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HEALTH AND SAFETY
(TID-4500)

PROCEEDINGS
of the
ROCKY FLATS SYMPOSIUM ON SAFETY IN PLUTONIUM HANDLING FACILITIES

April 13-16, 1971

Sponsored by The Dow Chemical Company
Rocky Flats Division
Golden, Colorado

(Prime Contractor of the U. S. Atomic Energy Commission)

DISCLAIMER

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FOREWORD

On April 13 through 16, 1971, a symposium was held at the Rocky Flats plant of The Dow Chemical Company to discuss safety in plutonium handling facilities. The purpose of the meeting was to review and discuss innovations for the safety of plutonium operations. Approximately 225 persons attended. They represented the AEC, AEC contractors, the AEC national laboratories, universities, and private companies. All those attending were concerned with and involved in the safe handling of plutonium. The symposium was divided into four general areas: (1) glovebox design and operation, (2) ventilation systems, (3) fire detection and suppression systems, and (4) environmental considerations. This volume is a record of the 42 papers that were presented. It should provide an indication of the state-of-the-art of plutonium safety in various facilities and a starting point for studies to further improve the safety of plutonium facilities. At the conclusion of the meeting, those who attended were polled as to their opinions on the value of the meeting and their opinions about the usefulness of future meetings of this type. The majority of those responding felt that this meeting was of value to them and to their operation and that further meetings of this type should be held on an approximately yearly basis.

M. A. Thompson

ACKNOWLEDGMENT

The organizers of this meeting appreciate the help and encouragement of the many people at Rocky Flats and elsewhere within the AEC complex for their help and assistance in making this a successful symposium. Special thanks are due to the many people within the AEC who helped and encouraged us in setting up and conducting the meeting and who participated in the meeting. The staff of the Rocky Flats Technical Information Department assisted in the physical arrangements for the meeting and in the preparation of the symposium proceedings. Joan Morris was responsible for coordinating all the arrangements and taking care of the many administrative details necessary for a successful symposium. A special acknowledgment is made to all those who presented talks and prepared papers, and to those who attended and participated in the discussions.

M. A. Thompson

INTRODUCTION

William H. Lee
Environment Control Manager

The Dow Chemical Company
Rocky Flats Division

Good morning and welcome to Rocky Flats. It is a real pleasure to see that so many of you from so many different companies and organizations share our interest and concern about safety in plutonium facilities. When this symposium was first suggested, we were concerned that there wouldn't be enough interest to put together a worthwhile program. As the time for the meeting approached, this worry changed to one of "will everyone who wants to come fit into our auditorium?"

The large response to this symposium indicates that there is a real concern by all those working with plutonium to have an operation that is both safe for those working directly with the material and safe for the surrounding environment. It is particularly appropriate that a group such as this gather together to discuss their mutual problems, because it is very apparent that what affects one of us affects all of us. Two significant examples of this were the May 1969 fire at the plutonium fabrication facility here at Rocky Flats and the recent increase in interest and concern by the general public in the environment. Because of these, we at Rocky Flats have received a lot of attention and advice from many sources. I'm sure that if you are working with plutonium, you have also received your share of attention.

Even without this recent increased interest in our type of business, we have a very big responsibility to insure that we don't injure our workers or damage our environment. To accomplish this, we all want to operate in the safest possible way. Therefore, a meeting such as this, in which different approaches to safety can be discussed and evaluated, is particularly valuable. It provides an opportunity for those who have had considerable experience in handling plutonium and have used different approaches, to accomplish the same objective, to come together and discuss these different methods and techniques. It should be possible to use some of the suggestions and ideas to be discussed during the next few days to upgrade the safety of existing facilities. For new facilities, it may be possible to take the best ideas and suggestions and incorporate them into the design and construction to make a facility that is safer than any of our existing facilities. Additions to any operation designed to increase the safety will add significantly to the cost. However, this increased cost may be small when you consider the results of a potential accident occurring in a facility that is not the safest possible.

If there is anything we can do to make your visit to Rocky Flats more profitable and enjoyable, please let us know.

Thank you.

SECTION I.

GLOVEBOX DESIGN AND OPERATION

HUMAN ELEMENT DESIGN CONSIDERATIONS WITH GLOVEBOXES AT THE ATLANTIC RICHFIELD HANFORD COMPANY

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April 13, 1971

HUMAN ELEMENT DESIGN CONSIDERATIONS WITH GLOVE BOXES AT THE ATLANTIC RICHFIELD HANFORD COMPANY

Since September 1966, Atlantic Richfield Hanford Company, or ARHCO, has had the responsibility for operating the chemical-processing portion of the Atomic Energy Commission's work conducted at Hanford. In prior years, this same work was performed by other contractors. This presentation spans the era of several contractors, and represents the considerations we have given for the human element, radiation control and plant production needs. In our terminology, we define glove boxes and hoods as being synonymous.

Hanford has been involved with processing of nuclear materials since 1944. This paper will consider the era starting in 1961. At this time a detailed study entitled "Human Engineering Consideration for Glove Box Design" was conducted. The study was made to define the limitation of an individual working through the glove ports in a hood adapted to specific requirements of a new facility. Equipment and radiation shielding could then be designed so that it would be compatible to dimensional limitations of the human being, minimizing accessibility problems which had previously plagued hood utilization while adequately controlling radiation exposure to the operator.

It would be necessary for the employee to work in the hood for a good share of an eight hour shift. Because of this, the safety and comfort of the individual was important. The limitations of an individual in this case was an important factor in his ability to work in a safe and efficient manner, with the least tiring effect.

Individuals involved in glove box work were consulted as to the important factors which are necessary for the safety, comfort and efficiency of the person doing the work. Fundamental motion patterns and limitation for an "average operator group" were developed for the specific requirements of the proposed facility. Particular effort was expended in an attempt to define the maximum accessible areas of the human hands when working in glove boxes. As a result of the consultation and the motion study, a list of basic criteria was established from which initial dimensions and drawing layouts could be developed. Attachments to this paper contain drawings of these motion patterns which were developed, one of which is being projected. (Figure I) This is a reach pattern considering a 15-1/2 inch work area from the glove port. The single and two hand work areas are evident.

The following criteria were established for this specific facility and have been successfully utilized in other support areas.

REACH PATTERN

PLANE 15 1/2" FROM FACE OF GLOVE PORT

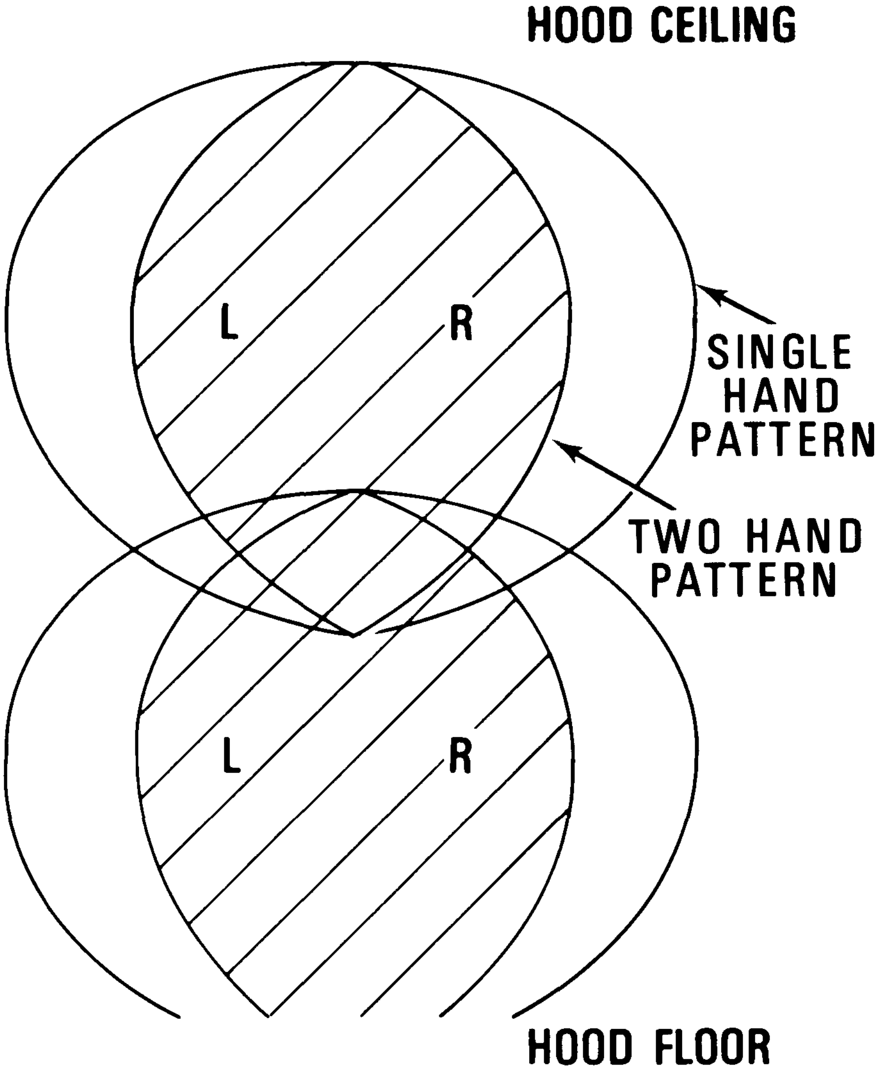


FIGURE I

1. The maximum accessible area is to be provided for an operator in a standing position, using both hands concurrently in a "clutch-grip" fashion.

2. As near as practical, there is to be 100 percent access to the hood when using one hand at a time.

3. The hoods are to be accessible from the sides with a "desirable" maximum of two tiers of glove ports.

4. Maximum accessible hood width is desirable depending on the size of the installed equipment.

5. Eye ports should be provided rather than large transparent panels.

Basic dimensions and work area limitations were taken from "average" man data. Experiments were conducted under simulated conditions using a flexible glove port mock-up to determine the specific restrictions in each case. For the "average man" the most comfortable position, considering maximum forward reach requirements, existed when the glove ports measured 16 inches between centers and 56 inches (maximum) to the floor or step. With a two handed grip, a slightly wider coverage area developed when the glove ports were closer than 16 inches because the crossover arm is the controlling length. Considering the variations in operator size, coupled with potential area coverage, the 16 inches on center and 56 inches height dimensions were selected.

The pattern generated through the concurrent use of both hands can be grouped to obtain the best relationship for maximum coverage. To insure the absence of voids in the two hands pattern, a hood depth of 18 inches is recommended. The maximum hood depth should not exceed 26 inches and if possible, should not exceed 24 inches. In a hood where there is access from both sides, a depth of 36 inches is recommended. The maximum distance from the horizontal centerline of the nearest row of glove ports to the top or bottom of a hood should not exceed 18 inches. These reach patterns are further illustrated in Figures II and III, showing the reach pattern as the depth of the work area increases.

One fundamental rule which must be applied is that all equipment mounted within the hoods must be accessible from the glove ports for operation, maintenance, replacement or decontamination. Since the hoods are greater in depth than can be reached with full coverage from the glove ports, equipment and glove ports must be located so that the above functions can be adequately carried out.

The ellipse formed by two hands held together in motion, requiring grasping with lever force or holding and turning, is called the two hand working area. An example of equipment which might have to be located in this area would be a valve and valve bracket where one hand would be required to hold the valve and the other hand required to fasten it to the bracket.

REACH PATTERN

PLANE 21 1/2" FROM FACE OF GLOVE PORT

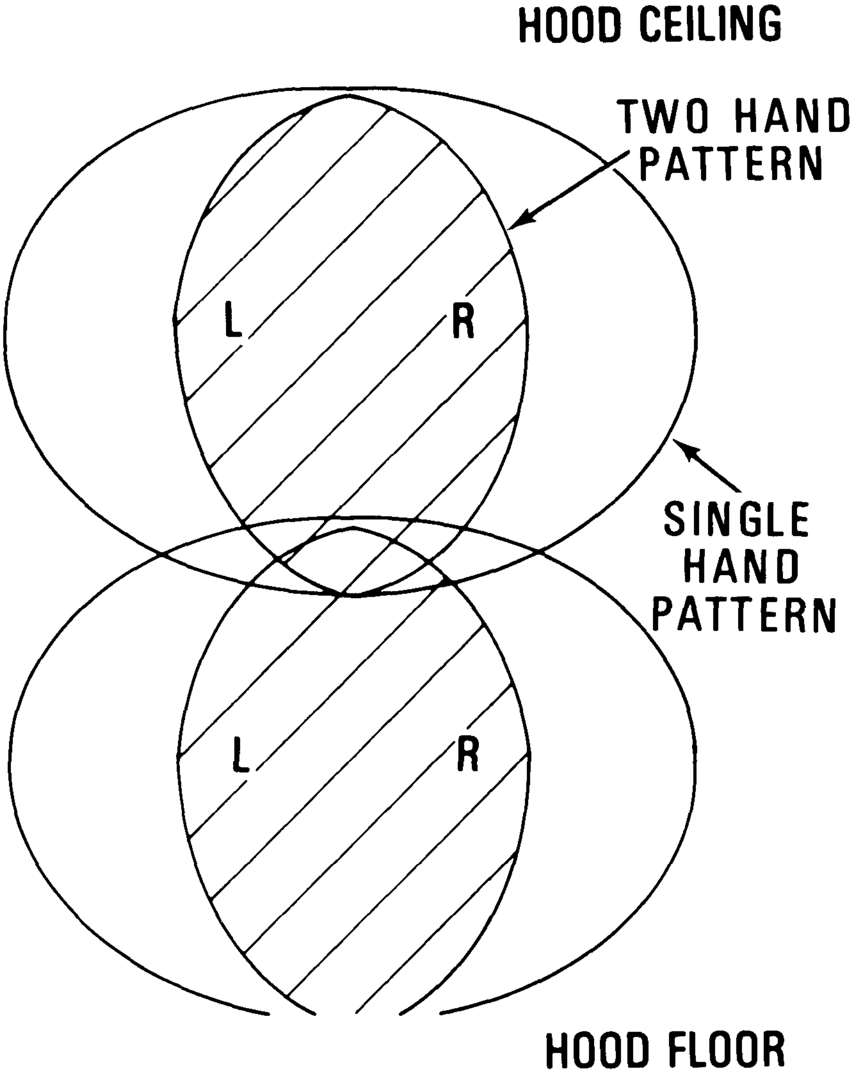


FIGURE II

REACH PATTERN

PLANE 24" FROM FACE OF GLOVE PORT

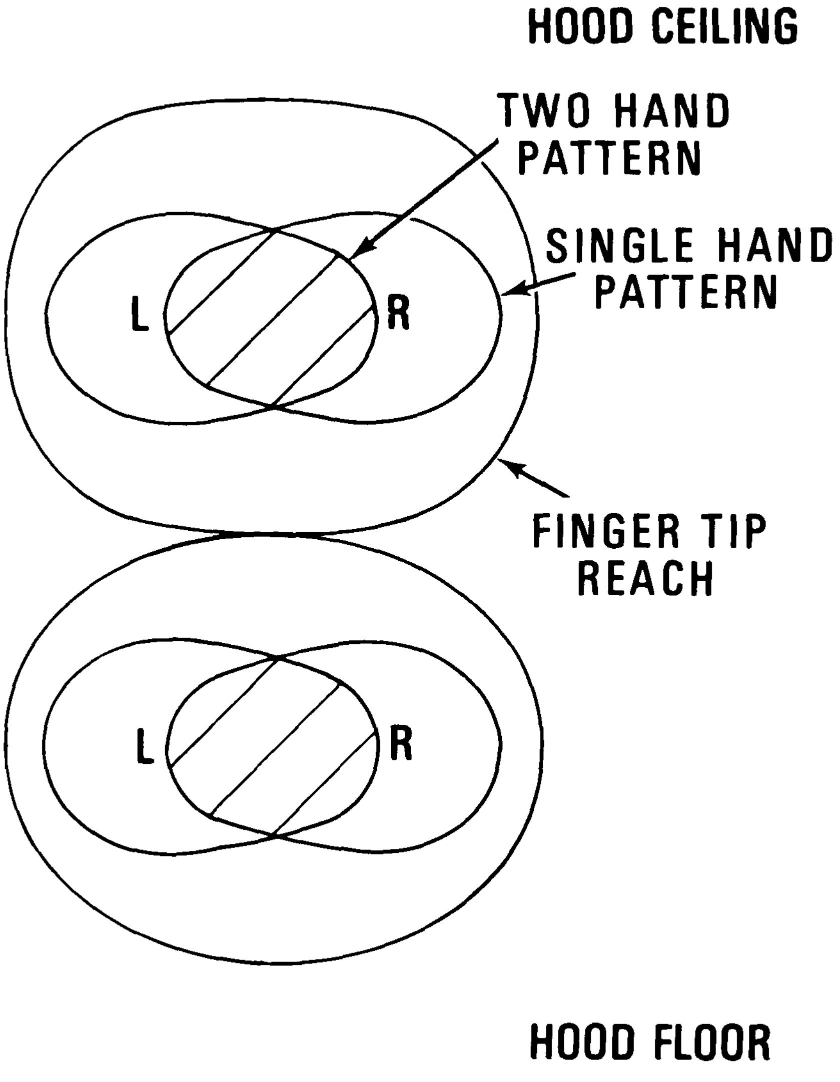


FIGURE III

The ellipse formed by one hand in motion requiring grasp and turning, but working independently of the second hand, is the one hand working area. There are actually two one hand reach zones in a hood. One case is when both hands are in a hood, each doing work independently with shoulders parallel to the hood face. The other case is when only one hand is in the hood. In the latter case, a part of an individual's shoulder enters the glove port so that his reach is actually extended approximately 2 inches. The reach obtainable by this technique is about the limit of the glove length. As the hood depth is increased beyond 18 inches, measured from the front of the glove port to the back of the hood, there are increasing areas within the hood which are not accessible without "arm extenders."

Work locations have a considerable effect on the location of viewing windows. Actual demonstration on the mock-up brought to light the following:

1. As the work location becomes increasingly higher above the centerline of the glove port, the distance between the horizontal center- of the glove ports and eye level decreases.

   Conversely, as the work location becomes increasingly below the horizontal centerline of the glove ports and more distant, the distance between the centerline of a glove port and eye level increases.

2. As the work location becomes more distant from the front face of the hood, the distance between the horizontal centerline of the glove ports and eye level decreases. There appears to be an actual hunching of the shoulders and neck of an individual.

3. As the work location shifts to the right, the eye position shifts to the left.

Further, it was determined that the triangle viewing window should be used for the top row of glove ports. These windows should be 14 inches on a side and mounted so that the apex of the triangle nestles between the glove ports. Our next two slides (Figures IV and V) picture an actual facility utilizing the viewing window placements. Please note the square, diamond and triangular window placements.

Square viewing windows should be used with other rows of glove ports. The dimensions of these windows should be 10 inches on a side. The windows should be mounted in a "diamond" pattern so that a line drawn through two opposite corners will be 7-1/2 inches above and parallel to the centerline of a pair of glove ports.

Where work location can normally be any place within the hood, some compromise must be made to permit adequate viewing of hood interiors while maintaining hood construction integrity. Proper location of viewing windows requires full consideration of the factors listed below:

1. Work Location

FIGURE IV

FIGURE V

2. Reach Phenomenon

3. Shielding Window Depth

Windows of practically no thickness such as unshielded glass do not cause viewing problems. As window thickness increases, one cannot look at any angle to the same degree as with a glass window.

Where possible, 3/8 inch safety glass is used in all hood viewing windows. The windows are kept small to obtain a flat seating surface. Window gaskets are the channel type made from neoprene or silastic LS-63. For small standard size windows, the gaskets are the one piece molded type.

In this presentation, we have made no attempt to discuss hood gloves or the types of hood atmospheres. We utilize room air, dry air and inert atmospheres as required. Furthermore, inlet and exhaust filtration utilizing HEPA fire resistant filters are a part of our design except in those cases where corrosive exhaust atmospheres demand CWS type. Fire detection equipment is installed and will be discussed in a paper scheduled for tomorrow. However, most existing glove boxes and all new glove boxes are equipped with heat detectors and fire alarm systems. Hand operated fire extinguishers are available at all glove box locations and are fitted with a bayonet type nozzle for easy penetration of the gloves. Glove port fire covers are provided near all glove boxes.

In closing, I would like to show you various other views of the facilities we have been discussing. Figure VI illustrates hood depth. Figure VII and Figure VIII illustrate a variation of glove and viewing window placement, and are shown to illustrate shielding concepts.

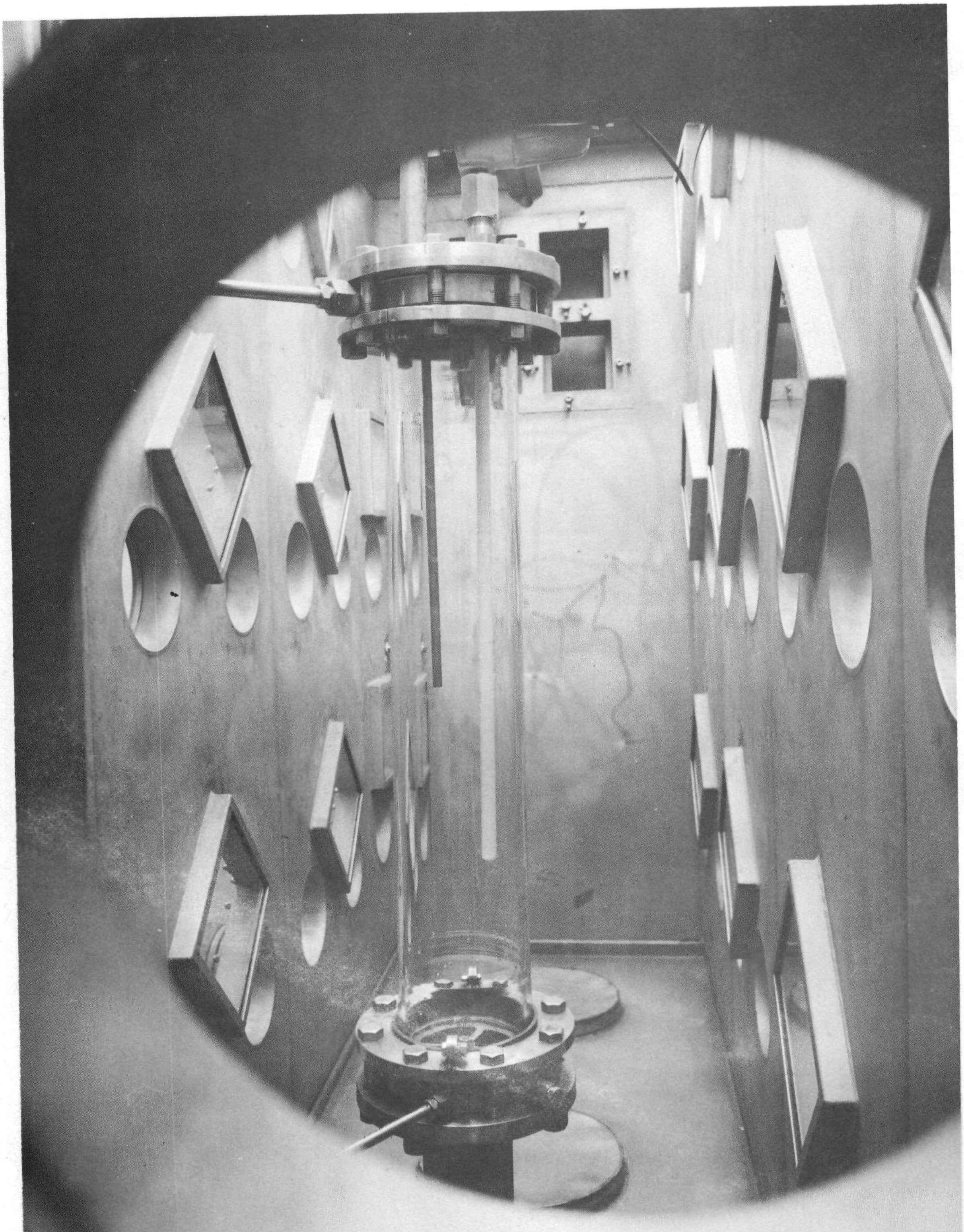


FIGURE VI

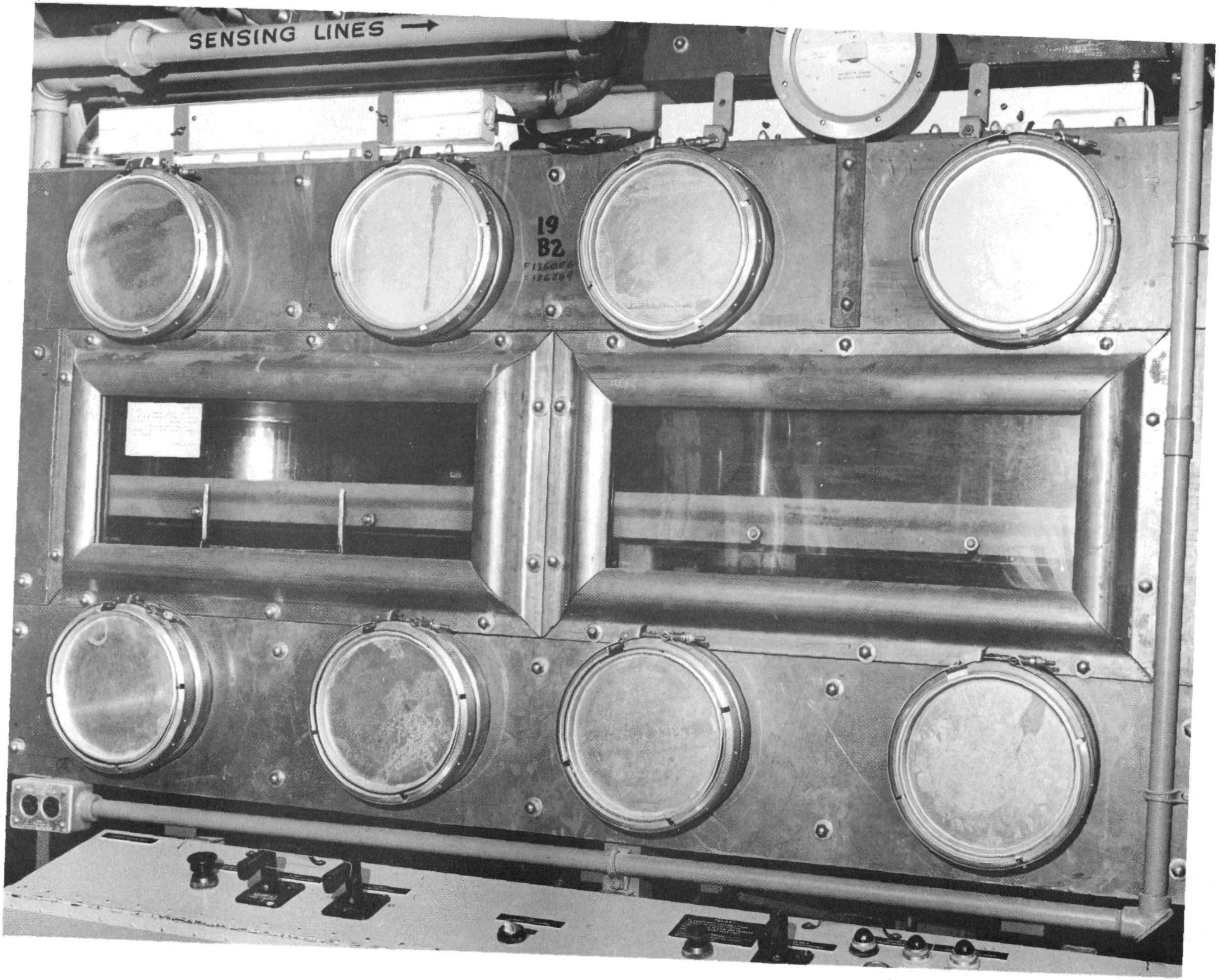


FIGURE VII

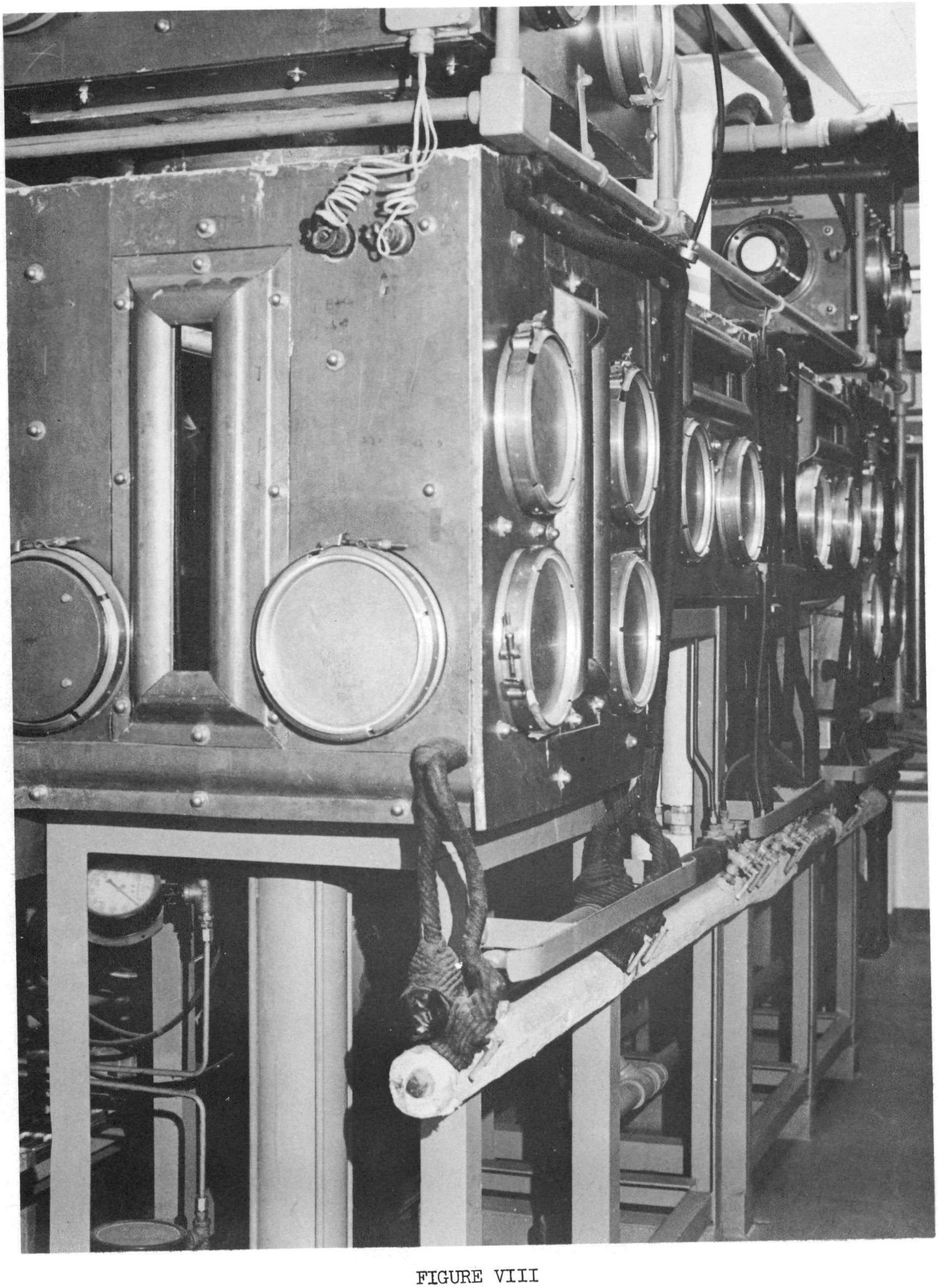


FIGURE VIII

GLOVEBOX FIRE-RESISTANT MATERIALS AND FIRE SUPPRESSION TESTS

F. D. Fisher

Nuclear Materials and Equipment Corporation
Apollo, Pennsylvania
A Subsidiary of Atlantic Richfield Company

April 13, 1971

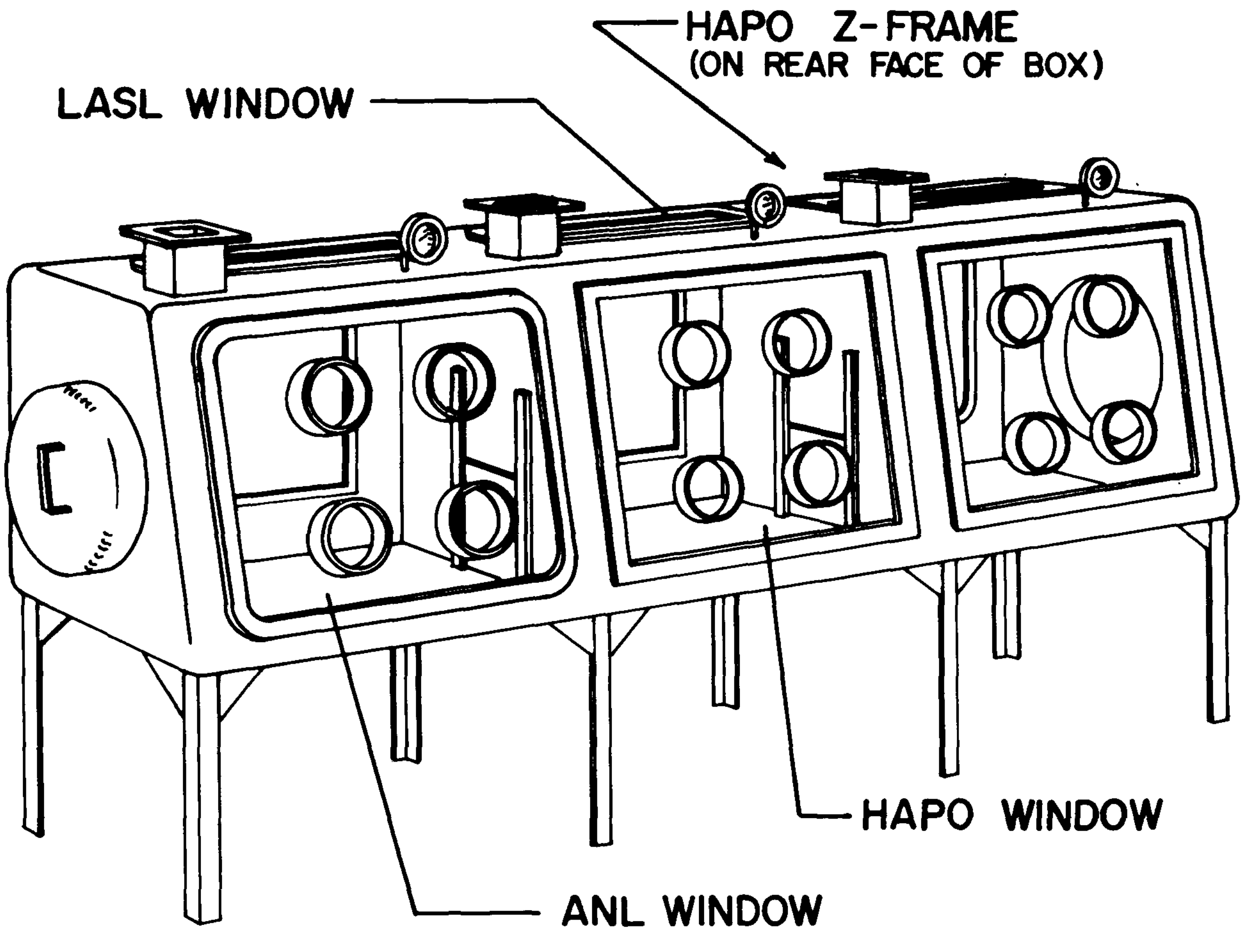
GLOVE BOX FIRE-RESISTANT MATERIALS AND FIRE SUPPRESSION TESTS

An industry-funded, AEC-sponsored "Ad Hoc" Committee has been formed to test and report on glove box window materials and fire suppression systems. The committee's study will include input from glove box users, glove box fabricators, glazing materials experts, fire suppression systems suppliers, a professional fire protection engineer and representatives from the AEC, Division of Operational Safety. The testing program is intended to complement those studies done here at Rocky Flats and elsewhere. Since our testing program is primarily oriented around the manufacture of PuO_2-UO_2 fuels for power reactors, rather than the manufacture of metallic plutonium assemblies, the emphasis of our testing program is expected to differ somewhat from similar programs done here at Rocky Flats.

The actual fire testing will be done at facilities provided by Overly Manufacturing Company, Greensburg, Pennsylvania, who are building a three-bay glove box, one side slanted and one side straight, to be used for the burning tests, c.f., Figures 1 and 2. The glove box will be equipped with a typical glove box ventilation system, c.f., Figure 3, consisting of an inlet filter, a metal mesh exhaust prefilter, and an exhaust damper for each of the three bays of the glove box. The three individual exhausts are manifolded together through an absolute filter into an exhauster which can function at very high temperatures to provide "glove box vacuum" even at the height of a fire. The inlet nozzles and exhaust nozzles of each of the three bays of the glove boxes are positioned such that their functions may be interchanged by simply relocating the exhaust manifold, the exhaust prefilters and the three inlet filter assemblies. Thus, the box can be run with either upflow through each bay when the ventilation is connected as in Figure 4, or with downflow when the ventilation system is connected as in Figure 5.

The glove box fire-resistant materials portion of this study is, at least for this first phase, limited to studies of the relative performances of different glazing materials. All commercially available vendor-recommended glazing materials will be tested under realistic in-fire conditions. Four window mounting schemes will be tested. They are as shown in Figure 6. I have labelled them, somewhat subjectively and largely arbitrarily, as ANL Type, HAPO Type, HAPO Z-Frame Type and LASL Type. The ANL-Type and LASL-Type will be tested both with and without restraining clamps necessary to provide mechanical support after the gaskets burn away. It is intended to test the various type window materials and window mounting means in different locations and with the two different ventilation schemes in the testing program.

Fire suppression systems to be tested will be largely confined to Halon 1301-based systems as provided by Safety First Products, Inc. Testing of those systems will be done both with simulated representative glove box "trash" to determine system effectiveness with such materials, and with n-hexane fuel since it is virtually a standard test fuel within the fire fighting systems industry.



THREE BAY GLOVE BOX
FOR FIRE TESTING
FIG I

NUMEC

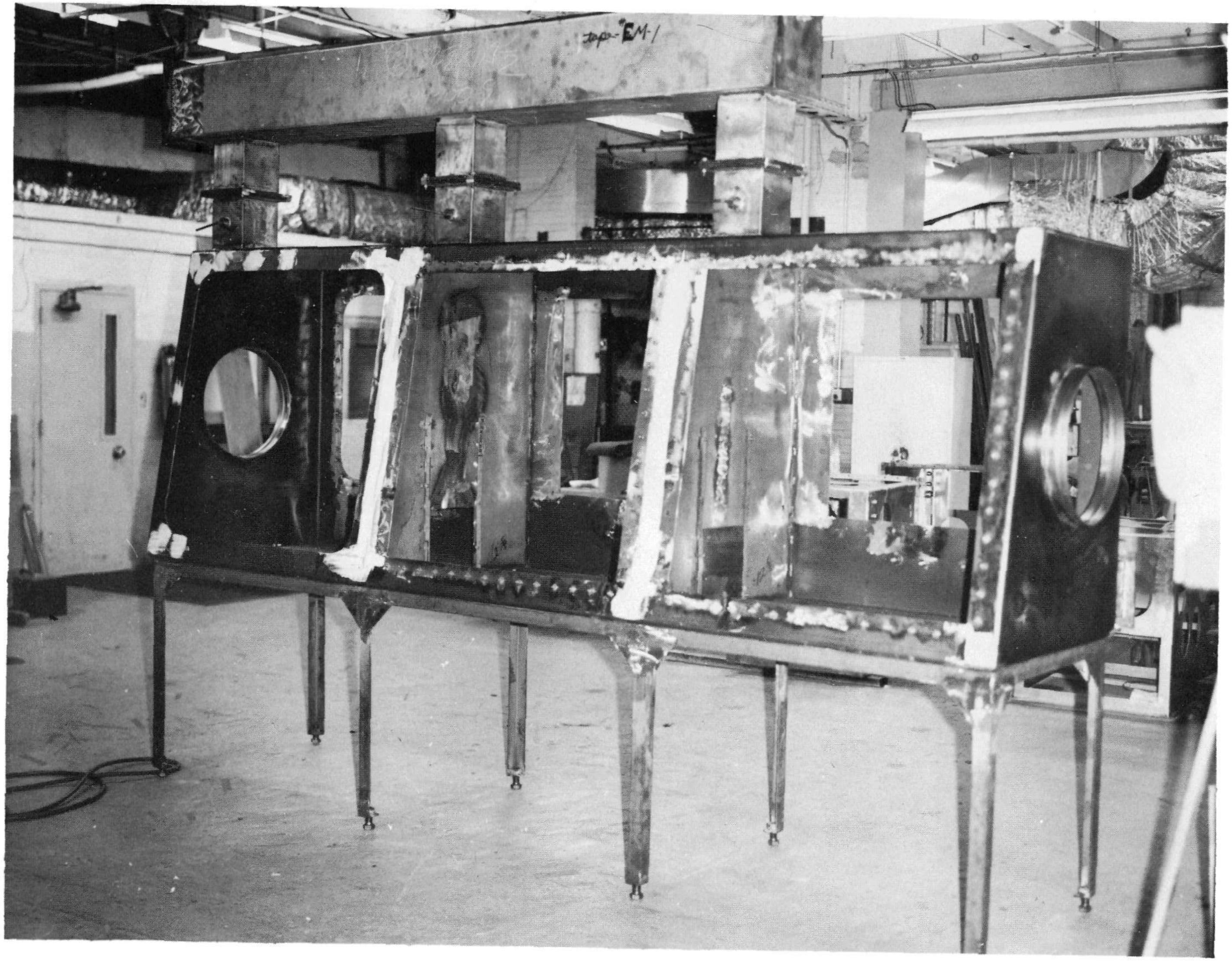
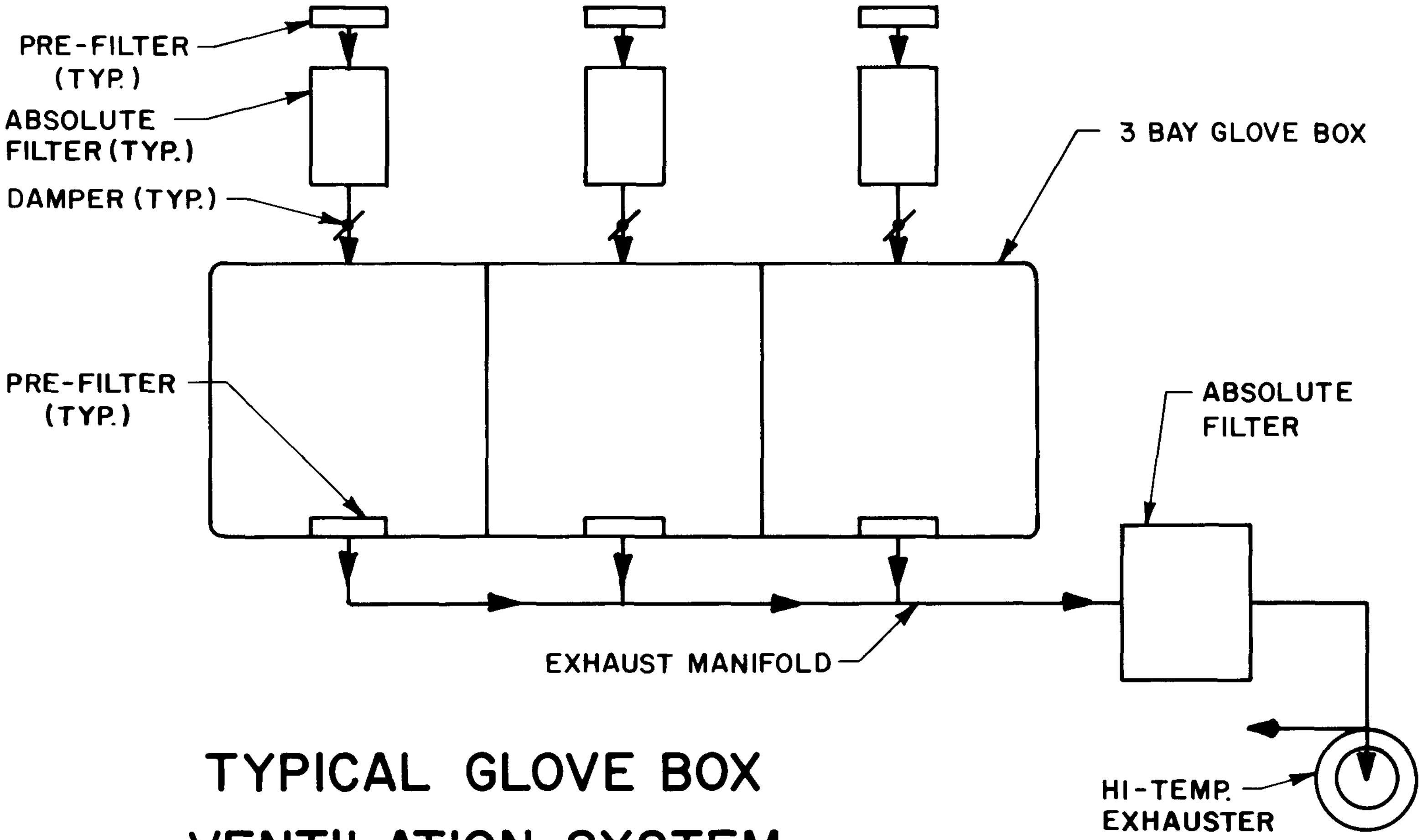


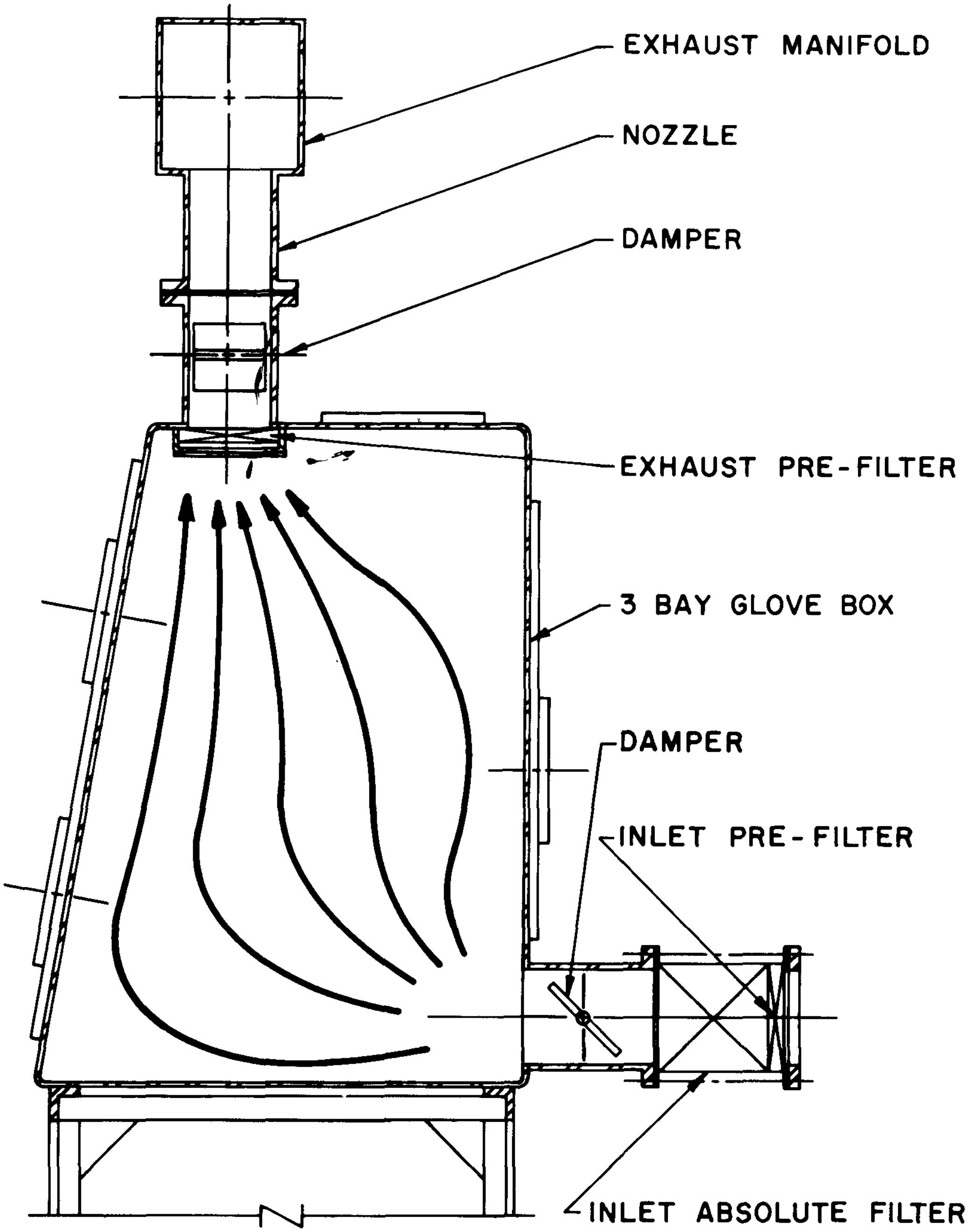
FIG 2 THREE BAY GLOVE BOX DURING CONSTRUCTION



TYPICAL GLOVE BOX VENTILATION SYSTEM SCHEMATIC

NUMEC

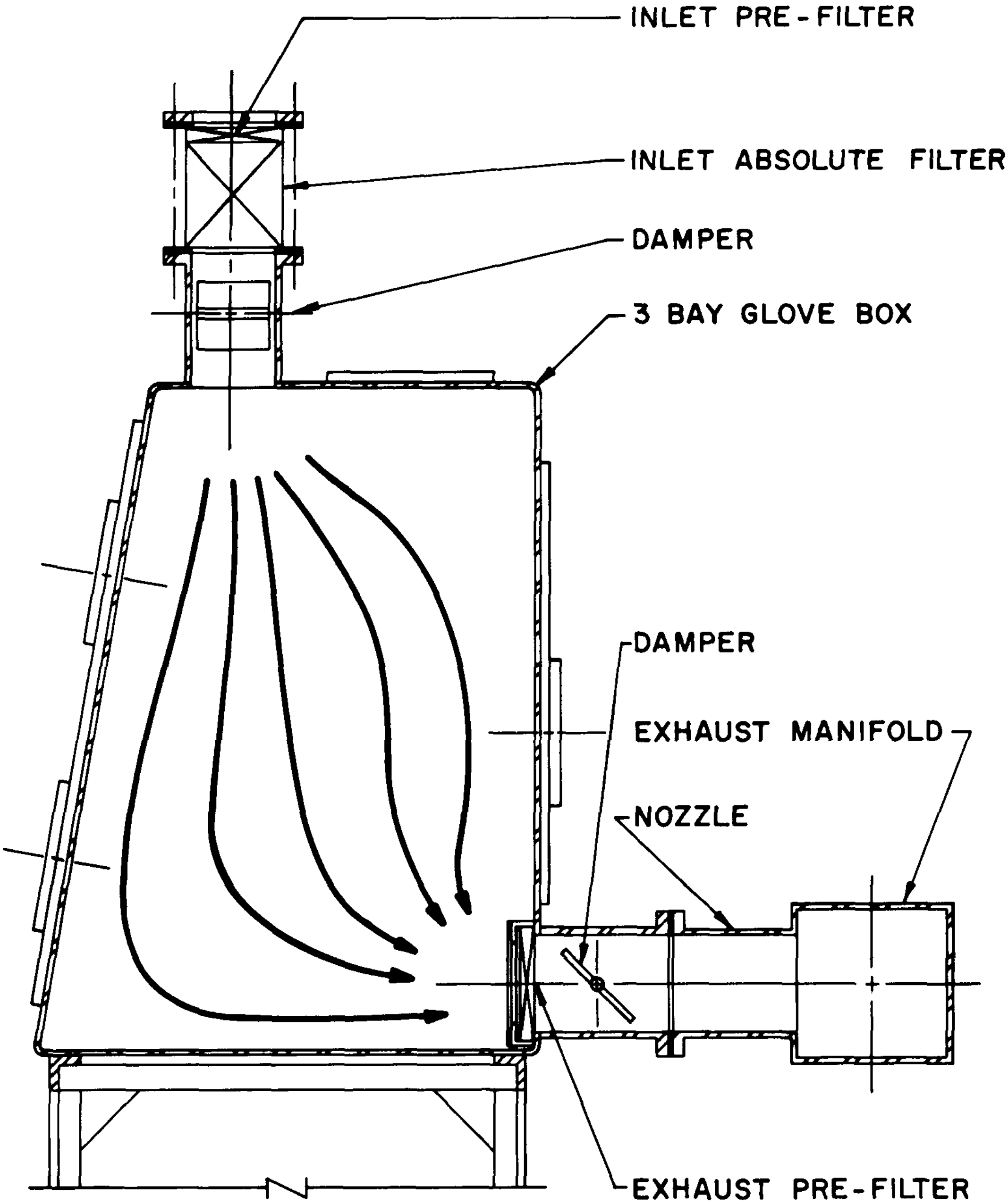
FIG 3



UPFLOW VENTILATION
ARRANGEMENT

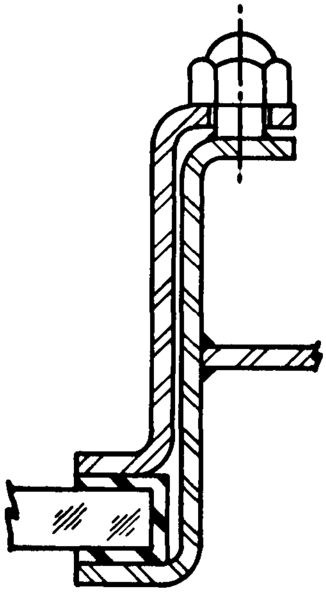
FIG 4

NUMEC

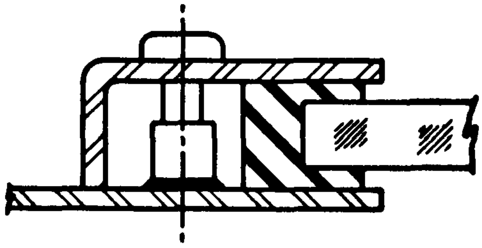


DOWNFLOW VENTILATION ARRANGEMENT
FIG 5

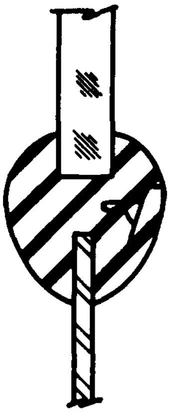
NUMEC



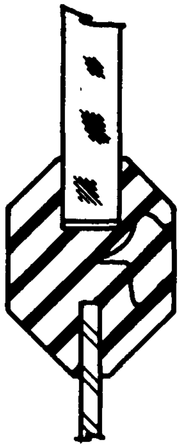
HAPO Z-FRAME



HAPO



ANL



LASL

GLOVE BOX WINDOW MOUNTING SYSTEMS

FIG 6

NUMEC

The fire detection portion of the fire suppression systems are, for the present anyway, regarded as beyond the scope of this study. Hence, the fire suppression systems in the study will be manually rather than automatically "triggered" during the testing program. However, they will be designed such that automatic activation could be readily incorporated into future real glove box installations.

