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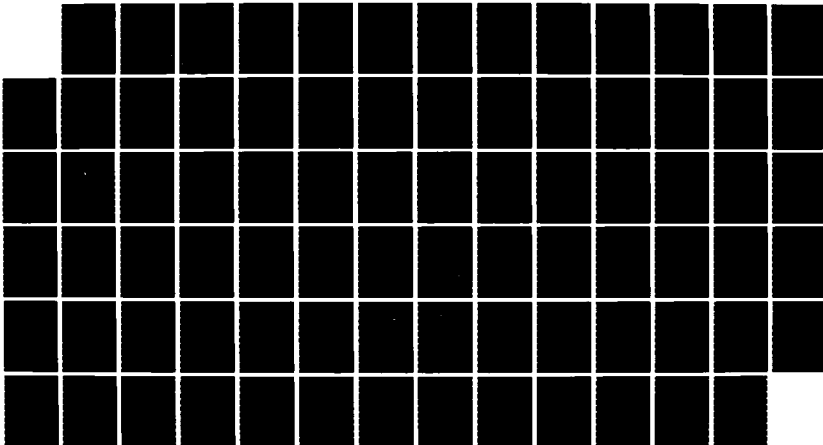
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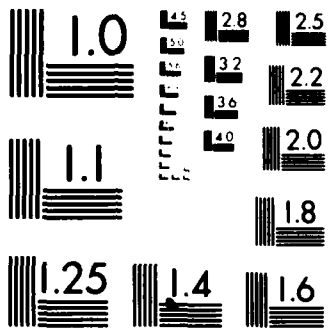
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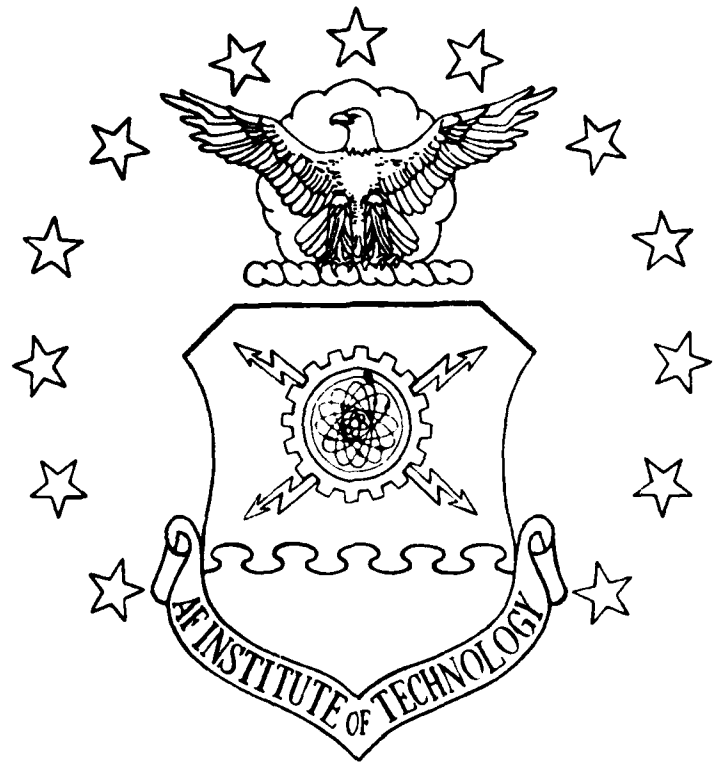




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 Richard C. Runchey
 Second Lieutenant, USAF
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A TREATISE ON THE MEASUREMENT OF
RADIOACTIVE ARGON IN THE ATMOSPHERE

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

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March 1984

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Preface

Large amounts of radioactive argon are produced in an underground nuclear explosion by neutron interaction with the surrounding soil. If the underground explosion is not properly contained, radioactive argon, along with other gaseous radionuclides and, in severe cases, particulate radionuclides, will escape (vent) into the surrounding atmosphere and disperse. Thus, a study has been undertaken to determine the feasibility of using ^{37}Ar and ^{39}Ar activity measurements from atmospheric samples as a means of monitoring the venting of radionuclides from underground nuclear explosions.

This study involved months of literature search, study and review of pertinent information dealing with many topics of interest. I would like to thank my advisor, Major John Prince, for his continual guidance and thoughtful discussions on many topics which were not clearly understood at first. I am very grateful to Linda Stoddart, reference librarian, for her assistance in locating scarce literature materials. Finally, I would like to thank Mom and Dad for their continual encouragement during the difficult times of this project.



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Abstract

A study was carried out to determine the feasibility of using ^{37}Ar and/or ^{39}Ar activity measurements from remote atmospheric samples for monitoring the possible venting of radio-nuclides from underground nuclear explosions. The study was broken into two parts; 1) a study of the processes for extracting and purifying argon from the remote air samples, and 2) an investigation of the use of mass spectrometry and low-level counting techniques for measuring ^{37}Ar and ^{39}Ar in the purified argon samples.

Argon can be extracted from air using a combination of gas-solid chemical reactions and low-temperature gas chromatography. It was found that mass spectrometry cannot be used to measure ^{39}Ar in the atmosphere. At present, low-level internal gas proportional counting is the only feasible means for measuring atmospheric levels of ^{37}Ar and ^{39}Ar .

It is concluded that it is not currently feasible to use ^{37}Ar or ^{39}Ar measurements for monitoring purposes. Further studies are needed on radioargon production sources and release rates into the atmosphere.

A TREATISE ON THE MEASUREMENT OF
RADIOACTIVE ARGON IN THE ATMOSPHERE

I. Introduction

Purpose

The measurement of the specific activity of radioactive argon isotopes present in the atmosphere presents a difficult challenge. If the challenge were successfully met, the results could reveal important information concerning current and prior world-wide nuclear processes. The main purpose of this thesis is to investigate the techniques needed to detect and measure ^{37}Ar and ^{39}Ar in remote (far away from source) atmospheric samples. A second goal is to determine the feasibility of using ^{37}Ar and/or ^{39}Ar measurements for monitoring the possible venting of radionuclides from underground nuclear explosions.

Background

Argon is a member of the chemical group known as the inert gases. Dry atmospheric air contains about 0.94 percent by volume of argon (29:B-6). Naturally occurring argon consists of three stable isotopes of mass number 36, 38 and 40 with abundances of 0.337%, 0.063% and 99.6%, respectively. In addition, nine unstable isotopes of mass number ranging from 33 to 44 are known to exist. Only ^{37}Ar , ^{39}Ar and ^{42}Ar have half-lives long enough to be present in remote atmospheric samples (28:22).

The most important sources for the production of argon radioisotopes found in the atmosphere are (14:69):

- cosmic radiation and secondaries interacting with air
- nuclear reactors and reprocessing plants
- atmospheric and underground nuclear explosions

The argon radioisotopes ($A=37,39,42$) are produced mainly from neutron interactions with ^{40}Ar atoms in the air and from neutron interactions with soil. The amount of ^{42}Ar produced relative to ^{37}Ar and ^{39}Ar is negligible. For this study, it is assumed that the ^{37}Ar and ^{39}Ar isotopes are produced from neutrons from underground nuclear explosions which interact with soil through the nuclear reactions $^{40}\text{Ca}(n,\alpha)^{37}\text{Ar}$ and $^{39}\text{K}(n,p)^{39}\text{Ar}$. These radioisotopes subsequently vent from the explosion site into the atmosphere and are dispersed by atmospheric circulation processes.

Both ^{37}Ar and ^{39}Ar are produced continuously by cosmic-ray interactions with the atmosphere. The half-lives of these isotopes (35.02 days and 269 years, respectively (12:81,87)) are long enough so that a steady-state production level is attained. Although ^{37}Ar production is somewhat dependent on solar activity, the specific activity of ^{37}Ar in the atmosphere is estimated to be 0.003 (\pm factor of 3) dpm/l-Ar at standard temperature and pressure (11:600). For ^{39}Ar , the specific activity is estimated to be about 0.10 dpm/l-Ar (21:486).

Problem and Scope

To measure ^{37}Ar and ^{39}Ar in the atmosphere, a system is required which will optimize both sample preparation and measurement procedures. The system should be able to operate in the laboratory in a production-line fashion so that argon measurements can be made on many given samples in as timely and inexpensive a manner as possible. The problem in developing this system can be broken into two parts. The first part deals with the processes that can be used to extract and purify the argon contained in the atmospheric sample. The second part deals with the actual detection and measurement of the ^{37}Ar and ^{39}Ar in the purified argon sample.

To measure ^{37}Ar and ^{39}Ar in the atmosphere, the argon contained in the air sample must first be extracted and purified. Impurities in the argon sample (mainly oxygen and nitrogen) must be removed for the detection system to operate properly. The extraction and purification processes to be reviewed generally fall into two categories, physical and chemical. Physical methods include gas chromatographic adsorption processes and low temperature gas separation by liquefaction and rectification. Chemical means include gas-solid chemical reactions and catalytic processes (31:29).

Radioactive isotopes can be detected by either directly measuring the number of atoms present in the sample or by directly observing the radiations emitted by their decay processes. In this study, mass spectrometry will be investigated as a means of measuring ^{39}Ar by counting the number of atoms

present in a sample. Low-level counting techniques will be investigated as means of measuring ^{37}Ar and ^{39}Ar by direct observation of their radiative decays.

Mass spectrometry has been suggested as a possible alternative to counting methods for the measurement of ^{39}Ar in the atmosphere (24). Mass spectrometry is used in many cases when the radioisotope to be measured has a long half-life and the sample size is small. In these cases, the specific activity is often so low that even low-level counting techniques are not sensitive enough to measure the isotope. However, given the maximum abundance sensitivity of conventional mass spectrometers, it will be shown that mass spectrometry cannot be used to measure low activity levels of ^{39}Ar present in remote atmospheric samples.

Because of the expected low specific activity levels of ^{37}Ar and ^{39}Ar in the atmosphere, low-level counting techniques must be employed. The term low-level counting is commonly used whenever small activities of radioisotopes are measured by direct observation of radioactive decay. Low-level counting systems must be applied to the problem of measuring ^{37}Ar and ^{39}Ar in the atmosphere since measurements of sufficient accuracy cannot be obtained in reasonable counting times by conventional counting systems. The major reason for this is that the source count rate (^{37}Ar or ^{39}Ar) is much smaller than the normal background count rate of conventional systems. Thus, low-level counting systems are designed to maximize the ratio of source count rate to background count rate.

Sequence of Presentation

Chapter II includes information dealing with the physical properties of argon and the production mechanisms and decay characteristics of ^{37}Ar and ^{39}Ar . Chapter III contains a review of some common extraction and purification processes for obtaining argon from air samples. The detection and measurement of ^{37}Ar and ^{39}Ar is discussed in detail in Chapter IV. Topics to be discussed include 1) the possible use of mass spectrometry for measuring ^{39}Ar in the atmosphere, 2) principles of low-level counting, 3) criteria used for choosing a low-level detection system for ^{37}Ar and ^{39}Ar , and 4) a survey of experimental work dealing with the measurement of ^{37}Ar and ^{39}Ar in the atmosphere. Chapter V concludes the thesis with a brief summary and recommendations for possible future use of ^{37}Ar and ^{39}Ar measurements in the atmosphere.

II. Characteristics of Argon

Introduction

Argon, from the Greek term argos (inactive), is the third member of the family of inert gases. It is located in the Periodic Table as chemical element number 18 and is commonly written with the symbol Ar. Natural argon has an atomic weight of 39.948 and consists of three stable isotopes of mass numbers 36, 38 and 40 which occur in abundances of 0.337%, 0.063% and 99.6%, respectively. In addition, nine unstable isotopic species of mass numbers ranging from 33 to 44 are known to exist (28:22).

Argon now comprises about 0.94% by volume of dry atmospheric air (29:B-6). Argon originates mainly in the upper layers of the earth's crust, from which it gradually escapes and mixes with the atmosphere (2:51). Of the stable argon isotopes, ^{36}Ar and ^{38}Ar are mostly primordial in origin, although they are produced in extremely small amounts by various nuclear reactions brought about by cosmic radiation and other sources of high-energy particles. The most abundant argon isotope, ^{40}Ar , is continuously being formed in the earth by the radioactive decay of ^{40}K , an isotope of potassium that is widely distributed throughout the earth's crust. The concentration of atmospheric argon is slowly increasing, although the increase amounts to only about 1.5×10^{-10} % per year of the amount of argon now present in the atmosphere.

Production of ^{37}Ar and ^{39}Ar

Table I presents the most important production mechanisms and their associated nuclear reactions for ^{37}Ar and ^{39}Ar . The half-lives of ^{37}Ar and ^{39}Ar (35.02 days and 269 years, respectively) are long enough so that the specific activity produced by cosmic-rays is at a steady-state level in the atmosphere. These levels have been estimated using various models of production mechanisms and atmospheric mixing processes. For ^{37}Ar , the specific activity in the troposphere is estimated to be from 0.002 dpm/l-Ar to 0.003 dpm/l-Ar with uncertainties up to a factor of three (17:25;11:600). For ^{39}Ar , the specific activity level in the atmosphere is estimated to be about 0.10 dpm/l-Ar (21:486).

