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EVALUATION OF A HIGH PRESSURE
PROPORTIONAL COUNTER FOR THE DETECTION
OF RADIOACTIVE NOBLE GASES

THESIS

Roy R. Hedtke
Major, USA

AFIT/GNE/PH/86M-7

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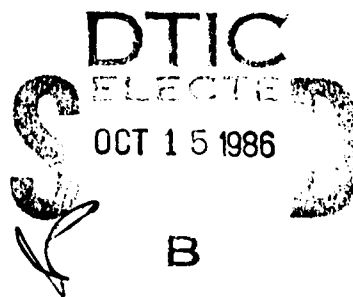
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EVALUATION OF A HIGH PRESSURE PROPORTIONAL COUNTER
FOR THE DETECTION OF RADIOACTIVE NOBLE GASES

THESIS

Presented to the Faculty of the School of Engineering
of the Air Force Institute of Technology

Air University

In Partial Fulfillment of the
Requirements for the Degree of
Master of Science in Nuclear Engineering

Roy R. Hedtke, B.S.

Major, USA

March 1986

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Preface

This thesis continues the work of three previous master's students in an attempt to find a high pressure proportional counter which is capable of analyzing radioactive noble gas samples. Although this system did not accomplish all that I had expected, the test results did indicate that it has the potential to make the desired analysis of radioactive xenon. The entire experiment was valuable from the standpoint of what I learned and what I was able to accomplish on my own.

I am very grateful to Dr. George John, my advisor, for his support and confidence in my abilities. I would especially like to thank Bob Hendricks, the physics lab assistant, for his help, his moral support, and his patience.

Finally, I want to express my heartfelt gratitude to my wife, Kathy, for her continued support; and to my children; Traci, Chris, and Jonathon, for their patience and understanding during my absence.

— Roy R. Hedtke
Major, U.S. Army



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Abstract

→ The performance of a modified high pressure proportional counter to detect and resolve ^{131m}Xe and ^{133}Xe in a gaseous mixture is evaluated. Modifications to the previous system included the installation of field tubes in the detector, and the use of titanium and zirconium turnings in the purification system.

Three external sources were used to determine operational characteristics of the detector, ^{241}Am , ^{109}Cd , and ^{57}Co . The fill gases used were pure argon and pure xenon at pressures of 5, 10, 20 and 50 atmospheres. Optimum resolutions were measured, as a function of anode voltage, for each fill gas at the various pressures tested.

Resolutions, with argon fill gas, varied from 3.56 keV for the 60 keV peak of ^{241}Am to 30.2 keV for the 88 keV peak of ^{109}Cd . Resolutions with xenon as the fill gas ranged from 4.4 keV for the 60 keV peak of ^{241}Am to 22.3 keV for the 122 keV peak of ^{57}Co . Efficiencies in argon were approximately 1 percent and they varied for xenon from 1 to 45 percent. An internal source consisting of ^{133}Xe , ^{131m}Xe , and stable xenon was used in the final phase of the study.

The system modifications functioned well; however, the ultimate goal, of resolving the xenon radionuclides,

was not conclusively achieved. Some experimental results indicated the presence of the ^{131m}Xe in the ^{133}Xe gas, however, more research is necessary to confirm this capability.

EVALUATION OF A HIGH PRESSURE PROPORTIONAL COUNTER
FOR THE DETECTION OF RADIOACTIVE NOBLE GASES

I. Introduction

Purpose

The purpose of this study is to test and evaluate a high pressure proportional counter previously studied by Lucyshyn, as part of his Master's Thesis, and further modified by George John. Evaluations of the detector's operating characteristics include studies of resolution, efficiency, and stability, using different fill gases at various pressures. Finally, it had to be determined whether or not the detector could be used to qualitatively and quantitatively determine the amounts of xenon-133 and xenon-131m in a gaseous mixture, especially when the xenon-131m is the lesser constituent by one or more orders of magnitude.

Goals

Inherent in the stated purpose are the following goals:

1. Test the detector, with its modifications, to determine utility in resolving and analyzing mixtures of two radionuclides of xenon.
2. Determine detector resolution as a function of voltage, pressure, and fill gas.

3. Determine the detector's sensitivity for detection, i.e., what is the lowest level of radiation that can be detected and what is the lowest activity which can be analyzed.

4. Determine the applicability of this type detector, and these procedures, for "production-type" operations.

5. Determine the results of the modifications to the detector.

Background

Radioactive noble gases, which are normally found naturally and in low abundances, can be released into the atmosphere in larger, more significant quantities, by nuclear reactors, detonations of nuclear devices, and processing plants for spent nuclear fuels. Of particular concern are the radioisotopes of xenon and krypton. Any detector, which can detect and accurately identify small concentrations/low activities of these noble gas radio-nuclides, would be extremely beneficial in the areas of licensing and monitoring nuclear facilities. Additionally, such a device would have many applications for environmental studies.

Scintillation and semiconductor detectors have both been used effectively in these studies. In general, scintillators have excellent efficiency but have poorer resolution (5 to 10 percent for gamma spectroscopy). In

comparison, the semiconductor detector has better resolution but has a lower efficiency. The use of proportional counters attempts to take advantage of its good resolution and a higher efficiency (because an internal source can be used).

Several prior Master's students have attempted to build and test detection systems which could accomplish the goals of this study. The earliest investigators explored the use of a Si(Li) semiconductor to analyze radioactive xenon (Rowe, Hunt, and Andrews) (16:4-5). This detector had good resolution but was plagued with poor sensitivity and problems with self-absorption of the internal conversion electrons. This system was successful in detecting xenon-131m in the presence of xenon-133. Typical resolutions and efficiencies were, respectively: approximately 0.8 keV and 0.5 percent for the 81 keV gamma photopeak of xenon-133; approximately 0.6 keV and 5.0 percent for the 30 keV x-ray of xenon; and between 6 and 10 keV and 0.55 percent for the 129 to 159 keV internal conversion electron peaks (1:45-49).

Studies of a liquified noble gas ionization chamber were then made by Gibbons and Berggren. They encountered problems with electronegative impurities and recombination in the liquified carrier gas which resulted in poor resolution of the radiation from the xenon radionuclides, xenon-133 and xenon-131m (18:1). Typical resolutions and

efficiencies were, respectively, on the order of 27 keV and approximately 70 percent for the americium-241 full energy peak (60 keV) (7:35-42).

Finally, a suggestion to use a high pressure proportional counter to increase resolution, while maintaining efficiency, resulted in the first design of a detector which led to the current detector. Although the proportional counter cannot equal the semiconductor for overall resolution, it does offer good resolution and high intrinsic efficiency for internal sources of higher pressures. Higher pressures serve to increase the absorption of photons within the detector's sensitive volume and improve the confinement of charged particles. In addition, proportional counters are well suited to the examination of lower-energy spectra. A gaseous source can be used, thus removing source mount difficulty; the solid angle is usually large, 2π or 4π , and weak activities can be measured. Finally, the background can be reduced to very low limits (21:323).

Three prior Master's students have worked with high pressure proportional counters in an attempt to resolve radioactive noble gases: Knapp, Lackey, and Lucyshyn. Each individual has offered contributions in the development of a detector which has, ultimately, led to the one which was evaluated. Problems encountered by one researcher were usually corrected by his successor. The last experimenter,

Lucyshyn, was unable to run tests using radioactive xenon because of major problems with the purification system and end effects inside the detector (18:52).

Modifications have been added to the proportional counter which are designed to solve these problems. Field tubes were installed inside the detector and the purification tube has been replaced by one with a mixture of titanium and zirconium turnings. This system was used for further study and research.

Scope

The ultimate goal of this study was to determine, once and for all, the feasibility of using a high pressure proportional counter to detect and resolve specific mixtures of xenon radionuclides. Argon and xenon were used as the principal detection media. High pressures of 5 to 50 atmospheres were used with external sources of americium-241, cadmium-109, and cobalt-57. Finally, a sample of radioactive xenon, containing xenon-133 and xenon-131m mixed with stable xenon, was used as an internal source in an attempt to quantitatively and qualitatively differentiate the two xenon radionuclides.

Assumptions Used

During this study, Diethorn's relationship for determining the gas amplification was assumed to be valid for the voltages and pressure ranges used. There is

evidence that this relationship is not valid for the highest pressures and voltages (25:234-235).

Approach

Since this project was a follow-on thesis, the general approach was to follow the same plan as Lucyshyn, i.e., attempting to repeat his procedures and calculations, in order to produce similar results. The modifications to the system made it possible to conduct tests in the areas in which Lucyshyn was unable to test.

The first step was to retrace Lucyshyn's steps in setting up the system and calibrating the equipment. The testing conducted during this portion was accomplished using P-10 gas as the detecting media at a pressure of one atmosphere.

The second phase of the research involved collecting spectra and determining system operating characteristics using pure argon as a fill gas. The detector was operated at pressures of 5, 20 and 50 atmospheres. The external sources were used that were mentioned previously.

The third phase of the study involved the use of xenon as a carrier gas. Initially, grade 3 xenon (99.9 percent pure) was used, and later, grade 4.5 xenon (99.995 percent pure) was used with the external sources to characterize the detector. Pressures of 5, 10 and 20 atmospheres were used with the xenon.

During the final stage, the xenon fill gas was replaced with a mixture of xenon-133, xenon-131m, and stable xenon. The detector was tested at pressures of 5, 10 and 20 atmospheres.

Plan of the Report

The sequence of this report is as follows: Chapter II contains a discussion of the principles of proportional counters and the theory related to their operating characteristics. The characteristics of the noble gas source and the external sources are presented in Chapter III. Chapter IV contains a description of the equipment and briefly discusses the procedures involved with each item. The results of the study and their analysis is discussed in Chapter V. Finally, Chapter VI contains the conclusions drawn plus recommendations for future work.

II. Theory of the Proportional Counter

General

A proportional counter is a type of gas-filled chamber in which the output pulse is proportional to the energy deposited by the incoming radiation. Radiation entering the detector interacts with the fill gas, ionizing it, producing charge carriers. These negative and positive ions drift toward the detector anode and cathode, respectively, and are collected to produce electric pulses. Gas filled detectors, in general, and specifically the proportional counter, usually operate in the pulse mode; that is, they produce an output pulse for each particle that ionizes the gas. Several different types of operation result as the voltages applied to the detector is varied. At very low potentials, the charge collected is less than the charge produced by the ionizing radiation because of recombination. When minimum voltage is applied to collect all of the ions, the detector operates as an ionization chamber in the ion-saturation region if additional increases in voltage do not increase the pulse height. As the voltage is raised further the counter pulse increases in size because the collection process produces additional ionizations and the pulse amplitude remains proportional to the original number of ion pairs created by the incident

radiation. This is the region of true proportionality. As voltage is increased even further, the region of limited proportionality begins. In this region the pulse amplitude still increases with increasing number of initial ion pairs, but effects such as space charge cause this increase to be nonlinear with respect to energy deposited. Finally, when voltage is increased to the point where pulse size is independent of the amount of initial ionization produced in the counter, no information as to the nature of the primary radiation can be obtained purely from pulse size measurements. This is called the Geiger-Mueller region of operation (6:4). The detector used in this study was designed to be used in the region of true proportionality.

Gas Multiplication

A proportional counter normally produces electric signals several orders of magnitude larger than the signals caused by the initial ion pairs. This process is called gas multiplication. The original ionization of the fill gas is intensified as the electrons move through the fill gas in an electric field with sufficient intensity to enable them to create secondary ions through collision. The secondary electrons are also accelerated and cause additional ionizations. This process of cascading electrons is called the Townsend avalanche. In this manner, the pulse height can be increased considerably, by up to

10^5 times, without loss of proportionality (21:303).

Ideally, the multiplication effect will be independent of where the original ion pairs are formed inside the detector.

The original number of ion pairs, n_o , is determined from the energy deposited in the absorbing material (here the fill gas) and from the average energy required to produce an ion pair. Then:

$$n_o = E_{dep}/W \quad (1)$$

where

n_o = original number of ion pairs created

E_{dep} = energy deposited in the detector (eV)

W = average energy required to produce an ion pair (eV/ion pair)

The resultant pulse height, out of the detector, is proportional to the energy deposited as follows:

$$PH = n_o * e * M * G \text{ (total)} \quad (2)$$

where

PH = pulse height (volts)

n_o = original number of ion pairs

e = electronic charge (coulombs)

M = gas multiplication factor

G = gain of the amplifier(s) (volts/coulomb)

If the detector is operated in the proportional region and the amplifier gain is fixed, then the gas

multiplication factor, M, will be proportional to the strength of the electric field. Several individuals have derived quantitative expressions for the value of M. Of these expressions, two of the more widely accepted are the Diethorn and Zastawny relationships (10:309). According to the Diethorn relationship (14:193):

$$\ln M = \frac{V}{\ln(b/a)} * \frac{\ln 2}{\Delta V} * \left[\ln \frac{V}{P a \ln(b/a)} - K \right] \quad (3)$$

where

M = gas multiplication factor

V = applied voltage (volts)

b = cathode radius (cm)

a = anode radius (cm)

P = gas pressure (atmospheres)

ΔV = a constant, unique to each fill gas, which is proportional to the potential through which an electron moves between ionizing events (eV)

K = minimum value of the electric field, divided by the pressure, below which multiplication cannot occur {volts/(cm-atm)}

This study was based on the assumption that the Diethorn relationship was valid over all values and pressure ranges used. There is evidence that, at higher voltages, gas multiplication increases at a slightly higher rate than predicted by Diethorn (9:207). When the other values are known, the gas amplification factor can be determined from either Equation (2) or (3).

Fill Gases

The fill gas plays an important role in obtaining spectra. The gas multiplication process relies on collecting free electrons at the anode. Consequently, whatever fill gases are used should have small cross-sections for electron attachment (14:190). The noble gases xenon and argon have such small cross-sections and therefore, are widely used. Argon is more frequently used because of its natural abundance and lower cost. However, xenon is a heavier noble gas which offers greater stopping power and increased photon interactions, therefore, it was selected for use with the internal sources.

Gas multiplication inside the detector is a result of secondary ionization of the gas molecules or atoms by electrons. Ultraviolet photons, produced when collisions between the electrons and gas molecules result in excitation and not ionization, can cause multiple pulsing. The addition of small amounts of polyatomic gases to the fill gas will result in the absorption of the ultraviolet photons, quenching the avalanche, or inhibiting multiple pulsing. The polyatomic gas prevents the detector from behaving as a Geiger-Mueller counter.

Pure noble gases and their mixtures can be used successfully as proportional gases if the multiplication factor is kept below a value of about 100 (23:389). Beyond this value, polyatomic gases should be added.

The argon used in this study was grade 4.5 or 99.995 percent pure. Xenon was available in grade 3 (99.9 percent pure), and grade 4.5 (research purity).

Efficiency

Counter efficiency is generally defined as either absolute or intrinsic. Absolute efficiency is defined as the number of pulses recorded divided by the total number of radiation quanta emitted by the source. Intrinsic efficiency is the number of pulses recorded divided by the number of quanta which enter the sensitive volume of the detector (14:93). The intrinsic efficiency depends on such things as detector material and thickness, the energy of the radiation and, to a small extent, on the distance between the source and the detector. Intrinsic efficiency is cited more often because it has less of a geometrical dependence than does absolute efficiency. The intrinsic efficiency is calculated by (12):

$$\text{Eff}_{\text{in}} = \frac{\# \text{ counts recorded}}{f_g f_t f_i A t_c f_\tau} \quad (4)$$

where

f_g = geometry factor

f_t = transmission factor

f_i = branching ratio of the i th quanta emitted

A = activity of the source

t_c = collection time

f_T = dead time correction

This method of calculating efficiency includes the approximations that, for the photons entering the detector, the counting time is very short compared to the half life of the radionuclide. In addition, if the count rate is kept small (less than about 1000/sec), the effect of dead time, for a proportional counter, can be neglected.

Geometry Factor. The geometry factor for an external source is given by Knoll as (14:95):

$$f_g = .5[1-d/(d^2 + r^2)^{0.5}] \quad (5)$$

where

d = the distance from the detector to the source

r = radius of the circular detector surface (window)

Lucyshyn used a model for the source well. This same model and the parameters for calculating the geometry factor are accurate for this study. The geometry factor is 0.00145 ± 0.00025 (18:47).

Transmission Factor. The transmission factor (for this study) was assumed to be caused solely by absorption of the photon in the window, or well, drilled in the side of the detector case. The effects of the intervening air

were neglected. The transmission factor for photons can be calculated from the narrow beam attenuation law since we want the fraction transmitted without any interaction.

$$f_t = \text{Exp}[-\mu/\rho * \rho * x] \quad (6)$$

where

μ/ρ = the total mass absorption coefficient for iron (cm^2/gm)

ρ = the density of iron (7.86 gm/cm^3)

x = the thickness of the detector ($0.0932 \text{ cm} \pm 0.03$)

The calculated transmission factors for the detector used in this study are found in Table I.

TABLE I
TRANSMISSION FACTORS

Gamma Energy (keV)	μ/ρ (Fe) (Ref 19)	f_t
59.537	1.174	0.4232 ± 0.117
88.03	0.4941	0.6963 ± 0.08
122.06	0.2896	0.8088 ± 0.06

Theoretical Intrinsic Efficiency. A theoretical intrinsic efficiency can be calculated for a narrow beam and a comparison with experimental efficiencies can be made. The calculation computes the fraction of photons

which interact by the photoelectric effect within the detector. This theoretical intrinsic efficiency is given by (18:49):

$$\text{Eff}_T = \tau/\mu \{1 - \text{Exp}(-\mu/\rho * \rho * x)\} \quad (7)$$

where

τ = the photoelectric cross-section for the gas

μ = the total absorption cross-section for the gas

ρ = the density of the gas

x = the diameter of the detector (4.128 cm)

The factors used to calculate the theoretical efficiencies for argon and xenon are shown in Tables II and III. A comparison of theoretical and experimental intrinsic efficiencies can be found in Table XI, Chapter V.

Resolution

The resolution reflects the detector's ability to differentiate between various energies in the spectra of the incident radiation. It is normally expressed as the full width at half the maximum height (fwhm) for a given peak in the pulse height spectrum and sometimes as a percent of fwhm to the energy of the radiation producing the peak. A fundamental limitation of the resolution arises from the statistical variation in the number of electrons arriving at the anode to form the voltage pulse. There are two main contributory causes to this limitation: one is the

TABLE II
FACTORS FOR THEORETICAL EFFICIENCIES IN ARGON

*Cross-Section (Ref 19)	Gamma Energies (keV)		
	59.537	88.03	122.06
τ/ρ (cm ² /gm)	0.2690	0.0791	0.0288
μ/ρ (cm ² /gm)	0.4566	0.2371	0.1692
Pressure (Atm)	**Density (gm/cm ³)		
5	0.0081		
20	0.0308		
50	0.0851		

*These were obtained from a log-log interpolation of values.

**These were calculated using Van der Wall's equation.

TABLE III
FACTORS FOR THEORETICAL EFFICIENCIES IN XENON

*Cross-Section (Ref 19)	Gamma Energies (keV)		
	59.537	88.03	122.06
τ/ρ (cm ² /gm)	7.5727	2.5474	1.0045
μ/ρ (cm ² /gm)	7.9275	2.7814	1.1899
Pressure (Atm)	**Density (gm/cm ³)		
5	0.0256		
10	0.0491		
20	0.0916		

*These were obtained from a log-log interpolation of values.

**These were calculated using Van der Wall's equation.

variation from the mean in the number of ion pairs formed, and the other is the variation in the multiplication factor (21:305-306). The latter involves the practical design of the detector including such items as amplifier noise, anode wire uniformity, end effects, and other, less-important, effects (25:330).

