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A NOBLE GAS SCINTILLATION COUNTER

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Submitted in partial fulfillment
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by

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Austin R. Frey
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ABSTRACT

A gas scintillation counter has been built to detect the scintillations caused by the passage of charged particles through the gas. The counter consists of a photomultiplier tube which looks into the scintillation chamber through a quartz window. The output of the photomultiplier is amplified and sent to a 12-channel pulse-height analyzer. The scintillation chamber contains the radioactive source and the noble gas for scintillation. The inside walls of the chamber are coated with a good light-scattering material. The outside of the chamber is surrounded by a heating element and is immersed in a dewar of liquid nitrogen. Associated control equipment permits the noble gas to be kept in any one of the three physical states.

Unfortunately pulse height is quenched, as a function of time, by the presence of contaminants in the noble gas. Water vapor is removed by the use of a cold trap. A dynamic purification system of finely divided metal was built to supply pure gas to the scintillation chamber. The effect of contamination is most evident when the noble gas scintillator is in the gaseous state. The effect is less noticeable in the liquid and solid states.

The scintillation counter was first used with argon, but it is felt that the instrument could be used with any of the noble gases. It is planned that future investigations will be made using xenon, krypton, and helium.

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INTRODUCTION

A gaseous scintillation counting system involves detecting the light emitted from gas molecules that are either ionized or excited by a charged particle passing through the gas. In 1953, Carl Muehlhause[1] made the first counter. He detected a light output from argon and from helium when these gases were excited by Pu alpha particles. Later in the same year, Wiegand and Segrè [2] increased the light output of an argon counter by applying an electric field. The apparatus was operated as a proportional counter, but in this case, light instead of electrons was used to measure the events. C. Eggler and C. M. Huddleston [3] built an argon counter which had a Pyrex window at one end through which a photomultiplier looked. A wave shifter (tetraphenylbutadiene) coated on the inside of the window converted the ultraviolet light into visible light. The wave shifter, however, introduced a contaminant in the system and progressively reduced pulse height with time. It was reported that a 15% resolution was obtained for 5.47-Mev Pu^{238} alpha particles. Pulse rise time was about 10^{-8} sec. The counter was notably insensitive to gamma rays.

J. A. Northrop and R. Nobles [4] investigated the use of xenon as a scintillator. They suggested ways of eliminating the quenching of pulse height caused by the outgassing of the wave shifter. The problem could be overcome by cooling the chamber, continuously purifying the gas, or interposing some quartz between the argon and wave shifter.

In building the counter, it was my purpose to devise means of eliminating the use of a wave shifter; to provide a means for purifying the gas; and to be able to investigate pulse heights while the scintillator is in the gaseous, liquid, or solid state. This report describes the means by which my purpose was accomplished.

I wish to express my thanks to Dr. Burton J. Moyer for his supervision and Dr. Roger Wallace for his guidance and criticism. In particular, I thank Theodore M. Jenkins, Technician, who worked constantly with me on the construction and experimental phases of the project.

This work was done in part under the auspices of the U.S. Atomic Energy Commission.

GENERAL ARRANGEMENT OF APPARATUS

Gaseous scintillation counting involves the use of a photomultiplier "looking" at the light that comes from an ionized or excited gas molecule. Ionization and excitation are caused by the passage of a charged particle. Figure 1 shows a general schematic arrangement of the equipment used in this experiment. Attached to this equipment are the associated electronic circuits, liquid argon control system, and the argon-purification system; these are described later.

Other workers [3, 4] have applied a wave shifter, such as tetraphenyl-butadiene, to the walls of the scintillation chamber and to the glass window. The wave shifter became a contaminant in the system and reduced the pulse heights with time. Attempts were made to remedy the situation by continually flowing purified gas into the chamber. In this work the need for a wave shifter or complicated gas-purification system was eliminated by using a quartz window on the scintillation chamber. The photomultiplier used was a special DuMont K1306, which has a quartz end window. Dow-Corning 200 fluid (viscosity 1000 centistokes) was used between the quartz chamber window and the phototube. This completed the optical path and provided good transmission of the ultraviolet scintillations of the argon gas.

The inside of the chamber was coated with aluminum oxide, which is a good light-scattering surface. The source was mounted in a stainless steel holder at the bottom of the chamber. The chamber also contained thermocouples and a liquid-level indicator, which are described later. A satisfactory high-vacuum seal for the chamber assembly was obtained by use of lead seals and special Radiation Laboratory vacuum-pipe fittings. Light seals were maintained by covering all joints with black photographic masking tape. The necessary thermal insulation was made of styrofoam, and completely surrounded the assembly.

The chamber was constructed to withstand 150 psi. This permits investigations to be made with the scintillator in the gaseous state at from 0 to 10 atmospheres pressure.

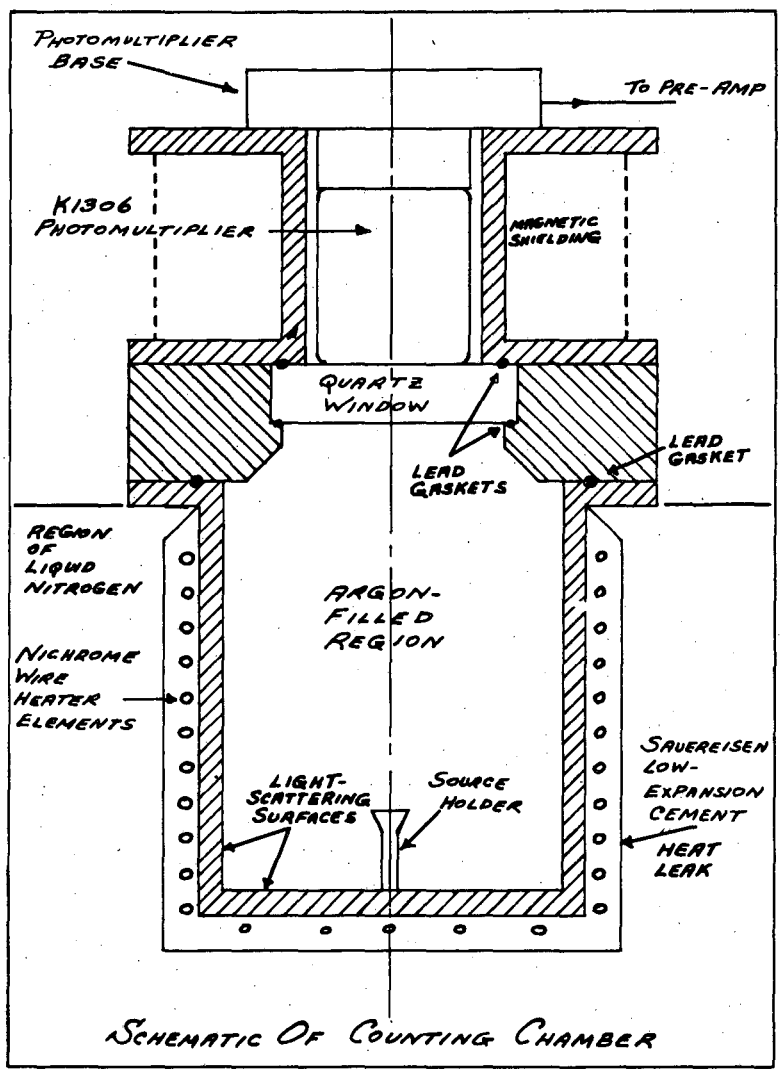

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Fig. 1. Schematic diagram of scintillation chamber showing heat leak made of Sauereisen low-expansion cement. Chamber is surrounded by a dewar of liquid nitrogen (not shown). Cooling for the photomultiplier is provided in the region of magnetic shielding. Thermocouples and liquid level indicator are shown in Fig. 6.

ELECTRONICS

The electronics used was that normally associated with scintillation counting. As shown in Fig. 2, the signal from the K1306 photomultiplier was sent to a low-noise preamplifier (Rad. Lab. Dwg. 1X4353C). From the preamplifier the signal went to a Model V Linear Amplifier (Rad. Lab. Dwg. 1X5334F). The signal was then sent to the 12-channel pulse-height analyzer after having been shaped properly by the timing-pulse generator and pulse shaper (Rad. Lab. Dwg. 1X4385). The circuit diagrams for the electronic units used are attached as an appendix to this report.

The noise level of the K1306 photomultiplier was reduced by cooling the tube and tube base with liquid nitrogen. The liquid nitrogen was introduced into the annular spaces of the magnetic shielding surrounding the tube. After about two hours' cooling the tube temperature became constant, as did the noise level. Once this condition was reached noise no longer presented a problem during the scintillation observations.

The noise level of the preamplifier (Rad. Lab. Dwg. 1X4353C) was also reduced by cooling the chassis with dry ice.