Table I
Principal Production Mechanisms and Related
Nuclear Reactions for Ar37 and Ar39

Production Mechanism	Nuclear Reaction	
Cosmic radiation and secondaries	$^{36}\text{Ar}(n,\gamma)^{37}\text{Ar}$	$^{40}\text{Ar}(n,2n)^{39}\text{Ar}$
	$^{40}\text{Ar}(n,4n)^{37}\text{Ar}$	$^{40}\text{Ar}(\gamma,n)^{39}\text{Ar}$
Atomic reactors and atmospheric nuclear explosions	$^{36}\text{Ar}(n,\gamma)^{37}\text{Ar}$	$^{40}\text{Ar}(n,2n)^{39}\text{Ar}$
Underground nuclear explosions	$^{40}\text{Ca}(n,\alpha)^{37}\text{Ar}$	$^{39}\text{K}(n,p)^{39}\text{Ar}$

(13:191,196;14:69-70)

In addition to the natural cosmic-ray production, ^{37}Ar and ^{39}Ar can also be produced artificially in nuclear reactors and nuclear explosions. Much greater quantities of ^{37}Ar and ^{39}Ar are produced from an underground nuclear explosion than are produced from an atmospheric nuclear explosion of equivalent yield or from nuclear reactors (17). It is believed that fluctuations observed in the measured ^{37}Ar activity of air samples taken world-wide from 1969 to 1973 were due to the venting of ^{37}Ar from underground nuclear explosions (17:26-27). No such study has been carried out dealing with ^{39}Ar , although it has been concluded that the contribution of ^{39}Ar to the global atmospheric specific activity level from underground nuclear explosions up to 1967 is less than 0.005 dpm/l-Ar (13:193).

Nuclear Decay Data

The decay schemes of ^{37}Ar and ^{39}Ar are shown in Figures 1 and 2. With a half-life of 35.02 days, ^{37}Ar decays by electron capture to the ground state of ^{37}Cl which then emits 2.8 keV x-rays and Auger electrons (12:81). With a half-life of 269 years, ^{39}Ar emits β^- -particles which have an endpoint energy of 565 keV (12:87).

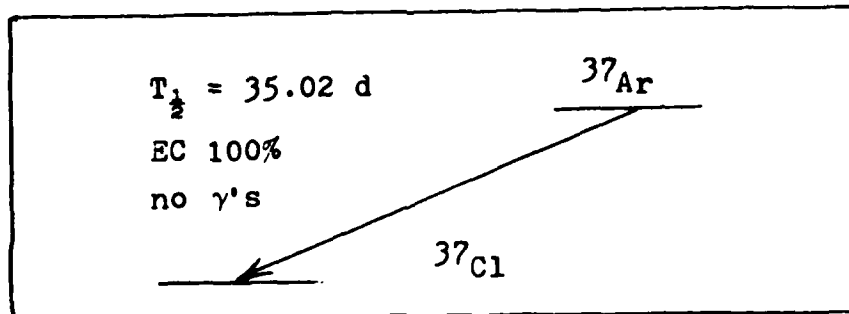


Fig. 1. Argon-37 Decay Scheme (12:80)

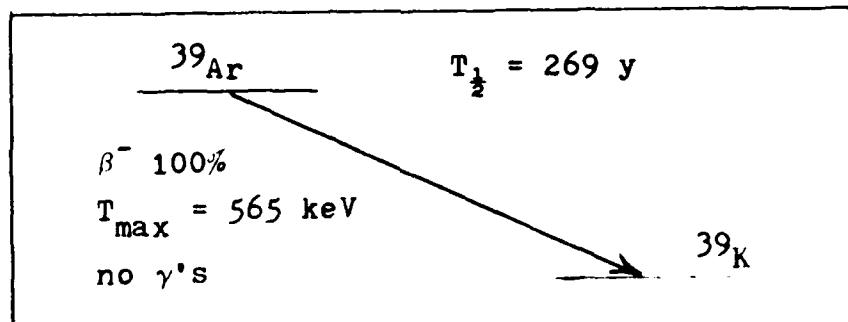


Fig. 2. Argon-39 Decay Scheme (12:88)

III. Extraction and Purification of Argon

Introduction

To measure radioactive argon in atmospheric samples by mass spectrometry or by counting methods, it is necessary to first extract the argon from the air sample and then remove trace impurities. The final system should operate in a production-type mode, extracting and purifying argon with as high a yield as possible from a number of given atmospheric samples. In addition, the system should not be overly time consuming or expensive.

For measuring ^{39}Ar in the atmosphere by mass spectrometry, any atomic or molecular impurity in the extracted argon sample will reduce the sensitivity for measuring abundance ratios for adjacent masses (30:26). The main mechanism for this reduction in sensitivity is the small-angle scattering of the primary ion beam from impurities in the sample or in the vacuum analyzer.

For radioactive counting methods based on ionization, it is important to remove electronegative impurities (mainly oxygen and nitrogen) from the extracted argon sample (1:186). Electronegative gases can bind the primary electrons produced by ionizing radiation interacting in the sensitive detector material. Negative ions are formed which have much smaller drift velocities than the electrons (roughly 1000 times smaller). These do not contribute to the buildup of the pulse at the anode of the detector, resulting in smaller

pulses and apparently lower gas amplification. The net sample count rate will be reduced. For measuring ^{37}Ar and ^{39}Ar in the atmosphere, it will be shown in Chapter IV that the net sample count rate must be kept as high as possible.

In this chapter, the most common processes used for extracting and purifying argon from air are briefly discussed. As one can see from Table II, the major problem is separating the small amount of argon from the larger amounts of oxygen and nitrogen. The processes used generally fall into two categories, chemical and physical (31:29). Chemical methods

Table II
Components of Dry Atmospheric Air

Component	Content (%) by volume	Content (ppm) by volume
N_2	78.084 \pm 0.004	
O_2	20.084 \pm 0.002	
Ar	0.934 \pm 0.001	
CO_2	0.003 \pm 0.001	
Ne		18.18 \pm 0.04
He		5.24 \pm 0.004
CH_4		2
Kr		1.14 \pm 0.01
H_2		0.5
N_2O		0.5 \pm 0.1
Xe		0.087 \pm 0.001

(29:F-172)

include gas-solid chemical reactions and catalytic combustion. Physical methods include gas chromatographic adsorption and low temperature separation by liquefaction and rectification of air. Following this brief discussion of common means for extracting and purifying argon from air, two actual laboratory apparatuses used for obtaining pure argon from air are reviewed.

Gas-Solid Chemical Reactions

A common chemical method used for eliminating oxygen and/or nitrogen from argon is metal "gettering" reactions (31:29-30). Gettering refers to a column or tube usually filled with a metal (sometimes a liquid). An impure gas sample is then injected into the column resulting in chemical reactions between the metal and the impurities of the sample.

Alkali and alkaline earth metals have the greatest affinity for oxygen. These metals (usually heated to high temperatures) will be oxidized when brought into contact with oxygen, forming oxides which are stable and refractory. Nitride formation runs similarly although alkali metals and barium tend to form unstable (explosive) azides rather than nitrides. Generally, the affinity of metals for nitrogen is less than that for oxygen and thus increased temperatures are required in the process of nitrogen removal than is needed for oxygen removal.

Gibbs, Svec and Harrington (7) investigated some 15 metals and alloys for their relative efficiencies in the

removal of oxygen and nitrogen from argon. They found that aluminum, cast-iron, titanium and zinc were not satisfactory for removing both oxygen and nitrogen. Barium, calcium, calcium-10% magnesium, magnesium, thorium and zirconium satisfactorily removed nitrogen. Uranium was found to be efficient for removing oxygen but not nitrogen.

Catalytic Combustion

Some commercial air separation facilities remove oxygen from argon by catalytic combustion with hydrogen (2:421). The oxygen-argon mixture is placed in a column and hydrogen is then added. The column is heated causing catalytic combustion which produces water vapor. This process could be used on a laboratory scale for removing small impurities of oxygen, although gas-solid chemical reactions or physical adsorption processes are generally preferred.

Gas Chromatography - Molecular Sieves

Gas chromatography principles are very similar to gettering reactions discussed previously. A column is filled with a suitable material, and an impure gas sample is injected into the column. The main difference is that no chemical reactions take place to remove impurities of the gas. The principle of gas chromatographic adsorption is that different chemical elements will pass from one end of the column to the other at different speeds. Argon can be effectively separated from oxygen and nitrogen by choosing proper adsorbing materials (8:82-84).

Synthetic crystalline zeolites (Type A) are the best materials for separating argon from oxygen and nitrogen by gas chromatography. These zeolites are crystalline aluminosilicates which have been dehydrated (2:225). After dehydration the crystals are interlaced with regularly spaced channels on the order of molecular dimensions. A given crystal will permit some molecules or atoms to enter the channels, or pores, but not others, thus giving the zeolites the common name "molecular sieves." The properties of the atom or molecule which determine whether or not it can enter the pores are its size, shape and structure. Molecular sieve Type 5A (a calcium zeolite) is widely used in the separation of argon from oxygen and nitrogen. At low temperatures, the argon atoms are able to enter the pores and pass through the column at a faster rate than oxygen or nitrogen. Evacuated chambers collect the various gases as they are eluted from the end of the column.

Liquefaction and Rectification

Low temperature liquefaction and rectification of air for producing large quantities of pure oxygen, nitrogen and argon is carried out by commercial air separation plants (2:413). Laboratory-scale production of argon by this technique is difficult because of the low temperatures involved (around -200°C for liquid nitrogen) and because the extraction yield of argon would be very small. However, liquefaction and rectification can be used to enrich the argon concentration

of the air sample by removing the constituents of air having a lower boiling point than argon (e.g. nitrogen and hydrogen).

The basic principle underlying rectification is that if a liquid and a vapor having the same composition are brought into contact, the composition of the liquid and gaseous phases will move apart. The liquid will contain more of the component of the mixture with the higher boiling point and the vapor will contain more of the component with the lower boiling point (25:74-75). For example, in the case of air, nitrogen has a boiling point of -195.8°C , argon -185.7°C and oxygen -183.0°C (29:back fly-leaf). Rectification in the laboratory can be used to separate nitrogen from an argon-oxygen mixture. The process is usually carried out in a vertical rectifying column. A current of gas (nitrogen) flowing upwards and a stream of liquid (argon-oxygen) flowing downwards are brought into contact on the surface of material in the column. The vapor, as it flows upwards, will become enriched in nitrogen, while the liquid flowing downwards will become enriched in argon and oxygen.

Laboratory Systems for Purifying Argon

In this section two laboratory systems for extracting and purifying argon from air samples are reviewed. Both systems employ chemical and physical means for obtaining pure argon from an air sample. The first system is used at the University of Washington (UW); the second system is used at the National Bureau of Standards (NBS) in Washington D.C.

UW Argon Purification System. Schell and co-workers at UW have developed a system for argon extraction and purification from air which uses cryogenic rectification for removal of nitrogen, chemical reactions with copper for removal of oxygen, and final purification by low-temperature gas chromatography (26:456-458). A block diagram of the apparatus is shown in Figure 3.

The air sample is bled into an evacuated condensation trap under the cryogenic still and liquefied by a liquid nitrogen flask surrounding the trap. The still is four feet long and consists of 1) an inner-tube 1-in. in diameter, filled with stainless steel wool, 2) a concentric 3/8-in. annulus containing liquid nitrogen, and 3) a vacuum jacket to minimize heat losses. Argon enrichment begins when the liquid nitrogen flask surrounding the condensation trap is removed, allowing the liquefied air to boil. Nitrogen is vaporized first and as it rises up the still is condensed. The condensing liquid flows downward interacting with the rising vapor. A 10 psi relief valve allows nitrogen arriving at the top of the still to be removed. A sample of the vented nitrogen is measured every 15 min by the gas chromatographic columns. The process is completed (about 2 hours) when a decrease in the N_2 peak or when an increase in the Ar- O_2 peak is observed out of the columns. The gas in the still is recondensed into the condensation trap which now contains an enriched argon-oxygen mixture and some nitrogen. This mixture is passed through a heated ($500^{\circ}C$) copper furnace for removal of oxygen and is

