

GAS SCINTILLATION COUNTERS

by

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## ABSTRACT

Construction details of a 5" diameter 1.7 litre gas scintillation counter are given. The behaviour of the counter when containing  $\text{He}^4$  gas and a mixture of  $\text{He}^4$  and 10% Xe at various total pressures has been investigated. Two 5" photomultiplier tubes and a variety of different gas purifiers were tested in an attempt to obtain the best possible resolution.

The pulse height was found to depend sensitively on the purification procedure adopted. A resolution of 10% for the  $\text{Po}^{210}$   $\alpha$ -peak has been attained in two different chambers containing  $\text{He}^4$  plus 10% Xe. The voltage pulses rose in  $\leq 300$  nsec. Intense irradiation of a counter with 6 Mev  $\gamma$ -rays caused only a slight increase in the maximum noise level already present from the photomultiplier tube.

The behaviour of the counter as a fast neutron detector has been investigated using an uncollimated beam of 4.1 Mev neutrons. Pulses from recoiling protons and  $\text{C}^{12}$  nuclei were observed as well as the  $\text{He}^4$  recoils.

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## I. INTRODUCTION

### 1. The advantages of noble gas scintillation counters

Pulses from a gas counter are faster than those from ion chambers and some solid scintillators. Rise and fall times of the order of  $10^{-7}$  sec to  $10^{-8}$  sec have been reported but are dependent on the type and purity of gas used. The pulse height is a linear function of energy and is independent of both the charge and mass of the exciting particle. Continuous purification of the gas maintains a constant pulse height indefinitely. By varying the pressure the stopping power of the counter gas may be varied over a wide range without serious alteration of the pulse characteristics. Signal to noise ratios of 150:3 and a corresponding resolution of 4.8% have been attained using a 2" quartz window photomultiplier (Sayres and Wu 1957), however pulse height and resolution are dependent on the type of gas in the counter. Almost complete insensitivity to gamma radiation makes these counters extremely useful for photodisintegration experiments.

Because of the simplicity of its construction and operation, the size and shape of the gas chamber may be adjusted over a wide range to meet the requirements of a particular experiment. All counters reported in the literature have been 2" diameter and the 5" diameter chamber reported in this thesis is at present the largest. The cost of a counter is determined primarily by the photomultiplier tube and electronics and in some cases by the counter gas.

A number of applications for gas scintillation counters have been suggested.

- a) The fast pulses could be clipped to reduce  $\alpha$  pileup during fission counting.
- b) Gas scintillators provide fast pulses for counter telescopes.
- c) Using a mixture of 10% Xe in He<sup>3</sup> a gas scintillation neutron spectrometer of high efficiency using the He<sup>3</sup> (n,p) H<sup>3</sup> reaction could be built.

- d) The counters are ideal for observing the photodisintegration of the noble gases.
- e) Gamma ray promptness measurements could be made using the gas counter as a fast detector of the recoils.
- f) A  $\text{He}^4$  filled counter may be used as a polarizing analyser for fast neutrons.

## 2. Relative efficiencies of counter gases

The efficiency of a counter is dependent on chamber geometry, reflectivity, type and thickness of waveshifter and many other factors so inter-comparison of results from different counters is difficult. The following is a list of successful counter gases with an estimate of their relative efficiencies.

Xe (6 psia) - Pulse height 105, increased to 145 with quartz tube.

Kr (8 psia) - 50, increased to 83 with quartz.

A (10 psia) - 15, increased to 28 with quartz.

He (45 psia) - 38, increased to 40 with quartz.

He + Xe - 10% Xe in He increases pulse height to 125.

Other mixtures of noble gases are reported by Northrop and Gursky (1958).

He +  $\text{N}_2$  -  $\text{N}_2$  in small quantities increases the pulse height.

## II. A SURVEY OF PREVIOUS WORK

Most of the photons emitted in a noble gas following the passage of a charged particle were found to have wavelengths in the ultraviolet (Muehlhause, 1953, Egglar and Huddleston 1956, Northrop and Nobles 1956 and Sayres and Wu 1957). These photons have been detected either by using an ultraviolet-sensitive photomultiplier (Nobles 1956, and Sayres and Wu 1957) or by using a waveshifter which absorbs ultraviolet radiation and re-emits photons in a spectral region more nearly matching the sensitivity of conventional photomultipliers. Northrop (1958) lists the relative efficiencies of a representative group of organic-phosphor waveshifters which have been used by various experimenters. He found diphenyl stilbene most efficient whereas Sangster (1952) found that quaterphenyl combines the highest efficiency with the shortest decay time. ( $\sim 8 \times 10^{-9}$  sec) Northrop's results on quaterphenyl and diphenyl stilbene used with He + Xe are in disagreement with further work published by Northrop and Gursky. It is possible that this discrepancy could be resolved by using MgO and waveshifter instead of aquadag on the chamber walls. Forte (1956) used sodium salicylate successfully. The phosphors were applied either by vacuum evaporation (Northrop and Nobles 1956, Sayres and Wu 1957) or by painting with a solution of phosphor-plastic mixture (Egglar and Huddleston 1956).

The counters were found to be susceptible to quenching of the light pulses by contaminants present in the gas or arising in the counter walls and gaskets. Out-gassing from the counter system has been controlled by using a clean, out-gassed metal system and Teflon gaskets (Sayres and Wu 1957). For low-priced counter gases fresh gas was allowed to flow through the counter at a rate which was sufficient to overcome the rate of evolution from the system. (Boicourt and Brolley 1954). The third method which also

removed contaminants present in the gas used to fill the counter was to circulate the gas in a closed system which included a purifier containing a reactive material such as hot calcium (Sayres and Wu 1957), uranium (Northrop and Nobles 1956), calcium magnesium alloy (Forte 1956), or barium (Bennett 1957). These purifiers effectively remove  $H_2$ ,  $O_2$ ,  $N_2$  and  $CH_4$ , the contaminants suggested by Northrop and Gursky (1958). Forte (1956) observed that extremely small quantities of  $N_2$ ,  $O_2$  and  $CO_2$  poisoned the pulses from argon.

To obtain maximum energy resolution, the interior walls of the chamber were coated with a light reflector. The most commonly used material was smoked Magnesium oxide (Northrop and Nobles 1956, Sayres and Wu 1957, Pasma 1957, Rubbia and Toller 1958 and Baldin et al. 1957). Egger and Huddleston (1956) used silvered and aluminized surfaces and Forte (1956) used white enamel. In all cases the resolution was reasonable. Because the reflectivity of these materials falls off in the vacuum ultraviolet, more efficient operation resulted if the reflector was coated with a layer of waveshifter. Northrop and Nobles (1956) found that the optimum thickness for tetraphenylbutadiene and quaterphenyl was  $30 \mu g/cm^2$ . This is in agreement with theoretical predictions.

Quantitative measurements of the properties of gas scintillation counters by various investigators are not in agreement. The decay of the scintillation light has been reported as being fast ( $10^{-9}$  sec) by Nobles (1956), having more than one decay period and having a decay period inversely proportional to pressure (Northrop and Nobles, 1956, Avivi and Cohen 1957) and in the case of "nitrogen as a waveshifter", being a function of the fractional amount of nitrogen present (Egger and Huddleston 1957). Nobles (1956) reported a voltage rise time of  $3.5 \times 10^{-9}$  sec for  $\alpha$ -particles in Xenon. Forte (1956) reported a rise time of  $1.5 \times 10^{-7}$  sec for  $\alpha$ -particles in pure argon and  $5 \times 10^{-8}$  sec in "tank" argon.

Unlike solid scintillators, noble-gas pulse heights are a linear function of energy and independent of the charge and mass of the exciting particles. Nobles (1956) reported measurements of pulse height in xenon gas versus energy for protons, deuterons and  $\alpha$ -particles from 2 to 5 Mev for which a straight line could be drawn through all the points. An energy intercept of 0.5 Mev was unexplained. Boicourt and Brolley (1954) measured fission and  $\alpha$ -particle spectra in Krypton gas and found the proper ratio between the energies of the light and heavy fission fragment mass groups, but each was too large with respect to the 4.76 Mev  $\alpha$  group.

Few measurements of light output have been made. Northrop and Nobles (1956) list relative light outputs for  $\alpha$ -particles on NaI (Tl) and a number of noble gasses, and Northrop (1958) found Xe gas and diphenyl stilbene waveshifter as efficient for  $\alpha$ -particle detection as NaI (Tl). Sayres and Wu (1957) found  $\alpha$ -particle pulse heights in CsI, Anthracene, and He + 10% Xe to be in the ratio of 160:22:105. All results show that even the most efficient combination of gas and waveshifter results in a less efficient particle detector than solid NaI (Tl).

Resolution variations with gas pressure have been observed, however this effect was attributed to "concentration of  $\alpha$  tracks" at higher gas pressure (Sayres and Wu 1957) and no extrapolation to large-volume counters or to distributed source conditions can be made. Baldin et al. (1957) report an almost linear increase in pulse height with pressure in the range from 10 to 80 atmospheres for pure He and for He + 3% Ne. In the range from 1 to 10 atmospheres the pulse height was constant.

Binary gas mixtures have been extensively investigated by Northrop and Gursky (1958). All 10 mixtures studied have the same general features. There is a large drop in light output for a small amount of the heavy gas in a large fraction of the lighter one. For example, the minimum of the He + Xe

curve extends over fractional Xe concentrations from  $10^{-4}$  to  $10^{-3}$ . A peak occurs at 10% Xe 90% He in which the light output was found to be slightly larger than for pure Xenon. It has been suggested that this mixture, because of its large light output and relatively high stopping power, would be of use in constructing a fast  $\text{He}^3$  counter for neutron detection (Batchelor, Aves and Skyrme, 1955). For all binary mixtures a large fraction of lighter noble gas in a heavier one did not seriously decrease the light output of the heavier gas.

Baldin et al. (1957) and Egglar and Huddleston (1956) have both found an intensity peak in the light output from He when  $10^{-1}\%$   $\text{N}_2$  is added. The pulse height drops quickly as the concentration is increased to 1%  $\text{N}_2$ . Baldin et al. (1957) report a similar peak at  $10^{-2}\%$   $\text{O}_2$  in He with very rapid quenching of the pulses as  $\text{O}_2$  percent is increased.

### III. THE THEORY OF SCINTILLATIONS PRODUCED BY CHARGED PARTICLES IN A GAS

#### 1. Excitation and Ionization Produced by the Slowing of Charged Particles

The slowing of a fast charged particle in a gas leaves energy in the form of ions, neutral excited atoms, and ionized and excited atoms. For 1 Mev of  $\alpha$ -particle energy expended in Xe, in He<sup>4</sup> and Argon the following division of energy was calculated.

i) Xe  $W_o = 21.4$  ev spent in the formation of one ion pair. (Rossi & Staub)

$$N_{xe} = \frac{10^6}{W_o} = \text{Number of ion pairs formed per Mev expended.}$$

$E_{xe} = 12.13$  ev ionization potential. (Richtmyer, Kennard & Lauritson)

$$N_{xe} = 46,800 \text{ ion pairs}$$

$$\text{Total energy expended in ionization} = N_{xe} E_{xe} = 570 \text{ Kev}$$

$$\text{Total energy used in excitation} = 1000 - 570 = 430 \text{ Kev}$$

ii) He  $W_o = 31.0$  ev  $E_{He} = 24.58$  ev

$$N_{He} = 32,200 \text{ ion pairs}$$

$$\text{Total energy expended in ionization} = 790 \text{ Kev}$$

$$\text{Total energy lost in excitation} = 210 \text{ Kev}$$

iii) Argon results in 40,000 ion pairs with 630 Kev ion pair energy and 370 Kev excitation energy.

These results cannot be directly correlated with the observed pulse height from each gas, thus they indicate that the photon emission and absorption processes are sensitive to more than just the relative energy of ion pairs to excitation energy, or to the number of ion pairs created.

#### 2. Photon emission

The energy level diagrams of the neutral and singly ionized noble gases are characterized by a very high first excited state, typically of the order of two-thirds of the ionization energy. Since the transition probability for allowed transitions between any two levels is proportional to  $(\frac{h\nu}{mc})^3$ , (Heitler) the

majority of the radiation will be from ground state transitions. From the relation  $\lambda(\text{A}) = \frac{12,380}{E(\text{ev})}$  and using  $E = \frac{2}{3} E_{\text{ionization}}$ .

$$\lambda_{\text{H}_e} = 750 \text{ A}$$

$$\lambda_{\text{X}_e} = 1530 \text{ A}$$

Both these primary emission lines lie in the vacuum ultraviolet and cannot be observed directly with glass envelope photomultipliers. The speed of the light pulses from a gas scintillator will, along with other factors, depend on the speed of these ground state transitions. A theoretical estimate of the lifetime of the lowest resonance state in He, for example, gives a value of  $5 \times 10^{-10}$  sec (Wheeler 1933). Measurements made on allowed lines in the visible region vary from  $10 \times 10^{-9}$  sec to several  $\mu$  sec, and it is known that ultraviolet transitions are faster.

Unfortunately the speed of this primary process is not observed in the scintillation light because any allowed transition ending on the ground state of the neutral atom produces resonance radiation. (Mitchell and Zemansky) There is a very large probability for subsequent re-absorption of such a quantum before it can escape from the gas and be detected. This phenomenon, called resonance trapping, requires such quanta to be absorbed and re-emitted many times before escaping from the gas or being lost due to collision processes. Radiation from allowed jumps between the higher levels as well as all radiation from excited ions and atoms has a much lower probability of being re-absorbed due to the low density of states involved in the transition.

Competing collision processes could provide important modes of de-excitation. A collision with an impurity atom may quench the photon or, in the case of a nitrogen atom, may shift the wavelength of the primary radiation as discussed in a following section. The effects of collisional transfer of charge in binary gas mixtures is of major importance in understanding their

light emission processes. To obtain a grasp on the relative importance of collisions, the collision frequency and collision period were calculated for He and Xe at standard temperature and pressure.

