into the design of such a unit has rejected the commercially popular three-discriminator analysis system for the basic reason that the instantaneous dynamic range explorable by the discriminator system is considerably less than that usually needed for good dual isotope counting conditions. In the existing Los Alamos system the discriminators are allowed to remain at a fixed optimum slit ratio while switching is performed in either the high voltage supply or the amplifiers, the latter being preferable in principle because of superior tracking.

The stability of the Los Alamos counter has been checked against the criterion of counting statistics in the course of a routine research problem, which required accumulated counting times of about 6 hr for each sample.

Fluctuations in repeated counts were found to be within the statistical prediction for total counts with standard deviations as small as 0.2%.

Figure 5 is a photograph of a complete liquid scintillation counting apparatus. Switches and connectors are provided on the discriminator chassis to furnish monitoring outlets from the amplifier or pulser inputs to the discriminators for alignment and checking. The arrangement of chassis in the short ventilated rack allows operator control from a seated position.

FUTURE TRENDS

It would appear at the present time that the type of electronic circuitry described thus far in this paper has reached a state of equilibrium. However, the state of the art in photomultiplier development is bringing continuous improvements in tubes, and these improved devices may be applicable to liquid scintillation counting.

The new, multi-alkali photocathodes[4] are more efficient and have the same order of thermionic emission as the Cs–Sb cathodes. The RCA developmental tube type C7237 with multi-alkali cathode has a cathode luminous sensitivity of 150 μA/lm, two and a half times that of most Cs–Sb cathodes, but tentative data gives an equivalent anode dark-current input of 10^{-9} lm which is a little higher than the figure given for recent tube types with conventional cathodes. This tube type would probably give improved performance in our existing circuitry, but its rather low amplification figure would not lead to a radically different type counter.

Tentative specifications for the RCA developmental type C7232A multiplier phototube are so interesting that mention of its possibilities should be made here. This tube is a sixteen-stage, head-on type with S-11 response and 60 μA/lm cathode sensitivity. It has fast response, very high current gain and a rather low equivalent anode dark-current input of 6×10^{-10} lm at a luminous sensitivity of 60,000 A/lm to the sixteenth dynode.

If one considers the proposal of using these new tubes to feed directly to the coincidence circuit, using a non-integrated last dynode output so that coincidence resolving time is limited by the speed of the liquid scintillator and transit time spread in the photomultiplier, one can calculate the following. Assuming an average of 2.9 e emitted from the photocathode per average tritium disintegration in the scintillator,[5] and a photomultiplier gain of 10^9 to the last dynode, then an output current pulse of 10 mμsec duration will have an amplitude of 46 mA. If one fed this current pulse through terminated 200Ω cable directly to coincidence, a 9.2 V positive pulse would result. This amplitude would probably be adequate for pulse height bounds selection, but an additional factor of two might be gained by passing the pulse through a suitable impedance transformer at the photomultiplier. Thus one has the interesting possibility of an efficient, low-level tritium counter without voltage amplifiers or photomultiplier refrigeration.

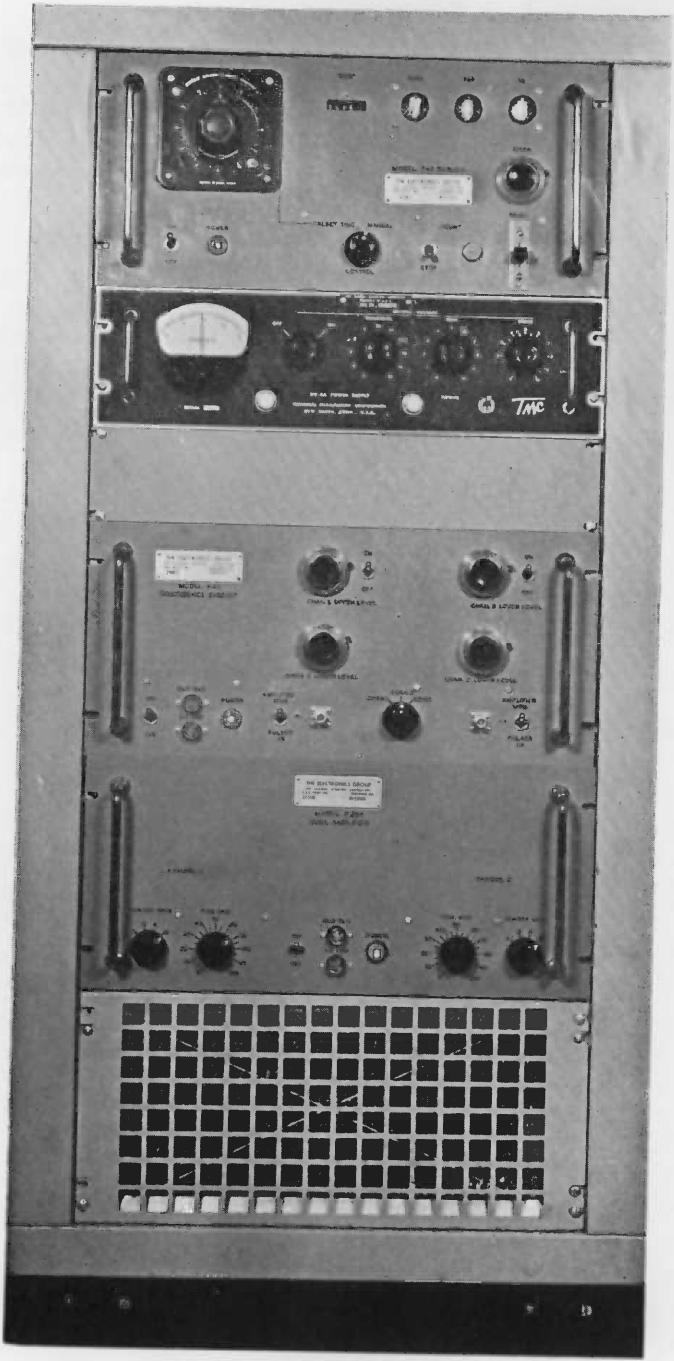

Fig. 5. Complete liquid scintillation counting apparatus.

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REFERENCES

[1] R. D. HIEBERT and R. J. WATTS. Fast coincidence circuit for H^3 and C^{14} measurements. *Nucleonics*, **11**, No. 12, 38–41 (1953).

[2] W. A. HIGINBOTHAM, Non-overload amplifiers. *Brookhaven National Laboratory Report* 234 (T–36), (1953).

[3] A. B. VAN RENNES. Pulse amplitude analysis in nuclear research, Pt. II. *Nucleonics*, **11**, No. 8, 26 (1952).

[4] A. H. SOMMER. Multi-alkali photocathodes. *IRE Trans. Nucl. Sci.*, **NS3**, No. 4, 8–12 (1956).

[5] F. N. HAYES and D. G. OTT. Liquid scintillators, IV. The small volume internal-sample counter. *Los Alamos Scientific Laboratory Report* LA–2095, 4.