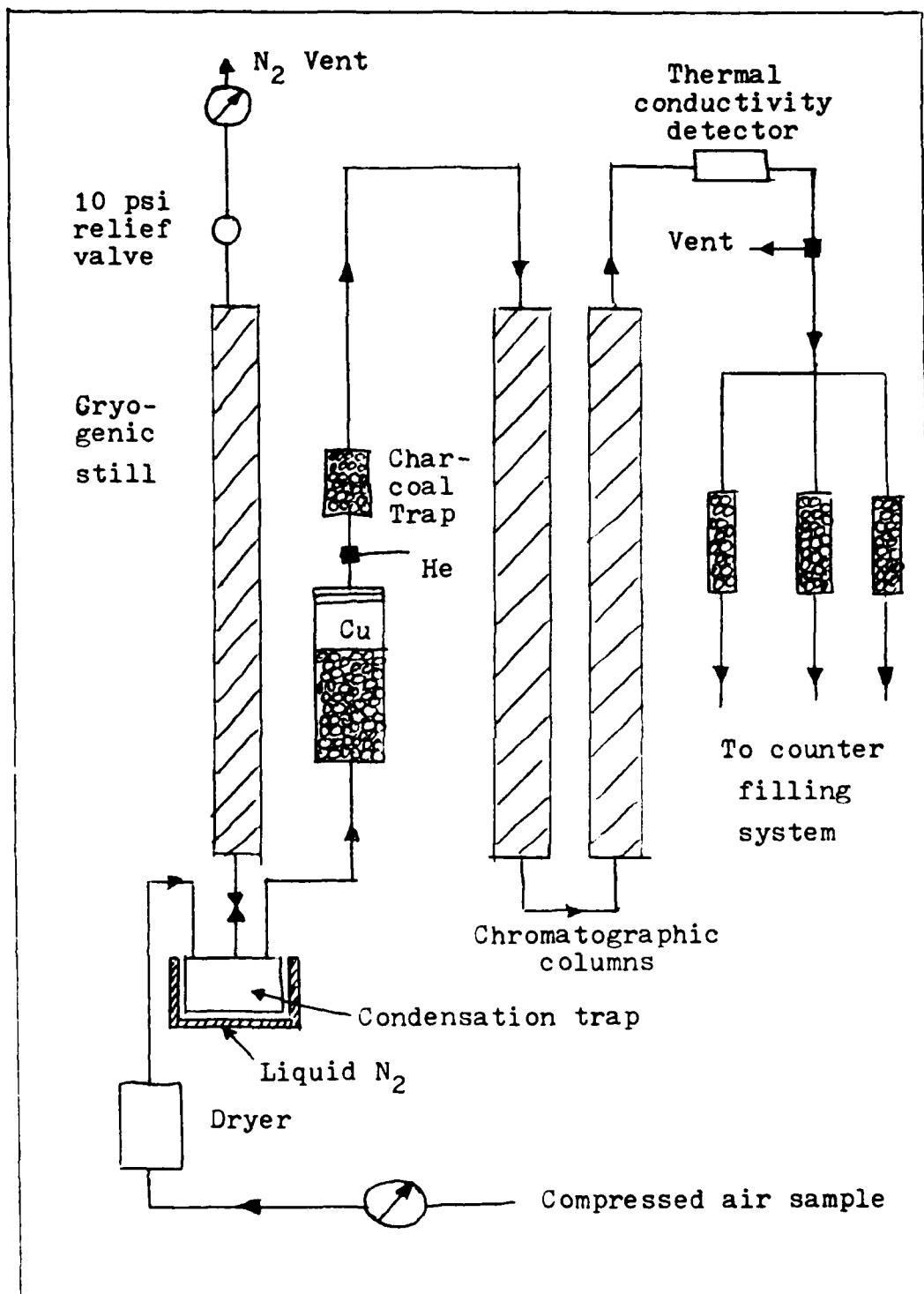


Fig. 3. UW Gas-Purification System for Argon (26:456)

then condensed onto a charcoal trap. The final purification is done with two high-pressure gas chromatographic columns. The first column contains 400g of Type 5A molecular sieve; the second column contains 70g of charcoal and 300g of Type 5A molecular sieve. The flow path is from top to bottom of the first column and from bottom to top of the second column. A sample gas injection is made by either evacuating the first column and absorbing the sample or by passing helium through the heated charcoal trap. The columns are initially cooled to liquid nitrogen temperatures. The helium elution pressure is adjusted to 50 psi and elution of the sample starts as the columns begin to warm under a temperature control program. This program reduces the time needed to elute the purified argon from the columns. The argon is effectively separated from oxygen and nitrogen at low temperatures in about 100 min. The purified argon fraction is collected in a charcoal trap activated when the argon peak is first detected by a thermal conductivity detector placed at the end of the second column. The argon is then transferred to a counter filling system for radioactive counting.

NBS Argon Purification System. The NBS system for extracting and purifying argon from air samples uses chemical reactions with calcium carbide (CaC_2) and titanium for removal of both nitrogen and oxygen followed by gas chromatography for separation of the noble gases. A diagram of the system is shown in Figure 4 (4:48).

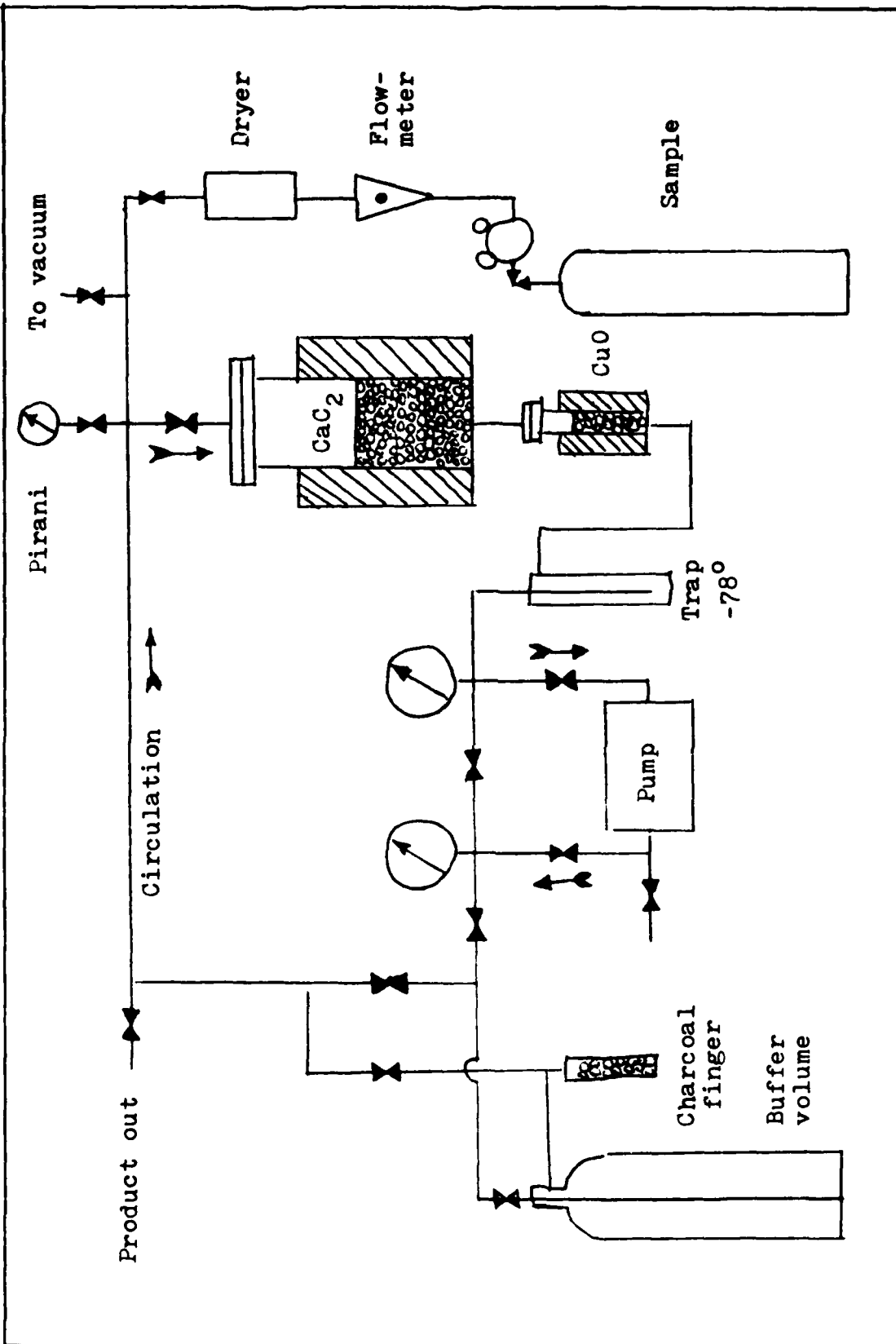


Fig. 4. NBS Gas-Purification System for Argon (4:48)

A 320 l sample of air enters slowly from the sample tank through a flow meter and molecular sieve dryer into a CaC_2 reactor bed. The bed contains 2.5 kg of finely ground CaC_2 which is degassed overnight while heating to an operating temperature of 800°C . The flow rate of sample gas into the reactor bed is moderated at 300 l per 4 hr. After interacting with the CaC_2 , the gas flows through a hot copper-oxide (CuO) reactor to remove hydrogen and hydrocarbons. The gas, which now contains about 84% argon, is collected into a buffer volume. Final purification of the argon is accomplished by contact with titanium cooling from 800°C to 25°C followed by gas chromatography using activated charcoal at -30°C to -20°C to separate the argon from the other noble gases.

Summary

Both the UW and the NBS argon purification systems can be used in the laboratory in a production-type fashion. Both employ a combination of chemical and physical separation processes. To measure low activity levels of ^{37}Ar and ^{39}Ar in atmospheric samples, it is necessary to extract and purify as much of the argon from the samples as possible. In the case of remote air samples, the quantity of air collected will often be limited, and thus the extraction yield for argon must approach 100%. In these cases, the NBS system is preferred because very little argon is lost during the process. In cases where given air samples are large and extraction yield not important, the UW system is preferred over the NBS system since the time needed for the process is much less.

IV. Measurement of ^{37}Ar and ^{39}Ar

Introduction

The detection and measurement of low activity levels of ^{37}Ar and ^{39}Ar in the atmosphere presents a difficult challenge. If successfully met, the results of this challenge could reveal important information concerning current and prior world-wide nuclear processes. The purpose of this chapter is to investigate the techniques needed to measure ^{37}Ar and ^{39}Ar present in atmospheric samples.

In this chapter, mass spectrometry and low-level counting techniques are investigated as methods for measuring radioactive argon in remote atmospheric samples. Mass spectrometry is commonly used to detect radioisotopes which have long half-lives and are present in small quantities in small samples. In these cases, the specific activity of the isotopes is so low that measurement by low-level counting techniques is not feasible. The major problem to be overcome in using mass spectrometry for measuring ^{39}Ar is the interference caused by the high abundance of ^{40}Ar atoms (about 99.6%) present in the purified argon sample. This problem has yet to be overcome.

Although difficult, the specific activity levels of both ^{37}Ar and ^{39}Ar in the atmosphere can be measured by low-level counting techniques. These techniques require the combined optimization of counting procedures and sample preparation. In most low-level counting systems, this usually involves increasing the ratio of sample count rate to background count

rate. The sample count rate can be increased by either increasing the amount of sample in the counter or increasing the counting efficiency. Increasing the sample size, however, means that more time must be spent in extracting and purifying argon from air. For remote atmospheric samples, the argon sample size will usually be limited to some degree and the counting efficiency will be close to 100%. Thus, background reduction becomes the major requirement in the development of a low-level detection system for ^{37}Ar and ^{39}Ar .

At the present time, low-level internal gas proportional counting is the most feasible means for measuring atmospheric levels of ^{37}Ar and ^{39}Ar by radioactive counting methods. The majority of attention of scientists world-wide has dealt with the measurement of ^{37}Ar since it is much easier, although still difficult, to measure than ^{39}Ar . Most of the discussion and review of experimental counting methods in this chapter will thus deal with the detection and measurement of ^{37}Ar .

The topics presented in this chapter include 1) a discussion on why mass spectrometry cannot be used to measure ^{39}Ar in the atmosphere, 2) a review of principles of low-level counting, 3) some criteria for choosing a detection system for measuring ^{37}Ar and ^{39}Ar , and 4) a survey of experimental work dealing with the measurement of atmospheric ^{37}Ar and ^{39}Ar . The information and knowledge obtained in these studies will then be used in the next chapter to determine the feasibility of using ^{37}Ar and/or ^{39}Ar measuring techniques for monitoring venting of radionuclides from underground nuclear explosions.

Mass Spectrometry

As discussed previously, mass spectrometry is commonly used to detect radioisotopes of very low specific activity which are present in small samples. It was thought that mass spectrometry could be a more sensitive technique than low-level counting for the measurement of ^{39}Ar in the atmosphere. The advantages of mass spectrometry are that smaller sample sizes can be used, measuring times are much reduced, and the purification of the extracted argon fraction may not have to be as extensive as in low-level counting (24).

The following calculations were then carried out which showed that present-day mass spectrometric techniques cannot be used to measure ^{39}Ar in the atmosphere:

Given	10^9	= S	: maximum abundance sensitivity (i.e., one atom of Ar-39 can be measured in the presence of 10^9 atoms of Ar-40)
	300.0	= V	: Volume of air sample at S.T.P (l)
	4.9×10^{-9}	= λ	: Ar-39 disintegration constant (min^{-1})
	0.94 %	=	percent by volume of argon in air
	99.6 %	=	percent abundance of Ar-40 in argon

Assuming that all the argon is extracted from the air sample and using the ideal gas law $PV = nRT$ along with the percentages given above for argon in air and ^{40}Ar in argon, the number of ^{40}Ar atoms in a 300.0 l air sample is

$$N_{40} = (P \cdot V_{40} \cdot N_a) / (R \cdot T) = 6.9 \times 10^{22} \text{ atoms}$$

where V_{40} is the volume of ^{40}Ar in the sample and N_a is

Avagadro's number. The corresponding minimum number of ^{39}Ar atoms needed in the sample to be measured by mass spectrometry is

$$N_{39} = N_{40}/S = 6.9 \times 10^{13} \text{ atoms}$$

This number of ^{39}Ar atoms can be related to a minimum specific activity level of ^{39}Ar in the given air sample which is

$$SA_{\min} = N_{39} \cdot \lambda / V_{\text{argon}} = 1.2 \times 10^5 \text{ dpm/l-Ar}$$

The expected specific activity level of ^{39}Ar in remote atmospheric samples is about 0.1 dpm/l-Ar. This is a factor of 10^6 smaller than the minimum measurable level of the mass spectrometer. It is clear that with a maximum sensitivity of modern mass spectrometers of 10^9 , mass spectrometry cannot be used to measure low activity levels of ^{39}Ar in the atmosphere.