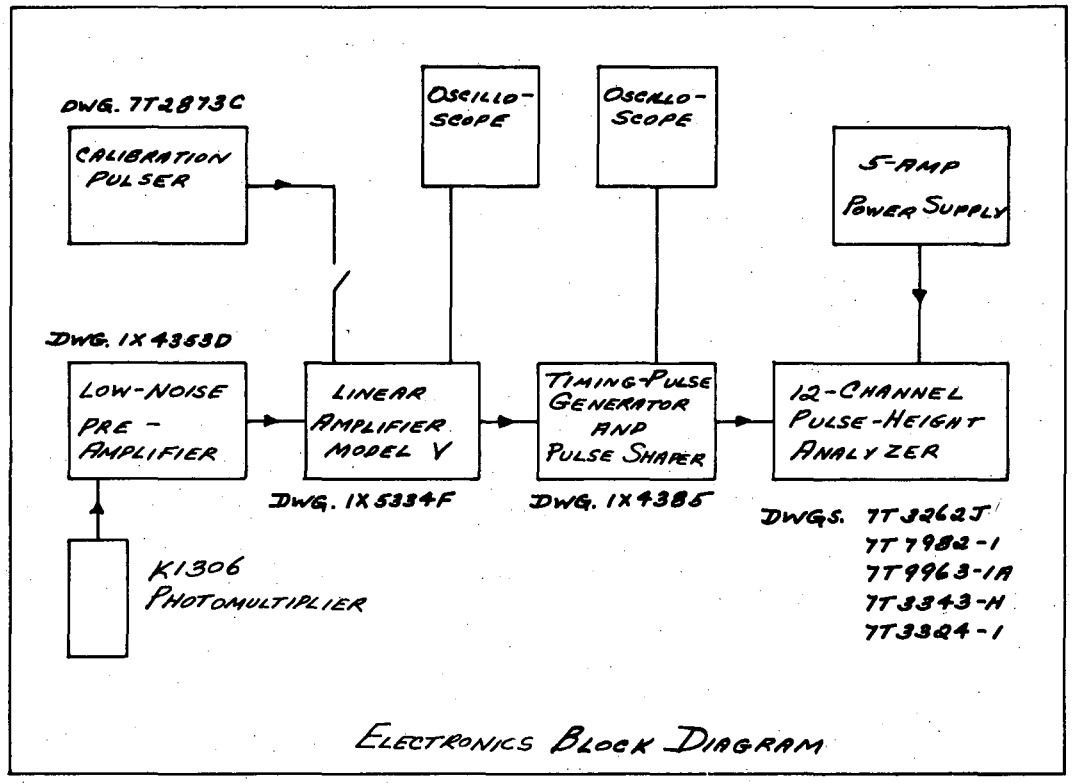

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Fig. 2. Block diagram of electronic components associated with noble-gas scintillator. Circuit diagrams of the individual components are in the appendix. K1306 photomultiplier is a special quartz-window tube which eliminated the use of a wave shifter.

ARGON-PURIFICATION SYSTEM

Because of the quenching effect on pulse height caused by contaminants in the argon, it was decided to build a purification system. A dynamic purification system was selected because it would provide a continuous supply of pure gas to the scintillation chamber and would also permit the storage of pure gas until it was needed. A flow diagram of the purification system is shown in Fig. 3. In order to insure that purification would take place it was necessary to control three physical conditions:

(a) particle size of metal to be used as purifying agent,

(b) temperature of the purifying agent,

(c) flow rate through the purifying agent by the impure feed gas.

D.S. Gibbs, H. J. Svec, and R.E. Harrington [5] recently conducted experiments in the purification of the rare gases. After referring to their results I decided to use small calcium metal turnings as the purifying agent. Calcium would effectively remove nitrogen and oxygen from the argon. Water vapor could be removed from the gas in a water trap cooled by dry ice. Mass-spectrometer analysis of purified argon in the system devised by D.S. Gibbs showed that the oxygen was completely removed and that only 0.07% nitrogen remained.

From the flow diagram it can be seen that the argon is first passed through a water trap. The trap is a copper container filled with glass wool to prevent moisture carry-over. The trap is cooled by crushed dry ice in the surrounding dewar.

The argon flow rate was controlled by the use of a Fischer and Porter tri-flat low-flow Flowrator. A flow of 830 cc/min was used, and resulted in purification of the gas as reported by D.S. Gibbs [5].

The purification tube was made of Vycor tubing. The tubing was surrounded with

(a) a sheet copper sleeve to prevent hot spots,

(b) a nichrome wire heating element,

(c) an iron-constantan thermocouple,

(d) sheet asbestos for insulation.

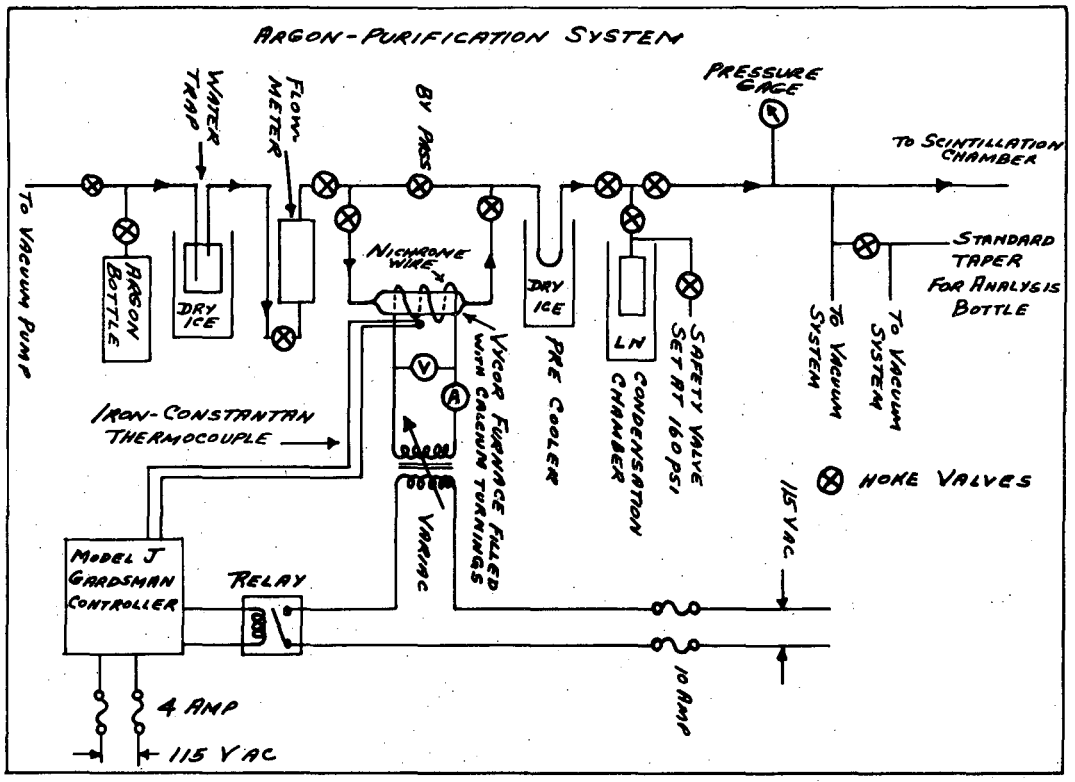

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Fig. 3. Flow diagram of the argon-purification system, showing Vycor furnace filled with calcium turnings. Gardsman controller maintained furnace at 650°C. Water trap removed moisture from unpurified gas. Flowmeter indicated flow rate of argon over the calcium purifying agent. Purified gas stored in condensation chamber until needed.

Inside the tube was the "charge" of calcium turnings. The calcium, which was more than 97% pure, was supplied by Nelco Metals, Connecticut. The calcium turnings were constrained in the Vycor purification tube by a wad of quartz wool at each end. The quartz wool was loosely packed and did not interfere with the proper flow rate of the argon. Each charge of calcium turnings can be used to purify approximately 1300 liters of argon before it all becomes calcium oxide or calcium nitride. The Vycor tube was maintained at 650°C by a Gardsman Model J Controller.

The purified argon is then passed through a copper-coil precooler. The precooler coils are surrounded by crushed dry ice. The precooler was put in the system to lower the temperature of the argon before it entered the condensation chamber. This reduced the amount of liquid nitrogen required to condense the argon. The condensation chamber is a copper vessel fitted with suitable valves so that purified argon from it can be sent to the scintillation chamber as needed. Keeping the argon condensed materially reduced the size of storage chamber needed as the argon is purified.

Provision is also made for putting a standard glass gas-analysis bottle in the system. With this arrangement it is possible to check the purity of the argon without stopping the purification system. The entire system is connected to a vacuum system consisting of a forepump, diffusion pump, and associated equipment.