The collision frequency for a gas atom is given by the equation

$$Z = \frac{4d}{3\eta} \frac{RT}{M} \quad (\text{Glasstone}) \quad Z = \text{Collision frequency (sec}^{-1}\text{)}$$

$d$  = Gas density ( $\text{g/cm}^3$ )

$\eta$  = Gas viscosity (Poises)

$R$  =  $8.31 \times 10^7$  (erg/deg C Mole)

$T$  = Absolute Temperature

$M$  = Atomic weight of the gas

Values for  $d$  and  $\eta$  obtained from the Handbook of Physics and Chemistry,  
29th edition

yield:

$$Z_{\text{Xe}} = 2.05 \times 10^9 \text{ collisions/sec}$$

$$Z_{\text{He}} = 2.3 \times 10^9 \text{ collisions/sec}$$

The corresponding collision periods,  $\tau$ , are:

$$\tau_{\text{Xe}} = 5.0 \times 10^{-10} \text{ sec}$$

$$\tau_{\text{He}} = 4.3 \times 10^{-10} \text{ sec}$$

The viscosity of a gas is independent of pressure except for very high or very low pressures, hence the collision frequency varies directly with pressure and the collision period varies inversely.

Because of this short collision period, energy transfer between excited and unexcited atoms in inelastic collisions competes strongly with direct radiative transitions. This high collision frequency combined with the large number of times that a resonance photon is emitted and re-absorbed indicates the way in which minute quantities of contaminants can act as poisons in gas scintillations.

A contaminant molecule absorbs energy either in an inelastic collision with an excited noble gas atom or by direct photon absorption. The energy is then degraded into thermal energy or into the infrared region where detection is not possible. Parenthetically it may be noted that a waveshifter is essentially a poison except that re-emission is in the visible region rather than the infrared.

Bennett and Wu (1957) have investigated the ultraviolet spectra due to the passage of  $\alpha$ -particles through helium and through argon. It was found that even with a nitrogen content of less than  $10^{-5}$ , the most intense lines observed were due to nitrogen. This effect has been attributed to collision excitation of  $N_2$  molecular band heads. In helium they observed the first negative system of the  $N_2^+$  molecule with the strongest band head at  $3914.4 \text{ \AA}$  and all nitrogen emission between  $3900 \text{ \AA}$  and  $5300 \text{ \AA}$ . In argon, the second positive system of the neutral  $N_2$  molecule was excited with the strongest band head at  $3371.3 \text{ \AA}$ . In a similar study, they found that the optical spectrum from xenon gas was a continuous distribution from  $2300 \text{ \AA}$  to  $6000 \text{ \AA}$ .

In addition to the levels in excited atoms that combine with the ground state in allowed transitions, there are also many metastable levels excited for which dipole radiation to a lower level is forbidden (Phelps and Molnar 1953). Excitation of these levels by the primary charged particle is not a forbidden process and a significant amount of energy may be deposited in these states which have long radiation life times. Fortunately there is frequently a close resonance level. With the collision frequency which was calculated for the gas there is a rapid exchange of energy between allowed and metastable levels, and the decay of the metastable level is controlled by the lifetimes of these nearby resonance levels and not by its radioactive lifetime. Sayres and Wu (1957) have explained  $N_2$  excitation in helium as being collisions with the  $2^1S_0$  (19.8 eV) metastable state of helium. This collision process

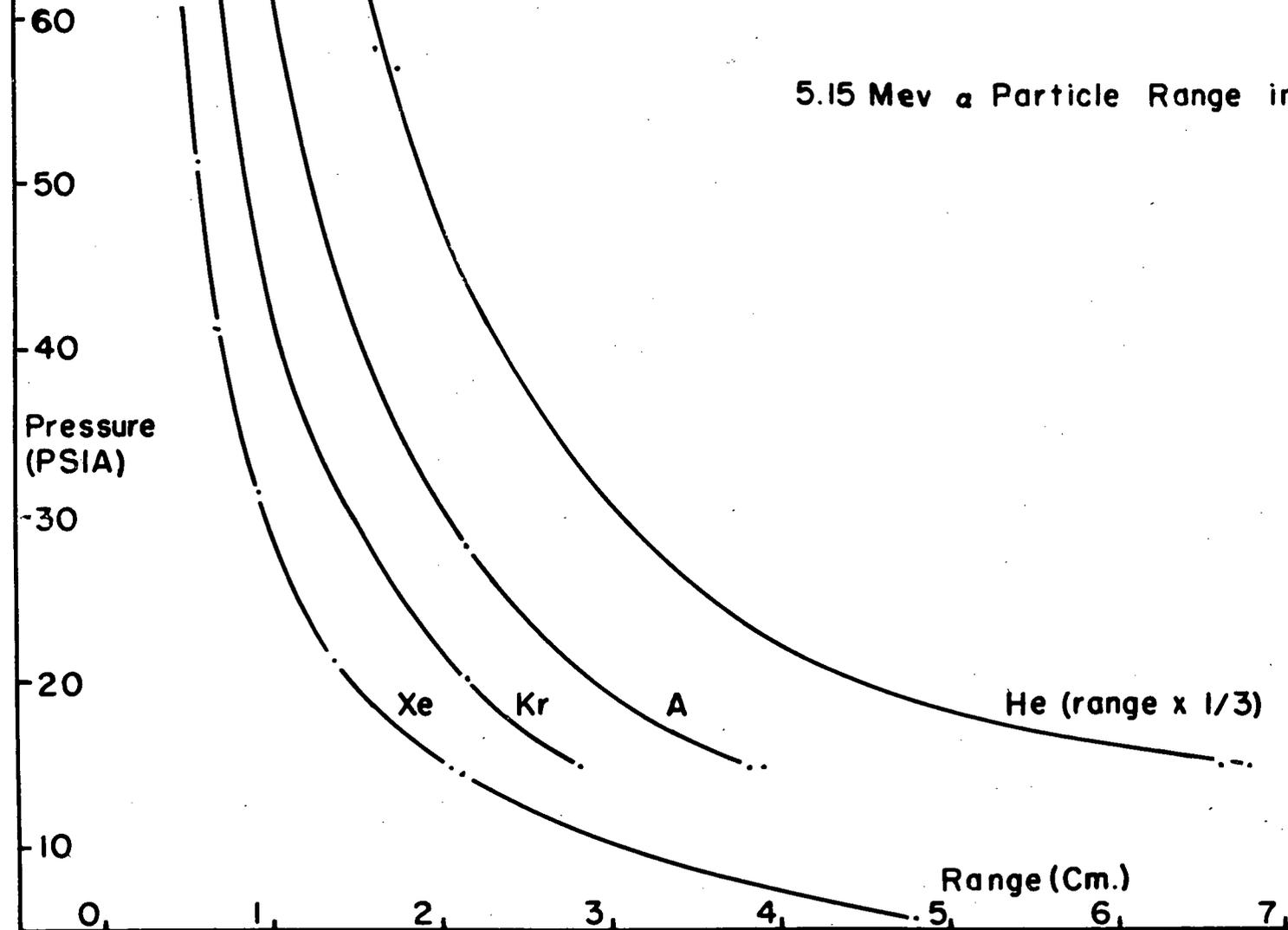
with the metastable states may explain the observed fact that the rise time of a noble element scintillator containing nitrogen is dependent on the nitrogen concentration.

No consideration has yet been made of the light coming from the primary particle energy expended in ion formation. This light arises when the ions recombine with thermal electrons. A study of this effect was made by Biondi et al. (1951-1957). Using microwave techniques, the rate of disappearance of electrons following an electrodeless discharge at several mm Hg pressure was observed. Recombination rates far higher than those theoretically estimated for the direct radiative capture process were observed. A subsequent investigation of mixtures of noble gases showed that the usual recombination process in a pure noble gas was as follows. First a stable molecular ion was formed in a radiationless collision, then the radiationless capture of an electron left the molecule unstable and it split into two excited atoms. Biondi estimated that the atomic ion recombination rate is at least a factor of  $10^3$  smaller than the molecular ion coefficient, thus the majority of the light emitted by ions results from the formation and de-excitation of molecular ions.

In the mixture of He + 0.1% A studied, another process predominates in which collisions between excited or ionized helium atoms and argon atoms results in a nearly complete transfer of the charge in the gas to the argon atoms because a heavier noble gas always has a lower ionization potential. Since the low density of argon makes argon-argon collisions rare, there is essentially no molecular ion formation. Subsequent recombination with electrons takes place only in a radiative collision and hence has a much smaller probability than the radiationless splitting of unstable molecules. This effect was used by Northrop to qualitatively interpret the dip in light

Figure 1. The Range of 5.15 Mev  $\alpha$  particles in Noble Gas

5.15 Mev  $\alpha$  Particle Range in Noble Gas



intensity observed in binary gas mixtures. He concluded that the light was still emitted, however the decay time was much longer than the electronic resolving time and hence was not observed. No measurements with resolving time greater than  $0.5 \mu$  sec were made.

The stopping time of the primary particle places an inherent limitation on the rise time of light pulses in a noble gas. Using the ranges listed in Figure 1 and assuming constant deceleration, the stopping times for 5.1 Mev  $\alpha$ -particles in He and in Xe at various pressures were calculated.

- a) He (45 psia) -  $7.5 \times 10^{-9}$  sec
- He (60 psia) -  $6.0 \times 10^{-9}$  sec
- b) Xe (5 psia) -  $6.2 \times 10^{-9}$  sec



#### IV. CONSTRUCTION DETAILS OF GAS SCINTILLATION COUNTERS

##### 1. Mechanical specifications for He<sup>3</sup>-filled scintillation counters

###### a) Size

To obtain an adequate count rate three litres of He<sup>3</sup> will be used in the gas chamber. Assuming that the maximum length of the cylindrical chamber is twice its diameter, the following He pressures were calculated.

Table 1

Chamber diameter	Pressure (0°C)	Volume
2 inches	220 psia	210 cm <sup>3</sup>
3	64	700
5	14	3200

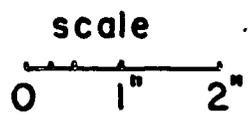
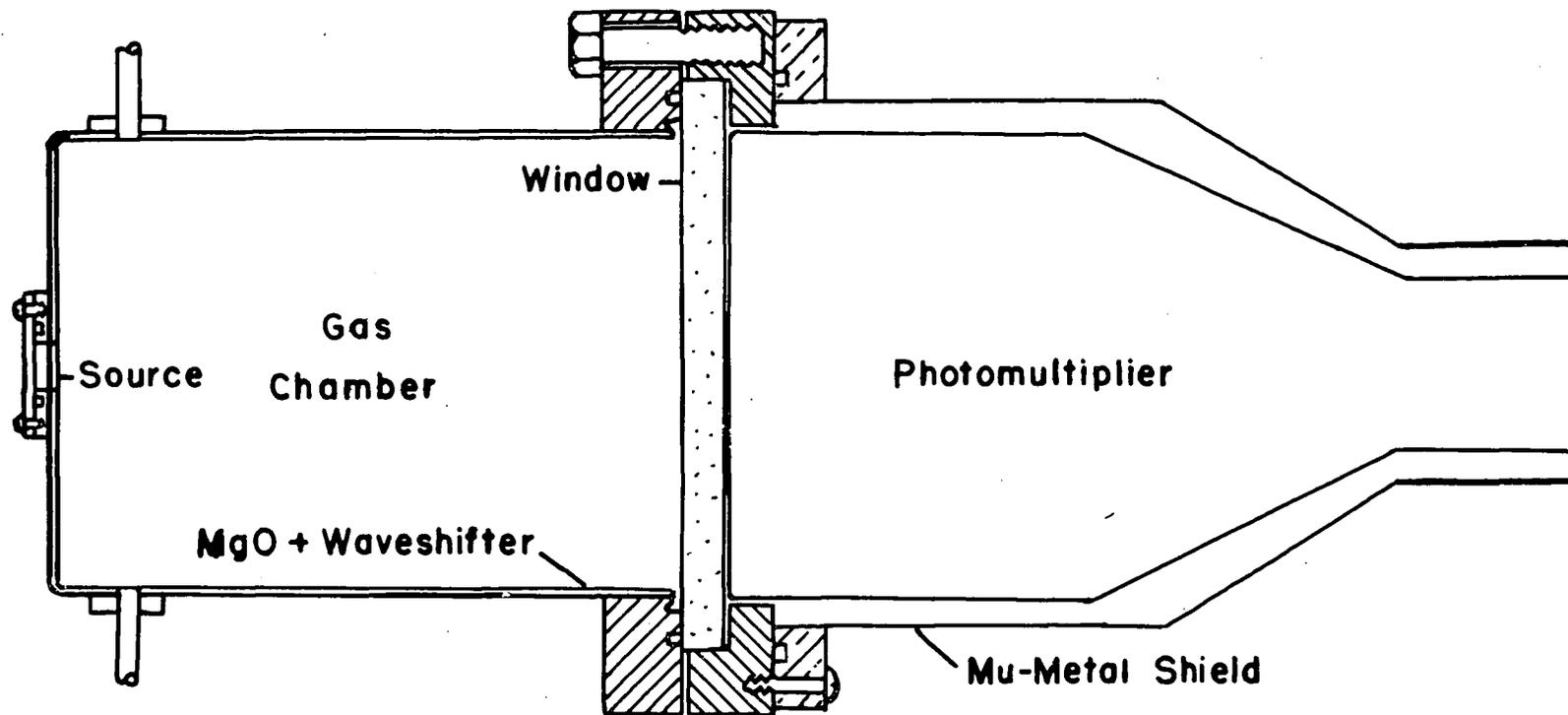
An internal pressure of 220 psia was considered excessive. Also a  $\frac{1}{2}$ " diameter source tube must be inserted through one end of the chamber and this would use up a large fraction of the interior volume in a 2" or even 3" chamber. Three 5 inch photomultipliers were available but 3 inch tubes were not, thus a 5 inch chamber diameter was chosen.

- b) The chamber and purifier must be vacuum and pressure tight.
- c) The walls of the sensitive volume should be thin to allow  $\gamma$ -ray penetration.
- d) All metal parts must withstand 150 psia internal pressure.
- e) Materials used must have low vapour pressures to reduce out-gassing, and consequent quenching of the light pulses.

##### 2. The Gas Chamber

A cross-sectional view of a typical chamber is given in Figure 3. The walls and end are a thin wall stainless steel beaker manufactured by Vollrath.

Figure 3. Cross Section of a 5" Gas Scintillation Counter



The top flare on the beaker was removed and a flange containing an o-ring was fastened on. Designs incorporating Vallrath beakers were limited by an inherent stress in the beaker wall near the top. All holes drilled in the wall cracked the beaker when heat was applied to silver-solder the joints, but holes very near the bottom of the beaker caused no trouble.