A single report is planned. One which, in addition to the results of these tests, will include fire-related physical and chemical properties data for the various window materials, as developed by other studies such as those conducted here at Rocky Flats and/or as supplied by glazing materials manufacturers. Data and conclusions from other fire testing programs and related programs, including suitable fire detection systems studies, will be included with, of course, due credit given to the originator(s). We intend that the report shall serve as a useful handbook for the industry and, particularly, for the glove box designers and fabricators whose prior experience with glove box applications may have been somewhat limited. The testing program is being monitored by the AEC, who will also publish the report.

My own personal opinion concerning this testing program has been that one of its major contributions would be the opportunity for glove box users to witness an accurately simulated glove box fire. In that regard, Milt and the other people here at Rocky Flats will be "stealing my thunder", so to speak, when they conduct burning demonstrations next Thursday afternoon. However, we still intend to have our tests open to any interested observers. We are now projecting commencement of actual burning the week of May 3, 1971. If I don't already have your name in my correspondence file and if you want to attend, please contact me:

Dr. Fred D. Fisher
NUMEC
609 North Warren Avenue
Apollo, Pennsylvania 15613
Phone: 412-842-0111

and I will see that you are notified of the testing schedule as soon as it is firmed up. I don't like to be so indefinite about the schedule, however, this testing program is being done completely on a volunteer basis and, thus, various unforeseeable exigencies of any of the participants can cause the actual testing to be delayed.

GLOVEBOX DESIGN AND OPERATION
GLOVEBOX MATERIALS STUDIES

John W. Lindsay

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April 13, 1971

U. S. Atomic Energy Commission Contract AT(29-1)-1106

GLOVEBOX DESIGN AND OPERATION

GLOVEBOX MATERIALS STUDIES

ABSTRACT

Because of the many environmental implications associated with plutonium, provision of maximum protection to plutonium handling facilities is of utmost importance. Such protection begins with the design of as failproof a glovebox system as possible. Proper design of a glovebox requires that potential hazards associated with the processes and materials it is intended to contain be well known. Once these hazards have been elucidated, the best available materials of construction for the system can be chosen, and the safest possible glovebox configuration designed.

INTRODUCTION

Building a safe glovebox system requires several important considerations. The first task facing the designer is one of determining the potential hazards associated with the materials and processes the box is intended to contain. Such questions as, what is the probability of occurrence and nature of potential flame and heat sources must be answered prior to choosing the best materials for construction.

Choice of the best possible material for use in a particular segment of a box is often a difficult task because of the many factors which must be considered. Determination of materials resistance to burn, for example, cannot be simply measured through a single standard test since phenomenon determining its resistance are numerous and complex. Use of a series of well thought out tests can however lead to a compiliation of data which will help in characterizing the materials thermal behavior. Because of the many variables involved, the tests chosen must simulate as closely as possible the actual conditions anticipated in the environment in which the material is to be used.

Unfortunately, some of the components of a glovebox are by necessity composed of organic based materials which will burn under sufficiently severe exposure to heat and oxygen. Protection of thermally vulnerable segments of a glovebox then becomes another important consideration in designing a safe glovebox system. Such protection can come in the form of thermal shielding, minimizing exposure, and in the reduction in the oxygen content of the atmosphere. Since the best material for a given situation will often be one that resists burning for the longest time, additional protection for the glovebox can be gained through the use of adequate thermal event detectors and extinguishment techniques.

This paper is concerned with some of the considerations which should be made prior to choosing a material for use in a glovebox and in analytical techniques which can aid in the choice. A new glovebox design which increases neutron shielding efficiency as well as lowers the total fuel content of the box is also discussed.

POTENTIAL HEAT AND FLAME SOURCES IN A PLUTONIUM HANDLING FACILITY

Most glovebox systems contain several categories of potential heat sources. Fires arising from ignition of rags, papers, and organic solutions can lead to rapid heating of the environment through both conductive and radiative heat transfer. Normally this type of fire is of a relatively short duration. Use of good flame retarding polymers in the glovebox system should prevent this type of fire from becoming too widespread.

Additional potentially hazardous materials,commonly found in a glovebox, include certain ion exchange resins and lead oxide impregnated drybox gloves[1] which have been exposed to nitric acid. Exothermic reaction from both of these latter materials can occur in the absence of oxygen at temperatures as low as 200 °C.

In the case of a fire involving massive amounts of metal, the demands on a polymeric construction material are more severe. Ignition of various forms of plutonium metal can occur over a wide range of temperatures. Metal fires involving plutonium can contribute sizeable quantities of heat to the environment by both conductive and radiative means. Conversion of plutonium to its dioxide releases 1.06 Kcal per gram of metal and can result in maximum temperatures in excess of 1000 °C. Table I shows the results of work by Felt[2] on the burning characteristics of plutonium metal in various configurations. These tests were made using a transite-board base for the metal and a carbon arc as the initiator. The ignition temperature of plutonium is a function of several factors. Those factors which seem to have the greatest effect on the ignition temperature include metal purity, configuration, and surface composition. Heat transfer between the metal and its environment should also be added to this list since ignition may occur readily in insulative environments and only with difficulty in the presence of a large heat sink.

CHOICE OF MATERIALS TO BE USED IN A GLOVEBOX SYSTEM

Once the potential hazards associated with processes and materials to be contained in a glovebox are known, a proper choice of construction materials can be made. Those segments of a glovebox which are most vulnerable to a thermal event are listed in Table II. Most of the materials suitable for use in the various categories fail at relatively low temperatures, compared to those which could theoretically be reached in the event of a large-scale metal fire.

The job of determining the most suitable material for a given situation requires knowledge of the materials behavior over a wide range of conditions. One way of choosing the optimum thermal characteristics is by looking at the burning process which can be roughly broken into three stages as shown in Table III. In the initial stage, heat from an external source raises the temperature of the material. Materials having high specific heats and low thermal conductivities are desirable because of their slower rises in temperature and rate of heat transfer. During this stage, materials having higher glass transition or melting temperatures will retain physical characteristics longer.

TABLE I THERMAL LIMITATIONS OF GLOVEBOX MATERIALS

Metal Configuration	Ignition Times Required	Temperature Profile	Comments
Alpha-phase massive cylinders, 2 in. dia. by 2 in. high; total weight 1 to 3 kg	60 to 70 sec., spread of burn took 12 to 15 min. to completely encompass sample.	850 °C peak temperature reached during the first 12 to 15 min. Temperature dropped to 600 °C in next 30 min. and in the final stages showed a gradual decrease of 20 °C/hr.	During burn, the metal melted and showed a viscosity similar to molten lead. Average burning rate 180 g/Pu per hour.
Alpha-phase small, 200 to 1000 g	40 to 45 sec., burn spread completed in ~6 min.	Initial peak temperature 850 °C followed by a rapid decrease to 350 °C.	Generally similar to massive pieces with a shorter burn time.
Delta-stabilized small, ~700 g	50 t0 60 sec. longer than similar alpha with similar burn spread.	Initial peak temperature 850 °C with a final burn rate somewhat slower than seen for alpha metal.	Similar to alpha with longer ignition times.
Delta-phase metal turnings, 200 to 215 g.	Ignition instantaneous with very rapid burn.	Initial peak temperature of 850 °C reached in 4 min. Following 30 min. showed a rapid decrease to 415 °C.	Following peak, turnings assumed a semi-molten mass. Complete oxidation to 120 min.
Misc. hood wastes, fillings, oxides, sweepings with hydride possible 200 to 350 g	Ignition very rapid.	Peak temperature of 1300 ° to 1350 °C reached in 3 to 5 min. Rapid drop over next 30 min. to 450 °C.	Molten mass at white heat.
Casting skulls 150 to 265 g	Ignition instantaneous.	Peak temperature of 620 ° to 935 °C reached in 3 to 4 min.	Small amount of sparking noted.

TABLE II THERMAL LIMITATIONS OF GLOVEBOX MATERIALS

Box Segment	Material	Approximate Temperature at which Material Fails or Becomes Hazardous
Glovebox Gloves	Neoprene/Hypalon	200 °C
Window Materials	Polycarbonates	300 °C
	Polyesters	300 °C
	Polymethyl methacrylates	200 °C
Gasket Materials	Fluorel®	200 °C
	Viton®	200 °C
	Silicone Rubber	200 °C
Shielding Materials	Benelex®	300 °C
	Polyethylene	100 °C
Glovebox Filters	HEPA	200 °C
Miscellaneous	Teflon®	400 °C
	Polyvinyl Chloride	100 °C

Fluorel® Minnesota Mining and Mfg. Co. St. Paul, Minn.
Viton® E. I. du Pont de Nemours and Co., Wilmington, Dela.
Benelex® Masonite Corp., 29 Wacker Dr., Chicago, Ill. 60606
Teflon® E. I. du Pont de Nemours and Co., Wilmington, Dela.

TABLE III DESIRABLE BEHAVIOR OF MATERIAL PRIOR AND FOLLOWING IGNITION

A. Heating Stage

1. High specific heat.
2. Low thermal conductivity.
3. High glass transition temperature and melting point.

B. Decomposition Stage

1. Minimal evolution of flammable toxic and corrosive gases.
2. Minimal evolution of smoke.
3. Absence of liquid pyrolysis products.
4. Maximum retention of configuration.

C. Ignition, Combustion, and Propagation Stages

1. High flash temperature of evolved gases.
2. High autoignition temperature.
3. High limiting oxygen index.
4. Low flame spread.
5. Maximum retention of configuration.

In the second stage, the material reaches its decomposition temperature. Desirable characteristics of a material during this stage include a minimal evolution of flammable gases, since in most cases ignition and combustion occur in the gas phase. Evolution of corrosive and toxic gases as well as smoke should also be minimal because of the effect on environment and fire fighting efforts. Evolution of smoke and corrosive gases is normally increased in flame retarding plastics so that gaining time prior to ignition may in some cases be tempered by adverse conditions following combustion. Many plastics form liquids on decomposition. This phenomenon is undesirable because of the potential hazard of spreading the fire to unaffected areas. Materials which retain a certain degree of configuration through a charring process, on the other hand, impede mixing of air with combustible cases and serve to slow flame spread to adjacent materials. Retention of configuration is an important consideration in gloveboxes where containment of radioactive materials is desirable.

Following decomposition of the material, ignition and combustion will occur providing sufficient heat and oxygen are available. Evolution of gases having a high flash temperature during the decomposition stage is certainly desirable, as is a high temperature of autoignition. Materials having high values of limiting oxygen concentrations (> 0.21) will not ignite under normal conditions in air. Once combustion has occurred, spread of the fire will occur most readily along exposed surfaces. Materials having slow surface burning rates will delay propagation.

CHOICE OF TESTS TO EVALUATE A MATERIALS THERMAL BEHAVIOR

Importance of choosing the proper material raises the question of what types of tests are most valid. In the case of a glovebox, environmental conditions are somewhat unique because of the forced draft. Such movement of air can contribute large volumes of oxygen which will accelerate flame propagation and result in rapidly moving flame fronts to previously unaffected areas. The ultimate solution to the problem of testing materials, of course, is to construct a full-scale glovebox using prospective materials. In this manner, the material's thermal behavior can be tested under actual conditions, thus eliminating uncertainties, such as the effects of drafts, position, geometry and proximetry of materials, and actual temperatures. Such a series of tests has and is being carried out at Rocky Flats.[3] Figure 1 shows the test facility. Because of the expensive and time consuming nature of such large-scale tests, need for meaningful laboratory screening tests arises. Values of a material's specific heat, thermal conductivity, glass transition temperature, and melting point are useful in characterizing a material's behavior during the initial heating stage. This type of data is easily obtained in the laboratory and is often readily available from the manufacturer. Characterization of other aspects of a material's thermal behavior can be made through a series of appropriate ASTM and specifically designed tests.

Information on the decomposition of a material can be gained rapidly through the use of simultaneous thermogravimetric (TGA) differential thermal (DTA) and effluent gas analyses (EGA). Data from these analyses gives temperatures of stability, relative rate of decomposition as a function of temperature, types and quantities

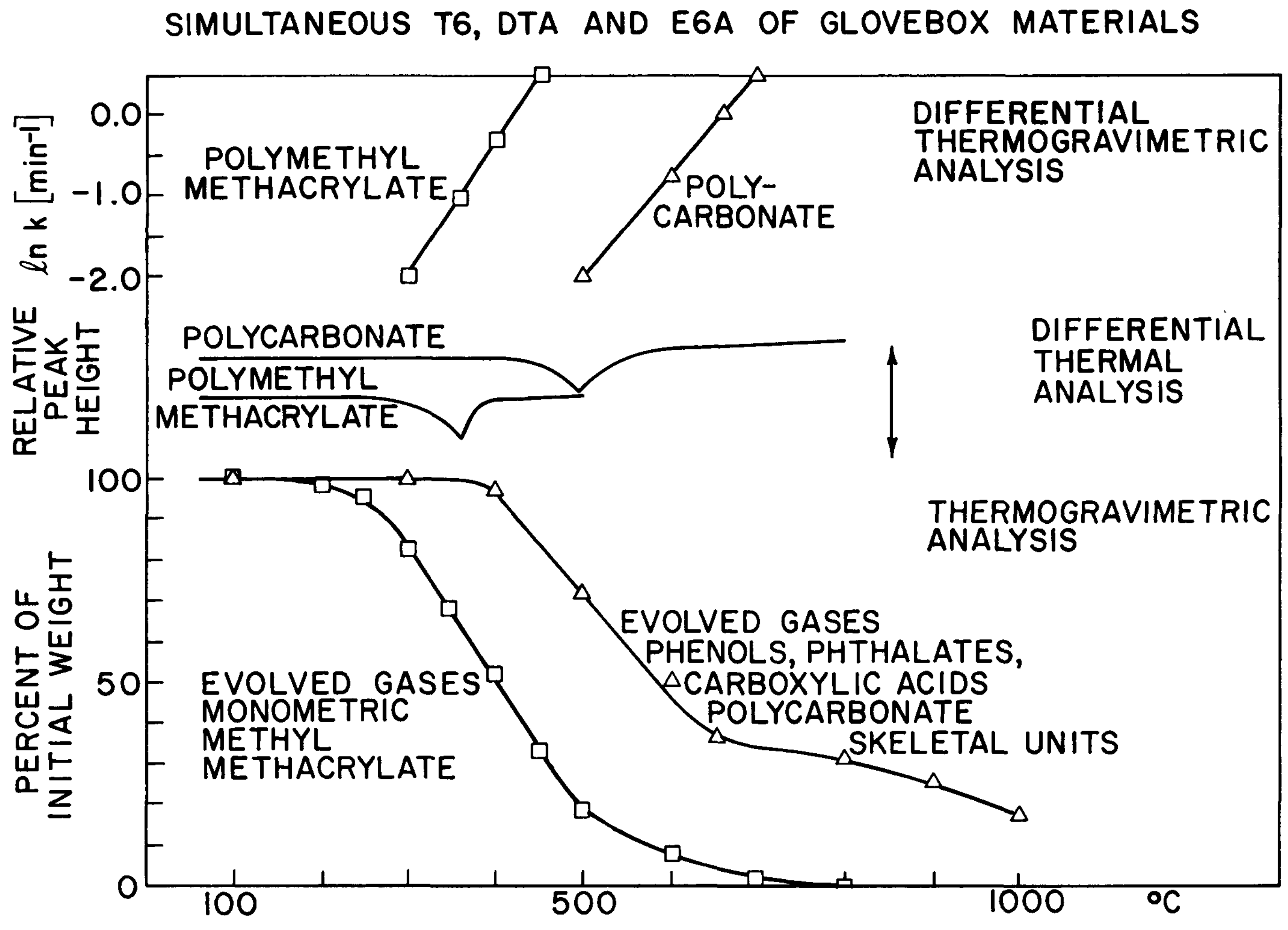

FIGURE 2

Simultaneous TG, DTG and EGA of Glovebox Window Materials

TABLE IV THERMOANALYTICAL DATA OBTAINED FROM VARIOUS GLOVEBOX MATERIALS

Material	Initial Decomposition Temperature	Gaseous Pyrolysis Products	Differential Thermal Analysis
Silicone Rubber Gasket	300 °C	Benzene Methane Ethylene	Exothermic
Neoprene Gasket	220 °C	High molecular wt petroleum products, styrene polymeric units.	Exothermic
Polyethylene	350 °C	Ethylene Ethane Methane	Exothermic
Teflon®	500 °C	Hydrogen Fluoride Tetrafluorethylene	Exothermic
Neoprene-lead Oxide Drybox Gloves	280 °C	-	Exothermic
Polyvinyl Chloride Bags	200 °C	Hydrochloric Acid various chlorinated hydrocarbons	Exothermic

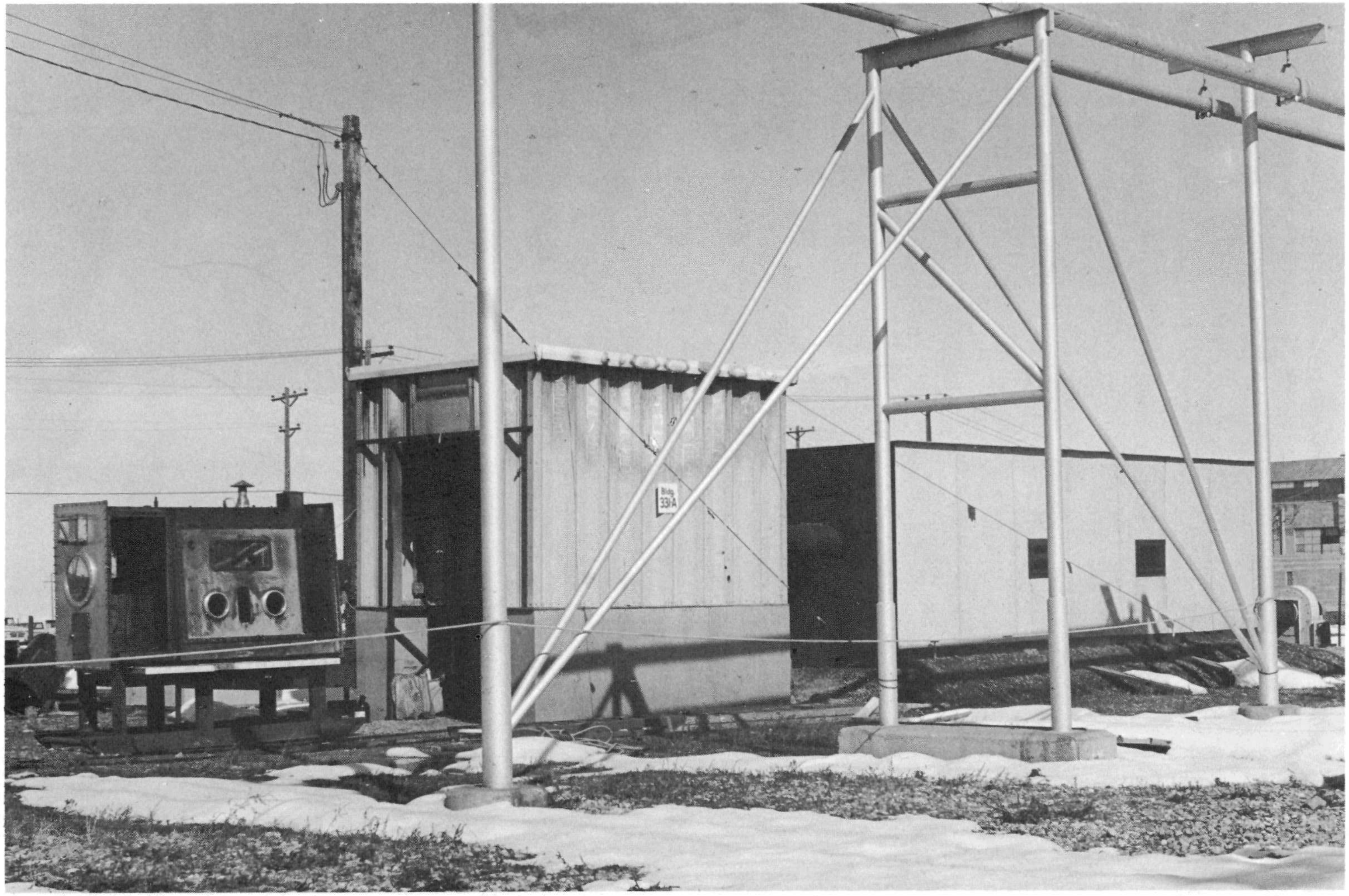

FIGURE 1

Fire Test Facility

of gaseous pyrolysis products, melting points, and information about the overall decomposition. Polymers shown in the example in Figure 2 are commonly used window materials. Approximate rate of the pyrolysis process can be determined simultaneously with the other analyses using an analog system available in many commercial thermoanalyzers. Examples of thermoanalytical data for other materials commonly used in gloveboxes are given in Table IV.

Many tests for the ease of ignition of a material are available. However, the limiting oxygen index or L.O.I. is probably the most reproducible and gives a good predictive index of a material's readiness to ignite and burn. L.O.I. values for polymeric materials common to gloveboxes are given in Table V[4] along with other data which can be readily obtained on a laboratory scale.

While data such as that shown in Tables IV and V provide a basic insight into the thermal behavior of a material, additional information such as flame spread, smoke evolution, effect of pyrolysis products on HEPA filters, etc., is still needed. Such data cannot as a rule be obtained from ordinary ASTM tests and applied to glovebox materials, due to the unique conditions of drafting in a glovebox system. Figure 3 shows a laboratory scale test unit which is being developed at Rocky Flats specifically for the evaluation of glovebox materials and plenum linings. This unit allows for a stringent test of materials under conditions which might be anticipated in an actual fire. The small scale unit which uses samples of 7" x 4" x 1/2" in size, offers both variable air flow and environmental temperature. Times to ignition and surface regression rates are measured automatically through the use of two microswitches which are activated by the burning of nylon threads across the sample surface. Heat for ignition is supplied by an impinging propane flame at one end of the sample.

Environmental temperature is preset prior to flame impingement through the use of an external heating element. Additional information on surface temperatures and regression rate is obtained from a series of thermocouples imbedded in the sample surface (Tc_1 to Tc_5). Relative amount of smoke evolved during the burning process is measured optically in the upper duct of the unit. Thermocouple Tc_6 measures exhaust gas temperature. Effect of the gaseous pyrolysis products on HEPA filter media is recorded by measuring pressure drop across a segment of the filter media held in the upper duct. Composition of the gases is determined through mass spectroscopy.

In general, tests of the type described can prove invaluable in the characterization of a materials thermal behavior over a wide range of conditions. This understanding is necessary along with knowledge of the potential hazards associated with materials and processes in a glovebox before a safe system can be constructed.

IMPROVED GLOVEBOX DESIGN

The final consideration in constructing a safe glovebox system is found in the design of the box itself. Minimizing the use of flammable materials, inert atmospheres, and thermal shielding of the flammable materials used are a few of the more important design criteria which should be considered. One of the largest contributors to the total fuel content of a glovebox is the neutron

TABLE V THERMAL PROPERTIES OF VARIOUS POLYMERS COMMON TO GLOVEBOXES

Material	Polyethylene	Teflon®	Polyvinyl Chloride	Polymethyl Methacrylate	Poly Carbonate
Decomposition Temperature °C	350	500	200	185	380
Specific Heat cal/g/°C	0.55	0.25	0.25	0.35	0.30
Thermal Conductivity 10^{-4} cal/sec^{-}cm^{2}	8.0	6.0	~ 5.0	5.0	4.6
Evolved Vapor Flash Temperature °C	340	-	340	280	420
Autoignition Temperature °C	350	530	454	450	N.A.
Limiting Oxygen Index	0.17	0.95	0.49	0.20	0.26

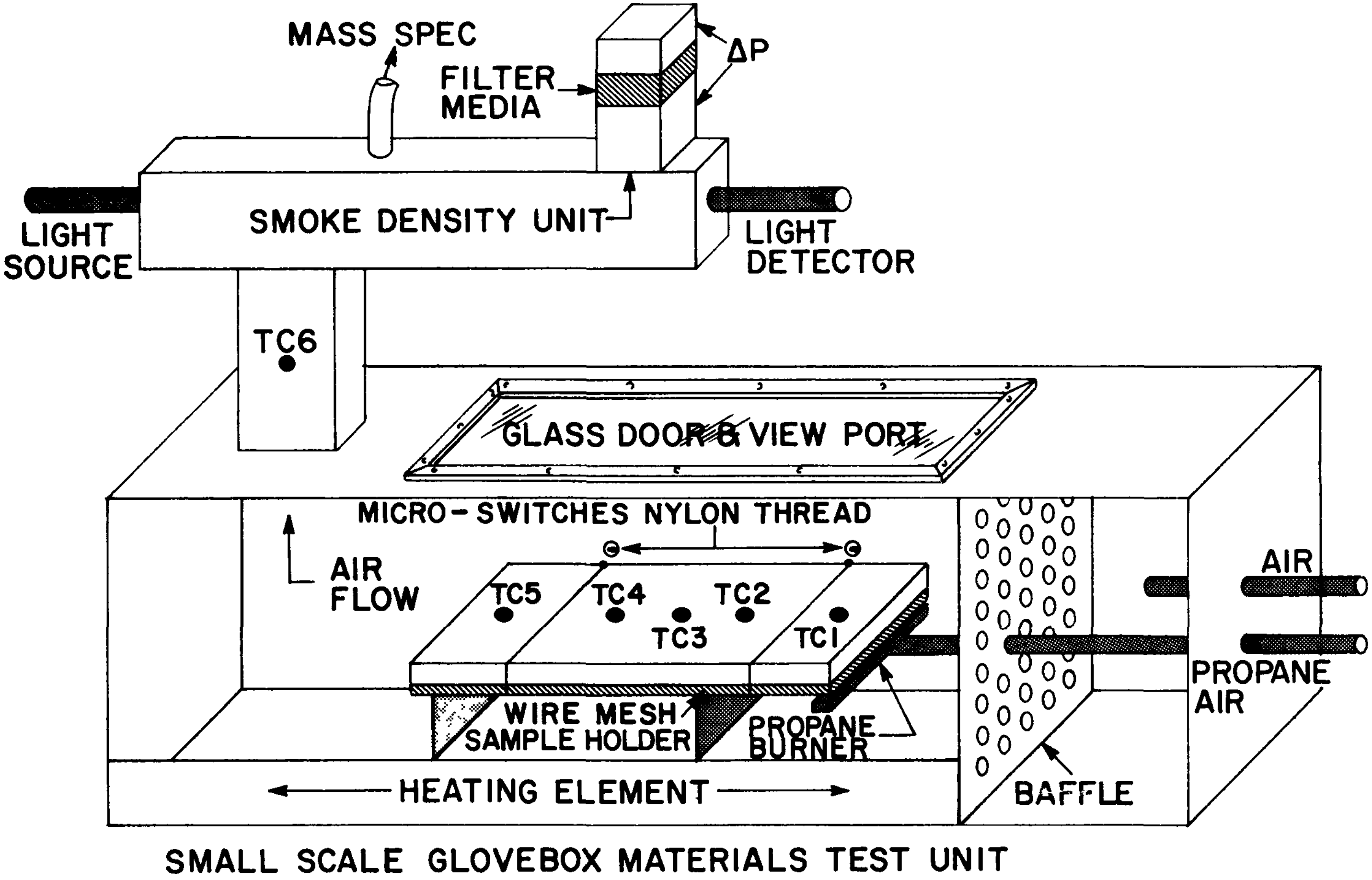

FIGURE 3

Small-Scale Test Unit for the Evaluation
of Glovebox and Plenum Materials

shielding. Massive pieces of Benelex® and Plexiglas G® are presently used at Rocky Flats. Figure 4 shows a new glovebox design which is presently being developed at Rocky Flats. Shielding in the box is contained between its double wall configuration and in specially designed windows. Spacing between the walls allows for 2 inches of shielding and eliminates the requirement for draping bulky shielding on the box exterior, as has been the practice.

Prospective shielding for the box consists of a honeycombed hydrophylic network type polymer which contains about 90 percent water by weight. Thermal behavior of the polymer is seen in Figure 5. Decomposition begins at about 100 °C and proceeds in two stages. The initial stage which is highly endothermic, results in the evolution of water. This stage is followed by pyrolysis of the remaining organic material at about 450 °C. No liquification of the polymer occurs either during its decomposition at elevated temperatures or when it is subjected to external pressure.

Because of its high hydrogen density, the polymer which is known at this time as "gelled water" or "Texgel," affords excellent protection against neutron emissions. Shielding efficiency against 1 mev neutrons emitted from a PuF_4 source is shown in Figure 6. Of the more commonly used shielding materials, only polyethylene has a higher efficiency. Degradation of the polymer by radiation appears to be minimal with some additional cross-linking actually occurring during extended periods of exposure.

Corrosion of stainless steel by the polymer appears to be minimal. Material stored in a type 304 stainless steel container for a period of eight months showed good retention of its physical properties and only small changes in the water content. Because of its high degree of clarity, lack of bubbles, and color, the polymer has been found suitable for use as a filler for glovebox windows of the type shown in Figure 4.

In addition to the increased shielding properties, use of the polymer should provide a large decrease in the total fuel content of the glovebox system as well as provide a sizable heat sink at the box faces in the event of a fire.

SUMMARY

Choice of materials for constructing a glovebox system should be made following a complete understanding of the potential hazards it is intended to contain. Because of the potential heat sources contained in most plutonium gloveboxes, flammable materials should be avoided where possible and afford maximum protection where their use is necessary. Evaluation of materials to be used in a glovebox should be made under realistic conditions. This, in most cases, will require large-scale testing in an actual glovebox. Screening tests such as the ones described can be used to lessen time and expense involved in the large scale tests. New glovebox designs such as the double walled box with "gelled water" shielding can be used to improve safety along with the choice of proper materials.

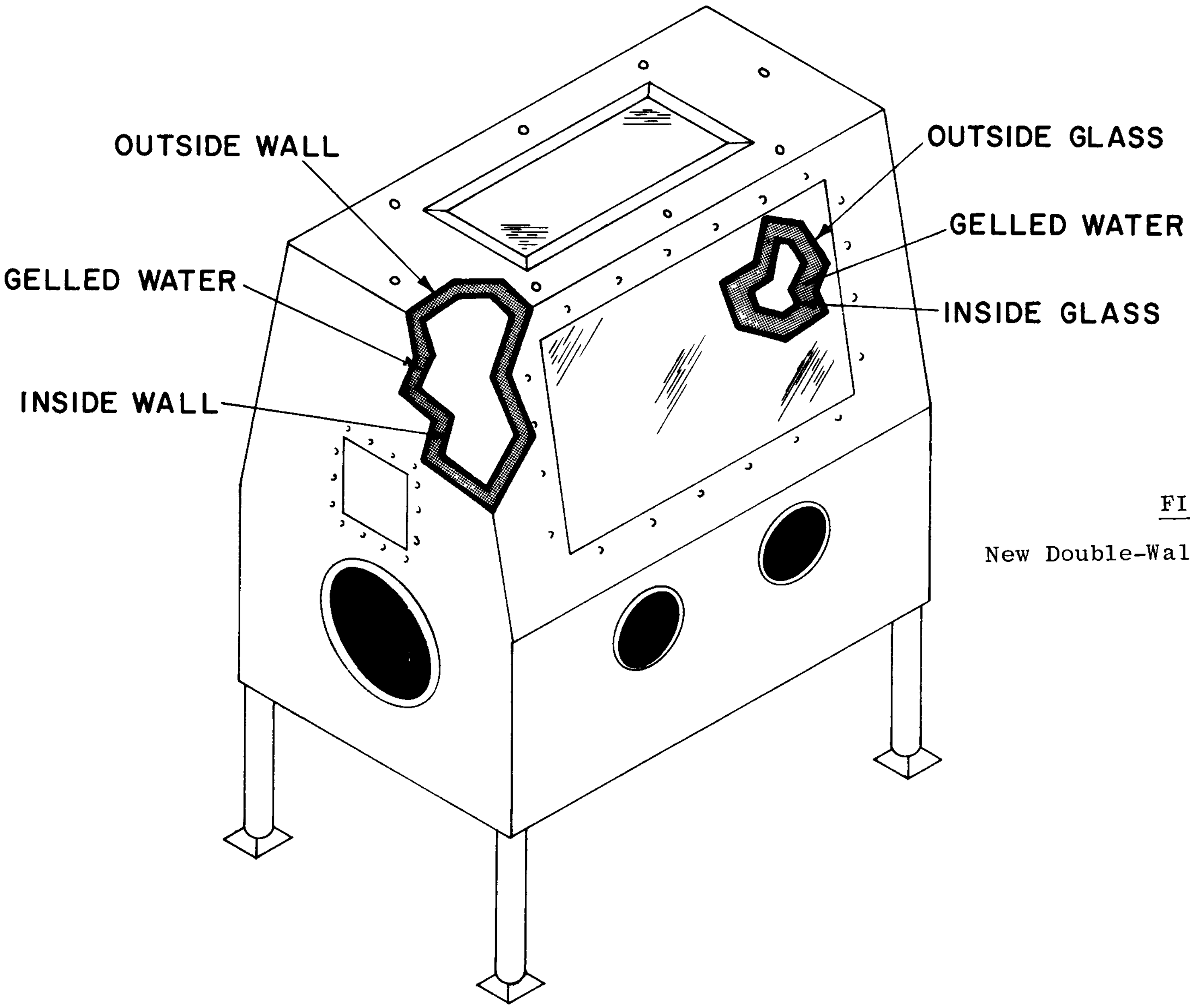

FIGURE 4

New Double-Wall Glovebox Design

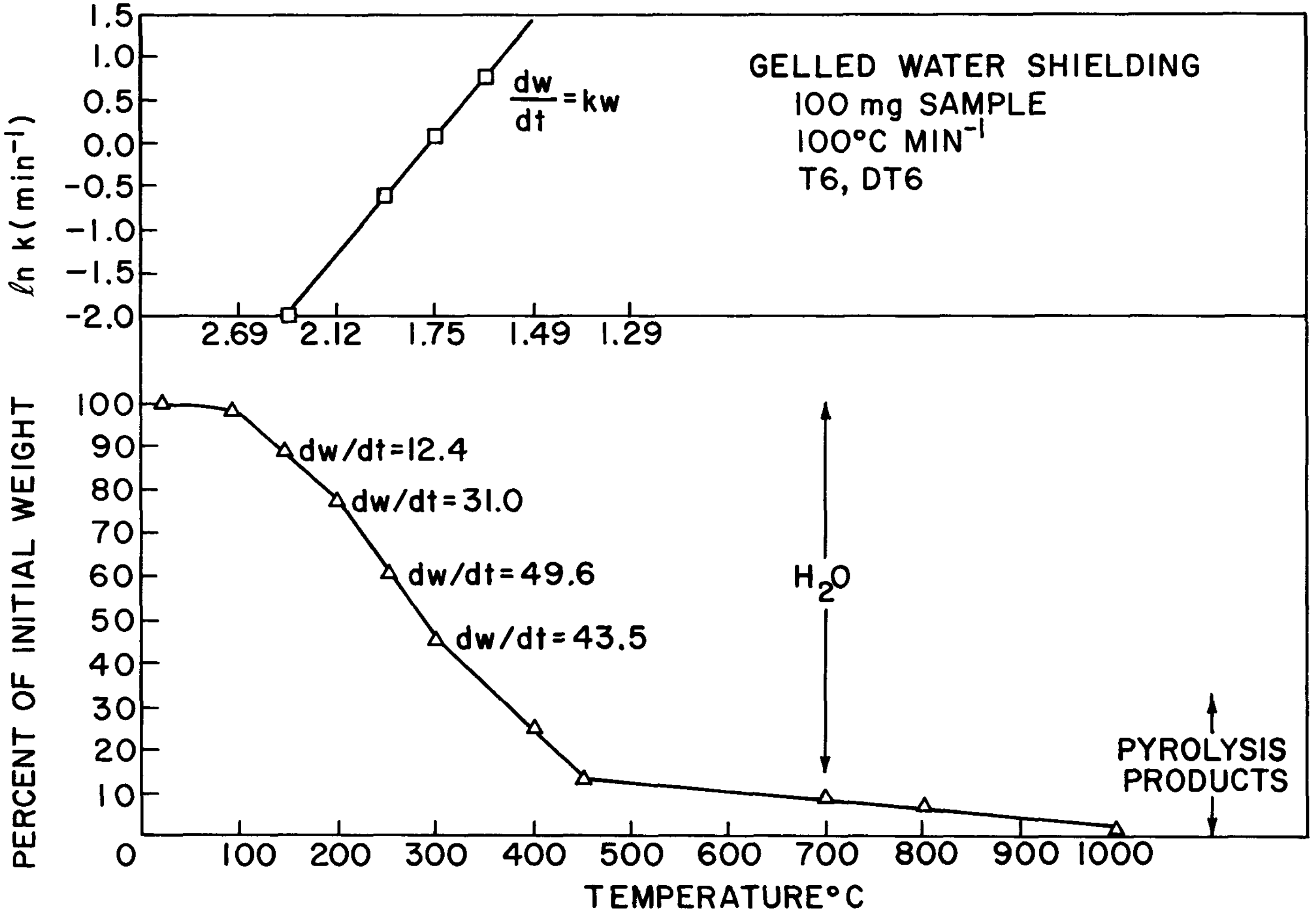

FIGURE 5

Thermal Behavior of Gelled Water Shielding Material

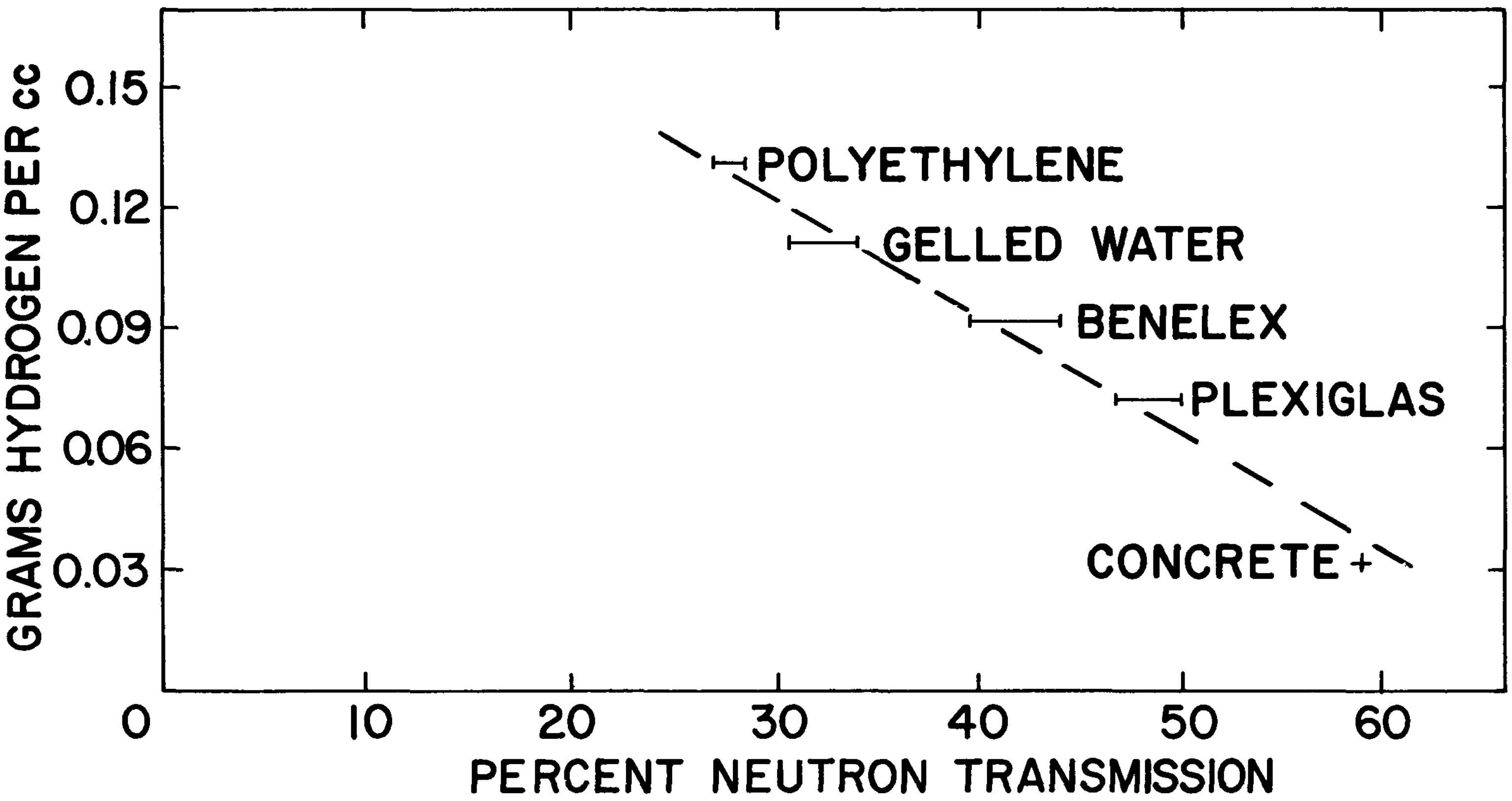

FIGURE 6

Comparison of Gelled Water Shielding with Other Materials

GLOVEBOX GLOVES

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ABSTRACT

Important or desirable properties of glovebox gloves are presented. A need to determine these properties has resulted in a program for glove evaluation and procurement. Gloves are most vulnerable to a glovebox fire. Burning properties and means of decreasing vulnerability are discussed.