THE LIQUID ARGON SYSTEM

At normal atmospheric pressure, argon melts at -189.2°C and vaporizes at -185.7°C. The scintillation chamber was equipped with a system that would keep the argon within the 3.5° range of the liquid state. (See Figure 4). The entire chamber was immersed in liquid nitrogen, which boils at-195.5°C. A nichrome wire heating element embedded in Sauereisen low-expansion cement was placed around the outside of the chamber. The Sauereisen cement formed a heat leak so that when the heating element was on the scintillation chamber would warm up and a minimum of liquid nitrogen would be boiled away. Control of the heating element was maintained with a Speedomax microvolt recorder and an associated copper-constantan thermocouple. The reference junction of the thermocouple was placed in liquid nitrogen.

Placing the reference junction in liquid nitrogen instead of in ice water made the system more sensitive to small temperature changes. With this system, the minute voltage change caused by a change in the temperature of the argon was a larger percentage of the total voltage difference than it would have been if the reference junction were at 0°C. The plot of voltage difference vs time as shown on the Speedomax recorder indicated that the combination of a liquid nitrogen temperature-reference junction and a Sauereisen heat leak controlled the liquid argon temperature very smoothly. The other junction was inside the scintillation chamber, at the bottom, so that it became immersed in liquid as soon as the argon began to condense. Because of the small voltage difference being measured, the junctions were fused and Belden 8422 shielded cable was used for the lead-in wires.

At liquid argon temperatures the voltage difference between junctions was on the order of 135 microvolts. Because the maximum range of the recorder was 100 microvolts, the voltage difference was sent through an attenuator before going to the amplifier of the recorder. The setting of the recorder was determined by using the graph on Fig. 5. The setting was the difference in microvolts between the liquid nitrogen

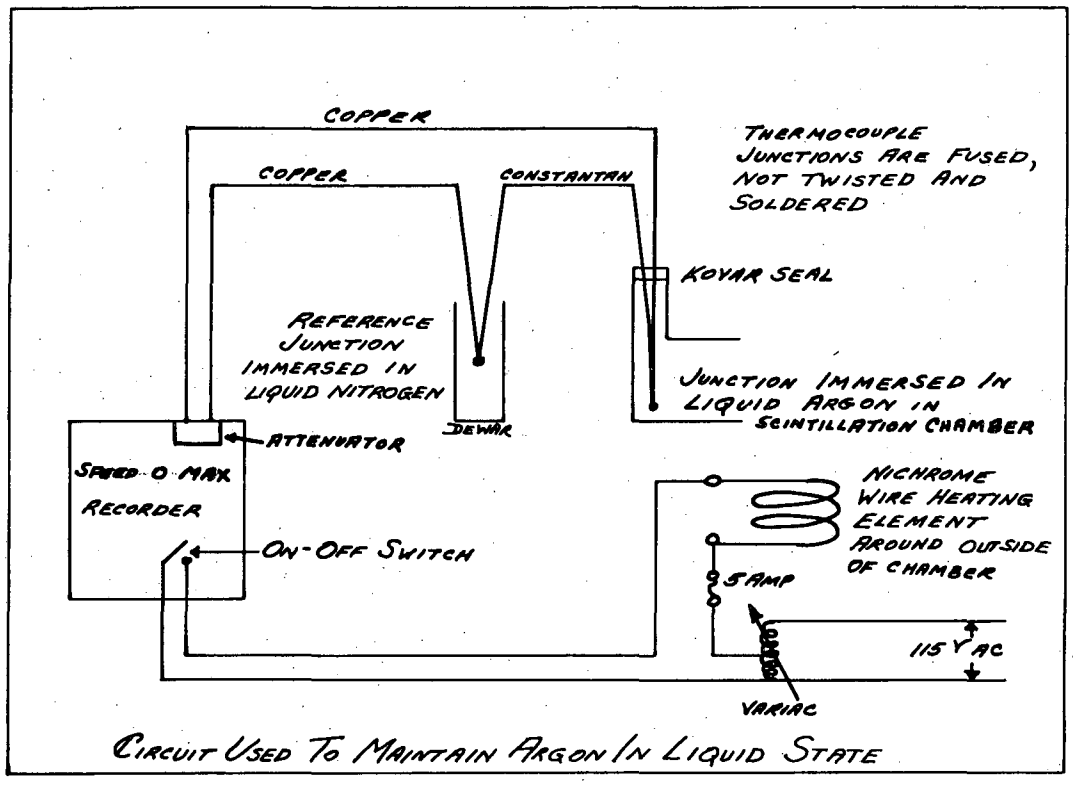

Fig. 4. Schematic diagram of system used to maintain argon in liquid state. Difference voltage from copper-constantan thermocouple actuates on-off switch of Speedomax recorder. Nichrome wire heating element is embedded in Sauereisen heat leak (not shown).

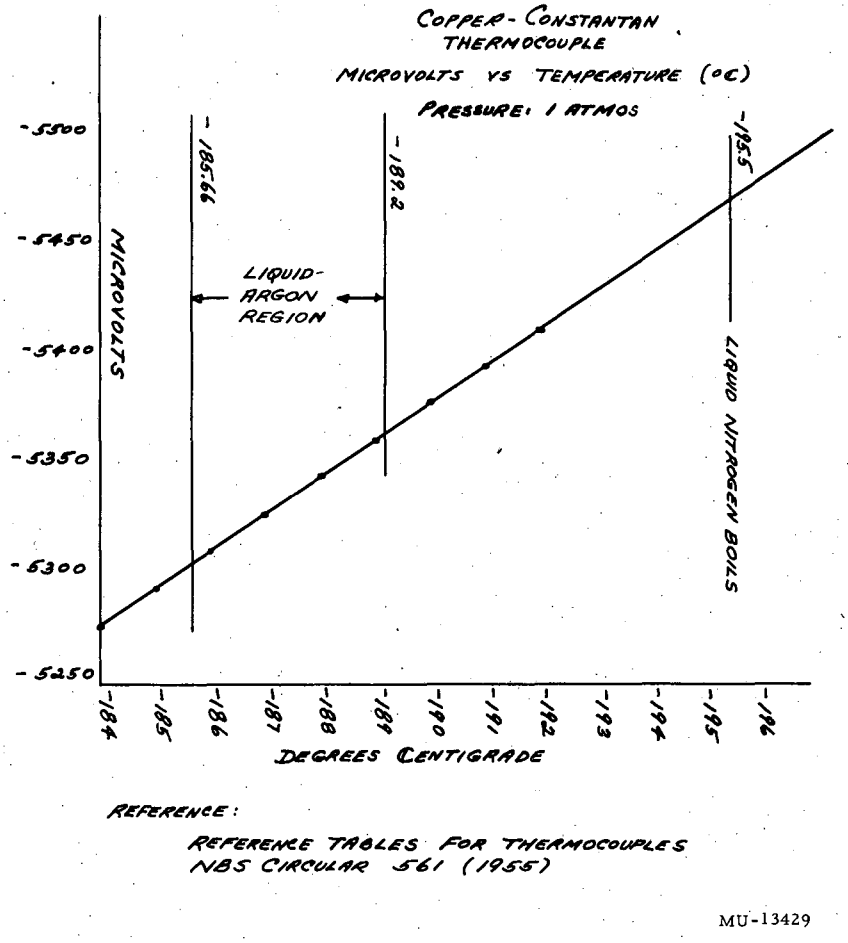

Fig. 5. Plot of microvolts vs degrees centigrade. Used to determine Speedomax setting for on-off switch in heating circuit of the liquid argon system.

temperature and a temperature in the middle of the liquid argon region. The heater switch remained open when the argon in the chamber was still in the gaseous state. The heater was able to keep the argon a liquid regardless of changes in the flow rate of argon into the chamber.

The level of the liquid argon was shown on the milliammeter of the liquid-level indicator. The schematic details of this system are shown in Figure 6. The liquid-level indicator was built by John J. Barale of the Radiation Laboratory and adapted to this system by the author.

(Other features of the system are shown in the figures that constitute the Appendix.)