Two gas chambers were built, one with a brass flange and the other with a steel one. Thermal stress was not serious in either chamber when heated to 200°C. The brass flange was fastened to the beaker with 1200°F silver solder flowed around the outside joint. This construction allowed the inlet holes to be drilled through the flange without serious cracking of the beaker but it also left a gas pocket inside the chamber. The steel flange was welded as shown in Figure 3. Because this construction gave no support to the beaker walls, the inlet holes had to be drilled near the bottom.

A window retainer ring was threaded onto the brass flange whereas the ring for the steel flange was bolted on. Both rings sealed the window but the threaded ring resulted in a smaller flange assembly. The front face of the retainer rings was machined smooth to insure a light tight seal with the photomultiplier shield.

A compound retarded bourdon gauge with a 2½" diameter dial read interior pressure to ± 3% in the range from 0 psig to 50 psig and also indicated approximate vacuum. The gauge was sealed to a 3" length of 3/16" diameter copper pipe with an annealed copper washer and the copper tubing was silver soldered to the chamber. This length of copper tubing allowed the gauge to be moved during vacuum evaporation.

The chamber is operated at both vacuum and pressure, thus o-ring grooves were cut to the exact outside and inside diameters of the o-rings and the

depth was chosen to allow the o-ring to just fill the groove when pressed very hard. Great care was exercised in polishing the o-ring grooves. Teflon o-rings<sup>\*</sup> were used for the coupling joints and the end plate seal on the chamber but a more elastic material such as Kel-F<sup>\*\*</sup> or Viton-A<sup>\*\*</sup> was used for the front window seal. Teflon has a low vapour pressure and withstands a temperature of 540°F, however the o-rings require a greater sealing pressure than neoprene o-rings and permanently take the shape of the groove. Viton-A and Kel-F are chemically similar to Teflon, have low vapour pressures and withstand high temperatures (Viton-A 500°F; Kel-F 300°F). Their greater elasticity allows them to be re-used and reduces the sealing pressure to that of neoprene. To eliminate contamination of the counter gas no grease was used on any of the o-rings.

The interior of the chamber was cleaned by swabbing it for 15 minutes with each of the solutions listed to remove scale from the stainless steel.

- |                   |                                   |
|-------------------|-----------------------------------|
| a) Scale loosener | Sulphuric acid (conc.) 1 part     |
|                   | Hydrochloric acid (conc.) 1 part  |
|                   | Water (65°C) 8 parts              |
| b) Scale Remover  | Hydrochloric acid (conc.) 5 parts |
|                   | Nitric acid (conc.) 1 part        |
|                   | Water ("hot") 14 parts            |

After a chamber had been thoroughly out-gassed by heating it to 200°C while the interior was evacuated, a thick layer of MgO was smoked onto the interior metal surface by burning a total of about 6 feet of Magnesium ribbon near the chamber mouth. The chamber interior was then coated with a reasonably uniform layer of waveshifter by two vacuum evaporations from a tantalum boat. To obtain a uniform layer of the correct thickness two boat

<sup>\*</sup> Obtained from Anchor Packing Company, Vancouver, B.C.

<sup>\*\*</sup> Obtained from Vinylloyd Company, 720 N. Broadway, Los Angeles 12, California.

positions were used, one coaxial with the chamber and at the mouth, and the second about 4 inches outside the mouth. The tantalum boat was covered with fine-mesh stainless steel screen when quaterphenyl was evaporated to stop it from "jumping" from the boat. Diphenylstilbene on the other hand did not "jump" at all and was very easy to apply.

Three types of window have been tested on the chamber. Destructive pressure tests were not made thus comparative strengths are unknown, but each type was run to 75 psia. The three types with their distinguishing properties are listed below.

Pilkington plate glass - available in Vancouver in  $\frac{1}{4}$ ",  $\frac{3}{8}$ " and  $\frac{1}{2}$ " thicknesses. Both faces are flat but the edge is chipped and the glass appears greenish. The strength of these windows is unknown.

Hobbs Herculite glass discs - available in any size but 1 month delivery time. The faces are flat and the edge is rough ground to the exact diameter. This glass is slightly green. The company supplied a  $\frac{1}{2}$ " thick plate for use at 150 psig with no guarantee. Both faces have two clamp marks  $\frac{1}{4}$ " from the edge which makes installation on the o-ring difficult.

Quartz discs - available in all sizes but the cost is high and delivery is at least one month. The faces are flat and the edge is accurately ground. Quartz is known to be transparent to visible and ultraviolet light and its strength is greater than that of glass.

Windows were coated with a thin layer of waveshifter by mounting them on a 5 inch inside diameter brass ring held 30 cm from a tantalum boat inside an evacuated bell jar. The thickness of the layer was controlled by the quantity of waveshifter in the boat. A measurement of the thickness was made by mounting 2 cm<sup>2</sup> of aluminum foil near the window and noting the increase in weight. This method was very inaccurate but it indicated two

filled boats consistently deposited less than  $100 \mu\text{gm}/\text{cm}^2$  at the distance used. If windows are mounted closer to the evaporation boat a non-uniform layer results.

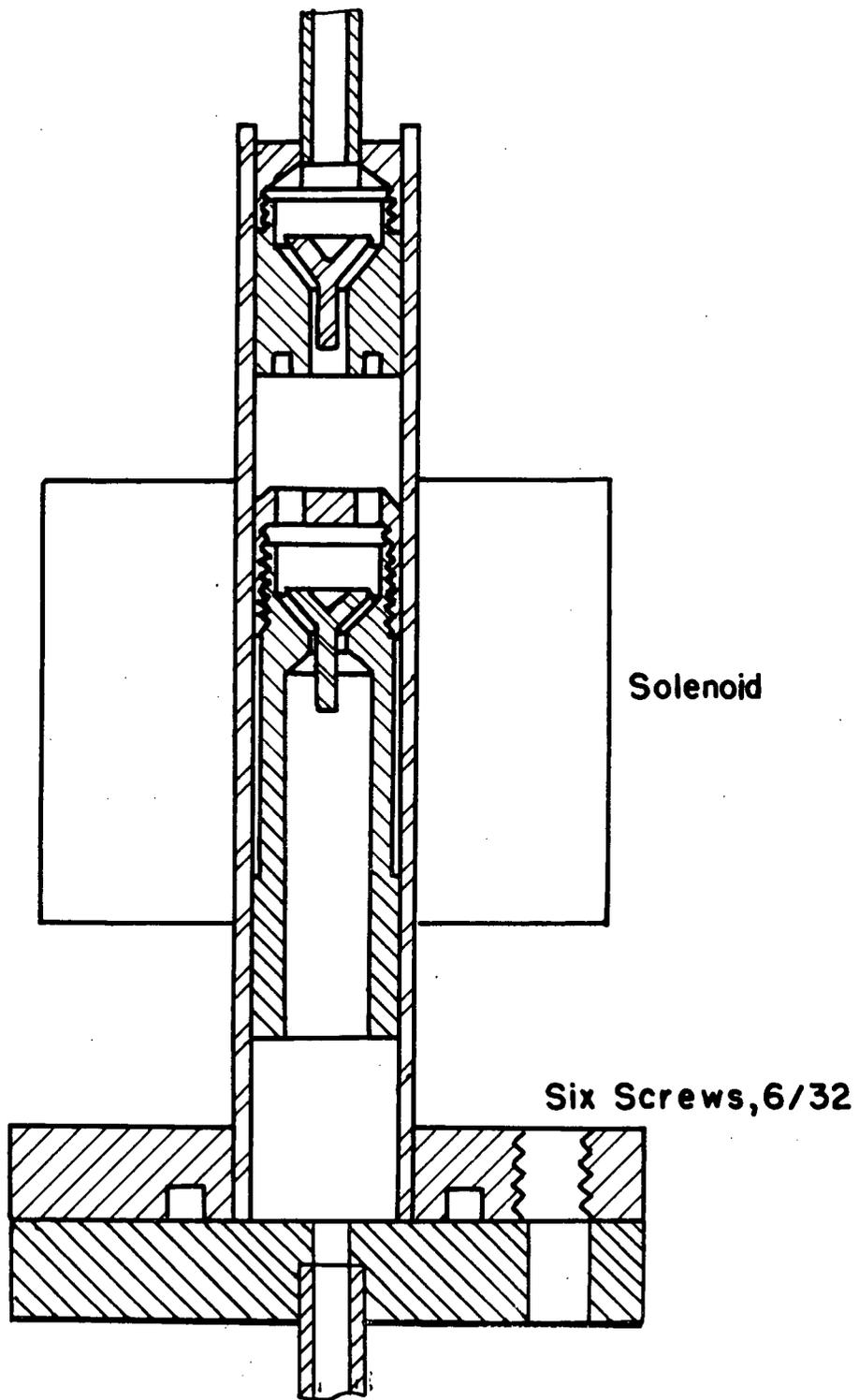
With careful attention to the o-ring seals, both types of chamber were vacuum and pressure tight and have withstood an interior pressure of 75 psia. The MgO and waveshifter layer is mechanically stable but is destroyed by moisture. During out-gassing, oil (possibly from the spinning of the beaker) was given off from the walls so a week of hard pumping at  $200^\circ\text{C}$  was necessary before MgO was applied. The end of the chamber snaps when changing from vacuum to pressure thereby limiting the type of apparatus which may be mounted on the end but no chipping of the MgO layer has been observed. In one of the chambers the inlet pipe from the circulator points toward the front window, and after continued operation a small mark shows on the waveshifter layer however no other deterioration of the waveshifter has been noted.

### 3. Circulation Pumps

The circulation pump continually forces chamber gas through the gas purifier, or through a cold trap to separate the Xenon from the Helium (see Figure 2). The gas flow rate should be variable to give flexibility in experimental runs, and the internal volume must be small.

#### a) Mylar diaphragm pulser with check valves

A 2 inch diameter 7 mil Mylar diaphragm as described by Sayres and Wu (1957) was mounted approximately as shown in Figure 1 of their paper. Two uni-directional check valves and a 3 way solenoid valve were connected as shown in this Figure. The 110 volt ac used to drive the 3 way valve was pulsed by a cam-activated microswitch and a Variac was used to control the speed of the ac-dc motor which drove the cam. High pressure air used to pulse the diaphragm was taken from the laboratory air line.



scale  
0 1/4" 1/2"

Figure 4. Cross Section of a Magnetic Circulation Pump

This pump was much too large and was very difficult to mount in the purification circuit. The 3-way valve made by General Controls needed frequent cleaning and at times chips from the valve scored the Mylar diaphragm. The interior volume of the pump was large relative to its pumping speed and it was difficult to increase the speed above 15 cc per minute. A number of the diaphragms ruptured allowing gas to leak out. This type of pump was used during preliminary work with the counters but was rejected in favour of the solenoid type described in the next section.

b) Magnetic solenoid pump

The construction of this type of pump is shown in Figure 4. Stainless steel and brass were used for all parts with the exception of the pump slug which was turned from magnet iron. Valve seats which were ground with emery dust were almost 100% efficient when operated vertically. A commercially manufactured solenoid of  $\frac{1}{2}$ " inside diameter and  $1\frac{1}{4}$ " long was mounted as shown. With a 30 volt dc supply it was capable of lifting the iron slug vertically  $1\frac{1}{4}$ ", but to insure reliable operation the pump stroke was kept to  $\frac{3}{4}$ ". When tested at atmospheric pressure the pump delivered 1.3 cc of air per stroke, and it could be pulsed at 150 cycles per minute.

This pump was found to be superior in the following ways.

- i) it may be operated with any interior gas pressure.
- ii) it is small and easily mounted in the circulation line. (Volume less than  $3 \text{ cm}^3$ )
- iii) it may be serviced easily.
- iv) the pumping speed can be greater than 130 cc per minute.
- v) it is reliably sealed and may be operated safely at high internal pressures.

One such pump has been circulating Helium at 30 to 60 psig for several months. The metal slug stuck in the stainless steel tube several times but this defect has been eliminated by polishing the slug with fine emery.

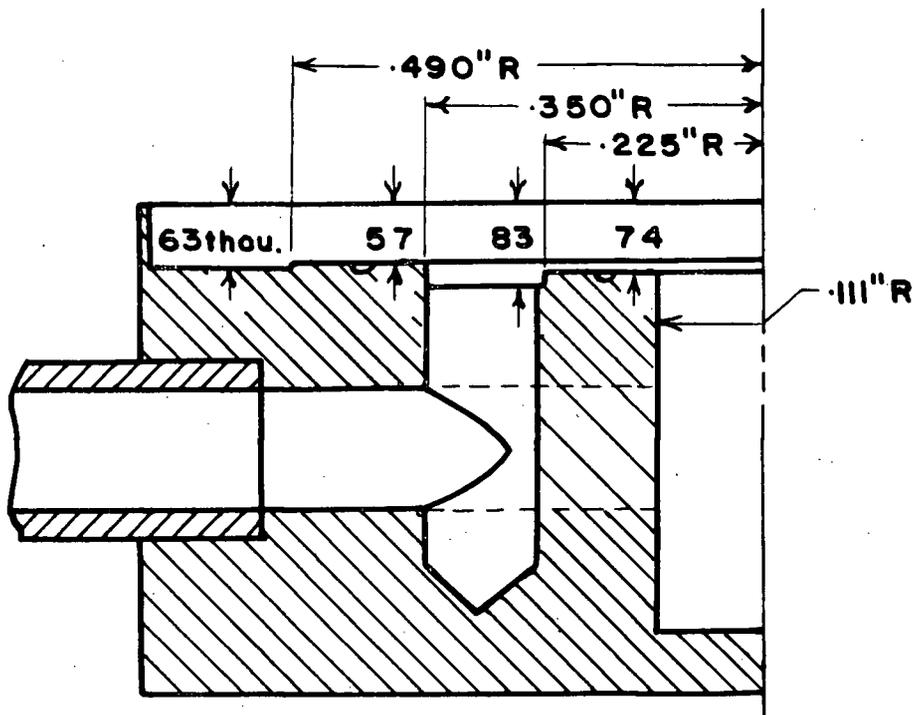
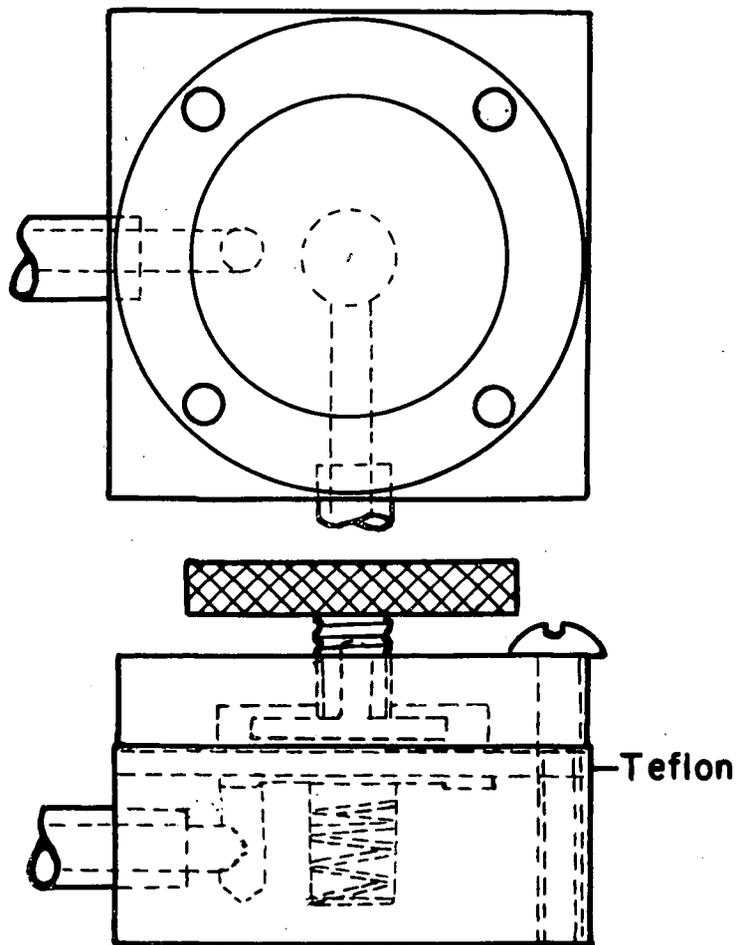