Statistical Considerations. The statistical variation in the number of primary and secondary ion pairs created in the detector is the most important limitation of the proportional counter. The assumption must be made that the formation of original ion pairs, n_o , and the secondary ionization produced from gas multiplication, M , are independent of one another and governed by the same probability distribution. Then, probability theory predicts the relative variance of the charge, Q , is given by (4:31):

$$\left(\frac{\sigma_Q}{Q}\right)^2 = \left(\frac{\sigma_n}{n_o}\right)^2 + \left(\frac{\sigma_M}{M}\right)^2 \frac{1}{n_o} \quad (8)$$

where

Q = the total charge produced

n_o = the mean number of original ion pairs formed

M = the mean multiplication factor

σ_Q σ_n σ_M are the standard deviations of those quantities

The variance of the number of primary ion pairs follows the dependence $(\sigma_n/n_o)^2 = (F/n_o)$ where F is the

Fano factor, a constant which is characteristic to each fill gas. The variance in the gas amplification is obtained by representing the variance of a single electron avalanche by the Polya distribution, $b = (\sigma_A / \bar{A})^2$. If one assumes a gaussian distribution of pulse heights, then the statistical limit can be expressed by the following relationship (22:251):

$$R(\text{stat. limit}) = 2.36[(F + b) * E_{\text{dep}} * W]^{0.5} \quad (9)$$

where

R is the resolution (keV)

F is the Fano factor (constant)

b is the variance from the Polya Distribution
(constant)

E_{dep} is the energy deposited from incident radiation
(keV)

W is the average energy required to create an ion
pair (eV/ion pr)

A more detailed derivation is found in reference 14, Knoll.

Other Factors Affecting Resolution

Several factors can affect the proportionality of the multiplication process by altering the electric field inside the detector. Some of these are uniformity (and eccentricity) of the anode wire, end effects, and space charge effect.

Other processes can reduce the number of primary electrons created, thereby reducing gas amplification.

These processes involve the operating parameters of the detector such as gas purity, gas pressure, and stability of the high voltage source.

Anode Wire Uniformity. The quality of the output pulse depends, to a large extent, on the independence of the multiplication factor to the energy deposited. Each primary electron created must undergo the same amount of amplification. Nonuniformities in anode wire diameter, eccentricities of its position, or even particulates on the wire itself can alter the electric field. One calculation on the effect of nonuniformity in the anode wire indicates that, for a detector with a limiting resolution of 4 percent (2.39 keV), a variation in the anode wire diameter of 0.5 percent is sufficient to cause the resolution to deteriorate to 5 percent (2.97 keV) (25:237). For the anode wire used in this experiment (diameter = $0.0003'' \pm 2$ percent) a variation of 7.5 percent can be expected in the multiplication factor (using Diethorn's relationship). Eccentricities such as kinks in the wire or particulates adhering to the wire can cause full energy peaks to be split into two separate peaks. This phenomenon was observed during this study.

End Effects. The anode wire inside the detector is supported at the ends by tubes which are thicker than the wire itself. These supports are not a part of the

collecting electrode but they can still introduce distortion of the electric field near the ends of the wire. The two main consequences of this are: the sensitive volume of the detector appears smaller due to electric field distortion and the gas multiplication factor is no longer constant along the length of anode wire (5:37). Lucyshyn encountered both problems in his thesis (18:52-53).

Field tubes were installed around the support tubes and ends of the anode wire to remedy these effects. When the voltages on the field tube are carefully adjusted, the end effects are reduced to negligible proportions and the sensitive volume becomes well-defined (5:37-38). The field tube voltage can be calculated from the following relationship (5:38):

$$V_f = V_a \frac{\ln (r_a/r_f)}{\ln (r_c/r_a)} \quad (10)$$

where

V_f is the field tube voltage

V_a is the anode voltage

r_c is the cathode radius

r_f is the field tube radius

r_a is the anode wire radius

The efficacy of correctly adjusted field tubes is depicted in Figure 1, which shows a plot of results obtained in pulse size as a function of distance along the anode wire.

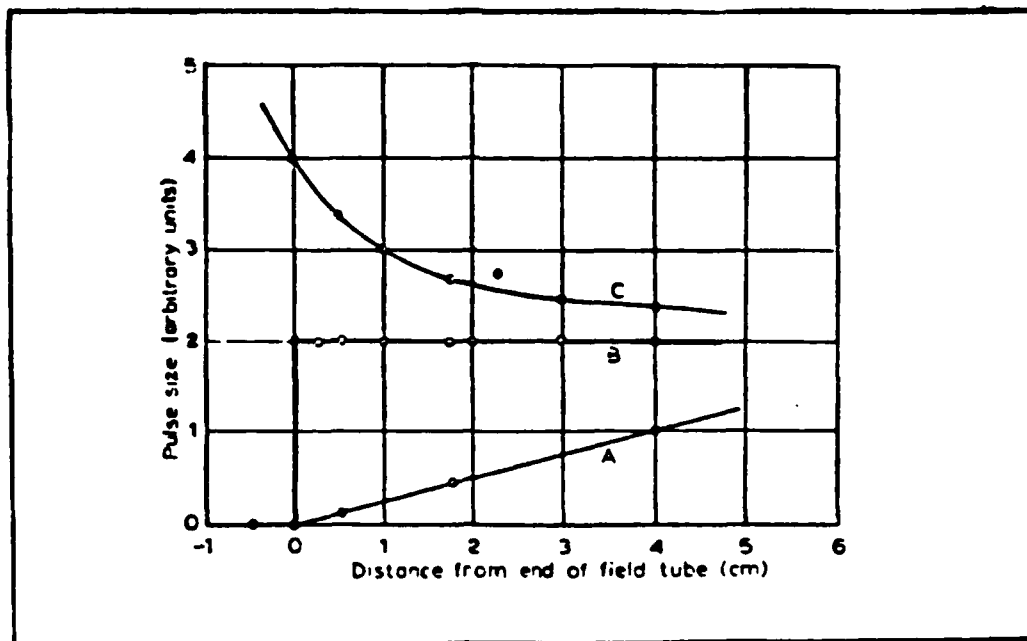


Fig. 1. Variation of Pulse Height Along the Wire. A, no end correction; B, end-correction tube at correct voltage; C, voltage 20 percent too high (21:326)

Space Charge Effects. Space charges, resulting from a buildup of positive ions, can appreciably reduce or distort the strength of the electric field at small radii. Self-induced effects are the result of a positive ion sheath from ions, formed in an avalanche, which have built up around the anode, reducing the number of electrons in other avalanches. The general space charge effect includes the results of many avalanches and produces nonlinearities in the electric field as all of these positive ions drift towards the cathode (14:196).

Electronegative Impurities. Electronegative impurities become especially important when using pure fill gases at higher pressures. These impurities are either present in the fill gas during the experiment, they degas from

epoxies and other materials, or they emanate from micro-cracks inside the detector. The most important impurities with a high affinity for electron capture are water vapor, oxygen, and nitrogen (8:289). The basic effect of impurities is to lower the overall charge collected. This can occur in one of two ways: the electronegative molecule can capture primary electrons, thus reducing the charge collected, or they can slow down the electron drift velocity. The effect of slowing down the drift velocity results in those electrons near the cathode being captured by electronegative molecules or lost to recombination (20:364,366). As the pressure inside the detector is increased, the effects of impurities become more pronounced. Therefore, special attention must be given to purity of gases, purification systems, etc., to prevent impurities from reducing the gas multiplication.

Lesser Factors. Any variable contained in the Diethorn relationship indicates a potential way in which gas multiplication can be reduced, thereby degrading resolution. Knoll discusses some of the detector operating parameters which can affect resolution. Small pressure changes as low as a few tenths of one percent can be significant (14:207). However, the use of a well sealed detector and higher pressures will usually solve this problem.

The gas multiplication factor is very sensitive to fluctuations in applied voltage. Again, Knoll indicates

that variations of 0.1 to 0.2 percent can seriously change gas multiplication. Today's well-regulated power supplies coupled with lower voltage requirements for thinner anode wire have helped make this problem negligible.

Purification

The effects of electronegative impurities were discussed in a previous paragraph. The three prior Master's students working with proportional counters all encountered problems with impurities (13:72; 16:98; 18:52). This subsystem is a critical component in the overall counting system, especially when using the detector at higher pressures. Lackey suggested the use of a convective flow purification system which Lucyshyn incorporated into his system. The current modification to the purification tube involved the replacement of the calcium turnings used by Lucyshyn with titanium and zirconium turnings. Titanium and zirconium have melting points of 1800 and 1700 °C, respectively (8:291). However, the quartz tube containing the titanium and zirconium has a strain point near 1070 °C, therefore, the operating temperature of the purification tube (oven) is kept below 1000 °C (16:61). Lucyshyn's purification system with its calcium turnings was operated at 600 to 650 °C. Gibbs et al. determined that the oxygen impurities were removed completely by the titanium and/or zirconium at temperatures above 600 °C. The titanium reduced the nitrogen impurities to approximately 1.0 percent at 1000 °C, and the zirconium reduced them to 0.1 percent at 1000 °C (8:295-296).

III. Characteristics of the Noble Gas and External Sources

General

This chapter includes a discussion of the production and nuclear decay data for radioactive xenon. It also presents a predicted spectrum for a mixture of radioactive gas, xenon-131m and xenon-133, and describes the source procured locally. Finally, a discussion is included concerning the external sources.

The element xenon has an atomic number of 54 and an atomic weight of 131.30 based on the carbon-12 scale (24:19,32-33). There are ten stable xenon isotopes and 27 radioactive isotopes and isomers. It is estimated that 0.5 percent of the xenon found in nature is the result of spontaneous fission, or of the neutron induced fission of uranium and thorium (13:10).

Production

Xenon and krypton are the primary noble gases produced during nuclear fission. Lucyshyn provided a table of the xenon isotopes resulting from a fission chain reaction (18:6). This table is reproduced as Table IV with updated half-lives (24:32-33).

There are only 12 isotopes created, but the only radioisotopes with half-lives greater than 48 hours are

TABLE IV
CUMULATIVE FISSION YIELDS FOR XENON (18:69; 24:32-33)

Isotope	Half-life	Fission Yield (%)
131m	11.92 days	0.017
131	Stable	2.770
132	Stable	4.130
133m	2.19 days	0.190
133	5.25 days	6.770
134	Stable	7.190
135m	15.3 min.	1.050
135	9.10 hrs.	6.720
136	Stable	6.120
137	3.84 min.	5.940
138	14.1 min.	6.240
139	40.0 sec.	4.960

xenon-131m, xenon 133m, and xenon-133. In spite of the fact that the xenon-131m has such a small fission yield, its half-life is twice as long as that of the xenon-133, and four times that of the xenon-133m. At later time, the xenon-131m will constitute a greater portion of total activity. The xenon-133m and other radioisotopes are of less concern because of their low fission yields, short half-life, or both.

Decay Data for Xenon-131m
and Xenon-133

Xenon-131m is an isomeric state of xenon-131, 163.93 keV above the ground state. It decays by internal conversion electron approximately 98 percent of the time and by gamma emission 2 percent of the time. Its decay scheme is shown in Figure 2.

Xenon-133 decays principally by beta emission (89.3 percent) to the first excited state (81 keV) of cesium-133. The excited cesium-133 then decays by internal conversion electron (63.4 percent) and by gamma ray emission (36.5 percent). The decay scheme for xenon-133 is depicted in Figure 3.

Table V contains a compilation of decay data for the two radiosotopes. It shows that the principal peaks in the pulse height spectrum for ^{131m}Xe should occur at 129.4 keV, 158.5 keV, from internal conversion electrons, and 29.8 keV from xenon K x-rays. For the ^{133}Xe inside

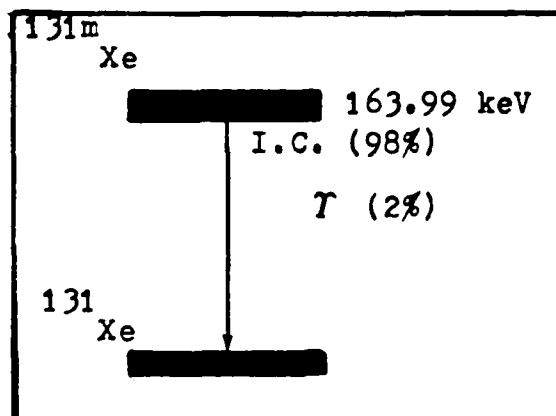


Fig. 2. Decay Scheme of ^{131m}Xe
(17:281)

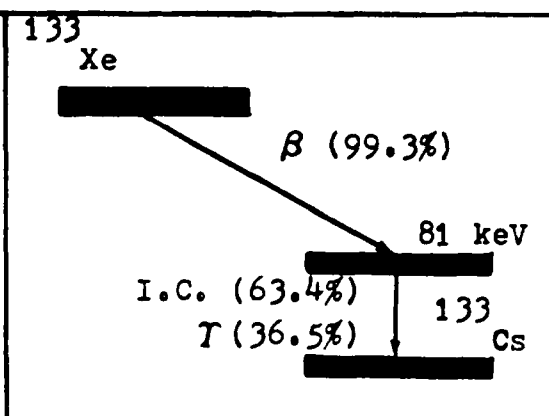


Fig. 3. Decay Scheme of ^{133}Xe

the detector we expect to see the beta continuum summed with internal conversion electrons so that the internal conversion electrons will not produce a peak. This will also occur when the 81 keV gamma ray is emitted coincident with a beta that deposits its energy in the sensitive volume of the detector.

Xenon Source Locally Procured

The radioactive xenon used in this study was obtained from the Nuclear Medicine Clinic of the Wright-Patterson Air Force Base Medical Center. Radioactive xenon is used for inhalation therapy in treatment of certain lung disorders. The source is obtained by the medical center in 2 milliliter bottles containing approximately 10 microcuries of radioactive xenon-133. Xenon-131m is contained as an impurity in this sample. A Ge(Li) semiconductor

TABLE V
CHARACTERISTIC RADIATIONS OF ^{131m}Xe AND ^{133}Xe (15:138)

Radiation Type	Energy (keV)	Fraction per Decay
^{131m}Xe		
Auger-L	3.43	0.75
Auger-K	24.6	0.068
ce-K	129.369	0.612
ce-L	158.477	0.286
ce-M	162.788	0.0650
ce-NOP	163.722	0.0178
X-ray L	4.1	0.080
X-ray K_2	29.458	0.155
X-ray K_1	29.802	0.287
X-ray K	33.644	0.102
Gamma	163.93	0.178
^{133}Xe		
Auger-L	3.55	0.497
Auger-K	25.5	0.056
ce-K	45.012	0.533
ce-L	75.283	0.081
ce-M	79.780	0.016
ce-NOP	80.766	0.004
Beta - max	346.3	0.993
avg	100.6	
X-ray L	4.29	0.061
X-ray K_2	30.625	0.136
X-ray K_1	30.973	0.253
X-ray K	35.0	0.091
Gamma	80.997	0.365

detector was then used to assay the samples and provide relative activities. Of the three sources obtained, the xenon-131m constituted between 0.4 percent and 6.2 percent of the total radioactivity of the sample depending on the sample itself and the amount of time which had elapsed. The activity of the three sources varied from approximately 5 millicuries to 5 microcuries. None of the sources was pristine. The first sample obtained was from the remains of a 10 microcurie dose vial sample which had some xenon drawn out for therapy. The second and third samples were withdrawn from a used 2 milliliter dose vial with a syringe, and injected into evacuated 10 milliliter dose vials. This process undoubtedly introduced air as an impurity. The radioactive xenon was later withdrawn from these dose vials and transferred into the detection system. This transfer equipment and this procedure will be described in Chapter IV.

Predicted Response from Radioactive Xenon

Both Knapp and Lackey predicted the spectrum from a mixture of xenon-133 and xenon-131m. Both assumed an activity ratio of approximately 50 to 1 (13:19; 16:56). Lackey assumed a 5 keV resolution for the 164 keV peak, that the most energetic beta particles (346 keV) and conversion electrons (164 keV) are stopped with 85 percent and 95 percent efficiency, respectively (for 50 atmospheres

of pressure), that less energetic electrons and x-rays are fully stopped or absorbed, and that only about 5 percent of the gamma rays are fully absorbed (16:56-57). Based on those assumptions Lackey predicted a spectrum which Lucyshyn altered by assuming that 10 keV resolution for the 164 keV peak was more realistic (18:10). This predicted spectrum is reproduced as Figure 4.

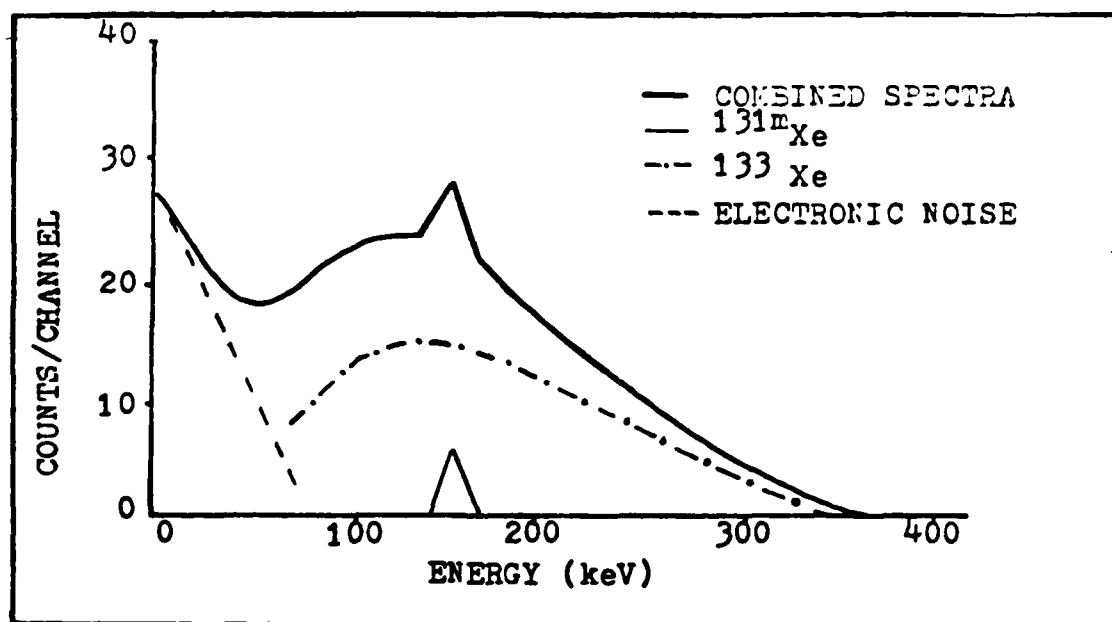


Fig. 4. Predicted Radioactive Xenon Gas Spectrum

External Sources Used

External sources, known to emit nearly monoenergetic gamma rays, are used to obtain precise energy and efficiency calibration data. The external sources in this study were americium-241, cadmium-109, and cobalt-57.

Americium-241 has a half-life of 432.2 years and decays by alpha emission to neptunium-237. Approximately 86 percent of these decays result in an excited state 59.537 keV above the ground state (17:430). Neptunium-237 returns to the ground state by gamma ray emission of this energy with a branching ratio of 0.359.

Similarly, cadmium-109, with its 453 day half-life decays by electron capture to an excited state of silver-109, 88.03 keV above the ground state. The branching ratio for emission of these characteristic silver gamma rays are 0.0361 (17:280).

Cobalt-57 decays by electron capture (99.8 percent of the time) to an excited state of iron-57, 136.42 keV above the ground state. From this state it emits two primary gamma rays, 122.06 keV with a branching ratio of 0.856, and 136.42 keV with a branching ratio of 0.106. When the 122.06 keV gamma is emitted, the iron-57 drops to a lower excited state 14.4 keV above the ground state (17:191).

These sources emit other x-rays, gamma rays, or auger electrons, but they are either unable to penetrate the detector wall or they are too energetic to be stopped inside the detector. Table VI contains the source activity data and Figure 5 depicts these sources' decay schemes.