Principles of Low-Level Counting

In this section some some basic principles of low-level counting statistics are presented which allow for an estimate of the minimum sample count rates that can be determined in the presence of a specified background count rate. It will be seen that for measuring ^{37}Ar and ^{39}Ar , the amount of background from the counting system must be minimized. Various sources of background are reviewed and methods of reducing their contributions to the overall background of the counting system will be discussed.

Counting Statistics. The goal of any counting system is to obtain measurements of sufficient accuracy within reasonable counting times. A widely used concept for evaluating counting systems is the figure of merit (FOM). The FOM of a detection system is defined as $1/T$ where $T = T_{S+B} + T_B$ is the minimum combined counting time that must be spent counting a source sample and a background sample to obtain a specified precision in the net source count rate (10:140).

As a first approximation, one may consider the case of measuring the source count rate, S , from an unknown source in the presence of a steady-state background count rate, B . The accuracy of the measurement is determined only by the Poisson error of counting and excludes other sources of error such as radioactive contamination, spurious counts, etc. The measurement is usually carried out by counting the source plus background count rate, $S+B$, for a time T_{S+B} , and then counting the background count rate alone for a time T_B . The net source count rate is equal to

$$S = N_1/T_{S+B} - N_2/T_B \quad (1)$$

where N_1 and N_2 are the total counts in each measurement. The standard deviation in S is then equal to

$$\sigma_S = [(S+B)/T_{S+B} + B/T_B]^{1/2} \quad (2)$$

Assuming a constant time is used to carry out the measurements, the above uncertainty can be minimized by optimally choosing the fraction of time spent on counting $S+B$ (or B).

By squaring and differentiating both sides of Equation 2 and setting $d\sigma_g$ equal to zero, the combined counting time becomes a minimum for any given counting precision when

$$T_{S+B}/T_B = [(S+B)/B]^{\frac{1}{2}} \quad (3)$$

The FOM can then be written as

$$1/T = \epsilon^2 S^2 / [(S+B)^{\frac{1}{2}} + B^{\frac{1}{2}}]^2 \quad (4)$$

where $\epsilon = \sigma_S/S$ is the relative standard deviation in S.

In the case of measuring ^{37}Ar and ^{39}Ar in the atmosphere with low-level counting systems, the source count rate will be much less than the background count rate (i.e. $S \ll B$). Thus, the FOM in Equation 4 reduces to

$$1/T = \epsilon^2 S^2 / 4B \quad (5)$$

The FOM generally used in low-level counting systems is $S^2/4B$ (20:429). From a statistical viewpoint, the better systems have higher values of $S^2/4B$. The FOM can be improved either by increasing the source count rate or by reducing the background count rate. The source count rate can be increased by improving the counting efficiency and/or increasing the amount of sample in the detector. For measuring radioactive argon from remote air samples, the amount of sample is usually limited. The FOM can be rewritten as $e^2 f^2 A / 4B$ where e is the efficiency of the detector, f is the yield of argon extracted from the air sample, and A is the activity of the sample. The product of (ef) is usually limited and close to

one. Thus, the FOM can be increased mainly by reducing the background count rate of the detection system.

To get a feel for the time required to measure ^{37}Ar in the atmosphere, one may consider first an ideal detector (i.e., one with zero background and 100% efficiency). Using Equation 4, the time required to measure the activity level of atmospheric ^{37}Ar ($S = 0.0025$ dpm/l-Ar) with a precision of 10% is

$$T = (\epsilon^2 S)^{-1} = 40,000 \text{ min/l-Ar}$$

Obviously this is an unreasonable counting time. It can be reduced greatly by using a larger sample in the detector; for example, a 10 liter sample will reduce the time to 4000 min. The addition of a background count rate and a counting efficiency less than 100% will greatly increase the counting time (e.g., for the above case of a 10 liter sample, 10% precision, 100% efficiency, but now with a background count rate of $B = 0.1$ cpm, the counting time goes up to 72,000 min). Thus, it can be seen that low background count rates are essential for measuring low activity levels of ^{37}Ar in the atmosphere.

From Equation 4, one may calculate the minimum source count rate which is needed to obtain a specified counting precision in a specified total counting time ($T_{S+B} + T_B$) in the presence of a background count rate:

$$S = (1/(\epsilon T^{\frac{1}{2}}) + 2B^{\frac{1}{2}})/\epsilon T^{\frac{1}{2}} \quad (6)$$

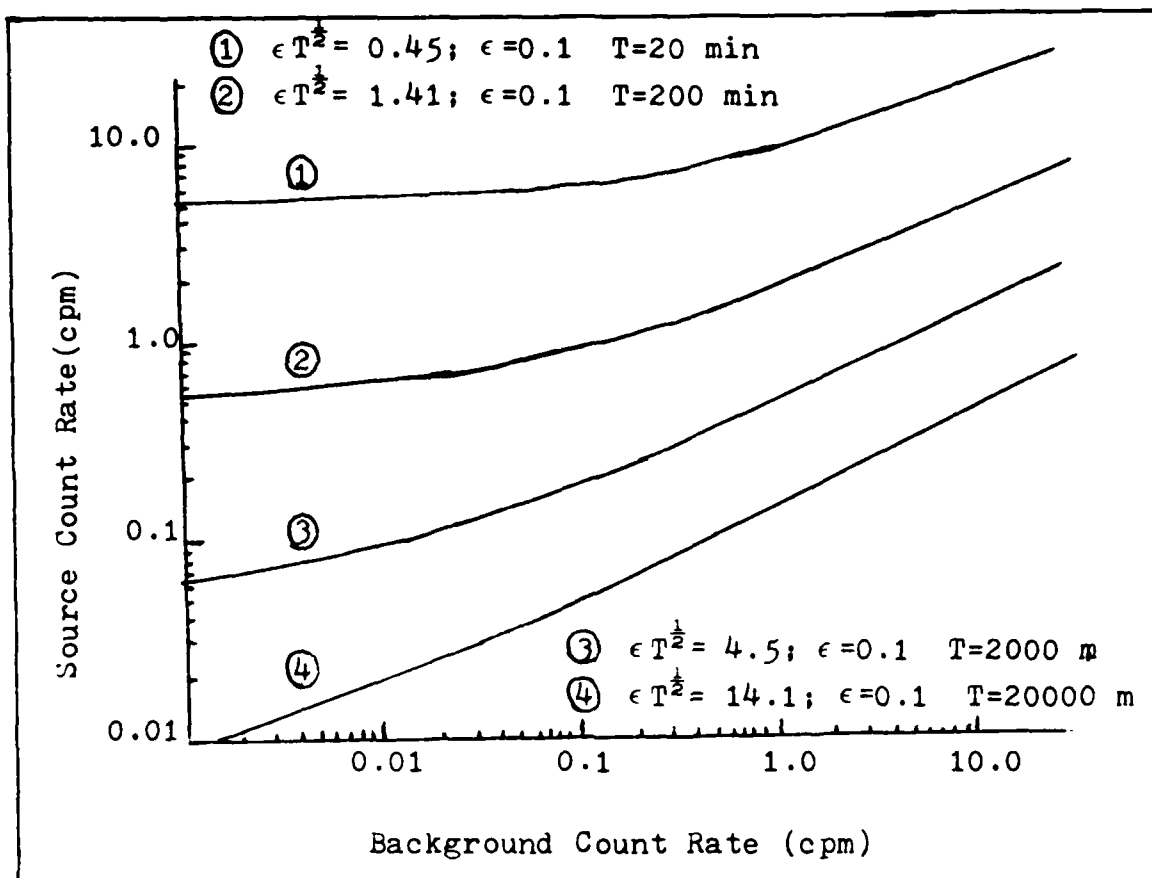


Fig. 5. Minimum Source Count Rate; $S=S(B,T,\epsilon)$ (20:431)

Using Equation 4, the above figure shows calculated source count rates as a function of background count rate for a given precision of 10% and counting times from 20 to 20000 minutes (20:431).

The statistical discussion so far has dealt only with pure poisson counting statistics. Non-Poisson errors can also be present in the detection system in the form of, for example, radioactive contamination, background variations in the muon flux, spurious counts, etc. The formal treatment of non-Poisson errors is difficult since the causes of the errors

are not distributed randomly in time. To demonstrate non-Poisson errors in low-level counting, one can assume that the non-Poisson error component in the background is distributed randomly in time with a standard deviation σ_{np} . For the case of $S \ll B$, the total standard deviation σ_{s-tot} of the net source count rate then becomes

$$\sigma_{s-tot} = [2(S+2B)/T + \sigma_{np}^2]^{\frac{1}{2}} \quad (7)$$

As long as $T \ll 2(S+2B)/\sigma_{np}^2$, the Poisson error dominates and the previous considerations hold. If, on the other hand, the total counting time is increased, the non-Poisson error begins to contribute more to the total error. Thus, increasing the counting time much above $2(S+2B)/\sigma_{np}^2$ is of no use.

To summarize this section on counting statistics, the FOM generally used for low-level counting systems is $S^2/4B$. For the case of a limited argon sample, the FOM is $e^2 f^2/4B$. Since (ef) is in most cases limited and close to one, it is essential to aim for a reduction in background of the detection system. In cases of very small activities requiring low background and very long counting times, the counting limits are determined by non-Poisson error contributions to the background count rate.

Sources of Background. All radiation detectors produce a background signal. There are two main sources of background - that resulting from ionizing radiation which interacts with the detector volume, and that resulting from pulses of differing origin (e.g., electronic noise, electromagnetic disturbances, breakdowns in high voltage insulators, etc.).

Ionizing background radiations can be grouped in the following categories (10:774-775):

- Primary and secondary components of cosmic radiation
- Natural radioactivity in the detector materials and surrounding structures
- Radioactivity in the air surrounding the system

For an unshielded counter, the cosmic radiation sets a lower limit to the total background count rate. In the case of gas chambers, the activity level from cosmic radiation is of the order of 1 cpm/cm² of detector cross-section (20:442). Muons are the major source of this cosmic background which is only slightly reduced by shielding.

A large component of the total background arises from γ -rays which interact with the detector volume and its walls. They can originate from the soft component of cosmic radiation, from radioactive impurities in the detector materials and shields, from bremsstrahlung and energetic electrons produced by charged cosmic particles, and from neutron capture by nuclei (20:443).

Natural radioactive isotopes of potassium, thorium,

uranium, and radium are often contained as impurities of ordinary construction materials of detectors, shields, and surrounding buildings (10:775). Such impurities can create severe limitations to detection systems designed for low-level measurements of radionuclides. In addition to material impurities, radioactivity in the air from trace amounts of radioactive gases or dust particles can also lead to a measurable background contribution.

Background Reduction. As mentioned previously, the most important consideration for measuring atmospheric levels of ^{37}Ar and ^{39}Ar by low-level counting is the reduction of background. General background reduction methods to be discussed in this section include the following:

1. Shielding
2. Anticoincidence
3. Pulse-Height Analysis
4. Rise-Time Discrimination
5. Underground Laboratory

1. Shielding. All background radiations except for muons arising from outside the detector volume can be significantly reduced by proper shielding (20:443). Shielding materials are used mainly to reduce the γ -flux incident on the detector. The materials used in systems for measuring ^{37}Ar and ^{39}Ar must be specially selected and radioactively pure substances (usually steel, lead, bismuth, and mercury). For example, 15 cm of lead shielding (or 25 cm of steel) can

reduce the γ -flux by at least a factor of 10^3 . Increasing the thickness further generally results in no additional background reduction because the radioactive impurities in the shield and those induced by cosmic-ray interactions in the shield become predominant. The neutron component of background can also be reduced by proper shielding. For example, evaporation neutrons produced in the γ -shields by proton and muon interactions are effectively eliminated by 15 cm of borated paraffin wax placed between the shields and the detector.

2. Anticoincidence. The highly penetrating muonic component of background can be effectively eliminated by the use of what is termed an anticoincidence shield. Actually, the anticoincidence shield is an additional detector (or array of detectors) surrounding the main detector. It is commonly referred to as a guard detector. If an incident radiation creates a pulse in both the main and guard detectors within a preset time span, the pulse out of the main detector is electronically blocked and not counted. When operated in conjunction with regular shielding, low background count rates can be achieved.

Most guard systems use Geiger-Müller (GM) tubes. If used in the proportional counting mode, these guard detectors have the advantages of much shorter rise times and virtually no losses of source counts due to dead time (20:444). If operated in conjunction with a proportional tube as the main counter, both have the same sensitivity to electrical disturbances (e.g., spurious pulses), which can thus be rejected.

Guard counters can also lead to a large reduction in the γ -induced background if additional pure shielding material is placed between the main and guard counters. A large part of the remaining background, after counts due to muons are eliminated, is the γ -rays produced by muons in the counter material and this inner shield. Pulses in the main counter from these γ -rays are accompanied by the muon pulses of the guard counter and are thus eliminated.

3. Pulse-Height Analysis. The background radiation which interacts with a detector has a wide band of energy deposition depending on the sensitive material in the detector. In comparison, the radiations from ^{37}Ar and ^{39}Ar will have a much more limited band of energy deposition. The FOM can be significantly increased by examining pulses from a counter that correspond only to the energy band of the source radiation.

The proportional counter is particularly useful for increasing the FOM by pulse-height analysis (3:207). For example, the x-rays emitted from the decay of ^{37}Ar will deposit energy in the small range around 2.8 keV compared to background radiations which have an energy deposition range up to 1 MeV. By limiting the pulses counted to the energy range from about 2 to 4 keV, a large increase in the FOM and the detection sensitivity for ^{37}Ar can be achieved.