Figure 5. Construction Details of a Harwell Valve

c) Electrical pulsers for the pumps

The cam-activated microswitch driven with an ac-dc stirring motor was found to be noisy and unreliable, primarily because the motor was old. A number of cams was made to allow greater variation of the pulse rate but without an unreasonable number of them it would not be possible to adjust the "on" and "off" times.

To drive the solenoid pump at about 100 cycles per minute, a multi-vibrator was built with a 10,000 ohm telephone relay as one plate resistor. A 6J6 tube in a free-running multivibrator circuit with a 300 volt plate supply switched sufficient current to activate the relay. Two 1 megohm potentiometers controlled the pulse rate and the "on" - "off" time. A full-wave bridge using Westinghouse TP25L rectifiers supplied 110 volts dc and a 500 ohm resistor in series with the pump solenoid limited the coil voltage to 30 volts dc. An 8  $\mu$ f capacitor was placed across the contacts.

4. Valves and Couplings

A large number of valves were required for use in conjunction with the 1/8" inside diameter (I.D.) copper tubing which connects the components in the purification circuit. These valves must be reliable when operated at a pressure of 150 psig and must have a gas flow rate equivalent to 1/8" I.D. copper tubing. To eliminate vapours in the counter they must either be packless or have a Teflon seat. The dead volume must be  $< 1 \text{ cm}^3$ . A number of commercial valves meeting these specifications were priced and found too costly.

A form of "Harwell" valve shown in Figure 5 was built. A 1/16" thick Teflon diaphragm efficiently sealed these valves. To overcome creep of the Teflon diaphragm, the modified valve shown in Figure 5 replaced this first design. The double seats "trap" a small bead of Teflon and thus reduces creep. This valve has been completely reliable until dirt accumulates on the brass seat.

Any number of holes may be drilled to the centre post of the valve thereby increasing its flexibility. The flow resistance from the centre hole to the outside hole is low and a series of these valves will not decrease gas flow noticeably.

To permit removal of components from the purification circuit and to connect valves to components, a small o-ring coupling was designed. Details are similar to a standard coupling used in this laboratory but were scaled to a number 5 o-ring. Either Teflon or Viton-A o-rings seal the couplings, but Teflon is badly distorted and should not be re-used to insure a vacuum seal. The o-ring seats must be carefully polished.

#### 5. Purifiers

Many different metals have been used to purify the noble gases in scintillation counters. Of these, hot calcium and calcium-magnesium alloy ( $\text{Ca}_3\text{Mg}_4$ ) were chosen. Both metals must be maintained at about  $400^\circ\text{C}$  while the gas flows through the purifier tube, and for efficient operation both must be out-gassed at  $500^\circ\text{C}$  or higher.

A  $\frac{1}{2}$ " outside diameter stainless steel tube  $2\frac{3}{4}$ " long contained the purifier metal. A bottom plate sealed with an o-ring allowed access to the interior, and gas flowed in from the top and out through the bottom plate. All joints were silver soldered with  $1200^\circ\text{F}$  solder. Two  $3/16$ " O.D. copper coolant coils were soldered to the top and the bottom of the stainless steel tube. Cold water flowed through the two coils in series to keep the outlet gas temperature low and to protect the o-ring and the soldered joints from high temperatures.

A Chromel-alumel thermocouple junction was strapped in contact with the stainless steel tube by wrapping the tube with four turns of 2" wide asbestos paper which was tied at the top and bottom with one turn of nichrome wire.

A close-wrapped coil of #28 nichrome wire was wrapped over the insulating layer of asbestos and an average coil resistance of 34 ohms at 20°C results. Four or five turns of asbestos paper over this coil thermally insulated the unit and the assembly is capable of reaching 700°C (surface temperature) with 2 amps ac flowing. An accurate measurement of the interior temperature is impossible, however with the thermocouple placed on the surface an out-gassing temperature at least 100°C higher than the maximum operating temperature is assured. Allowance should be made for the change from vacuum during out-gassing to pressure during purification.

A 3 inch long purifier barrel was found to be much more reliable than a 2" barrel and if the heating coil is at less than 70 volts with respect to the barrel four layers of asbestos provide adequate electrical insulation.

A charge of calcium consisted of four to five grams of clean calcium metal turnings cut into small pieces. A degreased stainless steel screen and steel wool baffle was inserted followed by the calcium chunks. The bottom was blocked with a second baffle held in place by the cover plate.

The calcium magnesium alloy used in the purifier was made as follows. Seventy percent calcium and thirty percent magnesium by weight were placed in an oxidized steel container, covered and placed on the bottom of a cylindrical steel vacuum chamber 3 inches in diameter. The top of the chamber was sealed with a cooled neoprene o-ring and when the chamber had been thoroughly evacuated with a rotary pump, heat was applied until the outside turned bright orange. This temperature was maintained for  $\frac{1}{2}$  hour. The resultant calcium-magnesium has a brilliant metallic lustre when cracked with a hammer. It is pyrophoric so must be handled cautiously, and storage under argon stops surface oxidation.

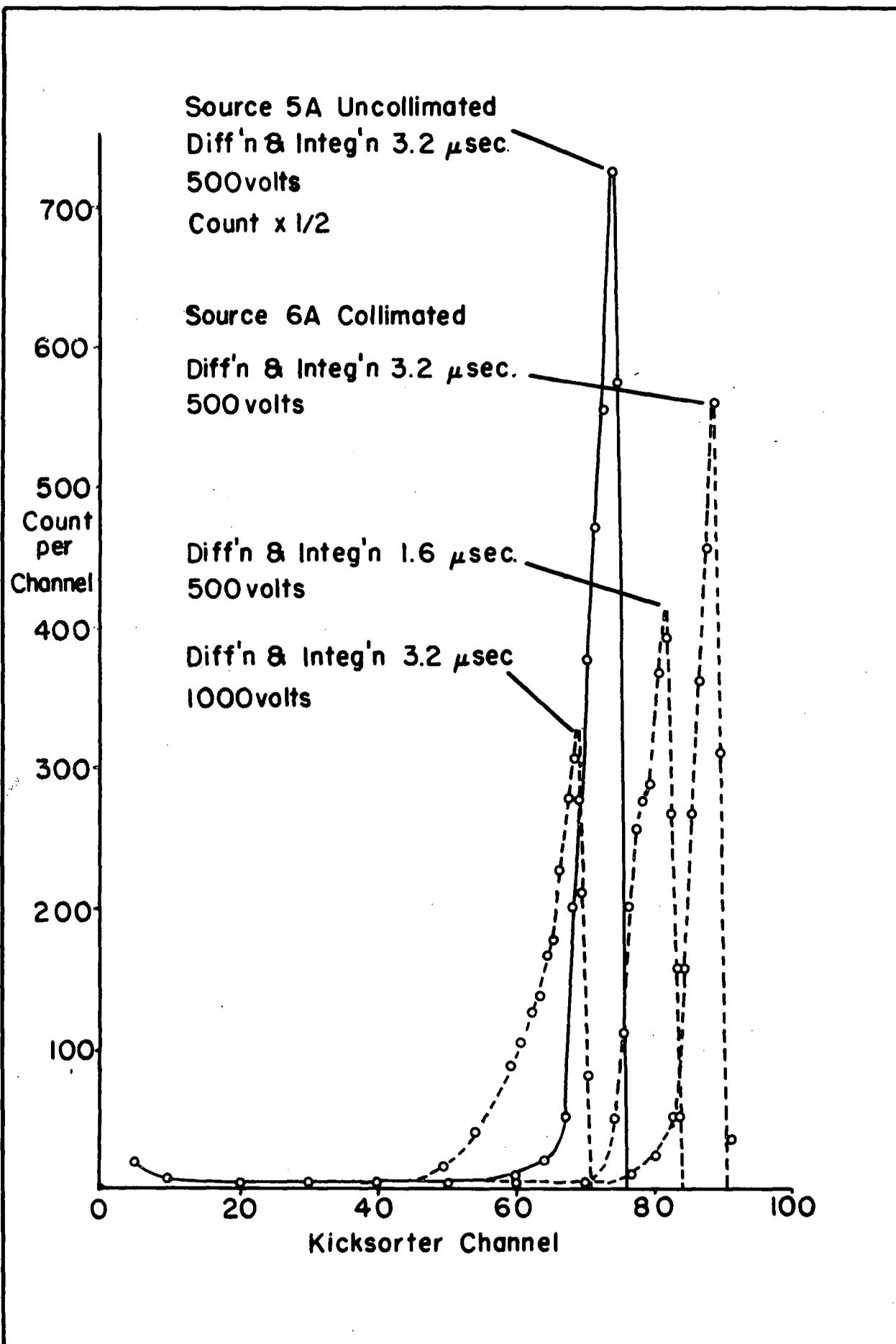


Figure 6. Ion Chamber Spectra of Two Po  $\alpha$  Sources

## 6. Calibration sources and source holders

To determine the resolution of the counter and to provide one point for energy calibration of the counter an internal  $\alpha$ -source was mounted on an iron holder which may be activated from the outside to blank off the  $\alpha$ -particles. The holder was inserted through the  $\frac{1}{2}$ " diameter hole in the end of the chamber and the source lay flush with the end surface. A typical source holder is shown in Plate 2.

Two sources were used for calibration. A Pu<sup>239</sup> source manufactured at Chalk River was used for all the preliminary runs with pure He<sup>4</sup>. The thickness of this source was measured as 1.4 Mev with the ion chamber described in Appendix A, thus resolutions measured with the source were in error.

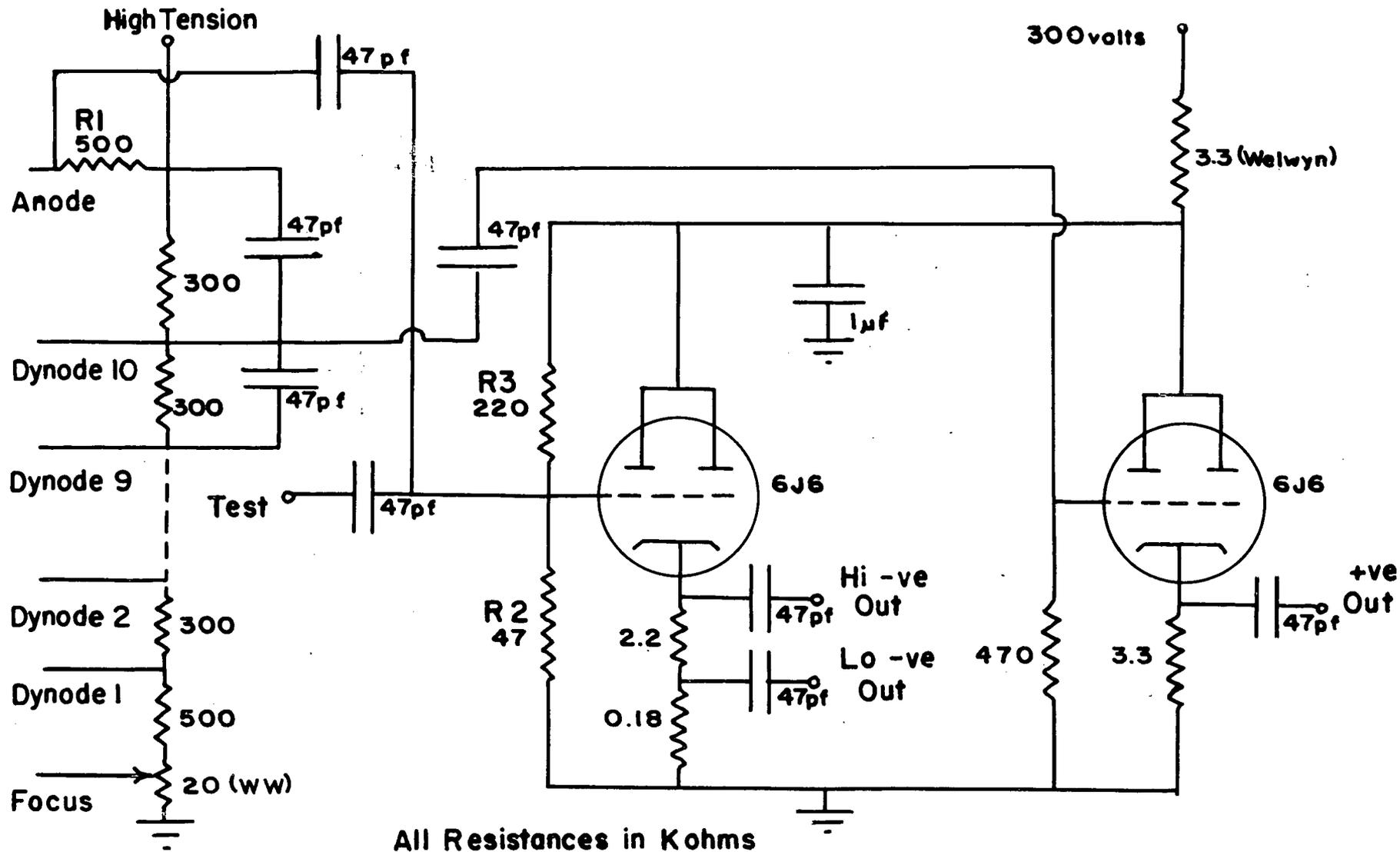
To replace this Pu<sup>239</sup> source a number of thin sources were made by dipping  $\frac{1}{8}$ " diameter polished silver buttons into a 0.5N HCl solution which contained a dissolved radium needle. Extremely high count rates were observed with a thin window geiger tube run as a proportional counter even with a dip time of less than 1 second. The ratio of  $\alpha$ -particles to  $\beta$ -particles was almost 1:1. The radium solution was diluted 10:1 with distilled water, and controlled dip times followed by vigorous washing immediately after dipping reduced the  $\beta$  to  $\alpha$  ratio and allowed the count rate to be chosen. Full width at half height of the 5.3 Mev  $\alpha$ -particle peak was better than 4% when measured with an ion chamber. Representative source spectra are shown in Figure 6.