TABLE VI
SOURCE INFORMATION

Source	Activity	Date	Uncertainty
Am	10 mCi	10 Mar 74	$\pm 10\%$
Cd	65 uCi	7 Nov 83	$\pm 10\%$
Co	97 uCi	9 Apr 84	$\pm 10\%$

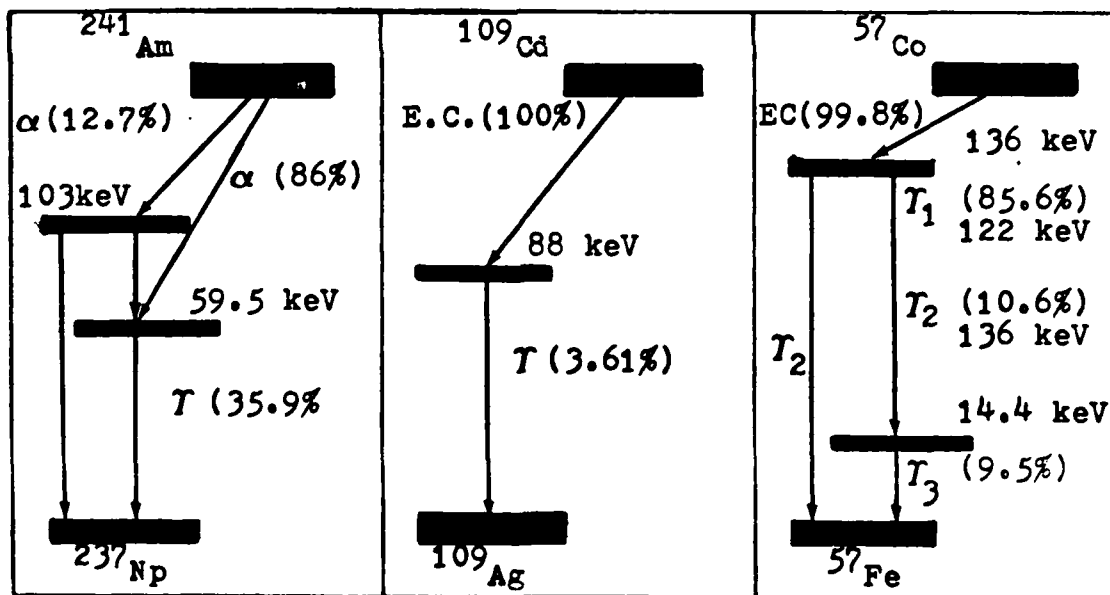


Fig. 5. External Source Decay Schemes (6)

IV. Equipment and Procedures

General

The equipment used in this experiment is identical to that used by Lucyshyn with the exception of the new purification tube and the field tubes in the detector.

This equipment can be most easily described by categorizing the counting system into three subsystems: the gas handling system, the detector itself, and the electronics. In addition, it is also necessary to describe the xenon transfer system which was used to transfer radioactive xenon gas from the dose vials into breakseal tubes for introduction into the counting system reservoir.

This chapter also presents those procedures associated with the various pieces of equipment.

Gas Handling System

The purposes of the gas handling system are: to allow for the evacuation of the proportional counter system (on the order of 10^{-7} torr), to allow for the correct amount of fill gas to be added, and finally, to allow for purification of the fill gas. This system was divided by Lucyshyn into two subsystems, the vacuum pumping section and the gas filling section (18:24-26). The vacuum pumping section consists of a forepump and a diffusion pump with a

cold trap. The forepump can evacuate the detection system to 10^{-3} torr, from which point the diffusion pump can further evacuate the system to 10^{-7} torr. Figure 6 shows a schematic of the gas handling system.

The gas filling section consists of the purification tube and a 23.6 liter gas reservoir, with sufficient inlet valves to introduce three different gases. A cold finger at the bottom of the reservoir allows the fill gas to be cryogenically pumped back into the reservoir. A Pace pressure transducer provides pressure readings from inside the reservoir and purification tube.

The purification unit consists of a quartz tube filled with titanium and zirconium turnings, which is heated by a tubular oven. Voltage is supplied to heat the oven by a DC transformer (powerstat). The purification tube is connected, by chromel-alumel thermocouples, to a potentiometer from which one obtains oven temperatures in milliamps. The milliamps can then be converted directly to degrees Celsius by a calibration table found in the Handbook of Chemistry and Physics. A more detailed description of the gas handling system can be found in Lucyshyn's thesis (18:24-26).

Gas Handling Procedures

The procedures used in the operation of the gas handling system are: charging the system with gas,

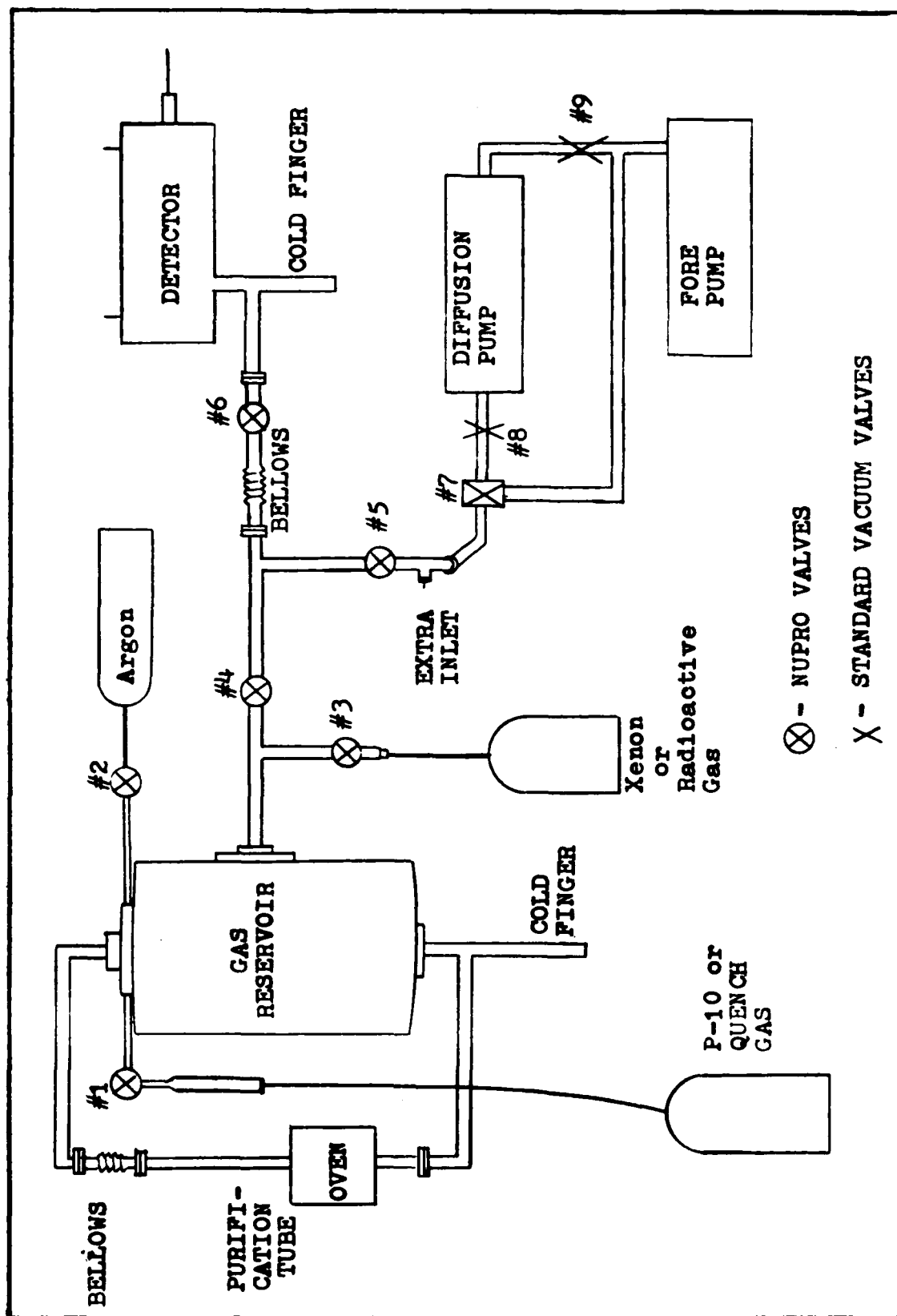


Fig. 6. The Gas-Handling System

purification of the fill gas, pressurization of the detector, and introducing a radioactive source.

Charging the System with Gas. The first required procedure is charging the gas handling system with the correct quantity of fill gas. This procedure involves three steps: evacuating the entire system, flushing the system, and injecting the fill gas into the reservoir.

The system must first be evacuated to approximately 10^{-7} torr using the forepump and the diffusion pump. Next, the system is flushed with the appropriate fill gas. The gas is introduced through one of the inlet valves to a pressure of about 100 to 200 torr. Then the system is evacuated, using only the fore pump, to a pressure below 30×10^{-3} torr. This flushing procedure is repeated three times. Finally, the fill gas is forced into the reservoir until the desired pressure is obtained. Vander Waal's equation of state is used to calculate the number of moles of fill gas required to achieve a desired pressure in the detector.

Purification. Once the system is filled with the correct amount of fill gas, the purification process can begin. Voltage is applied to the tubular oven to heat the titanium and zirconium turnings to approximately 950 °C (8:295-296). The purification system operates on the principle of convective flow. In addition, a dewar of

liquid nitrogen is placed around the reservoir cold finger to freeze the gas into the cold finger. The gas circulates as the fill gas thaws and expands, and helps to push gas through the purification tube. This process of cryogenically freezing the fill gas is accomplished three times during a 12-hour purification cycle and twice during a 3-hour cycle.

Pressurization of the Detector. As the fill gas is purifying, the detector is kept under vacuum (10^{-7} torr) until just prior to transfer of the gas. A small amount of purified gas is used to flush the evacuated detector. Before transferring the gas into the detector, the vacuum system is isolated and the fill gas is cryogenically pumped from the reservoir into the detector cold finger. When the gas condenses inside the cold finger, and the reservoir pressure drops to the vapor pressure of that particular gas (at liquid nitrogen temperatures), the detector is isolated from the remainder of the system. The vapor, which remains in the system and does not condense, is accounted for in the calculation for the number of moles of fill gas. Finally, the dewar of liquid nitrogen is slowly lowered, allowing the gas to thaw and expand into the detector at the desired pressure.

Introducing a Radioactive Source. Both external and internal radioactive sources were used during this

study. The external sources were affixed to plexiglass disks and placed over one of the three source windows (see Figure 7). Radioactive xenon gas is introduced into breakseal tubes in the xenon transfer system. The break seal tube is then attached to inlet valve #3 and a dewar of liquid nitrogen is raised over the reservoir cold finger to cryogenically pump the gas from the breakseal tube into the reservoir. After the radioactive gas is transferred, valve #3 is closed and fill gas is added. The fill gas mixture can now be purified and transferred into the detector as mentioned above.

The Detector

The detector is constructed from a stainless steel cylinder 6.25 inches long, with an outside diameter of 3.36 inches and an inside diameter of 1.625 inches. The detector is capable of withstanding pressures greater than 50 atmospheres. Inside the detector is the removable centerpiece which holds the anode wire, the support and the field tubes. The anode wire is made of 304 stainless steel and has a diameter of 0.0003 inches.

The field tubes, with an outside diameter of 0.369 inches, are also constructed of stainless steel and epoxied in place on the ceramic endpieces of the center section. Electrical contact is made through the side of the detector via narrow rods, soldered on the field tube, with spring

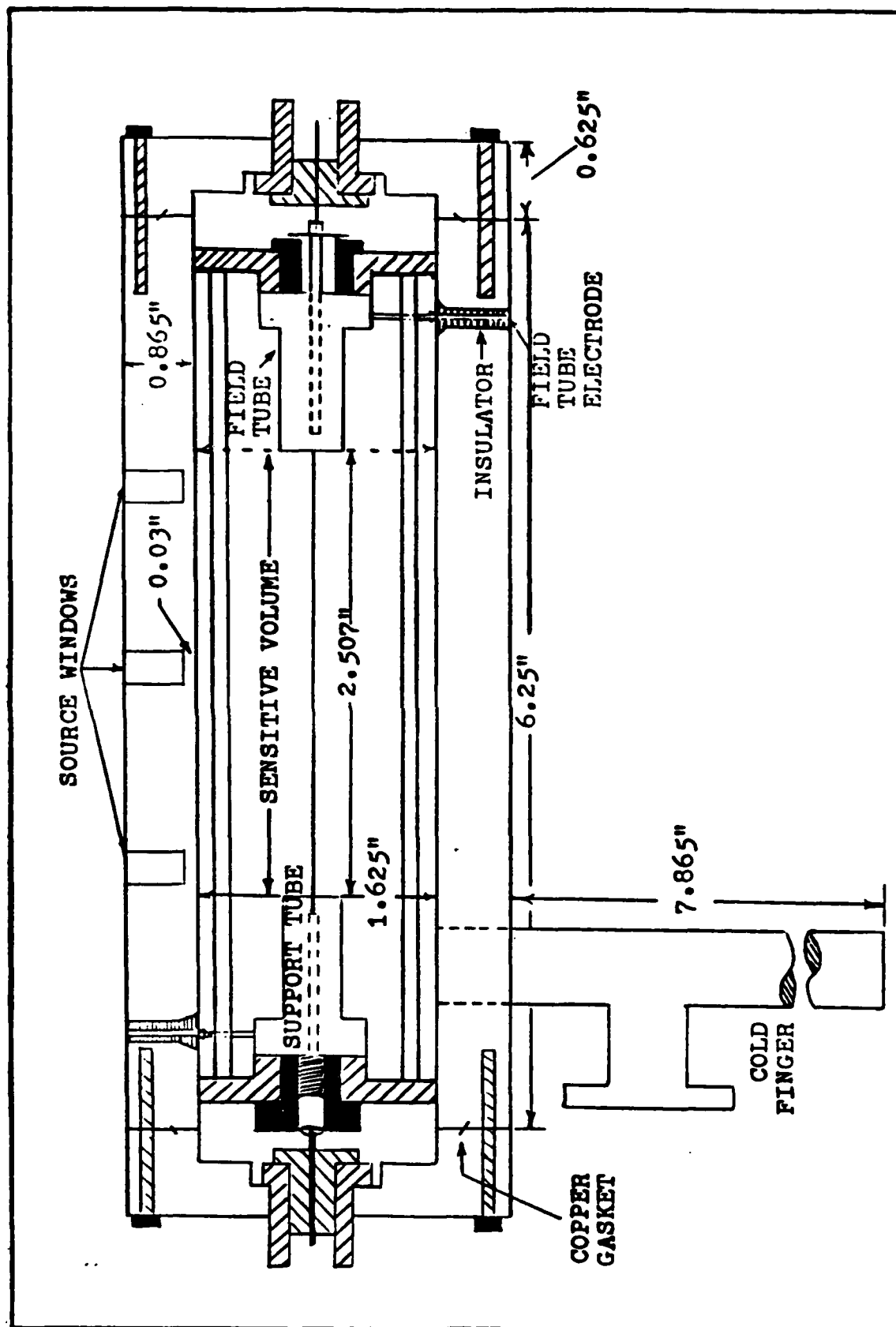


Fig. 7. The Detector

contacts on top. A locating pin on the bottom right inside of the detector ensures proper alignment of the field tube rods and the contacts. Figure 7 is a cross-sectional schematic of the detector. Figure 8 depicts the center-piece with its measurements. More detail concerning the detector can be found in Lucyshyn's thesis (18:27).

Procedures Involving the Detector

There are two specific procedures involving the detector which must be described: collecting data and changing the anode wire.

Collecting Data. After the source has been introduced and the counting gas has been transferred and expanded into the detector, the desired voltages are applied to the anode wire and the field tubes. The resultant pulses are monitored on the oscilloscope, the data is collected and analyzed in the multichannel analyzer, and the results are stored on 8-inch floppy disks.

Replacement of the Anode Wire. In the event that the anode wire becomes pitted or broken, it is necessary to replace it. This procedure had to be accomplished four times during the course of this study. The entire process can be performed in five phases: removal of the endcap(s) and centerpiece; replacement of the anode wire; glowing the anode wire in a specially made glass tube; replacing

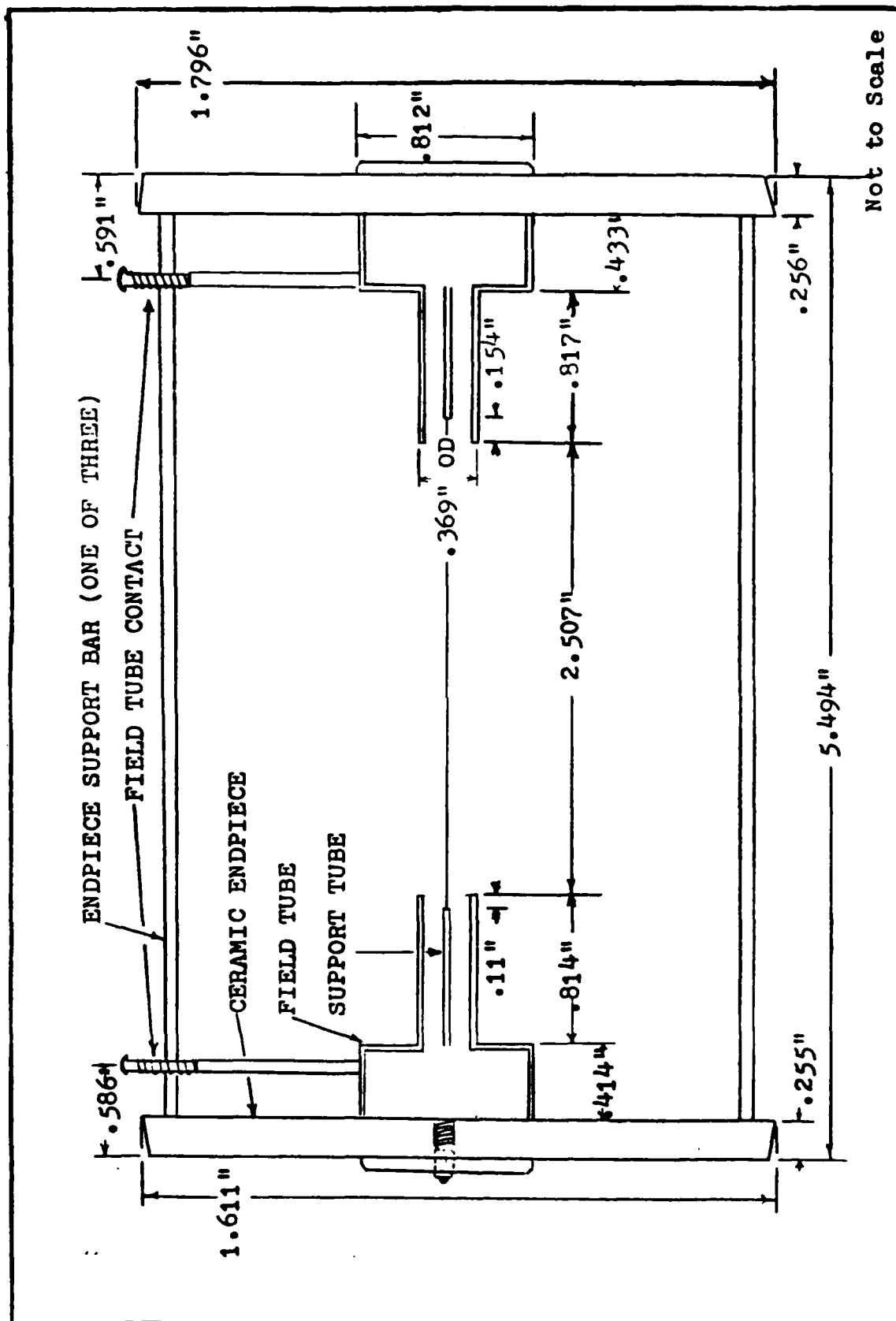


Fig. 8. Detector Centerpiece Measurements

the centerpiece and endcap(s); and finally, glowing the anode wire inside the detector. Because of the length of this procedure it will be described in Appendix B, Procedures.

The Electronics

The electronics are those generally used for pulse height analysis (see Figure 9). The high voltage power supply, which supplies the voltage bias to the anode, is a very stable, guarded power supply with a maximum output of 5000 volts. The only difference between the electronics used in this study and those used in Lucyshyn's experiment is the addition of a 3000-volt power supply for the field tubes (18:29).