INTRODUCTION

There are a limited number of ways in which plutonium can be handled by man. These might include programmed robots, enclosures with manipulators, and gloveboxes. Gloveboxes are in widespread use. The gloves used on these boxes must be functional, i.e. strong, flexible, comfortable, and protective. It is important that desirable properties of gloves be defined for procurement purposes and further, that capabilities be available for determining these desirable properties.

The burning properties of gloves are especially important. Loss of glovebox integrity usually occurs first through a gloveport during a fire. Protection of the port and the glove is necessary.

DISCUSSION

A problem at Rocky Flats is that currently we procure our gloves from a single source. It has been our experience in the past, and most hopefully is yours, that manufacturers will approach the glove user and say, "here, use my glove." As a government contractor, one must justify his choice of product; in this case, gloves. It therefore behooves the glove user to have desirable glove properties defined and further, to have a method to measure and compare these properties so that when a glove candidate becomes available, it can be properly evaluated for purchase and subsequent use.

Rocky Flats has such capabilities. A program for drybox glove evaluation and procurement was initiated about 8 years ago. It has progressed as follows:

First, testing procedures in the ASTM standards were reviewed. Then rubber testing facilities of rubber companies and glove manufacturers were visited. Finally an experimental program was initiated which resulted in a capability to test gloves. A procedure for evaluating gloves was established. Specifications were authored for each type of glove needed. Finally, plant acceptance and quality assurance procedures were established.

Glove properties which we deem important and which we choose to determine, are listed on a laboratory data sheet, (Figure 1). These include water vapor transmission, (a measure of film permeability), fire resistance, lead equivalency (shielding power), mechanical properties (tensile strength, ultimate elongation, modulus of elasticity, permanent set, puncture resistance), accelerated aging, dimensional properties, workmanship, packing, marking, economics (cost per day per glove), and finally chemical properties. A description of these test methods, a discussion of test results, criteria used for selecting gloves, and Rocky Flats' specifications for seven different types of gloves used at this plant are included in a topical report entitled "Drybox Gloves: Evaluation and Procurement." This report, RFP-1286, will be available in the near future.

At present, eight types of drybox gloves are stocked at Rocky Flats (Figure 2): 30-mil butyl, used occasionally for the ultimate in impermeability; 15-mil neoprene, used where good dexterity and touch are necessary; 30-mil neoprene, a general service glove; 30-mil lead neoprene (0.1 mm lead equivalency), widely used in plutonium areas where some gamma ray protection is needed; Hypalon-coated gloves, both leaded and non-leaded, used where glove failure is prominent from acid attack or ozone aging; 45-mil leaded (0.2 mm lead equivalency) Hypalon-coated gloves; and 80-mil leaded (0.36 mm lead equivalency) Hypalon-coated gloves of multi-layered construction, used in areas where high penetrating gamma radiation is present, such as in americium processing.

The drybox glove itself is usually the most vulnerable glovebox component to the effects of a fire. It is first to cause loss of glovebox integrity. To date, a flexible glove that cannot be breached by a major fire has not been developed.

We evaluate the burning properties of glove materials by burning strips held vertically. A burner flame is applied to the bottom of the strip. After the material has ignited and a flame is established, the burner flame is removed. Results are compared to the burning properties of 30 gauge neoprene glove material. The neoprene requires flame exposure for several seconds to accomplish ignition. After a good flame is established and the burner is removed, the neoprene continues to burn slowly. The burning properties of lead-loaded neoprene and Hypalon-coated gloves are not significally different from neoprene.

We are currently experimenting with a Dow Corning silicone elastomeric polymer as a material from which to manufacture drybox gloves. This polymer exhibits a resistance to fire superior to neoprene and it possesses the strength and elastomeric properties necessary for a flexible glove. The silicone material was self-extinguishing by our burning test. A glove of this material would not show much improvement in containing a major fire.

A number of means for protecting the present gloves have been evaluated. An aluminized asbestos blanket can be hung above the glove port (Figure 3). This is cheap, simple, but not too

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GLOVEBOX GLOVES

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Chemical Technology
The Dow Chemical Company
Rocky Flats Division
Golden, Colorado 80401

April 13, 1971

U. S. Atomic Energy Commission Contract AT(29-1)-1106

DATA SHEET FOR GLOVE TESTS

FIGURE 1

Glove Description:

Cost –

Sp gr – ________ g/cc

Water Vapor Transmission (Permeability):

0 percent to 50 percent Ralative Humidity Differential: ________ g/24 hr/M^2

Fire Resistance:

Lead Equivalence: ________ mm (X-Ray tube source)

Energy ______ kv; Current ______ Ma: Time ______ minutes;

Distance ______ inches

Mechanical Properties:

Tensile Strength: ________ psi

Force to Break: ________ lb.

Ultimate Elongation: ________ %

Modulus at ______ % Elongation: ________ psi

Permanent set at 70% U. E. ________ %

Puncture: ________ lb, ________ inches travel

Accelerated Aging:

	Heat	Nitric Acid
Tensile Strength:	________ psi	________ psi
Force to Break:	________ lb.	________ lb.
Ultimate Elongation:	________ %	________ %

Dimensional Properties:

Diameters: ________ in. at cuff

________ in. at wrist

Bead: ________ in.

Taper ________ in./in.

Length ________ in. overall

________ in. third finger

________ in. cuff to taper

Hand Size: ________

Thickness: ________ in. palm

________ in. finger

________ in. wrist

________ in. arm

________ in. shoulder

Workmanship:

Packing:

Marking:

Economics:

Other observations, opinions, and recommendations:

FIGURE 1 (CONT'D)

Chemical Resistance:	Observations			
	1 hour	4 hours	24 hours	72 hours
Chemical				
14 M HNO_3				
Red Fuming Nitric Acid				
3 M HNO_3				
12 M HCl				
48 percent HF				
10 M KOH				
10 M NaOH				
Acetone				
Benzene				
CCl_4				
Trichloroethylene				
Freon TF				
Hydraulic Oil				
KW Decontamination Solution				

	Weight Change[a]		Observations After Drying[b]	
	Dry for 10 minutes	Dry for 48 hours	Dry for 10 minutes	Dry for 48 hours
13 M HNO_3				
3 M HNO3				
Red Fuming Nitric Acid				
12 M HCl				
48 percent HF				
10 M KOH				
10 M NaOH				
Acetone				
Benzene				
CCl_4				
Trichloroethylene				
Freon TF				
Hydraulic Oil				
KW Decontaminating Solution				

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[a] Weight change in milligrams per square centimeter from original after immersion for 24 hours and drying for time indicated. Percent of original weight is also given where sample was completely penetrated

[b] After immersion for 24 hours and drying for time indicated.

FIGURE 2

DRYBOX GLOVE APPLICATIONS

TYPE	APPLICATION
30-mil butyl	For extremely low permeability to water, oxygen or solvents and for certain chemical environments such as bromobenzenes and methylene bromide.
15-mil neoprene	Optimum touch and flexibility, lead protection not needed.
30-mil neoprene	General application, no lead protection.
30-mil leaded neoprene	General application, lead protection needed.
30-mil Hypalon-neoprene	For oxidizing acids or where failure results from aging or stress cracking (ozone attack).
30-mil leaded Hypalon-neoprene	Lead protection needed plus oxidizing acids, such as nitric acid, or where failure is generally due to aging and stress cracking (ozone attack).
45-mil leaded Hypalon-neoprene	As above, 50% additional lead protection.
80-mil leaded Hypalon-neoprene multi-layered	Flexible, 0.36-mm lead equivalency for areas of high or penetrating radiation (such as americium processing).

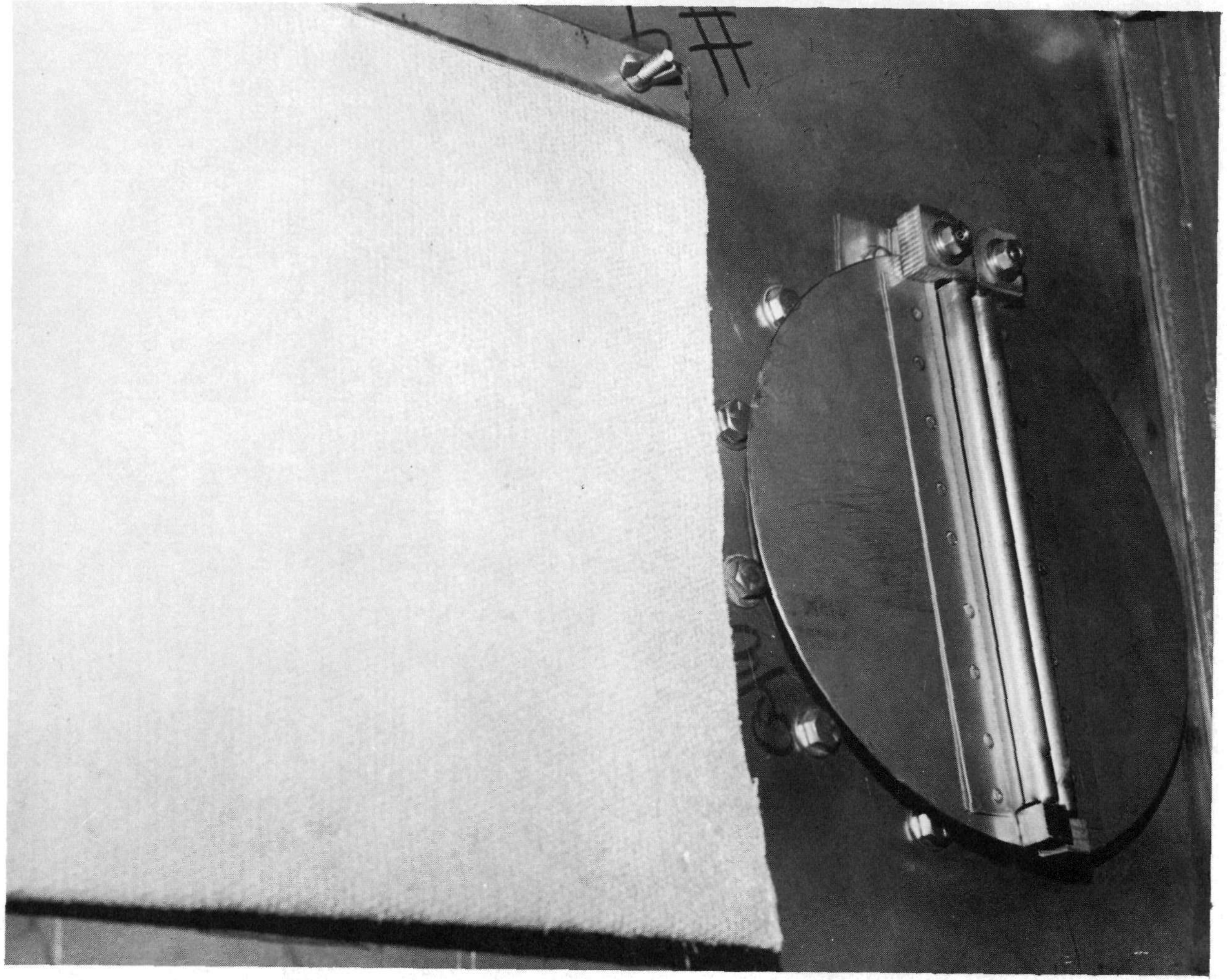

FIGURE 3

ALUMINIZED ASBESTOS BLANKET AND GUILLOTINE-TYPE STEEL PLATE ON INSIDE OF BOX FOR GLOVE PROTECTION effective. The cotton binder in the asbestos soon decomposes in the presence of a fire and the protection is lost. It buys some time. This is not currently being used anywhere at Rocky Flats except on our test facility.

A guillotine-type steel plate has been evaluated (Figure 3). It is placed in front of the glove port on the inside of the glovebox. Such a protective plate is much more effective than the asbestos curtain, but it is also breached by a bad fire because it does not form a seal over the port. The heat and flames can circumvent the plate and will eventually destroy the glove. This type of protection might be best used where metal spattering could cause loss of glove integrity, as when a pressure vessel ruptures in the thermite type reduction-to-metal facility. Such a protective plate is not in current use at Rocky Flats.

Another type of lead overlayed stainless steel plate is mounted on a spring loaded pivot outside the glovebox (Figure 4). It contains a neoprene O-ring cemented into a groove which forms a seal with the drybox glove. After being swung into place, it is drawn toward the port with two spring clamps. However, a good seal is not formed until a vacuum is drawn in the area between the plate and the glove. Argon can then be introduced into this area. This plate is used at Rocky Flats to prevent permeation of air through gloves in a plutonium hydriding facility. It is not used elsewhere specifically for fire protection.

A plug is available which can be placed in the port from either the inside or the outside of the glovebox. It forms an effective seal of the port (Figure 5). A major fire will not breach this plug. A few plugs of this type are currently being used at Rocky Flats in vacuum or inert systems in order to prevent undesirable diffusion into the glovebox. They are not being used for the purpose of fire protection.

Swinging lead doors are sometimes used as a radiation shield for ports when the gloves are not in use (Figure 6). Since these doors do not form a seal, they would not prevent a glove breach during a fire.

When a glove port is not going to be used for an extended period of time, a steel plate is bolted into place (Figure 7). The plate is overlayed with lead for gamma protection, and neoprene sheeting is bonded to the steel so that an air tight seal is formed.

In considering employment of any of these devices, one must weigh the risk of fire damage relative to the hazard present in that glovebox.

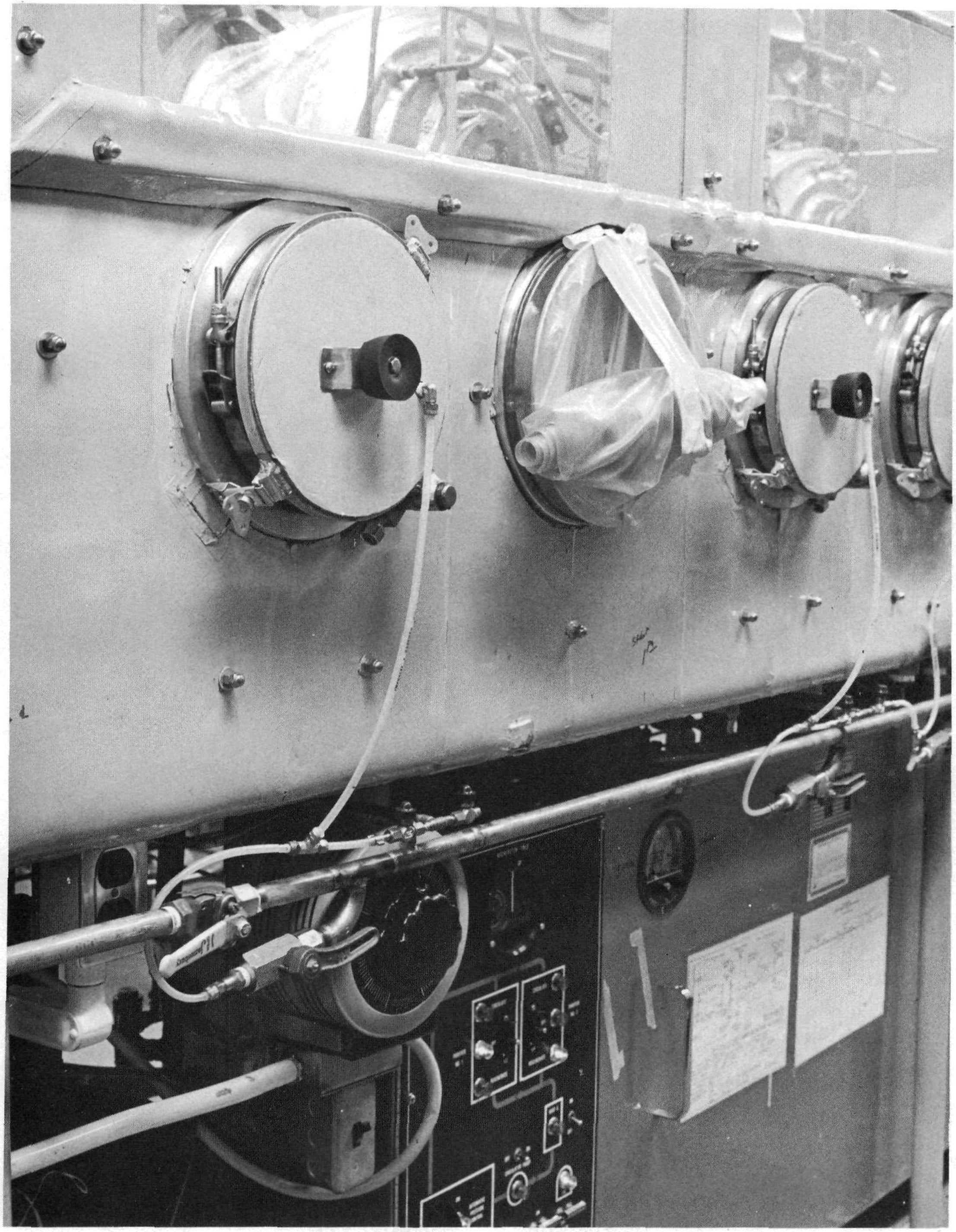

FIGURE 4

GLOVE PORT SEAL

FIGURE 5

PLUG-TYPE PORT SEAL USED INSIDE
OR OUTSIDE OF BOX--NO FIRE BREACH

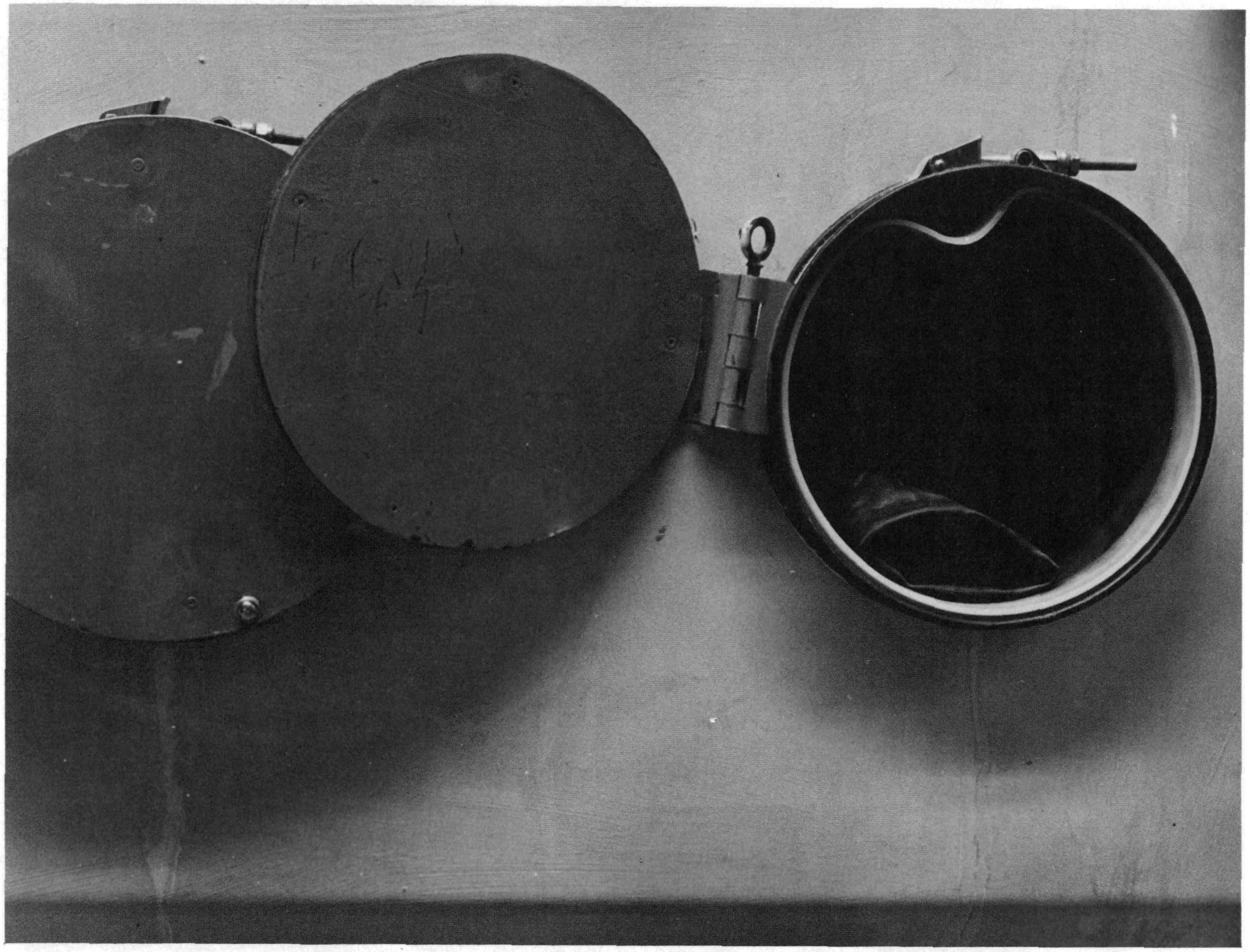

FIGURE 6

LEAD DOORS FOR RADIATION SHIELDING

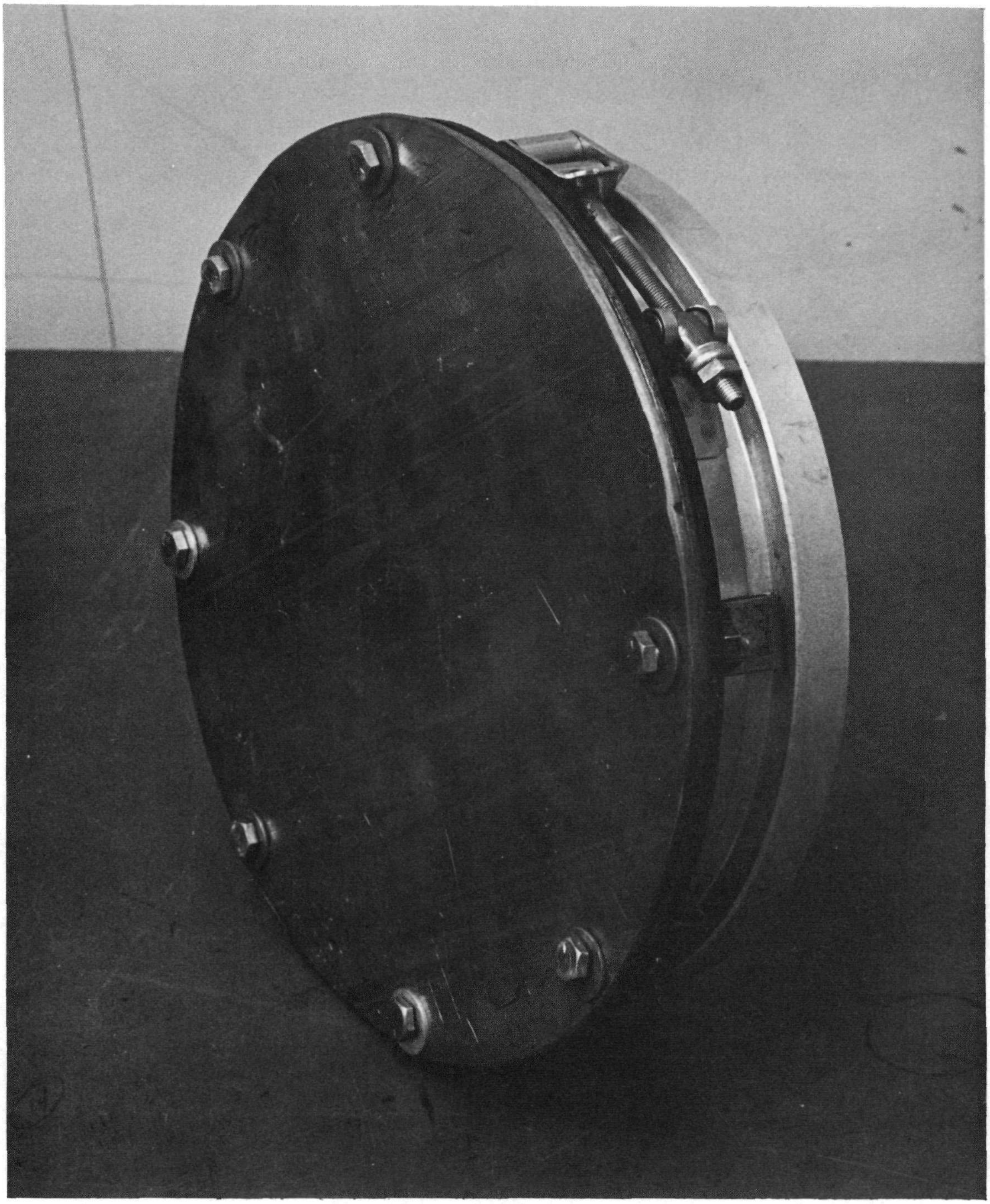

FIGURE 7

LEAD-OVERLAYED STEEL PLATE
BOLTED OVER PORTS NOT IN USE

DESCRIPTION OF ARHCO'S NEW COMPACT PLUTONIUM STORAGE FACILITY

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April 13, 1971

DESCRIPTION OF ARHCO'S NEW COMPACT PLUTONIUM STORAGE FACILITY

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ABSTRACT

A new plutonium storage facility has been constructed at Richland which uses a shielded cubicle arrangement for reducing radiation dose rates. Reduction in radiation exposure by a factor of ten is expected in the vaults from that experienced in plutonium storage vaults previously used. A products-of-combustion fire detection system throughout the facility and a sprinkler system in the scrap storage area provides continuous fire protection. A preliminary emphasis is placed on fuel reduction with all concrete construction. Compartmentation within cubicles reduces the amount of plutonium exposed to combustion if a fire should occur. A storage density of 2.1 spots per square foot is realized for the overall facility. The engineered construction of interconnecting precast concrete panels to form the basic structure and the shielded storage cubicles provides increased safety at decreased unit cost.

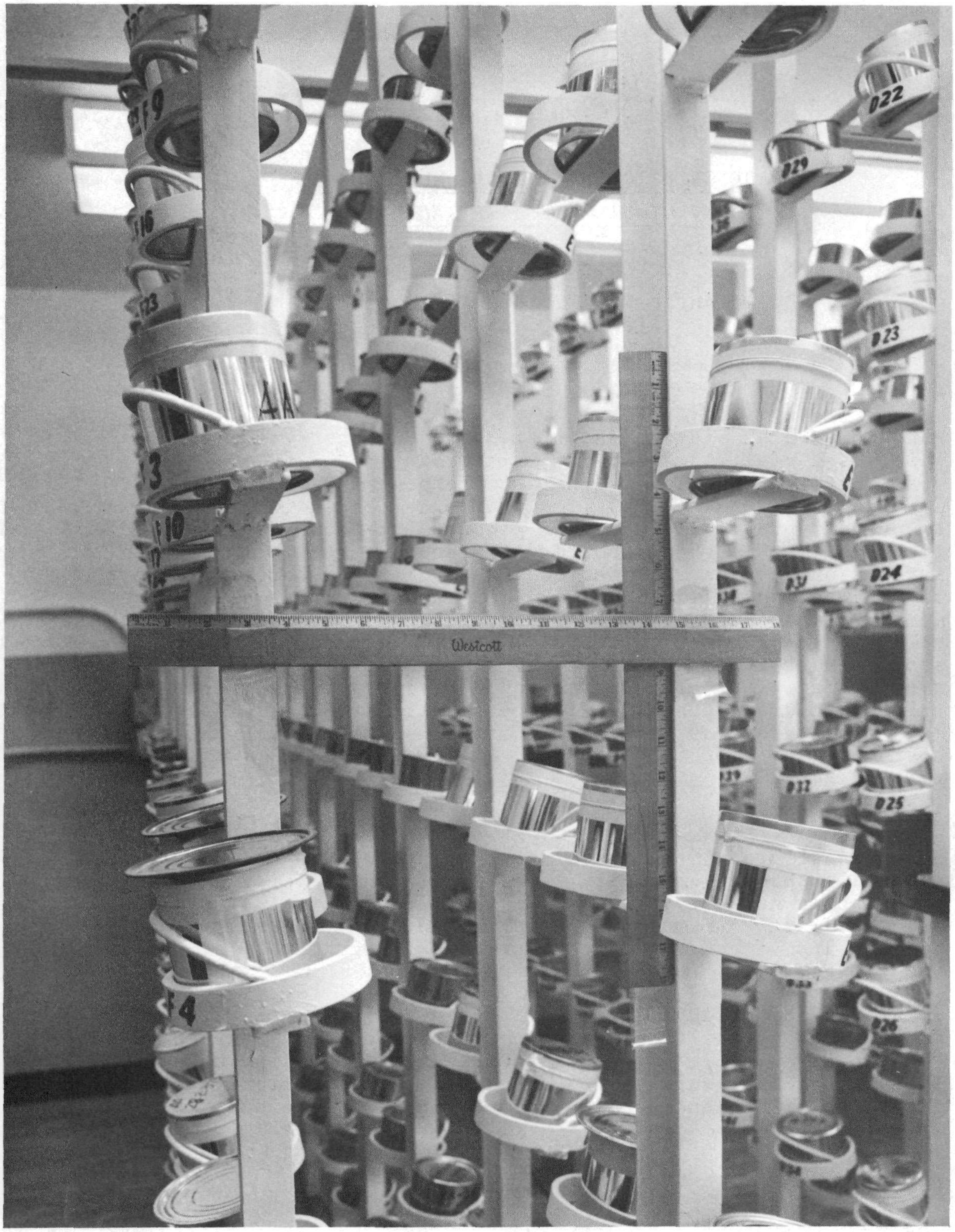
INTRODUCTION

Traditional practices in storage of plutonium have commonly used pedestals spaced vertically on poles within an isolated vault. These poles normally extend from the floor to the ceiling for stability with sufficient spacing between the poles to avoid any possibility of a critical concentration. Such an array may be seen in Figure 1.

Relatively high radiation dose rates are experienced with a plutonium storage vault of this type. This fact plus the need for additional plutonium storage space led to the design of a new storage facility during 1969 and 1970. A new plutonium storage facility at Richland has since been constructed and is now in operation.

DESCRIPTION

The 2736-Z Building, as accepted by Operating personnel in March 1971, represents a significant forward advance in improved storage



OPEN PEDESTAL PLUTONIUM STORAGE VAULT

FIGURE 1

of plutonium metal and oxides. Three criteria have governed design and construction of this plutonium storage facility in achieving this improvement. They are:

1. Reduction of radiation exposure to personnel,

2. A reduced amount of plutonium exposed to a fire in the event that one occurs, and

3. Minimum probability of contamination spread in the event of a spill or container rupture.

As a prelude to discussing the means by which these criteria were met, let us first take a look at the building.

Figure 2 shows the exterior of the 2736-Z Building. This storage facility is adjacent to the major plutonium processing building at Richland, on which it is dependent for ventilation supply and exhaust. The interconnecting ventilation ducts may be seen in the background.

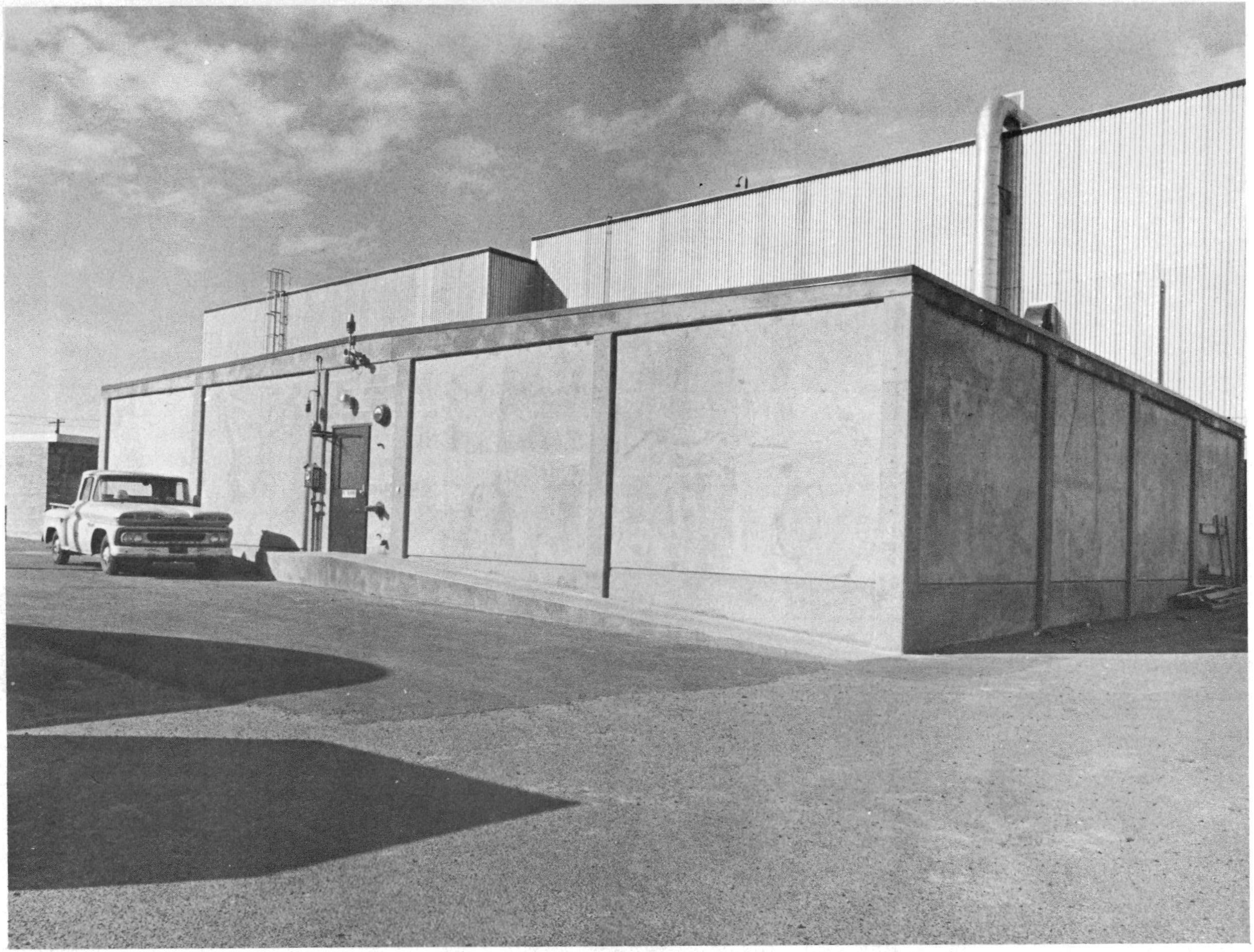
Figure 3 shows a plan view of the building. One enters it through airlocks at either end of a central corridor. In addition to stabilizing an internal negative air differential of 0.2 inch of water, these airlocks are also used to house electrical controls, fire protection controls, and a breathing air manifold for use in the event contamination should ever escape from the sealed plutonium containers. The fire protection equipment consists of a products-of-combustion or ionization type fire detection system and a wet pipe fire sprinkler system for the scrap storage vaults.

The doors from the central corridor each lead to one of the four storage vaults. Two of the vaults are open-bays as shown in Figure 4. Although presently reserved for storing scrap, provision has been made should the need arise, for adding cubicles as seen in Figure 5 for storing plutonium metal and oxide.

This provision was made while pouring the floor slab by anchoring parallel steel angles in the concrete with one web flush with the floor. Mating angles similarly anchored in the base of each precast cubicle panel are then aligned and welded together during erection of the cubicle. After erection, the cubicles are further stabilized by steel angles welded along the top edges of the cubicle panels. Two shielding doors, one bolted to the front of the cubicle panel and one hinged to it, completes the cubicle concrete construction.

Figure 5 shows the general arrangement of these cubicles within one of the two metal and oxide vaults. The view is of an aisle along either end of the vault showing the rows of cubicles back-to-back. In the background is a typical cubicle as seen from the front. The vertical eight inch wide slotted opening in the cubicle face permits placing a container of plutonium metal or oxide on pedestals that are located inside the recessed area of the cubicle. This opening may be enlarged by swinging open the hinged shielding door.

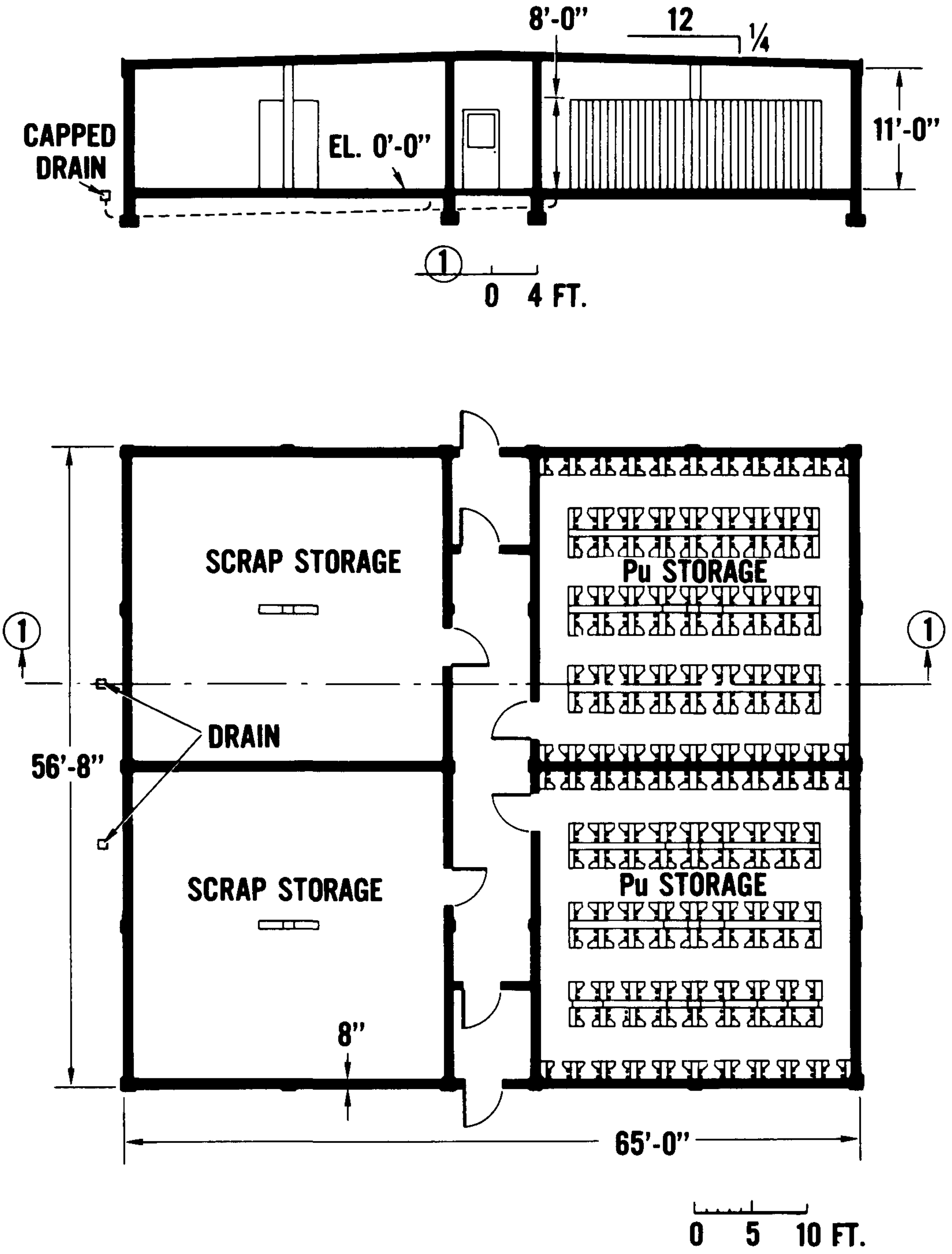
Nine variations of precast concrete panels along with precast roof slabs are combined to form the basic building structure and the storage cubicles. This permits appreciable increases in safety at decreased unit construction cost.



2736-Z BUILDING PLUTONIUM STORAGE FACILITY

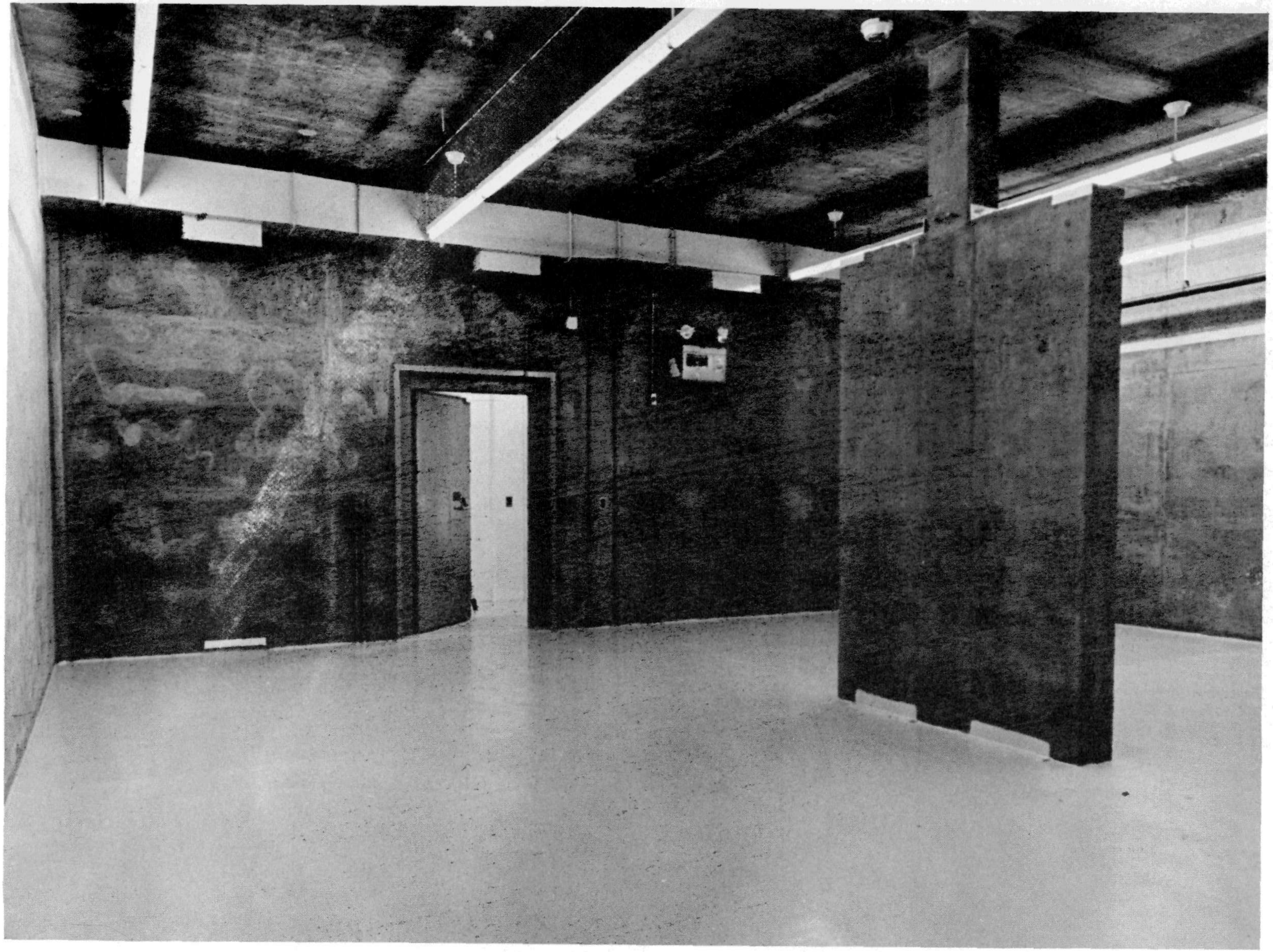
FIGURE 2

ARCHITECTURAL PLAN OF 2736-Z BUILDING



FLOOR PLAN

FIGURE 3



PLUTONIUM STORAGE VAULT WITHOUT CUBICLES

FIGURE 4

PLUTONIUM STORAGE CUBICLES IN VAULT

FIGURE 5

This major departure from previous vault design was motivated by need for substantial decrease in the radiation exposure receivable by personnel on entering the storage area. Exposures inside a vault of 150 to 200 mrem/hr were experienced as quantity of material stored in the vaults increased. In one case of a dose rate of 170 mrem/hr inside the vault, 130 mrem/hr was observed to be from neutrons and 40 mR/hr was from gamma radiation. Subsequent attempts to increase the loading capacity of vaults, coupled with storage of higher exposure plutonium, resulted in internal vault dose rates of approximately 500 mrem/hr. Distribution in one case was 400 mrem/hr from neutron radiation and 110 mR/hr from gamma radiation.

A number of concepts were considered for construction of future plutonium storage vaults which would greatly reduce internal dose rates. One concept involved rows of filing cabinets with critically safe internal spacing of storage positions. The remaining space within the cabinet drawers were to be filled with appropriate shielding materials. Still another concept considered was an array of holes located within a concrete-earth floor. Into each hole a vertical array of stored containers could be lowered for storage. The array from any one hole could be raised above floor level as desired. In either of these and similar concepts, the radiation would be limited to that received from the one storage component being used at the moment. Each of these approaches, although capable of adequate radiation reduction, entailed operating disadvantages which resulted in their rejection.

The layout sketch as originally conceived for the adopted design concept, consisted of an array of concrete blocks so stacked as to form cubicles which separated a group of four pedestal posts from all other similar groups of four. Each cubicle was accessible to a corridor by an eight inch wide vertical opening. This original array was ultimately modified to the design shown in Figure 6. This particular design was predicted to reduce in-vault dose rates to one-tenth or less that of earlier vault design. Although the vault has not, at this time, been fully loaded, dose rate measurements to date indicate that the goal of a tenfold reduction in radiation exposure within the vault will be realized.

The selection of concrete as the shielding material increases the probability of a fire being confined to a single cubicle. The eight inch concrete walls and six inch concrete roof of the building structure along with the installed fire detection and fire sprinkler systems combine with the cubicle compartmentation to achieve another of the facility goals - that of reducing quantity of plutonium exposed to a fire in the event that one occurs.

Fire prevention and fire fighting philosophy for the facility has employed two concepts. In the two scrap storage vaults and central corridor, automatic wet-pipe fire sprinkler systems are used. Here the miscellaneous nature of the material and its packaging requires allowance for significant quantities of combustible materials.

ARRANGEMENT OF STORAGE CUBICLE

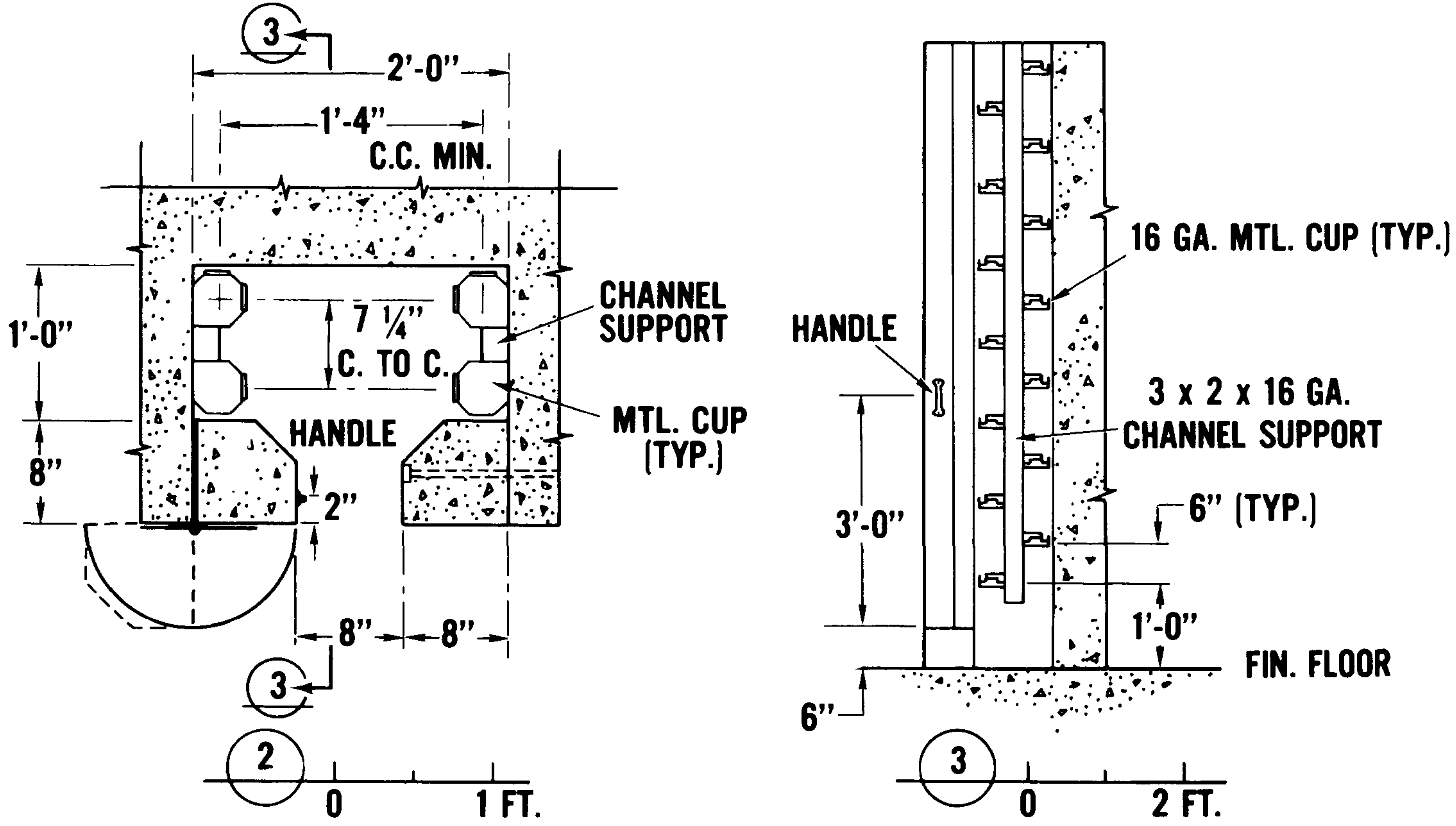


FIGURE 6

However, in the two compact vaults for storing plutonium metal and oxide, principal emphasis is placed on eliminating all fuel for a fire, other than the plutonium itself which is well compartmentized within the many separate fireproof cubicles. Here the only fuel other than plutonium is the small part of a plastic bag that is used in sealing the plutonium out of the glove boxes. Even with this, a program is underway for replacing the plastic with metallic foil. A system embodying products-of-combustion sensors located throughout the facility are used for early detection and alarming in the event of any fire.