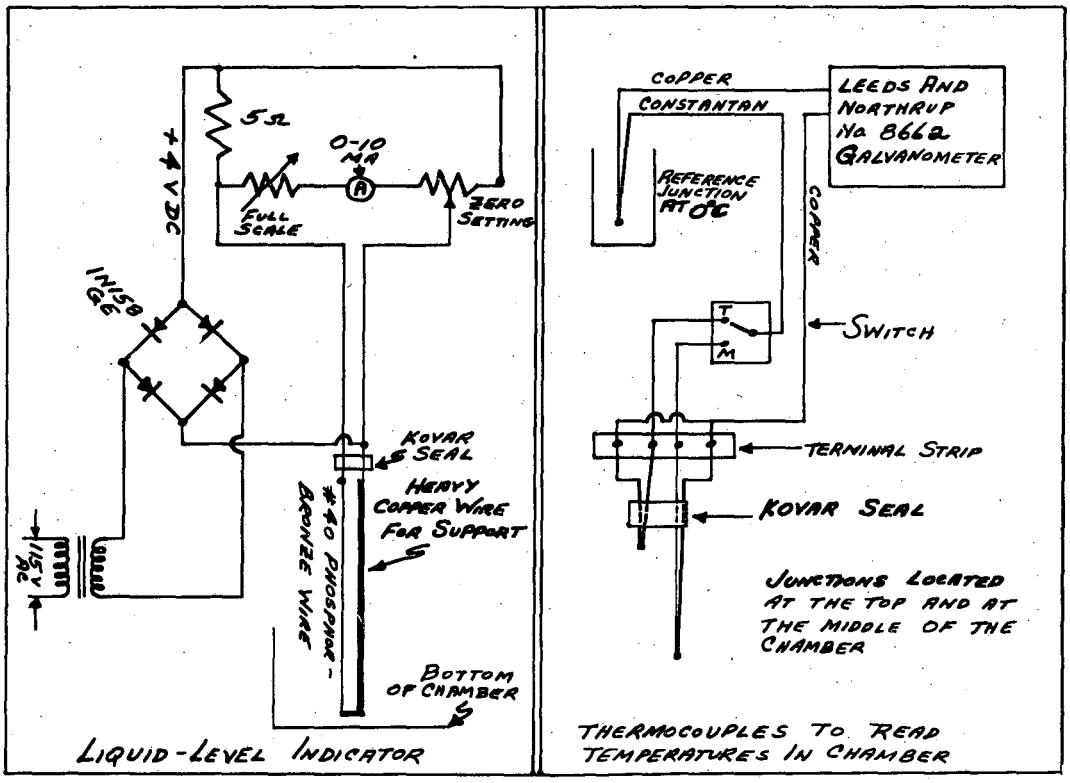

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Fig. 6. Schematic diagrams of liquid-level indicator and thermocouples in scintillation chamber. The 0-10 milliammeter in the level indicator was calibrated to read in inches of liquid argon.

CONDITIONS INSIDE THE SCINTILLATION CHAMBER

The inside of the scintillation chamber was coated with aluminum oxide to provide better light scattering. The aluminum oxide was applied to the chamber walls with sodium silicate. The sodium silicate was necessary to keep the aluminum oxide from being washed off by liquid argon.

The pressure inside the chamber was indicated by two means. First a pressure gage with a range from 30 inches vacuum to 300 psi was used. A vacuum thermocouple was used to indicate pressure conditions in the system before the argon was admitted to the chamber.

Because of the quenching effect of contaminants, the system was pumped down to a vacuum and refilled with new gas used whenever the setup was being changed for investigating scintillations at a different pressure.

Thermocouples were used to measure temperature conditions at the top and at the middle of the chamber. Figure 6 shows the wiring of this system. Figure 6 also shows the level indicator used to measure the liquid level of the argon. The indicator actually measured current flow through a fine (3-mil) phosphorbronze wire whose resistance changes as the liquid level rises in the chamber.

INTENDED USES OF THE SCINTILLATOR

The gas scintillator has been built in order that the problems indicated as follows may be investigated.

1. J.A. Northrop [4] reported a linear relationship between energy release in the scintillating gas or liquid and pulse height. When the plot of particle energy for particles stopping in the gas vs pulse height is extrapolated to the abscissa, the intercept is at 0.5 Mev instead of 0.0 Mev as might be expected. The reason for this is not, as yet, understood.

2. The scintillation chamber permits investigations with the scintillator in any one of the three physical states. The relationship between pulse height and physical state is to be determined.

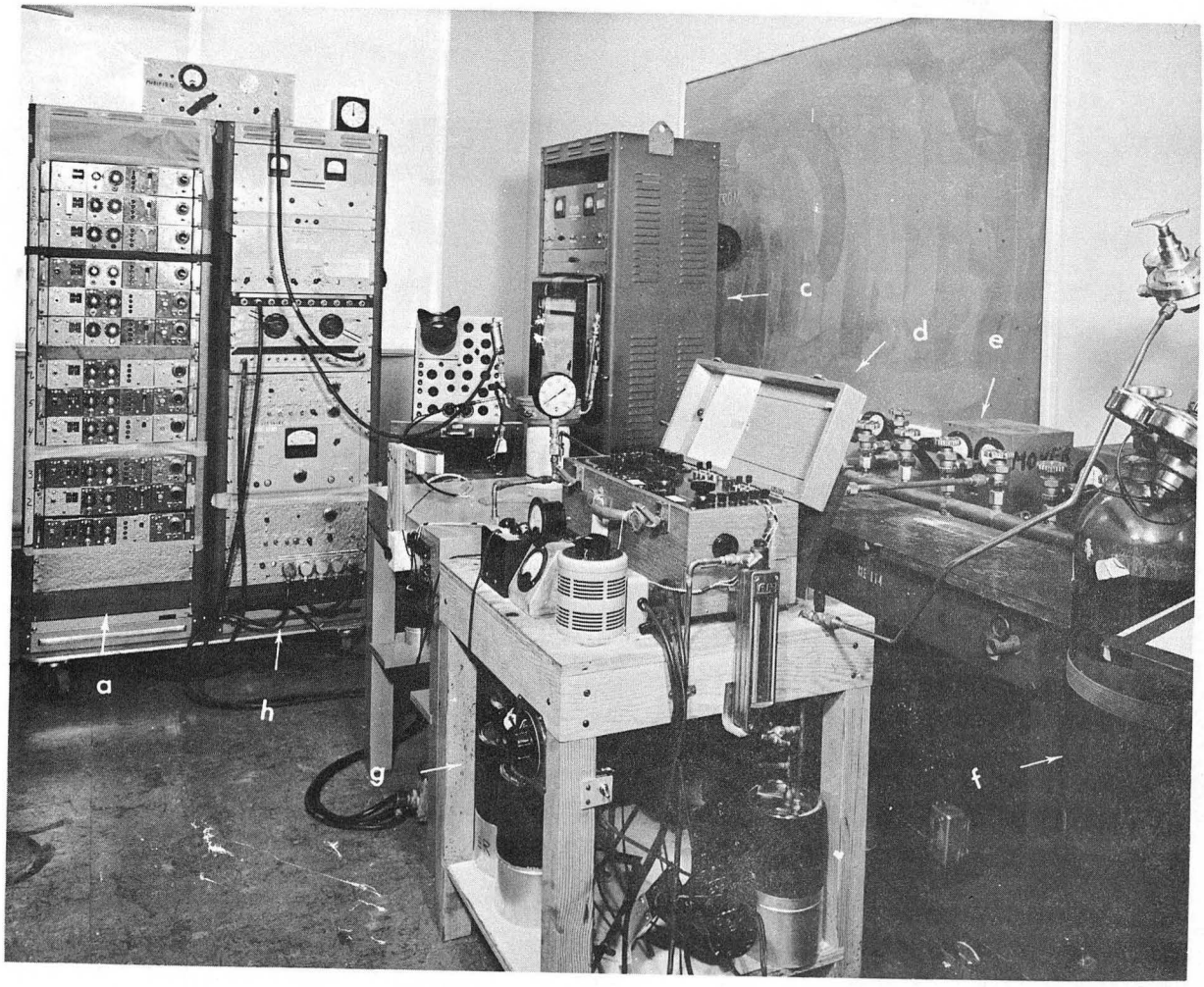
3. J.A. Northrop [4] reported that lower-Z noble gases give smaller pulse heights when the scintillator is in the gaseous state. The relationship between pulse height and Z for the liquid and solid states is to be investigated.

4. The effects of contaminants in the scintillating gas are to be determined. It is expected that contaminants will affect the gaseous state the most, the liquid state less, and the solid state the least. In the solid state the crystalline surface structure of the scintillator must be such as to pass the scintillations to the photomultiplier. The method for obtaining this surface is not clear as yet.

5. The best geometric shape for the scintillation chamber so that light is reflected to the photomultiplier efficiently is to be determined. The optimum scattering surface on the scintillation chamber is also of concern.