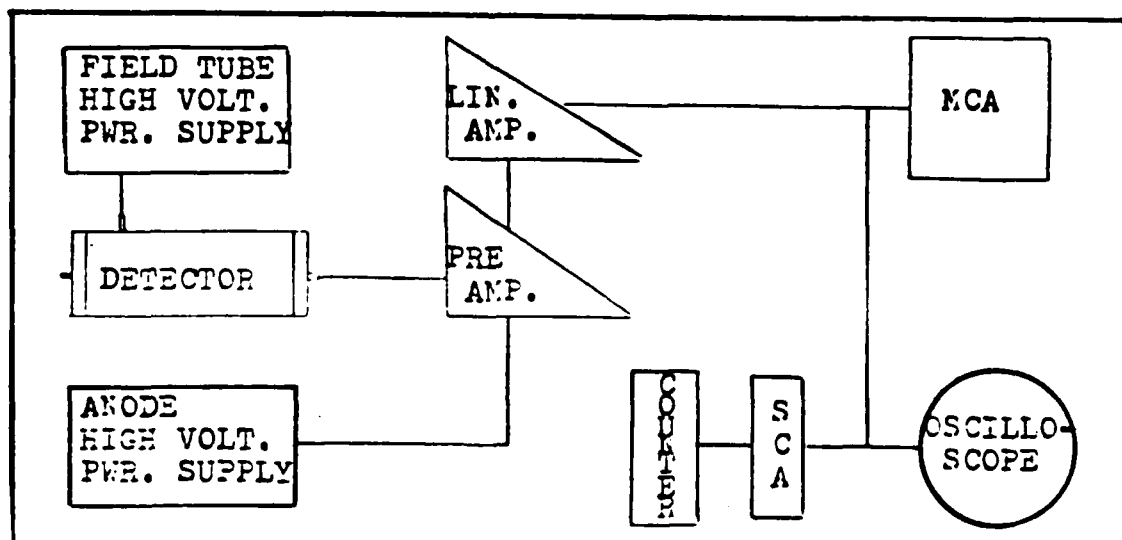


Fig. 9. The Electronics

The Xenon Transfer System

The xenon transfer system is a separate gas handling system which allows radioactive xenon to be moved from dose vials into the system, mixed as necessary, and then pumped into breakseal tubes for transfer into the counting system. The system is a complicated series of stainless steel tubes separated by Nupro valves and is depicted in Figure 10. It consists of 11 valves (V1 - V11); a sample chamber (not used in this study); fore and diffusion pumps; a pressure transducer; two cold fingers (F1 and F2); and three external connections (EC1 - 3). The first external connection (EC1) was never used and its valve (V3) was never opened. The syringe was attached at EC2 for extracting radioactive gas from dose vials. The breakseal tubes were also attached there to transfer radioactive xenon gas to the counting system. Stable xenon was introduced into the transfer system through EC3 for mixing with radioactive gas.

Xenon Transfer Procedures

There are two basic procedures with this system: extracting radioactive xenon gas from dose vials and cryogenically pumping radioactive gas mixtures into breakseal tubes for introduction into the counting system.

Extracting Radioactive Gas from Dose Vials. The dose vial must be prepared before any gas can be extracted from it. A layer of catalyzed silicon rubber is molded to

FRONT VIEW

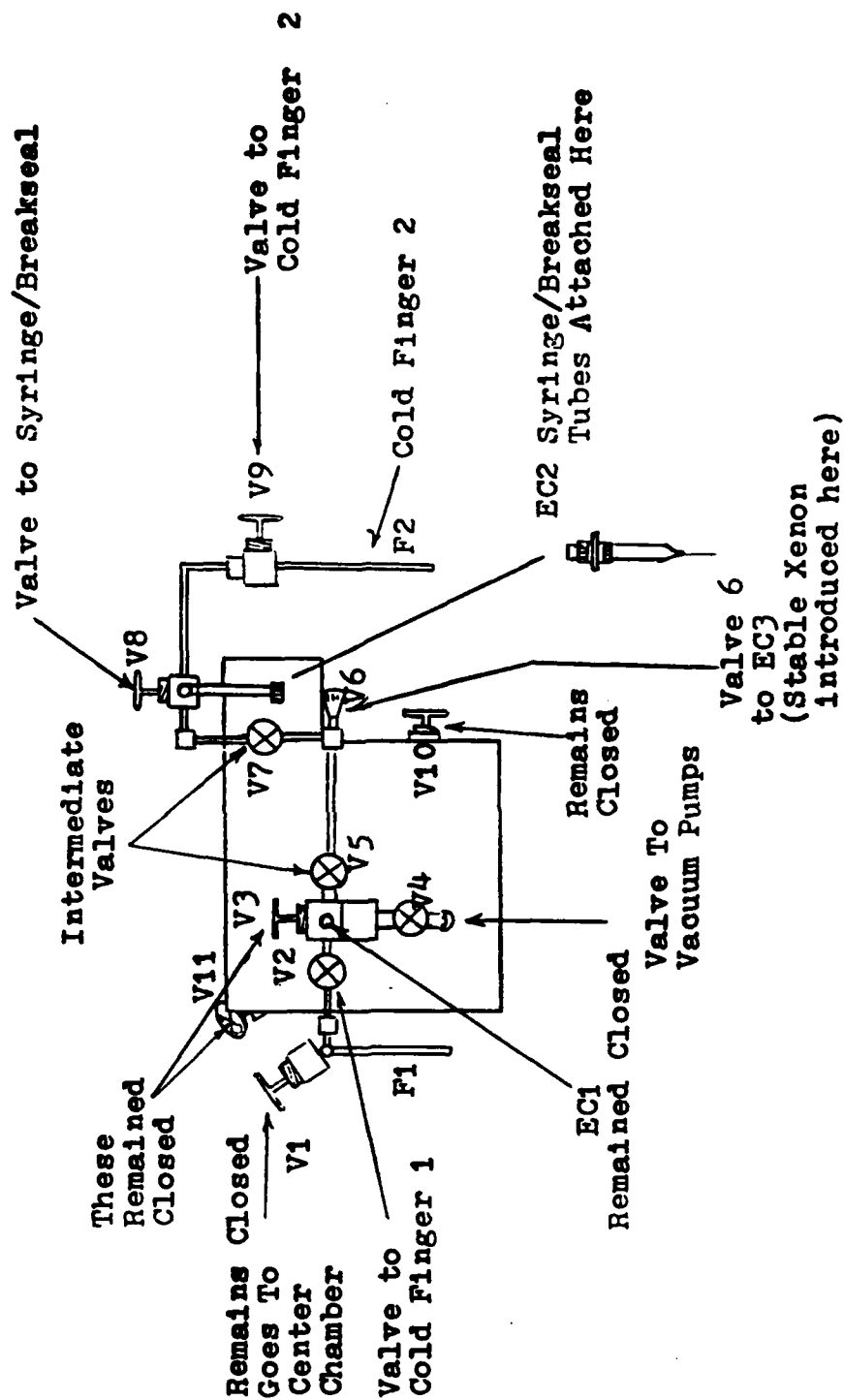


Fig. 10. Xenon Transfer System

the rubber septum on top of the dose vial. The syringe, attached at EC2, is inserted into the catalyzed silicon rubber and the system is evacuated to approximately 10^{-7} torr. A small amount of high vacuum grease should be placed at the junction of the syringe and the catalyzed rubber to prevent leaking. Once a good vacuum is obtained, valve #4 is closed, the system is isolated from the vacuum pumps, and the dose vial is forced upward until the tip of the syringe penetrates the rubber septum. Cryogenic pumping alone, on either of the cold fingers, was insufficient to extract the small amounts (5 uCi) of radioactive xenon from the dose vials. It became necessary to transfer stable xenon into the dose vial, and then cryogenically pump the mixture out in order to extract radioactive gas. By pumping the xenon gas mixture into coldfinger #1 and closing valve #2, a pressure reading can be obtained on the Pace pressure transducer indicator. Valve #8 can then be closed, the dose vial removed, and a second assay can be obtained from the dose vial. The difference indicates how much radioactivity is contained inside the xenon transfer system.

Filling the Breakseal Tube. The radioactive gas mixture must first be cryogenically pumped into one of the two cold fingers and isolated from the remainder of the system. The breakseal tube is attached at EC2 and the

system, minus the isolated cold finger, is evacuated to approximately 10^{-7} torr. A dewar of liquid nitrogen is slowly raised over the breakseal tube and the radioactive gas is cryogenically pumped into it. When the xenon gas has been frozen inside, a torch is used to seal off the pinched top of the breakseal tube trapping the xenon inside. Finally, the tube can be removed from the system, assayed in the Ge(Li) detector, and introduced into the counting system.

V. Results

General

This project was a continuation of three previous studies concerning high pressure proportional counters. Since this system was basically the same as Lucyshyn's, and the approach of the study was to follow his plan, certain measurements and calibration calculations are compared to his findings. The first results, therefore, compare volumetric measurements and equipment calibration. Once these values were determined, the detector response was studied using external sources. Finally, an attempt was made to resolve xenon-133 and xenon-131m using an internal source. The fill gases used and the pressures at which the detector was studied are listed in Table VII.

TABLE VII
FILL GASES AND PRESSURES TESTED

Fill Gas	Pressure (Atm)				
	1	5	10	20	50
P-10	X				
Argon		X		X	X
Xenon (Grade 3)		X	X	X	
Xenon (Grade 4.5)		X	X	X	
Radioactive Xenon		X	X	X	

The remainder of this chapter will present the results of the experimental multiplication factors, experimental Diethorn constants, resolution, and intrinsic efficiency at optimum voltages. This chapter will also contain a discussion of the modifications to the purification system and the field tubes. Finally, a brief discussion of detector stability will be presented.

Experimental Multiplication Factors

The multiplication factors were calculated directly using a form of Equation (2) and Equation (11). Experimental multiplication curves are depicted in Figures 11 through 14 for testing conducted with argon, xenon (grades 3 and 4.5), and radioactive xenon. The argon multiplication factors determined from this study are consistently less than those of Lucyshyn (18:39-40). There are no comparative multiplication values for studies using pure xenon as a fill gas.

The argon spectra were collected with the source over the center window. At that stage of the study, accurate field tube voltages had not been determined. However, all source windows were used during collection of xenon spectra.

Once multiplication factors had been calculated, they were used to calculate the Diethorn constants ΔV and K . Experimental values for the Diethorn constants

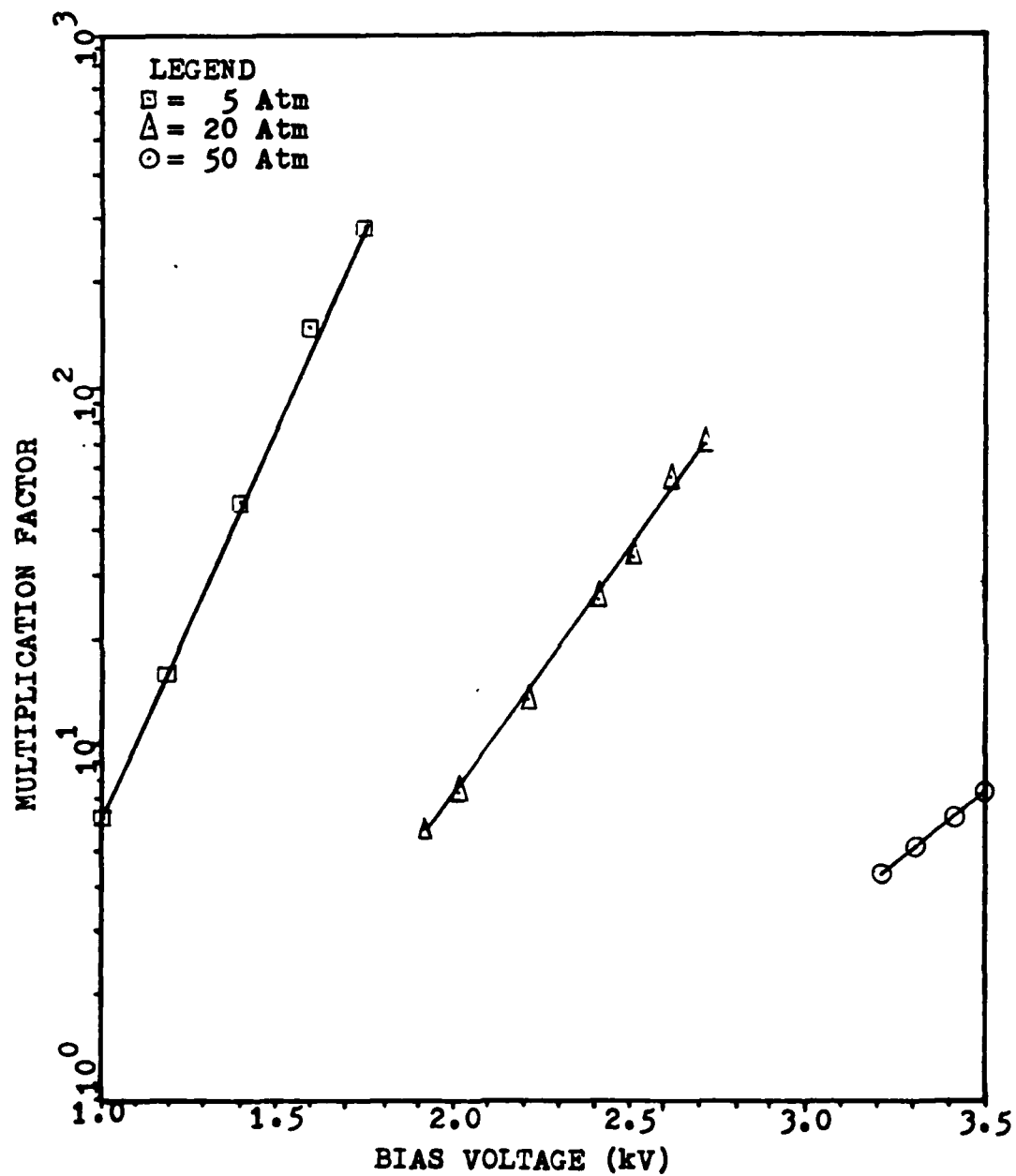


Fig. 11. Experimental Multiplication Curves for Argon

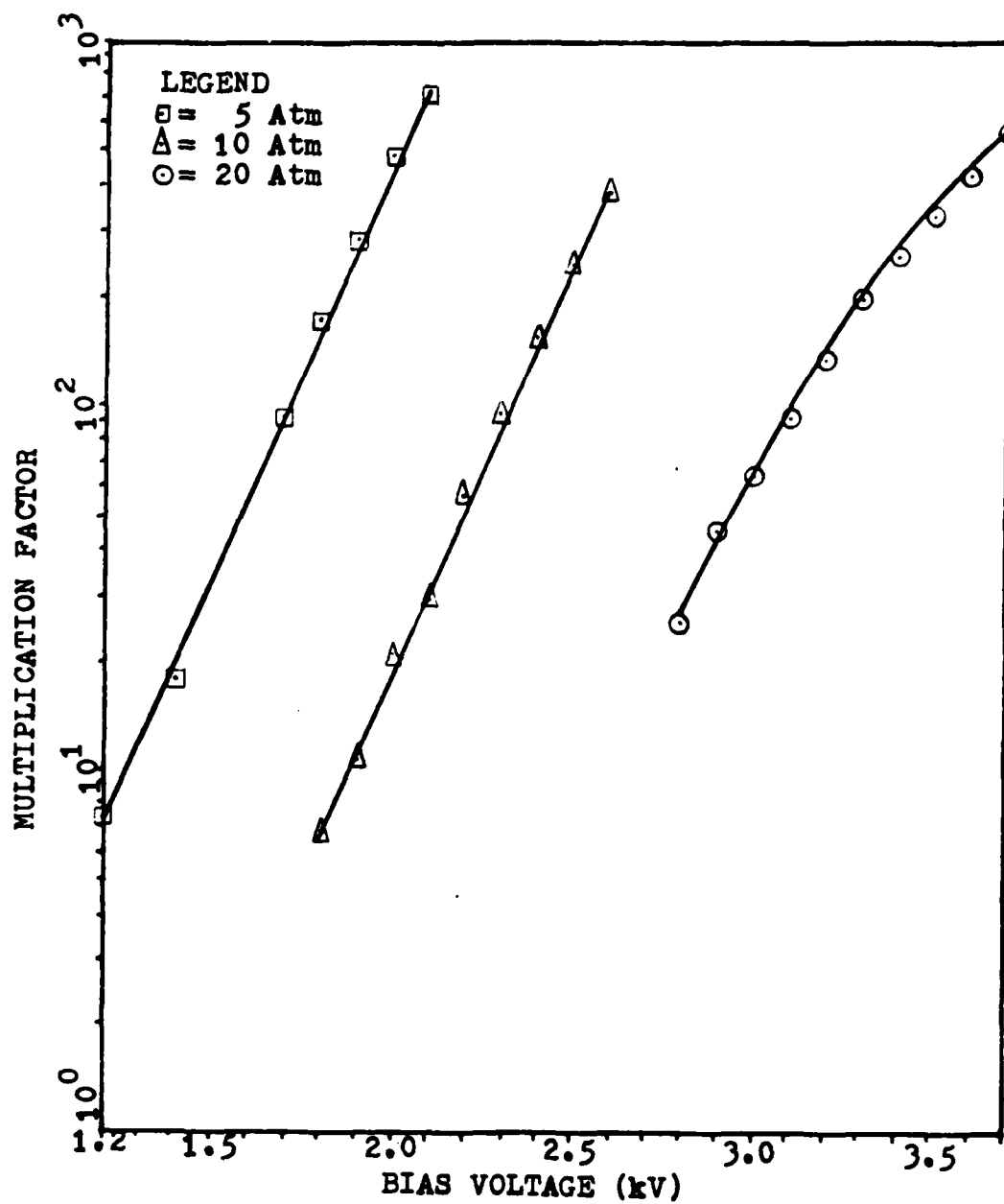


Fig. 12. Experimental Multiplication Curves for Xenon (Grade 3)