The facility goal of minimizing probability of contamination spread in event of a fire is achieved by a heating, ventilation and air conditioning design that permits an air change every four minutes. This flow is considered adequate also to compensate for the self-heating from the plutonium stored within the cubicles. The possibility of contamination being spread by an outflow of water from the fire sprinkler system in the scrap storage vaults during a fire is minimized by retention of water up to a two inch depth within the building before running outside. A scupper as may be seen at floor level in Figure 4, permits water up to one-half inch depth to be retained in the vault before flowing into the corridor. In this way, the potential for escape of waterborne plutonium particles is greatly reduced by settling and initial retention of water.

In addition to meeting the design objectives of radiation and increased safety factors in case of fire or contamination spread, a further design consideration was retention of the compactness associated with the open pedestal vault of Figure 1. Compactness being a function of storage arrangement, it varies in the new facility depending on whether one is considering only the inside of the cubicles themselves, the central area of a vault, or the total area including service rooms and corridors. Figure 7 shows the compactness of this plutonium storage facility as expressed in storage spots per square foot of floor space and per cubic foot of the building space. The vault area value of 2.3 spots per square foot or 2.1 for the overall facility is essentially the same as that prevailing in the earlier vault storage areas where dose rates exceeded 500 mrem/hr. Faced with the alternative of greatly expanding the plutonium storage areas to reduce radiation exposure, this new design promises to permit retention of previous compactness through judicious source spacing and the economical and effective use of in-vault shielding.

In summary, the advanced design of ARHCO's new compact plutonium storage facility has brought into reality the 1969 goals of a tenfold reduction in radiation exposure, reduced fire hazard, and minimal probability of contamination spread. It has been particularly satisfying to achieve these objectives while retaining compactness and at the same time improving such fundamentals as criticality control and operating efficiency.

STORAGE SPOT DENSITIES OF 2736-Z PLUTONIUM STORAGE VAULT

Building Area	Description	Storage Spots per sq. ft. of Area	Storage Spots per cu.ft. of Space
Cubicle	Cubicles only not including aisle ways	5.09	0.463
Central Area of Vault	Cubicles back-to-back between 36 inch wide aisles with no cross aisles	2.84	0.258
Interior of Vault Including Walls	Cubicles back-to-back between 36 aisles with 32 inch cross aisles at ends of vault	2.29	0.208
Overall Building	The vault interior plus central service corridor and airlocks	2.07	0.188

FIGURE 7

SECTION II.

VENTILATION SYSTEMS

APPLICATION OF AN INERT ATMOSPHERE TO A PLUTONIUM PROCESSING SYSTEM

K. W. Calkins and C. F. Bogard

The Dow Chemical Company
Rocky Flats Division
Golden, Colorado 80401

April 13, 1971

U. S. Atomic Energy Commission Contract AT(29-1)-1106

APPLICATION OF AN INERT ATMOSPHERE
TO A PLUTONIUM PROCESSING SYSTEM

BY

K. W. Calkins, Project Manager
Manufacturing Department
and
C. F. Bogard, Project Engineer
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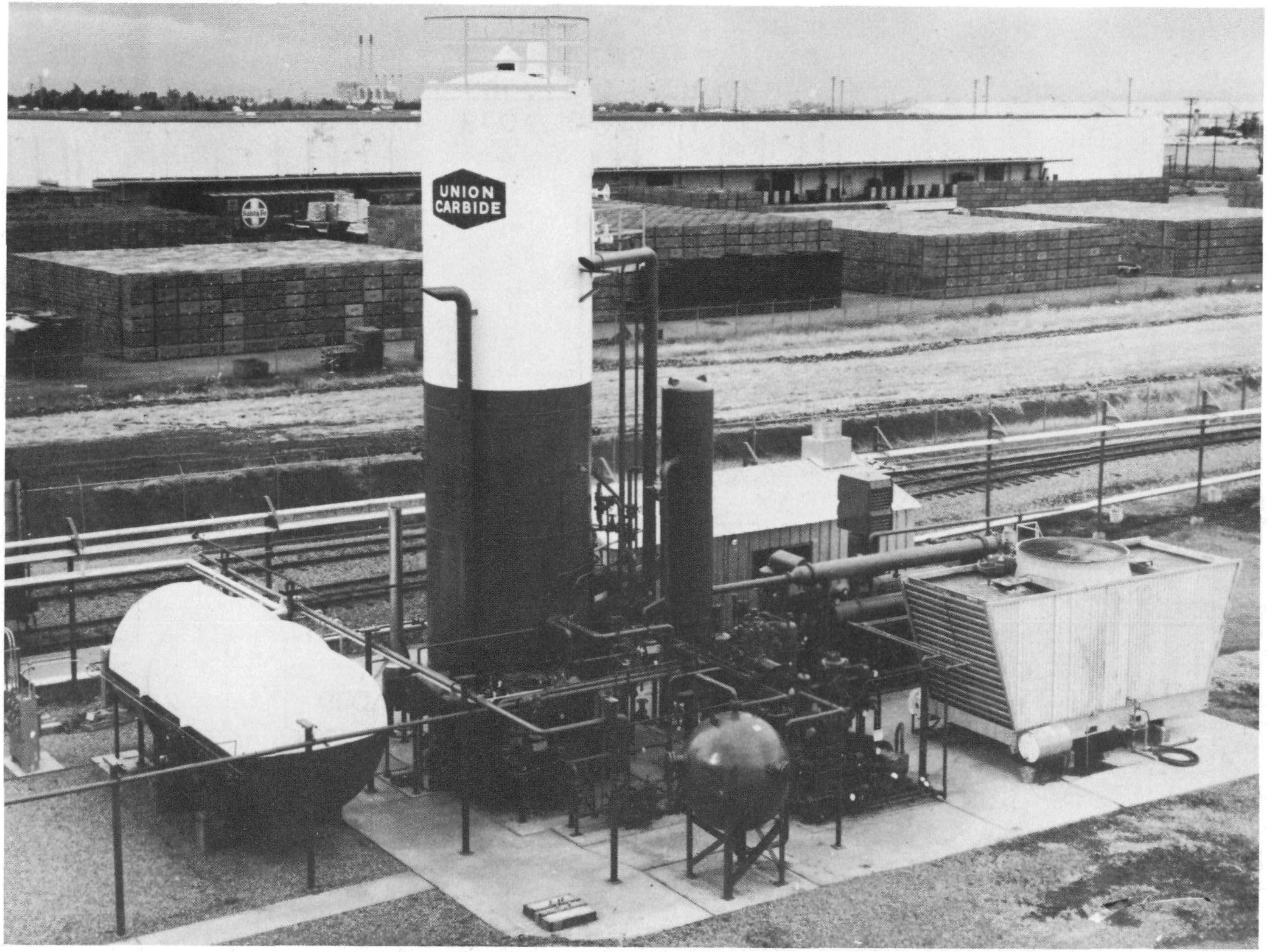
Use of an inert atmosphere for plutonium processing work was considered for Rocky Flats during initial design of the plant in 1951 and again in 1961 when most of the plutonium operations were further dehumidified. The inert atmosphere was rejected both times for the same reasons. Research at that time indicated that plutonium corroded faster in an oxygen-free atmosphere of the same humidity as an air atmosphere. Since the dry air atmosphere was also cheaper, it was selected. The minor chip fires that occurred were not considered a serious risk at that time. Procedures and materials for fire control were a part of normal operations.

In May 1969 interest in an inert atmosphere for metallic plutonium operations was renewed. A committee was established to investigate feasibility of inerting and related subjects. That committee, headed by Dr. L. A. Matheson, concluded that inerting of large production facilities at Rocky Flats was feasible, and both Dow and USAEC management have given positive backing to the project.

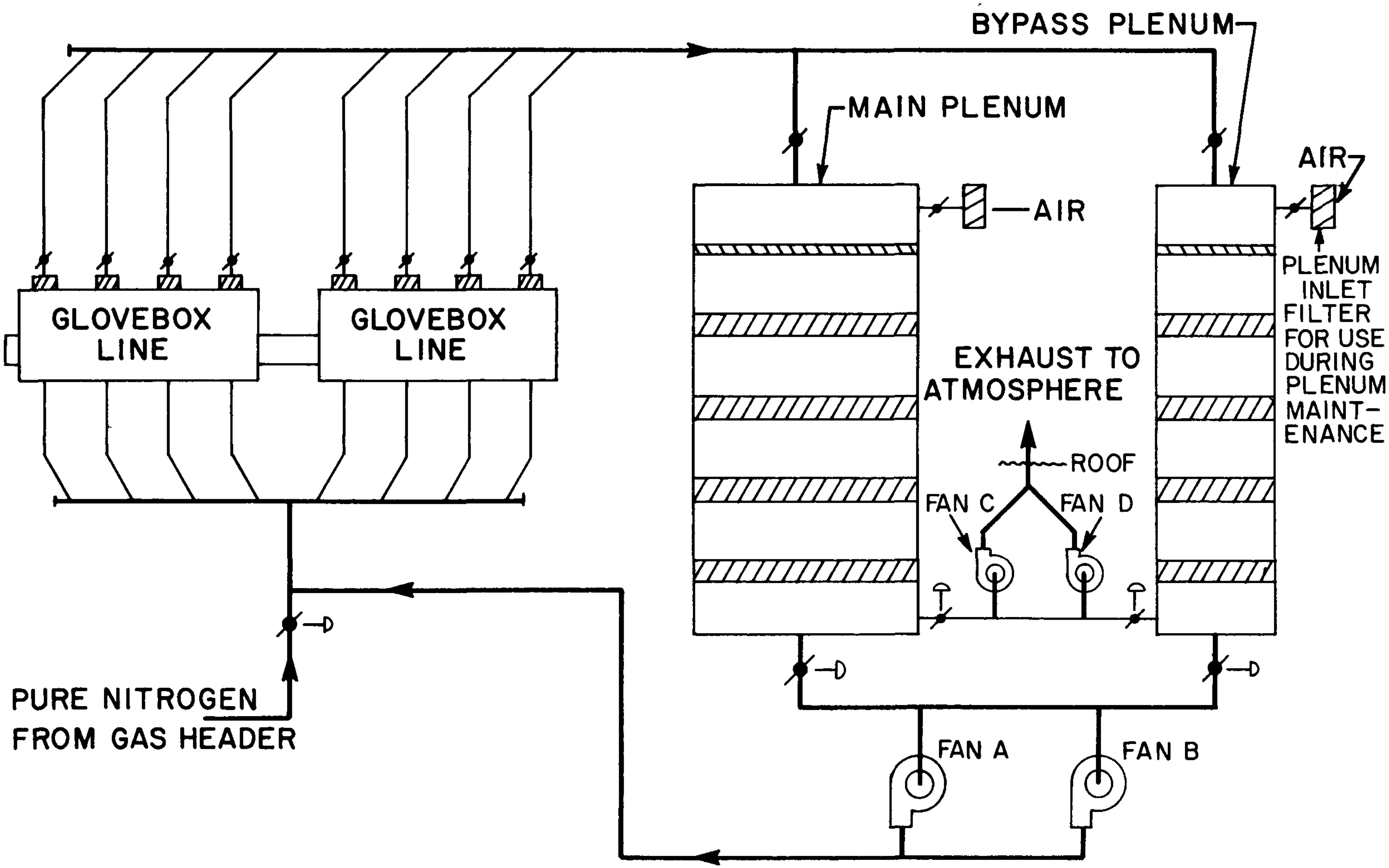
We selected nitrogen as the inerting gas due to price and availability. We plan to limit oxygen levels in the nitrogen systems to 5% by volume maximum, which will prevent plutonium chip fires, as will be discussed later by L. E. Musgrave. Incidentally, no other materials in the glovebox lines burn at less than 10% oxygen.

Our present plans during pre-Title I engineering are to use pure nitrogen from a vendor-owned and operated liquefaction-fractionation (SLIDE 1) plant such as this one by Linde. We anticipate a capacity requirement of 140,000 SCFH of gas at about 30 psig from the plant. We will locate the plant about 1,000 feet away from our main production buildings, and pipe the gas to the desired locations. Reliability of supply to our systems will be guaranteed by the vendor through use of a large liquid nitrogen backup dewar.

The glovebox systems to be inerted in each building will be converted to recirculating ventilation systems as shown here (SLIDE 2). Gas will be purged from each system to overcome in-leakage and maintain the system at a negative pressure around 1/2 inch water



SLIDE 1



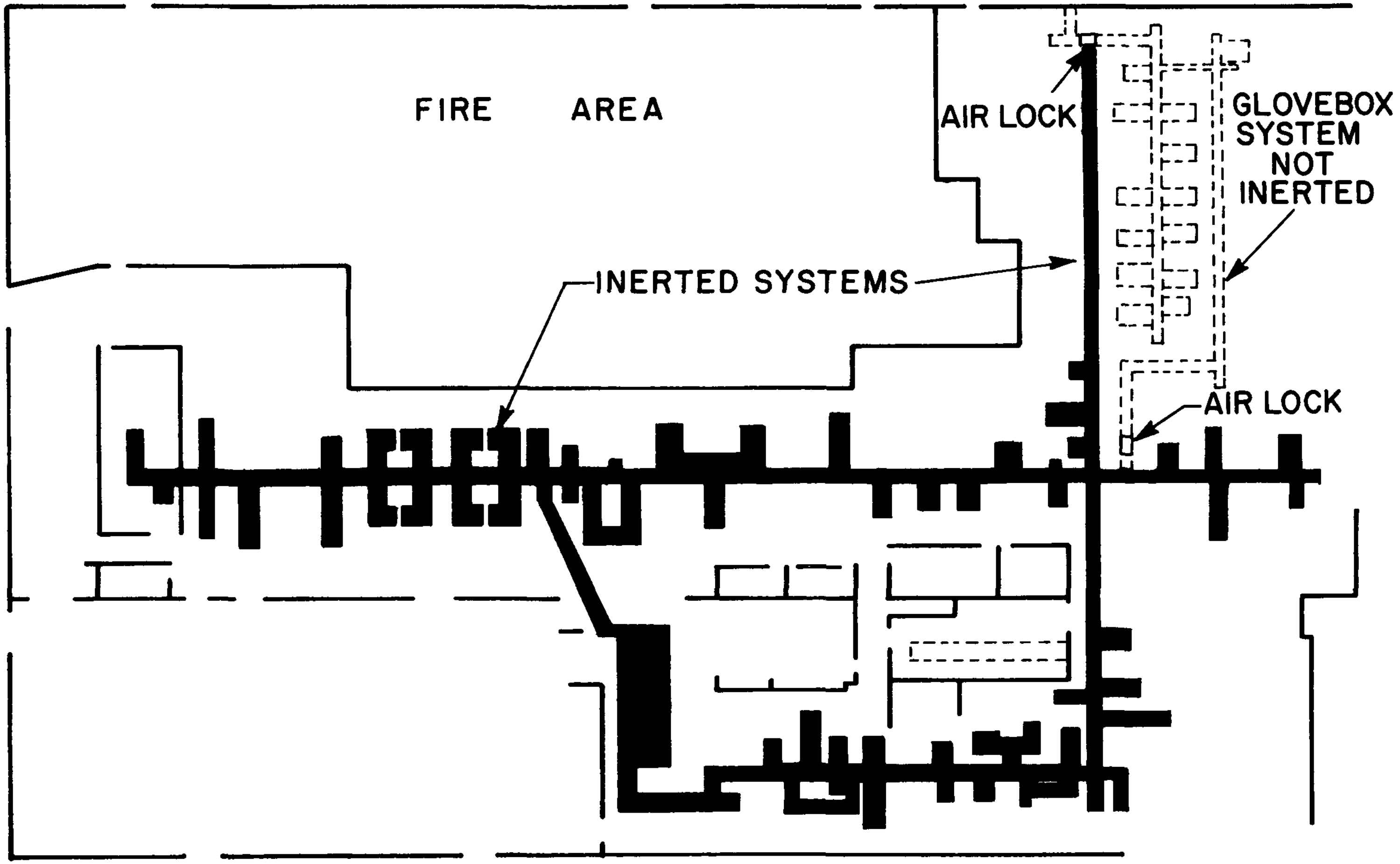
FLOW DIAGRAM
RECIRCULATING NITROGEN
SYSTEMS
SLIDE 2

column. Additional gas will be exhausted to permit control of oxygen or water levels in the system by dilution with pure nitrogen from the gas plant. Our flow rate through the recirculating system will be comparable to the present air flows in each glovebox. You're probably curious about the other plenum shown on the schematic. We will have a bypass plenum so that nitrogen flow from either of our systems can be bypassed into a spare plenum. The main plenum then would have air bled through it to provide an air atmosphere for plenum maintenance. This has several advantages in addition to providing a plenum for emergency use. First, we would not degrade our nitrogen atmosphere by the inevitable leaks through open plenum doors during maintenance; and second, we can change filters and check filtration efficiency, exactly as at present, with respirators in the third and fourth stages, and supplied air hoods in first and second stages.

I previously mentioned that inert operations were rejected in 1961 and a dried air atmosphere having 200 ppm of moisture was selected as providing better corrosion protection. Since that time, however, Rocky Flats' Research and Development group has found no apparent difference in corrosion rates between plutonium stored in our present dried air atmosphere and oxygen depleted nitrogen atmospheres having a minimum of 1% oxygen. We've also found that nitriding of the plutonium will be no problem at any of our operating temperatures, provided the 1% oxygen is maintained.

So far I've been talking about the generalities of inerting at Rocky Flats. Now I'd like to talk about our actual systems. (SLIDE 3)

Building 776 is one of our main production buildings. The existing glovebox system will be served by three separate ventilation systems. The colored systems, which have a total enclosed volume of about 30,000 cu. ft., will be inerted with nitrogen. The adjoining gloveboxes, shown dotted, will remain on a dried air system. Building 776 is typical of the other building, 707, which will also have two inert systems adjoining an existing dried air system. The total volume to be inerted in both buildings is about 90,000 cubic feet, which includes ductwork, filter plenums, and a 22,000 cubic feet storage vault in Building 707. Our main reason for having two inert systems in each building is to permit different oxygen level controls, and less nitrogen usage, in the systems. One system will control oxygen to about 4% by volume with humidity approaching that of the room. The other system will control humidity (by dilution with dry nitrogen) to present levels in these gloveboxes (i.e., less than 700 ppm of moisture) with resulting oxygen levels of about 2%. The existing ventilation systems in these buildings can be conveniently divided to provide these two systems.



EQUIPMENT LAYOUT
BLDGS. 776-777

SLIDE 3

Where the nitrogen systems connect to adjoining air systems, either the room air or glovebox air, we will use a two-door airlock with power operated doors interlocked to prevent both doors being open at the same time. We feel no need for vacuum airlock operation considering the large volume of our system, the recirculation and mixing of the atmosphere, and the oxygen levels we will have. The operating requirements for our production personnel working in these areas will not be appreciably different from the present situation.

One of our major problems in preparing for this inerting project is that no one within the AEC complex has anything quite comparable in size or system design to our proposed system. Our own experience is limited to several argon, helium, or nitrogen inert boxes, inert to the low ppm range of oxygen, and either using a small regenerator at each box or a vacuum pumpdown/purge system. The most comparable facilities are two isolated inert systems having a 3% oxygen in nitrogen atmosphere, one with 180 cu. ft. and the other 550 cu. ft. volume. Both of these are chip briquetting operations. Previously when the operation was done in air we averaged about one burning incident every day. In the 17 months of inert operation there have been no fires. The leak rate of these boxes is presently running about 0.2 CFH/cu. ft. of glovebox volume. These are new gloveboxes, probably having a lower leak rate than other boxes in the building. They do have a large bellows and a bag port which has more leakage potential than for our planned total system.

One problem our large inert system will correct for these individual inert briquetting boxes is material movement in and out of the box. The need for airlock or bagging operations at the box will be eliminated when it is absorbed into the large system.

The leakage problems we anticipate in our existing gloveboxes are of three types; (SLIDE 4) diffusion and unintentional and deliberate open leak paths. Compared to open leak paths the diffusion problem is negligible, and no changes to the system will be made solely to eliminate diffusion. The unintentional open leak paths will be a problem. We presently plan to attack this problem after the systems are converted to nitrogen by using a simple leak test instrument inside the line, the E.F.I. W-6 Wind Noise Test Set. Major leaks detected will be caulked, or the joints tightened.

The main leakage into our systems, though, is from the deliberate open leak paths. (SLIDE 5). This is a cross-section of typical glovebox leaks at Rocky Flats. Some of these items - tool drops, air locks, sample take-outs, and bag rings - present such a large source of leaks that they will be modified or, sometimes, eliminated from the existing gloveboxes. Others - window mountings, gasketing, shaft seals, etc. - will be corrected on new gloveboxes added to inert systems, but existing boxes will only be tightened or caulked. (SLIDE 6). The gloveboxes at Rocky Flats have win-

SLIDE 4

TYPES OF GLOVEBOX LEAKAGE

DIFFUSION

- Gloves
- Bags
- Windows
- Gaskets

OPEN LEAK PATH

UNINTENTIONAL

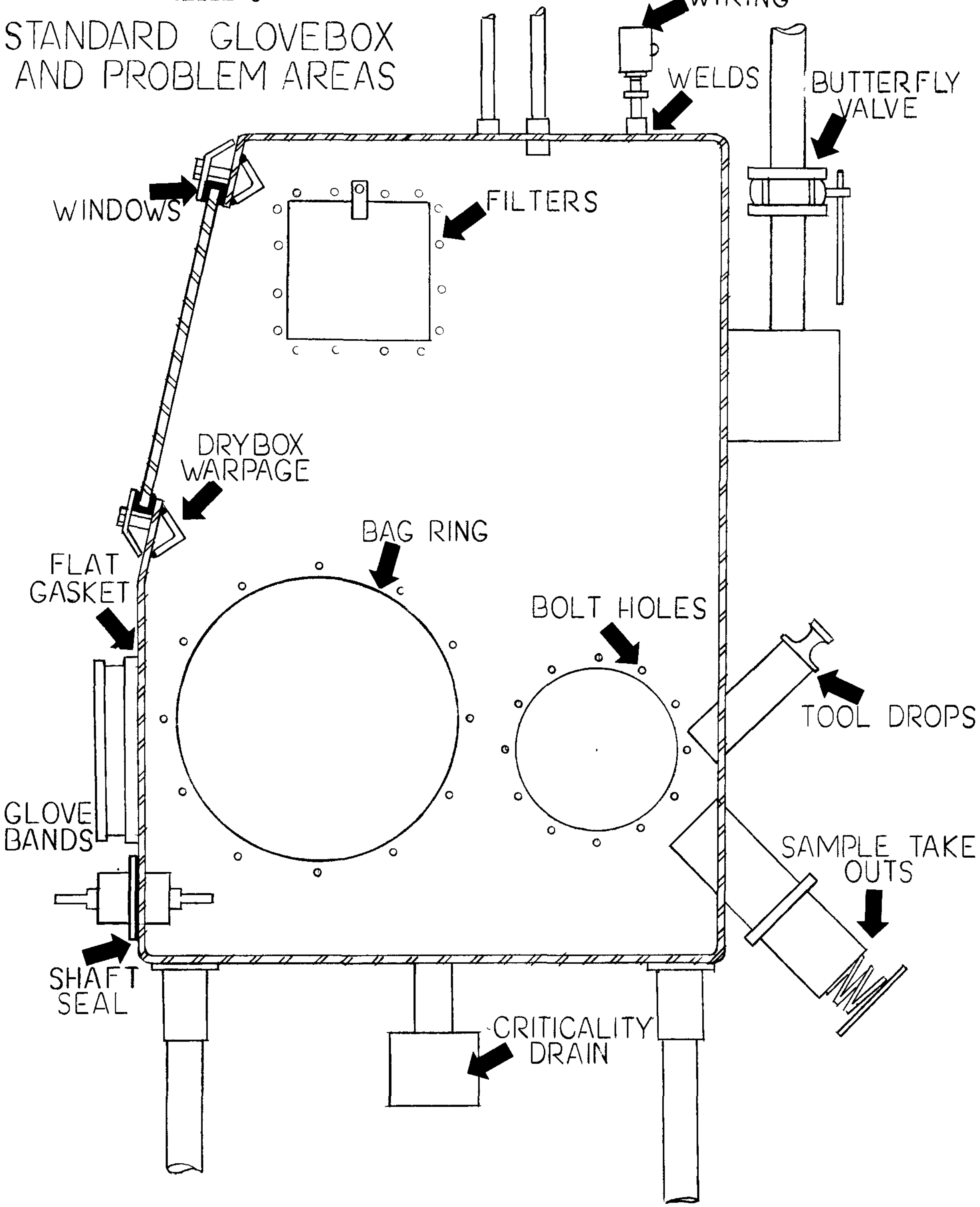
- Gloves
- Bags
- Window Gaskets
- Electrical Passthroughs
- Closed Airlocks
- Closed Tool Drop
- Rotary Shafts
- Criticality Drains
- Oil Drains

DELIBERATE

- Tool Drop
- Airlock
- Glove Change
- Bag Change

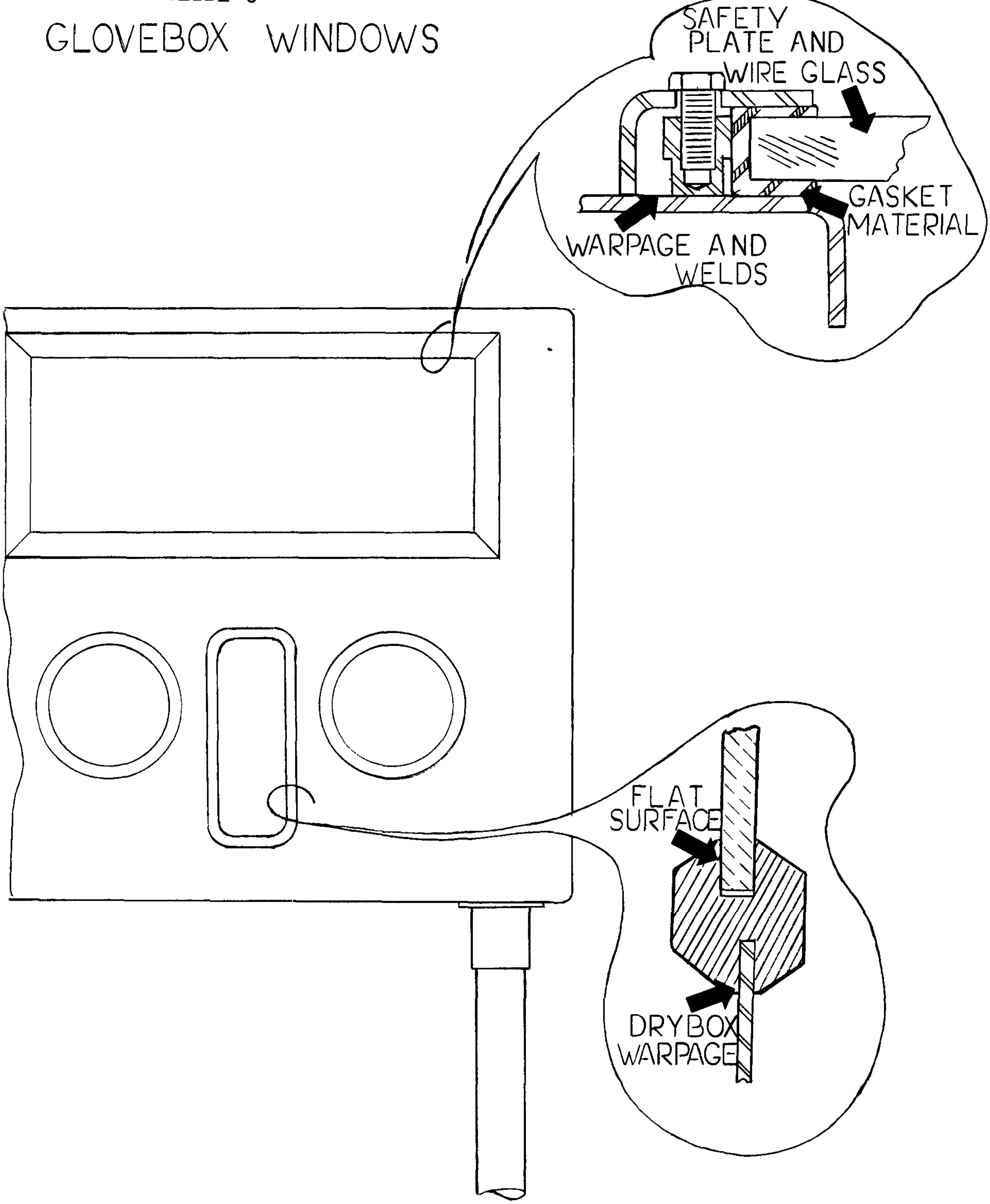
SLIDE 5

STANDARD GLOVEBOX AND PROBLEM AREAS



SLIDE 6

GLOVEBOX WINDOWS



dow seals common to other AEC sites. We anticipate no change in the design other than use of caulking compounds on the gasket surfaces at installation.

Airlocks to the atmosphere in the existing systems will be replaced with ones having (SLIDE 7) vertical lift, hydraulic powered doors as shown on this slide. The doors on each airlock will be interlocked so that both doors cannot be open at the same time.

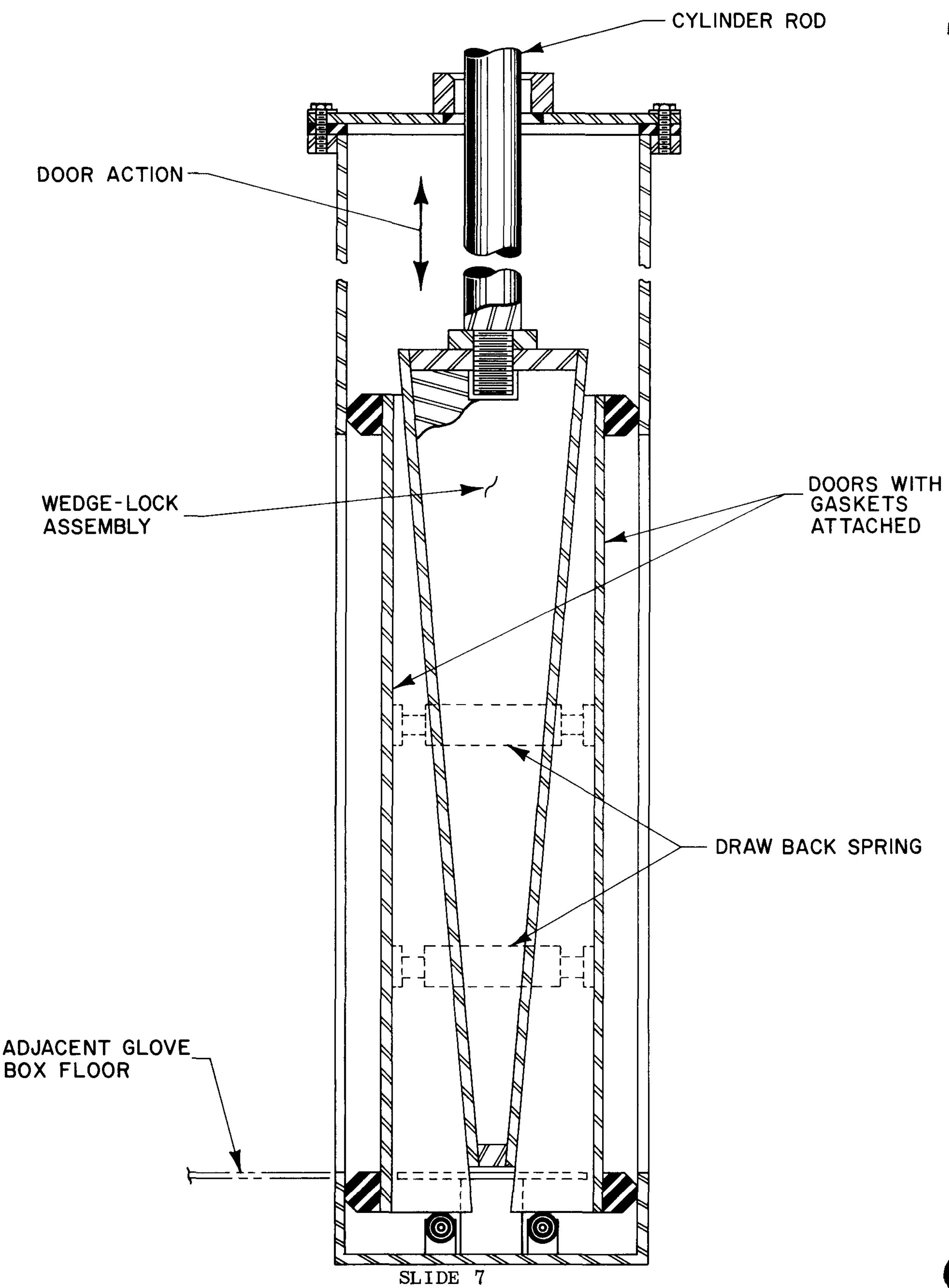
Tool drops (SLIDE 8) now flow 60 to 70 CFM of air into our system as measured at one of our normal 2-inch diameter drops. This graph does demonstrate the effect of a lower differential negative pressure in the box. If we decrease the differential by .2 inches of water, the in-flow of air is reduced by about 10 CFM. We will try to operate our inert systems at about .5 in. negative pressure. We propose to minimize this flow (SLIDE 9) by use of plugs at both ends of the drop tube making an enclosed space and a limited air volume. To hold the "drop" at a negative pressure, we will need a small bleed hole in the tube.

The inerting system will not be fool-proof. For instance, if one or two gloves were accidentally torn off somewhere in the glovebox system, the oxygen level would rise above the 5% control point. As mentioned before, other materials in the glove box system would not burn in less than a 10% oxygen level, so we have a safety factor. We cannot rely on this to provide an absolute protection against fire in the gloveboxes so we will provide fire detection and protection in all inert systems identical to that provided in air glovebox systems.

This project presently is just getting "off the ground". We are in pre-Title I engineering now. Our preliminary construction plans call for having our first system inert by mid-1972 with all construction complete by mid-1973.

I've given you a very brief survey of the inerting project and have probably left some items of specific interest to you unanswered. Are there any questions?

-FINIS-



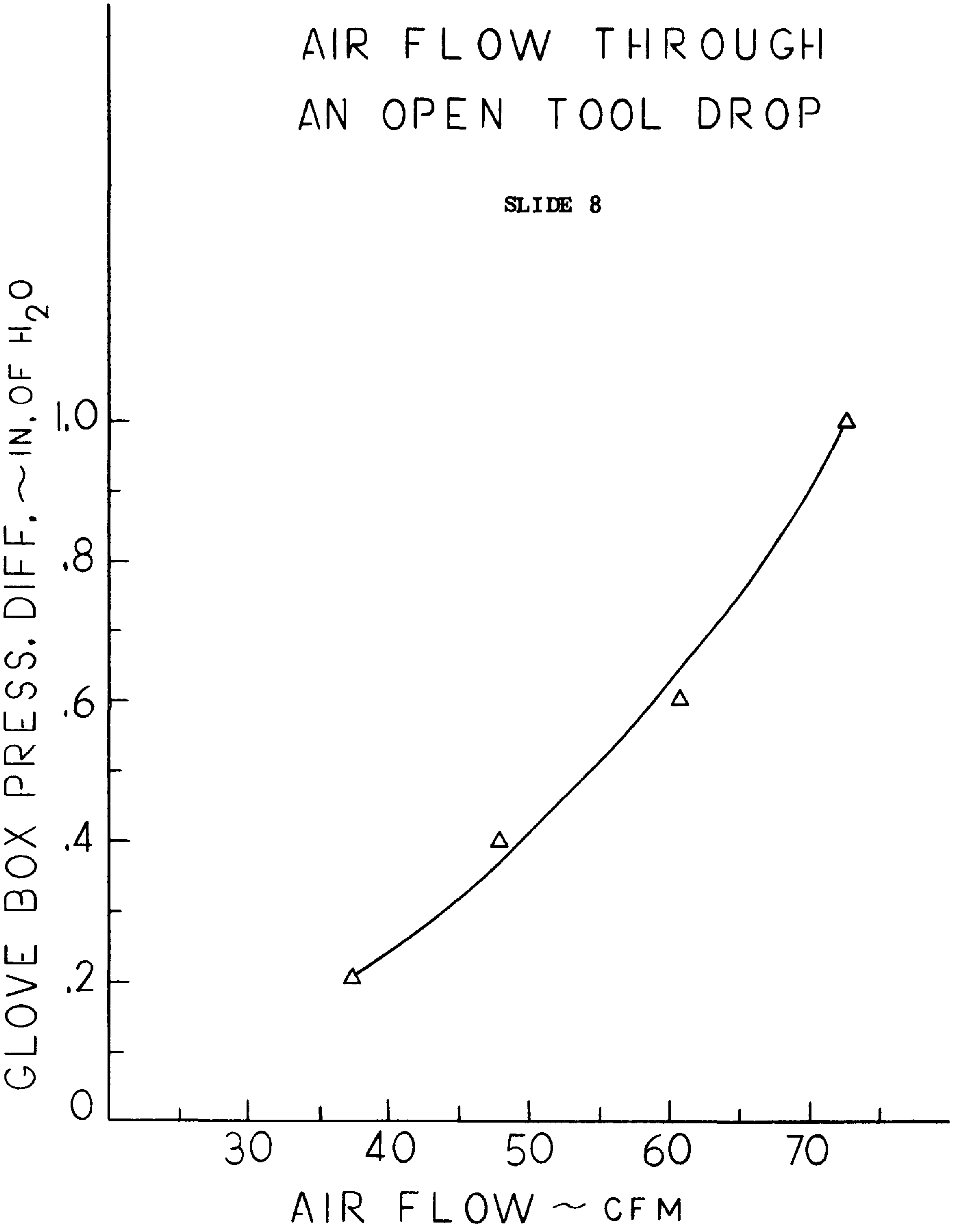
SLIDE 7

WEDGE ACTION AIR LOCK DOOR

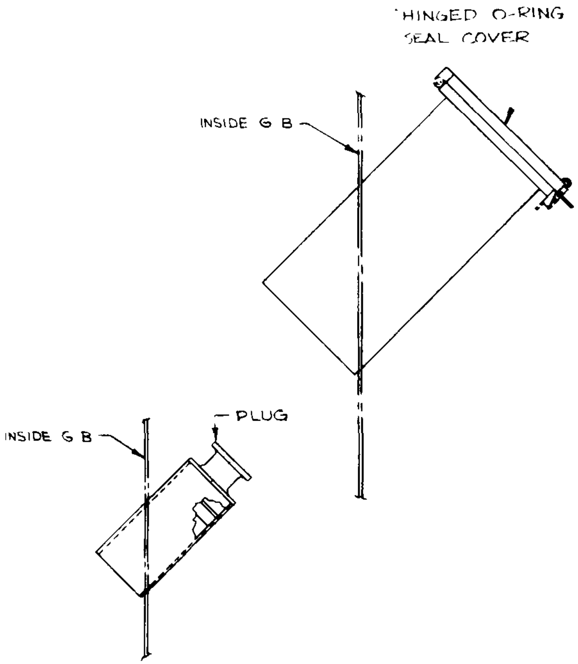
WEDGE FORCES GASKETS AGAINST FRAME FOR SEAL

AIR FLOW THROUGH AN OPEN TOOL DROP

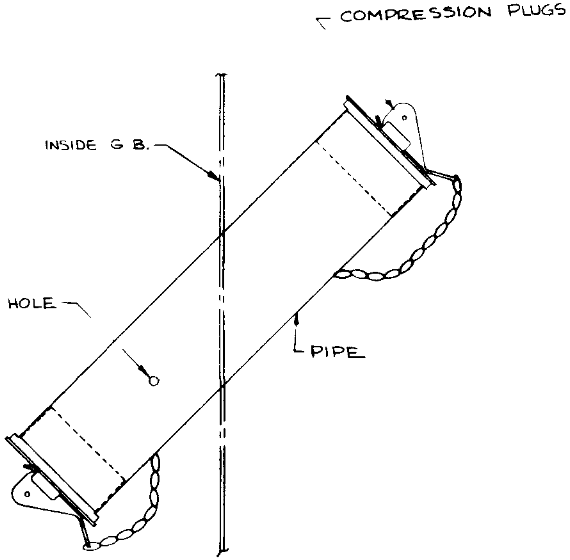
SLIDE 8



TOOL DROPS



EXISTING



PROPOSED

SLIDE 9

AIR LEAKAGE INTO PLUTONIUM GLOVEBOXES AND ITS EFFECT ON INERT OPERATIONS

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April 13, 1971

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AIR LEAKAGE INTO PLUTONIUM GLOVEBOXES AND ITS EFFECT ON INERT OPERATIONS

INTRODUCTION

The Rocky Flats Plant plans to substitute an inert gas, nitrogen, for the dried air now used in many of the present plutonium gloveboxes at Rocky Flats (1). The reason for this is to suppress the ignition and burning of plutonium metal and organic materials inside the gloveboxes. Oxygen concentrations below 5% in nitrogen do not support combustion of plutonium(2), and oxygen concentrations below 10% do not support combustion of hydrocarbons, paper, wood, and most plastics(3). It is planned to maintain the oxygen content in nitrogen below 5% in routine operation of the gloveboxes.

The nitrogen supply would be several times more expensive per unit volume than the dry air supply currently used. Several characteristics of the glovebox system must be understood before an economic analysis of nitrogen usage can be made and the nitrogen supply system designed. The most significant characteristic of the system that will affect nitrogen usage is the amount of air leakage into the gloveboxes. Air leakage into gloveboxes will increase both the oxygen concentration and water vapor concentration in the gloveboxes, both of which must be controlled.

To develop quantitative information on air leakage, a material balance method has been developed to relate air leakage through openings into gloveboxes to resultant increases in water vapor or oxygen concentration. This allows estimation of the leakage into gloveboxes and also the probable nitrogen supply requirements. Using water vapor concentration measurements, this material balance method has been applied to a large glovebox system at Rocky Flats. This paper describes the analysis of the nitrogen requirements using the water vapor material balance method to determine leakage, and other considerations that arise during conversion of a glovebox system to an inert atmosphere.

DISCUSSION

Where highly radioactive material such as plutonium is handled in gloveboxes, a slight negative pressure of about 1/2-inch of water is maintained in gloveboxes relative to room or box external to prevent radioactive material being carried into the room. Under these conditions, any hole in the gloves, windows, window seals, gaskets, etc., allows passage of room air into the gloveboxes with its 21% oxygen and room moisture content. This is called open path leakage. Oxygen and water vapor also diffuse through the solid rubber or plastic of the gloves, windows, etc., but this can be shown to be negligible compared to the open path leakage in our gloveboxes.

DEVELOPMENT OF MATERIAL BALANCE EQUATIONS

The relationship between open path air leakage, atmosphere flow and moisture or oxygen gain may be deduced from a material balance around a glovebox or glovebox system such as shown in Figure 1. Equating the total quantity of the species entering the system to that leaving gives

$$IC_I + LC_R = (I + L)\ C_o$$

Rearranging, the equation becomes

$$L\ (C_R - C_o) = I\ (C_o - C_I)$$

giving a leakage rate

$$L = \frac{C_o - C_I}{C_R - C_o} \times I$$

Expressed in words, the equation for a water vapor balance is

$$\text{Leakage CFM} = \frac{\text{Outlet } H_2O \text{ conc} - \text{Inlet } H_2O \text{ conc}}{\text{Room } H_2O \text{ conc} - \text{Outlet } H_2O \text{ conc}} \times \text{Inlet CFM} \quad (1)$$

A similar equation may be written for an oxygen balance,

$$\text{Leakage CFM} = \frac{\text{Outlet } O_2\% - \text{Inlet } O_2\%}{\text{Room } O_2\% - \text{Outlet } O_2\%} \times \text{Inlet CFM} \quad (2)$$

The above may be applied to the whole or to a part of the recycle circuit. When it is applied to the whole of the recycle circuit being fed pure nitrogen the equation then becomes

Leakage CFM = Inlet CFM X Outlet $O_2\%/(21\% - \text{Outlet } O_2\%)$, (3)
because Inlet O_2 concentration is essentially zero.

NITROGEN SUPPLY REQUIREMENT AT VARIOUS OXYGEN CONCENTRATIONS

The amount of nitrogen supply required will increase as the oxygen concentration to be maintained in the gloveboxes decreases. The oxygen balance formula allows calculation of the ratio of nitrogen supply to leakage into gloveboxes at various oxygen concentrations in the gloveboxes. This is shown in Table I.

The data in Table I show, for instance, that 3.2 volumes of nitrogen are required to dilute 1 volume of air at 21% oxygen, to 5% oxygen. A glovebox system operating at 5% oxygen can have its leakage trebled and the oxygen concentration will only increase to 10%, at which concentration plastics, paper and hydrocarbons still do not burn. This is an extra safety factor in preventing glovebox fires. Table I shows that as oxygen

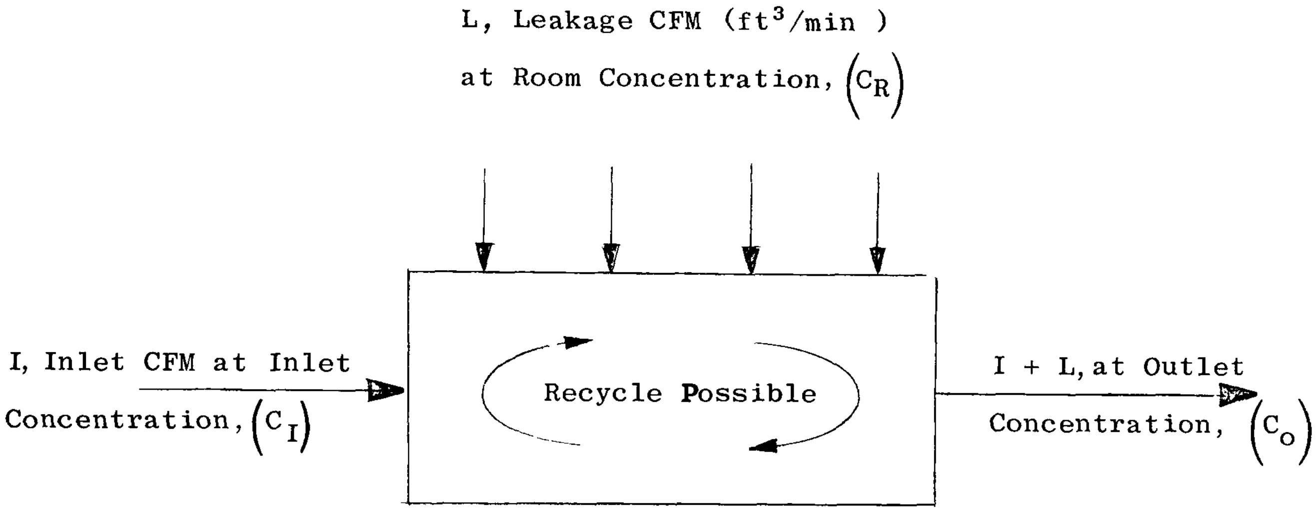

FIGURE 1

OPEN PATH LEAKAGE EFFECT ON GLOVEBOX ATMOSPHERE

TABLE I

EFFECT OF GLOVEBOX OXYGEN CONCENTRATION ON NITROGEN SUPPLY REQUIREMENT

Oxygen Concentration in Gloveboxes (Volume Percent)	Ratio of Nitrogen Supply Requirement to Leakage Into Gloveboxes
10	1.1
5	3.2
4	4.25
3	6.0
2	9.5
1	20.0

concentration decreases, leakage becomes more important in determining nitrogen usage.