4. Rise-Time Discrimination. A recently developed technique used for background reduction in proportional counters is rise-time discrimination. When radiation interacts

with the sensitive volume of the detector, an ionization track is produced. For a given incident radiation, a lower incident energy will result in a larger linear energy transfer (dE/dx) compared to a higher incident energy. Thus, lower energy radiations produce shorter track lengths. Rise-time discrimination makes use of the fact that the rise-time of the pulses in a proportional counter varies with track length. For example, the low energy x-rays and Auger electrons following electron capture of ^{37}Ar create very short ionization tracks compared to background radiations which produce extended tracks. The rise-times of these pulses will be different even though the same amount of energy may have been deposited. If one measures the pulse rise-times along with the pulse-heights of a certain radioisotope, one should be able to effectively discriminate between the source pulses and background pulses (20:448-449).

5. Underground Laboratory. The background count rate can be further reduced in all radiation detectors when they are operated underground. In fact, most low-level counting systems are usually operated in the basements of buildings or in specially constructed underground laboratories. The main components of background which are reduced are those resulting from γ -rays and muons (22:149). The background of low-level proportional counters have been reduced by factors of 2 to 4 when operated underground (about 70 m water equivalent below surface). The remaining background is due to radioactive impurities of the counter construction materials

Choosing a Detection System

The goal of the detection system is to measure the activity levels of ^{37}Ar and ^{39}Ar in the atmosphere with the best possible accuracy within reasonable counting times. The best system will optimize both the counting procedure and the sample preparation. The major criteria for choosing a system for measuring ^{37}Ar and ^{39}Ar include the following (20,432):

- Activity levels to be measured
- Radiations to be detected
- Nature and quantity of sample to be collected
- Counting efficiency and background levels to be needed

The minimum specific activity levels to be measured are about 0.0025 dpm/l-Ar and 0.10 dpm/l-Ar for ^{37}Ar and ^{39}Ar , respectively. The radiations to be detected include 2.8 keV (total energy) x-rays and Auger electrons from the decay of ^{37}Ar and the spectra of energetic β^- -particles ($T_{\text{max}} = 565\text{keV}$) from the decay of ^{39}Ar . The sample to be collected is pure argon gas. The quantity of the sample is usually limited and is assumed to be from a 1 m³ atmospheric sample (24) (i.e., a maximum quantity of about 10 liters of pure argon). The counting efficiency must be as high as possible and background levels as low as possible for a given sample size and detector.

The major consideration concerns the nature of the sample. Argon can easily be prepared for use as the sensitive volume of an ionization detector. For example, P-10 gas (90% Ar:10% CH₄, methane) is commonly used as the counting gas of proportional and GM counters. In addition to the argon

sample, the low specific activity levels of ^{37}Ar and ^{39}Ar to be measured means that a high counting efficiency must be obtained to minimize the amount of sample needed for the measurement. Thus, external detection systems (i.e., the sample material does not form part of the sensitive material of the detector) have been disregarded as possible detection systems because of their inherent low counting efficiency.

Internal counting (i.e., the sample is used as the sensitive material of the detector), on the other hand, offers many advantages. The argon sample, when mixed with small amounts of CH_4 , can be used as the sensitive material of the detector. The counting efficiency for detecting ^{37}Ar will thus be determined exclusively by the amount of dead volume of the detector which is usually less than 20% of the total volume. The efficiency for detecting ^{39}Ar is somewhat less since the range of β^- -particles is much greater than the volume of the detector (19).

Gas ionization chambers and scintillator detectors can both be used as internal counters. The sample material is prepared so that it can be used as the counting gas of a gas chamber or as all or part of the sensitive liquid of a liquid scintillation counter. However, argon is not easily prepared for use in liquid scintillation counters (9:201). Heavier noble gases are known to be appreciably soluble in aromatic hydrocarbons such as toluene and xylene which are commonly used as sensitive materials for liquid scintillators. Krypton and xenon have both been dissolved in a toluene liquid

scintillation solution and have been detected with counting efficiencies up to 100%. Argon, though, has very limited solubility in toluene, and even when dissolved the counting efficiency is low and any quenching reduces the efficiency drastically.

Internal gas counters offer the best means of measuring ^{37}Ar and ^{39}Ar in the atmosphere. All work currently being done on the detection and measurement of ^{37}Ar and ^{39}Ar uses low-level internal proportional counters. The main reason for choosing proportional counters over GM counters is that background count rates can be reduced to a much greater extent. For example, the background count rate can be reduced by considering only events which occur in the energy spectra of interest (i.e., pulse-height analysis). In addition, the anticoincidence system is much more effective with proportional counters than GM counters because pulse rise-times are shorter and there is virtually no loss of source counts resulting from dead time. Another advantage is that proportional counting allows for the identification of the radiation detected and verification of possible contamination of the sample gas (23:240).

Although internal proportional counters, when used in conjunction with the methods of background reduction described earlier in this chapter, have low background count rates, a lower background is still desired for cases of measuring very low specific activity levels of radionuclides such as ^{37}Ar and ^{39}Ar . In this light, various detectors have been designed

which have inherent low backgrounds without much shielding. When combined with shielding, the resulting backgrounds make it possible to measure ^{37}Ar and ^{39}Ar activities approaching those of cosmic-ray produced levels. Three common low background detectors include quartz-tube counters and Oeschger-type counters (both internal proportional counters), and a recently developed, but untested, plastic scintillation counter (6:576).

Both the quartz-tube counter and the Oeschger-type counter have been used (with various modifications) to measure ^{37}Ar and ^{39}Ar in the atmosphere. The newly developed plastic scintillation counter does not yet have the sensitivity to measure ^{37}Ar and ^{39}Ar in the atmosphere. It has been used to measure these isotopes at much higher activity levels (greater than 130 dpm/l-Ar) as effluents from nuclear reactors (23:241).

These three counter construction to be reviewed here are designed to have a low inherent background. In addition, the following properties are also desired (6:576):

- Simple and robust construction
- Small overall dimensions of the main and guard counters
- Large effective volume
- Optimal counting properties
- High radioactive purity of construction materials
- Small memory effect

Quartz-Tube Counters. The quartz-tube counter, sometimes referred to as the Groningen counter (5), is merely a common proportional counter with a metallically-coated quartz tube encased on the inner walls of the cathode. The basic construction of the quartz-tube counter is shown in Figure 6. The inner tube consists of transparent quartz. Two annular end plates are melted onto the inside of the quartz tube. The whole surface except the surfaces of the copper flanges in the end plates to the ends of the tube is coated with a transparent layer of tin oxide to get a conducting surface. The high voltage is applied to the inner wall of the quartz tube. The quartz tube is placed inside a copper tube. The counter, as designed in 1959, could operate at pressures up to 3 atm. Surrounding the counter was an anticoincidence ring counter and lead shielding. The effective volume of the detector is 85% of the total volume. The counter was used for measuring ^{14}C activity (β^- emitter, $T_{\frac{1}{2}} = 5730$ years, $T_{\text{max}} = 156$ keV).

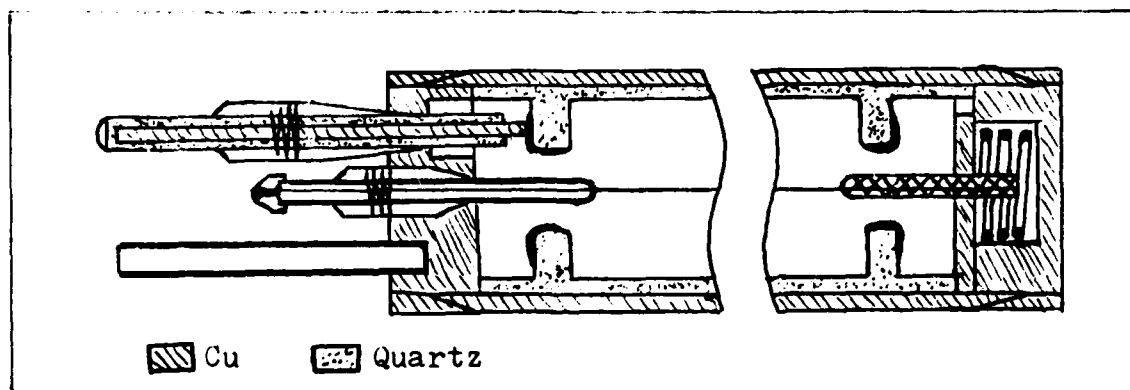


Fig. 6. Quartz-tube Counter (Groningen) (5:112)

The background obtained under the 156 keV energy range was about 1.33 cpm at 3 atm pressure.

The principle behind the use of the quartz tube is the absorption of the α - and β -particles originating from the outer copper tube. Thus, the background is lower than if a simple proportional tube was used with the same shielding and anticoincidence ring counter. The quartz-tube counter has excellent counting properties and is superior to the common proportional tube since it has a lower background (6:577-578).

Oeschger-Type Counters. The Oeschger-type counter became famous because of its extremely low background (6:580). Its basic construction is shown in Figure 7 (27:175). Oeschger counters, also known as membrane detectors, use an aluminized polystyrene or Mylar foil placed into a vacuum-proof cylindrical casing. In the area between the cylinder wall and the foil, a ring of center wires is strung (see Figure 8) which

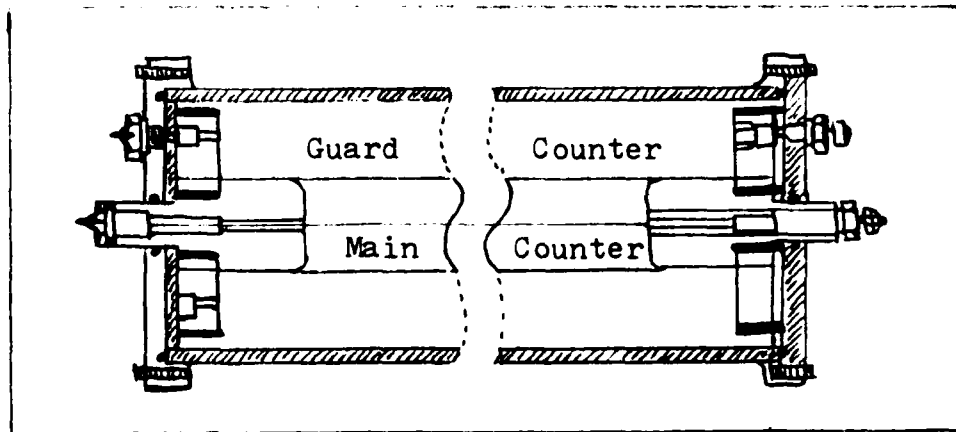


Fig. 7. Oeschger-type Counter (27:175)

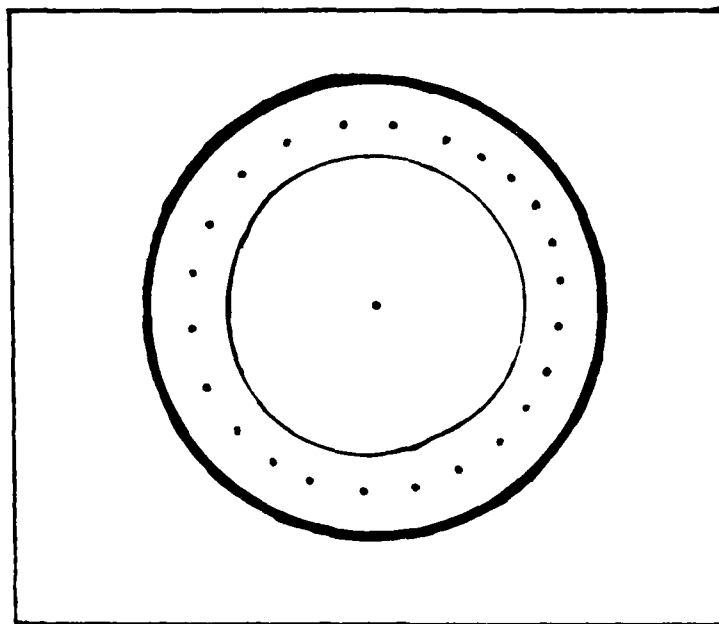


Fig. 8. Cross-Sectional View of Oeschger-type Counter

serve as anode wires for the outer chamber. A single wire is strung in the center of the inner chamber. The foil acts as the cathode for both the inner volume (main detector) and the outer volume (guard detector). The foil has holes which allow for equal pressure on both sides of the membrane, and the sample gas thus acts as the counting gas for both the main and anticoincidence guard counters.

The overall dimensions of the Oeschger counter are small. The major design disadvantage is that part of the sample gas is used in the anticoincidence counter and so the maximum efficiency of the main counter is about 75%. The counting properties are still good. The major achievement of this counter is the background reduction because the cathode

can be made very thin and the component of background produced by γ -rays interacting with the foil is eliminated by the anticoincidence guard counter (6:580).

Two slight modifications of the Oeschger counter are often used. One modification replaces the foil by a ring of cathode wires. The outer ring of anode wires are modified so that every other wire acts as a cathode wire (see Figure 9). An important advantage of this modification is that it is relatively insensitive to radioactive contamination of construction materials (18:439).

Another modification of the Oeschger counter is done by making the foil leak-proof between the two chambers. Sample gas is then contained only in the inner volume and non-radioactive background gas is used in the guard counter. The major advantage of this modification is the increase in the counting efficiency of the detector (27:175).