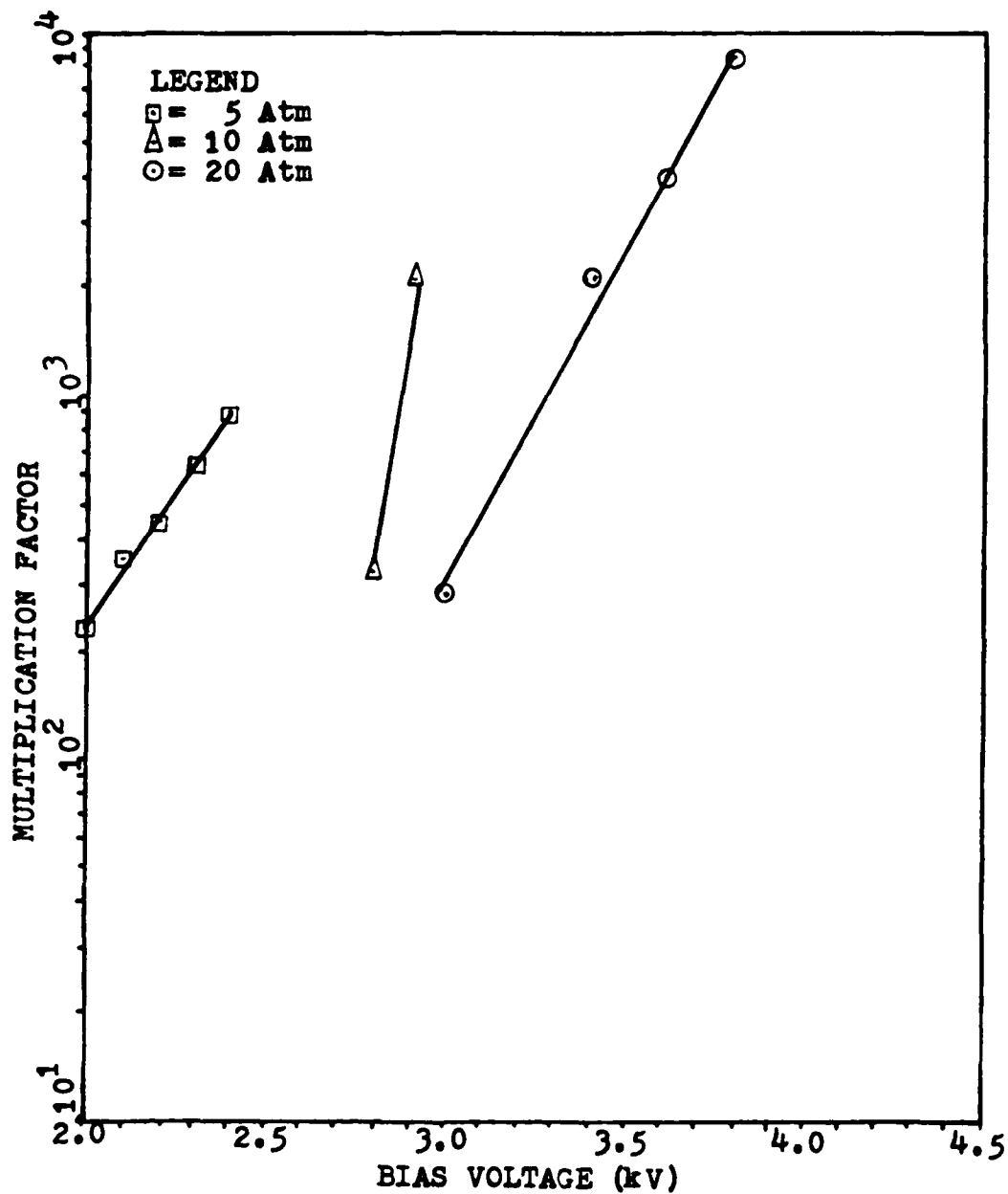


Fig. 13. Experimental Multiplication Curves for Radioactive Xenon

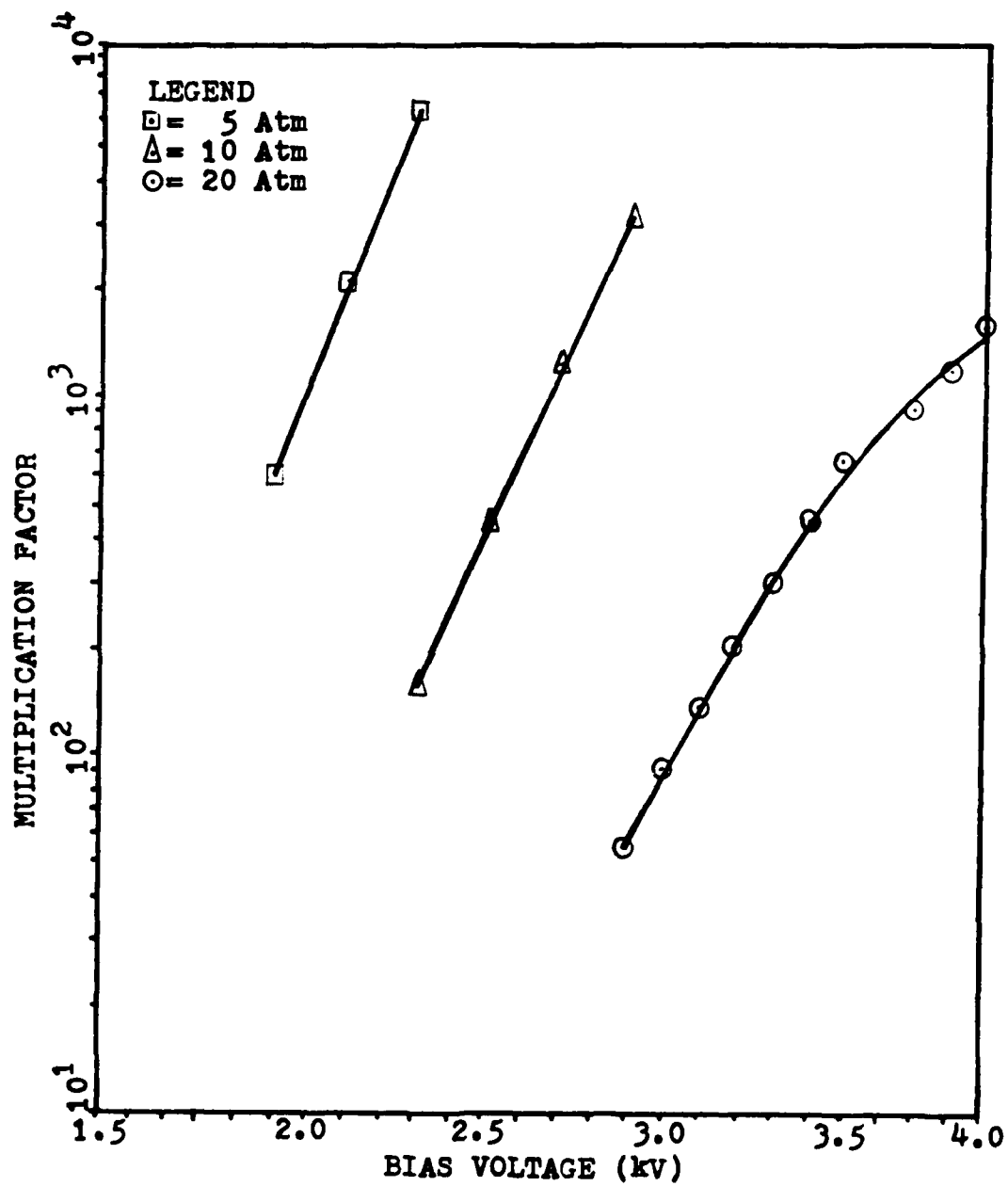


Fig. 14. Experimental Multiplication Curves for Ultra-Pure Xenon

TABLE VIII
EXPERIMENTAL DIETHORN CONSTANTS

Fill Gas	Pressure (atm)	ΔV (ev)	K (V/cm-atom)
Argon	5	28.45	32,141
	20	37.24	21,621
	50	58.39	15,657
Xenon (gr 3)	5	26.41	44,022
	10	23.80	39,653
	20	43.18	21,947
Xenon (gr 4.5)	5	30.8	31,500
	10	33.8	29,909
	20	48.2	19,477
Xenon (rad.)	20	38.22	18,355

are listed in Table VIII. The Diethorn relationship (Equation (3)) can be manipulated to produce the following:

$$\frac{\ln(M) * \ln(b/a)}{V} = \frac{\ln 2}{\Delta V} * \ln \left[\frac{V}{P_a \ln(b/a)} \right] - \frac{\ln 2 * \ln K}{\Delta V} \quad (11)$$

A plot of $V/\ln\{P_a \ln(b/a)\}$ along the x axis and $\{\ln M - \ln(b/a)\}/V$ along the y axis produces an almost straight line with a slope of $\ln 2/\Delta V$ and an intercept of $-(\ln 2)(\ln K)/\Delta V$ (14:194). This allows ΔV and K to be calculated easily. A linear-correlation coefficient, r, was determined for each set of data to test its straight-line fit. The value of r ranges from 0, where there is no correlation, to ± 1 , where there is complete correlation. Of the 10 data sets listed in Table VIII, six values of r were 0.98 or above,

three values were 0.89 and above, and the last value had low correlation at 0.45. This indicates the results did follow the straight line described by Knoll with the exception of the one data set. A least squares fit to the above-mentioned line will determine both the slope and intercept. The data should produce the same line for different detectors using the same gas.

There are no comparable constants for pure argon or pure xenon except for those values determined by Lackey and Lucyshyn. Lackey found a ΔV of 28 eV and a K of 14,500 V/cm-atm for both 25 and 50 atmospheres of argon (16:80). Lucyshyn found values of 31.5 and 40.8 (eV), and 20,729 and 14,915 (V/cm-atm) for 20 and 50 atmospheres, respectively. For comparison, one author determined the Diethorn parameters for a detector using a fill gas of 90 percent Xe and 10 percent CH₄ of 33.9 eV and 36,200 V/cm-atm. Another of his detectors with a fill gas of 95 percent Xe and 5 percent CO₂ had a ΔV of 31.4 eV and a K value of 36,600 V/cm-atm (10:311). These two detectors were operated at pressures of 0.5 to 2 atmospheres. The smallest anode wire used in that study was 25.4 μ m in diameter compared to a diameter of 3.8 μ m for this study. Lucyshyn's calculated values are 15 and 30 percent less for the two comparable ΔV values and the K values are about 4 percent less.

Resolution

The resolution is defined as the full width at half the maximum height (fwhm) for a given peak in the pulse height spectrum (see Chapter II).

The limiting theoretical resolutions for argon were computed by Lucyshyn in his thesis (18:45). They are reproduced here for ease in comparison with optimum experimental resolutions for each particular fill gas and pressure combination. Table IX contains the limiting theoretical resolution calculated using data from Knoll and the three principal gamma rays of the external sources. An assumption was made in calculating theoretical resolutions for testing with xenon fill gas. Although Knoll provides an experimental value of the Fano factor for xenon, 0.17, no value of multiplication variance, b , could be found in the literature. Knoll does, however, state that b is approximately equal to 0.5 for large values of the multiplication factor (> 100) (14:199). Since the experimental xenon multiplication factors ranged from 10 to 1000, a value of b equal to 0.5 was deemed appropriate for use.

Experimental resolutions were obtained at the optimum operating conditions for each combination of pressure and fill gas. A test to determine the impact on electronic noise resulted in a fwhm of 2 channels, as it did for Lucyshyn (7:44). The noise contributed between 0.05 and 0.4 keV depending on the energy deposited. The following

TABLE IX
LIMITING THEORETICAL RESOLUTION

<u>ARGON</u>					
Source	Edep (keV)	F*	b*	W(eV) *	R(%) [keV]
Am	59.537	0.17	0.50	26.2	4.05 [2.41]
Cd	88.03	0.17	0.50	26.2	3.33 [2.93]
Co	122.06	0.17	0.50	26.2	2.83 [3.45]
<u>XENON</u>					
Source	Edep (keV)	F*	b*	W(eV) *	R(%) [keV]
Am	59.537	0.17	0.50	21.5	3.67 [2.19]
Cd	88.03	0.17	0.50	21.5	3.02 [2.66]
Co	122.06	0.17	0.50	21.5	2.56 [3.12]

*These values were assumed applicable at all pressures.

equation was used to determine the fwhm without the noise (14:92):

$$(\text{FWHM})_{\text{Tot}}^2 = (\text{FWHM})_{\text{Det}}^2 + (\text{FWHM})_{\text{Noise}}^2 + (\text{FWHM})_{\text{Other}}^2 \quad (12)$$

The detector fwhm can now be determined separate from the noise by assuming the other contributions were negligible. Table X lists the optimum experimental resolutions corrected for noise.

TABLE X
OPTIMUM EXPERIMENTAL RESOLUTIONS

Fill Gas	Pressure (Atm)	Voltage (Volts)	Resolution in % and (keV)		
			Am	Cd	Co
Argon	5	1600	5.98%(3.56)	—	—
Argon	20	2700	7.79%(4.64)	11.1%(9.97)	—
Argon	50	3500	25.6%(15.2)	34.3%(30.2)	21.9%(26.8)
Xenon	5	2000	7.3%(4.4)	6.5%(5.7)	5.4%(6.6)
Xenon	10	2800	12.2%(7.3)	20.6%(18.2)	18.2%(22.3)
Xenon	20	3500	14.5%(8.6)	10.5%(9.1)	15.7%(19.1)

Intrinsic Efficiency

The method of computing intrinsic efficiency, both theoretical and experimental, was described in Chapter II. The efficiencies for the spectra, obtained at the optimum operating conditions, were calculated using the information contained in Tables I, II, and III (Chapter II). The various factors used in calculating the experimental efficiencies are contained in Appendix D. The net counts for cobalt-57 include those counts from both the 122 and 136 keV gamma rays.

For tests conducted with argon at 5 atmospheres of pressure, only the americium full energy peak could be resolved. Therefore, no experimental data was available for calculating the efficiencies for the cadmium-109 and cobalt-57 sources.

The comparable efficiencies calculated during this study are slightly better than those computed by Lucyshyn with the exception of americium-241 as a source for argon at 50 atmospheres. The experimental efficiencies for argon were, at worst, within a factor of 2 to 4 of the theoretical efficiencies.

The experimental efficiencies for xenon, especially for the americium-241 photons, are approximately 2 orders of magnitude less than the theoretical values. Since no references or comparable data can be found in the literature for studies using pure xenon, no other comparisons will be made. Many problems were encountered while testing with xenon as a fill gas. The count rate was so high at times that lead was used to collimate sources. The use of lead will definitely change the geometry factor and thus reduce efficiency. Also, the high count rate can effect a change in efficiency by the dead time factor. With no quench gas added, the effect of multiple pulsing is a loss of counts under the full energy peak, further reducing the intrinsic efficiency. Finally, the method in which net counts are obtained could significantly affect the resultant efficiency. Table XI lists the theoretical intrinsic efficiencies (calculated by using Equation (7)) and the experimental intrinsic efficiencies (calculated by using Equation (4)).

TABLE XI
EXPERIMENTAL AND THEORETICAL EFFICIENCIES

Fill Gas	Pressure (Atm)	Energy (keV)	Efficiencies	
			Experimental	Theoretical
Argon	5	59.537	0.003 ± .001	0.0089
	5	88.03	--	--
	5	122.06	--	--
Argon	20	59.537	0.010 ± .003	0.0332
	20	88.03	0.007 ± .002	0.0099
	20	122.06	--	--
Argon	50	59.537	0.021 ± .007	0.0873
	50	88.03	0.025 ± .006	0.0267
	50	122.06	0.017 ± .004	0.0098
Xenon	5	59.537	0.022 ± .007	0.5419
	5	88.03	0.132 ± .003	0.2332
	5	122.06	0.061 ± .013	0.0997
Xenon	10	59.537	0.021 ± .007	0.7637
	10	88.03	0.256 ± .059	0.3947
	10	122.06	0.087 ± .018	0.1809
Xenon	20	59.537	0.019 ± .005	0.9076
	20	88.03	0.447 ± .103	0.5959
	20	122.06	0.185 ± .039	0.3059

Testing with Radioactive Xenon

Tests were conducted with a mixture of radioactive xenon and stable xenon as the fill gas. It was hoped that these tests would result in the successful resolution of the xenon-131m from the xenon-133. This result was not achieved. One test, however, did give an indication of the presence of xenon-131m. This test was conducted with an anode voltage of 2900 volts, a field tube voltage of 651 volts, and at a pressure of 10 atmospheres. The detector had a combination of xenon-133, xenon-131m, and stable xenon (grade 3) as the fill gas and internal source. The activity of the xenon-133 was approximately 1.031 uCi and the activity of the xenon-131m was approximately 0.044 uCi. The xenon-131m activity comprised about 6.2 percent of the total sample activity. This was the best ratio of xenon-131m to total activity during this study. In addition, an external source was used to provide reference peaks. It consisted of americium-241 covered with a thin foil of holmium. This provides both the 59.5 keV gamma (from americium) and the 47.5 keV K_{α} fluorescent x-ray from holmium. An additional peak was obtained for the xenon x-ray escape peak for the 59.5 keV gamma. The resultant spectrum (see Figure 15) had several identifiable peaks. The two most prominent peaks were assumed to be the 60 keV full energy peak for americium and the 29.7 keV K_{α_1} escape peak for americium. These were used to calibrate the MCA

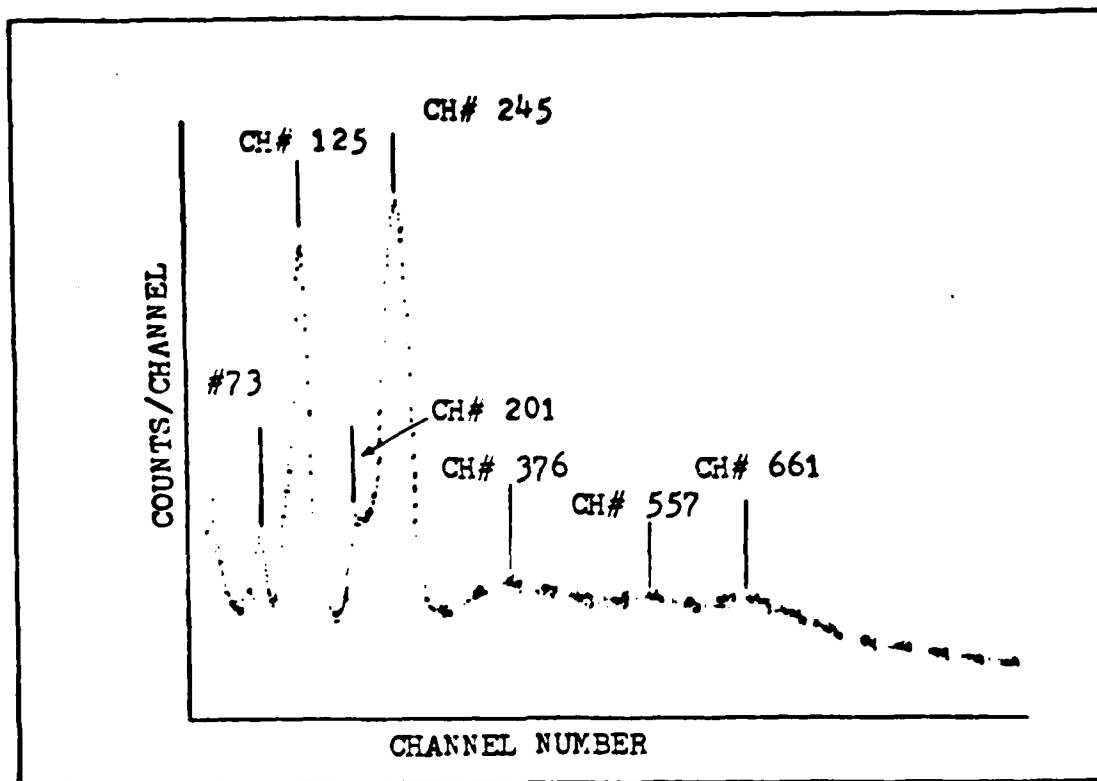


Fig. 15. Radioactive Xenon (Internal) Spectrum with Americium-241 on Holmium Foil (External)

screen. This resulted in a calibration of 0.248375 (keV/Ch)
 * Ch# - 1.312 keV. Using this calibration, Table XII is a listing of peaks of and the energies observed for this test. The internal conversion electrons of xenon-131m form the peak near channels 661 and 557. The peak near channel 376 corresponds to an x-ray escape peak for xenon-131m's conversion electrons.

The remaining tests with an internal source met with less success. The general result, using an internal source alone, was five well-defined peaks with a beta continuum (see Figure 16). This result occurred with the

TABLE XII
SPECTRAL RESULTS USING AN INTERNAL AND EXTERNAL SOURCE

Observed Energy (keV)	Identification of Peak		Actual Energy (keV)	Channel Number	δ (keV)
163.9	ICE	xenon-131m	163.98	661	-0.08
137.0	ICE	xenon-131m	129.4	557	7.6
93.0	XEP for 131m ICE		99.6	376	-6.6
59.537	FEP americium-241		59.537	245	0.0
48.61	holmium K_{α}	x-ray	47.55	201	1.06
29.735	Xenon K_{α}	XEP (Am)	29.735	125	0.00
24.52	Xenon K_{β}	XEP (Am)	25.896	104	-1.376
16.81	Xenon K_{α}	XEP (Ho)	17.7	73	-0.89

SYMBOLS: FEP - Full Energy Peak, XEP - X-ray Escape Peak, ICE - Internal Conversion Electrons.

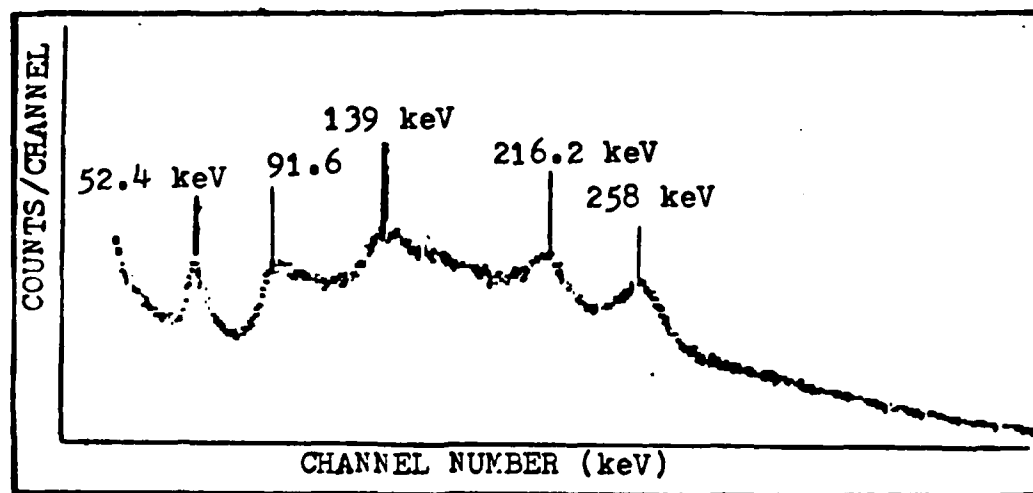


Fig. 16. Radioactive Xenon Spectrum (Internal Source Only) Calibrated with Americium-241 (External) FEP and XEP

use of grade 3 xenon only. When anode voltage was higher than 2400 volts (for 5 atmospheres) and 3000 volts (for 10 atmospheres) the peaks in the spectra smoothed out. Grade 4.5 xenon was only used in one test with an internal source, 20 atmospheres, and this test produced indications that the anode wire had an area of non-uniformity. Time was not available to replace the anode wire and conduct more tests using grade 4.5 xenon. In addition, the local source for xenon-133 had temporarily become unable to supply radioactive xenon. Sample spectra from the various tests can be found in Appendix C.