LEAKAGE MEASUREMENTS ON EXISTING FACILITIES

Few inerted boxes were available to check leakage by the oxygen balance formula, so the water vapor balance formula was used to determine present leakage rate of glovebox systems at Rocky Flats. Leakage rate can be expressed as a term called "Leakage Factor." The leakage factor (LF) is the rate of leakage into a glovebox divided by the volume of the glovebox.

$$\text{Leakage Factor} = \frac{\text{Leakage } (ft^3/hr)}{\text{Glovebox Volume } (ft^3)} \left[=\right] hr^{-1}$$

Leakage data for the plutonium fabrication glovebox dry air system prior to May 11, 1969 are shown in Table II. Leakage factors for recent operation of the same system are shown in Table III. Little effort had been expended to decrease the leakage factor at the time these values were determined.

OTHER CASES

Several other cases from Rocky Flats and other sites are considered in Table IV. Most cases are considered on the basis of 100,000 ft^3 of glovebox volume, near to the Rocky Flats present estimated glovebox volume to be inerted. Hanford has reported a leakage factor of 0.050 hr^{-1} on the Building 234-5 nitrogen inerted line. Their leakage factor was applied to the projected inert system at Rocky Flats, for comparison purposes.

The two cases of 70,000 and 140,000 SCFH nitrogen supply are given because these are standard size liquid air separation plants. Applying these nitrogen supplies to the 100,000 ft^3 of glovebox anticipated to be inerted gives the leakage factors which must be attained to satisfactorily operate with each plant size. These are 0.29 hr^{-1} for the 70,000 SCFH plant and 0.58 hr^{-1} for the 140,000 SCFH plant. However, a small nitrogen supply increases the hazard from system breach or upset and increases the time required to recover from it.

It should be mentioned that even the large nitrogen plant, 140,000 SCFH, cannot withstand a very large leakage hole total area. The allowable leakage rate of 56,000 ft^3/hr can pass through an area of about 0.5 ft^2 under maintained pressure differential of 0.5 inches of water.

The last row shows data on an individual glovebox made ultra tight to manufacture PuN via the hydride process in ORNL. A flow of 0.3 ft^3/min of nitrogen with 10 - 15 ppm oxygen is fed into a 30 ft^3 glovebox with a gain of about 5 ppm in oxygen while traversing the box. Covers are provided for gloveports and take-out bags. Accurate pressure control and tightening of the system allow a leakage factor of 1.4×10^{-5} to be attained. This is about 10^{-4} times less than what is anticipated for the new Rocky Flats system. The calculations were made using oxygen data. The figures illustrate how oxygen-tight an individual glovebox may really be made. Water data was not taken. It would have undoubtedly shown much higher values of leakage because of

TABLE II

PLUTONIUM FABRICATION
GLOVEBOX DRY AIR SYSTEM LEAKAGE FACTORS
(1969 Values)

Date	Leakage Factor* (hr^{-1}) Low	High
5-1-69	0.61	4.02
5-2-69	0.23	1.29
5-3-69	0.36	1.94
5-4-69	0.36	0.50
5-5-69	0.32	0.73
5-6-69	0.41	0.83
5-7-69	0.48	0.72
5-8-69	0.33	0.50
Average	0.39	1.32
Overall Average	0.85	

* Based on system volume of 45,000 ft^3

TABLE III

PLUTONIUM FABRICATION

GLOVEBOX DRY AIR SYSTEM AVERAGE LEAKAGE FACTORS

Time Period		Leakage Factor* (hr^{-1})
October	1970	0.65
November	1970	0.79
December	1970	1.13
January	1971	0.78
October 15, through January 31,	1971	0.86

*Based on system volume of 47,000 ft^3

TABLE IV

LEAKAGE INTO GLOVEBOXES AND N_2 REQUIREMENTS TO MAINTAIN 5% O_2 IN GLOVEBOXES

Case	Leakage Factor (ft^3/hr ft^3)	Leakage Rate (ft^3/hr)	Atmosphere Changes/hr	Recovery Time, (hr to 90% avg)	N_2 Requirement SCFH	Calculated CCl_4 Concentration (ppm by volume)
Rocky Flats Avg 10-15-70 to 1-31-71, 47,000 ft^3	0.86	40,400	21	0.14	300,000 130,000	9 28
Hanford Rate on Rocky Flats Boxes, 100,000 ft^3	0.05	5,000	0.21	14.3	12,000	2040 5840
70,000 SCFH N_2, 100,000 ft^3	0.29	29,000	1.2	2.5	70,000	350 1000
140,000 SCFH N_2, 100,000 ft^3	0.58	58,000	2.4	1.0	150,000	176 500
Inerted Chip Box in Pilot Line, 180 ft^3	0.11 to 0.26	20 to 47	0.45	6.7	120@3% to 200@4%	
PuN Glovebox ORNL, 30 ft^3	1.4×10^{-5}	4.3×10^{-4}	0.8	3.7	18	

some 500 times higher permeability of elastomers and plastics for water than oxygen.

Explanation of the methods used to obtain the values in the last four columns of Table IV follow:

Atmosphere changes per hour:

This is calculated from the total system oxygen balance using an outlet oxygen concentration of 5%.

Outlet volume = nitrogen usage + leakage @ 5% O_2 = 4.2 X leakage. So atmosphere changes per hour = 4.2 X leakage factor to maintain an oxygen atmosphere of 5%.

Time to 90% recovery:

This is the time required for the system to recover 90% toward the equilibrium concentration if the system is disturbed, e.g., by a large air leak of short duration. This was taken as the time for passage of three glovebox volumes from the system outlet. A recirculation system approaches a perfectly mixed system which requires about three volumes. A once-through system might take less than two volumes.

Nitrogen requirement:

These are given or calculated from the total system oxygen balance and converted to standard cubic feet. One SCF (standard ft^3) = 1.34 CF (ft^3) at the Rocky Flats glovebox temperature and pressure.

Carbon tetrachloride concentration in boxes:

The two figures in Table IV for each situation were calculated from two different estimates of CCl_4 evaporation into gloveboxes. These were from 1968 data - 380 gallons of CCl_4 evaporated per month, and 1963 data - 1000 gallons of CCl_4 evaporated per month. The CCl_4 in the glovebox atmosphere was considered to be present in only one-half of the gloveboxes because the machining area, from which most evaporation occurs, includes about one-half of the gloveboxes in the system.

CONTROL OF WATER VAPOR IN GLOVEBOXES

Two of the four systems to be inerted at Rocky Flats also require control of the water vapor concentration in the glovebox to minimize corrosion. In these two systems the maximum allowable water vapor concentration is approximately 200 ppm by volume or 0.85 grains H_2O/pound dry air. Humidity in the room surrounding the gloveboxes is typically about 12 grains H_2O/pound of dry air in summer, and about 9 grains H_2O/pound of dry air in winter.

Oxygen and water vapor enter the gloveboxes and occur in gloveboxes proportional to their room air concentration,

$$\text{i.e.,}\ \frac{H_2O\ \text{Outlet}}{O_2\ \text{Outlet}} = \frac{H_2O\ \text{Room}}{O_2\ \text{Room}} = \frac{12\ \text{grains/lb. dry air}}{21\%}$$

Table V shows the values of various parameters which result. The fourth parameter is fixed by specifying three.

At 0.85 grains H_2O/pound dry air, the water vapor has the higher ratio of room concentration to glovebox concentration. (See Table VI.) Therefore, in maintaining the proper water vapor concentration in the gloveboxes the oxygen concentration will be reduced to a value well below the required level. (See Table VII.) This illustrates that both oxygen concentration and water vapor concentration must be considered when analyzing the quantity of nitrogen required to inert gloveboxes for plutonium processing.

LEAK CHECKING AND LOCATION

The oxygen balance or water vapor balance may be used on small sections of glovebox line, e.g., one box. Some oxygen indicators may be sensitive and rapid and may help in locating leaks. Extension rubber tubes inside boxes should aid by getting close to the leak. Portable, inexpensive oxygen or water vapor indicators that could be installed inside gloveboxes could help greatly. An inexpensive rapid system of conveniently locating leaks can influence the cost of inerting. Measurement of ultrasonic transmission through holes using an ultrasonic source inside the glovebox or ultrasonic noise generation by leakage have been very helpful(4).

ECONOMICS

Extra labor will be required to keep the inert boxes tight and to operate them in that condition. It was estimated by maintenance and production personnel that 10% extra operating and maintenance labor would be required for this. This cost could amount to several hundred thousand dollars per year, which is of the same order of magnitude as the cost of the nitrogen necessary for the inerting. The optimum point for operation would be at the leakage level where the sum of the two costs is the minimum. This type of analysis will require more data than is available at present.

CONCLUSIONS

1. The material balance method developed for glovebox leakage analysis provides an empirical method for determining leakage into existing glovebox systems.

2. Water vapor leakage into gloveboxes instead of oxygen will determine nitrogen usage for machining operations which are maintained at low humidities.

3. Optimum economic operation will be a balance between extra maintenance and operating labor and the cost of the nitrogen supplied to the system. Improved equipment, operation, and leak detection will lower leakage factors and minimize nitrogen cost. A larger nitrogen supply increases the safety of the inerting by assuring an adequate supply under a greater range of circumstances.

TABLE V

RELATION OF OXYGEN AND WATER VAPOR CONCENTRATIONS IN ROOM AND GLOVEBOXES

Room O_2 Concentration (%)	Room Humidity $\left(\frac{\text{Grains } H_2O}{\text{Pound Dry Air}}\right)$	Box Outlet O_2 Concentration (%)	Box Outlet Humidity $\left(\frac{\text{Grains } H_2O}{\text{Pound Dry Air}}\right)$
21	12	4	2.3
21	12	1.5	0.85
21	9	2.0	0.85

TABLE VI

RELATION OF ROOM AND GLOVEBOX CONCENTRATION OF OXYGEN AND WATER VAPOR

	Water $\left(\frac{\text{grains}}{\text{lb. air}}\right)$	Oxygen (vol %)
Room Concentration	12.0	21.0
Glovebox Concentration	0.85	4.0
Ratio $\frac{\text{Room Concentration}}{\text{Glovebox Concentration}}$	14.1	5.3

TABLE VII

OXYGEN CONCENTRATION LEVEL IN GLOVEBOX WHEN WATER VAPOR CONCENTRATION IS MAINTAINED AT 200 PPM BY VOLUME

$$\text{Outlet } O_2 \text{ Concentration} = \frac{\text{Outlet } H_2O \text{ Concentration}}{\text{Room } H_2O \text{ Concentration}} \times \text{Room } O_2 \text{ Concentration}$$

$$\text{Outlet } O_2 \text{ Concentration} = \left(\frac{0.85 \ \frac{\text{grains } H_2O}{\text{lb. air}}}{12.0 \ \frac{\text{grains } H_2O}{\text{lb. air}}} \right) \times (21\%) = 1.5\%$$

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INERT ATMOSPHERE MANAGEMENT AND SURVEILLANCE AT THE ARGONNE NATIONAL LABORATORY FUELS TECHNOLOGY CENTER

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April 13, 1971

INERT ATMOSPHERE MANAGEMENT AND SURVEILLANCE AT THE ARGONNE NATIONAL LABORATORY FUELS TECHNOLOGY CENTER*

Introduction

Argon and nitrogen atmosphere gloveboxes and nitrogen atmosphere hot cells are in operation at the Fuels Technology Center at Argonne National Laboratory to evaluate plutonium alloys, compounds, or mixtures, and the techniques necessary to produce or investigate these materials. A two-cell cave facility and approximately 80 gloveboxes with inert atmosphere are currently in use. This equipment is installed in three separate areas of the building and operated independently. All enclosures in which plutonium is handled are maintained at a pressure that is negative with respect to the room and are ventilated with inert gas on a low-flow, once-through basis. This technique is possible because of the extremely low leak rate of the enclosures.

The following discussion describes some of the design features, administrative controls, and techniques employed that provide and maintain inert atmospheres for the main purpose of fire prevention. As a secondary benefit, a high-purity atmosphere gives the scientist a significant technical advantage when working with reactive materials such as sodium, NaK, or plutonium-bearing materials.

Atmosphere Source and Purity

Argon and nitrogen are purchased and delivered as cryogenic fluids. Separate vendor contracts are solicited for a one-year term on the basis of estimated consumption and, of course, low bid. Liquid nitrogen is routinely delivered to the facility on Monday, Wednesday, and Friday, which is convenient for both the Laboratory and the vendor because of the large amount of liquid nitrogen delivered for other purposes each day. Liquid argon is delivered on demand by calling the vendor when a low-liquid-level alarm is activated, which is approximately once a month.

The Laboratory specifications require that liquid nitrogen be 99.997% pure. Maximum oxygen concentration is limited to 20 ppm of the total 30 ppm impurities. The mositure content is not specified, nor does it seem customary for vendors to recognize moisture as an impurity. This may seem disturbing and inconsistent, but in practice, a high moisture content in a cryogenic fluid processing system cannot be permitted. Normally, 1 to 2 ppm moisture is observed in the delivered product, and only minor variations occur from year to year. Oxygen concentrations usually vary between 3 and 8 ppm, with occasional variations between 1 and 15 ppm.

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*This work was performed under the auspices of the United States Atomic Energy Commission.

Specifications for liquid argon are that it also be 99.997% pure. The maximum oxygen concentration is limited to 5 ppm, nitrogen is limited to 20 ppm, and other gases are limited to 5 ppm total. Oxygen monitored during the past two years has not exceeded 5 ppm except for a few brief excursions to 30 ppm caused by improper techniques used in liquid transfer between trucks and the converter. These excursions actuate alarm devices but are not cause for serious concern.

Quality Control

Source gas quality control is assured by a four-step process:

(1) Product purity (99.997%) and maximum specific impurities (O_2 and N_2) are specified by the Laboratory. The specifications are realistic with respect to vendor capabilities and the end use of the product.

(2) A vendor-performed analysis of each delivery is specified, and this analysis is submitted to the Laboratory with each load.

(3) A "go-no go" test is performed by the Laboratory's Fire Protection Department for each delivery. Samples of the liquid phase are taken from the vehicle and analyzed for oxygen content before the product is dispensed. Occasionally liquid nitrogen quality does not quite meet the specified purity. These deliveries can usually be accepted for use in noncritical facilities where the product is used primarily for cold-trap operations. When a wide variation between specification and analysis is apparent, delivery is not accepted. Variations from the liquid argon specification are never permitted. Analysis is performed with a Hot Filament Oxygen Detector, A.P.C. 13-493-99-0020, the calibration of which is verified with cylinder gas of known oxygen content.

(4) Argon and nitrogen gas supply systems within the facility are continuously monitored for oxygen and moisture content. Instrumentation consists of Beckman and Consolidated Electrodynamics electrolytic type moisture monitors and Analytic Systems Company series 300 trace oxygen analyzers. Alarm systems connected to these instruments alert responsible persons in the event of high impurity levels.

Conversion and Distribution

Liquid argon and liquid nitrogen converter systems are installed adjacent to the building (see Figure 1). The two systems are similar except that the liquid nitrogen system has a steam-to-liquid nitrogen vaporizer in series with an ambient vaporizer. The liquid products are contained in the storage vessels at 90 psig, continuously dispensed on demand through vaporizers and three-stage pressure reduction stations, and distributed throughout the building as gases at 60 psig. Pressure reduction to approximately 10 psig is made at the facility being serviced. Both converter systems include pressure-building coils, gas (boiloff) recovery systems, and low-level alarms. The converters are not dependent upon any utility. The liquid nitrogen steam vaporizer mentioned above is not essential, but it has eliminated ice formation problems characteristic of a high and continuous demand, ambient vaporizer system.

The liquid product is not dispensed for any other purpose from these systems nor do the gas distribution systems serve any apparatus other than glovebox or hot-cell facilities. The gas distribution systems can be manually cross connected so that each system is a backup for the other. Cylinder gas reserve manifolds are included only in the argon system but are not really considered necessary. The cylinder manifolds supplied the facilities before installation of the cryogenic system and will probably be removed if other needs for manifolds develop.

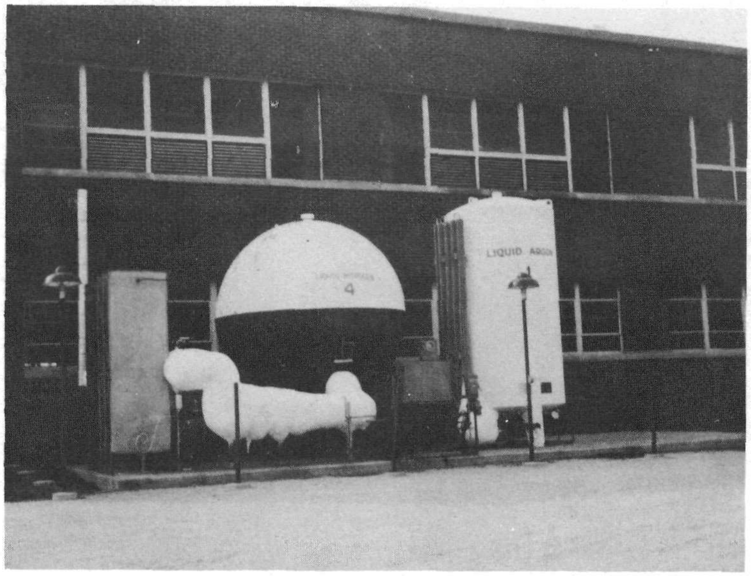

Figure 1. Liquid Nitrogen and Liquid Argon Converters

Gas connections to individual gloveboxes or hot cells are made through pipes, check valves, and metering hardware of limited size and capacity. The combination of fixed line pressure and built-in pressure drop guarantees finite limits for individual flow rates. Gloveboxes are limited to approximately 2 scfm per glovebox, and the hot cells are limited to approximately 15 scfm each. Normal flow rates are 0.2 scfm per glovebox and from 5.0 to 8.0 scfm per hot cell.

Gloveboxes

Inert atmospheres are contained in approximately 80 gloveboxes, located in 17 laboratories in separate areas of the building, and in two separate cells of an Alpha-Gamma facility.

The gloveboxes vary in size from 8 to 87 cubic feet and are maintained at a 1/2-inch water-column pressure with respect to the room. Negative pressures are used for plutonium facility gloveboxes and positive pressures for nonplutonium gloveboxes.

Structures consist of extruded aluminum frames, safety glass windows, aluminum panel sides and baseplates, and O-ring seals between the frame and each panel or window (see Figures 2 and 3). This design results in gloveboxes having a leak rate less than 0.02% of the box volume per hour measured (calculated) at a differential water-column pressure of 4.0 inches, before gloves and pouches are applied. The glovebox structure is proof tested at a water-column pressure of +6 inches. Large gloveboxes (87 cubic feet) will fail by window breakage at a water-column pressure of approximately 10 inches, positive or negative. Because smaller gloveboxes are more rigid structures, they can withstand higher differential pressures.

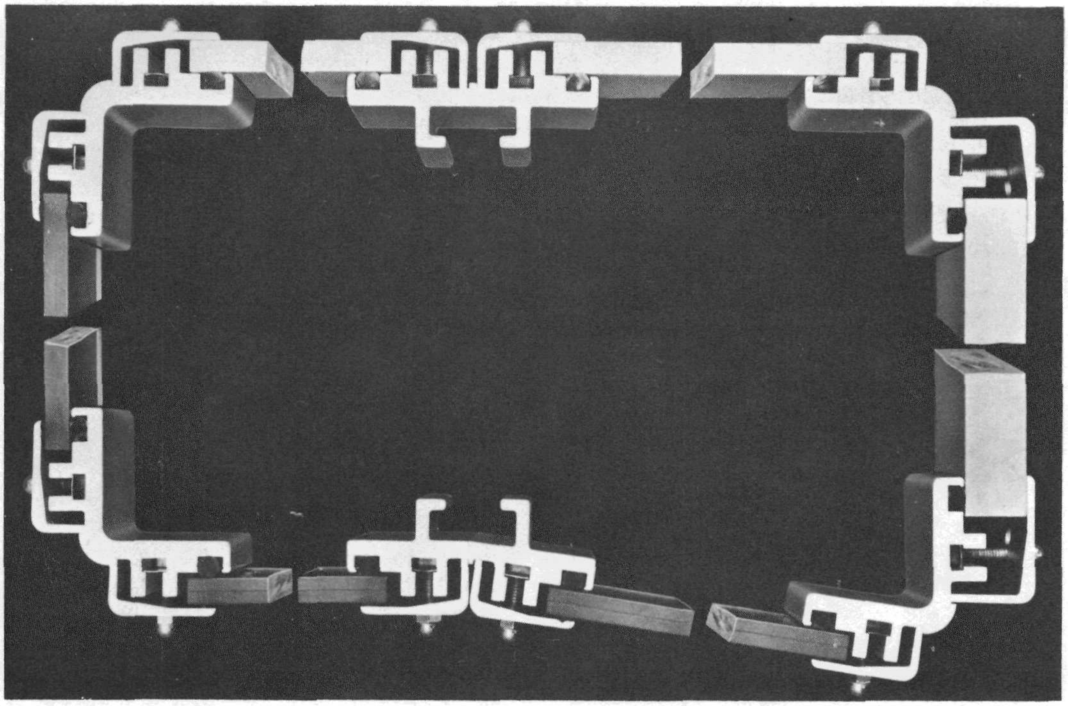

Figure 2. Cross Section through Assembled Glovebox Components

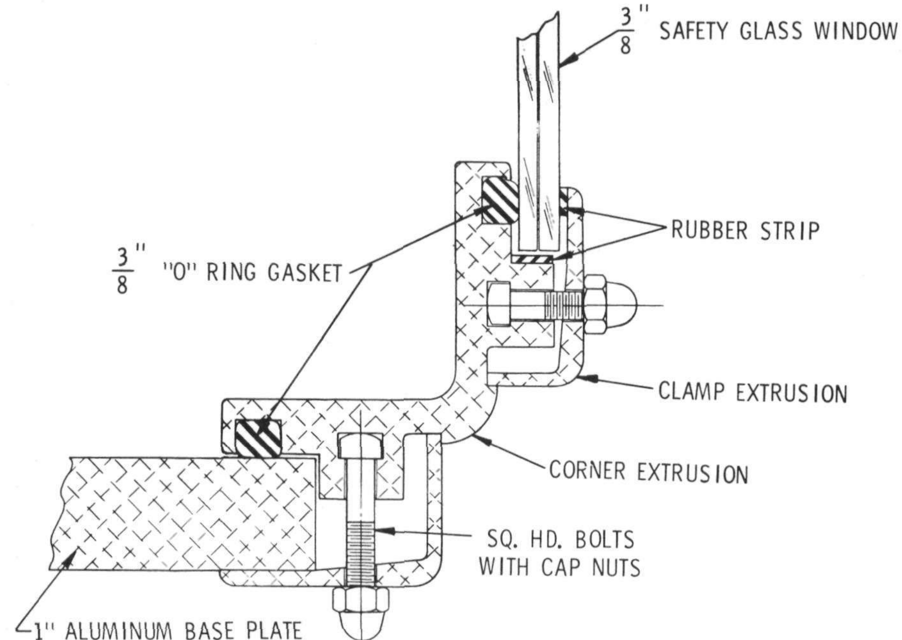

Figure 3. Section through Corner Extrusion Showing Window and Panel Seals

Leak testing is performed as a four-phase project and extends throughout the lifetime of the glovebox. Normally, the first leak test is by a helium mass spectrometer "sniffing" technique. The second test is an air-leak rate test performed at a water-column pressure of +4 inches. Pressure and temperature changes of the closed glovebox are carefully measured and the leak rate calculated from data accumulated over a period of several hours (sometimes days). This very sensitive procedure requires regulation of other activities in the room and stable weather conditions during the test.

After the first two tests the glovebox may be opened, windows removed, and equipment installed and tested. This process sometimes requires several months of effort. When all equipment is ready for use, the windows are reinstalled, gloves and pouches applied, and the box purged with inert gas.

The third leak test is performed under operating conditions. When the glovebox has been purged of air and connected to the pressure control system, it is ventilated (with inert gas) at a rate of 0.2 scfm for two or three days. During this time no work (glove manipulation) is performed in the box. Then the oxygen content of the box atmosphere is measured. An oxygen level of 50 to 75 ppm is considered acceptable, and the glovebox is placed in operation.

The fourth phase of leak testing is a continuing process. Twice each year, usually over a three- or four-day weekend, all glovebox gas flow rates are reduced to 0.1 scfm (half the normal nominal flow). On the next workday, before the facilities are used, all gloveboxes are carefully monitored for oxygen content. Approximately six hours are required to test all gloveboxes in the plutonium facilities. Leaking gloveboxes are easily identified by the accumulated data. In a glovebox train with one leaking glovebox, an oxygen gradient from box to box can be observed. Glove deterioration is the usual cause of glovebox leakage, but a few persistent leaks have been detected and repaired in other pieces of glovebox hardware.

The Working Atmosphere

An average glovebox, purged at a rate of 0.2 scfm, in which normal transfer techniques are used (PVC bagging and push-through can entries) will have an inert atmosphere containing less than 200 ppm oxygen and approximately 100 ppm moisture. Carefully performed transfer techniques and a flow rate of about 1 scfm will result in an atmosphere containing less than 50 ppm oxygen and about 25 ppm moisture.

Leaking gloveboxes are usually detected through oxygen analyzer alarm systems before oxygen levels reach 0.5%. A large leak can result in a 3-5% oxygen concentration before corrective action is taken, but oxygen levels above 1% are rare.

The two hot-cell atmospheres vary in oxygen level with transfer operations and leak rate changes. One cell, designed and tested for inert gas application, normally contains less than 200 ppm oxygen when ventilated with nitrogen at a rate of 5 scfm. The other cell, designed for air ventilation and recently converted to nitrogen atmosphere, contains approximately 2.5% oxygen when purged at 8 scfm.

Instrumentation and Alarms

Continuous oxygen analysis is made of the manifolded glovebox exhaust gas. Two laboratories, each housing up to 10 gloveboxes, are monitored by one indicating, recording instrument. When a high-oxygen level is observed or causes an alarm, the "problem lab" is easily identified by manipulating sample line valves (see Figure 4). Once the problem area is defined, each glovebox may be monitored through an individual sample line connected to a manifold at the analyzer location. Instrument troubleshooting and glovebox sampling are performed outside the laboratory space. Glovebox troubleshooting requires laboratory entry, but only after the specific leaking glovebox has been identified.

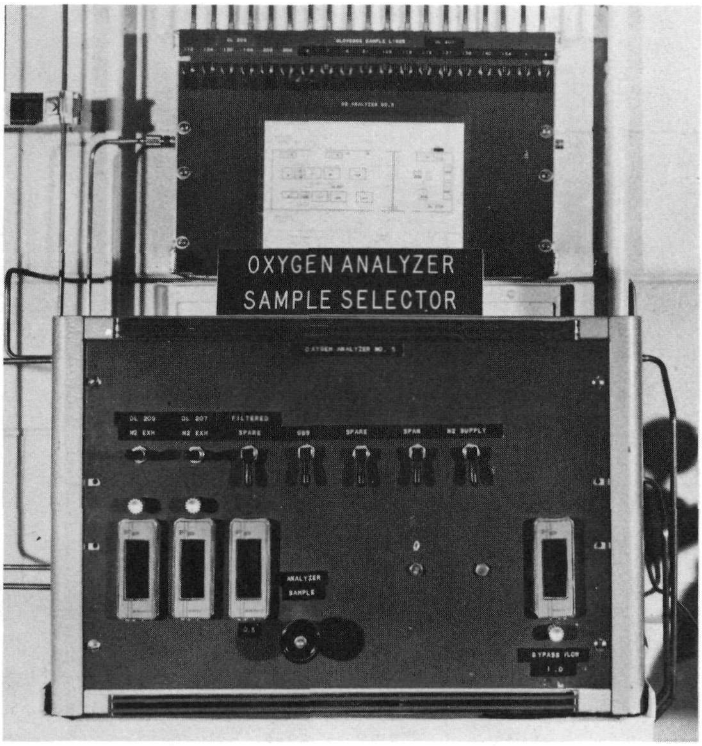

Figure 4. Oxygen Analyzer Sample Selection Panel

Hot-cell installations are similar, but locating a leak in one large enclosure is accomplished by moving the sensing point (the end of a long plastic hose) to various locations within the cell.

Instrumentation consists of dual range, paramagnetic-type oxygen analyzers, ANL-developed sample-selector systems, and indicating recorders equipped with alarm contacts. Instrument ranges of from 0 to 1% and 0 to 5% oxygen are most common. The Hays Corporation Model 632 is preferred for this low-range application. Recorders are 24-hour circular-chart devices with contacts set to alarm at 25% of full scale (0.25% or 1.25% for 1% and 5% ranges, respectively).

Local alarms sound in affected areas immediately, and signals are relayed to a central building annunciator system if not manually silenced within 1-1/2 minutes by area occupants. If the building annunciator is not responded to within 20 minutes, the signal is relayed to the Laboratory telephone switchboard, which is always manned.

Response to alarms during off-hours is accomplished through emergency call lists available to the persons on duty. Initial troubleshooting is sometimes performed via phone instructions. This procedure usually results in either establishing the fact that an instrument or sample system has malfunctioned or that a real glovebox leak has been announced. Approximately three real leaks per year have been detected during the off-hour periods in the past three years. Recorder charts usually reveal that a small leak developed during the late afternoon hours, ultimately actuating the alarm sometime between 7:00 p.m. and 3:00 a.m.

Summary

In conclusion, the main points of atmosphere management and of surveillance may be briefly listed.

Management Techniques

(1) Source-gas purity is specified by the Laboratory.

(2) Vendor-gas analysis is provided with each delivery.

(3) Liquid conversion and gas distribution are not dependent on utilities.

(4) Distribution pressures and components are designed to limit flow at the point of use.

(5) Gas usage is restricted to glovebox or hot-cell atmospheres.

(6) Low glovebox leak rates are obtained before use as plutonium handling devices.

(7) Gas flow rates are reduced semiannually and increased only as needed by the glovebox user.

(8) Circular recorder charts "demand" daily attention, which results in detecting problems before they become serious.

Surveillance Techniques

(1) Each delivery is tested by the Laboratory for oxygen content.

(2) Gas supply systems within the facility are continuously monitored for oxygen content.

(3) Glovebox and hot-cell exhaust systems are continuously monitored for oxygen content.

(4) Cryogenic reservoirs are equipped with liquid-level alarms.

(5) Gas distribution systems are equipped with pressure alarms.

(6) Oxygen analyzer charts are changed, and, therefore, are observed daily.

(7) Instruments are calibrated weekly, mainly to assure that they do respond to oxygen.

(8) Leaking gloveboxes are identified at least twice each year.

The techniques listed above are not in a priority sequence, nor would the same priority apply to both argon and nitrogen systems for a given technique.

PLUTONIUM IGNITION IN INERT ATMOSPHERES

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PLUTONIUM IGNITION IN INERT ATMOSPHERES*

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ABSTRACT

The prevention of plutonium ignition is an important consideration in handling plutonium. In order to determine fire safe, inert atmospheres in which plutonium can be handled, the ignition of plutonium has been studied in nitrogen-air and carbon dioxide-air mixtures. The results show that such mixtures are effective in preventing plutonium ignition when the oxygen concentration is below about 5 percent. The maximum allowable oxygen concentration in which plutonium ignition is prevented is higher as the specific surface area decreases. It is also higher in nitrogen-air than in carbon dioxide-air mixtures.

INTRODUCTION

Experience with plutonium metal shows that under certain conditions the metal is pyrophoric. Plutonium has been involved in serious fires both in England and in the United States. In addition to the direct fire hazards, plutonium fires offer severe toxilogical and radiation hazards. The maximum permissible dose of plutonium is about 1 microgram. Therefore, the spread of plutonium aerosols by fire is extremely undesirable. In cases where such aerosols are spread, much time and effort must be spent in decontamination.

These factors require that great effort be expended in preventing plutonium fires. Much progress has been made in understanding plutonium ignition and burning.[1-5] It appears, however, that only inert atmospheres are completely effective in preventing plutonium ignition and burning. Thompson[1] observed plutonium ignition in argon with greater than 6.5 percent oxygen. Felt[2] found that argon containing about 5 percent oxygen was adequate to extinguish burning plutonium. Schnizlein and Fischer[3] incorrectly reported that Rhude[6] observed plutonium burning in 5% oxygen in nitrogen and to be extinguished only in 1% oxygen in nitrogen. Rhude was using magnesium to simulate plutonium due to hazards involved with handling plutonium.

The cost of inerting a large plutonium handling area with argon is high. Economic substitutes of partially inert gases such as nitrogen or carbon dioxide would be desirable, if the necessary

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*Work performed under the auspices of the U. S. Atomic Energy Commission Contract AT(29-1)-1106.

degree of protection could be attained. Since little information is available on the ignition of plutonium in nitrogen or carbon dioxide atmospheres, this study was carried out.

The goal of this study was to determine protective atmospheres for a large plutonium processing area. For this reason, plutonium lathe turnings were most comprehensively studied. The turnings are most likely to cause serious fires in the area because they are generated in large amounts and have a sufficiently high surface area to ignite easily. Supplementary studies on more massive coupons and more finely divided filings were carried out to determine the effect of surface area in such atmospheres.

EXPERIMENTAL

The ignition temperatures of the samples were determined by the burning curve method.[7] Figure 1 shows a schematic of the apparatus. The sample was placed on a spoon inside a Vycor tube. It was heated at a 30°C temperature rise per minute with a tube furnace. The sample temperature was monitored with a platinum/platinum-13% rhodium thermocouple placed inside the spoon. At ignition, the sample temperature rose rapidly. The heating rate curve and the temperature curve following ignition were graphically extrapolated as shown in Figure 2. The intersection of these curves is the ignition temperature. Gases surrounding the sample were controlled by continuous flow of the proper mixture through the Vycor tube.

In a supplementary experiment to determine the effect of oxygen concentration on burning plutonium the equipment shown in Figure 3 was used. Plutonium coupons were placed on a heater maintained at 500°C and ignited. A Vycor bottle through which a gas mixture flowed was placed over the sample until glowing ceased. The time between placing the bottle over the sample and the cessation of glowing is the "glow" time.

Plutonium used in these studies was either unalloyed or delta-stabilized (Pu/1 wt % Ga). All samples weighed about 0.5 g. Coupons were wire brushed and cut to size (about 1.0 x 0.5 x 0.1 cm^3). The delta-stabilized lathe turnings were about 2.0 x 0.1 x 0.02 cm^3. The unalloyed lathe turnings were about 0.2 x 0.2 x 0.02 cm^3. When necessary oil was removed from the lathe turnings by washing in carbon tetrachloride. The filings were obtained from a clean delta-stabilized coupon. The filing was done in a dry air glovebox (200-700 ppm moisture). They were collected between 300 and 140 mesh screens. All filings were tested within 24 hours to reduce surface oxidation.

Gases used were compressed air and bottled nitrogen. Flow was regulated and measured with a flow meter. Mass spectrometry was used to check the accuracy of the flow measurements.

RESULTS

Initially there was concern to compare experimental data observed in nitrogen-air mixtures with that previously published in argon-air mixtures. There was particular interest in comparing this work with the data of Felt[2] because Felt had used much larger

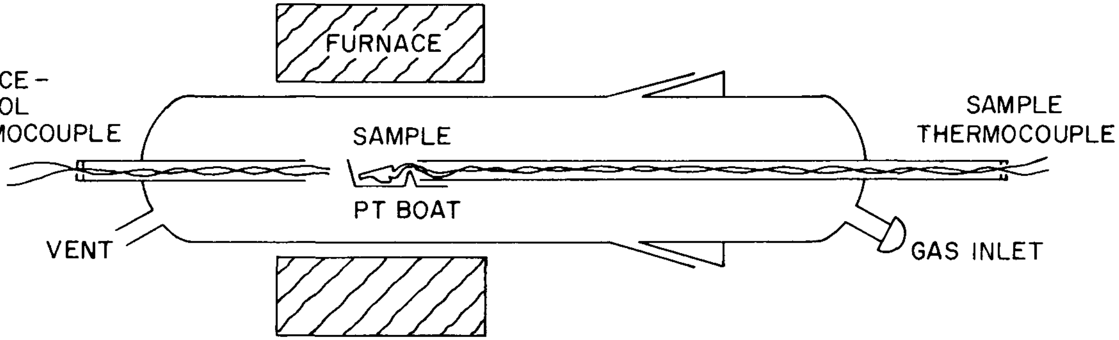

Figure 1. Apparatus for determining the ignition temperature by the burning curve method.

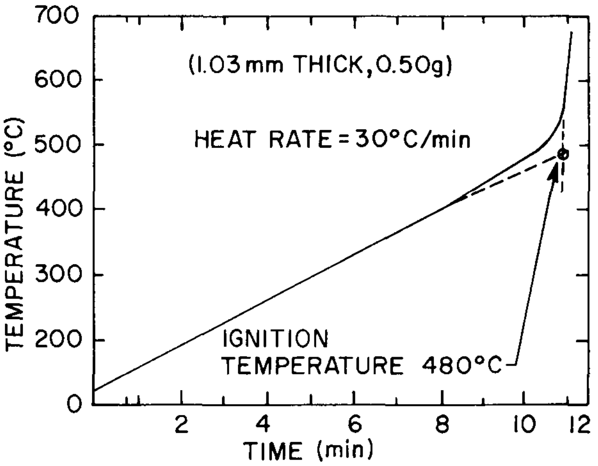

Figure 2. Burning curve of a 0.5-g delta-stabilized coupon in air showing extrapolations used to determine the ignition temperature.

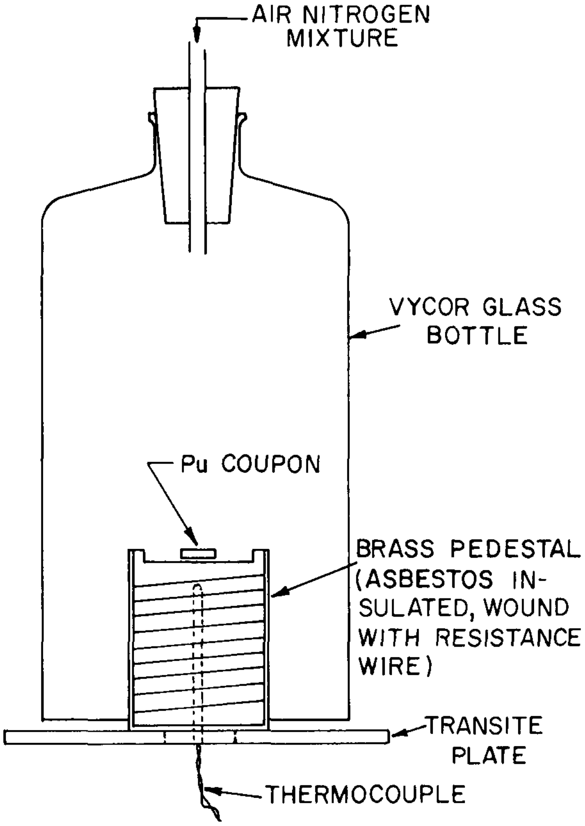

Figure 3. Apparatus for determining the glow time of burning plutonium.

amounts of plutonium which more realistically represented a fire situation. Therefore, delta-stabilized plutonium was ignited in nitrogen-air mixtures to determine at which oxygen concentration ignition would cease. It was observed that the temperature rise following ignition gradually decreased as the oxygen concentration decreased making it difficult to distinguish between ignition and non-ignition. Arbitrarily a definition of ignition was developed. Ignition was defined as a sudden temperature rise of at least 50°C in the burning curve. The data gained with the 0.5 g delta-stabilized coupons is shown in Table I. The slight reactions indicated at 5 and 6 percent oxygen concentrations indicate a sudden temperature rise below 50°C. This rise occurred at about 510°C. Hence, ignition was stopped in oxygen concentrations less than 6 percent, a value similar to that observed by Thompson[1] in air-argon mixtures and consistent with Felt.[2] A supplementary measure of glow time shown in Table II confirms that oxygen concentrations of up to 7 percent could readily extinguish the burning delta-stabilized plutonium coupons.

Similar data was gathered with delta-stabilized plutonium lathe turnings shown in Table III. It is noted that ignition is prevented in nitrogen-air mixtures containing less than about five percent oxygen.

During tests with delta-stabilized plutonium filings it was found that the maximum temperature following ignition could be reproducibly measured to within ±20°C by careful placement of the sample thermocouple in the middle of the powder bed. This gave further information on the effect of reduced oxygen levels on plutonium ignition. Figure 4 shows that the ignition temperature changed little as the oxygen concentration was reduced. However, the maximum temperature during ignition was lowered as the oxygen concentration was reduced. This demonstrated that plutonium powder would not ignite in oxygen concentrations below about 4 percent if the previous definition of a 50°C temperature rise was used. Furthermore, the reaction of the powder was incomplete. Samples which reacted in three or five percent oxygen concentrations in nitrogen could be cooled and re-ignited in air at 200°C--about 30°C greater than the ignition temperature of unreacted samples in air.

It was also found that the maximum temperature of lathe turnings could be reproduced if the sample thermocouple was carefully placed. Figure 5 shows the ignition temperature and maximum temperature of unalloyed plutonium lathe turnings in air-nitrogen mixtures. The ignition temperature is nearly constant and the maximum temperature declines as the oxygen concentration is reduced. Ignition is prevented in oxygen concentrations less than about six percent.

Figure 6 shows the ignition temperature and maximum temperature for unalloyed plutonium lathe turnings in air-carbon dioxide mixtures. Again the ignition temperature is nearly constant and the maximum temperature declines as the oxygen concentration is reduced. There is a unique, reproducible double ignition phenomena at about 11 percent oxygen. The metal ignites at about 300°C, heats to about 500°C, then cools to the furnace temperature. The furnace continues heating and the sample re-ignites at about 500°C

Table I. Ignition temperature of 1 wt % Ga alloyed plutonium coupons in various concentrations of oxygen.

Ignition Temperature (±10°C)	Oxygen Concentration (vol % O_2)
505	20 ± 1
510	10 ± 1
no ignition (slight reaction)	6 ± 1
no ignition (very slight reaction)	5 ± 1
no ignition	<1

Table II. "Glow" time of 1 wt % Ga alloyed plutonium coupons in various concentrations of oxygen.

"Glow" Time (±.01 min)	Oxygen Concentration (vol % O_2)
.03	<1
.03	4 ± 1
.04	6 ± 1
.06	7 ± 1
.10	9 ± 1
1.0	10 ± 1
1.5	20 ± 1

Table III. Ignition temperature of 1 wt % Ga alloyed plutonium lathe turnings in various concentrations of oxygen.

Ignition Temperature (±10°C)	Oxygen Concentration (vol % O_2)
265	20 ± 1
270	12 ± 1
400	8 ± 1
slight reaction at 270°C	5 ± 1
no reaction	3 ± 1
no reaction	<1

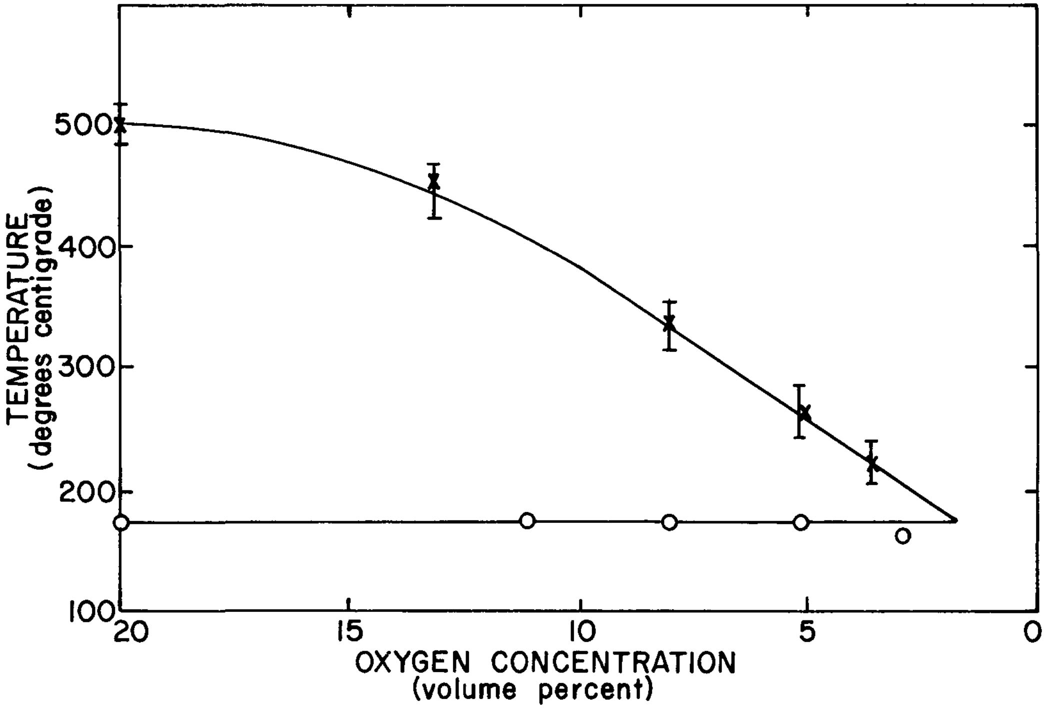

Figure 4. The ignition and maximum temperatures of 140 mesh delta-stabilized plutonium powders in nitrogen-air mixtures.

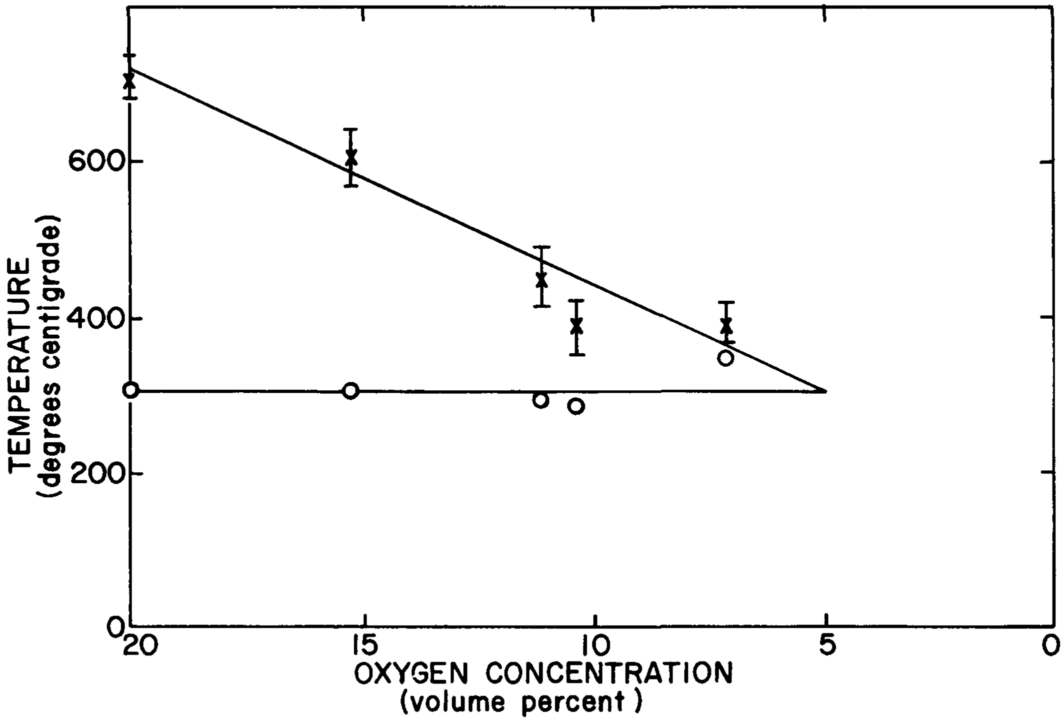

Figure 5. The ignition and maximum temperatures of unalloyed plutonium lathe turnings in nitrogen-air mixtures.