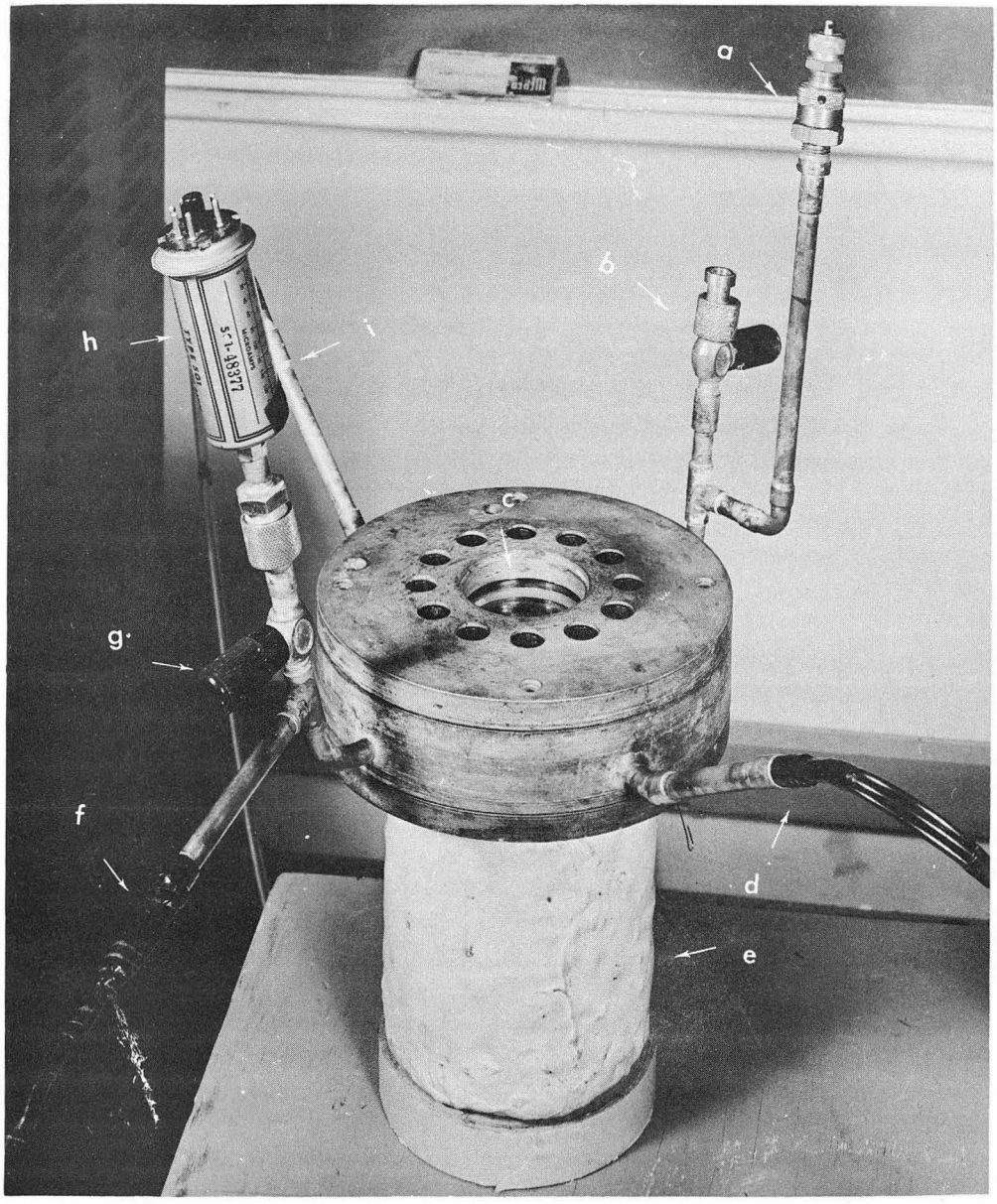
6. The best noble gas to use to produce usable pulse heights (keeping in mind cost and ease of handling) is to be determined.

Experiments are now in progress to collect the necessary data to help solve some of the above problems. When these data have been analyzed they will be published as a supplement to this report.



ZN-1712

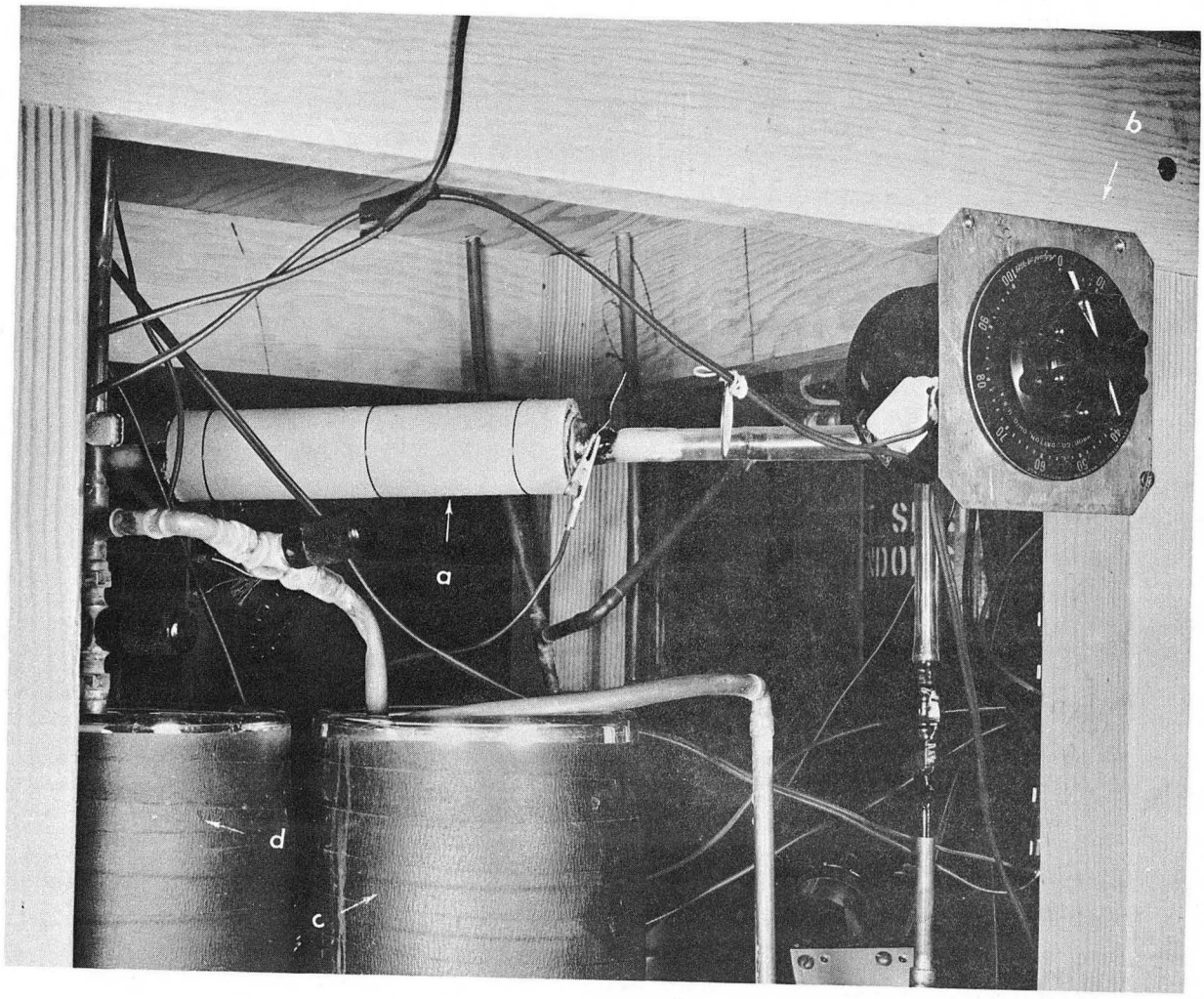
Fig. 7. General arrangement of equipment.
- a. Twelve-channel pulse-height analyzer
- b. Calibration pulser
- c. Speedomax recorder
- d. Leeds and Northrup Model 8662 galvanometer
- e. Vacuum-system manifold
- f. Argon supply bottles
- g. Argon-purification system
- h. Electronics stack: pulse shaper, linear amplifier, coaxial switch, power supplies



ZN-1711

Fig. 8. Exterior of scintillation chamber

a. Safety value set at 160 psi
b. Radiation Laboratory vacuum fitting and Hoke valve
c. Two-inch-diameter quartz window
d. Kovar seal with connections for top and middle thermocouples inside chamber
e. Sauereisen jacket surrounding nichrome heating element
f. Kovar seal with connections for thermocouple used to control heating element
g. Hoke valve
h. Thermocouple vacuum-gage connection
i. Kovar seal with connection for liquid-level indicator



ZN-1710

Fig. 9. Argon-purification system
a. Vycor purification tube showing asbestos jacket and electrical connections to nichrome heating element
b. Variac for heating element around scintillation chamber
c. Purified gas precooler. Dewar contains crushed dry ice
d. Condensation chamber, in dewar of liquid nitrogen