## 7. Mounting of the components

The most efficient plan for connecting the basic components of a gas scintillation counter is shown in Figure 2. With these connections a number of advantages are apparent.

- a) the chamber may be sealed from the purifier circuit.
- b) the gas purifier may be isolated and evacuated to out-gas the purifier metal.

Figure 7. Circuit Diagram of a Photomultiplier Head Amplifier



- c) when filling the gas chamber, incoming gas is passed through the gas purifier
- d) the xenon in the gas chamber may be removed by circulating the chamber gas through a cold finger with the circulation pump.

Mechanical positioning of the components is shown in Plate 3. The gas scintillation counter was mounted 41" from the floor on a dexion rack. All components including pumps and power supplies were mounted on lower shelves. The unit can easily be moved into a position with the gas chamber very close to a target on the Van de Graaff.

## 8. Electronics

### A. Components

The head cathode follower shown in Figure 7 is used to match the output voltage signal from the photomultiplier to a 100-ohm coaxial cable which carries the pulses to the cathode follower input of a Dynatron wide band amplifier. The cable is terminated with a 100 ohm resistor at the cathode follower. Both positive and negative pulses are amplified by the head amplifier but all experimental results were taken using a negative output signal. With negative input pulses to the Dynatron amplifier, the positive output pulses must be inverted before activating the 100 channel C.D.C. kicksorter. This is done with a standard inverter circuit using a 403B tube.

A Northeast Scientific Supply Company regulated voltage supply, serial 82, provided high voltage for biasing the photomultiplier dynodes. Both a DuMont #6364 and an E.M.I. #6099B five-inch end-window photomultiplier tube were used to observe scintillations in the gas. The bias resistor circuit for the DuMont tube is shown in Figure 7. The circuit for the E.M.I. tube is similar (with the exception of the 20 K $\Omega$  focus control which is unnecessary) but was constructed with  $\frac{1}{2}$  Watt, 10% resistors.

The stability and linearity of the amplifiers and the kicksorter were measured by injecting precise pulses at the negative test input of the head cathode follower. The pulses were formed by discharging a 0.1  $\mu$ fd capacitor through a 34 ohm resistor. A Westinghouse 276D mercury switch driven by 60 cycle line current was used to charge and discharge the capacitor, and the precise charging voltage was supplied from the wiper of a 100 K $\Omega$  Helipot with 15 volts dc across it (Robertson, 1955).

B. Observed behaviour of the components

a) Photomultipliers

The photomultiplier tube was expected to limit the resolution attainable with the scintillation counter and also to be the major source of electronic noise, consequently an intensive investigation of the properties of the photomultiplier tubes was carried out under the following headings.

i) Photomultiplier noise

DuMont Two #6364 tubes were tested. Both were darkened for two days with 1700 volts on the dynode bias chain. The maximum noise pulses from the tube which was discarded were twice the height of those from the tube which was used. A Pye Scalamp galvanometer placed in series with the anode load measured the dark current. This current increased as the voltage was raised and at 1700 volts the dark current was 0.18  $\mu$ amp. A more useful measure of the noise for scintillation counter applications is the pulse height spectrum. Typical noise spectra are included in the calibration spectrum plots

(Figure 9). Numerous noise spectra taken during the investigation showed that:

The tube was very noisy immediately after it had been exposed to room light. After two or three hours of operation on the gas chamber it returned to normal.

Even when the tube had been darkened with the voltage off, it was more noisy immediately after the voltage was turned on.

Two trivial observations were that the maximum tube noise decreased to about three-quarters of its value at room temperature when the tube and the gas chamber were cooled with liquid air, and that tube noise increased with increasing bias voltage.

E.M.I. The current noise in the #6099B tube was  $0.037 \mu\text{amp}$  at 1700 volts. This is in agreement with published data and indicates that the tube has not been roughly handled. The noise behaviour was similar to that of the DuMont tube and typical pulse height spectra are shown.

ii) Photomultiplier gain

A two inch diameter NaI (Tl) crystal was optically coupled to the centre of the photocathode of both photomultiplier tubes. The voltage was varied from 800 volts upward for both tubes and the peak height of  $\text{Co}^{60}$  gamma pulses was plotted against voltage.

DuMont The pulse height varied as  $V^{7.0 \pm 0.2}$  where V = voltage on the dynode bias chain

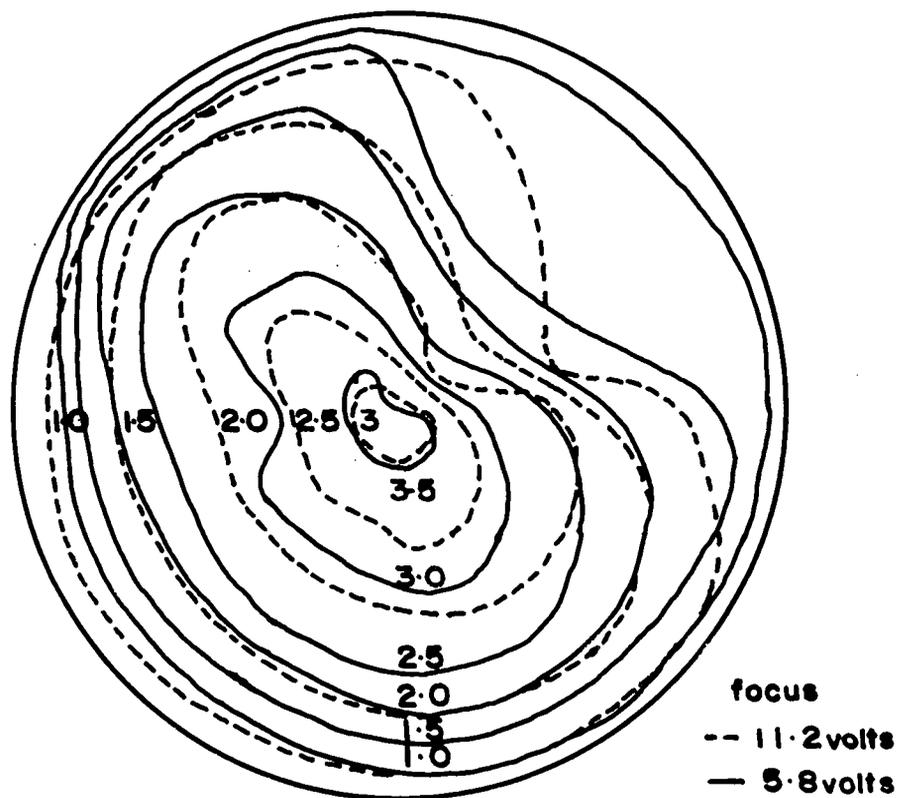
E.M.I. The pulse height varied as  $V^{8.0 \pm 0.2}$

Intercomparison of gas scintillation pulse heights at 1500 volts and 1700 volts showed that

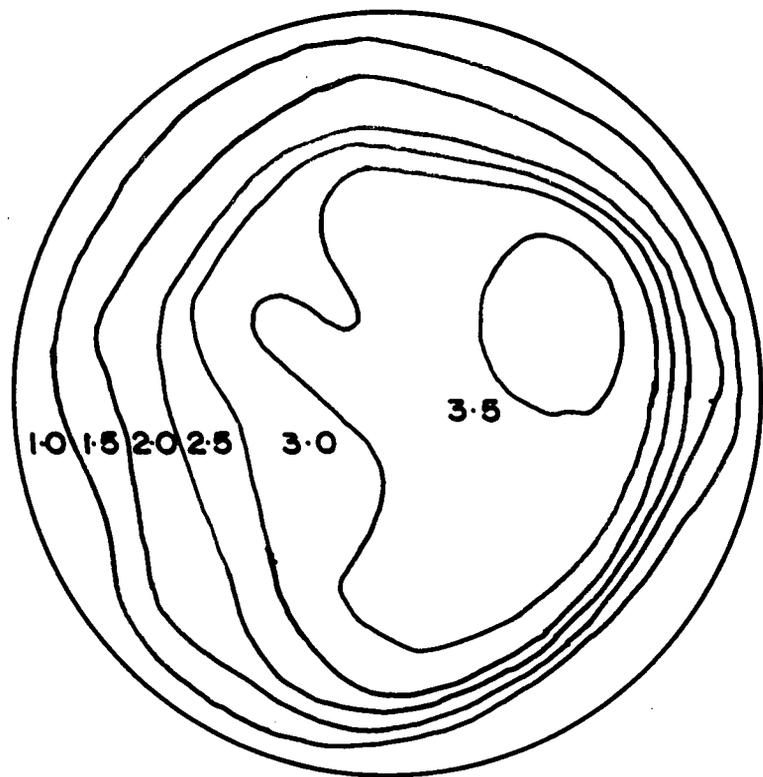
$$\frac{\text{E.M.I. Pulse Height}}{\text{DuMont Pulse Height}} = 3.2$$

Both tubes exhibit a gain shift during the first hour of operation. The apparent pulse height changes as much as 10% with time.

No stability measurements have been made on the high voltage supply for the photomultiplier. A gradual shift in this voltage would alter the pulse height and cause an increase in apparent resolution. Count times ranging from two minutes to three hours taken with clean gas resulted in constant resolution thus voltage drifts, if they occur, must be over a longer period than 6 hours.



DuMont Photocathode



E.M.I. Photocathode

Figure 8. Photocathode Sensitivity of the Two 5" Photomultipliers Used

iii) Photomultiplier resolution

A two-inch diameter NaI (Tl) crystal was optically coupled to the centre of the photocathode of the E.M.I. tube and the  $\text{Co}^{60}$  gamma peaks were counted using the electronics described in this chapter. A resolution of 8.5% was obtained for the most energetic gamma peak at both 1000 volts and 1200 volts. A larger crystal was not available, however Sharpe (1957) reports 8% resolution with a  $4\frac{1}{2}$  inch NaI (Tl) crystal coupled to a 5 inch #6099B tube.

iv) Photocathode uniformity

The uniformity of both photocathodes was measured by moving a  $\frac{1}{4}$ " diameter anthracene crystal and  $\text{Cs}^{137}$  beta source over the photocathode and observing the pulse height with a Tektronix #541 oscilloscope connected to the high output of the head cathode follower. Two one-gallon paint cans were used for the light-tight container and the photomultiplier tube neck was light-sealed through the centre of the bottom can thereby allowing the associated head cathode follower to be mounted outside the dark volume. A mu-metal shield placed around the photomultiplier reduced stray magnetic fields. The tube was mounted vertically in the bottom can with the photocathode facing up. The top can was sealed to the bottom one with an o-ring which allowed it to rotate, and the source and crystal hung downward from a  $3/16$ " rod which passed horizontally through the centre of it at a height which allowed the crystal to rest on the photocathode when the crystal and source were hanging downward. By sliding the rod and rotating the top can the crystal could be moved accurately over the whole photocathode.

Results of a number of tests are shown in Figure 8. The effect of varying the focus voltage of the DuMont tube is clearly seen.

b) Head cathode follower

Rise time

The input time constant of the load resistor and anode stray capacity

is very much longer than the fall time of the current pulse on the anode, thus the output voltage pulse will have a rise time equivalent to the fall time of the exponential current pulse, the order of  $10^{-9}$  second. The cathode-to-ground stray capacity of the paralleled 6J6 tube is the major limitation of pulse rise time. Assuming 25 pf stray capacity, the rate of change of cathode voltage for a negative step input on the grid is

$$\frac{dv}{dt} = \frac{i}{C} = \frac{18 \times 10^{-3} \text{ amp}}{25 \times 10^{-12} \text{ farad}} = 0.7 \text{ volts/n sec}$$

For a 2 volt output pulse the rise time is  $\sim 3$  n sec.

#### Fall time

The differentiation time constant of this circuit may be adjusted by varying either the anode load resistor (R1) or R2 (in this case R3 must also be adjusted to maintain the bias voltage on the grid). The fall time was set at  $1.5 \mu$  seconds. This choice eliminates non-linearity in the C.D.C. kicksorter caused by pulses which are less than  $\sim 1 \mu$  sec wide. The wideband amplifier settings used throughout the investigation were  $0.16 \mu$  sec. integration time and  $8 \mu$  sec differentiation time.

#### c) Linearity and stability of the components

With a 100 ohm cable connected to the Lo output jack, the head amplifier was linear for input pulses up to at least 18 volts. The stability of the head cathode follower, Dynatron amplifier, inverter and kicksorter was  $\pm 1.5\%$  in absolute pulse height over a period of 4 days.