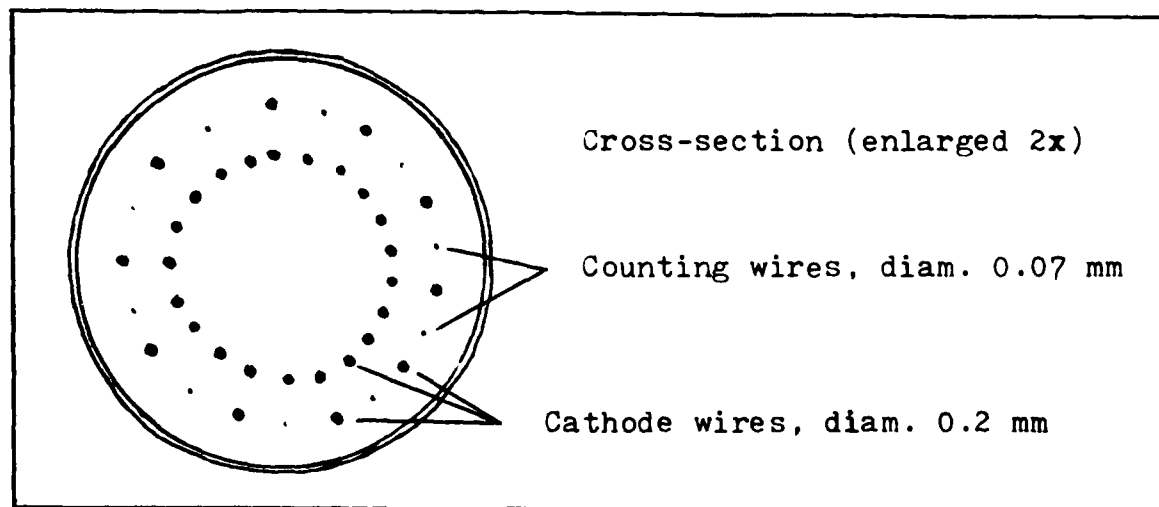


Fig. 9. Cathode Foil Replaced by Ring of Wires (18:438)

Plastic-Scintillation Counters. A third low background detector which has only recently been constructed and not yet tested is the plastic scintillation counter. The basic construction of one type of this counter is shown in Figure 10 (6:584). The counter consists of a cylindric plastic scintillator covered with an outside reflecting layer. A smaller, cylindrical-shaped tunnel has been dug out of the plastic scintillator. A proportional tube can then be inserted into this hollow tunnel. This arrangement is similar to an Oeschger counter except that the plastic scintillator now serves as the guard counter. The detection of background radiations (mainly muons) by the plastic scintillator is very efficient and the counting properties of the main counter are excellent. However, certain problems do exist. Although the use of plastic materials ensures a high radioactive purity, there could be

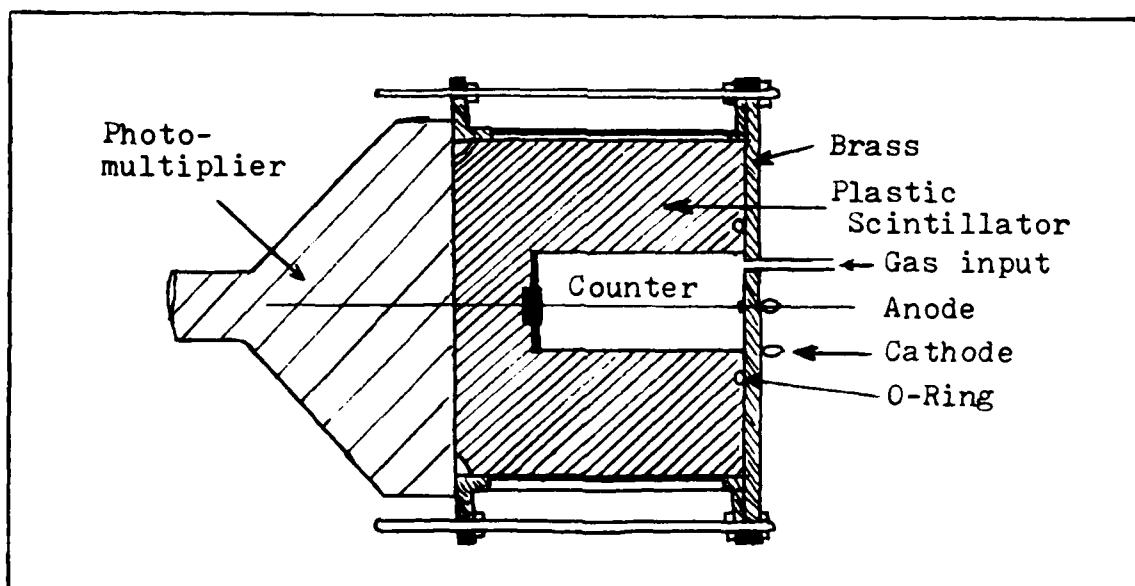


Fig. 10. Plastic Scintillation Counter (6:584)

problems with a memory effect. In addition, the main counter must be optically separated from the plastic scintillator.

Initial measurements of background count rates for this counter approach the low background rates obtained by Oeschger counters. It appears that the plastic scintillator could be much better for low-level detection applications. No work on the measurement of atmospheric levels of ^{37}Ar or ^{39}Ar using this technique has been found in the literature.

Survey of ^{37}Ar and ^{39}Ar Measurements in the Literature

The work of two groups of scientists who have worked on the detection and measurement of ^{37}Ar and ^{39}Ar in the atmosphere will be reviewed in this section. Both groups employ low-level counting techniques and use low background detectors. Table III provides information concerning counter characteristics and detection sensitivities.

Schell and associates at the University of Washington have measured ^{37}Ar in the atmosphere using a modified Oeschger counter in which the sample is confined to the main counter and background P-10 gas is used as counting gas of the guard counter. The actual counter is pictured in Figure 7 on page 40. It was developed at Isotopes, Inc., and is referred to in this text as the II detector (26,27).

Loosli, Oeschger and co-workers at Bern, Switzerland have measured both ^{37}Ar and ^{39}Ar in the atmosphere. They have designed a low background Plexiglas proportional counter with a wall thickness of 5 mm to contain the β^- -particles of

Table III
Detector Characteristics

	UW (³⁷ Ar)	Bern, Switz. (³⁷ Ar, ³⁹ Ar)
Detector	Proportional (Modified Oeschger)	Proportional (β ⁻ -thick Plexiglas)
Shielding	Cadmium-paraffin (2 in 4 in) 8 in old lead	2 cm old lead Ring anticoin- cidence 5 cm old lead 10-20 cm "Boli- den" lead Flat anticoin- cidence
Volume	1.14 liter	1.0 liter
Operating pressure	Up to 20 atm	5.2 atm
CH ₄ content	10%	2%
Efficiency	74%	67%
Counting Time (T _{S+B} = T _B)	1000 min	5000 min
Background (cpm)	0.531 ± 0.024	0.08 ± 0.004 (Ar-37) 1.6 ± 0.18 (Ar-39)
Detection Sensitivity (dpm/l-Ar)	0.051 (+ 10%)	0.009 (+ 20%) (Ar-37) 0.16 (+ 5%) (Ar-39)

(13,14,15,16,26,27)

^{39}Ar decay (13,14,15,16). Counters of this type were used in a systematic study of monthly tropospheric activity levels of ^{37}Ar from 1969 to 1973 (17). Results of this study will be highlighted in a later section of this chapter.

II Detector. ^{37}Ar has been measured by Schell and associates at the University of Washington using a modified Oeschger counter designed at Isotopes, Inc. The II detector consists of a 1.14-liter effective volume inside a larger 9-liter outer volume. The two chambers are separated by a metallized Mylar foil which is leak-proof between the two chambers. The anodes of both the main and guard counters are made of stainless steel of 0.001-in diameter. The detector is surrounded by a cadmium-paraffin shield and 8-in of old lead. Metallic parts of the detector were made of commercial high purity nickel as it was easier to obtain than oxygen-free high-conductivity copper. Nylon was used instead of Teflon because it sealed to the Mylar foil better (27:175-176).

Both chambers are filled with their respective counting gas simultaneously; the inner chamber with a P-10 mixture of sample gas containing the extracted argon, the outer chamber with P-10 background gas. A simple pressure control unit keeps the pressure differential on the Mylar foil less than 5 mmHg, allowing the chambers to be filled separately with minimal danger of breaking the foil. The filling pressures used during the measurements were 5.2 atm and 10.0 atm.

The block diagram of the electronic components of the II detector system is shown in Figure 11. They are specially prepared to allow the proportional spectra to be measured directly from the amplifier output. Two separate single channel analyzers are used as the upper and lower discriminators, respectively. The discriminator levels are easily set by using a radioactive standard gas in the detector and recording the events on the pulse-height analyzer (27:178).

Different possible counting modes for the II detector system are listed in Table IV. All these modes are not necessary for the ^{37}Ar measurement, but they are good to have for systematic checks of detector operation and gas purity.

The important factors in measuring the low specific activity of ^{37}Ar in the atmosphere are counting efficiency, background count rate and background reproducibility. The

Table IV
Counting Modes for the II Detector

Mode	Counts
Net I	Anticoincidence-lower window
Gross I	Non-anticoincidence-lower window
Net II	Anticoincidence-upper window
Gross II	Non-anticoincidence-upper window
Guard	---

(27:179)

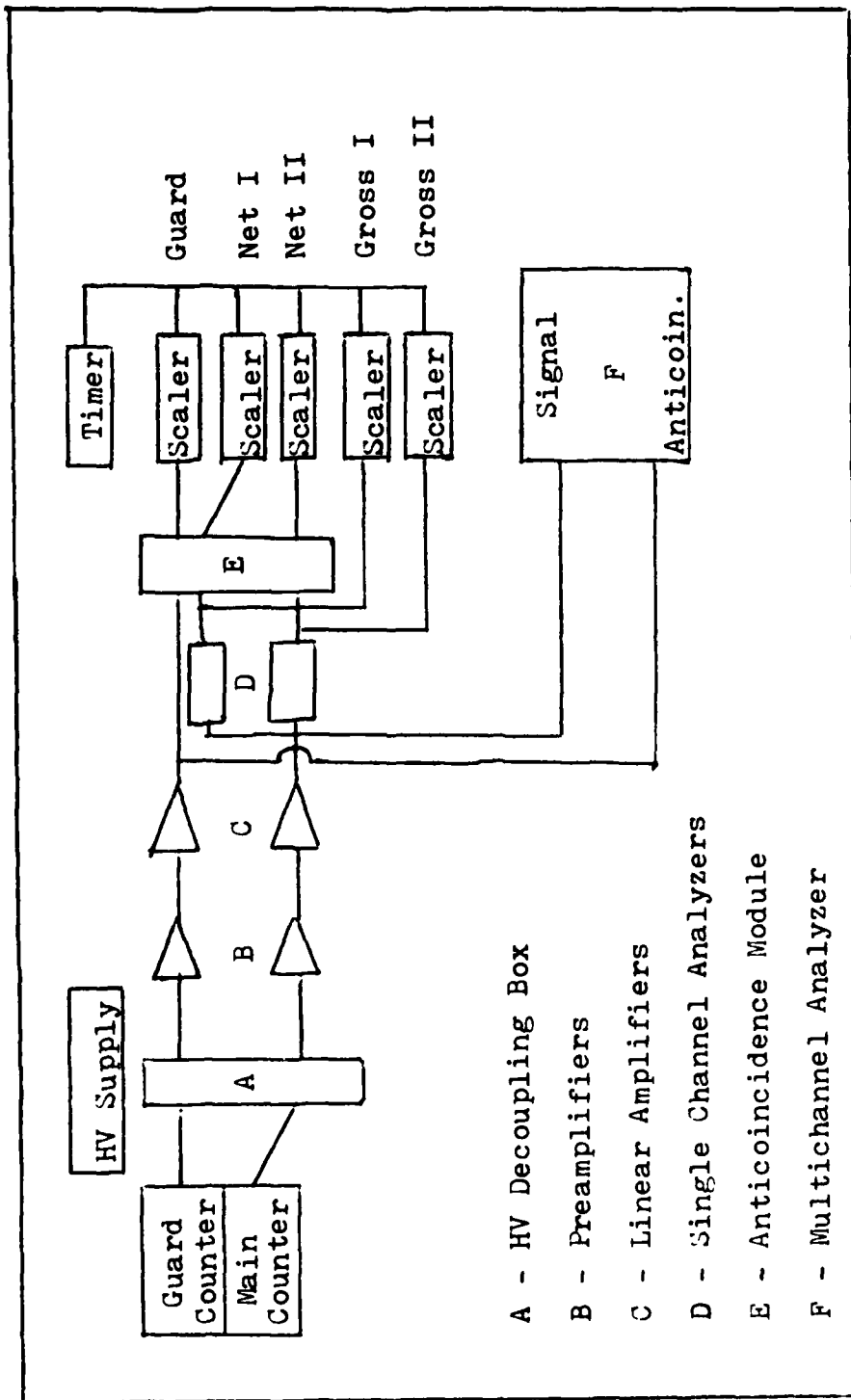


Fig. 11. Electronic Components of II Detector (27:179)

counting efficiency for ^{37}Ar x-rays in the II detector is 74% (27:198). Figure 12 shows a background spectra obtained for a mixture of P-10 background gas at 10 atm pressure (27:189). It is noted that a low energy peak is present

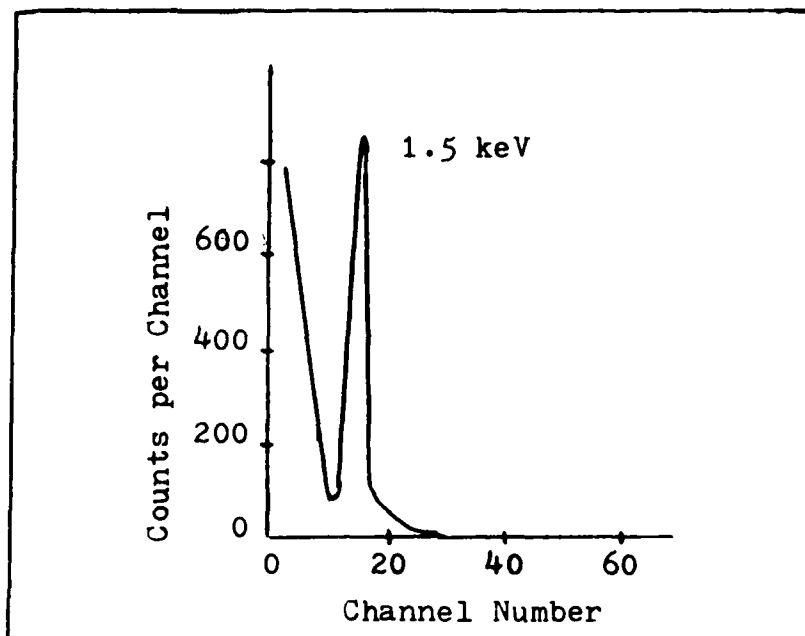


Fig. 12. Background Spectrum of II Detector (27:189)

present with an energy of about 1.5 keV. Apparently, the aluminized foil is being excited by radiation in the detector which is not being detected by the guard counter. The peak could arise from either radioactive contamination from the foil, from inefficient anticoincidence guarding, or from radiation entering from the ends of the detector.