APPENDIX

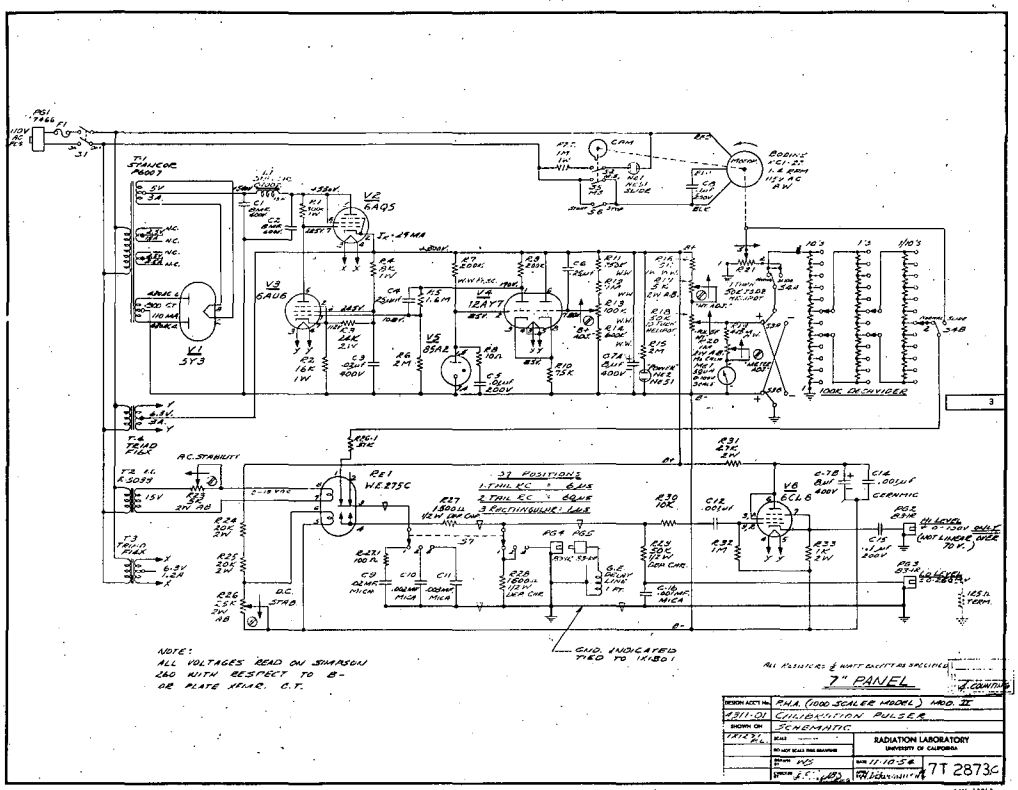


Fig. 10 Calibrating pulser for 1000-scaler Model II.

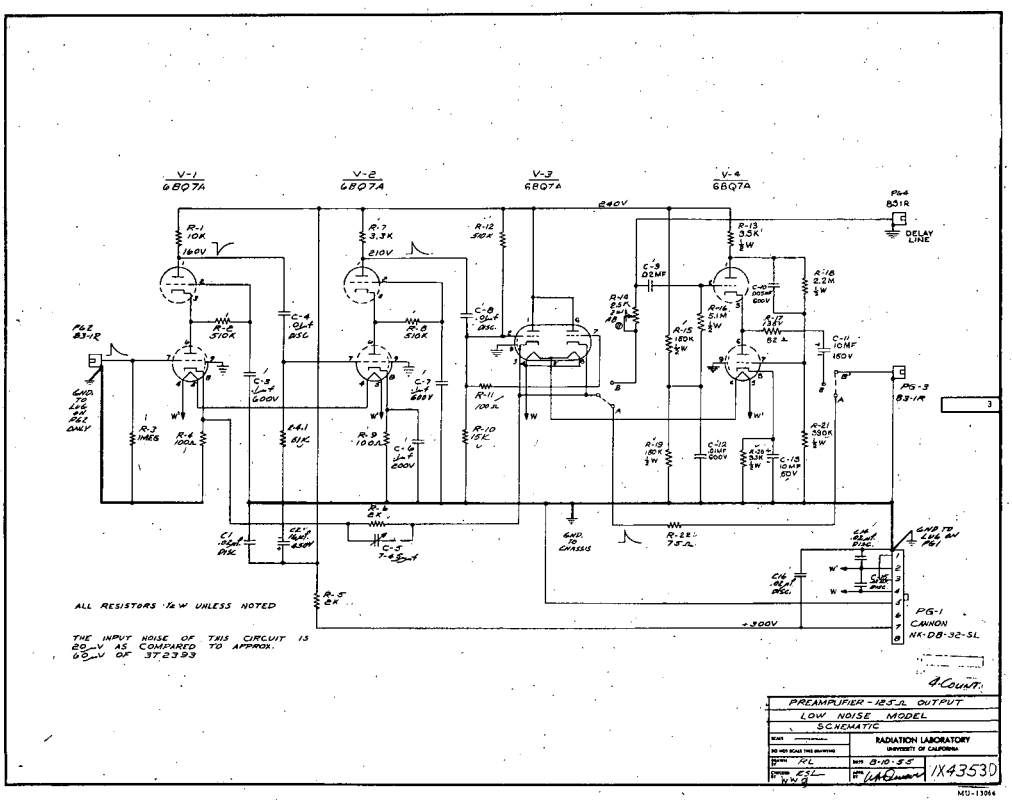


Fig. 11. Low-noise model preamplifier (125 Ω output).

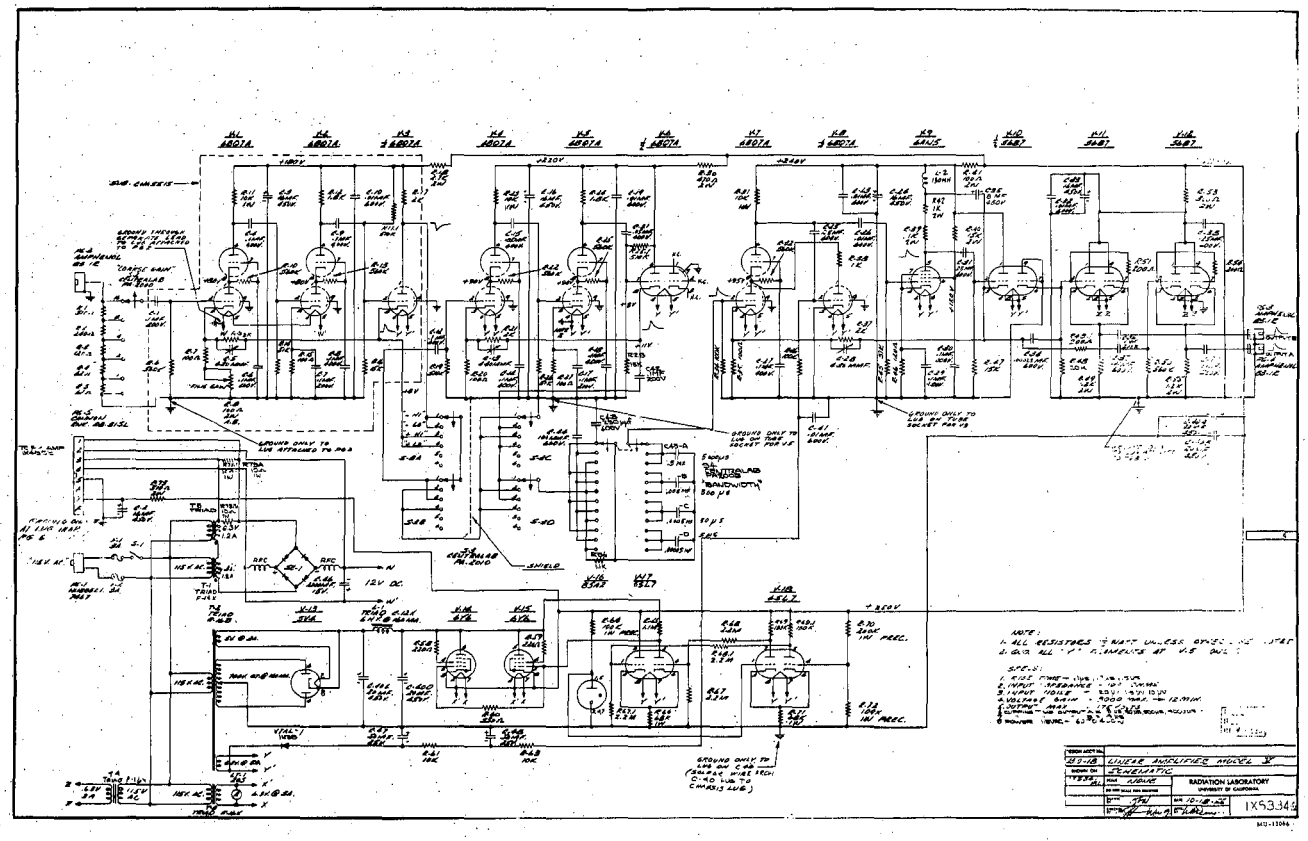


Fig. 12. Linear amplifier, Model V.