## V. THE BEHAVIOUR OF HE<sup>4</sup> GAS SCINTILLATION COUNTERS

### 1. Preliminary measurements with 5" diameter chambers

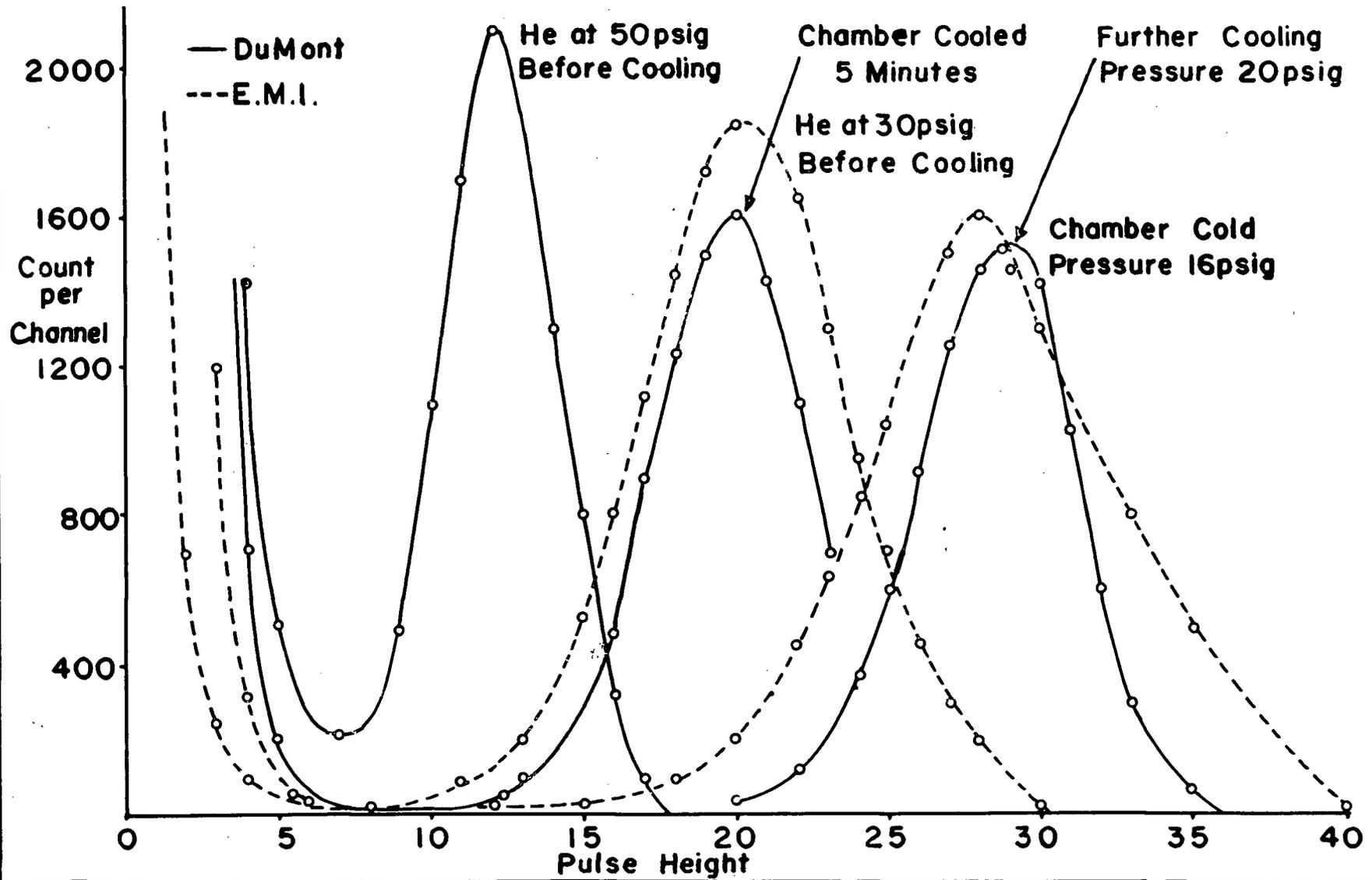
The first counter system tested consisted of a Plutonium  $\alpha$ -source mounted on the end of the brass-flanged chamber. The chamber interior was coated with MgO and quaterphenyl and the  $\frac{1}{2}$ " thick glass window was excessively and unevenly coated with quaterphenyl (possibly as much as 1,000  $\mu\text{gm}/\text{cm}^2$  at the centre). The chamber and purifier circuit were first evacuated with a rotary pump which was connected to the chamber with a rubber hose. A filling of He<sup>4</sup> from a Helium cylinder gave pulses slightly above the noise level of a 5" E.M.I. photomultiplier tube run at 1650 volts. The rise time of these pulses was  $\sim 40 \times 10^{-9}$  sec but the resolution was not measurable. Purification with calcium heated to 400°C resulted in a recognizable  $\alpha$  group which could be switched off, but the resolution was never better than 50%. Variation of the purifier temperature produced a radical change in pulse height (and resolution). The pressure was varied from 0 psig to 40 psig and wall effect occurred at 15 psig, however no significant change in resolution with pressure was observed beyond the wall effect region. The inclusion of an out-gassed charcoal trap in the filling line resulted in 50% resolution immediately after filling, but the calcium purifier caused a decrease in pulse height when heated to less than 400°C.

A substantial increase in resolution was obtained by replacing the calcium metal in the purifier with clean out gassed calcium chips. The ultimate resolution attainable with such a counter was 38% using the 5" E.M.I. photomultiplier tube. The  $\alpha$  peak was a factor of 3 higher than the average height of the photomultiplier noise.

### 2. The effect of cooling the scintillation counter

The chamber and mu-metal photomultiplier shield were cooled to liquid

Figure 9. The Effect of Cooling the Gas Chamber



nitrogen temperature by covering the metal surface with a soft cloth and carefully pouring on liquid nitrogen. The chamber gas pressure dropped due to leaks in the o-ring seals, but this was counteracted by flowing Helium in directly from the cylinder. The effect on pulse height and resolution is shown in Figure 9 for both a 5" DuMont photomultiplier tube and the E.M.I. tube. The pulse heights were adjusted so that both tubes gave peaks in approximately the same kicksorter channel. A similar run was made in which only the gas chamber was cooled and the pulse height increased without a decrease in photomultiplier noise, thus the effect is not caused by a gain shift in the photomultiplier. The pulse height in a chamber whose gas has been more completely purified shows little increase on cooling.

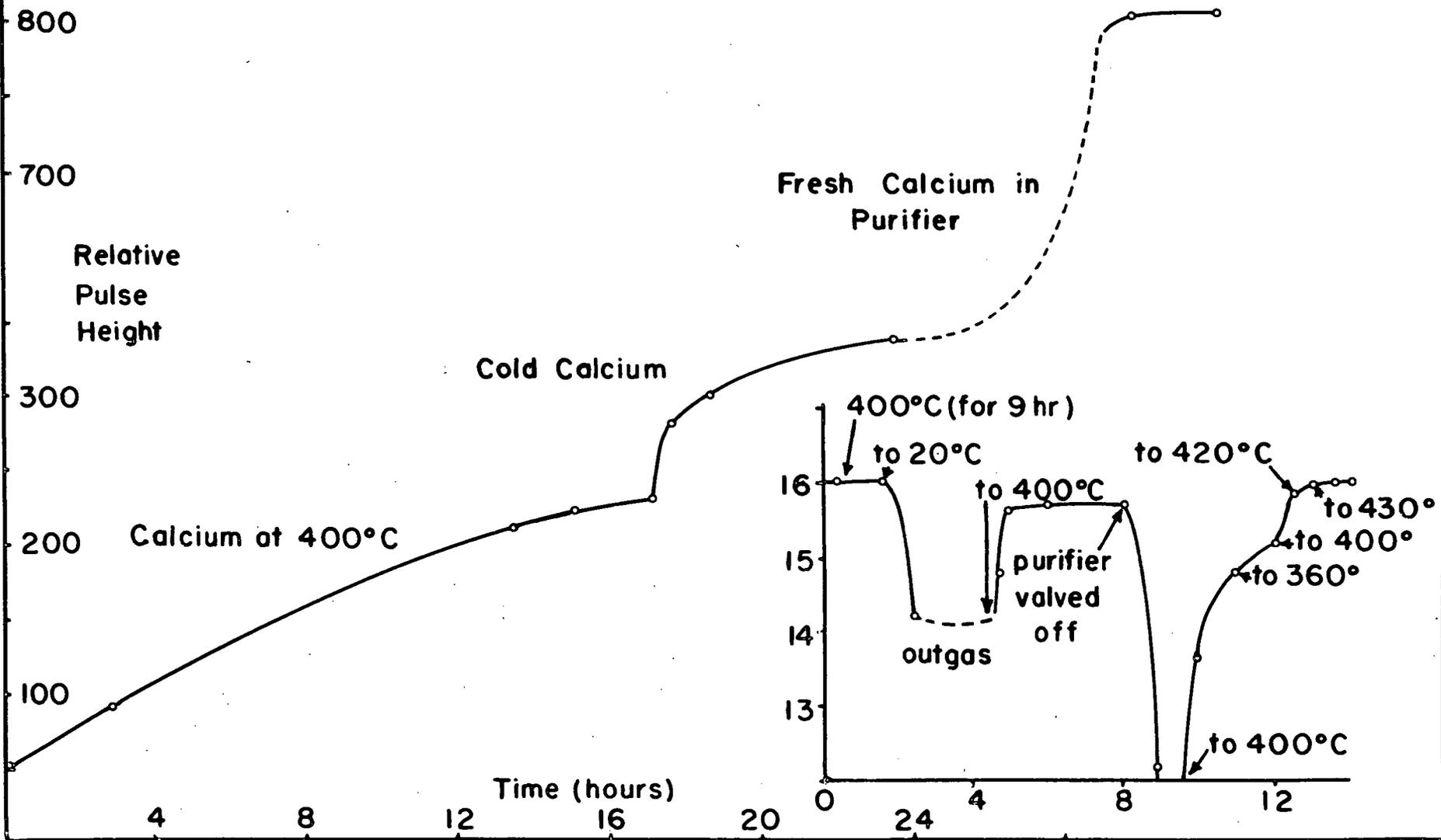
The increase in pulse height has been attributed to "freezing out" of impurities, however an attempt to duplicate these results using a cold finger attached to the chamber was unsuccessful. Gas circulation through a cold finger was not tried. It is conceivable that the increased pulse height was partially caused by a decrease in collision frequency at the lower gas temperature thereby reducing the possibility of quenching collisions. Unfortunately this cooling procedure may not be used with  $\text{He}^4 + \text{Xe}$  in the counter because the Xe would liquify, but it may be of use in producing  $\text{He}^4$  counters without external purifiers.

### 3. Gas scintillation pulses from Helium plus Xenon

The addition of 10% by pressure of Xe to the  $\text{He}^4$  increased the pulse height by a factor of three. The corresponding increase in resolution revealed a thick source tail on the  $\alpha$  peak. The counter resolution for the Pu  $\alpha$ -particles as measured from the high energy edge of the peak decreased to 16% and the signal to noise ratio increased to 38:3.

Following purification with hot calcium the gas chamber was sealed off and the pulse height decreased 0.6% per hour during the first 24 hours and

Figure 10. The Effect of Calcium Purification on He + 10% Xe



0.2% per hour during the second 24 hour period. This decrease was not caused by wet gas because cold calcium purification caused no increase in the pulse height.

Thin Po  $\alpha$  sources with a resolution of less than 4% were used for subsequent investigations because thin Pu sources of the size required were not available. One unfortunate consequence of using Po sources is contamination of the chamber and source holder caused by the well known phenomenon of "creeping" of Po-alpha sources. To counteract this effect as much as possible all Po-coated silver discs were mounted within a 0.125 inch inside diameter brass cup. With a source holder in the off position the shutter covered the top of the brass cup thereby stopping all  $\alpha$  particles.

With Po source #5A in the chamber a preliminary filling of 30 psig He<sup>4</sup> which had been passed through the charcoal purifier resulted in a resolution of 32% and a signal to noise ratios ( $\alpha$  peak height: average photomultiplier noise height) of 26:3. The ultimate resolution obtained with He<sup>4</sup> plus 10% Xe at a total pressure of 22 psig was 12% and the signal to noise ratio with the tube operating at 1600 volts was 90:3. Recent work with a source mounted midway on side wall of the chamber indicated that a resolution of less than 10% may be obtained. The source mount caused some broadening of the peak and an exact resolution could not be calculated.

The effectiveness of the calcium purifier and solenoid pump was investigated using source #5A with 30 psig He plus 10% Xe. The resultant relative pulse height graph, Figure 10, shows the importance of using fresh well out-gassed calcium operated at the correct temperature. The sharp rise in pulse height when the calcium was cooled to room temperature was a result of moisture removal from the gas by cold calcium. When the calcium has become ineffective for removing impurities from the gas, the surface layer of the

initially metallic calcium chunks is black and may be crumbled. Removal from the purifier barrel showed that only the calcium near the central region of the coil has deteriorated, thus a more effective heater could be developed in order that a larger percentage of the calcium would react to remove  $N_2$  from the gas more quickly. An alloy of 70% calcium -30% magnesium was used in the purifier. No comparison with calcium was made, however  $Ca_3Mg_4 + Ca$ . (the resultant mixture when 70% Ca and 30% Mg are mixed) has given equally good resolution in  $He^4$  and Xe gas mixtures. Cold calcium-magnesium alloy maintained the scintillation pulse height for 10 days following purification of the gas with hot  $Ca_3Mg_4$ . Before re-using the purifier at a high temperature it must be out-gassed.

#### 4. Further factors affecting the resolution of $He^4$ and $He^4 + Xe$ counters

##### a.) Filling procedure

Following a chamber evacuation using a rotary pump, two flushes of helium to a vacuum of ~15 inches Hg before the final filling of helium greatly increased the initial pulse height. This procedure was found unnecessary when the chamber was pumped to  $10^{-6}$  m Hg through a  $\frac{1}{2}$ " diameter tube. The decay time of the chamber was also lengthened by higher vacuum before filling. There was no measurable improvement when a cold trap was placed in the pumping circuit of the rotary pump which seems to indicate that the impurities are occluded on the chamber walls and are not due to back streaming from the forepump.