With so many unknown peaks in the radioactive xenon spectra, it is extremely difficult to interpret the results. Therefore, an examination of where the peaks occurred was undertaken. More precisely, the number of channels between corresponding peaks was calculated to see if there were any trends. Seven different spectra were examined from tests at 5 and 10 atmospheres of pressure. The average number of channels between peaks and the predicted dispersion from the mean are as follows: peaks 2-1, $52.4 \text{ channels} \pm 1.33$; peaks 3-2, $68.1 \text{ channels} \pm 2.12$; peaks 4-3, $95.3 \text{ channels} \pm 4.48$; peaks 5-4, $60.7 \text{ channels} \pm 1.77$; and between peaks 5 and 1, $276.6 \text{ channels} \pm 6.65$. These results indicate that the radioactive gas samples had a very similar makeup and, quite possibly, a similar cause for the multiple peaks. The radioactive xenon tests were fraught with problems such

as: too high a count rate, too low a count rate, and many anomalous or, at least, unexplainable peaks. An attempt to complete an energy calibration using the americium FEP and XEP was unsuccessful. The use of cobalt-57 and cadmium-109 with the internal source resulted in a spectra with the same 5 peaks characteristic of the internal source alone.

Purification of Fill Gases

The modification to the purification system was evaluated with argon in three ways: a 12-hour purification cycle, a 3-hour purification cycle, and no purification at all. The results of the three methods are listed in Table XIII.

TABLE XIII
COMPARISON OF RESULTS USING DIFFERENT
LEVELS OF PURIFICATION

Gas	Pressure (Atm)	Method	Resolution (60 keV, Am) %[keV]	Intrinsic Efficiency
Ar	5	12-hr cycle	5.98% [3.56]	0.0032 ± .00108
Ar	5	3-hr cycle	7.9% [4.72]	0.0022 ± .00076
Ar	5	no purification	11.24% [6.69]	0.0022 ± .00074

The best results were obtained with the 12-hour purification cycle. During the 3-hour purification test discharge occurred at 1600 volts. In comparison with the

theoretical statistical limit of $\sqrt{.05}$ percent, the best resolution from the test of the 12-hour cycle was 1.5 times larger, the resolution from the 3-hour purification cycle was about two times larger, and the resolution from the test with no purification was about 2.8 times larger. Electronic noise accounted for about 0.4 keV.

An evaluation of the purification method using xenon as a fill gas was not conducted. However, during one test with grade 4.5 xenon, the gas had been purified the first night and left in the detector without further purification the second night. The resultant spectra after the second night were unuseable because the resolution had deteriorated. Further purification (12 hours) resulted in spectra which were useable again. This result indicates that either the detector leaks (which is not likely if it is at high pressure) or that there are still sources of impurity inside the detector.

Field Tubes

The addition of the field tubes to the detector corrected the problem of distortion of the electric field cited by Lucyshyn (18:41). An example of the results of a test with argon as the fill gas at a pressure of 5 atmospheres is found in Table XIV. The anode voltage was 1600 volts, the field tube voltage was 335 volts, and the external source was americium. In this test the location

TABLE XIV
RESULTS OF FIELD TUBE TEST

Window	Peak Channel	Resolution % [keV]	Efficiency
Left	82	8.9% [5.30]	0.0010 + .0003
Center	83	7.29% [4.34]	0.0014 + .0005
Right	84	7.95% [4.73]	0.0013 + .0004

of the FEP, the resolution, and the efficiency were obtained with the source at 3 different locations along the center wire. As the results of table XIV indicate, peak location and efficiency are essentially constant. However, the resolution at the left and right windows (which are close to the field tubes) are poorer by 0.4 to 1.0 keV.

During a later test, using grade 3 xenon at 5 atmospheres, with an anode voltage of 2000 and a field tube voltage of 425 volts, the resultant peaks (from left to right window) were in channels 253, 253, and 254.

Detector Stability

The detector was pressurized and allowed to remain in that condition for 24 to 96 hours in order to determine whether or not the pressure was remaining constant inside the detector. Upon venting the pressure back into the system, it was evident that small losses of gas had occurred. A summary of these results is contained in Table XV.

TABLE XV
DETECTOR PRESSURE LOSS

Gas	Press (Atm)	Time in Det. (hours)	Press. Before (torr)	Press. After (torr)	% Change
Ar	5	24	206	199	-3.4
Ar	20	96	355	352	-0.8
Ar	50	96	628	611	-2.7
Xe	5	24	38.4	37.2	-3.1
Xe	10	24	73.3	71.3	-2.7
Xe	20	24	139.0	136.9	-1.5

The period of data collection was always less than 8 hours. These results amount to a loss of approximately 1 percent (maximum) during any testing period. The pressure, therefore, was assumed to remain constant during data collection.

VI. Conclusions and Recommendations

Conclusions

The purpose of this study was to test and evaluate a high pressure proportional counter to qualitatively and quantitatively analyze the amounts of xenon-133 and xenon-131m present in a gaseous mixture.

Resolution with pure xenon was better than with argon by about 1 keV at 5 atmospheres and about 3 keV at 20 atmospheres for the 59.5 keV full energy peak for americium. Resolutions became poorer as pressure was increased from 5 to 50 atmospheres. The increase in fwhm was not linear with pressure and no single functional relation was found between resolution and pressure.

A disparity was found in the intrinsic efficiencies of xenon. Experimental efficiencies for the xenon trials using americium-241 as the source were lower by factors of 9 to 47. The remaining efficiencies were all within one order of magnitude of the experimental values. This disparity was most likely caused by one of two factors: use of lead to collimate the source (without altering the geometry factor) and/or the possibility of too-high a dead time rate.

Outgassing problems were experienced with both argon and xenon. However, typical times for the subsequent

deterioration in resolution were about 12 hours compared with 3 to 4 hours used for obtaining data.

Considering all the experimental data from this study, it is probable that this system can perform the type of detailed analysis desired. However, in spite of numerous separate experiments with xenon as a fill gas (over 200 spectra were taken) the requisite quality of data, to make the desired analysis, is missing. The problem with lack of quality data stems as much from inexperience in working with xenon as from problems with the system itself. The initial problems of anomalous peaks, while testing with xenon, seemed to dissipate with the use of grade 4.5 xenon. However, time on the project ran out before grade 4.5 xenon could be used more extensively.

The tests conducted with an internal source were too few in number. Grade 4.5 xenon was used only in the tests conducted at 20 atmospheres. Unfortunately, this was the last experiment as time ran out, and the results, showing a peak found only in results from the center source window, indicated that the anode wire had some type of non-uniformity.

The only positive indication that the detector can resolve the 164 keV peak of xenon-131m had the problem of almost 75 percent dead time. However, the fact that the radioactive gas, xenon-133, has only minute amounts of xenon-131m as an impurity, coupled with spectral results

indicating the presence of the 164 keV peak, leads one to believe that the detector is capable of performing the desired analysis.

The goal of determining the results of the new purification system were successful in regards to using argon as a fill gas. The purification system does improve resolution. The results indicated that the full width at half the maximum obtained with gas that was purified for 12 hours was one-half that of a test using no purification. However, the effect of the purification system on resolution, with xenon as a fill gas, was not fully quantified. While using xenon as a fill gas, purified xenon was left inside the detector overnight. The next morning (about 20 hours later) the spectra were unuseable. However, after a 12-hour purification the spectra became useable again. Because of this, it is felt that the system does improve the resolution by eliminating many impurities in the xenon fill gas.

The field tubes were tested and they are functional. The distortion in the electric field inside the detector has been reduced to almost negligible proportions. Results of one test indicated the mean resolution from each of the three source windows was 8.05 percent and $\sigma_n = 0.47$ percent. This indicates that the field tubes are effective. In addition, the sensitive volume of the detector now occupies

43.7 percent of the detector volume (minus the coldfinger) as compared to Lucyshyn's value of 37 percent.

The detector was not characterized as to its level of sensitivity. The activity of the internal source for the last test conducted was assayed as 0.0081 uCi (maximum). However, a rough calculation based on the number of counts registered at the MCA predicted an activity of 0.0025 uCi. The detector was responding with 25 to 40 counts per second during this test. For this level of activity, the detector response indicated the presence of a beta continuum. However, problems with the anode wire, combined with the fact that this was the last experiment, left further questions unanswered.

The question of whether this detector has promise for "production-type" operations cannot be answered at this time. The major problems seen with this detector are the anode wire, the impurities in the system, and the loss of pressure through leaks. The current anode wire is extremely fragile and can become broken or pitted easily. A different method of anode wire replacement would be foremost among considerations for a production-type model. Realizing it would be easy to require factory replacement, under more suitable conditions, the anode wire would require greater durability and tensile strength. Further research must be accomplished concerning types of anode wire available, characteristics of such a wire, and its

cost. This could prompt further research as to suitability for this type detector. The second problem, that of impurities in the system, could be controlled through the production process. The same is true of the third problem mentioned, that of leaks in the system.

In summary, the study of this detector has indicated the potential to qualitatively and quantitatively analyze small amounts of xenon-131m and xenon-133 in a gaseous mixture. However, further testing and more research are necessary in order to draw more definite conclusions. The tradeoff, in determining whether or not further research is accomplished, will be the amount of available resources (in time, money, and personnel) compared to the relative importance of the outcome.

Recommendations

It is recommended that the study of this high pressure proportional counter, and its capability to detect and analyze noble gases, be continued.

There are no major system faults that demand immediate correction, however, some additional modifications should aid in system performance.

The purification system should be further modified to add an in-line air-flow pump. This pump would increase circulation inside the gas reservoir and help to ensure that all the fill gas, inside the reservoir, passed through

the purification tube. One type of air flow pump has already been constructed and is available for use. It is made of glass and is approximately 8 inches in length. It has a glass rod with two propeller-like blades to force gas through the system. The pump would be powered by a variable magnetic field applied at one end of the rod. This pump could fit in the system between the top of the purification tube and the gas reservoir.

It is also recommended that a pressure transducer be installed in the detector. A transducer was ordered, prior to the start of this study, but it never arrived. The installation of the transducer will provide an accurate means of monitoring fill gas pressure in the detector.

Further study of this system should use grade 4.5 xenon instead of grade 3 xenon. Approximately 70 percent of all testing with xenon as a fill gas was conducted using grade 3 xenon. The improved results would be worth the cost of the research grade xenon.

Consideration should be given to finding a different type of anode wire. The present wire lacks the durability and strength to be used at higher voltages. Cost and availability will be key factors in choosing and obtaining a stronger, more durable, wire while maintaining a small diameter.

Finally, binary mixtures of argon and grade 4.5 xenon should be studied as potential fill gases. Lucyshyn

had limited success in his study with such mixtures, and such a combination should prove effective with this detector.

Appendix A: Equipment Calibration

Volume Measurement

Table XVI depicts the comparison of volumetric measurements between this study and Lucyshyn's results (18:56).

TABLE XVI
COMPARISON OF DETECTOR SYSTEM VOLUMES

Volume Between Valves	<u>THIS STUDY</u>		
	Name	Vol. (cm ³)	Uncertainty (cm ³)
#4, #5, & #6	"T" Section	44.08	± 0.77
#6	Detector	252.02	± 1.35
#1, #2, #3, & #4	Reservoir	23610.70	± 40.6
Total		23906.80	± 40.2
<u>LUCYSHYN'S</u>			
#4, #5, & #6	"T" Section	44.89	± 0.95
#6	Detector	257.60	± 1.2
#1, #2, #3, & #4	Reservoir	29051.12	± 171.5
Total		29353.61	± 86.04

All of the volumetric measurements taken during this study proved to be smaller than Lucyshyn's measurements. Lucyshyn's reservoir volume was larger by 18.7 percent. His "T" section volume was larger by only 2.0 percent. His detector volume was larger by 2.2 percent, but, this was predictable since the field tubes were added to the detector after his study was completed.

The results of system calibrations are found in Table XVII.

TABLE XVII
RESULTS OF SYSTEM CALIBRATION (18:56-59)

<u>Validyne Pressure Gauge</u> This Study	Lucyshyn's Study
<u>High Scale</u>	
$P = P(\text{Val}) * 1.0017 - 0.02113$	$P = P(\text{Val}) * 1.00475 - 0.28147$
slope: ± 0.00135	slope: ± 0.00289
intercept: ± 0.06117	intercept: ± 0.1325
<u>Low Scale</u>	
$P = P(\text{Val}) * 0.99879 - 0.03912$	$P = P(\text{Val}) * 0.98222 - 0.03057$
slope: ± 0.00133	slope: ± 0.00102
intercept: ± 0.01828	intercept: ± 0.1045
PA Gain (x10) = 1845.13 (mV/pC) ± 42.9	Gain (x10) = 2107.4 (mV/pC) ± 35.5

Appendix B: Procedures

Replacement of the Anode Wire

The procedure to replace the anode wire is a lengthy process which can be completed in five basic phases: removal of the endcap(s) and centerpiece; replacement of the anode wire; glowing the anode wire in a glass tube; replacing the centerpiece and endcap(s); and finally, glowing the anode wire inside the detector. This entire process can be accomplished in 2 to 3 hours if everything goes smoothly; however, it usually takes in excess of 8 hours because of the many problems encountered during the process.

Removal of the Endcap and Centerpiece. It is first necessary to adjust the pressure in the detector to approximately one atmosphere. When xenon was in the detector it was cryogenically pumped into the reservoir and isolated, and the less-expensive argon was used to backfill the detector. This was introduced into the detector by an inlet valve just below valve #5. The endcap can be removed once the detector is at atmospheric pressure and isolated from the remainder of the system. The ceramic endplate of the centerpiece has three holes in it. The L end of a small allen wrench can be inserted into one of the holes and the centerpiece can be slowly pulled out of the detector.

Some resistance will be felt as the spring contacts for the field tubes will be in contact with the inside of the detector. If the centerpiece cannot be removed with the allen wrench, it is necessary to remove the left endcap as well, and push the centerpiece out of the detector.

Replacement of the Wire. The anode wire is stainless steel 0.0003 inches in diameter. This wire is not rigid, is thinner than human hair, and has less tensile strength than hair. The use of a dark cloth, as a background, significantly improves one's ability to see the wire. First, a piece of 0.005 inch stainless steel wire must be threaded through both support tubes. This wire is more rigid and threads through readily. Next, a tiny drop of cyanoacrylic (super glue) is placed on the larger wire to attach the top of the thinner anode wire. The tension spring is placed in the recess on the left end of the centerpiece before the lead is pulled through. A sufficient amount of anode wire must be pulled through the end to allow for attaching the wire to a brass endpiece which fits in the recess behind the spring. The anode wire is pulled through the brass endpiece and glued to its side with another small drop of cyanoacrylic. The tiny brass pin is then inserted into the brass endpiece and seated inside the left end recess. The right end of the anode wire is attached to a 2 gram weight with a piece of scotch tape.

This weight is slowly lowered until the spring has tension applied and the anode wire is taut. Then, the right-hand pin must be inserted very carefully to avoid breaking the fragile wire. Finally, the excess anode wire must be cut off.

Glowing the Anode Wire in Glass. The centerpiece is now inserted into the specially-made glass tube and the electrical leads are connected from the end caps on the tubes to the ends of the wire. The glass tube can then be inserted into the leak detector and the tube evacuated to at least 10^{-5} torr. Next, a regulated DC power supply and an ammeter are connected in series with the glass tube. The voltage on the power supply is slowly raised while observing both the ammeter and the anode wire. When the anode wire begins to sag and glow in the evacuated glass tube, the amount of amperage and voltage should be noted for subsequent use when the wire is inserted inside the detector. The anode wire should be glowed in this fashion for approximately 5 seconds. This procedure helps to remove particulates on the wire itself, but, more importantly, it provides information as to what voltage and amperage are required to glow the wire. After the voltage is turned back down, the glass tube can be vented and the centerpiece removed. The wire must be visually checked at this point to ensure that it becomes taut again when cooled. If it does not tighten then the wire must be replaced.

Replacing the Centerpiece and Endcap. When the centerpiece is re-inserted into the detector, care must be taken to align the field tube contact pins correctly. There is a guide pin located at the bottom of the right side of the detector. This will help ensure that the alignment is correct. A thin copper shim is used to depress the spring-loaded field tube contacts when the centerpiece is inserted into the detector. This prevents the spring-contacts from binding at the inside edge of the detector. Finally, the endplate(s) are bolted back on. New copper gaskets are used that have been carefully cleaned. The gaskets should be handled with clean forceps or lintless gloves. The copper varian gasket(s) must be mounted correctly or detector leaks could develop. The endplate bolts should be tightened in a cross-type pattern to ensure that the copper gasket is seated evenly. Once the endcaps have been replaced, the anode wire can be glow inside the detector.

Glowing the Anode Wire in the Detector. With the detector reassembled, the fore pump and diffusion pump are used to evacuate the detector to approximately 10^{-7} torr. After evacuation is complete it must be maintained while the anode wire is glow. The regulated DC power supply and ammeter are again attached, in series, to the two outer anode wire contacts. The voltage and amperage are slowly

raised to the same settings used with the glass tube. The wire should again be glowed for approximately 5 seconds. After glowing the wire, perform a continuity check to ensure that the anode wire has not burned through during the glowing process.

Appendix C: Sample Spectra

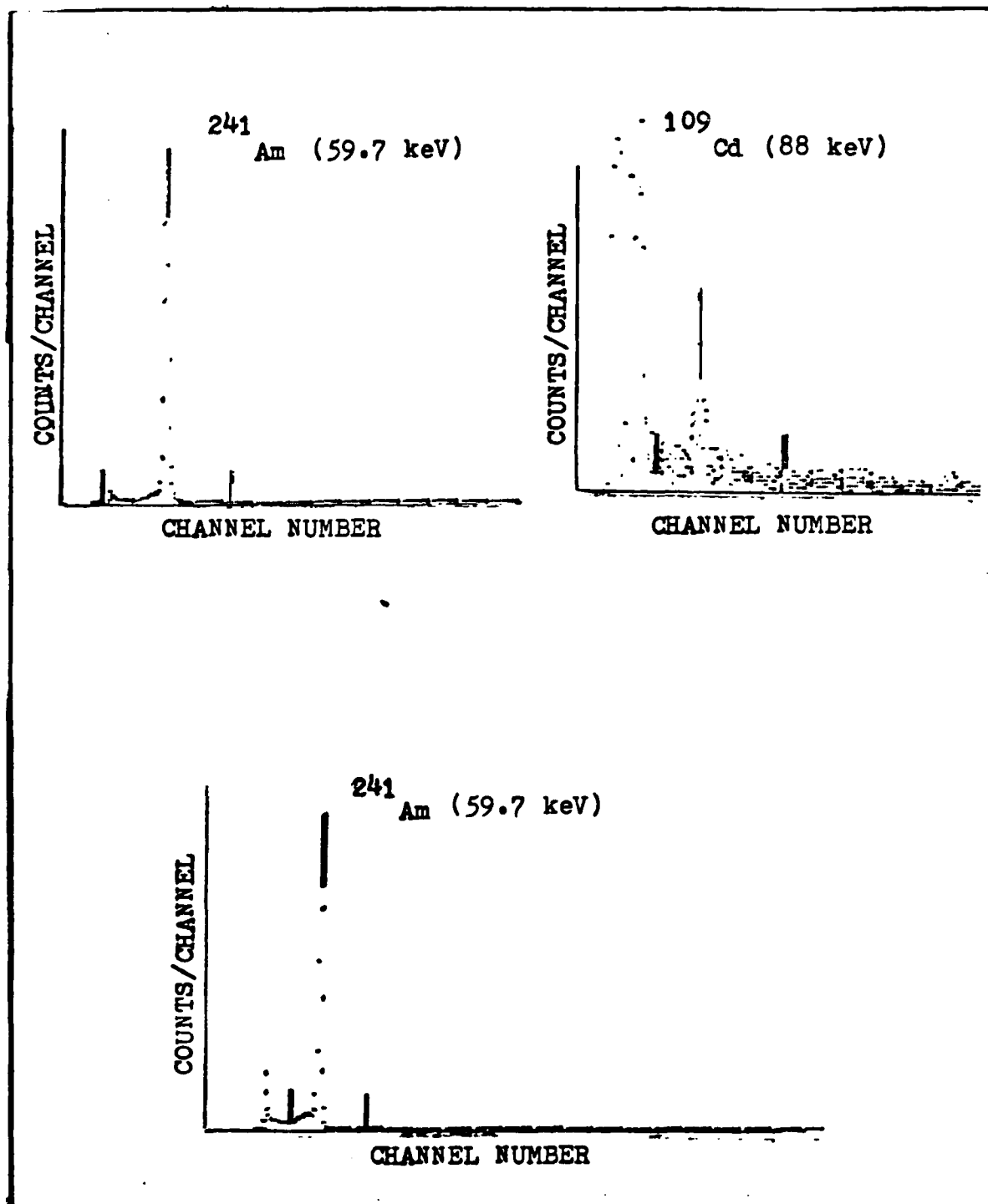


Fig. 17. Argon Fill Gas. Top: 20 Atm and 2700 Volts;
Bottom: 5 Atm and 1600 Volts