If external radiation which interacts directly with the foil were the cause of the peak, there should be no change in the background recorded for different gas densities (27:189).

The background was, however, found to increase in rough proportion to the gas density. Therefore, the cause of the peak was believed to be due to external radiation which interacts with molecules of the counting gas, exciting electrons or photons that then excite aluminum which emits characteristic x-rays. This 1.5 keV energy peak will cause a slight interference in detecting ^{37}Ar . Gold-coating on the Mylar foil could be substituted for aluminum as it also has high radioactive purity and the characteristic K x-rays have energy of about 66 keV.

Table V shows measurements made on background counting gas in the ^{37}Ar window at 10.5 atm pressure. It can be seen that the background of the II detector under the ^{37}Ar window is fairly low and reproducible (26:461-462).

Table V
Background in the ^{37}Ar window of the II Detector

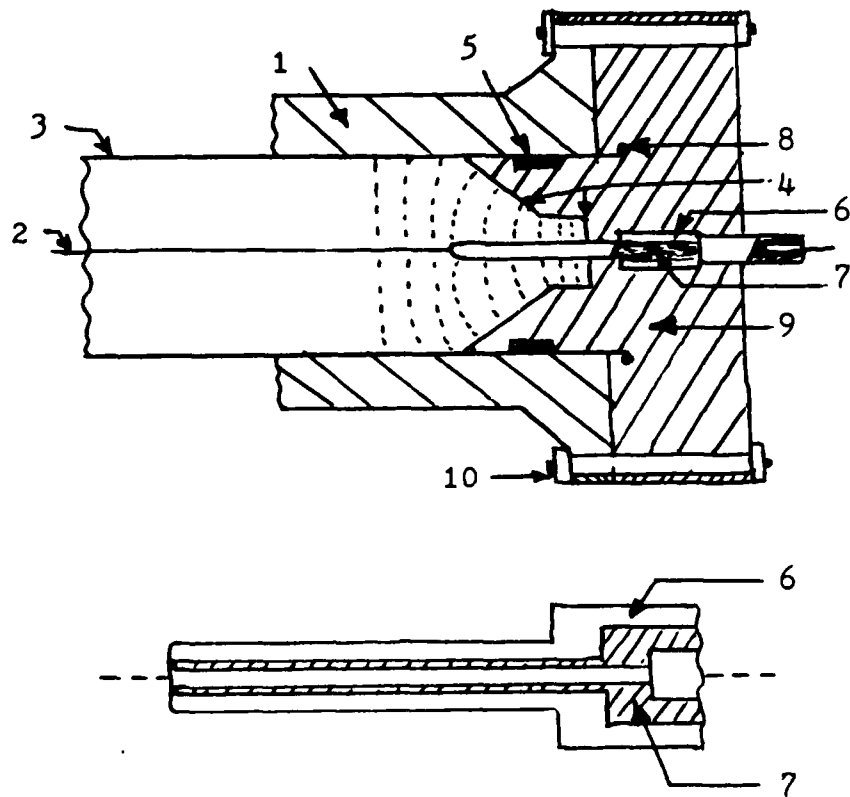
Date	Counting Gas	Background (cpm \pm σ)
23 Sept. 1968	1966 P-10 gas	0.531 \pm 0.024
		0.502 \pm 0.024
24 Sept. 1968	1967 P-10 gas	0.502 \pm 0.023
		0.502 \pm 0.024
24 Sept. 1968	1967 Ar+CH ₄	0.563 \pm 0.025

(26:461-462)

The detection sensitivity for ^{37}Ar at 10.5 atm pressure and with a total counting time of 2000 min can be determined using Equation 6 from page 27. With a background of 0.531 cpm, a counting efficiency of 74%, and a 10-liter argon sample in the detector, the minimum measurable specific activity of ^{37}Ar is 0.051 ($\pm 10\%$) dpm/l-Ar. This level is about twenty times the estimated cosmic-ray produced level. A single sample measurement from 12 December 1967 yielded a measured ^{37}Ar level of 0.16 dpm/l-Ar, roughly 64 times the estimated cosmic-ray produced level.

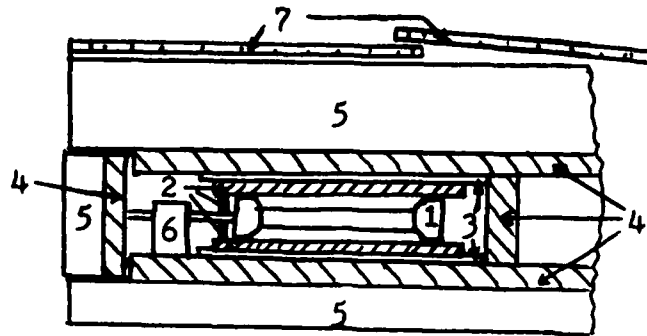
Plexiglas Detector. Loosli, Oeschger and associates have constructed a detector capable of measuring both ^{37}Ar and ^{39}Ar in the atmosphere. The counter construction is illustrated in Figure 13 (15:596) and its characteristics are shown in Table III on page 45. Plexiglas was chosen as the material for the counter because of its high radioactive purity and because it is easily machined. The plexiglas walls are 5 mm thick and are termed "beta-thick" meaning the β^- particles emitted from ^{39}Ar are stopped in the walls and do not penetrate into the surrounding guard counter. The cathode is metallized Mylar foil with the end pieces evaporated with gold. The special arrangement of the metallized end pieces result in a small dead volume and reduced end effects.

Because of the sensitivity of the background count rate to any secondary electrons produced by γ -interaction with the detector walls and with the gas, special care was taken in



1. Plexiglas wall 5 mm thick
2. Central wire (0.04 mm diam.)
3. Metallized foil (Mylar)
4. End pieces evaporated with gold in the area between the end points of the arrows
5. Metal ring for mechanical support of the foil. Also makes electrical contact with gold surface of the end piece
6. Guard electrode of stainless steel
7. Electrical insulation of Plexiglas
8. O-ring
9. End piece of Plexiglas
10. Bolt
11. Lines of force shown schematically

Fig. 13. Construction of Plexiglas Counter (15:596)



1. Main counter of plexiglas with a volume of 1-liter filled up with Ar + 2% CH₄ at a pressure of 6 atm.
2. 2 cm of radioactively very pure old lead reducing especially the background component induced in the shield (4,5) by muons and nucleons.
3. Anti-coincidence ring counter reducing the background component of charged particles.
4. 5 cm old lead
5. 10-20 cm lead from "Boliden."
6. Pre-amplifier and filter for high voltage.
7. Flat anti-coincidence counters with a similar background reduction mechanism as shield no. 2.

Fig. 14. Arrangement of Plexiglas Counter and Shields
(21:492)

developing effective shielding. The final arrangement of the shields is shown above in Figure 14. A cylindrical guard counter and big flat guard counters on five sides of the shield are filled with CH₄ at 1 atm pressure and are operated in anticoincidence with the main counter. The electronic components of the system are shown in Figure 15.

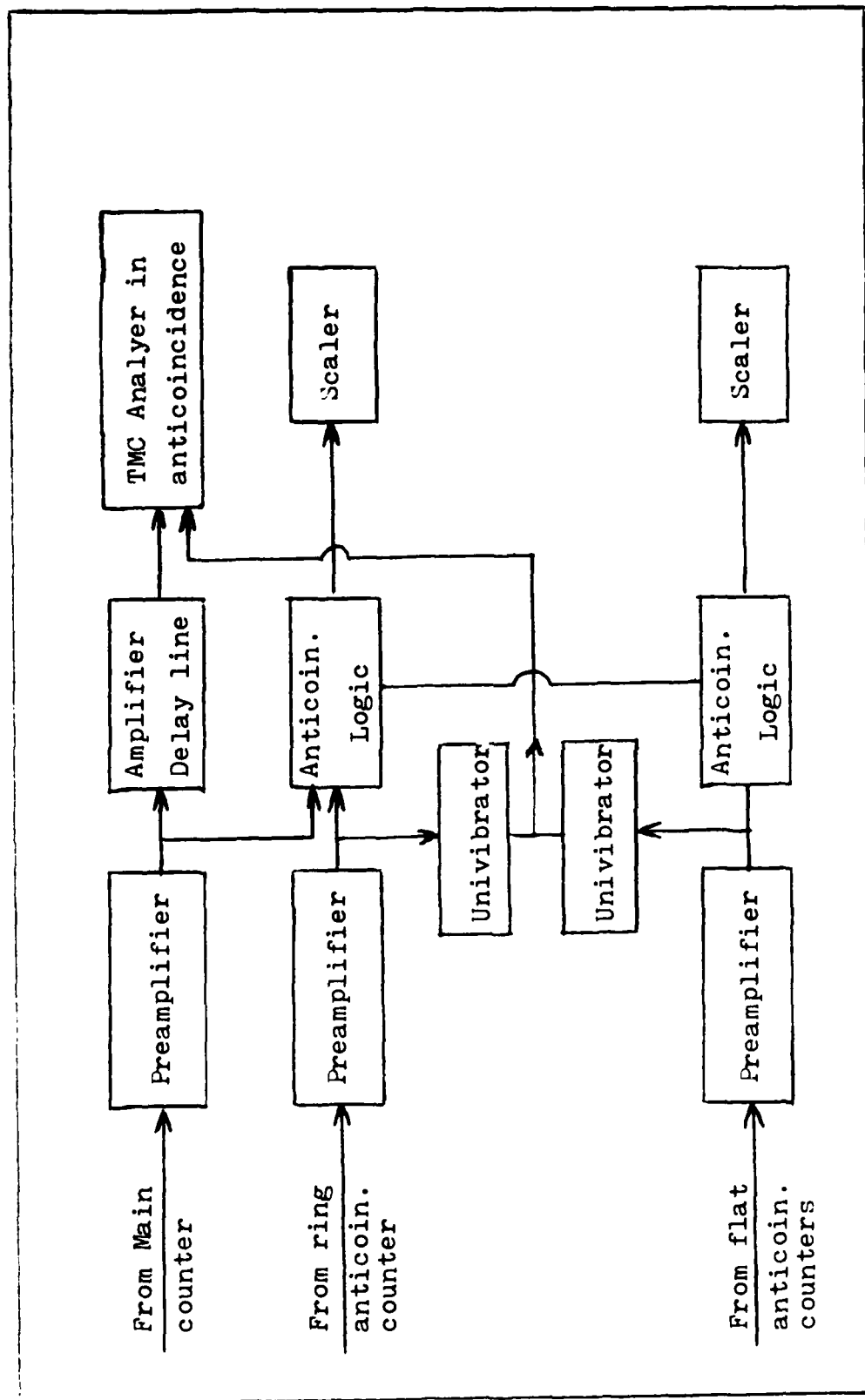


Fig. 15. Electronic Components of Plexiglas Counter (15:598)

1. ^{37}Ar Measurements. The first ^{37}Ar measurements carried out are summarized in Table VI. The first sample was from Oakland, Ca. and was measured by Schell as discussed previously. All other samples were collected and measured by Loosli and co-workers in Bern, Switzerland. All samples were collected near the ground (16:2896;17:24).