##### b.) Waveshifters

One unsuccessful attempt was made to observe visible light pulses from  $He^4$  without waveshifters on the interior of the chamber and the window. Following this, both quaterphenyl and diphenyl stilbene were tried as waveshifters. No apparent difference was found, so to standardize the investigation quaterphenyl has been used exclusively. The thickness of waveshifter layers

on windows must be greater than  $20 \mu\text{gm}/\text{cm}^2$  and less than a few hundred  $\mu\text{gm}/\text{cm}^2$  to obtain maximum pulse height. Two full tantalum boats deposit about  $40 \mu\text{gm}/\text{cm}^2$  at a distance of one foot (with an allowance for loss due to jumping). Such a layer when deposited on a glass window shows interference rings  $\frac{1}{2}$ " wide.

c) Windows

A He + Xe filled counter with a quartz window has attained a resolution of  $\leq 10\%$  with the DuMont tube whereas the ultimate resolution with a Pilkington glass window counter is  $\sim 10\%$  with the DuMont tube and a side-mounted source. Because of the large number of factors affecting resolution, it is not certain that this increased resolution resulted from the quartz. There is no measurable difference in resolution between the two types of glass window.

d) Impurity content in Helium counters

Immediately after filling a counter with purified helium the pulse height is unsteady. After one filling it rose 19% above its initial value (resolution  $15\frac{1}{2}\%$ ) over a period of 4 hours then fell to 10% below and remained constant during purification. The pulse resolution changed accordingly, thus the increase was not caused by a photomultiplier gain shift. If, on the other hand, the initial filling is dirty and the resolution is poor, the pulse height increased with purification but no maximum has been observed or if the chamber was sealed off the pulse height dropped. The effect is most probably caused by small variations in the nitrogen content of the gas. If, for example, the initial filling of helium had a nitrogen content of  $5 \times 10^{-3}\%$  then further nitrogen removal would cause the pulse height to rise and then fall to less than the initial height (Egglar and Huddleston 1956 - Figure 2).

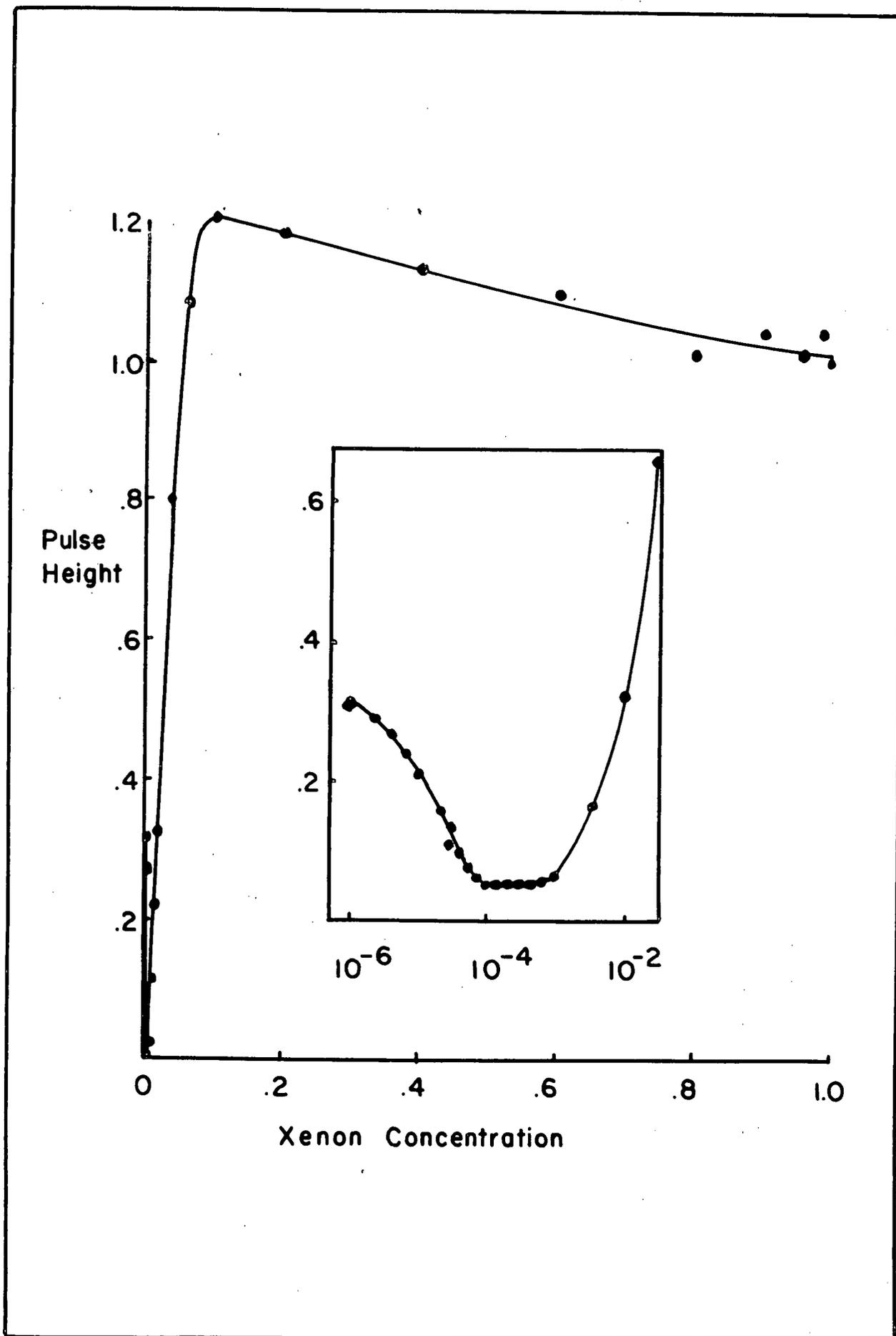


Figure 11. Light Output from He Plus Xe as Observed by Northrop and Gursky (1958)

e) Electronics voltage levels

The high tension voltage on the DuMont tube was varied from 1300 volts to 2000 volts. The resolution and signal to noise are shown in Table 2 .

Table 2

High Tension	Resolution	Signal to Noise	Comments
1300 volts	17%	29:3	peak in low kicksorter channel
1500 volts	13.5%	30:3	
1700 volts	13.5%	39:3	
1800 volts	13.5%	37:3	
1900 volts	13.5%	39:3	
2000 volts	13.5%	-	observed at Bias 5

Variation of the focus volts on the DuMont tube over the range shown in Figure 8 had no effect on the  $\alpha$  peak shape or height, hence photocathode sensitivity is not a serious limitation on chamber resolution.

f) Xenon concentration

As Xe is added to the He, the pulse height rises linearly when observed with the Tektronix oscilloscope, however a well defined maximum pulse height is not apparent because of the relatively slow mixing rate of the Xe in He. No decrease in pulse height was observed when Xe concentration was raised from 10% to 16%. This is in agreement with Northrop and Gursky who found a maximum in pulse height for a 10% Xe in He mixture followed by a small decline with increasing Xe concentration (Figure 11).

5. The statistics of photon observation in gas scintillation counters

About  $10^5$  photons/Mev expended in a gas should be given off, however the

efficiency of the photocathode, waveshifter, reflector and window will reduce this by a factor of about  $10^3$ , thus a 5 Mev  $\alpha$ -particle results in roughly 500 photoelectrons being emitted from the photocathode. One expects a Poisson distribution of these photoelectrons about the mean number,  $m$ , hence the standard deviation,  $\sigma$ , of the distribution is  $\sqrt{m}$  and the full width at half maximum is  $2\sqrt{2 \ln 2} \sigma = 2.354 \sigma$  (Evans). Assuming that the peak of the photomultiplier voltage pulse is a linear function of the number of photoelectrons emitted by the photocathode, the resolution,  $R$ , of a pulse height distribution would be:

$$R = \frac{\text{width at half maximum}}{\text{pulse height}} = \frac{2.35\sqrt{m}}{m}$$

$$R = \frac{2.35}{\sqrt{m}} \quad \text{or} \quad m = \frac{5.52}{R^2}$$

This treatment has neglected the statistical nature of the secondary emission process which Morton (1949) estimated would increase the width less than 15%.

Maximum noise pulses in a photomultiplier tube correspond to  $\sim 8$  photoelectrons emitted from the photocathode. Using the relationship between  $m$  and  $R$ ,  $m$  was calculated for a representative number of  $\alpha$ -particle peaks. Assuming a linear relation between  $m$  and the channel number, the channel corresponding to 8 photoelectrons was calculated. There was close agreement with a resolution of 40% but with a resolution of  $\leq 20\%$  the channel corresponding to 8 photoelectrons was a factor of 2 below the maximum of the photomultiplier noise spectrum for the E.M.I. tube and 4 below for the DuMont tube.

#### 6. The counting efficiency for $\alpha$ -particles

The areas of twenty-five representative  $\alpha$ -particle spectra were measured

and converted to count rates. The count rates for a variety of gas conditions including pure and impure  $\text{He}^4$  and  $\text{He}^4 + \text{Xe}$  were found to be within  $\pm 5\%$  of the value obtained by integrating the source peak obtained with an ion chamber. Errors in plotting and measuring the curves explain this deviation, thus the gas scintillation counter has been consistently 100% efficient.

#### 7. Photon collection efficiency

The ultimate resolution attainable with a gas scintillation counter is strongly dependent on the photon collection efficiency of the photomultiplier and chamber. The  $\alpha$ -pulse height was increased by a factor of as much as 1.5 when the photocathode was optically coupled to the chamber window with a thin layer of glycerine and Russian oil.

The ratio of centre to average photocathode efficiency for the DuMont tube is 2.0 thus some variation in voltage pulses out for a constant light pulse near the photocathode is expected, however the path of photons left by charged particles and the diffusing by the walls will average the light over a larger region of the photocathode.

#### 8. The effect of $\gamma$ -radiation on gas scintillation counters

Both a thin wall chamber and a recently built thick wall chamber were intensely irradiated with 2 Mev and 6 Mev  $\gamma$ -rays ( $\sim 10^7$  photons). The resultant low energy pulses increased the count in the noise channels but had no effect on channels higher than 500 Kev on the energy scale. The shape of the  $\gamma$ -ray spectrum was similar to that of the noise pulses.

## VI. A HELIUM GAS SCINTILLATOR USED AS A DETECTOR OF FAST NEUTRONS

### 1. Types of fast neutron counters

Neutrons produce very little direct ionization in their passage through a gas, and so they cannot be detected directly by Geiger counters, ion chambers or cloud chambers whose operation depends on ions produced by the entry of a particle. Nevertheless, instruments of this kind have been adapted to detect and count neutrons by using secondary effects of these neutral particles.

The following types of detectors are successfully used for fast neutron detection. These counters have a wide range of characteristics and no one type can be chosen as superior for all fast neutron applications.

#### a) Ionization detectors

i) Boron trifluoride proportional counter ("long counter") with moderator: - This counter efficiently detects slow, intermediate and fast neutrons, but there is a considerable time delay due to moderation and diffusion effects as well as the pulse rise time of the proportional counter ( $\sim 1 \mu\text{sec}$ ). There is very good discrimination against  $\gamma$ -rays but neutron energy is not measured.

ii)  $\text{He}^3$  filled proportional counter: - This counter has been used by Batchelor et al. as a neutron spectrometer by the analysis of the pulse height distribution from the reaction  $\text{He}^3 (n,p) \text{H}^3$ . Fundamentally there is no limitation on the neutron energy or the number of neutron groups which may be detected, however above a neutron energy of 1 Mev the  $\text{He}^3$  recoil spectrum tends to obliterate the spectrum of groups from the (n,p) reaction. The quantity measured by such a counter corresponds unambiguously to one neutron energy. The rise time of the pulses obtained is limited to  $\sim 1 \mu\text{sec}$  by the electron collection time.

iii) Proton recoil counter: - The primary neutron target in such a counter may be either H<sub>2</sub> gas or a thin plastic layer. This counter may be used as a fast neutron spectrometer by the analysis of the pulse height spectrum of the knock-on protons. Unfortunately the range of energetic protons even in heavy gas mixtures makes wall effects a serious limitation, and collection time is increased if the gas pressure is raised.

b) Scintillators

i) ZnS in plastic (Hornyak Button or Sandwich Counter): - This fast neutron counter detects proton recoils from the plastic by observing the resultant ZnS scintillations with a photomultiplier.

The ZnS limits the speed of the pulses to  $\sim 3 \mu\text{sec}$ . The height of the pulses is not linear with neutron energy and the overall efficiency is low ( $\sim 10^{-3}$ ). With the correct pulse discrimination level the counter is insensitive to  $\gamma$ -rays.

ii) Organic scintillators: - Small stilbene crystals have been used as fast neutron detectors. The pulses are very fast ( $\sim 6 \text{ nsec}$ ) and the efficiency of a  $\frac{1}{2}$  cm thick crystal for 1 Mev neutrons is  $\sim 5\%$ . Plastic scintillators give slightly faster pulses than stilbene and have the added advantage that a variety of sizes and shapes is available. A combination of plastic in a liquid scintillator provides even faster pulses (2 nsec), however mounting of the scintillator is more difficult. All of these organic scintillators have a high  $\gamma$ -ray sensitivity.

c) Time-of-flight selection of neutron energy

The determination and selection of fast neutron energy by velocity measurement is at present the most accurate method. Measurements of neutron flux can be carried out simultaneously provided the overall efficiency of the detecting system is accurately known. A method frequently used is to

trigger a timing device with the  $\gamma$ -ray emitted in a  $(d;n, \gamma)$  reaction and to observe the neutron reaction a measured time later. If the neutron path length is known, the neutron energy may be specified within limits set by the timing device and the  $\gamma$ -ray counter. In cases where no  $\gamma$ -ray is emitted at the same time as the neutrons from the group under investigation, it is possible to achieve the same result by using a fast "chopper" in the path of the neutron beam or by electrostatically pulsing the deuteron beam rapidly on the target. Mechanical "choppers" produce monoenergetic neutron beams of up to 1 Mev energy from intense neutron sources such as reactors. A precise path length is defined by the apparatus and only neutrons which traverse this distance in a specified time emerge from the "chopper".

In general the time-of-flight methods are extremely powerful and their application is limited only by the difficulty of obtaining a precise estimate of the overall efficiency.

## 2. A $\text{He}^4$ plus Xe gas scintillation counter used as a neutron spectrometer

### a) Discussion

The absence of complex electron energy bands in a gas makes the analysis of pulse spectra from gas counters more simple in principle than those from solid scintillators. The gas counter pulses should be more accurately proportional to energy expended than pulses from the solid scintillators but evidence on this point is not yet clear. Northrop (1956) found that the pulse height in a Xe gas counter varied linearly with energy but the graph published shows an energy intercept of 0.5 Mev. Further investigation of the pulse height in  $\text{He}^4$  or  $\text{He}^4$  plus Xe under many different conditions could be carried out by exposure of the gas counter to neutrons of various energies.