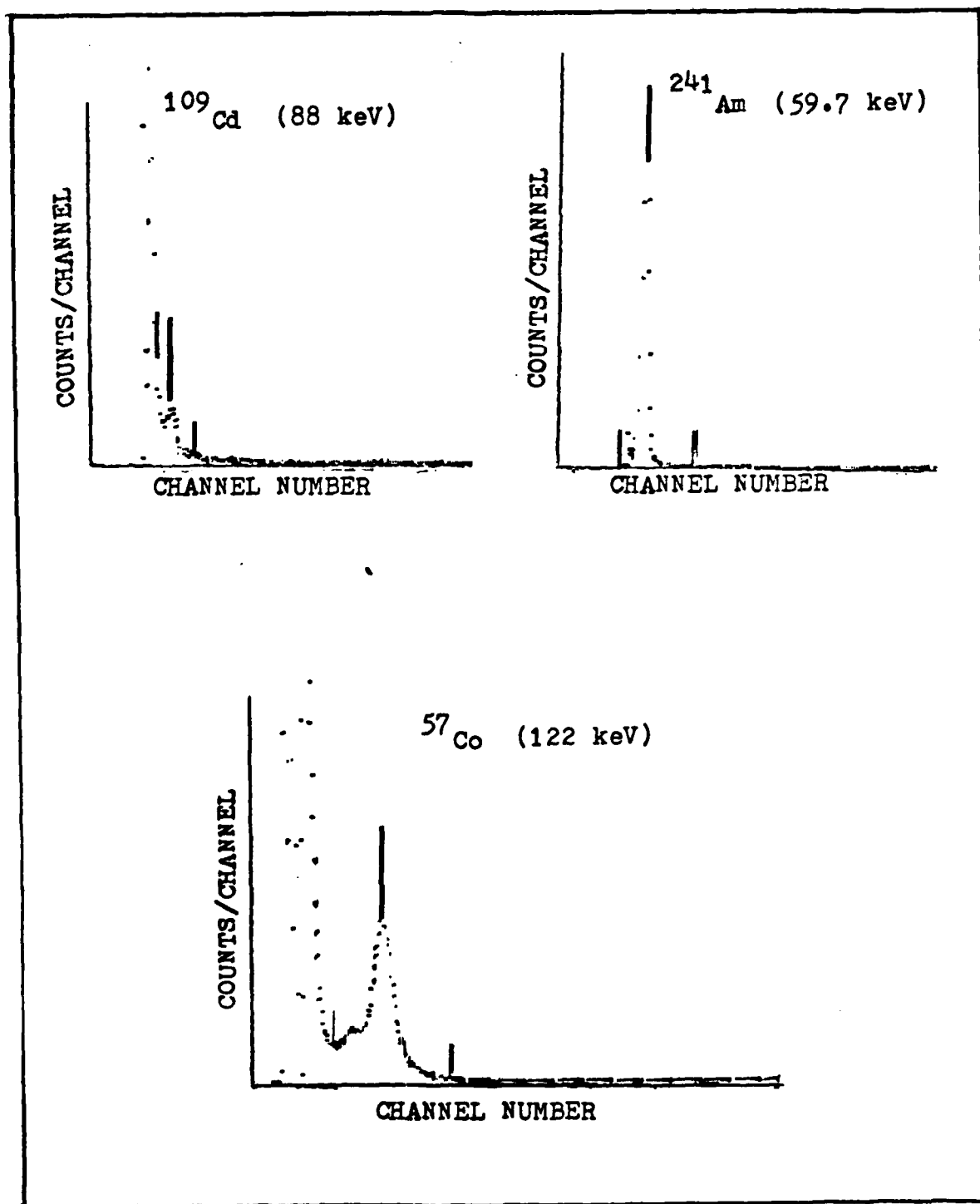


Fig. 18. Argon at 50 Atm and 3500 Volts

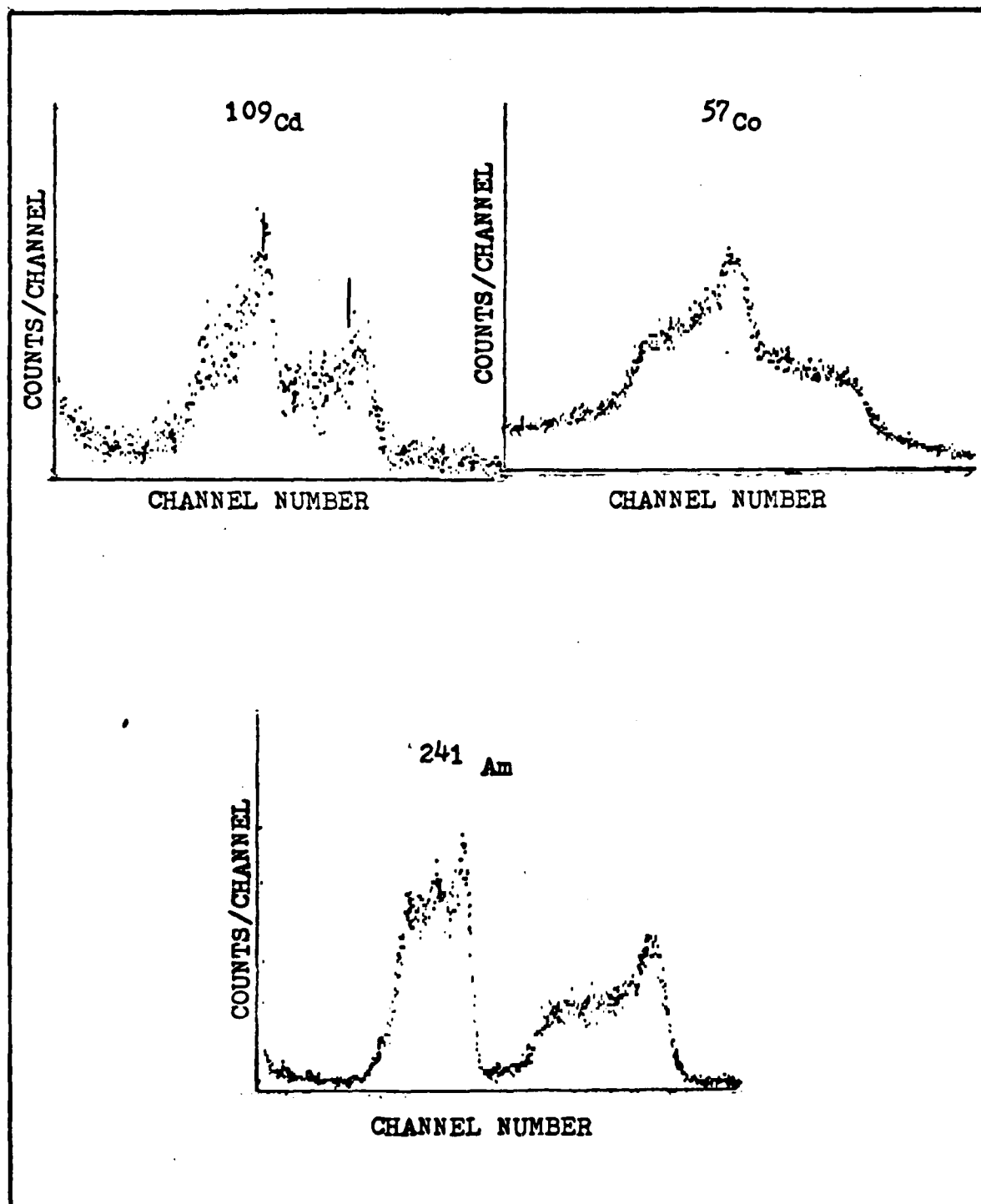


Fig. 19. Xenon at 5 Atm and 2000 Volts

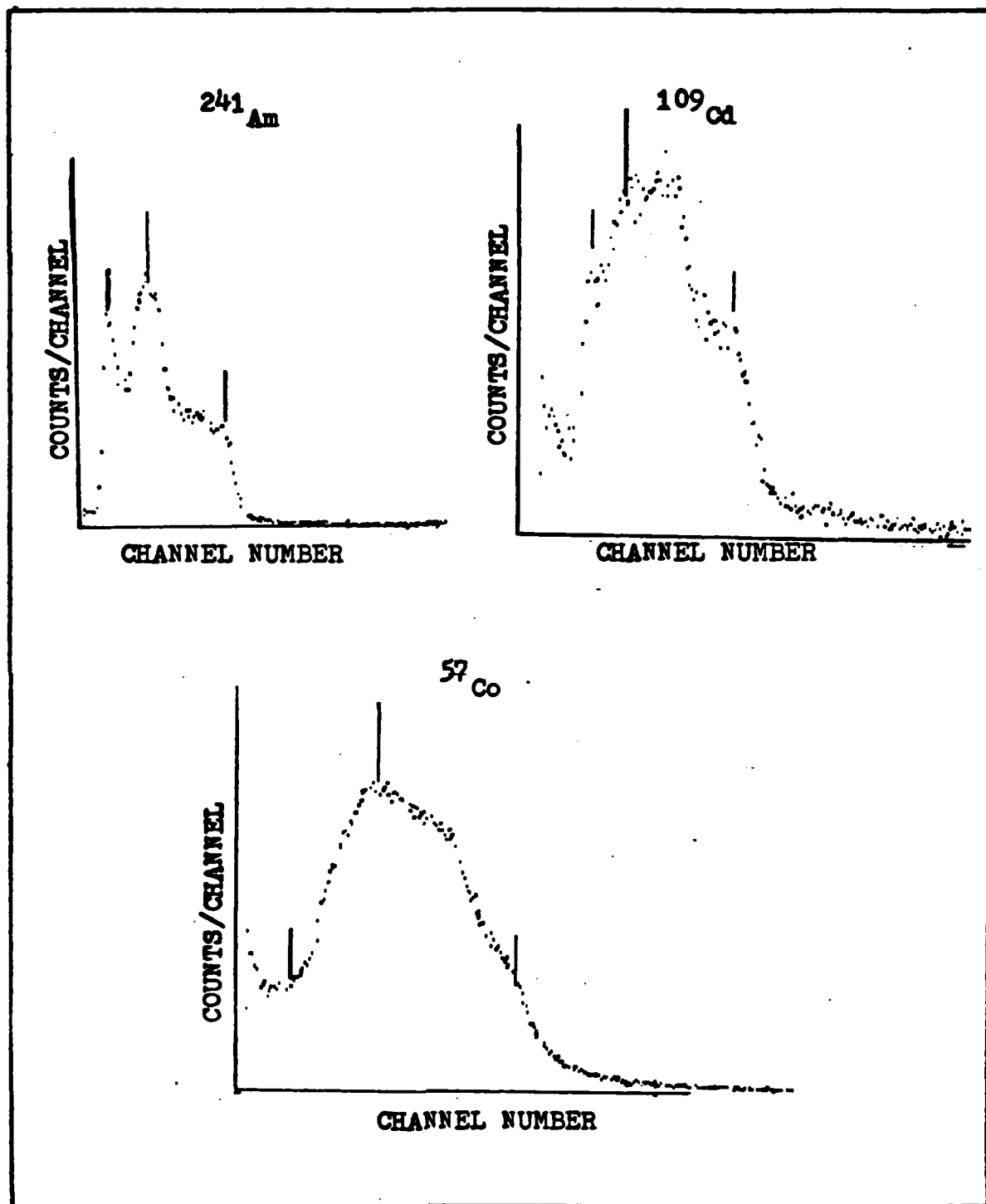


Fig. 20. Xenon at 10 Atm and 2800 Volts

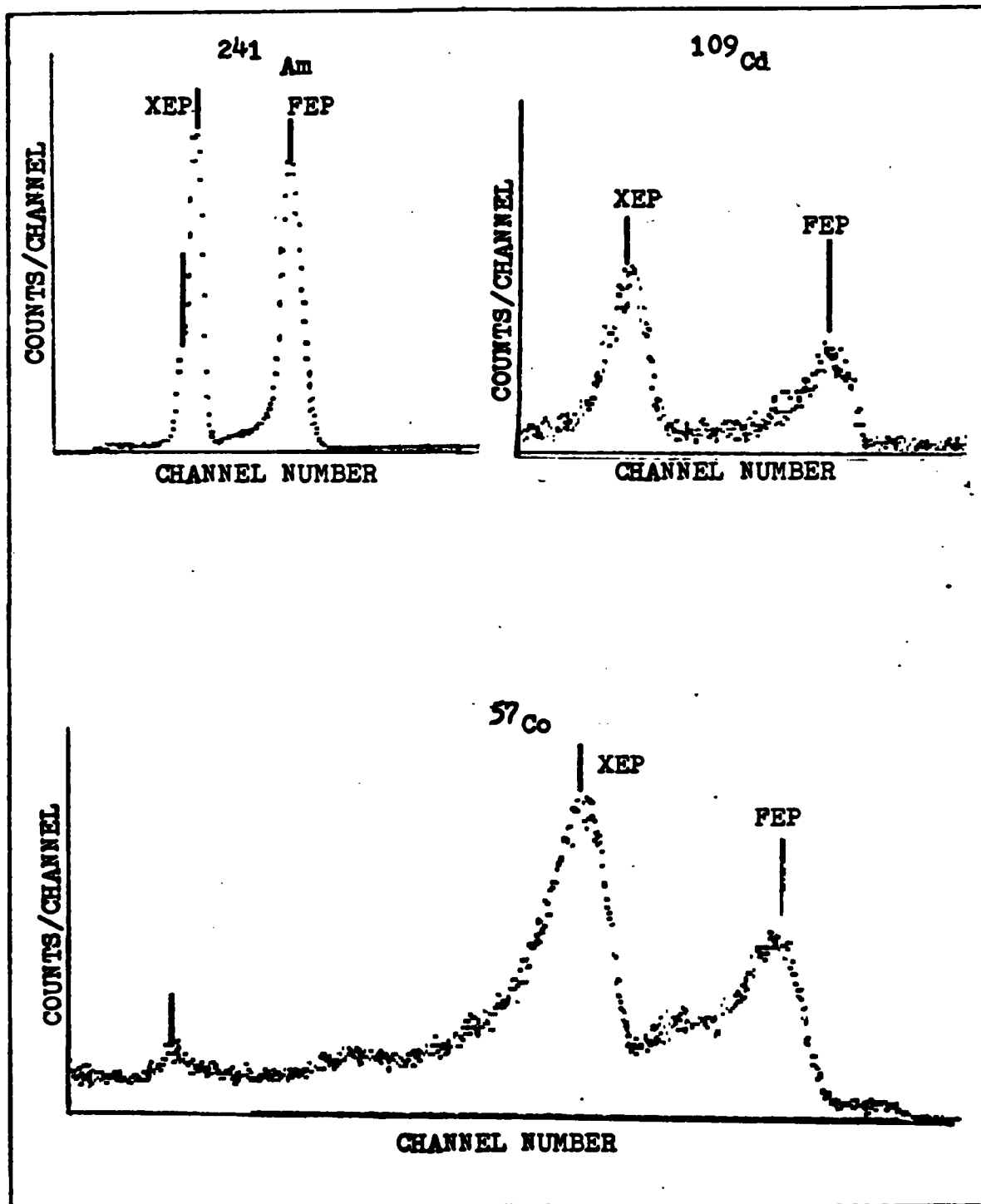


Fig. 21. Xenon (Grade 4.5) at 5 Atm and 2000 Volts

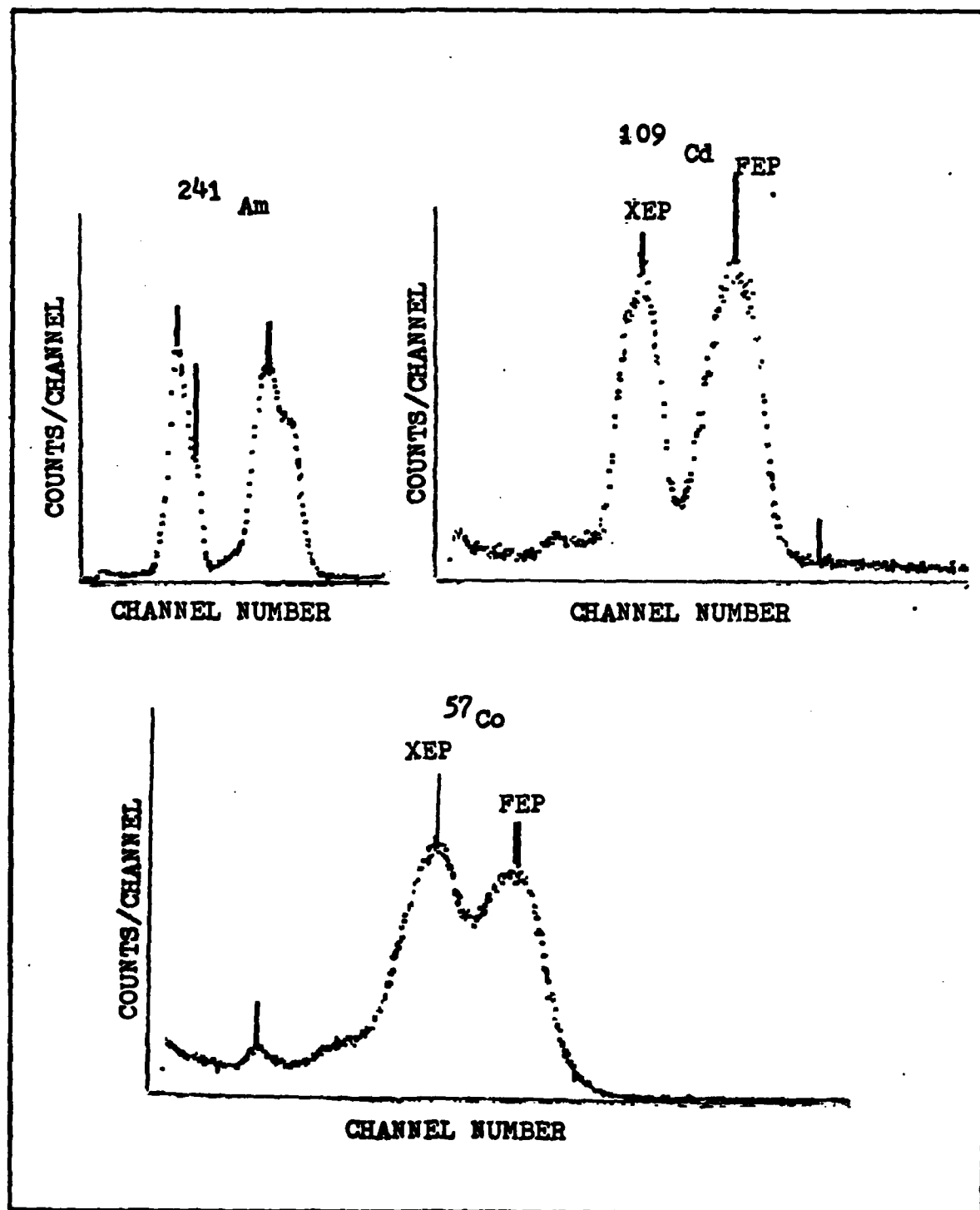


Fig. 22. Xenon (Grade 4.5) at 10 Atm and 2800 Volts

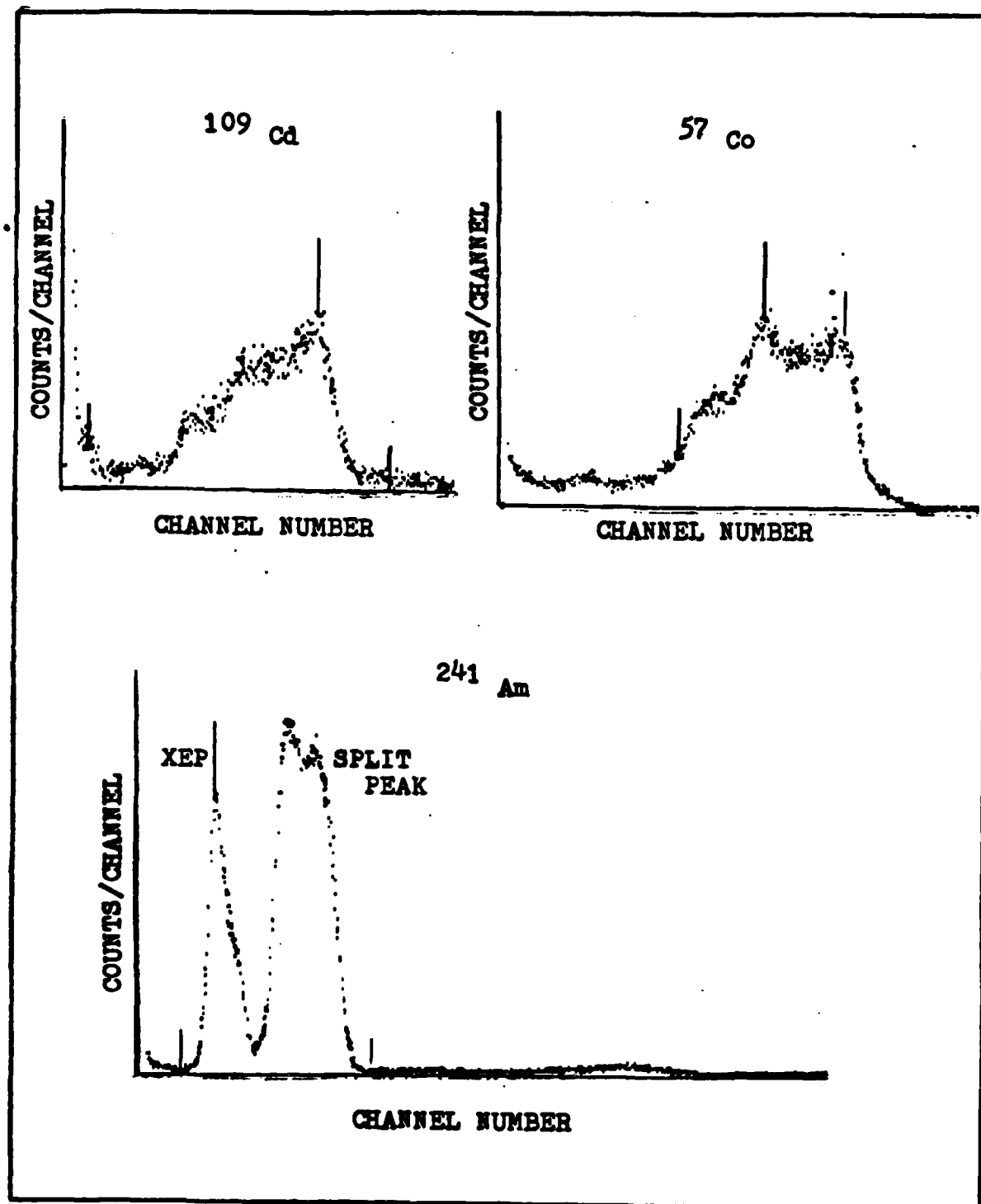


Fig. 23. Xenon (Grade 4.5) at 20 Atm and 3500 Volts

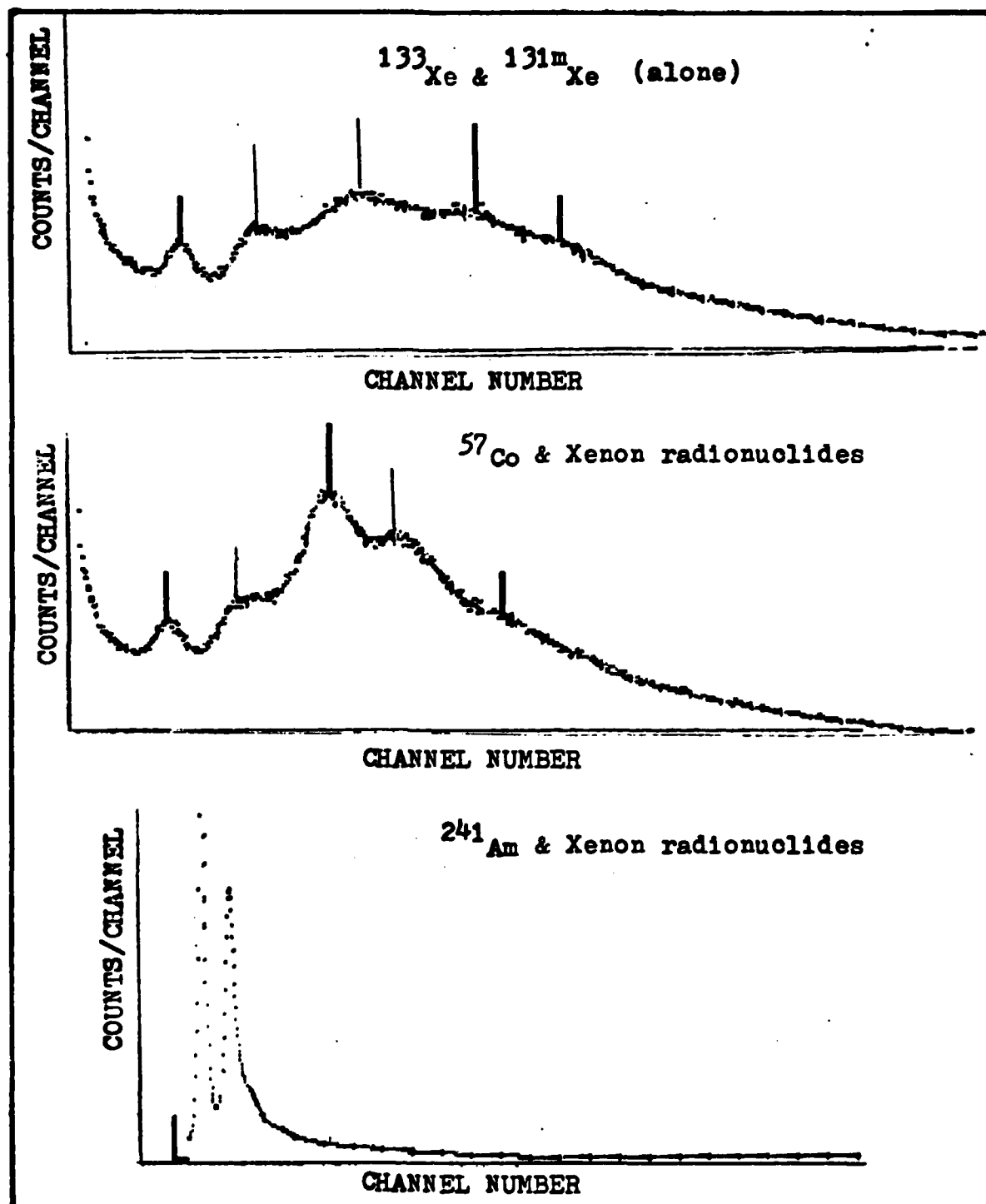


Fig. 24. Radioactive Xenon at 5 Atm and 2000 Volts with Various External Sources

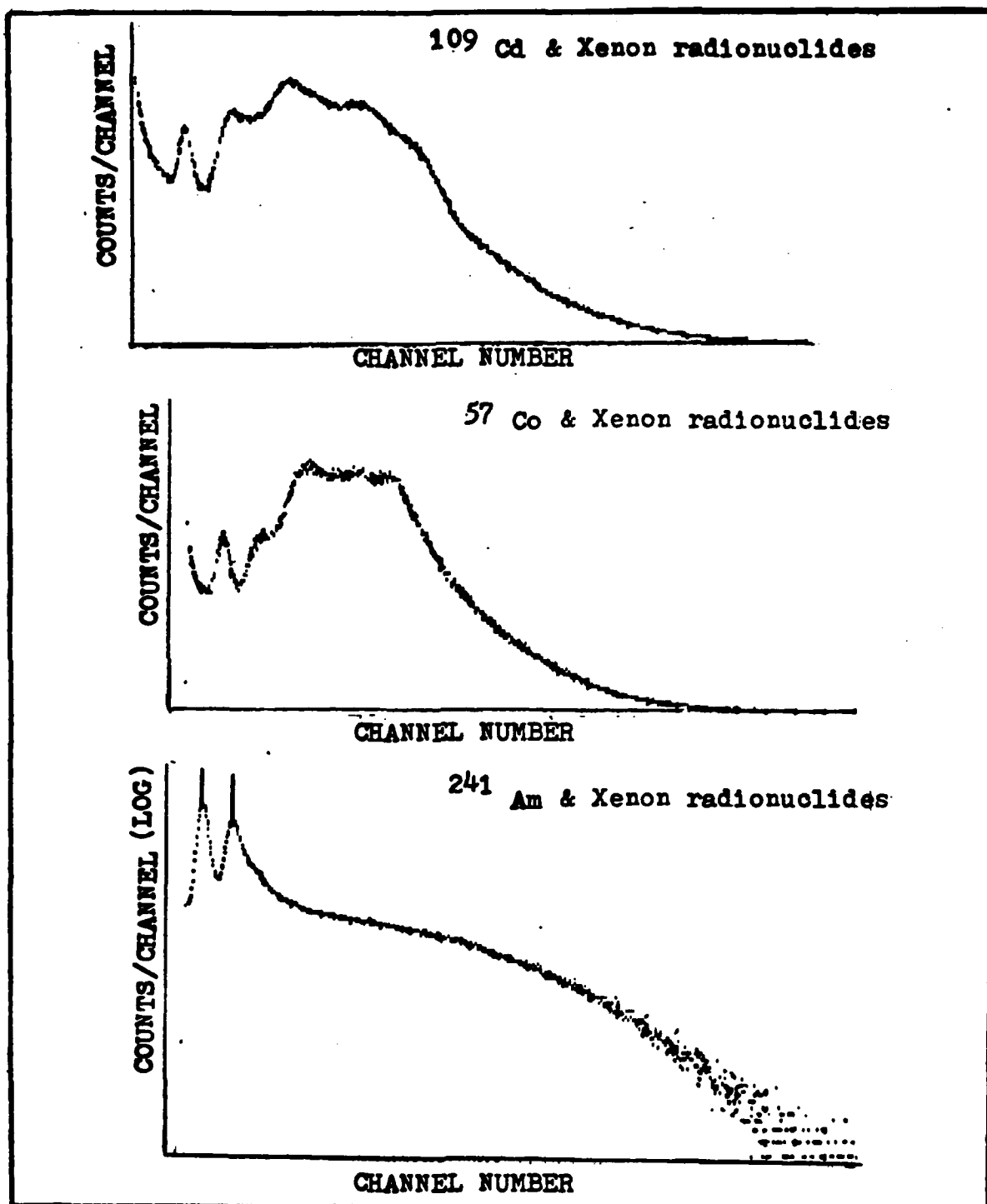


Fig. 25. Radioactive Xenon at 10 Atm and 2800 Volts with Various External Sources

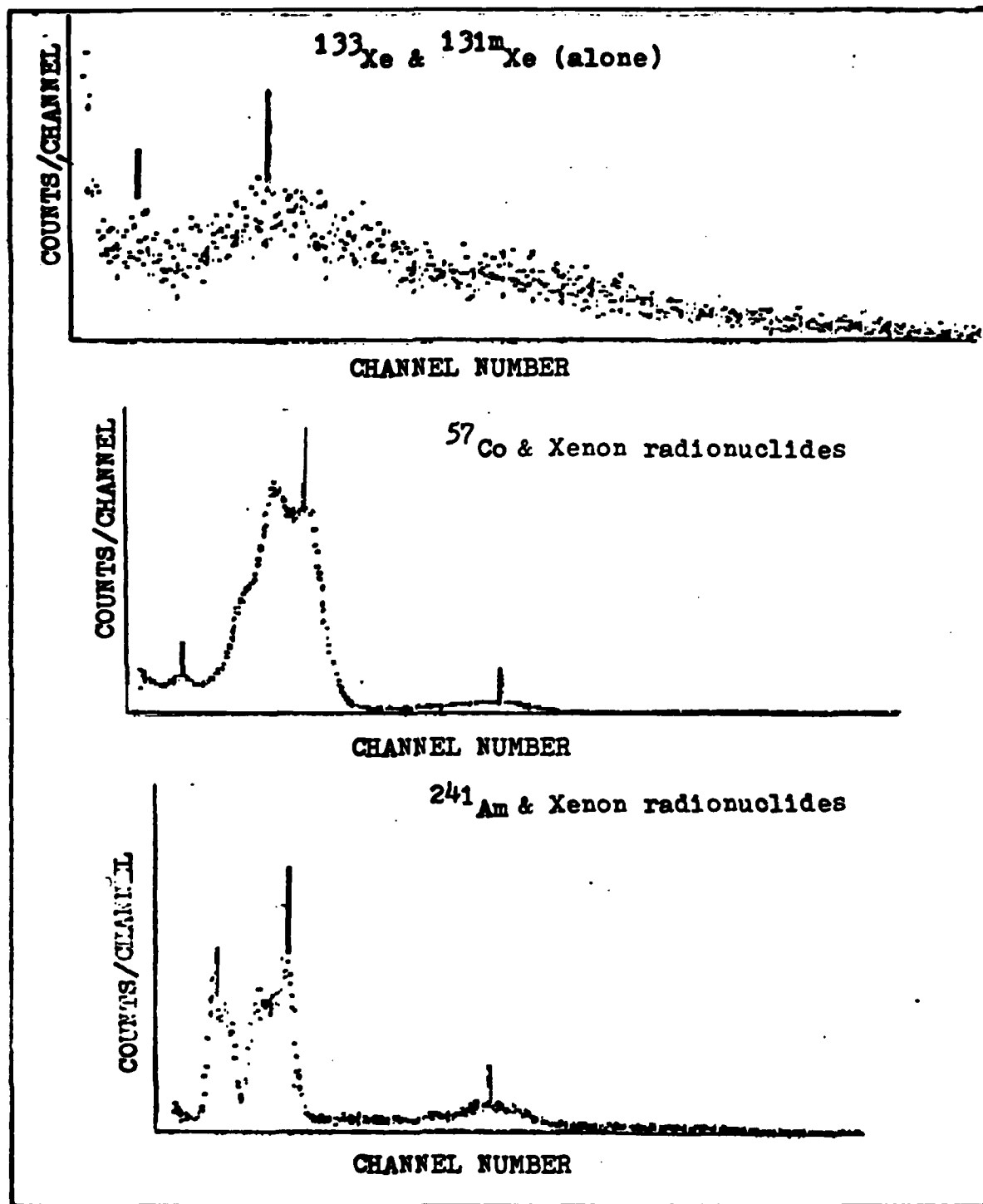


Fig. 26. Radioactive Xenon (with Grade 4.5 Stable Xenon) at 20 Atm and 3500 Volts with Various External Sources

Appendix D: Factors Used to Calculate
Intrinsic Efficiencies

<u>Gas</u>	<u>Pressure</u> <u>(Atm)</u>	<u>Volts</u> <u>(KV)</u>	<u>Source</u>	<u>Activity</u> <u>(dis/sec)</u>	<u>tc</u> <u>(sec)</u>	<u>Net Counts</u> <u>(source FEP)</u>
Ar	5	1.6	Am	3.6384E+8	400	101380
Ar	20	2.7	Am	3.6383E+8	400	308520
	20	2.7	Cd	9.0226E+5	400	181
Ar	50	3.5	Am	3.6379E+8	400	658483
	50	3.5	Cd	8.9016E+5	800	647
	50	3.5	Co	9.1715E+5	400	10694
Xe	5	2.0	Am	3.6368E+8	400	688604
	5	2.0	Cd	7.7779E+5	1200	4487
	5	2.0	Co	7.6720E+5	400	18916
Xe	10	2.7	Am	3.6368E+8	400	676085
	10	2.7	Cd	7.7896E+5	4000	29081
	10	2.7	Co	7.6916E+5	400	26823
Xe	20	3.5	Am	3.6369E+8	400	608340
	20	3.5	Cd	7.9429E+5	1000	12938
	20	3.5	Co	7.9509E+5	400	58942

The above factors were used in Equation (4) to
solve for the experimental intrinsic efficiency.

Bibliography

1. Andrews, Wayne L. Evaluation of a Detection System Employing Two Semiconductors for the Analysis of Radioactive Noble Gases. Unpublished MS thesis. School of Engineering, Air Force Institute of Technology (AU), Wright-Patterson AFB OH, March 1982.
2. Campion, P. J. "A Study of Proportional Counter Mechanisms," International Journal of Applied Radiation and Isotopes, 19: 219-234 (1968).
3. Charles, M. W. "Gas Gain Measurements in Proportional Counters," Journal of Physics E: Scientific Instruments, 5: 95-100 (1972).
4. Charles, M. W. and B. A. Cooke. "Proportional Counter Resolution," Nuclear Instruments and Methods, 61: 31-36 (1967).
5. Cockroft, A. L. and S. C. Curran. "The Elimination of End Effects in Counters," The Review of Scientific Instruments, 22: 37-42 (January 1951).
6. Curran, S. C. and J. D. Craggs. Counting Tubes. Academic Press, London, 1949.
7. Gibbons, M. R. Evaluation of a Gridded Ionization Chamber for the Detection of Radioactive Noble Gases. Unpublished MS thesis. School of Engineering, Air Force Institute of Technology (AU), Wright-Patterson AFB OH, March 1984.