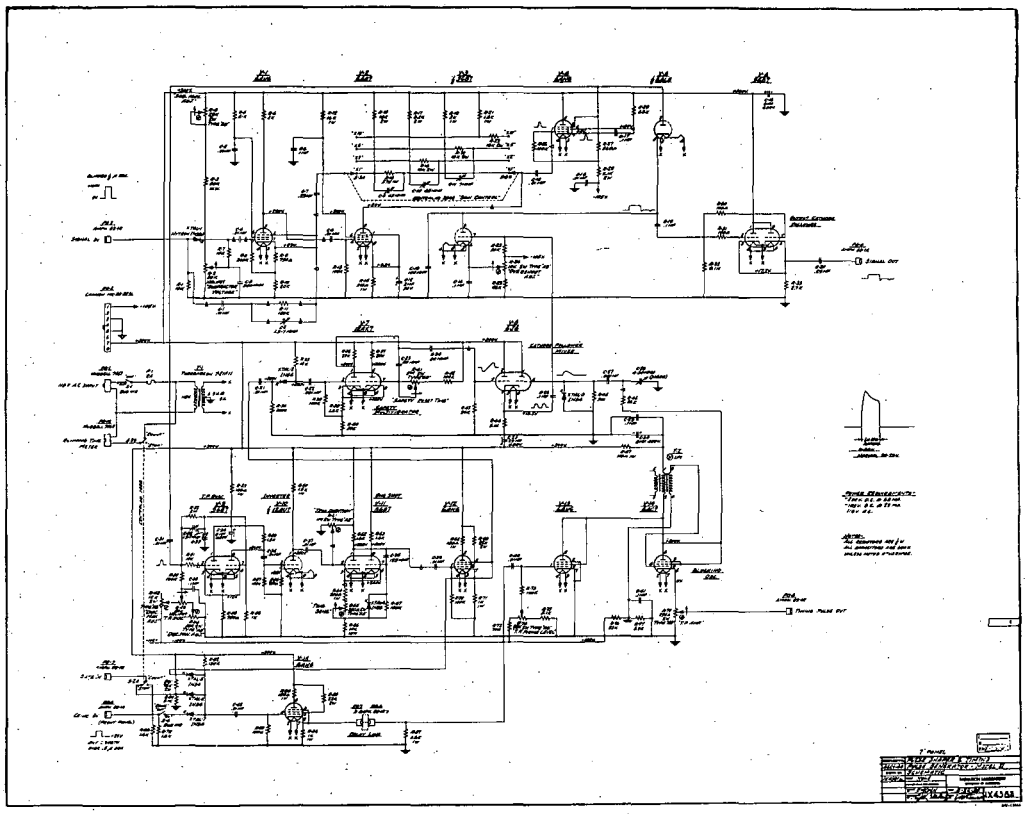


Fig. 13. Pulse shaper and timing-pulse generator, Model II.

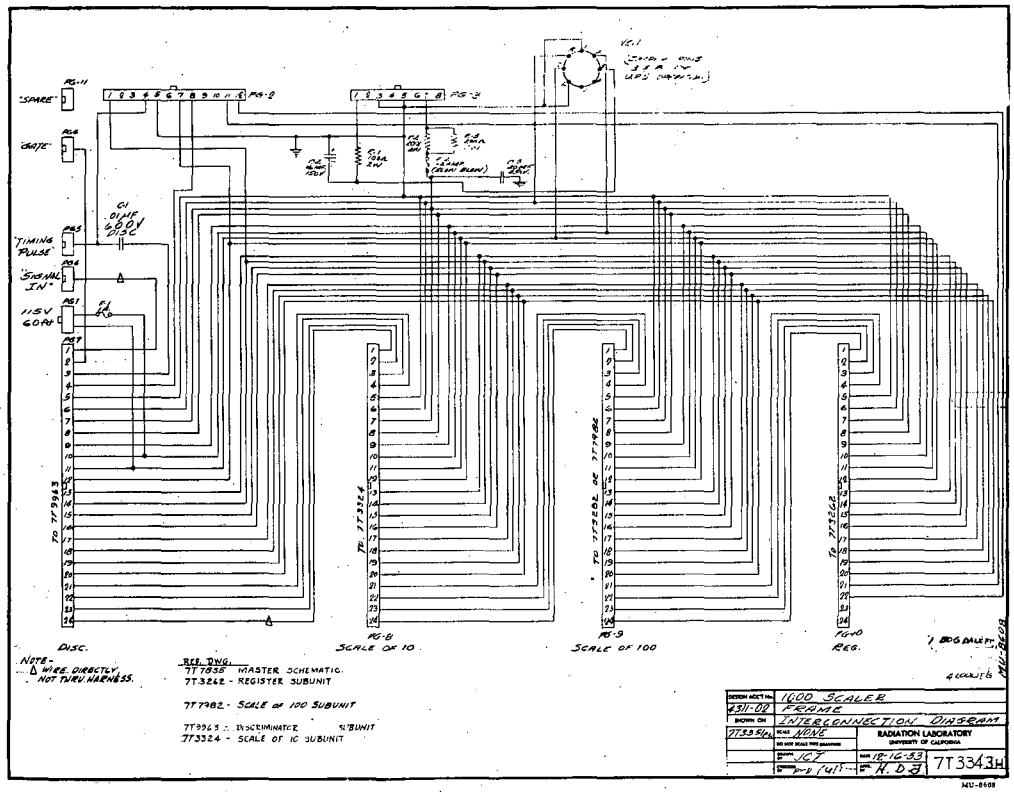


Fig. 14. Interconnection diagram for 1000 scaler.

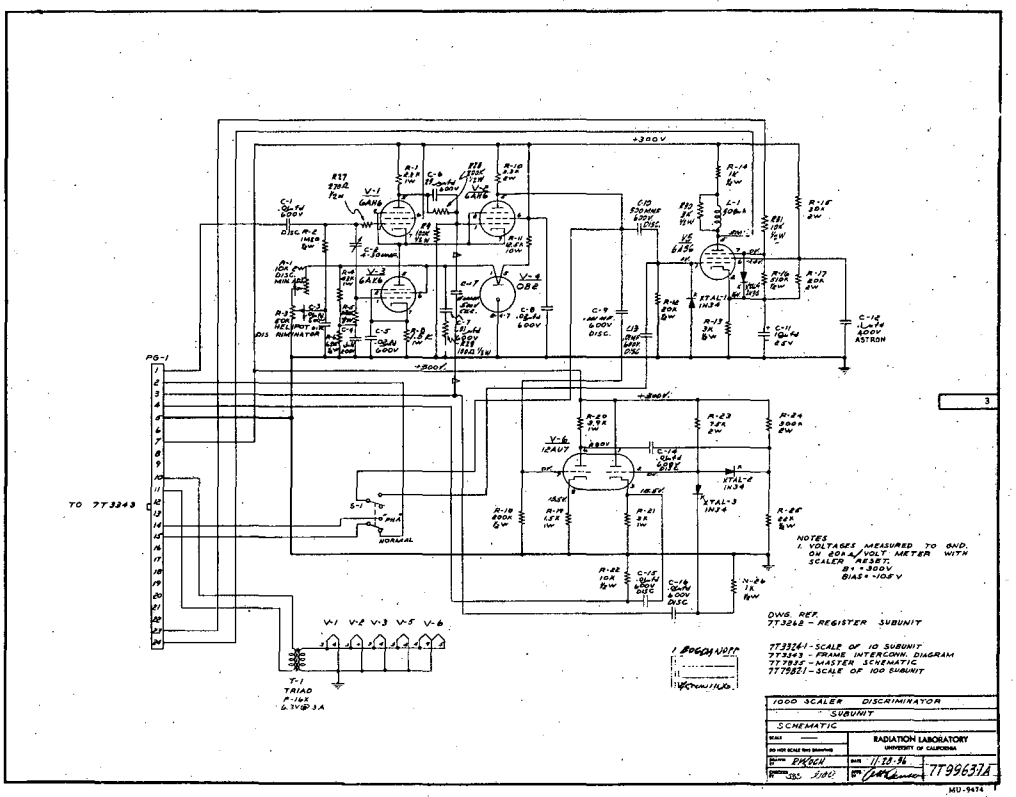


Fig. 15. Discriminator subunit of 1000 scaler.