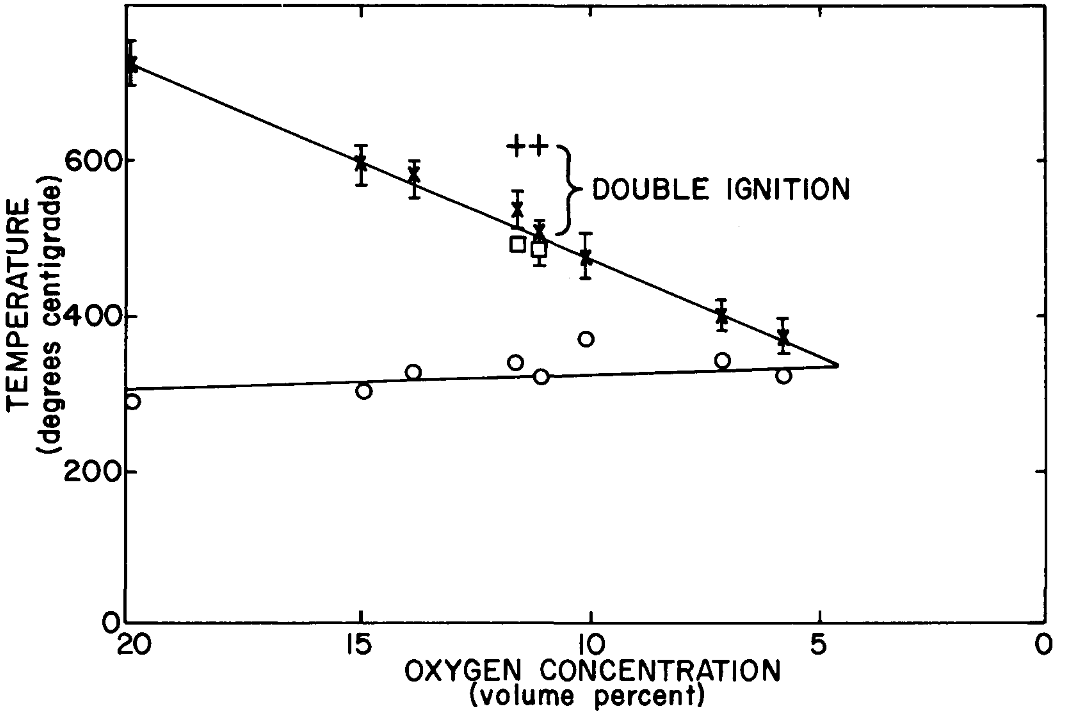

Figure 6. The ignition and maximum temperatures of unalloyed plutonium lathe turnings in carbon dioxide-air mixtures.

and heats to about 580°C. A similar phenomena has been observed with delta-stabilized plutonium chips in carbon dioxide-air mixtures at about 11 percent oxygen concentration. This phenomena is not generally observed with lower oxygen concentrations and ignition is prevented in oxygen concentrations of about 5 to 6 percent.

DISCUSSION

These results show that it is possible to significantly reduce the possible hazard of plutonium burning by inerting plutonium handling areas with either nitrogen or carbon dioxide. Plutonium, due to toxicity and radioactivity, is presently handled in gloveboxes operated at negative differential pressures. Such gloveboxes would likely allow some air leakage. However, since oxygen concentrations as great as five percent will prevent all but extremely finely divided plutonium from burning, some leakage would be acceptable, particularly if finely divided plutonium is kept to a minimum. Therefore, it is possible to achieve much greater fire safety by inerting present plutonium handling systems.

The data gathered here shows good agreement with previous studies in argon-oxygen mixtures. It also shows some agreement with the theory of Chatfield[8] who predicts that delta-stabilized plutonium will not burn in a 0.04 partial pressure of oxygen. However, the nearly constant ignition temperatures observed as the oxygen concentration was varied disagree with Chatfield's theory. Chatfield's prediction of a variation of ignition temperature with oxygen partial pressure is due to an assumption that the oxidation rate is pressure dependent. Recent experiments with plutonium show that this is not true. Larson and Cash[9] have observed oxidation rates with plutonium at a pressure of 5×10^{-2} torr similar to rates observed at atmospheric pressures. A theory by Musgrave[10] to be published shows that the ignition temperature of plutonium would not be expected to vary with varying oxygen concentration in inert gas-air mixtures.

The phenomena of double ignition in carbon dioxide-air mixtures can be explained by the following: plutonium oxidation undergoes a change in kinetics between 350°C and 400°C.[11] This change is likely due to the formation of a protective suboxide--possible PuO.[12] In preliminary experiments Larson[13] suggests that PuO is more easily stabilized in carbon dioxide. It is possible that the plutonium ignites normally, heats until sufficient PuO is formed to slow oxidation. Further heating raises the temperature sufficiently to overcome the protection of the PuO.

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FILTRATION FOR THE EXHAUST VENTILATION OF PLUTONIUM RECOVERY FACILITIES

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April 13, 1971

FILTRATION FOR THE EXHAUST VENTILATION

OF

PLUTONIUM RECOVERY FACILITIES

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Introduction

The primary component for exhaust ventilation of a plutonium recovery facility is the high-efficiency particulate air (HEPA) filter. Most exhaust ventilation systems also include additional components that perform functions supplementary to the HEPA filters. These components can extend the service life of the HEPA filters, reduce exhaust air temperatures, minimize particulate loading (dry and other), and protect the HEPA filter from fire and products of combustion that can result from a fire within the facility.

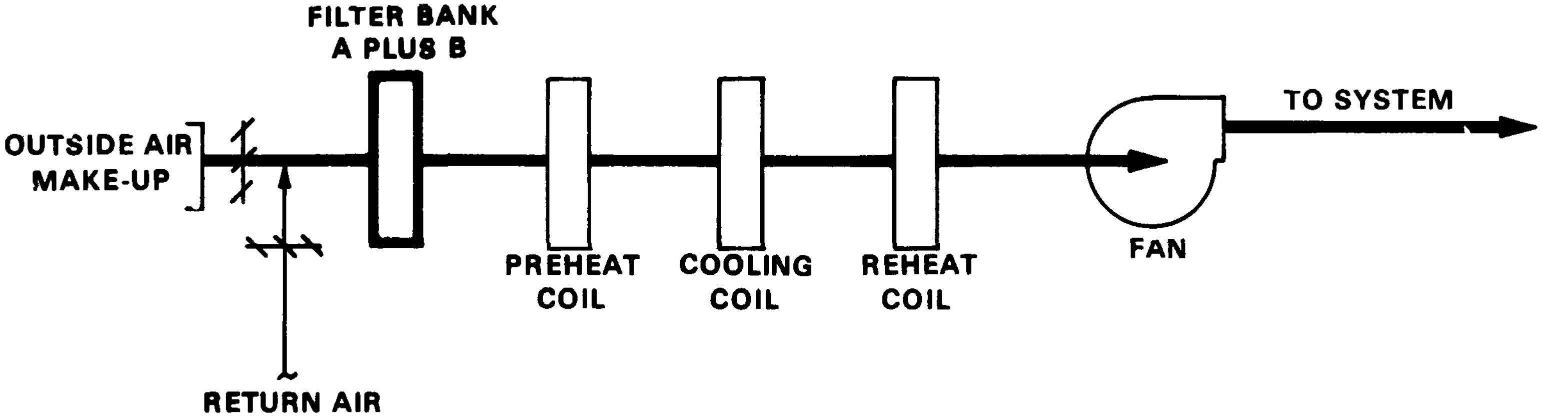
The following discussion is limited as closely as possible to these peripheral HEPA components and their application to plutonium recovery facility exhaust ventilation systems. It is apparent that the same components are also applicable to many types of nuclear plant exhaust systems to: maintain continuous operation under adverse conditions, hold radioactive effluents to a minimum, and provide for relatively simple servicing without interruption of operation.

Discussion

Figure 1 illustrates the first but not least important contribution to the exhaust system. All ventilation supply systems should include these filter components.

Prefilter A is a roll-type, fiber-glass pad media that is automatically renewable. The media advance mechanism is actuated by an adjustable timer control. A differential pressure timer override switch is advisable to compensate for unusually heavy air stream loadings (e.g., dust, water, precipitates, snow, and frost) that may be experienced.

FIGURE 1



FILTER BANK INCLUDES:

A. 8% NBS TEST RATED PREFILTER

B. OPTIONS:
  1. 50% NBS TEST RATED FILTER
  2. 80% NBS TEST RATED FILTER
  3. 93% NBS TEST RATED FILTER

An alternate Prefilter A can be a high-voltage electrostatic precipitator. This type functions as an agglomerator and requires less servicing than the roll-type prefilter.

Filter B is an extended surface type disposable unit filter. These filters are characterized by a pleated or pocket configuration.

Filter B is available for several degrees of cleaning efficiency. The determination of options B-1, B-2, or B-3 should be based upon the relative radioactivity level of the space or rooms to be served by the system.

The basic and minimal filter installation for a nuclear plant ventilation exhaust system is illustrated in Figure 2. The simplicity and economy of such a system is an advantage but it is far outweighed by many disadvantages, some of which will become apparent during further discussion.

Figure 3 illustrates an improved filter installation that incorporates a prefilter. The diagram shows an 8 per cent NBS test rated prefilter as the minimum installation. Higher efficiency prefilters are acceptable up to the rather indeterminate point where they may be overloaded and blinded by heavy particulate loading unless they are extended surface units with heavier allowable particulate loadings.

Figure 4 illustrates another improvement to the filter installation shown on Figure 3.

The second HEPA filter is installed solely to prevent escape of particulate in the event of a leak in or rupture of the first HEPA filter.

The screen is installed to prevent debris from the first HEPA filter from damaging or blinding the second.

The filter installation shown in Figure 5 further refines the installation shown in Figure 4. The additional screen shown located upstream of the first HEPA filter is installed to prevent debris from the prefilter from damaging the first HEPA filter in the event that it is ruptured.

Figure 6 illustrates a significant improvement to the filter installation. The prefilter shown on previous diagrams has been replaced with a prefilter and filter bank assembly similar to Prefilter A and Filter B shown on Figure 1, also with the alternative of an electrostatic precipitator for Prefilter A. The Filter B cleaning efficiency options are also applicable.

Up to this point, we have been concerned with the filter installation alone. However, there is the possibility of a seriously weak link in the systems we've reviewed here. The exhaust fans can be the weak link; there may be mechanical failure and they do require periodic maintenance.

The filter installation shown in Figure 7 uses two exhaust fans; they are not necessarily operated simultaneously. Each fan may be sized to deliver 100 per cent of the system's air volume. With this application (100 per cent of volume standby), the dampers upstream and downstream of the standby fan must be closed. Automatic change-over from one fan to the other is advisable.

FIGURE 2

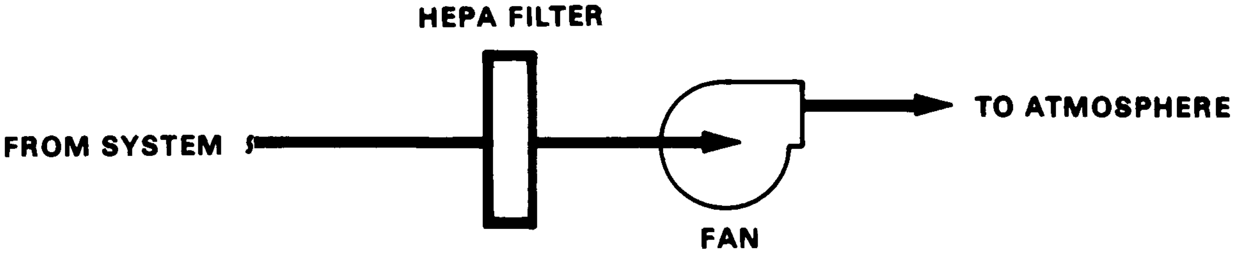
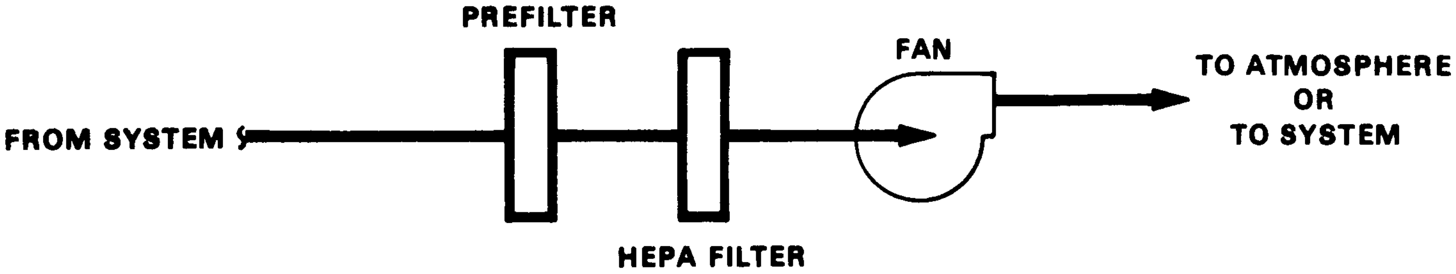
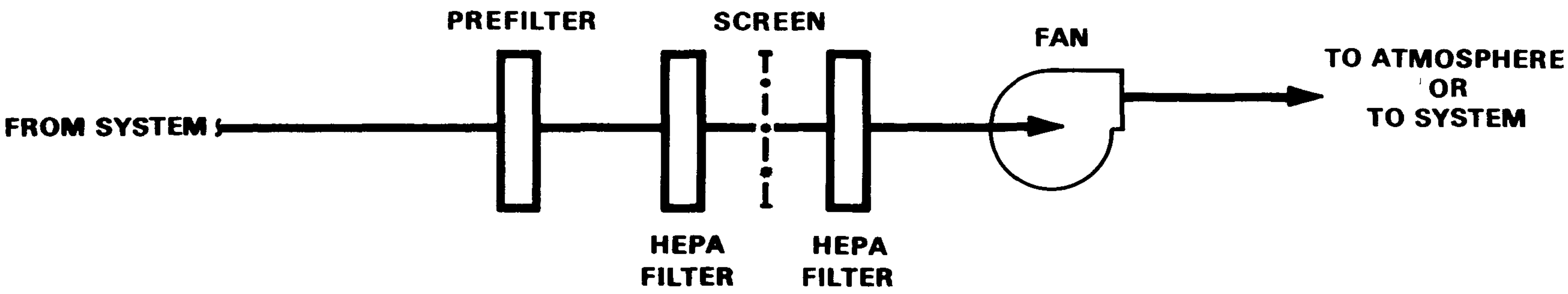


FIGURE 3



NOTE: PREFILTER IS 8% NBS TEST RATED.

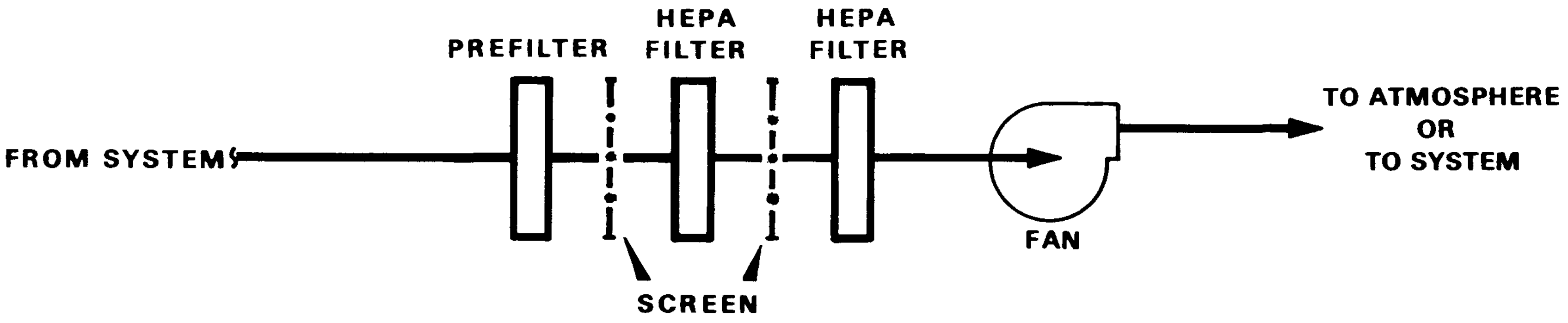
FIGURE 4



NOTES:

1. PREFILTER IS 8% NBS TEST RATED.
2. SCREEN IS 1" MESH 14 GAGE HARDWARE CLOTH.

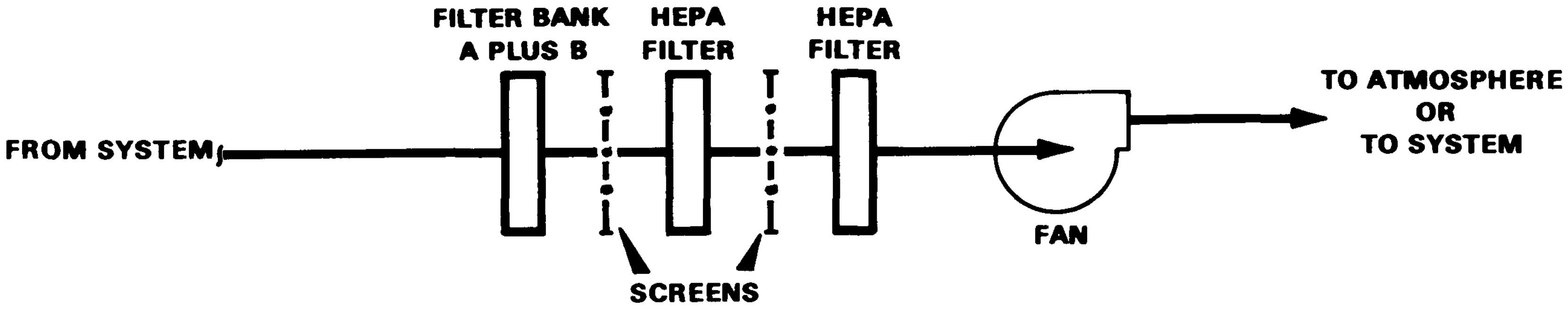
FIGURE 5



NOTES:

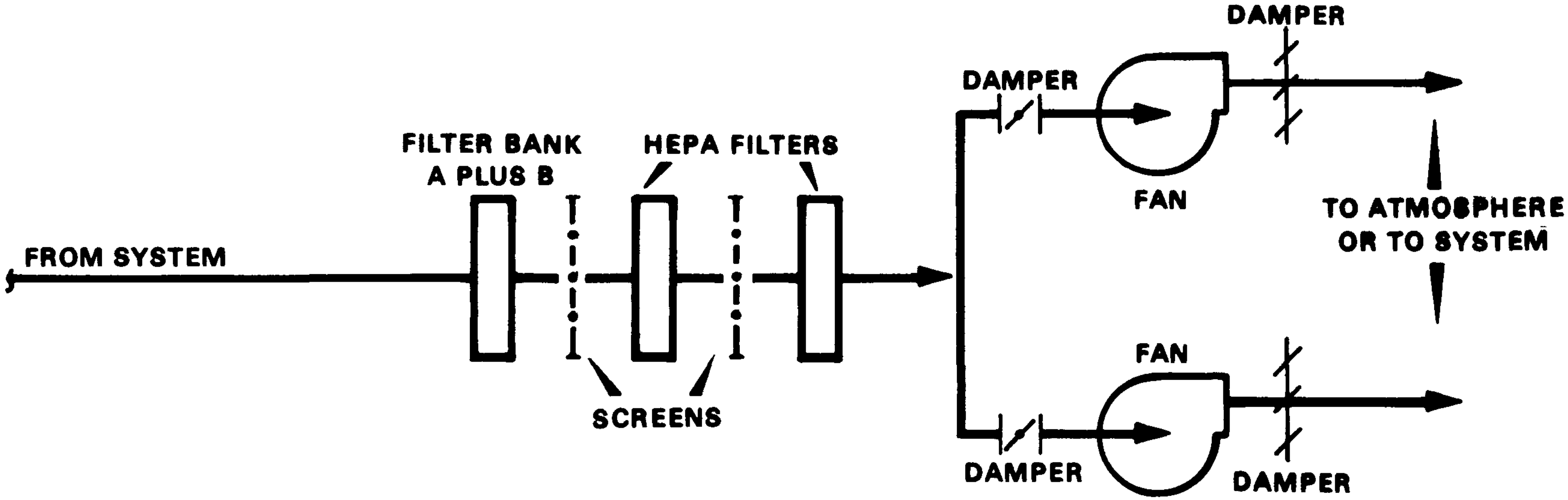
1. PREFILTER IS 8% NBS TEST RATED.
2. SCREENS ARE 1" MESH, 14 GAGE HARDWARE CLOTH.

FIGURE 6



1. FILTER BANK INCLUDES:
    A. 8% NBS TEST RATED PREFILTER
    B. OPTIONS:
        1. 50% NBS TEST RATED FILTER
        2. 80% NBS TEST RATED FILTER
        3. 93% NBS TEST RATED FILTER
2. SCREENS ARE 1" MESH, 14 GAGE HARDWARE CLOTH

FIGURE 7



NOTES:
1. FILTER BANK INCLUDES:
   A. 8% NBS TEST RATED PREFILTER
   B. OPTIONS:
      1. 50% NBS TEST RATED FILTER
      2. 80% NBS TEST RATED FILTER
      3. 93% NBS TEST RATED FILTER
2. SCREENS ARE 1" MESH, 14 GAGE HARDWARE CLOTH

The two fans can, however, be sized to deliver any volume between 100 and 50 per cent of the system air volume. In the event that either fan fails, the system will be capable of handling only that portion of the system air volume that each fan is sized to deliver. The dampers upstream and downstream of the failed fan should be automatically controlled to fail closed.

A filter installation at the state of evolution shown in Figure 7 will provide an appreciable degree of operational reliability, serviceability, and air cleaning efficiency. However, such installations should have protection from the balance of the building ventilation system. Abnormal conditions could include high exhaust air temperatures; fire and products of combustion; chemicals in liquid, fume, or precipitate states; and excessive particulate sizes and quantities. A filter and fan installation that does respond to such conditions is shown in Figure 8; a scrubber and separator have been installed upstream of the filter installation.

During normal conditions, the bypass around the scrubber and separator in Figure 8 would be open; and the fan sized to handle the system static plus the filter static would be operating. The dampers upstream and downstream of the scrubber and separator would be closed.

The scrubber would be a venturi-type, wet collector upstream of a wet centrifugal-type entrainment separator. This equipment is available with a capability to adjust throat gas velocities and scrubbing fluid quantities. Some specific applications might permit recirculation of all or a portion of the scrubbing fluid.

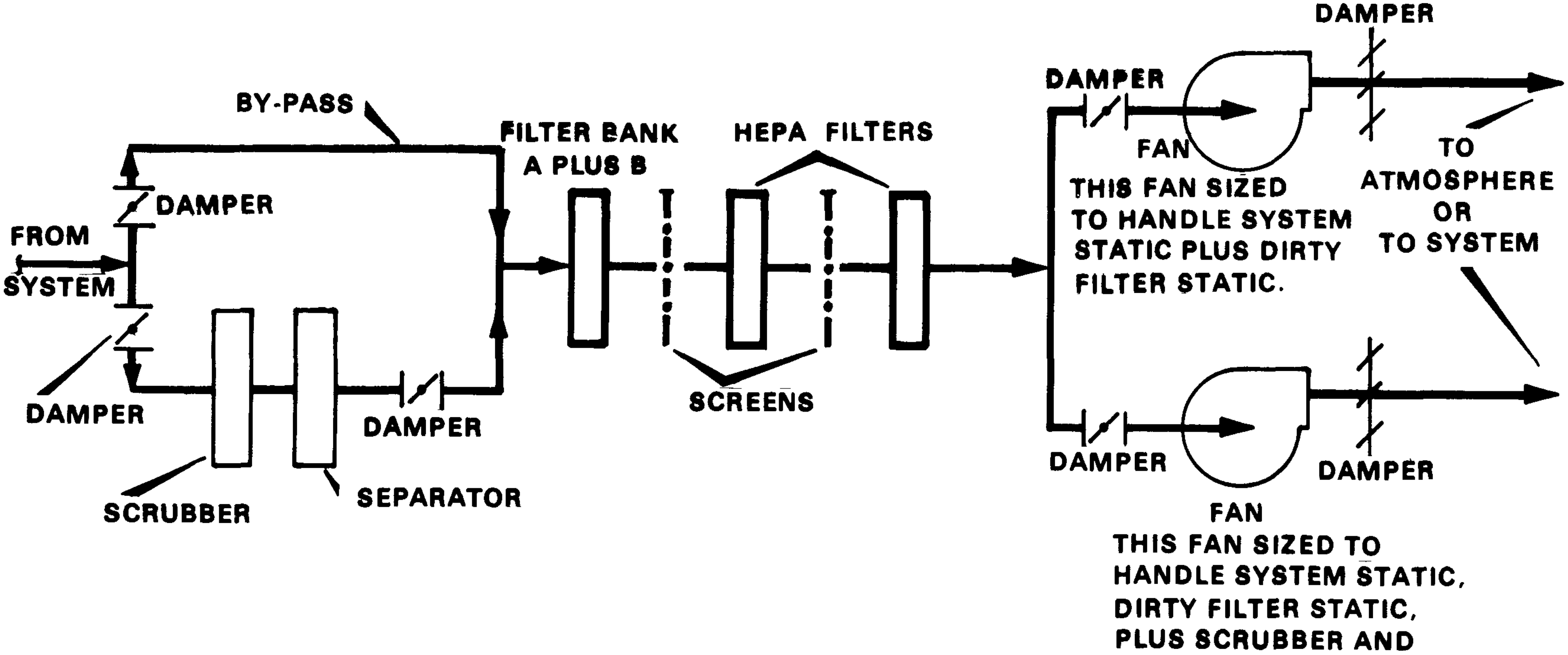
Abnormal exhaust air conditions would be detected and would automatically open the dampers upstream and downstream of the scrubber and separator; close the bypass damper around the scrubber and separator; stop the fan sized to handle the system static plus the filter static; close the respective dampers; and start the fan sized to handle the system static, plus filter static, plus scrubber and separator static, and open the respective dampers.

Figure 9 illustrates the scrubber and separator, filter, and fan installations as described in Figure 8; here the filter installation has been separated into three assemblies.

Each of these assemblies may be sized for 50 per cent of the system air volume. With this application of 50 per cent of volume standby, the dampers upstream and downstream of the standby assembly are closed. This installation will permit servicing or replacement of one filter assembly while two filter assemblies are operable. Manual changeover is advisable.

Figure 10 illustrates a desirable method of installing the HEPA filter; each filter in a bank is secured independently of adjacent units so that one unit can be replaced without disturbing others. The retaining frame angle is held in place by eight bolts so that proper gasket compression is assured. The gasket on the upstream or dirty face of the filter will properly compensate for irregularities of the filter unit and irregularities of the retaining frame. The recommended gasket compression is 25 pounds per square inch.

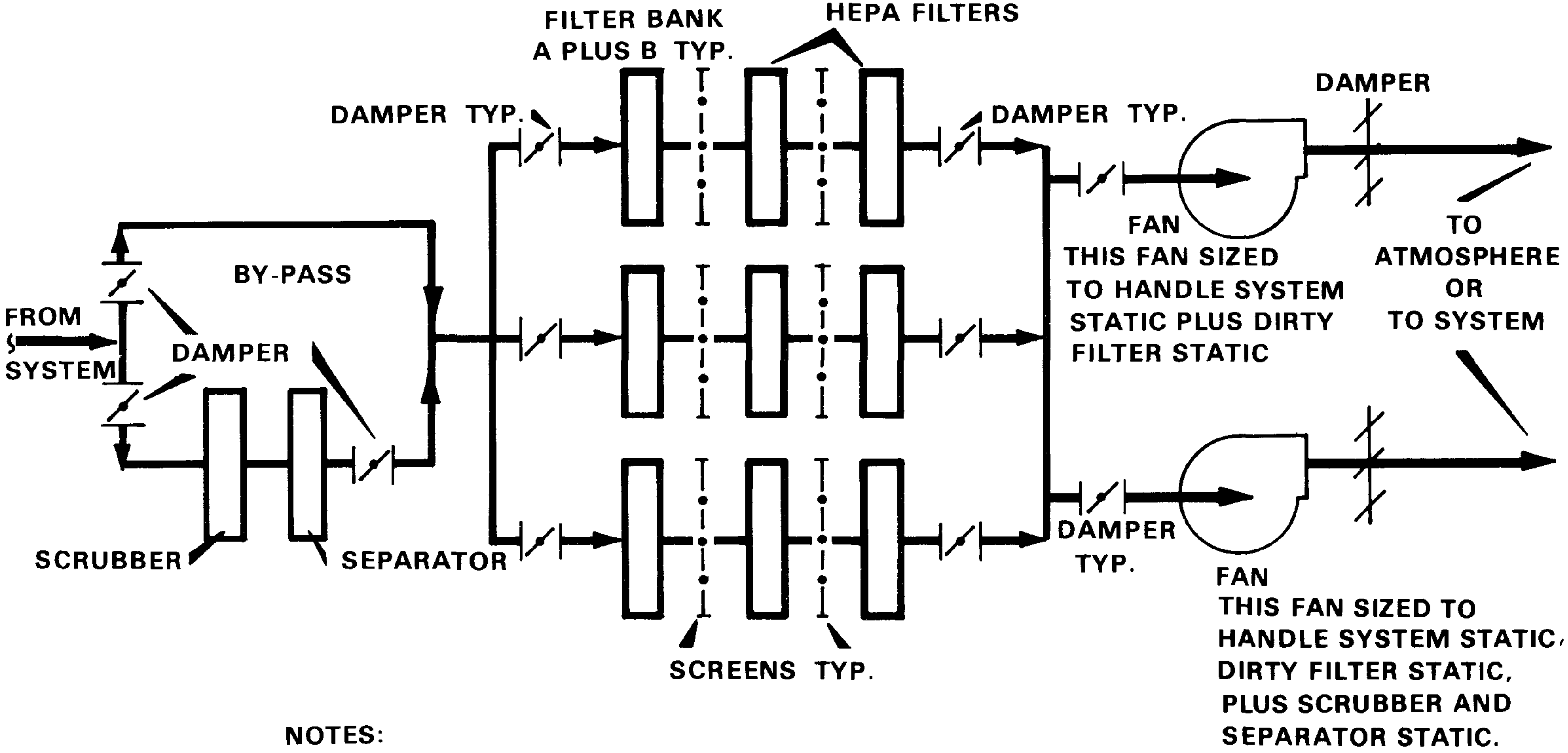
FIGURE 8



NOTES:

1. FILTER BANK INCLUDES:
   - A. 8% NBS TEST RATED PREFILTER
   - B. OPTIONS:
     1. 50% NBS TEST RATED FILTER
     2. 80% NBS TEST RATED FILTER
     3. 93% NBS TEST RATED FILTER
2. SCREENS ARE 1" MESH, 14 GAGE HARDWARE CLOTH

FIGURE 9



NOTES:
1. FILTER BANK INCLUDES:
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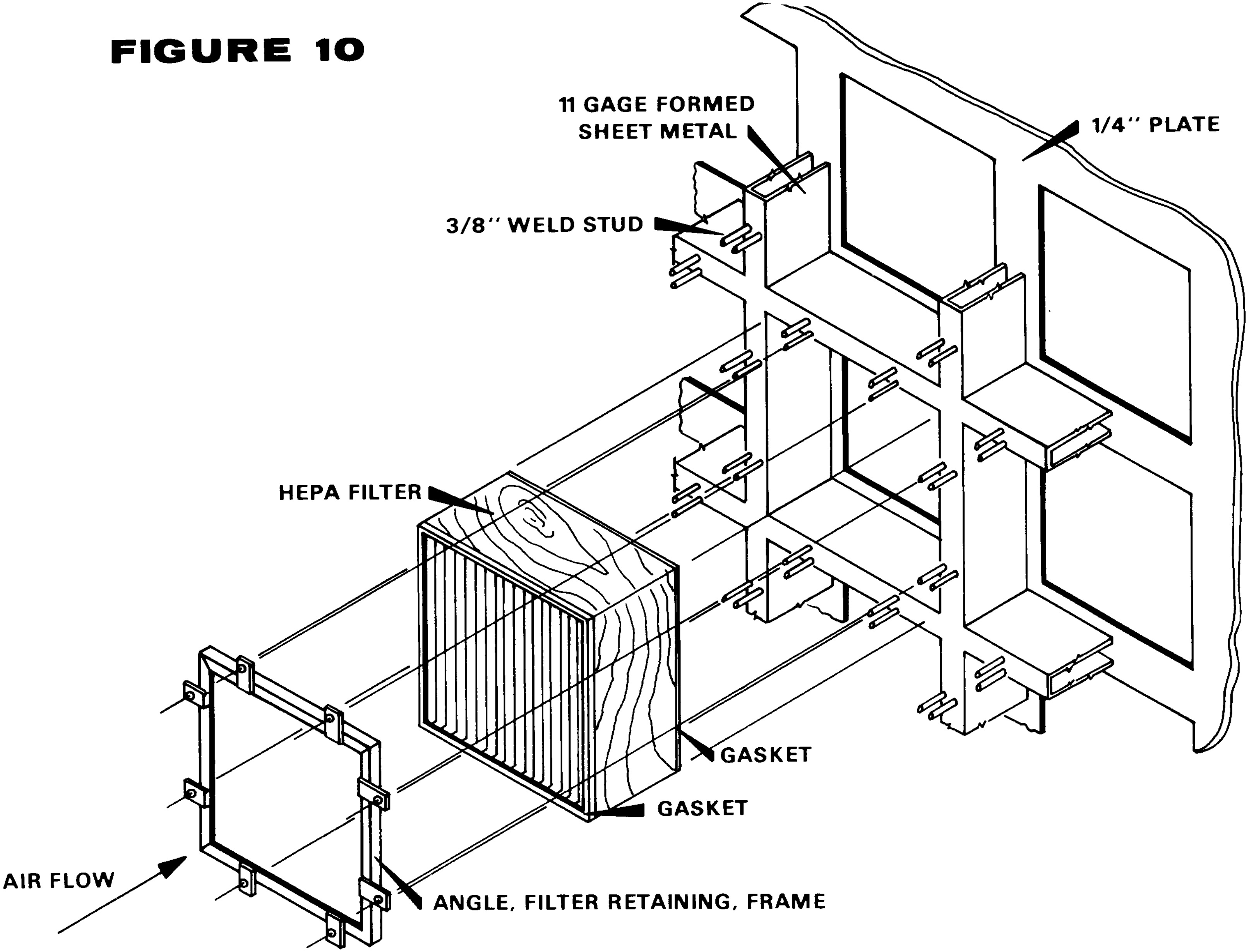
2. SCREENS ARE 1" MESH, 14 GAGE HARDWARE CLOTH

FIGURE 10


11 GAGE FORMED SHEET METAL
1/4" PLATE
3/8" WELD STUD
HEPA FILTER
GASKET
GASKET
AIR FLOW
ANGLE, FILTER RETAINING, FRAME

HEPA assemblies of a maximum of 20 units each are most desirable for most nuclear plant exhaust systems. A maximum of 1000 cubic feet of air per minute for each 24 x 24 x 11-1/2 inch unit is recommended for any HEPA filter bank configuration.

The foregoing description has been directed toward a single exhaust and a single supply ventilation system. In reality, it is advisable to install an exhaust and supply ventilation system for each building area. These areas may be determined by building configuration, radiation levels, processes, or a combination of all of these factors. A single supply and exhaust ventilation system is not recommended.

All discussion or description of detailed automatic control methods, aerodynamics, materials of construction, and construction details have been excluded from this description; but, along with good engineering judgment, they are most essential to all ventilation systems.

Conclusion

It is possible to design and construct, using present technology and equipment, a filtration system for exhaust ventilation of plutonium recovery facilities or for any nuclear plant exhaust system. It is just a question of "how badly do you need it?"

VENTILATION SYSTEMS
AT MOUND LABORATORY "PP" BUILDING

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April 13, 1971

VENTILATION SYSTEMS
AT MOUND LABORATORY "PP" BUILDING

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I. INTRODUCTION

In this paper I will discuss the Plutonium Processing Building ventilation systems at Mound Laboratory and the safety measures taken to ensure the integrity of the ventilation systems, thereby preventing the release of radioactive material to the environment. The paper will be divided into three main topics:

1. The differential pressures between the ventilation systems used to restrict the radioactive contaminants from spreading through the building.

2. The high-efficiency air filtration system used throughout the building. This air filtration system complies with the latest safety requirements recommended for nuclear high-efficiency filtration systems.[1]

3. The fire protection for glove boxes. The design and installation of a Halon 1301 automatic fire detection and protection system.

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* Mound Laboratory is operated by Monsanto Research Corporation for the United States Atomic Energy Commission under Contract Number AT-33-1-GEN-53.

[1] C. A. Burchsted and A. B. Fuller, Design, Construction, and Testing of High-Efficiency Air Filtration Systems for Nuclear Application, ORNL-NSIC-65 (January 1970).

II. VENTILATION IN THE PLUTONIUM PROCESSING BUILIDNG

The outside air for the ventilation system is pulled down through a face bypass damper and preheat coils into an air tunnel that services all the supply plenums for the building and ventilation systems (Figure 1). There are four air supply plenums in the building:

1. Access corridor plenum
2. Laboratory air plenum
3. Office area plenum
4. Cold area plenum

The four supply systems are basically the same with a bank of air conditioning filters upstream from the blower. Downstream from the blower, there is a back-draft damper, chilled-water cooling coil, and reheat heat coils. The design, installation, and construction of the supply ventilation system are the same as for any ventilation system.

The cold area supply and exhaust ventilation is a standard system. The only variation in the cold area system from any other ventilation system is that it connects to the laboratory exhaust system downstream and goes through the laboratory high-efficiency air filtration plenum.

The office area ventilation system services the offices and corridors of the building. The office rooms and corridors are maintained at atmospheric pressure (see Figure 2). The laboratories are maintained at a -.05 negative pressure by a control damper with a high-pressure sensing tap tieing into the office area and the low-pressure sensing tap located in the exhaust duct upstream from the damper.

Each laboratory room exhaust duct has a control damper. The ventilation system in the access corridors is maintained at a -.10 negative pressure by a control damper with the low-pressure tap tieing into the exhaust duct upstream from the control damper and the high pressure tap tieing into the laboratory room. The access corridor is the area in the back of the glove box. The laboratory is the area in front of the glove box or on the working side of the glove box. A metal wall divides the back side (the maintenance side) of the glove boxes from the work side (laboratory side) of the glove boxes.

The glove boxes receive their supply of air from the access corridor (see Figure 3). They have their own exhaust systems. The glove boxes are maintained at a -.65 negative pressure by a control damper in the exhaust system with the high pressure sensing tap tieing into the access corridor and the low pressure

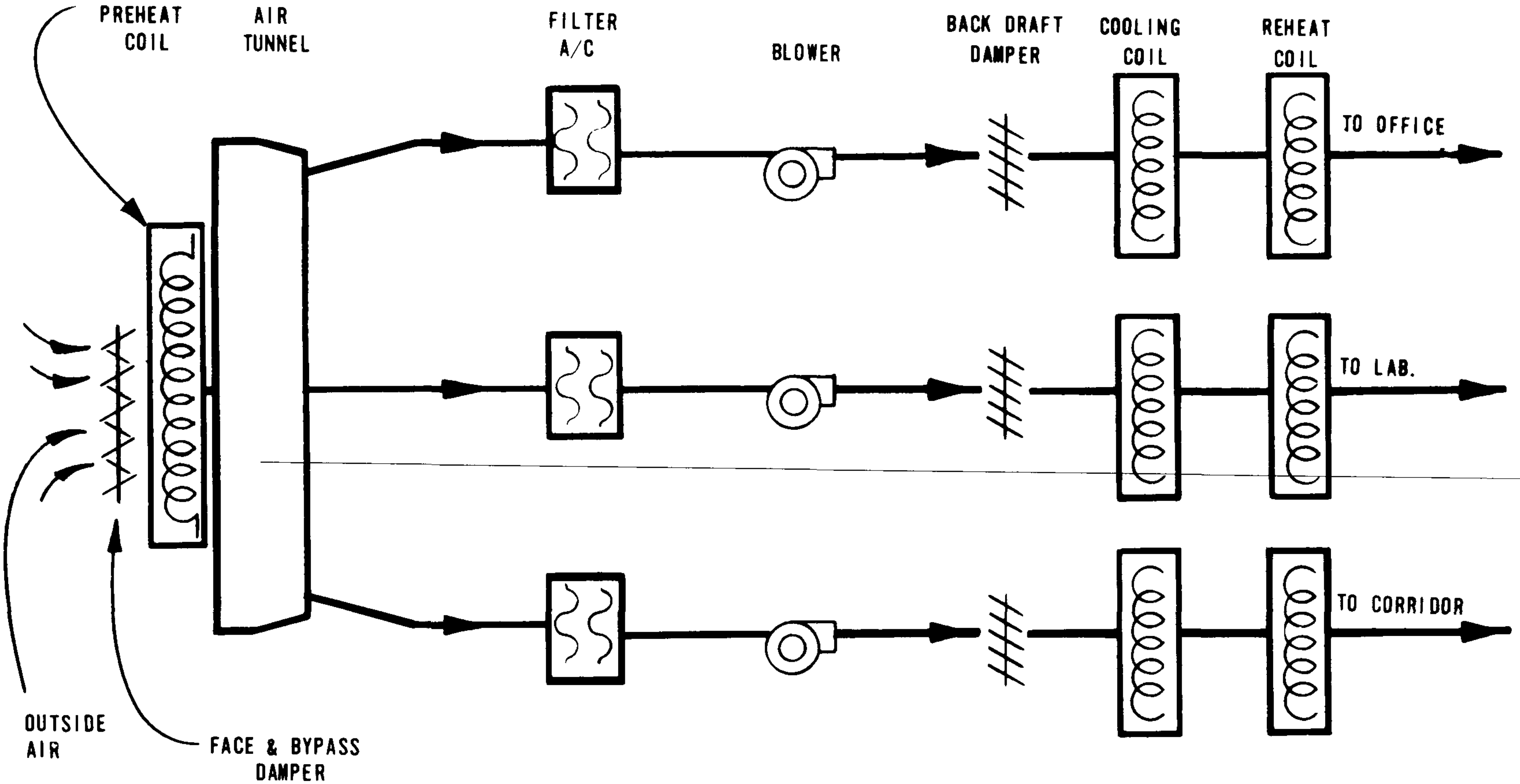

Figure 1 - Ventilation System, Plutonium Processing Building, Supply System.

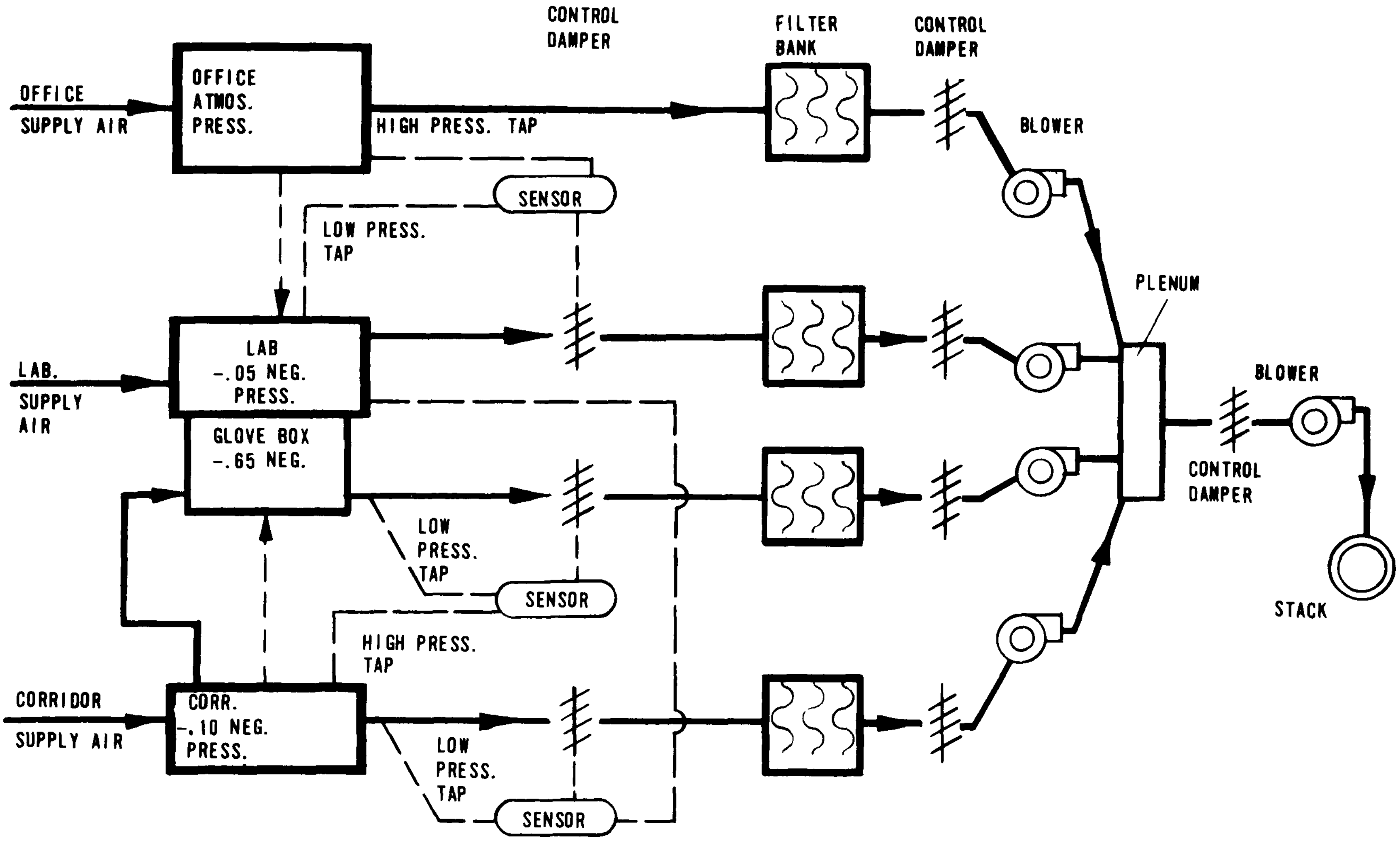

Figure 2 - Ventilation System, Plutonium Processing Building, Exhaust System.