At the same time, the properties of the gas scintillator as a fast neutron detector are worthy of examination. The choice of a gas to provide suitable recoils is important. Hydrogen would be preferable for purposes of pulse analysis because the n-p scattering is isotropic in the centre-of-mass system at moderate neutron energies. This results in an ideal spectrum from a single neutron group which is rectangular with a sharp upper limit at the incident neutron energy. On the other hand, the long range of proton recoils causes pronounced wall effects. A further limitation is set by the fact that hydrogen even in small quantities (<5%) reduces the light output of gas scintillators. He<sup>4</sup> recoils are much shorter for the same neutron energy, but in the 1 Mev - 5 Mev region the scattering is markedly anisotropic (Adair 1952), with the effect that the ideal pulse spectrum is peaked both at zero energy and at the maximum recoil energy ( $16/25 E_n$  as shown below).

The maximum transfer of neutron energy in a single elastic collision may be derived most easily by considering the collision in the centre of mass coordinates. The resultant laboratory expression is:

$$E_{\max} = \frac{4 M_n M_x}{(M_n + M_x)^2} E_n$$

where  $E_{\max}$  is the maximum energy of the recoil particle.

$M_x$  is the mass of the recoil particle

$M_n$  is the mass of the incident neutron

$E_n$  is the initial energy of the neutron

A gas scintillation counter filled with He plus Xe is a suitable counter for observing the recoil spectrum produced by neutrons. The pulses approach the speed of those from the organic scintillators while the He recoil spectrum maintains the relative simplicity of a gas counter.

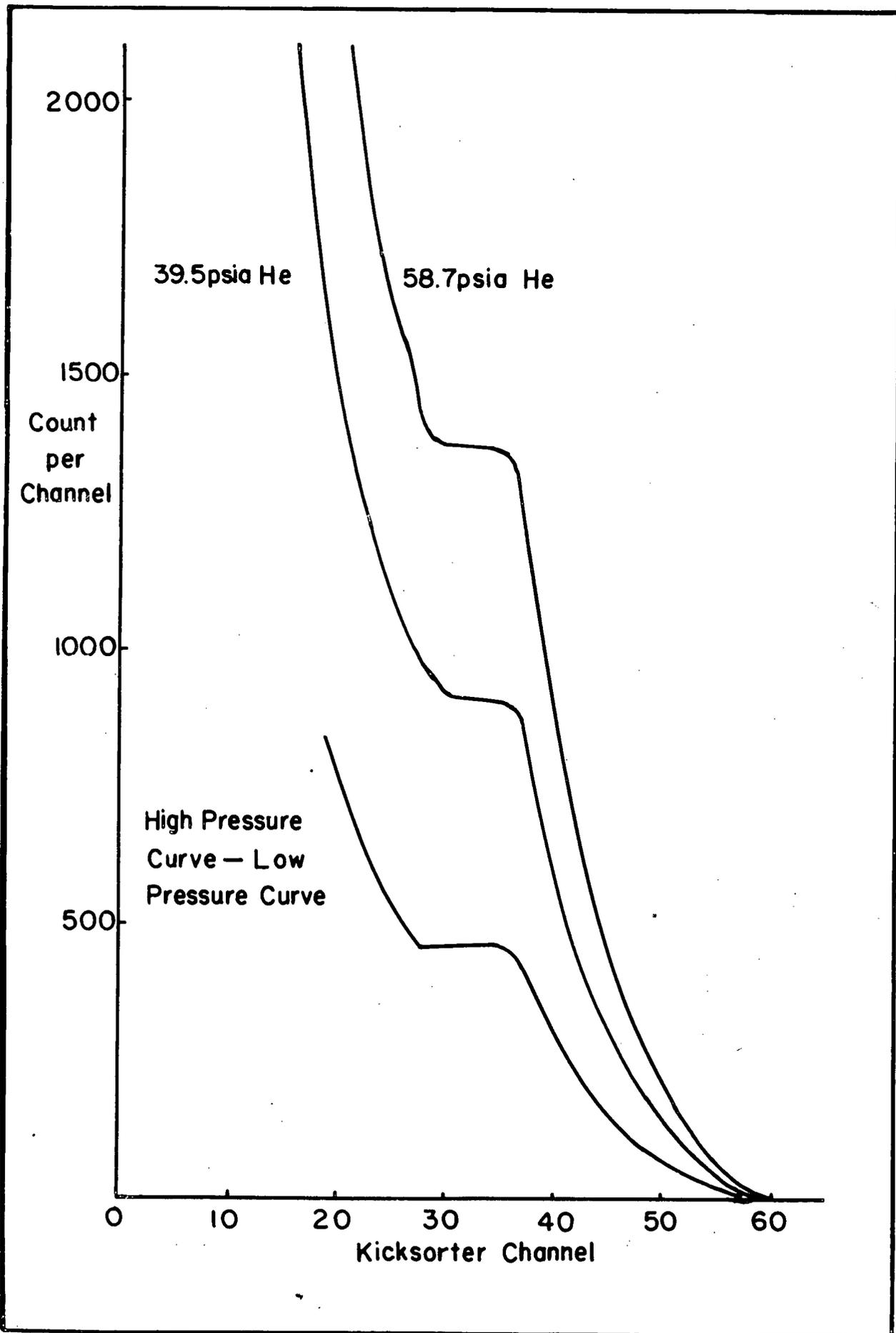


Figure 12. The Spectra of 4.1 Mev Uncollimated Neutrons at Two Helium Pressures

Almost complete insensitivity to  $\gamma$ -rays and the ability to vary the gas pressure without affecting the pulse rise time are further advantages.

The basic difficulty in operating such a counter lies in the presence of recoil protons and  $C^{12}$  nuclei from the waveshifter. In principle these can be eliminated by operating the counter at two He pressures then subtracting the low pressure spectrum from the high pressure one. The background pulses plus the photomultiplier noise will be equal for both spectra and thus will cancel. The subtraction process can succeed only if all wall effects and spatial distribution effects are absent from the recoil pulses obtained.

b) Neutron spectra obtained with a 5" diameter He plus Xe filled gas scintillation counter

The spectra shown in Figure 12 were obtained by irradiating a gas scintillation counter with uncollimated 4.1 Mev neutrons obtained by bombarding a 30 Kev thick heavy-ice target with a total of 1,060  $\mu$  coulombs of 1 Mev deuterons accelerated by the Van de Graaff generator. Initially the counter was filled to 39.5 psia  $He^4$  and 6 psia Xe (15%). Immediately after obtaining a number of low pressure spectra the Helium pressure was increased to 58.7 psia (10% Xe). A preliminary calibration with the 5.3 Mev  $Po^{210}$   $\alpha$ -source "on" established one point (Channel 86  $\equiv$  5.3 Mev) on the energy scale.

Assuming proportionality, the points on an energy scale corresponding to the maximum energy of recoil protons, Helium atoms, Carbon atoms and Oxygen atoms were found to be:

$H^1$  max = 4.1 Mev  $\equiv$  Channel 66

$He^4$  max = 2.6 Mev  $\equiv$  Channel 42

$C^{12}$  max = 1.2 Mev  $\equiv$  Channel 18

$O^{16}$  max = 0.91 Mev  $\equiv$  Channel 14

It can be seen from Figure 12 that the high energy tail caused by protons from the waveshifter layer produce maximum pulses in Channel 60 for both the high and the low helium pressure. A similar curve obtained with collimated neutrons showed the same maximum proton energy. The range of protons in the Xe plus He mixture was calculated to be  $\sim 13\frac{1}{2}$  cm whereas the chamber length is  $16\frac{1}{2}$  cm, thus protons up to Channel 66 should be observed. This discrepancy has not been resolved.

The peak effect of the  $\text{He}^4$  recoils is expected in Channel 42 whereas the sharp upper edge (knee) of the observed  $\text{He}^4$  recoils is in Channel 37. It is of interest to note that if a 0.5 Mev energy intercept is assumed as in Northrop's paper, the knee of the  $\text{He}^4$  recoils should occur in Channel 38.

The expected rise of the curve at low energy from  $\text{C}^{12}$  recoils occurs in a higher channel than predicted. This effect may be caused by a large number of low energy  $\text{He}^4$  recoils produced by wall effects.

The subtracted curve resulting from the two spectra is also shown in Figure 12. Neither proton subtraction nor carbon subtraction is satisfactory. A small shift in the observed  $\text{Po}^{210}$  pulse height, the lack of neutron collimation, probable wall effects and the unexplained poor agreement in the  $\text{C}^{12}$  recoil channels gave inconclusive results.

#### c) Counter efficiency

No measure of the neutron efficiency could be extracted from these preliminary results, however, considering a 20 cm long gas chamber and using a scattering cross section of 3 barns for 4.1 Mev neutrons, an efficiency of 0.15% per atmosphere of  $\text{He}^4$  was calculated.

#### d) Conclusion

An attempt has been made to analyze the pulse spectrum produced in the He plus Xe gas scintillator by an incident neutron beam of 4.1 Mev energy.

The results show clearly the presence of recoil protons and  $C^{12}$  nuclei as well as recoil  $He^4$  nuclei but the maximum energy of the latter is clearly visible in the pulse spectrum. Further work is required to test the proportionality of pulse height with energy in the counter before accurate neutron energy determinations are possible. Also gas mixtures of higher stopping power are needed to test the idea of subtracting a low He pressure spectrum from a high pressure spectrum to find the  $He^4$  recoil spectrum alone. If these difficulties can be overcome, the He gas scintillator should prove a valuable type of fast neutron detector.

CONSTRUCTION AND OPERATION OF AN-UNGRIDDED ELECTRON CHAMBER

To obtain an accurate measurement of the thickness of calibration alpha-sources used with the gas scintillation chambers, an argon-filled ungridded electron chamber was constructed. The chamber was designed to be rigidly connected to a Dynatron type 1430-A pre-amplifier, serial 1208.

a) The size of the chamber

To facilitate construction, the chamber was cylindrical with the source central on one end and the collector at the other. Five thin equipotential rings were mounted coaxially near the side walls to insure a uniform field gradient throughout the sensitive volume (see Plate 3).

At least 20 psia argon is used in the chamber to eliminate the inflow of contaminant gases, hence the required size of the sensitive volume is determined by the range of  $\sim 5$  Mev alpha-particles in Argon at 20 psia. From Figure 1 this range is 2.9 cm. The inside radius of the equipotential discs was chosen large enough to allow a small decrease in argon pressure without introducing wall effects. An additional  $\frac{3}{4}$ " was added to the  $\alpha$ -particle range for the distance from the end plate to the collector, because the source-to-collector distance is dependent on the thickness of the source mount.

b) The mechanical construction of the chamber

Four-inch diameter thin wall brass tubing was used for exterior side walls, and interior dimensions were adjusted to allow a press fit of the equipotential ring assembly. Electrical contact with the wire from the Kovar seal was obtained by tinning both surfaces and heating the metal plate from above when the unit was in position. A similar electrical connection was made on the collector.

Before placing the equipotential assembly in position, a resistor-and-bypass-capacitor chain was soldered between the electrodes near their outer edge so that the field lines in the sensitive volume were not disturbed.

The exterior part of the Kovar seals passed through holes cut in the end of the Dynatron pre-amplifier chassis and the chamber was rigidly screwed onto the amplifier chassis. In this way the lead from the collector to the input was kept very short.

c) The electrical behaviour of the chamber

The collector was connected to the amplifier input terminal and the equipotential lead to the terminal of the first capacitor of the dc line filter. This procedure isolated the collector from the equipotential plates, and also placed a 10 Megohm resistor in series with the bleeder resistors inside the chamber.

Ten-Megohm bleeder resistors were chosen to bias the equipotential discs in order that only a small fraction of the high tension voltage would be lost across the 10 Megohm resistor in the pre-amplifier. It was found necessary to bypass the resistors with 0.02  $\mu$ farad capacitors to eliminate bleeder noise which was fed to the plate by capacity coupling within the chamber.

With a typical collimated alpha source mounted in the chamber the characteristics of the voltage pulses from the Dynatron type 1430-A amplifier are shown in Table 3.

Table 3  
Pulse Characteristics

Argon Purity	High Tension	Amplifier		Pulse	
		Integration	Differentiation	Rise Time	Fall Time
Gas flushed through chamber 4 times	500 volts	0.08 $\mu$ sec	250 $\mu$ sec	2.0 $\mu$ sec	100 $\mu$ sec
Gas flushed twice chamber dirty	500 volts	0.08 $\mu$ sec	250 $\mu$ sec	>50 $\mu$ sec	-

The amplifier rise and fall times should be equal, and close to the rise time of the input pulse from the ion chamber (Gillespie 1953). The optimum

available amplifier settings for use with this chamber are thus: 3.2  $\mu$ sec integration time constant and  $\geq 3.2$   $\mu$ sec differentiation time constant. Amplifier pulses are fed through the inverter to the 100 channel C.D.C. kicksorter to obtain the energy resolution of the source. To properly activate the kicksorter the pulses must rise in less than 4  $\mu$ sec. This restriction is met with the above amplifier settings provided the argon is not dirty.

Increasing the high tension from 500 to 1000 volts caused a distortion in the pulse height distribution probably due to fringing fields near the collector. This effect is shown in Figure 6 . Because the chamber is ungridded, there will be a low energy tail on all curves. An exact measure of this effect has not been made, however it adds less than one channel in three to the peak width at half maximum. A more accurate resolution may be obtained by drawing a curve symmetrical with respect to the peak and the high energy tail as shown dotted in Figure 6 . 6

#### d) Summary

The ungridded electron chamber described above may be used to measure  $\sim 5$  Mev alpha source resolutions to within + 20%, -5%. Allowance must be made for the low energy tail due to the lack of a grid.

The chamber is filled by flushing it several times with tank argon, pumping it down with a small rotary vacuum pump, and then filling to  $\geq 20$  psia argon. The pulse height should remain constant for several hours, and the pulse rise time at the amplifier output should be less than 3.5  $\mu$ sec with Dynatron amplifier settings of 3.2  $\mu$ sec and high tension 500 volts. Pulses may be displayed on the 100 channel C.D.C. kicksorter to obtain accurate resolution measurements.

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