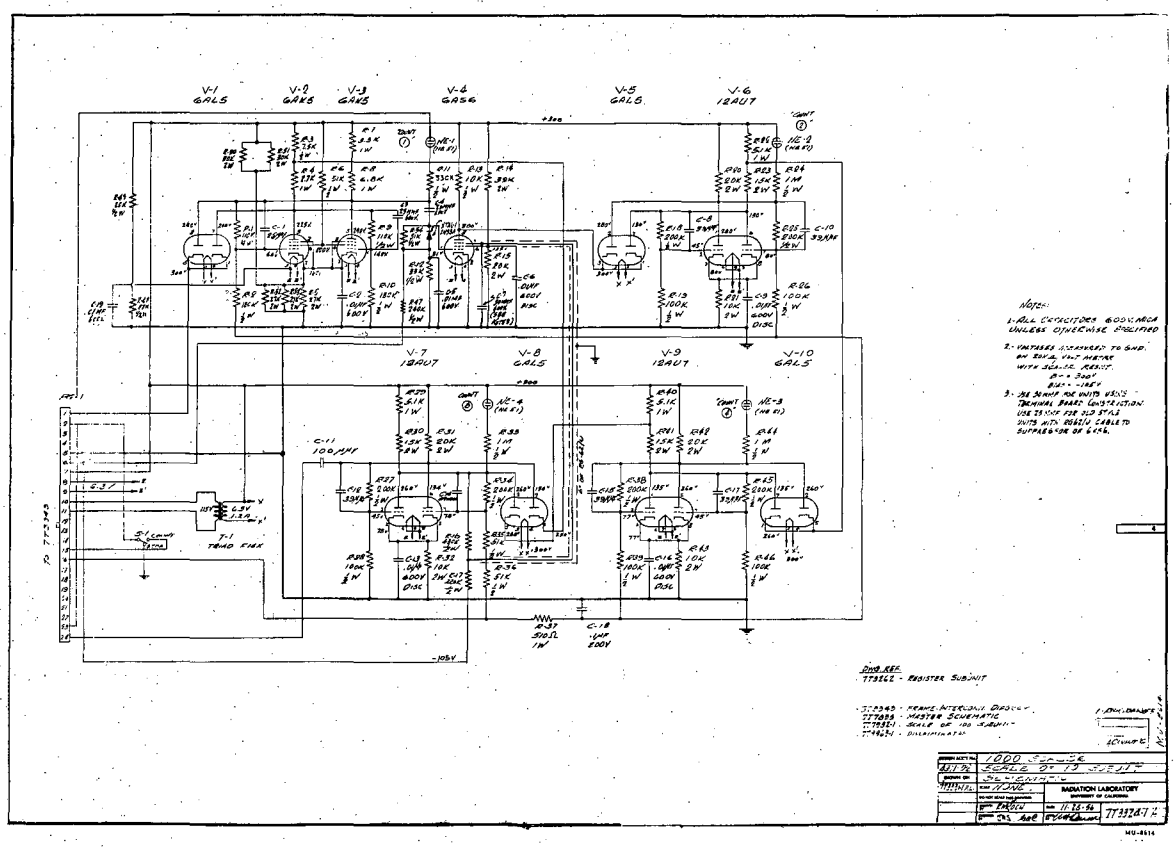


Fig. 16. Scale-of-10 subunit of 1000 scaler.

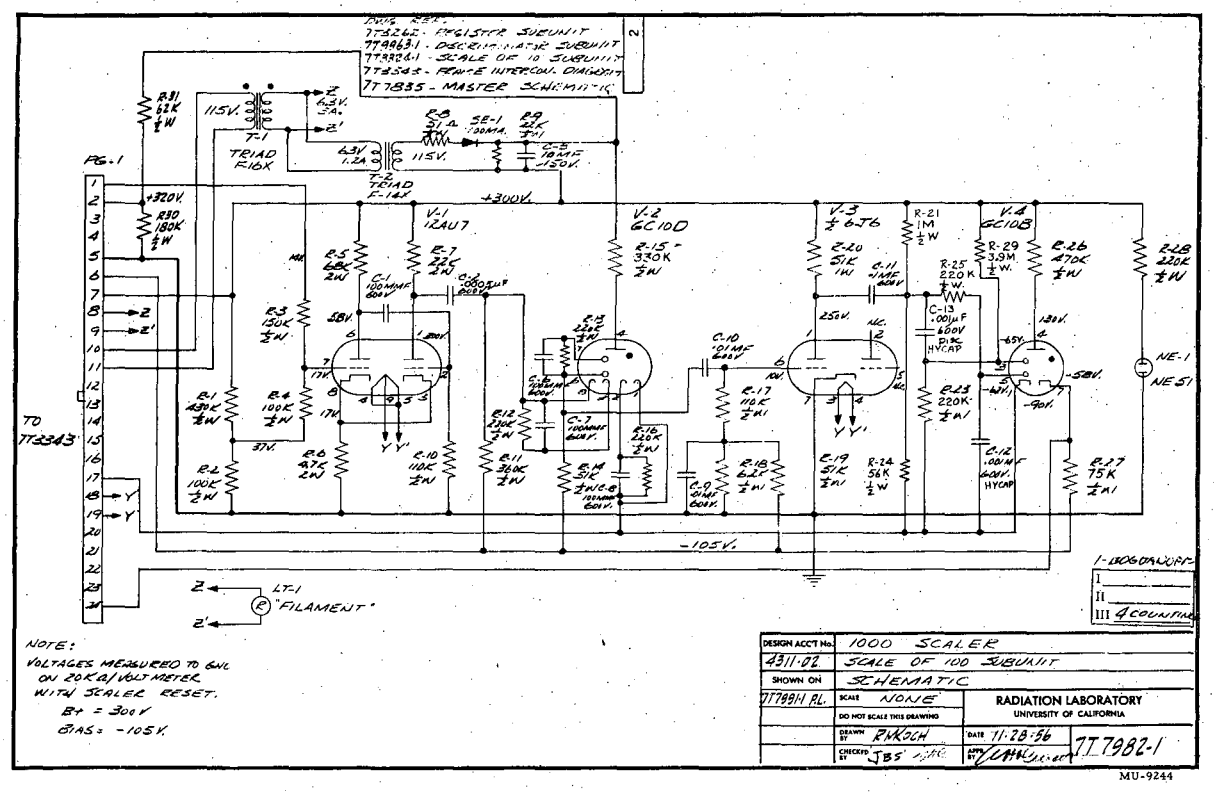


Fig. 17. Scale-of-100 subunit of 1000 scaler.

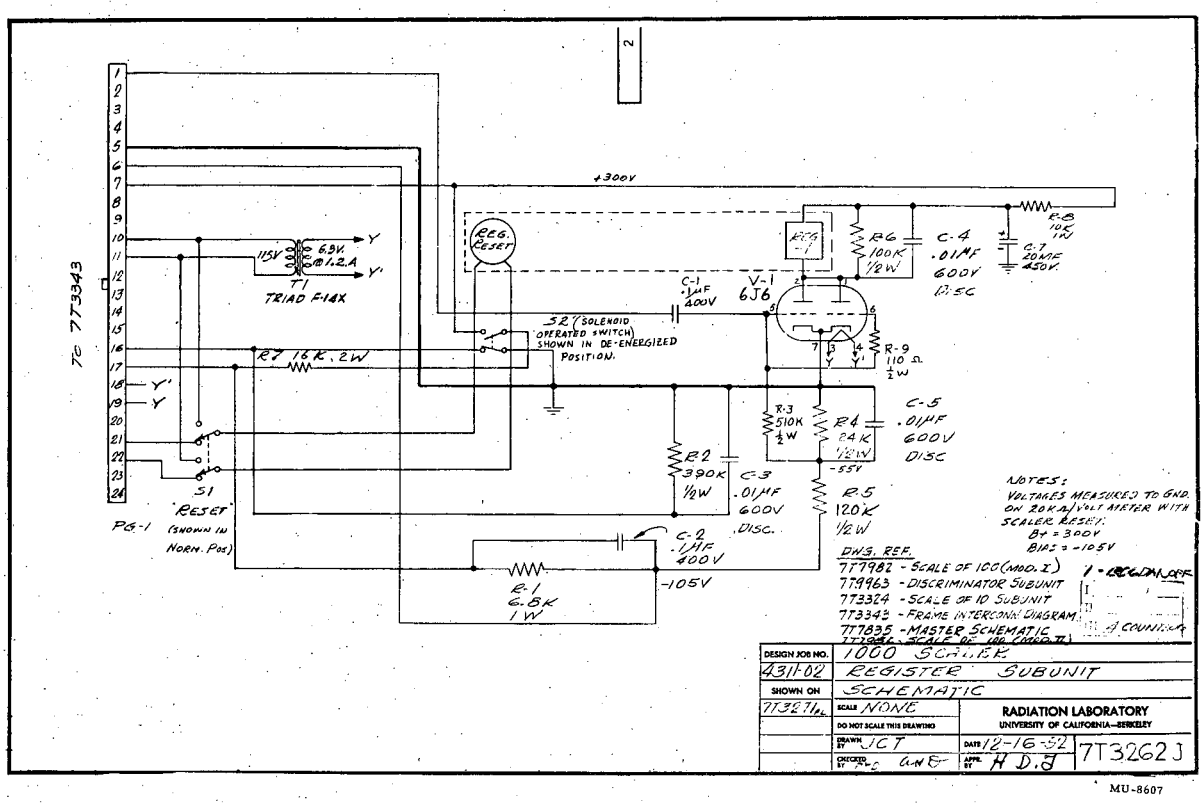


Fig. 18. Register subunit of 1000 scaler.

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