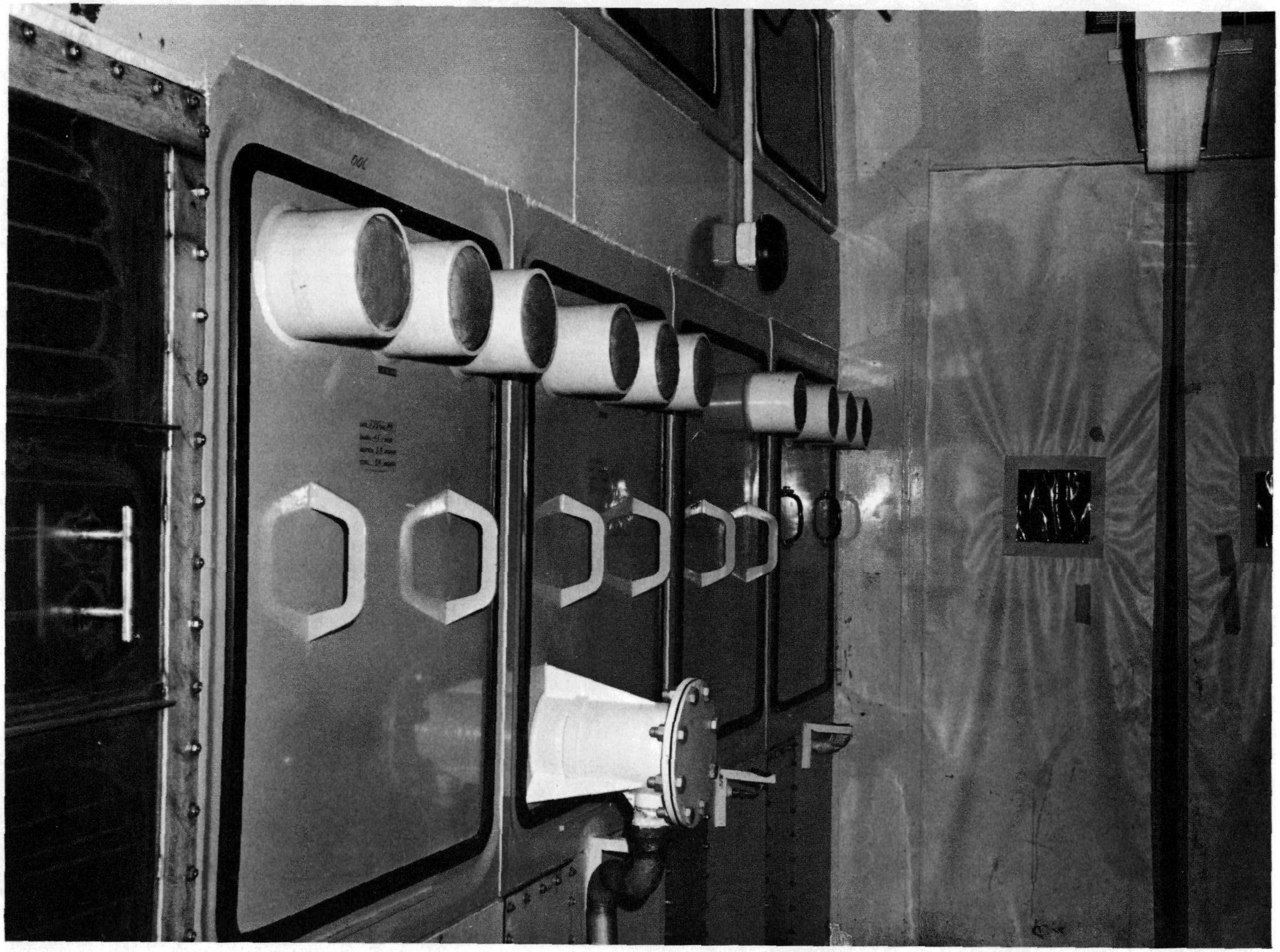

Figure 3 - Access corridor - glove box supply air from access corridor.

sensing tap tieing into the exhaust duct upstream from the control damper.

The system is designed so that all the air in the building will flow toward the next lower pressure area. With a pressure differential between offices and laboratories, air will flow toward the lower pressure area which is the laboratories. With a pressure differential between laboratories and the access corridor, the air will flow toward the lower pressure area which is the access corridor. With the glove boxes being lower in pressure with a -.65 negative pressure to the surrounding area, which is the laboratory on the front side and access corridor on the back side, the air will flow toward the glove boxes. Thus radioactive contamination will follow the air flow and stay in the glove boxes under normal working conditions.

The duct work in the exhaust systems is stainless steel 304L. The four exhaust systems in the Plutonium Processing Building have HEPA (High Efficiency Particulate Air) filter plenums and another control damper and blower prior to exhausting into the stack. The control damper arrangement is the same except the high pressure sensing tap is to the atmosphere.

This is a general description of the overall ventilation system in the Plutonium Processing Building. There are several smaller systems within the larger systems; but the principle of pressure differential is the same, and all the air is exhausted out one of the four HEPA filter plenums.

III. HIGH EFFICIENCY AIR FILTRATION

As discussed before, the supply air for the glove boxes comes from the access corridor area (see Figure 4). The supply air entering the glove box passes through a prefilter mat and two HEPA filters. The HEPA filters are cylinders 6-in. long by 6 in. in diameter with 99.97% efficiency for 0.3 μm particles (see Figure 4).

The exhaust from the glove box has a mat prefilter with two cylindrical filters 6-in. long by 6 in. in diameter with 99.97% efficiency for 0.3 μm particles. The access corridor has a prefilter and one HEPA filter. The prefilter is a 6-in. vertical 24 x 24 x 6 in. thick fire-retardant filter. The HEPA filter is a 24 x 24 x 11½ in. thick asbestos separator with a 99.97% efficiency for 0.3 μm particles.

The HEPA filters in the exhaust ducts for the glove boxes and access corridor keep the duct work free from any combustible material that could propagate into the filter plenum and help keep the radioactive particles confined to the area. Each glove box has its own supply and exhaust filters.

The access corridor (which is the area on the back side of the glove boxes) is divided into rooms with curtain walls, as shown in

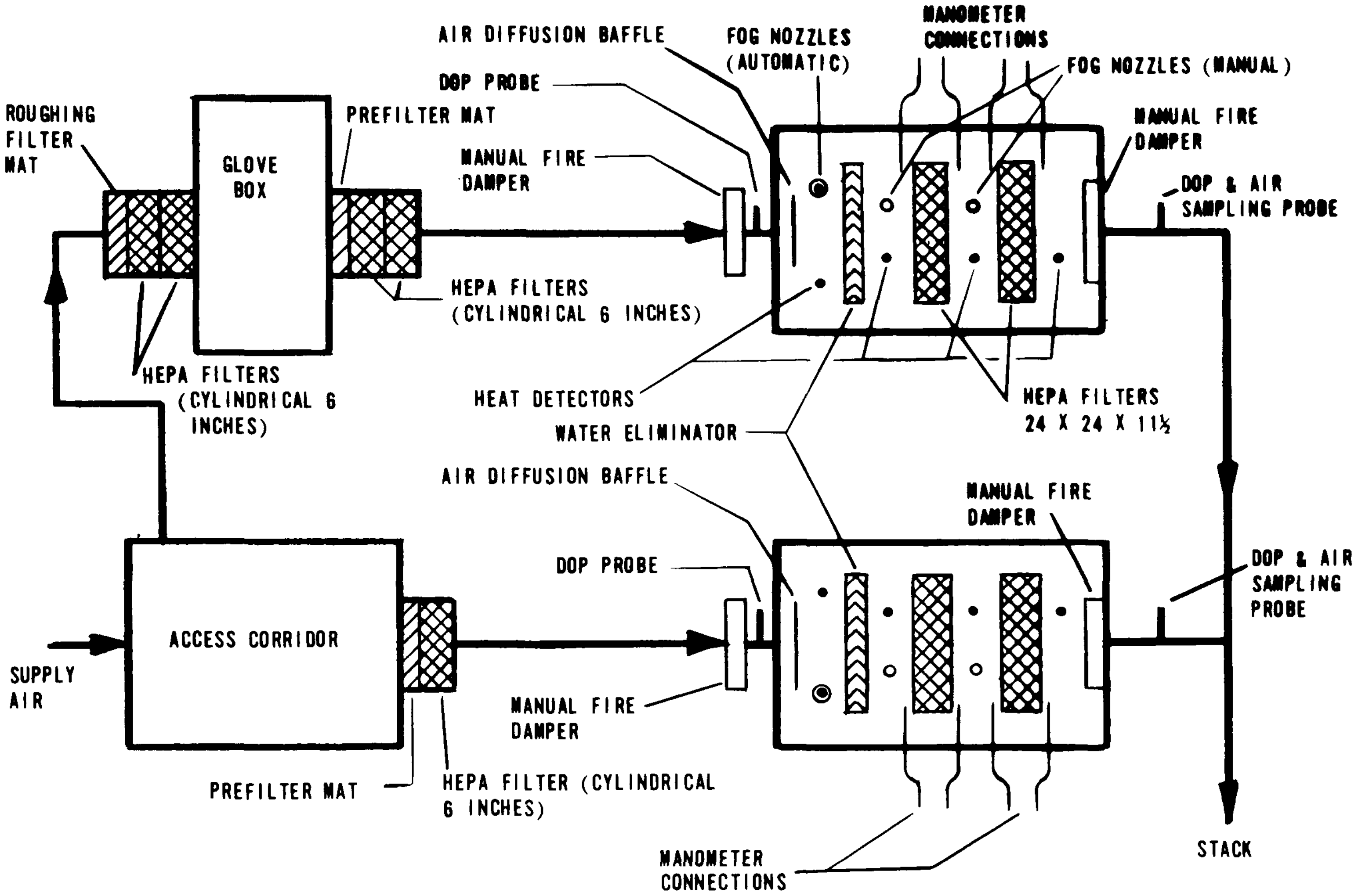

Figure 4 - Plutonium Processing Building High Efficiency Air Filtration System.

Figure 3. Thus maintenance or changes on glove boxes can be accomplished without getting the whole access corridor radioactively contaminated. The access corridors are approximately 110 ft long. Figure 3 also shows the supply ports into the glove boxes.

The original design of the filter plenum did not have a second bank of high-efficiency particulate air filters installed and did not include all the latest safety requirements for a high-efficiency air filtration system for nuclear application. A safety modification to the exhaust system in "PP" Building is now being installed and will have, when completed in June of this year, the following safety features:

- A manual fire damper installed in the duct in front of the plenum.

- An air diffusion baffle to prevent the inlet air from jet-streaming through the filter bank in a fire emergency.

- A water eliminator in front of the HEPA filter bank.

There are two banks of HEPA filters (24 x 24 x 11½ in. thick). The second HEPA filter bank added on the safety modification project is installed on the downstream side. In the glove box and the access corridor exhaust plenums, we do not have a prefilter. Although the glove boxes and the access corridor have prefilters and HEPA filters in the exhaust duct, we still wanted to put a prefilter in the plenum; but it was physically impossible to do so within the space. Provision for introducing filter testing aerosol, dioctyl phthalate (DOP) into the duct is located upstream from the filter bank a sufficient distance to promote thorough mixing for representative sampling of the aerosol reaching the filters. A fixed probe for sampling the representative filter testing is located at a point immediately ahead of the filter plenum. A second fixed probe for sampling DOP is located downstream from the filter bank in the exhaust duct.

Air samplers are located in every room, laboratory, corridor, and access corridor and are spaced so that the area is thoroughly covered. The air sampling probes are also located in the exhaust duct downstream from the blower and in the stack. All the air samples are taken daily and in some areas more frequently.

An automatic fog nozzle is located upstream from the water eliminator. The automatic water spray system is designed to supply 0.25 gal/min/ft^2 of filter face with all manual and automatic nozzles operating in one plenum. A heat detector, located in each chamber of the plenum, activates the deluge valve of the specialized fire protection pneumatic system (see Figure 5). In front of each HEPA filter bank is a manually operated fog nozzle. When the deluge valve of the specialized fire protection pneumatic system is activated, it sends a signal through the coded signal transmitter which will send a signal through the proprietary fire alarm

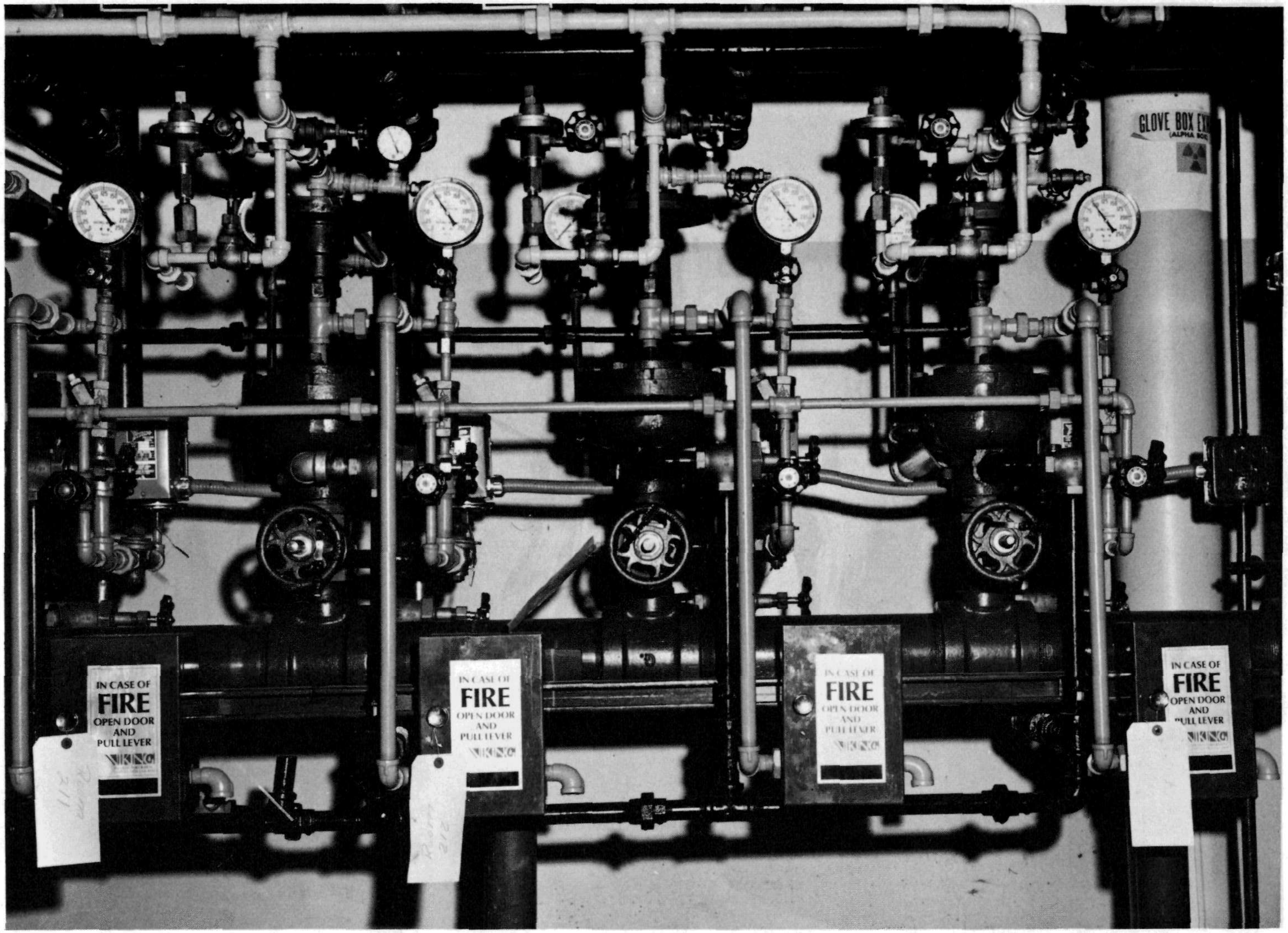

Figure 5 - The deluge valves of a typical specialized fire protection pneumatic system.

system to the central guard headquarters and the on-site fire department. An alarm bell is also activated in the area. The lab exhaust and office exhaust (Figure 6) are the same as the other two systems with the exception of a prefilter. The prefilter is installed in the exhaust plenum. There are no filters in the office or laboratory exhaust ducts where the air leaves the room.

IV. FIRE PROTECTION FOR GLOVE BOXES (HALON 1301 SYSTEM)

The need for fire protection in glove boxes has become more apparent in the last few years. At the present time, Mound Laboratory is installing an automatic fire protection system in all the air glove boxes in the Plutonium Processing Building.

The extinguishing agent is Halon 1301 (Bromotrifluoromethane). It is an odorless, colorless, electrically nonconductive gas that has proven to be an effective extinguishant for all classes of fires. Neither cooling nor oxygen exclusion satisfactorily explains the fire suppression action of Halon 1301. It is thought that Halon 1301 extinguishes by a chemical action. The halogen compound reacts with the transient combustion products which are responsible for rapid and violent flame propagation. This reaction terminates the combustion chain reaction and thereby stops the flame propagation.

Other important advantages of Halon are:

A. Effectiveness - An average concentration of less than 5% by volume of Halon 1301, as opposed to 35% by volume of CO_2, is required for extinguishing fires involving various hydrocarbons and commercial solvents.

B. Personnel Safety - Halon 1301 has exceptionally low toxicity. It is considerably safer than any of the vaporizing liquid fire extinguishing agents. Underwriters Laboratories classify Halon 1301 in Group 6 (least toxic).

C. Versatility - Halon 1301 is a safe and effective fire extinguishing agent for use on Class A (cellulosic material), Class B (flammable liquid), and Class C (electrical fires). It is equally effective for use in total flooding or local application systems when properly applied.

D. Cleanliness - Halon 1301 is a vaporizing agent. It leaves no residue, nor will it attack or react with normal materials of construction. It is particularly advantageous for use against fires involving delicate electrical, mechanical, or electronic equipment and for high value storage areas.

The glove box fire detection and protection system is designed around systems from several manufacturers which were modified to meet Mound Laboratory requirements (Figure 7).

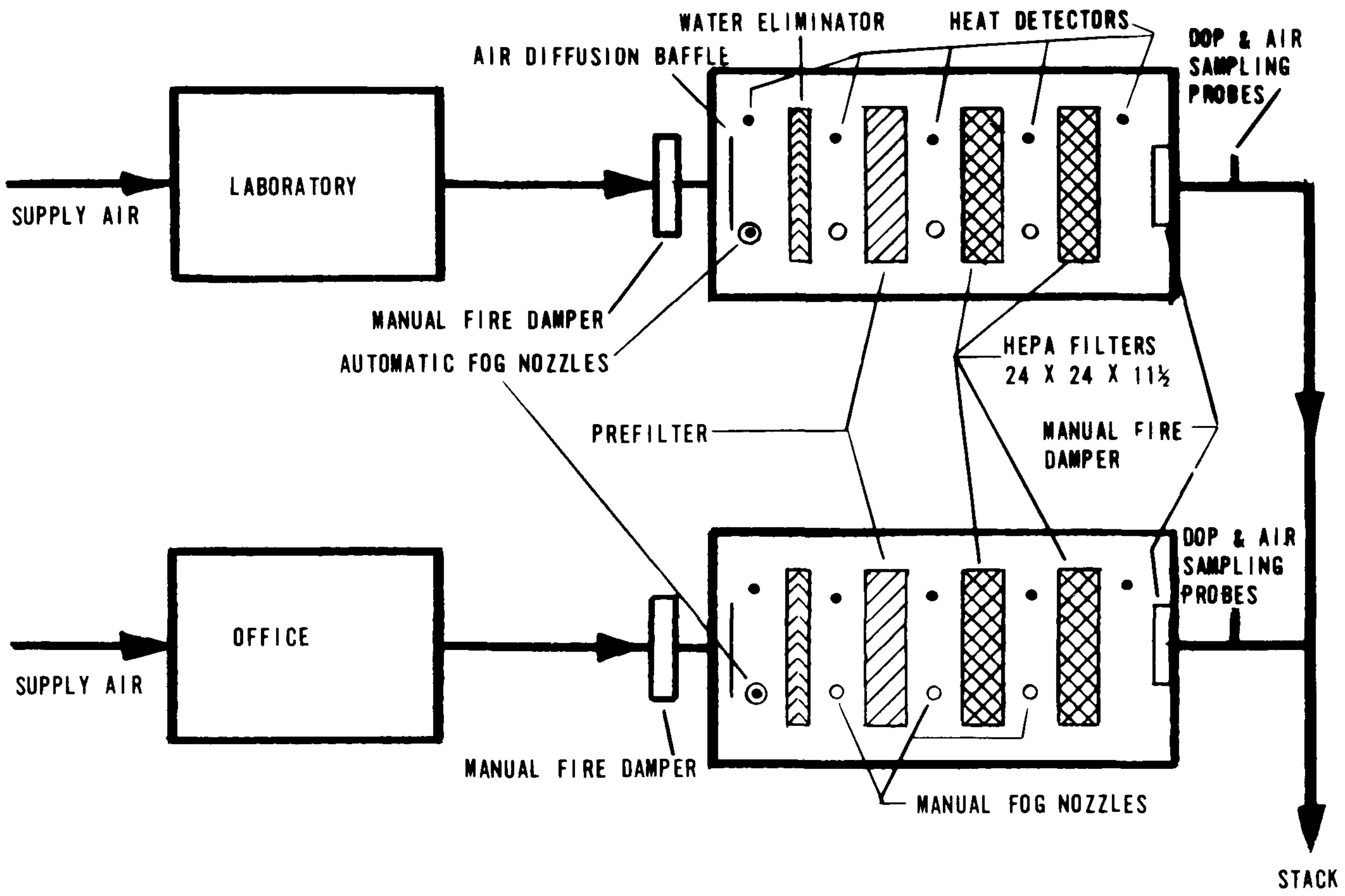

Figure 6 - Plutonium Processing Building, High Efficiency Air Filtration System.

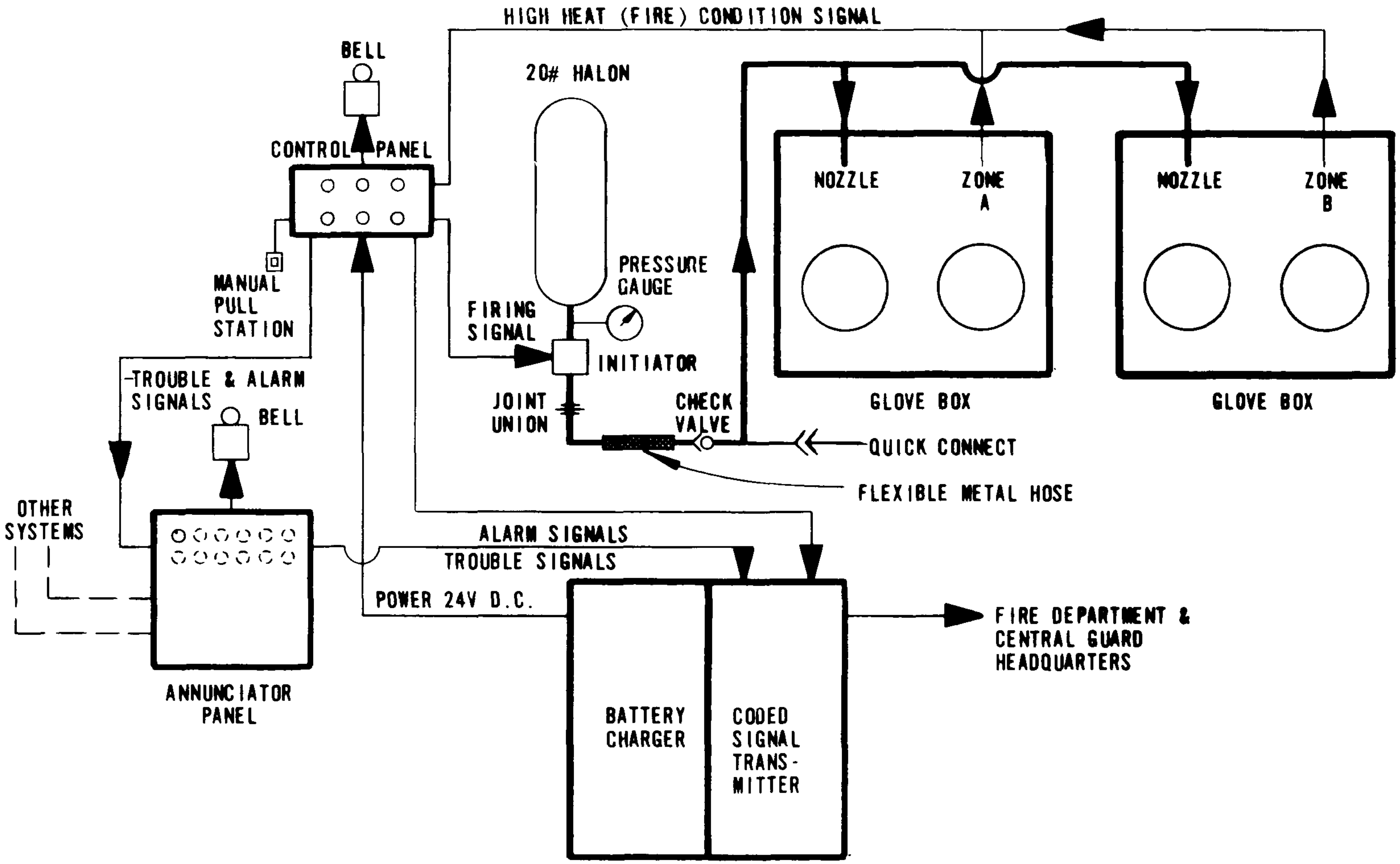

Figure 7 - Halon 1301 Automatic Fire Protection System for Glove Boxes.

The fire detection and protection system consists of the following:

- The heat sensor is a rate compensating, fixed temperature heat detector (140° F); normally contacts are open. The heat sensor selected is DETECT-A-FIRE manufactured by Fenwal Inc.; material: stainless steel. The gas nozzle is a one-piece design, non-clogging, hollow cone; material: stainless steel (type 303), Teflon, or hard rubber depending on the proces being performed in the glove box. A silicone rubber plastic or Viton cover will be installed over the nozzle and will be ejected when the gas is released.

- The agent storage container is a cylindrical vessel mounted with the valve down. The fill range of the agent storage container is 20 lb. The initiator cable assembly, mounted at the bottom of the storage container, receives the signal from the control panel and releases the agent. The agent in the storage container is under 360 psi pressure at 70° F. A quick-connect fitting is available in the line, so that if additional agent is required it can be quickly injected into the system.

- The control panel is mounted outside the laboratory room in the corridor (see Figure 8). The main functions of the control panel are to fire the initiator and to continually supervise all wiring, necessary components, and power. An alarm bell is located above the control panel which can be shut off by the fire protection personnel when the heat sensor contacts are open. There are five indicator lights on the control panel.

  1. Power-on light
  2. Alarm light
  3. Trouble light
  4. Zone A alarm
  5. Zone B alarm

  The trouble and alarm condition signals are transmitted to the central guard headquarters and on-site fire department. The trouble light will stay on until the initiator is replaced or the trouble has been corrected.

- The annunciator panel has all the control panels connected to it, with an indicator light for each control panel. It also has an alarm bell, a test button, and silence button. This annunciator panel is located in a corridor inside the main door, so that the fire protection personnel can readily locate the problem area. The test button is used to check the circuitry periodically to see that it is functioning properly. The silence button is used to turn off the bell at the annunciator panel. The alarm bell will sound for a fire, but not for a trouble signal.

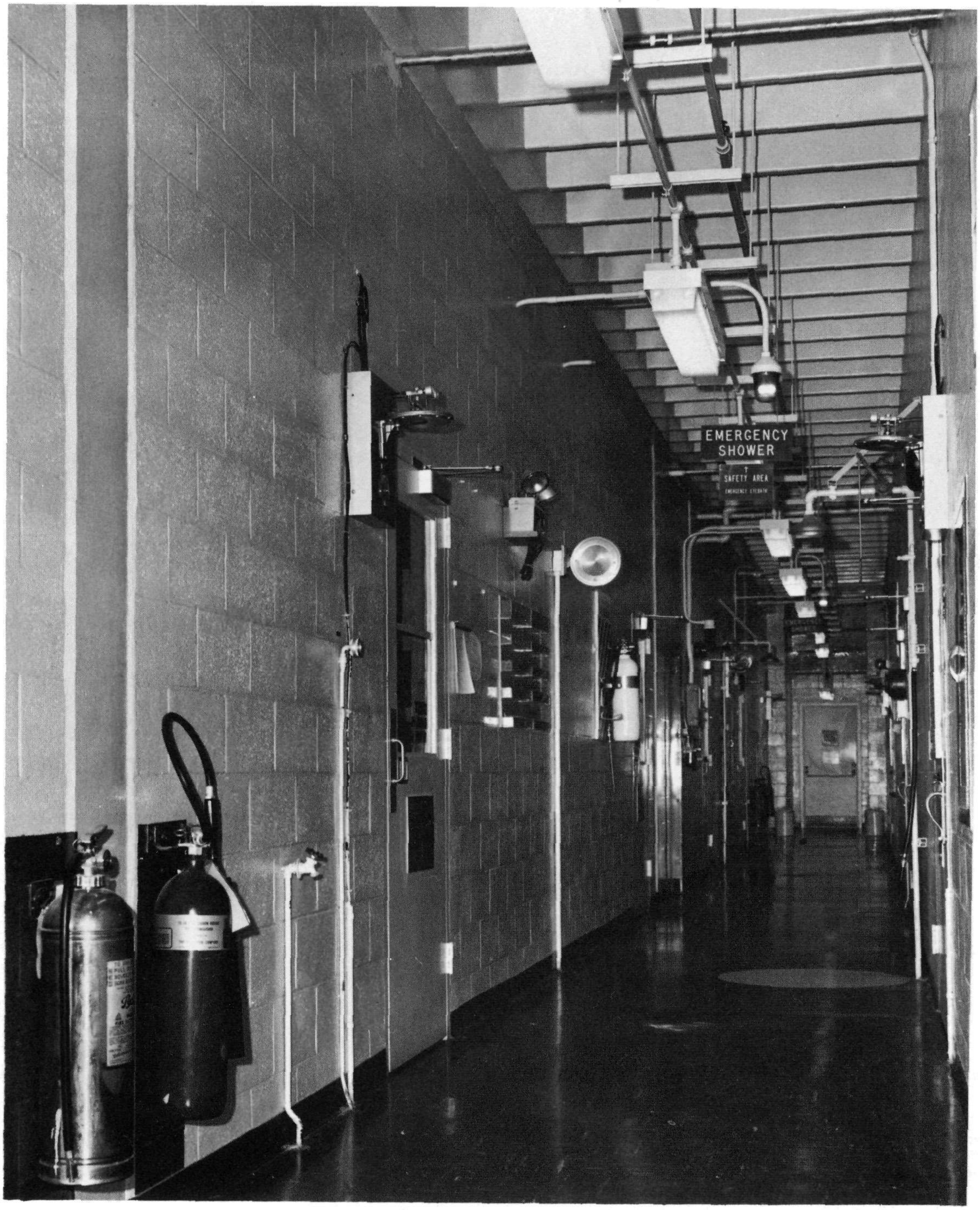

Figure 8 - Corridor between the laboratory showing the control dampers sensing mechanisms, emergency equipment, etc.

- The annunciator panel is connected to a coded signal transmitter which will send a signal through the proprietary fire alarm system to the central guard headquarters and the on-site fire department. The electrical system is 24 V DC with a central battery supply. The charger has the capacity to recharge the batteries for 24 hr while supplying the normal load. A battery supply system is used so that power is available during a power failure. The central battery supply was installed rather than an individual battery per circuit or per control panel so that maintenance of the equipment would be kept at a minimum. A battery charger is located in the battery panel to float the battery (keep the battery at full charge at all times).

The design criteria was that the Halon 1301 gas fire suppression system shall provide a minimum concentration of 5% (by volume) in each glove box or multiple of 27 ft^3, and this concentration shall be attained in less than 10 sec. The average air flow rate per glove box is 10 ft^3/min. The boxes normally operate at -0.5 to -1.0 in. of water column. During the discharge of the gas, the positive internal pressure in the glove box shall not exceed 2.0 in. of water column.

In order to test the integrity of the systems, a glove box line will be set up with a series of boxes simulating the largest line in the system. The cubic foot volume will be varied with the proper size nozzles and tested with all the variations needed to ensure that all systems will work within the design criteria set forth.

This project is scheduled to be completed the fourth quarter of fiscal 1972. This automatic and manual fire detection and extinguishing system will provide prompt detection and extinguishment at the exact location in the event of a fire in a glove box. This project, by correcting deficiencies in fire supervision and control, will provide better protection against fire and will minimize the possibility of spreading a fire and of releasing contamination resulting from a fire.

V. SUMMARY

The purpose of this paper is to supply information of the plutonium processing ventilation systems and the major safety projects in work at Mound Laboratory in the plutonium processing operation. Should an emergency situation develop, these improvements will assist in maintaining the integrity of the glove boxes and exhaust ventilation systems and thereby prevent the release of radioactive materials. Recent events at other AEC sites indicated that additional safety measures of this type should be incorporated without undue delay.

DIFFERENT TYPE GLOVEBOX FILTERS USED FOR PLUTONIUM-238 WORK AT MOUND LABORATORY

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April 13, 1971

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ABSTRACT

A review is presented of the different type glove box filters that have been used with plutonium-238 glove boxes at Mound Laboratory. Three filter installation types are discussed: (1) box-type filter mounted inside a small housing on top of glove box, (2) cylindrical filter with 2-in. diameter threaded pipe at one end designed to screw into exhaust line inside glove box, and (3) dual cylindrical filters with molded rubber gaskets designed to fit inside a stainless steel or fiber glass tube below glove box. Each type filter installation is discussed in terms of filtration adequacy, sources of leaks, ease of filter change, contamination control problems during filter change, and interference with glove box operations.

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*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Atomic Energy Commission under Contract No. AT-33-1-GEN-53.

INTRODUCTION

Mound Laboratory, Miamisburg, Ohio, has over 10 years' experience in large-scale glove-box operations with plutonium-238. The relatively high specific activity of this isotope, approximately 17 Ci/g, plus its high radiotoxicity have presented serious problems in containment and in the prevention of internal exposures to personnel.

One feature of glove-box design which is important, from the applied health physics point of view, is the primary glove-box filter installation at the exhaust of the glove box.

One of the early style primary filter installations used on plutonium-238 glove boxes at Mound Laboratory was a box-type housing mounted external to each glove box. An improved design for general use is a cylindrical filter housing mounted inside the glove box. A completely new design was placed into use when the Plutonium Processing Facility at Mound Laboratory went into operation in March, 1968. In this latest design two cylindrical filters in series are mounted in a stainless steel or fiber glass exhaust tube below the rear of the glove box.

In the following sections the different type primary filter installations used at Mound Laboratory are described and the major lessons learned by experience with each type are reviewed.

DESCRIPTION OF PRIMARY, GLOVE-BOX FILTER INSTALLATIONS

A. Box-Type Housing, External to Glove Box

An example of the box-type filter installation external to the glove box is shown in Figure 1. Four separate filter housings and part of a fifth may be seen in this photograph. Each filter housing serves the exhaust from one glove box. The housing has an upper and a lower chamber, each containing one absolute filter. This provides two filters in series at the exhaust of the glove box. There are four doors on each housing, two in the front and two in the rear. The rear doors are not visible in the photograph.

The absolute filter used in this installation is 8 in. x 8 in. x 5-7/8 in., gasketed on both faces. The filter is rated for 50 ft^3/min. The gaskets are compressed by sliding the filter into the chamber. The doors on the housing are gasketed and held shut by latches.

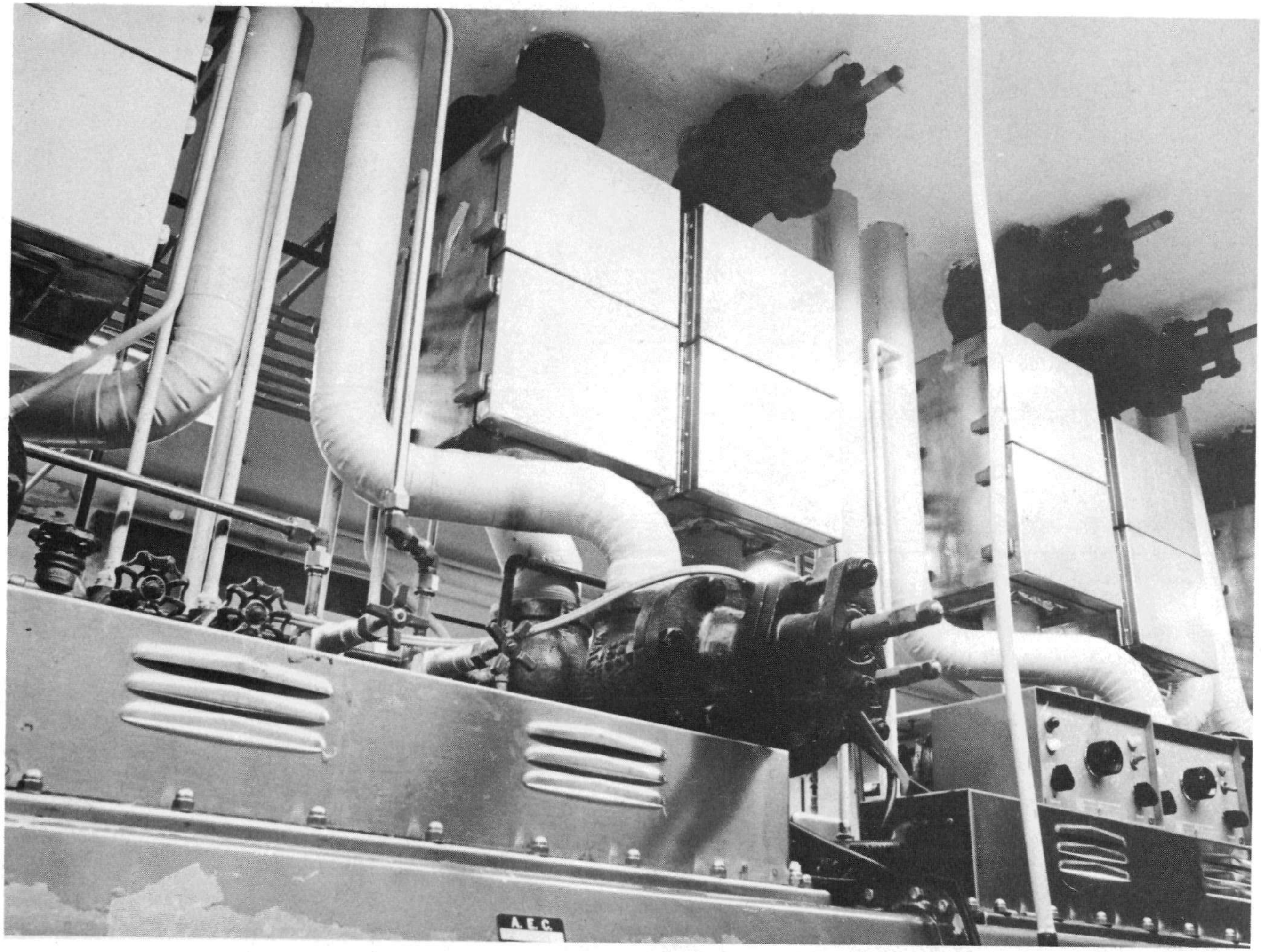

Figure 1 - Box-type housing, external to glove box.

B. Cylindrical Housing, Mounted Inside Glove Box

An example of the cylindrical filter housing which is mounted inside the glove box is shown in Figure 2. The filter housing is a steel canister 6 5/8 in. in diameter and 9 1/2 in. long. The outlet end of the housing screws directly into the exhaust line of the glove box. The inlet end is fitted with a small holder containing a roughing filter.

Inside the canister are two absolute filters in series. Each internal filter is 6 1/2 in. in diameter and 2 13/16 in. long. The complete assembly is rated for 25 ft^3/min.

There is no gasket associated with this type installation. The seal is dependent upon screwing the threaded pipe tightly into the exhaust line. Teflon tape is normally applied to the threads; however, care must be taken to minimize the amount of tape which will be in contact with high level contamination because (α, n) reactions with the fluorine in the Teflon can result in a significant neutron yield.

C. Dual Cylindrical Filters, Installed in Exhaust Tube

The newest type of primary glove box filter installation at Mound Laboratory is shown in Figures 3 and 4. Figure 3 is a photograph of the rear of a glove box in the new Plutonium Processing Facility, showing the access to the filter tube. Figure 4 shows a closeup of the top of the filter tube with the upper filter in position. The screen, coarse fiber material, and cap visible in this picture are placed over the top of the filter tube to serve as a roughing filter and to protect the face of the upper filter. Figure 5 shows one of the two absolute filters which are positioned in the tube, one behind the other. The filter itself is 5 1/4 in. in diameter and 6 in. long. The gasket around the top of the filter is a dual lip made of molded silicone rubber which is compressed when the filter is inserted into the exhaust tube. Three wooden spacers are attached near the bottom of the filter to keep the filter positioned straight inside the tube. The filter is rated at 30 ft^3/min.

EXPERIENCE WITH DIFFERENT TYPE GLOVE BOX FILTER INSTALLATIONS

A. Box-Type Housing, External to Glove Box

1. Filtration This type filter installation has proven very satisfactory in filtering the air exhausted from a glove box. Although such installations have never been tested "in place" by the D.O.P. technique, air samples collected downstream from the filters

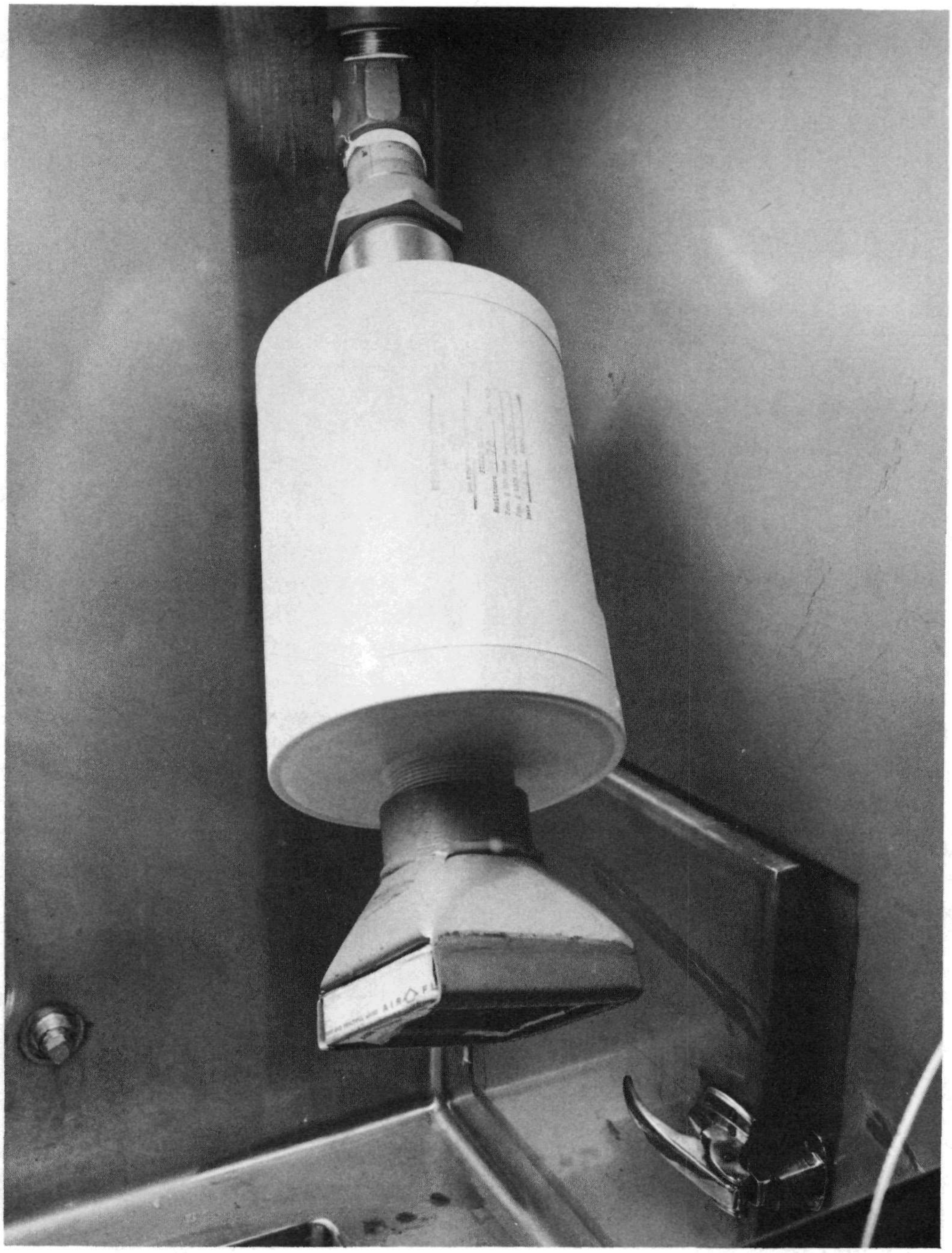

Figure 2 - Cylindrical housing, mounted inside glove box.

Figure 3 - New, primary filter installation. Rear of glove box showing access to filter tube.

Figure 4 - New, primary filter installation. Top of filter tube with upper filter in position.

Figure 5 - One of the two absolute filters which are positioned in the tube.

have confirmed that the installation does clean the airstream with a high degree of efficiency.

2. Source of Leaks If the exhaust air from the glove box contains acid fumes, corrosion of the metal filter housing may occur over a period of time. In some installations there may be enough damage sustained from acid corrosion that leaks develop around the gasket seal.

3. Ease of Filter Change As can be observed from Figure 1, access to these filter installations above the glove boxes for a filter change is awkward. Personnel changing filters must work on ladders and reach out to the filter housing. In some laboratories, the space above the glove boxes is even more cluttered with pipe lines and equipment than is evident in Figure 1. Some difficulty has been experienced in pushing the old filter out during the change procedure, but this is not normally the case.

4. Contamination Control During Filter Change Large plastic bags are sealed over the front and rear doors to permit bagging both the old filter out and the new filter in. Because of the awkward working conditions and the possibility of high level contamination being present on the lower filter, changing these filters can be a major contamination control problem. The fact that the filter housings are located out in the open laboratory could give some difficulties, since a large area is subject to contamination if anything goes wrong.

   The contamination control problem during filter changes was the main reason that Mound Laboratory largely abandoned this type filter installation on plutonium-238 glove boxes.

5. Interference With Glove-Box Operations The external type filter housing takes up no valuable space inside the glove box. It must be recognized, however, that normal operations in a glove-box line equipped with this type installation are interrupted if any contamination results from the filter change. As indicated above, releases of activity may occur during filter changes with this type installation and this, of course, results in some shutdown of glove-box operations during decontamination.

6. Other Considerations The processing of large quantities of plutonium-238 dioxide does pose an external radiation

control problem. The (α,n) yield of plutonium-238 dioxide is on the order of 4 x 10^4 n/sec/g. Some glove-box lines have to be shielded with several inches of a neutron shielding material. The accumulation of plutonium dioxide in the unshielded external filter housings along a box line raises the dose rate in the general work area.

B. Cylindrical Housing, Mounted Inside Glove Box

Because of the contamination control problems during filter changes with the box type filter housing, a cylindrical type housing mounted inside the glove box was adopted. A discussion of Mound's experience with this type filter installation is given below.

1. Filtration This type filter installation is very satisfactory, provided it is properly installed and does not leak. As in the case of the box-type housing, these cylindrical housings have never been tested "in place" using the D.O.P. technique, but air samples downstream from these installations have demonstrated that they can clean the air with great efficiency. Some trouble has been experienced with leaks in this type installation.

2. Source of Leaks With very few exceptions, the leak occurs at the pipe threads where the canister screws into the glove-box exhaust line. A leak at this point permits some of the glove-box atmosphere to be drawn directly into the exhaust line, bypassing the filters. The problem here is that the threaded fittings of the exhaust line become corroded and when the primary filters are changed, the new canister cannot be, or simply is not, screwed tightly enough into position to achieve a good seal. To correct this problem, the metal fitting of the exhaust line has been replaced in some cases with a PVC threaded fitting.

   Another source of leaks may be the corrosion of the canister itself. A hole in the canister after the last filter will also allow air to bypass the filters. In a few installations where this has been a problem, the regular canister has been replaced with one having a stainless steel or PVC casing. This has prevented corrosion holes in the canister itself, but makes recovery of plutonium dioxide from these canisters more difficult.

3. Ease of Filter Change As indicated, corrosion in exhaust line threads may make the physical act of changing the filter difficult, and this increases the chance that the canister will not be screwed in properly.

It should be mentioned that since this type installation takes up space inside the box, it is normally located in the rear or off to one side so as to interfere as little as possible with the normal use of the box. Because of their location inside the box, these canisters cannot be changed by using the main gloves and the box must be equipped with a set of upper glove ports in the viewing window.

4. Contamination Control During Filter Change All filter changes take place inside a glove box.

5. Interference with Glove Box Operations This type filter installation uses up about one cubic foot of valuable glove box volume. The change of filters in one glove box does not interfere with other glove-box operations in the laboratory.

6. Other Considerations A buildup of material on the filters does increase the dose rate at the operator's position; however, as these boxes are shielded, this problem is not too serious.

C. Dual Cylindrical Filters, Installed in Exhaust Tube

In order to eliminate the problem of leaks associated with the threaded type fitting and to retain the advantage of changing the filter inside the glove box, a new dual cylindrical filter and housing installation was developed. Our experience with this new system is described below.

1, Filtration Before the Plutonium Processing Facility became operational in March 1968, approximately 30% of the glove-box exhaust filter installations were selected at random and D.O.P. tested. With two exceptions, no penetration could be detected. This, of course, was with two absolute filters in series, each of which is rated at least 99.95% efficient. The lower limit of detectability in this D.O.P. test was 0.001% penetration. The two exceptions which showed some leakage were found to have defects in the exhaust tubes. These exhaust tubes were out-of-round so that the gaskets on the filters did not seat properly.