8. Gibbs, D. S. et al. "Purification of the Rare Gases," Industrial and Engineering Chemistry, 48: 289-296 (1956).
9. Gold, Raymond and Edgar F. Bennett. "Electron Multiplication Process in Proportional Counters," Physical Review, 147 (1): 201-213 (July 1966).
10. Hendricks, R. W. "The Gas Amplification Factor in Xenon Filled Proportional Counters," Nuclear Instruments and Methods, 61: 31-36 (1968).

11. Hunt, Kenneth K. Analysis of a Semiconductor Detection System for Measuring Radioactive Noble Gases. Unpublished MS thesis. School of Engineering, Air Force Institute of Technology (AU), Wright-Patterson AFB OH, December 1976.
12. John, G. Unpublished class notes on Nuclear Physics (NE 6.51); Wright-Patterson Air Force Base, Ohio: Air Force Institute of Technology, 1985.
13. Knapp, J. J. Study of the Characteristics of High Pressure Proportional Counters for the Detection of Radioactive Noble Gases. Unpublished MS thesis. School of Engineering, Air Force Institute of Technology (AU), Wright-Patterson AFB OH, March 1983.
14. Knoll, Glenn F. Radiation Detection and Measurement. New York: John Wiley and Sons, 1979.
15. Kocher, D. C. Radioactive Decay Data Tables. Technical Information Center, U.S. Department of Energy, 1981.
16. Lackey, R. E. Evaluation of a High Pressure Proportional Counter for the Detection of Radioactive Noble Gases. Unpublished MS thesis. School of Engineering, Air Force Institute of Technology (AU), Wright-Patterson AFB OH, March 1985.
17. Lederer, C. M. et al. Table of Isotopes (Seventh Edition). New York: John Wiley and Sons, 1978.
18. Lucyshyn, W. Evaluation of a High Pressure Proportional Counter for the Detection of Radioactive Noble Gases. Unpublished MS thesis. School of Engineering, Air Force Institute of Technology (AU), Wright-Patterson AFB OH, March 1985.
19. McMaster, W. H. et al. Compilation of X-ray Cross Sections. UCRL50174 Sec. II, Rev. 1. Livermore: Lawrence Radiation Laboratory, University of California, 1969.
20. Povinec, Pavel. "A Study of Proportional Counter Optimization for Long-Term Counting," Nuclear Instruments and Methods, 163: 363-368 (1979).
21. Siegbahn, Kai. Alpha-, Beta-, and Gamma-Ray Spectroscopy. Volumes I and II. Amsterdam: North Holland Publishing Company, 1966.

22. Sipila, Heikka. "Energy Resolution of the Proportional Counter," Nuclear Instruments and Methods, 133: 251-252 (1976).
23. Sipila, Heikka. "Inert Gas Mixtures for Proportional Counters," Nuclear Instruments and Methods, 140: 389 (1977).
24. Walker, F. W. et al. Chart of the Nuclides, 13th Edition, General Electric Company Nuclear Energy Operations. San Jose: The General Electric Company, 1984.
25. Williams, A. and R. I. Sara. "Parameters Affecting the Resolution of a Proportional Counter," International Journal of Applied Radiation and Isotopes, 13: 229-238 (1962).

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