The background count rate in the ^{37}Ar peak energy band was about 0.08 cpm and the net counting rates measured varied between about 0.003 and 0.1 dpm/l-Ar. To all the argon samples about 2% of inactive methane was added and the plexiglas counter was filled to a pressure of 5.2 atm. A counting efficiency of about 67% for the low energy x-rays was calculated using a given ^{37}Ar standard (17:24). The counting times

Table VI
 ^{37}Ar Measurements from Tropospheric Samples

Date	Activity (dpm/l-Ar)	Location
12 December 1967	0.16	Oakland, Ca.
1 February 1969	0.0276 ($\pm 15\%$)	Bern, Switz.
12 March 1969	0.0168 ($\pm 10\%$)	"
20 May 1969	0.0084 ($\pm 20\%$)	"
23 June 1969	0.0034 ($\pm 30\%$)	"
22 July 1969	0.0037 ($\pm 30\%$)	"
14 October 1969	0.0067 ($\pm 30\%$)	"
24 November 1969	0.1068 ($\pm 5\%$)	"
15 December 1969	0.0420 ($\pm 9\%$)	"

(16:2896;17:24)

generally lasted about 5000 minutes.

The most striking result of the ^{37}Ar measurements is the wide fluctuation in the specific activities between February and December of 1969. Additional measurements for ^{37}Ar were systematically carried out in monthly periods between 1969 and 1973 and the measured specific activities varied by as much as 3 orders of magnitude (17). This study and the possible explanations for the variations will be discussed after the next section on ^{39}Ar measurements using the plexiglas counter.

2. ^{39}Ar Measurements. The major difference between the measuring techniques of ^{37}Ar and ^{39}Ar arises from the fact that the background under the ^{39}Ar spectrum will be much greater than that under the ^{37}Ar peak. In addition, an argon background gas must be obtained in order to determine the background count rate of the detector. The best way of measuring ^{39}Ar at the present time is to compare recent atmospheric argon with argon originating from ^{40}K -decay in underground well gas (13:191). Although the procedure for obtaining well gas and extracting the argon is difficult, it has been accomplished. Another possible source for background gas would be argon occluded in 1500 year old ice (age found using ^{14}C dating techniques) in the polar ice caps (13:195).

The lowest background obtained in the ^{39}Ar spectrum (at a cutoff energy of 2 keV) was 1.6 cpm (16:1895). The counting times generally lasted 5000 min. The ^{39}Ar activity was measured in 7 atmospheric argon samples extracted in 1940

and between 1959 and 1967 (13). The specific activity obtained from a first set of measurements was 0.121 ± 0.005 dpm/l-Ar, whereas a second set of measurements on the same samples a few months later gave specific activities of 0.084 ± 0.004 dpm/l-Ar. The discrepancy was believed to have been due to a contamination of the background well gas sometime between the two sets of measurements. Calculations showed that the contribution of ^{39}Ar produced from nuclear explosions up to 1967 is less than 0.005 dpm/l-Ar (about 5% of the cosmic-ray produced level).

^{37}Ar Monitoring of the Troposphere

The ^{37}Ar activity in world-wide ground level air samples was monitored during the period from 1969 to 1973. The specific activity levels varied by as much as three orders of magnitude from a low value of 0.0025 dpm/l-Ar to a high value of 1.8 dpm/l-Ar (17:24). Most of the samples were measured by Loosli and associates with the plexiglas counter reviewed earlier. Some of the conclusions drawn from the study include the following:

1. The lowest ^{37}Ar specific activities (about 0.0025 dpm/l-Ar) probably represent the cosmic-ray produced activity found at ground level in a mixed atmosphere. Within the uncertainties, the measured values agree with the theoretical estimates.
2. High ^{37}Ar activities measured in the atmosphere could not have been caused by minor local releases, but very large quantities of Ar-37 must have been released into the northern troposphere. Possible sources for these massive releases include underground nuclear tests, nuclear reactors and nuclear reprocessing plants.

3. Sudden local influxes of stratospheric air into the troposphere could cause only minor variations in activity up to 0.05 dpm/l-Ar.
4. Release of reactor-produced ^{37}Ar is the reason for minor variations in activity.
5. Atmospheric nuclear explosions do not account for any measured fluctuation in activity of Ar-37.
6. Underground nuclear explosions can easily produce the large amounts of Ar-37 necessary to explain the highest activity levels measured (1.8 dpm/l-Ar).

Summary

The major points to be summarized in this chapter on the detection and measurement of ^{37}Ar and ^{39}Ar in the atmosphere include the following:

1. Mass spectrometry cannot be used to measure ^{39}Ar in the atmosphere at low specific activity levels approaching the cosmic-ray produced levels.
2. In the development of a detection system for measuring atmospheric levels of ^{37}Ar and ^{39}Ar , it is essential to aim for a low background and good background reproducibility.
3. At present, internal gas proportional counting is the only feasible means for measuring ^{37}Ar and ^{39}Ar in the atmosphere because counting efficiency is maximized and background can be reduced effectively through the use of shielding, anti-coincidence and pulse-height analysis. Further reductions can be achieved by employing rise-time discrimination and operating in an underground laboratory.

4. Both ^{37}Ar and ^{39}Ar have been measured at levels approaching the cosmic-ray produced levels. Because the background count rates are higher and because of the need to obtain an ^{39}Ar -free background gas to determine the background count rate, ^{39}Ar is much more difficult to measure than ^{37}Ar .

5. Specific activity levels of ^{37}Ar in the northern troposphere have been seen to fluctuate by three orders of magnitude. It is believed that the lowest levels measured (about 0.0025 dpm/l-Ar) represent the cosmic-ray produced level, whereas the highest levels (up to 1.8 dpm/l-Ar) were caused by ^{37}Ar produced in underground nuclear explosions.

V. Conclusions and Recommendations

Conclusions

The purpose of this thesis was to investigate the techniques needed to detect and measure ^{37}Ar and ^{39}Ar in remote atmospheric samples and to determine the feasibility of using these measurements for monitoring the possible venting of radionuclides from underground nuclear explosions. From the studies carried out in this thesis, the following conclusions have been made:

1. The final system which will measure ^{37}Ar and/or ^{39}Ar in remote atmospheric samples requires the combined optimization of sample preparation and measurement procedure. It will operate in a production-type mode, extracting and purifying argon from many given atmospheric samples and then measuring the activity of ^{37}Ar and/or ^{39}Ar in the argon samples.

2. In cases where the argon extraction yield from the remote sample is important, the NBS argon purification system is preferred over the UW system. This condition will usually apply to remote air samples of limited quantity.

3. At present, low-level internal gas proportional counting offers the only feasible means for measuring atmospheric levels of ^{37}Ar and ^{39}Ar . It is strongly recommended that any low-level counting system eventually chosen be very similar to the systems employed by Loosli or by Schell.

4. The possibility of using atmospheric ^{37}Ar and/or ^{39}Ar measurements for monitoring purposes remains questionable. One would like to determine whether or not the cause of observed variations in activity at the sampling location was due to a particular source (e.g., venting from an underground nuclear explosion). Thus, one needs to know the quantitative amounts of ^{37}Ar and/or ^{39}Ar produced by a specific source (and any other nearby sources). In addition, the rate of radioargon release into the atmosphere, as well as atmospheric mixing processes between the source and sampling locations, must be well known. At present, these needs are not fully met.

Recommendations

It is not currently feasible to use ^{37}Ar or ^{39}Ar measurements from remote air samples for underground monitoring purposes. To become feasible, a detailed study on quantitative amounts of ^{37}Ar and ^{39}Ar produced from various sources and their release rates into the atmosphere is needed. Along with fairly well-known atmospheric mixing processes, this information will allow one to determine whether or not observed variations in ^{37}Ar or ^{39}Ar measurements were the result of venting from a particular underground nuclear explosion.

Radionuclides other than ^{37}Ar and ^{39}Ar (mainly krypton and xenon) are currently more practical for monitoring purposes. Both krypton and xenon are produced as direct fission products of the nuclear device. Compared to ^{37}Ar and ^{39}Ar , they are easier to measure by low-level counting since they are

more easily extracted and purified from air samples. However, ^{37}Ar and ^{39}Ar measurements could become valuable monitoring tools in the event of the development of a pure fusion nuclear device.

In addition to the use of ^{37}Ar and ^{39}Ar measurements for monitoring purposes, other important scientific information could be gathered from these measurements (13,16,21,26). For example, ^{37}Ar could be used as an excellent tracer in the study of atmospheric mixing processes; ^{39}Ar could be used as a valuable tracer in oceanographic studies. Both ^{37}Ar and ^{39}Ar , along with other gaseous radionuclides (e.g., CO_2 , ^{131}Xe , ^{133}Xe , ^{85}Kr , and HT) can be used to study the constancy of cosmic-particle production rates and influences of, for example, solar cycles, solar flares and supernovae.

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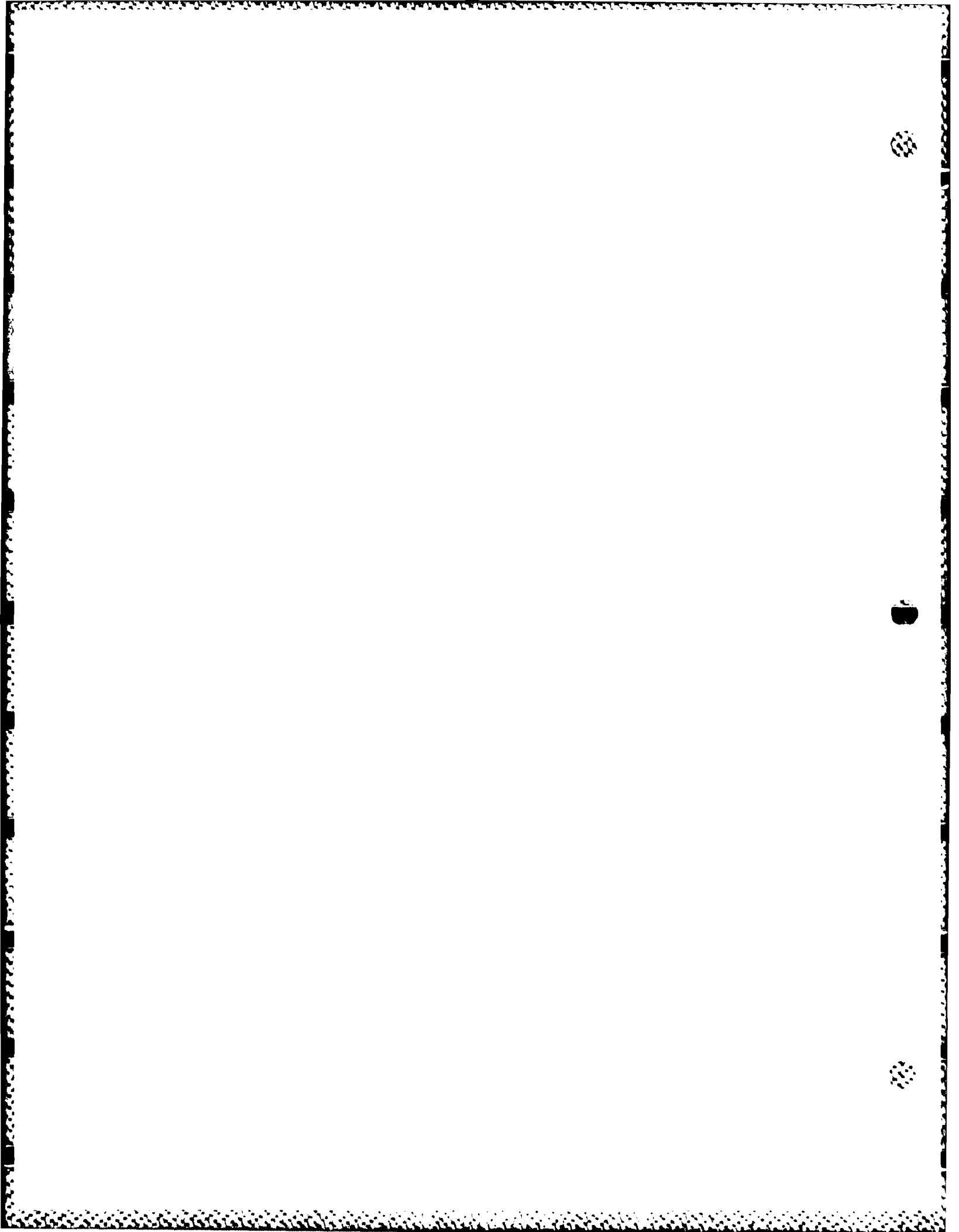


Vita

Richard Charles Runchey, son of Robert and June Runchey, was born on 22 February 1960 in Marshall, Minnesota. He graduated from Marshall Senior High School in 1978. Upon receiving a four-year Air Force Reserve Officer Training Corps scholarship he attended Creighton University, Omaha, Nebraska. He received the degree of Bachelor of Science in Physics from Creighton in May 1982. After receiving his commission in the United States Air Force in May 1982, he entered the Air Force on active duty in August 1982, enrolling in the School of Engineering, Air Force Institute of Technology.

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A study was carried out to determine the feasibility of using Ar^{37} and/or Ar^{39} activity measurements from remote atmospheric samples for monitoring the possible venting of radionuclides from underground nuclear explosions. The study was broken into two parts; 1) a study of the processes for extracting and purifying argon from the remote air samples, and 2) an investigation of the use of mass spectrometry and low-level counting techniques for measuring Ar^{37} and Ar^{39} in the purified argon samples.

Argon can be extracted from air using a combination of gas-solid chemical reactions and low-temperature gas chromatography. It was found that mass spectrometry cannot be used to measure Ar^{39} in the atmosphere. At present, low-level internal gas proportional counting is the only feasible means for measuring atmospheric levels of Ar^{37} and Ar^{39} .

It is concluded that it is not currently feasible to use Ar^{37} or Ar^{39} measurements for monitoring purposes. Further studies are needed on radioargon production sources and release rates into the atmosphere. (These)

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