   Since the facility has become operational, air samples collected from the glove box exhaust system has confirmed that this type installation does provide excellent filtration.

2. Source of Leaks There have been a few instances where high level contamination has been noted in the chamber below the exhaust tube. In these cases, these glove boxes had been used for processes where a large quantity

of acid fumes were generated. The Plutonium Processing Facility does have a separate acid fume exhaust system which is designed to collect the fumes locally inside the box and prevent them from being carried into the main glove box exhaust. In these isolated cases, apparently large volumes of acid fumes were carried to the glove-box filter installation. It is not known how the material penetrated the filters: however, one possibility is that the fumes condensed above the filter gasket, and liquid migrated past the seals to the lower chamber.

3. Ease of Filter Change The changing of filters in this type installation is much easier than in either of the other two cases described earlier. Refer to Figure 3. As may be seen in this photograph, there is a cubical chamber directly below the exhaust tube. This chamber is opened by removing a bolted, gasketed plate. Once opened, a new filter is placed in and pushed upward into the exhaust tube. Normally, the filter is forced upward into the tube with the aid of a small jack. As the new filter is forced upward in the tube, the top filter is forced out into the box and the lower filter moves up to the top position.

4. Contamination Control During Filter Change The design of this installation lends itself to good contamination control. The filter change is made in a controlled access service corridor behind the glove-box line. The removal of the cover plate over the chamber below the exhaust tube is carefully monitored. If any contamination is detected, a small plastic enclosure with glove ports is used to complete the job. To date, over 90% of these filter changes have been made without any significant contamination being detected.

5. Interference With Glove-Box Operations The protrusion of the exhaust tube into the glove box and the need for room to push the dirty filter out during a change uses up perhaps one-half of a cubic foot of glove-box volume.

6. Other Considerations As in the case of the internal canister-type glove-box filter, accumulation of material on the filter does increase the dose rate at the operator's position, but the filters are inside the shielded box so that the problem is less serious than in the case of the external filters.

DESIGN OF FILTER PLENUM HEAT EXCHANGERS

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April 13, 1971

U. S. Atomic Energy Commission Contract AT(29-1)-1106

DESIGN OF FILTER PLENUM HEAT EXCHANGERS

ACKNOWLEDGMENT

The author wishes to acknowledge the efforts of M. A. Thompson, D. R. Cartwright, C. M.Johnson, and D. M. Peck for assistance in performing the experiments reported here.

The concept and design for the heat exchanger was submitted by the heating and ventilating section of the Dow Engineering Department under the direction of C. L. Amos.

INTRODUCTION

Fires occurring within glovebox systems develop exhaust air temperatures in the range of 1600 °F.[1,2] Since the spread of radioactive contamination is controlled by an air flow inward toward zones of higher contamination as shown in Figure 1. The air within gloveboxes must be exhausted through a filter system.

The extent to which a filter is damaged by high temperature air is shown by Table I.[3] Therefore, several fire protection systems have been proposed to protect the relatively fragile filters from heat damage. The major portion of these systems have consisted of water spray deluge systems, installed with the spray field impinging upon the filter media. However, two questions regarding the adequacy of these protection systems have arisen: Do these systems damage the filters or otherwise allow contamination to escape? Can sufficient cooling be supplied by spray systems?

To answer these questions, a fire test facility was constructed at the Rocky Flats Division of The Dow Chemical Company. This facility consists of a glovebox, protective building, and filter plenum shown in Figure 2. The construction of this facility has

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[1] Domning, W. E., Woodard, R. W., "Glovebox Fire Tests," RFP-1557, The Dow Chemical Company. 1970.

[2] Smith, S. E., Et.al., "Protection Against Fire Hazards in the Design of Filtered Ventilation Systems of Radioactive and Toxic Process Building, AWRD 0-24/65, UKAE, 1965.

[3] Burchstad, C. A., Fuller, A. B., "Design Construction and Testing of High Efficiency Air Filtration Systems for Nuclear Applications," ORNL NSIC-65, Oak Ridge National Laboratory, 1970.

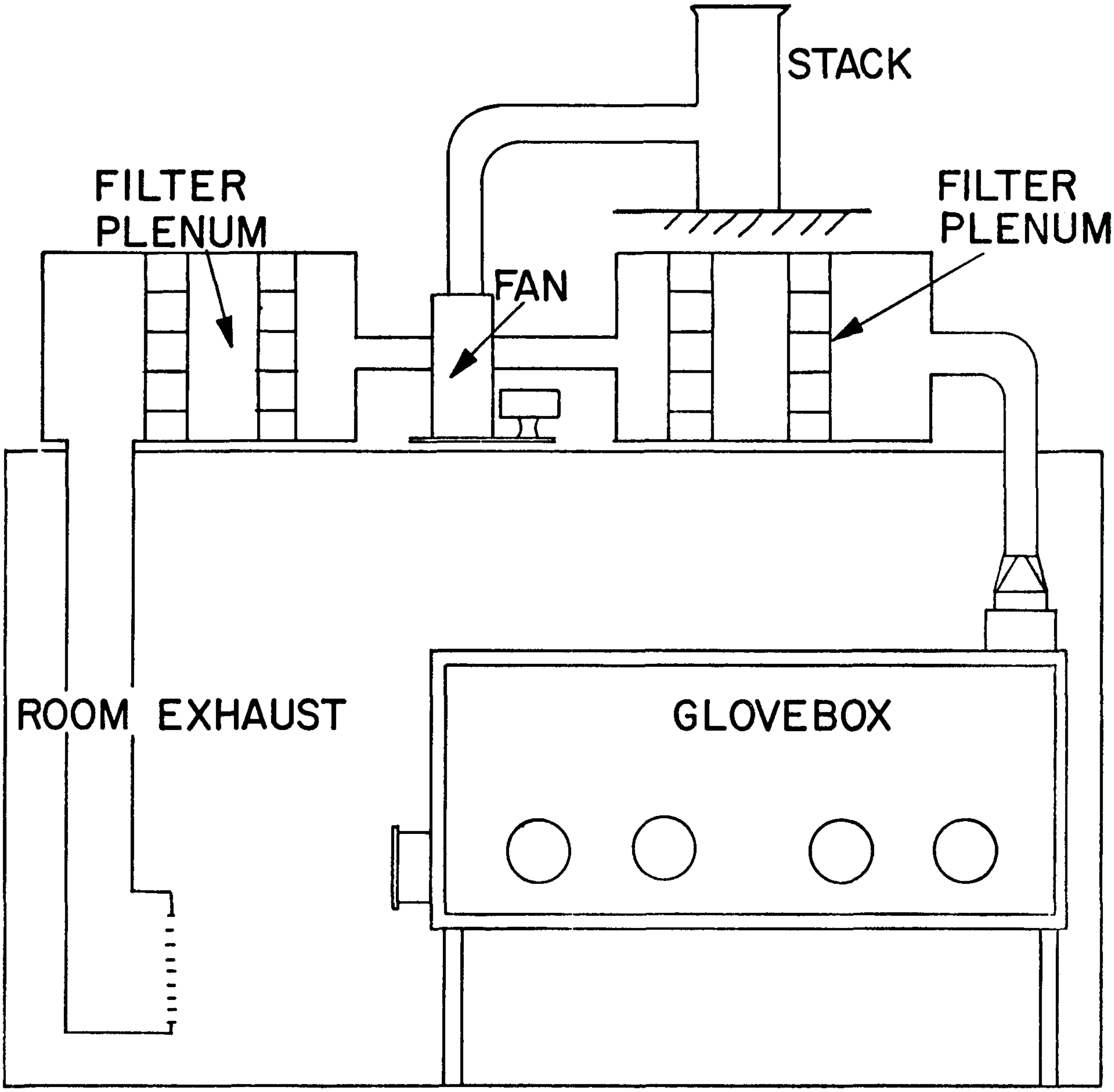

FIGURE 1
Building System

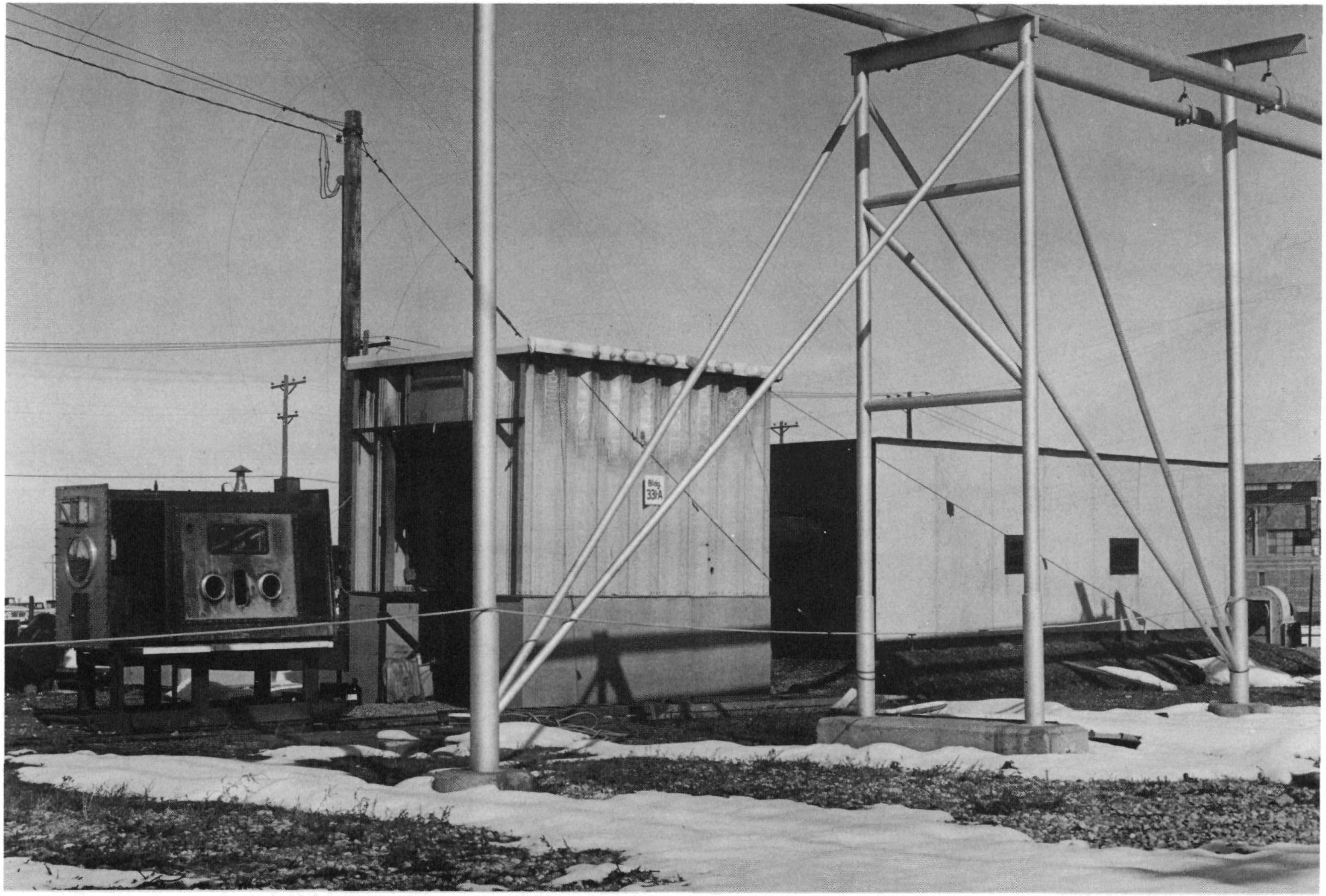

FIGURE 2

Fire Test Facility

been described elsewhere.[4] The purpose of this paper is to summarize the tests performed within the filter plenum of this facility.

TABLE I

RECOMMENDED LIMITING SERVICE TEMPERATURES
FOR WOOD-FRAMED FIRE-RESISTANT HEPA
FILTER UNITS

Frame Material	Temperature to Which Filter Was Exposed (°F)				
	Up to 10 min.	Up to 2 Hr.	Up to 48 Hr.	Up to 10 Days	Indefinitely
3/4-in.-thick Plywood	750	300	276	200	180

EXPERIMENTAL RESULTS

Initial experiments were performed to determine the water damage to a filter bank from a spray deluge system. The various spray heads shown in Figure 3 were mounted at various distances from the filter bank and tested at a water density of 0.25 gal/min-sq ft of the filter face. During these tests, there was approximately 6000 CFM of air flowing through the plenum system and the water impinged directly on the filter face. The results of these tests as shown by Figure 4 usually showed that one or more filters had been torn by the combined force of the water and air.

Examination of the flow distribution within the plenum showed a nonuniformity as illustrated in Figure 5. With the addition of a deflector, the flow became uniform.

Evaluation of a heat exchanger section within the plenum is illustrated in Figure 6. Cooling of the fire-heated air within this heat exchanger is done by a water spray section followed by a disengager or mist eliminator. The temperatures of consequence are shown as T_{G_1}, T_{G_2}, and T_i. In this heat exchanger, air flowing from the incinerator at a nominal rate of 6000 CFM, heated to temperature T_{G_1}, is contacted with a water spray. The gas is cooled to temperature T_{G_2} after passing through a moisture disengager.

The temperature T_i is the temperature of the gas-air interface as shown by a thermocouple located in the spray field at the upstream side of the disengager. Various combinations of deflectors and disengagers were evaluated.

The data, which will be used for calculations, consist of the average inlet and outlet temperatures and the average interface temperature. Experiments numbered 1, 2, and 3, in Table II were

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[4] Domning, W. E., "New Fire ProtectionSystems for Filter Plenums," CONF 700816, Harvard Air Cleaning Laboratory, 1970.

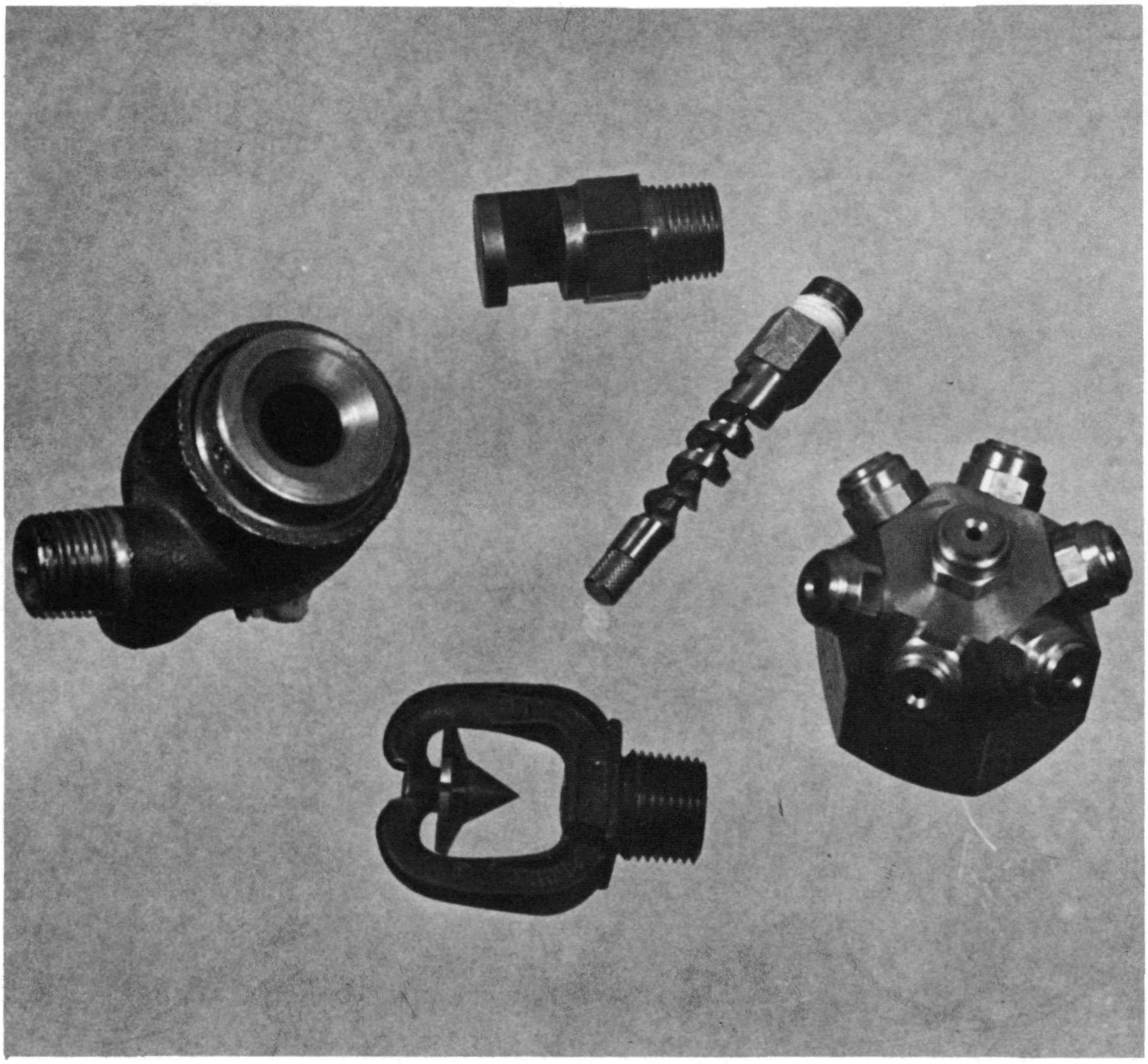

FIGURE 3

Nozzles Tested

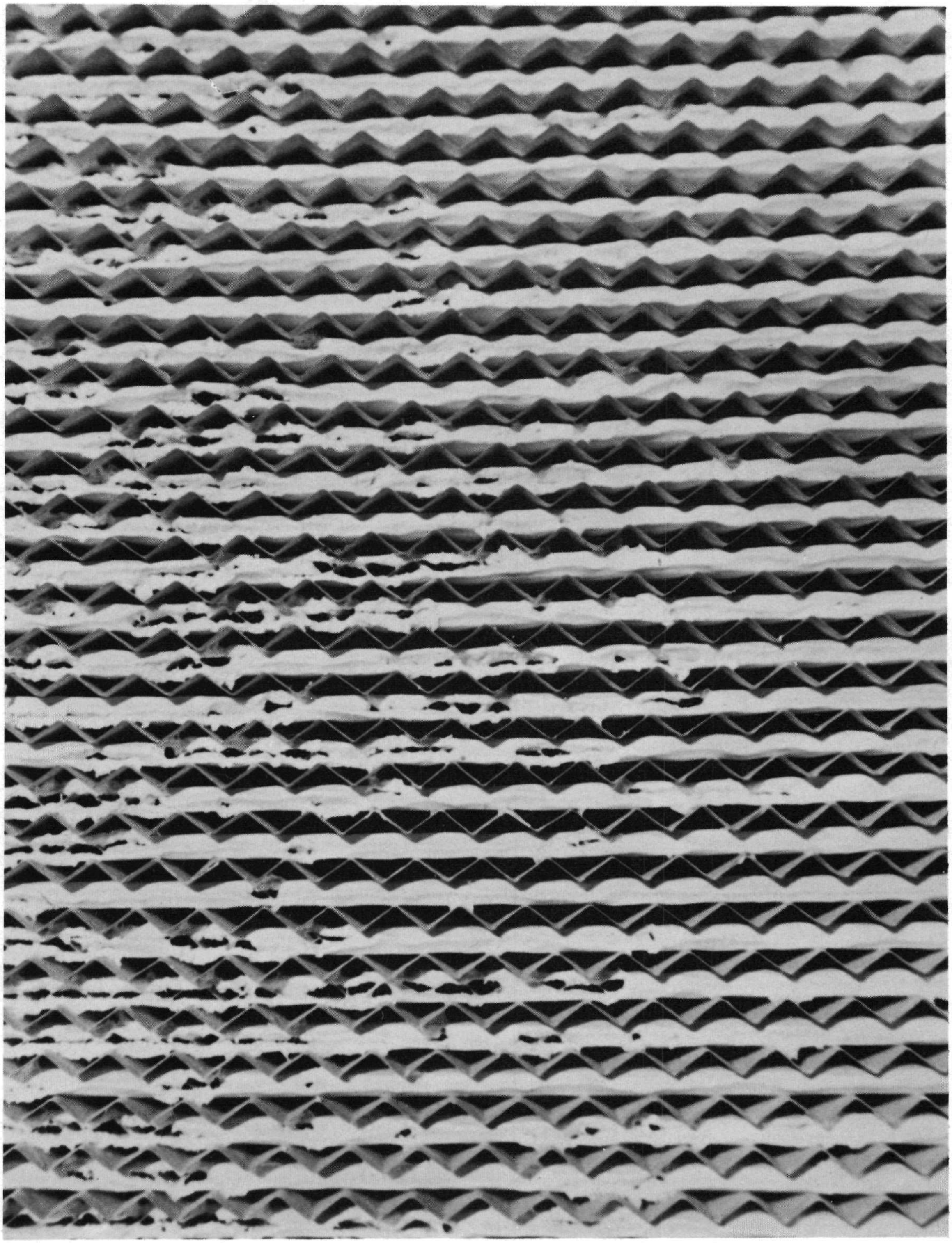

FIGURE 4

Torn Filter Media

110 440	100 480	100 540
175 400	2400 400	220 440
250 440	1200 460	195 440
110 420	120 420	100 400

Top - CFM without Deflector
Bottom - CFM with Deflector

FIGURE 5

Air Flow Distribution

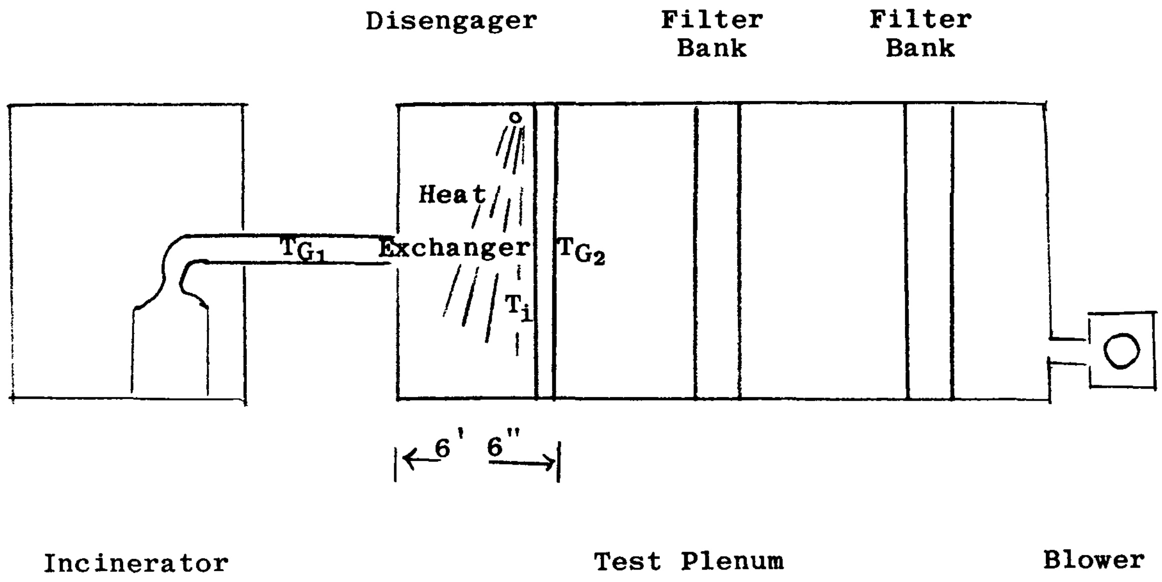

FIGURE 6

Test Schematic

those using a staggered channel iron baffle, shown in Figure 7, while Experiments 4 and 5 used a Farr mesh filter, shown in Figure 8.

TABLE II

DATA FOR HEAT TRANSFER CALCULATIONS

Experiment No.	Temperature °F		
	Inlet Air	Outlet Air	Interface
1	646	176	86
2	338	140	122
3	501	185	103
4	635	176	101
5	554	194	172
Average			116

DISCUSSION

The amount of heat transferred from a gas stream to a liquid is dependent upon the contact time between the two phases. By defining the air flow rate as a function of volume per unit area, it is possible to express the contact time as a length. A heat transfer unit is that length (contact time) required to transfer a specified quantity of heat. An equation for the heat transfer unit can then be developed from the usual heat transfer relationship $Q - hA\Delta T$

where: Q = amount of heat transferred

h = heat transfer coefficient

A = area of transfer

ΔT = temperature difference.

By assuming adiabatic conditions, the equation representing the exchange process can be simplified, and the dimensions of a heat exchange chamber can be specified for any inlet and outlet temperature.

The transfer of heat by direct contact of water and gas is typified by the equations used to design a cooling tower. For design purposes, the system has been considered as an adiabatically operated cooling tower. Based on an observed air temperature difference of 15°F for the plenum section downstream from the heat exchanger chamber, little heat is lost from the system by convection. In a normal adiabatic cooling tower, the water is recirculated, and the temperature of the water reaches an equilibrium value. This equilibrium water temperature is then representative of the interface temperature. In the experiments

FIGURE 7

Construction of
Channel Baffle

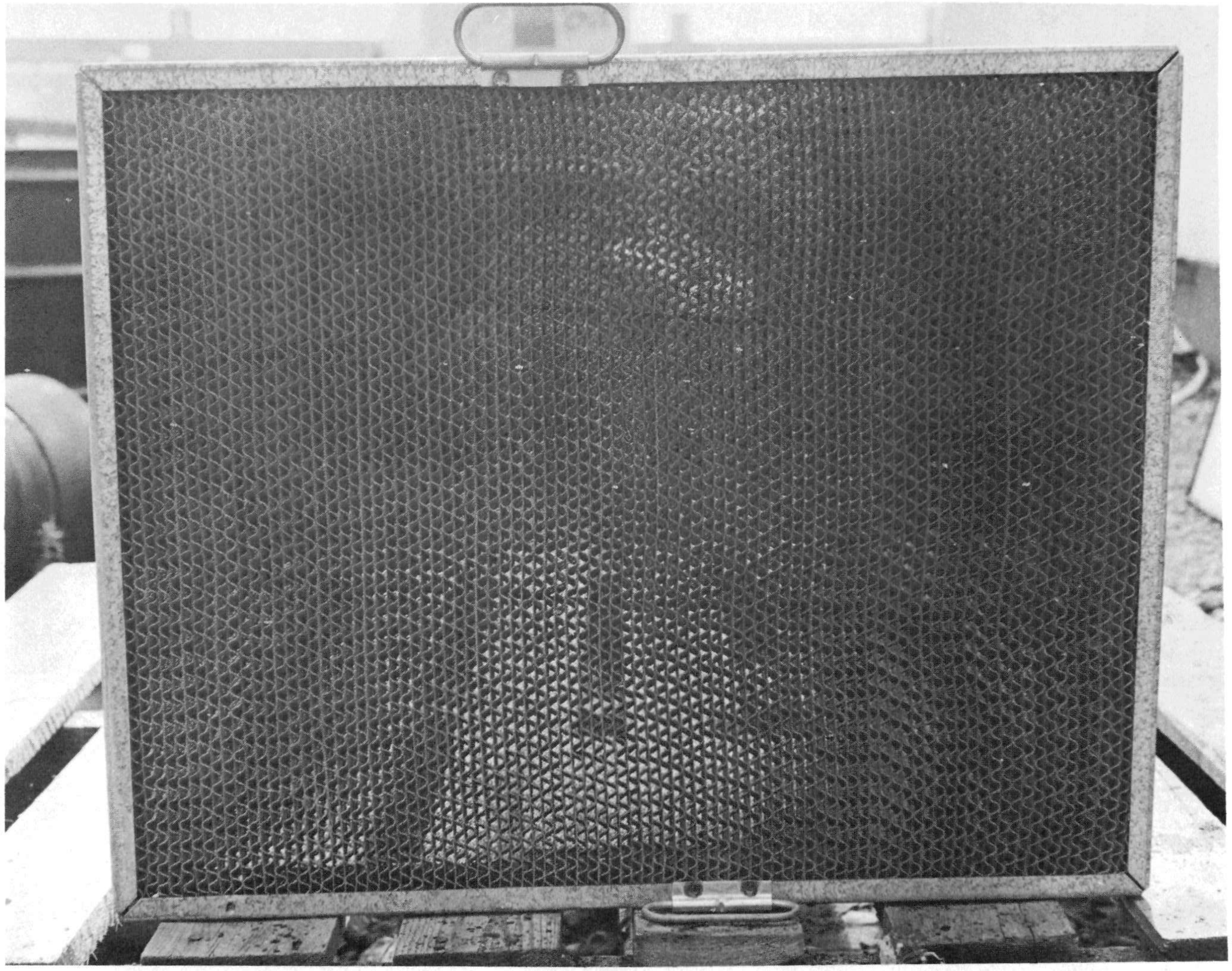

FIGURE 8

Farr Mesh Filter

performed, recirculation of the water did not occur and the interface temperature was obtained from thermocouples located in the spray field near the upstream face of the disengager. It is probable that, because of the need for a large critically safe tank to contain the water from the sprays, the design of these units will specify water recirculation and, therefore, the case for adiabatic design would apply.

It is possible that the interface temperature used for this calculation is low due to the nonrecirculation of the water. However, since several other arbitrary factors enter into the final performance of the heat exchanger, it is felt that the assumed interface temperature will not be the most uncertain assumption in the design.

There are five basic relationships that can be developed describing the overall transport phenomenon.[5] The equation applicable to adiabatic heat transfer is that describing the rate of transfer of sensible heat from the gas stream through the gas film to the air-water interface as follows:

$$W_G \; C_S \; dt_G = h_G \; a_H \; S_o \; dz(t_i - t_G) \qquad (1)$$

where: W_G = gas rate, pounds per hour

C_S = humid heat of gas Btu/(lb. gas)(deg. F)

t_G = bulk temperature gas stream, deg. F

h_G = mean coefficient of heat transfer Btu/(Hr.)(ft^2)(deg. F)

a_H = total transfer surface divided by volume of transfer unit

S_o = total cross section of transfer unit

z = height (length) of transfer unit

t_i = temperature of gas liquid interface.

Rearranging equation (1):

$$W_G \; C_S \int_1^2 \frac{dt}{(t_i - t_G)} = h_G \; a_H \; S_o \int_o^z dz \qquad (2)$$

Integrating (2) at adiabatic conditions:

$$\frac{h_G \; a_H \; S_o \; z}{W_G \; C_S} = \ln \frac{\Delta t_1}{\Delta t_2} \qquad (3)$$

---

[5] Mc Adams, W. H., "Heat Transmission," Third Edition McGraw-Hill Book Co., N.Y. 1954.

Defining a theroetical heat transfer unit as:

$$\frac{G_G C_S}{h_a a} = z_t \qquad (4)$$

where: G_G = the superficial mass velocity lb/hr ft^2

a = the total heat transfer area divided by the volume.

Combining equations (3) and (4) to obtain the ratio $\frac{z}{z_t}$, the relationship becomes:

$$\frac{z}{z_t} = \ln \frac{t_{G_1} - t_i}{t_{G_2} - t_i} \qquad (5)$$

Equation (5) can be used to find z_t since z is known to be 6.5 feet from Figure 6. The ratio $\frac{z}{z_t}$ can be used to find the downstream temperature corresponding to any upstream temperature (fire condition) for any heat exchanger distance.

Table III shows values for z_t and $\frac{z}{z_t}$ calculated by using equation (5). As noted earlier, there were two types of disengagers used in the experiments, and the average value of z_t for the staggered channel baffle was 3.42 feet, while the value of z_t for the mesh filter was found to be 2.80 feet. Therefore, the overall average value of 3.17 feet and the value of 2.80 feet for z_t will be used for determination of temperatures for assumed lengths.

Heat exchanger lengths of 6 feet, 3 feet, and 1.5 feet were assumed and temperatures of 1832°F, 932°F, and 572°F were chosen. The outlet gas temperatures were calculated using equation (5), and assuming the average value of 116°F for the interface temperature. The results of this calculation are shown in Table IV and Figures 9 and 10.

Equation (4) may be used to calculate the effect of a change in cross section or flow rate for any assumed length.

In order to optimize the design of the heat exchanger, the inlet and outlet temperatures must be known. In the case of a fire, they are usually not known as accurately as they would be for a process; therefore, suitable estimates must be made for these temperatures.

Studies have shown that some of the heat from a burning glovebox will be dissipated by convection through the exhaust duct walls. Figure 11 shows the temperature drop expected in air flowing in 12-inch diameter ducts at 1000 ft^3/min from burning gloveboxes.[6]

Several curves are given in the original report; however, the most realistic curve is that given for steady state heat transfer assuming heat is lost by natural convection and radiation. The curve has been replotted to reflect a starting temperature of 1590°F. The mass rate of flow for the above study is approximately equal to the average flow existing in ducting. To be conservative, assume 50 feet of duct between the glovebox exhaust and the heat exchanger inlet. Using Figure 11, the inlet temperature would be 1260°F.

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[6] Op. cit.[2]

TABLE III

LENGTH OF HEAT TRANSFER UNIT

Experiment No.		z_t (ft.)	$\frac{z}{z_t}$
1		3.55	1.83
2.		2.61	2.49
3		4.10	1.85
4		2.28	2.85
5		3.32	1.96
	Average	3.17	

TABLE IV

EXIT GAS TEMPERATURE AS A FUNCTION OF EXCHANGER LENGTH, z

z_t	Temperature °F	Outlet Air Temperature °F		
		1.5 ft.	3 ft.	6 ft.
	1832	1187	779	374
317	932	624	427	253
	572	400	293	184
	1832	1120	703	318
2.80	932	594	396	212
	572	382	272	170

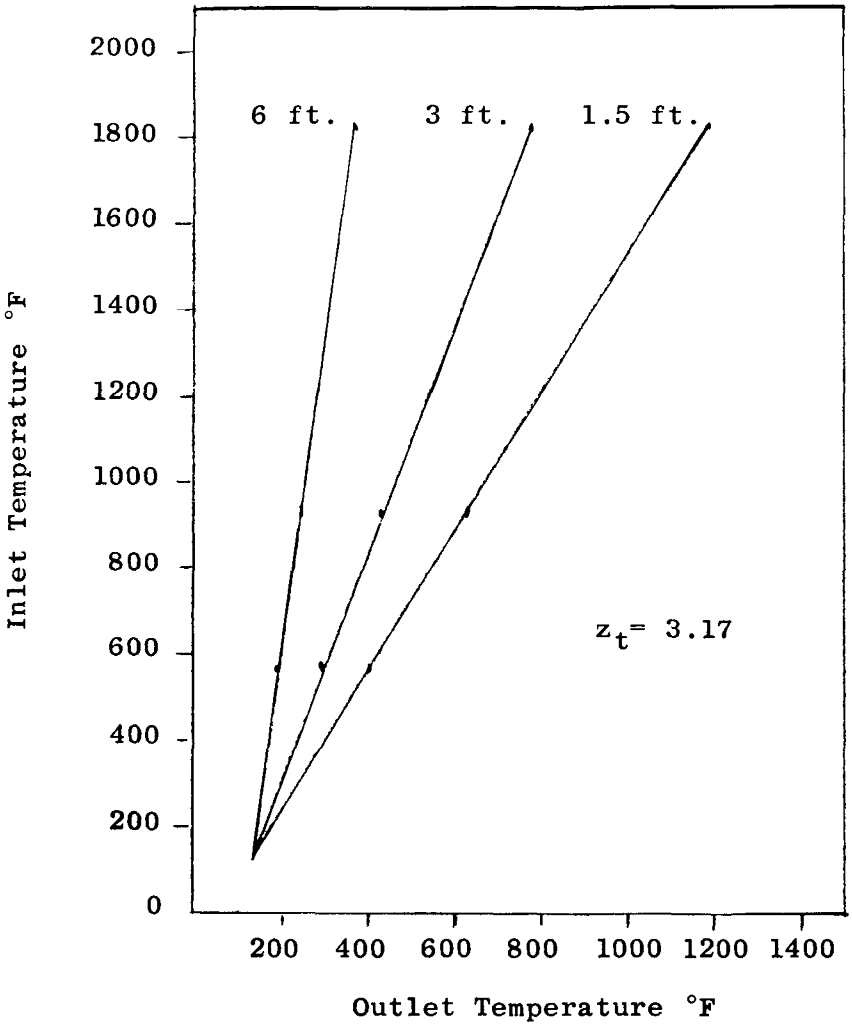

FIGURE 9

Exit Gas Temperature as a Function of Distance, z

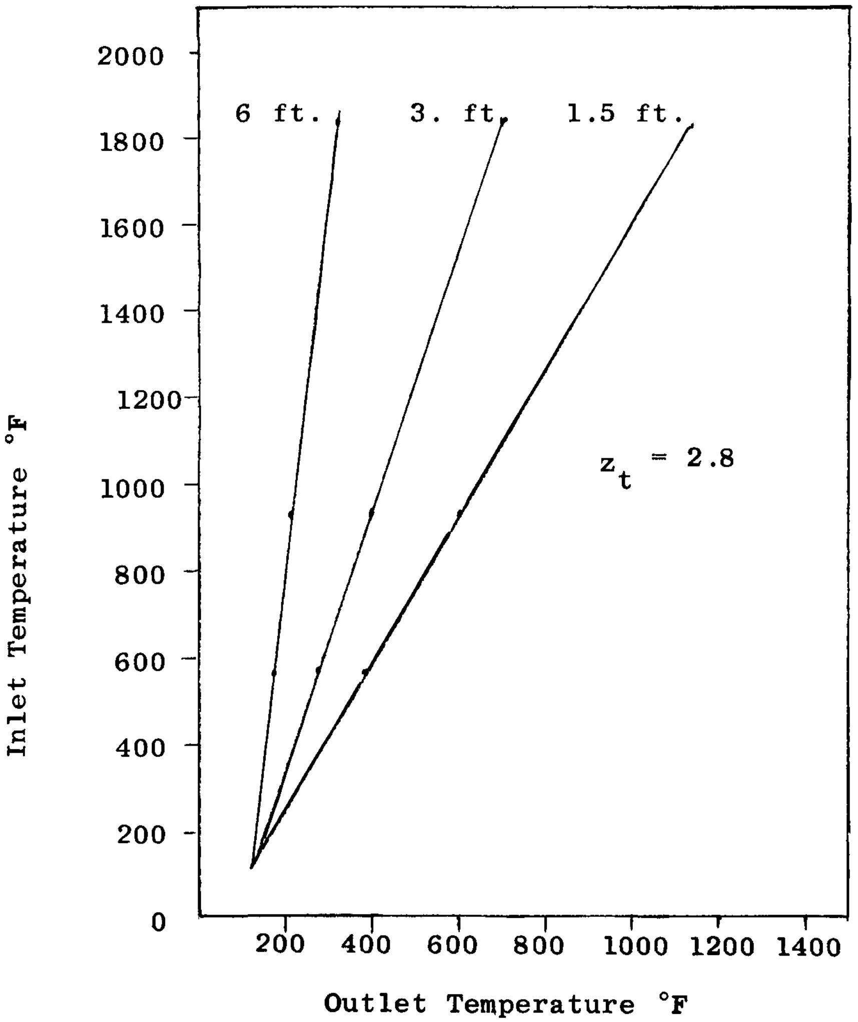

FIGURE 10

Exit Gas Temperature as a Function of Distance, z

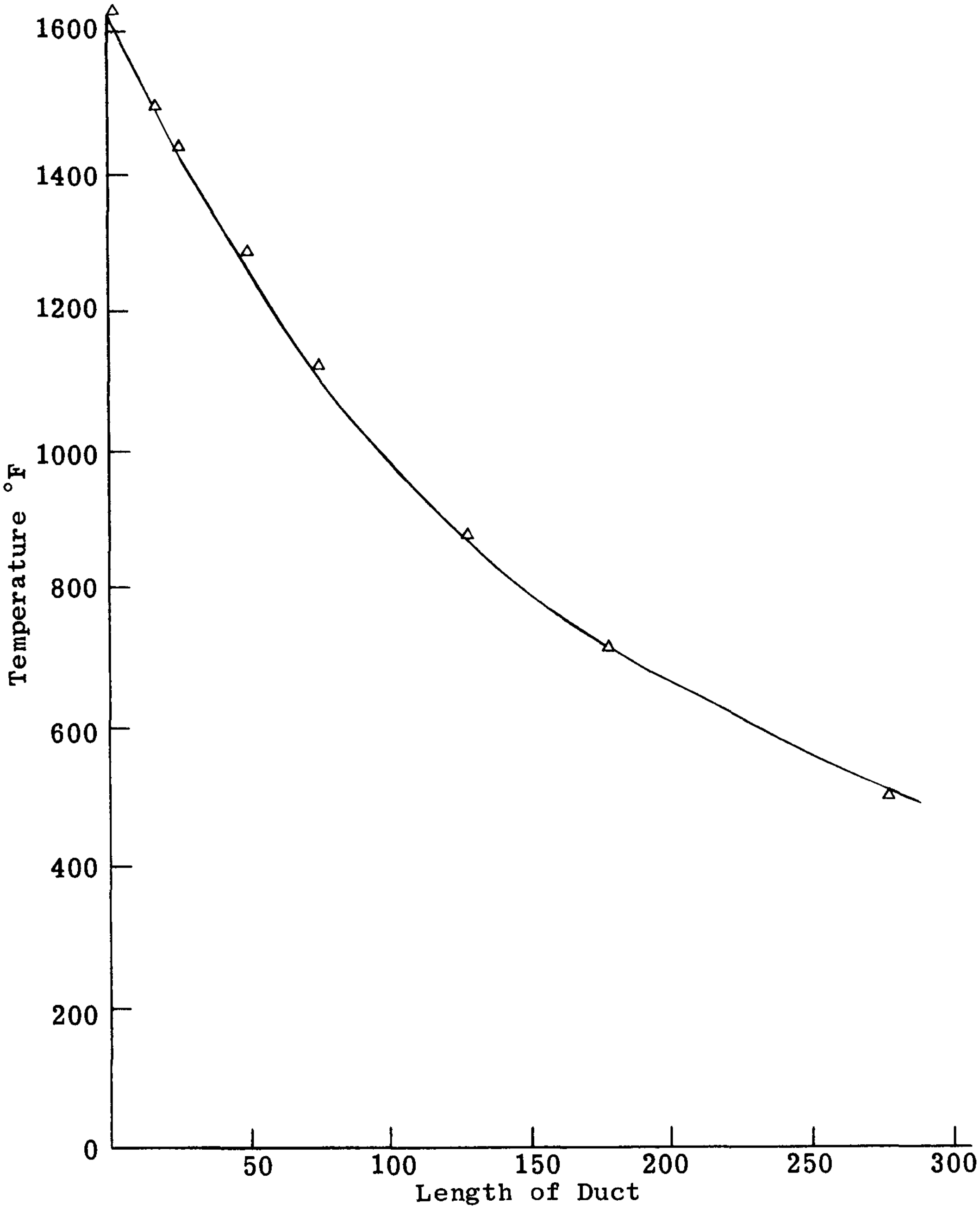

FIGURE 11

Heat Transfer Through a Duct Length

Table I suggested that for a one-hour exposure to heated air, the temperature of HEPA filter should not exceed 300 °F. Referring to Figure 9 and using 1260 °F and 300 °F as inlet and outlet temperatures, an exchanger 6-feet long is required, using the average value of 3.17 as the length of a heat transfer unit. Applying the same boundary conditions and using a more optimistic value, 2.8 for the length of a heat transfer unit, a length of about 5.5 feet is suggested by Figure 10.

CONCLUSION

Heated air generated by a fire within a radioactive processing building is likely to damage the exhaust filters. The application of water sprays to the face of HEPA filter banks is likely to result in breakthrough of contamination. Therefore, it is preferable to cool the heated air with a heat exchanger prior to filtration.

It is necessary that the distribution of the air flow be uniform so that the contact time within the spray field is maximized for all portions of the air stream.

The example, showing the use of the equations for adiabatic heat transfer of hot gas stream to a water spray applied as fire damage control, illustrated that the design parameters for an exchanger are subject to uncertainties. The greatest source of uncertainty is the temperature of the inlet air expected from a fire. It appears that the value for the theoretical heat transfer unit is independent of the disengager used, and because of the relatively high inlet temperatures expected, has little effect on the exchanger length required.

In order to optimize the heat exchanger design for each filter plenum, it is necessary to determine the exchanger inlet temperatures. This can be done by a survey of the combustible material in the area served by the filter plenum, and knowledge of the relative heats of combustion and burning rates. The design can be further optimized for the specific case by considering the effect of dilution air and knowing more accurately the heat loss through the ducting. Based on the assumptions used for the example which approximates a general case, a heat exchanger length of 5- to 6 feet appears necessary for protection of the filters for a 1-hour fire.

PERFORMANCE OF HIGH EFFICIENCY PARTICULATE AIR FILTER FOR FUME FROM BURNING PLUTONIUM METAL

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Rocky Flats Division
Golden, Colorado

April 13, 1971

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ABSTRACT

The primary objective of this study was to determine the efficiency of a commercial high efficiency particulate air (HEPA) filter in retaining plutonium aerosol from burning plutonium metal. Penetration of the test filter ranged from 0 to .0003 percent.

Particle samples collected on electron microscope grids by means of a point-to-plane electrostatic precipitator revealed a size range of .01 to .1µ for individual particles. The particulate matter was typically found in aggregates containing hundreds to thousands of particles. The cross section of aggregates measured 3 to 15µ.

Samples of particulate matter collected on membrane filters and transferred to electron microscope grids also revealed the preponderance of particles in the form of aggregates.

INTRODUCTION

High efficiency particulate air (HEPA) filters are used extensively to filter exhaust air from gloveboxes in which plutonium is processed. It is important to know how effective this type of filter is in retaining particulate matter containing plutonium. This paper reports on evaluation tests of a commercial 8 x 8 x 6-inch HEPA filter operated at flow rates up to the full rated flow while being exposed to the fume generated by burning plutonium metal.

Air was sampled using membrane filters before and after HEPA filtration. The membrane filter samples were then subjected to radiometric alpha-counting. The comparison of alpha activity of samples indicated the degree of retention of the plutonium particulate by the HEPA filter. The fume from the burning plutonium metal was sampled using a point-to-plane electrostatic precipitator to collect samples for the electron microscope to enable observation and sizing of the plutonium particulate.

DESCRIPTION OF EQUIPMENT

The major components of equipment included a test duct, sampling devices, an instrument panel, and a chip burning tray.

Test Duct

The test duct illustrated in Figure 1 consisted of two 8 x 8 x 6-inch filters positioned between short flanged sections of stainless steel duct. The plywood frames of the filters were an integral part of the duct to ensure that there would be no bypassing of the filter units.

The inlet end of the duct was adapted to fit into a glove port on a glovebox. The outlet end of the duct was provided with a butterfly valve to control air flow through the duct.

Couplings welded to the duct accommodated tubing connectors to Magnehelic® differential pressure gages and sampling tubes used to sample air passing through the duct.

Sampling Devices

Most samples were taken using a Millipore® membrane filter (AA 0.8μ White Plain 37 mm) held in a disposable plastic holder called a Field Monitor. The inlet end of the plastic holder was connected to the sampling tube and the outlet end to a rotameter which was in turn connected to a vacuum pump. Three filter holders were used. The first (P-1) was connected to the sampling tube preceding the first HEPA filter, the second (P-2) to the sampling tube between the first and second HEPA filters, and the third (P-3) to the sampling tube following the second HEPA filter.

A point-to-plane electrostatic precipitator was fabricated according to the instructions of Morrow and Mercer(1). It is shown in Figure 2 and consists of a plastic tube through which the air being sampled passes an electron microscope grid (the plane) held in apposition to a sharp electrode (the point) to which high voltage (8000 - 8500 volts) is applied. Any particulate matter not deposited on the grid is retained by a membrane back-up filter (17 mm .8μ AA Millipore) incorporated in the unit.

Instrument Panel

An instrument panel held four Magnehelic Gages to indicate the following:

M1 Pressure drop in inches of water across the first HEPA filter

M2 Pressure drop in inches of water across the second HEPA filter

M3 The negative pressure in the duct following the second filter

M4 The negative pressure at the outlet of the butterfly valve

Also mounted on the panel were four rotameters. Three of these were provided for regulating the flow of air through the membrane filter samplers. The fourth was for use with the electrostatic precipitator sampler.

Magnehelic® is the registered trademark of the F. W. Dwyer Manufacturing Company, Michigan City, Indiana.

Millipore® is the registered trademark of Millipore Corporation, Bedford, Mass.

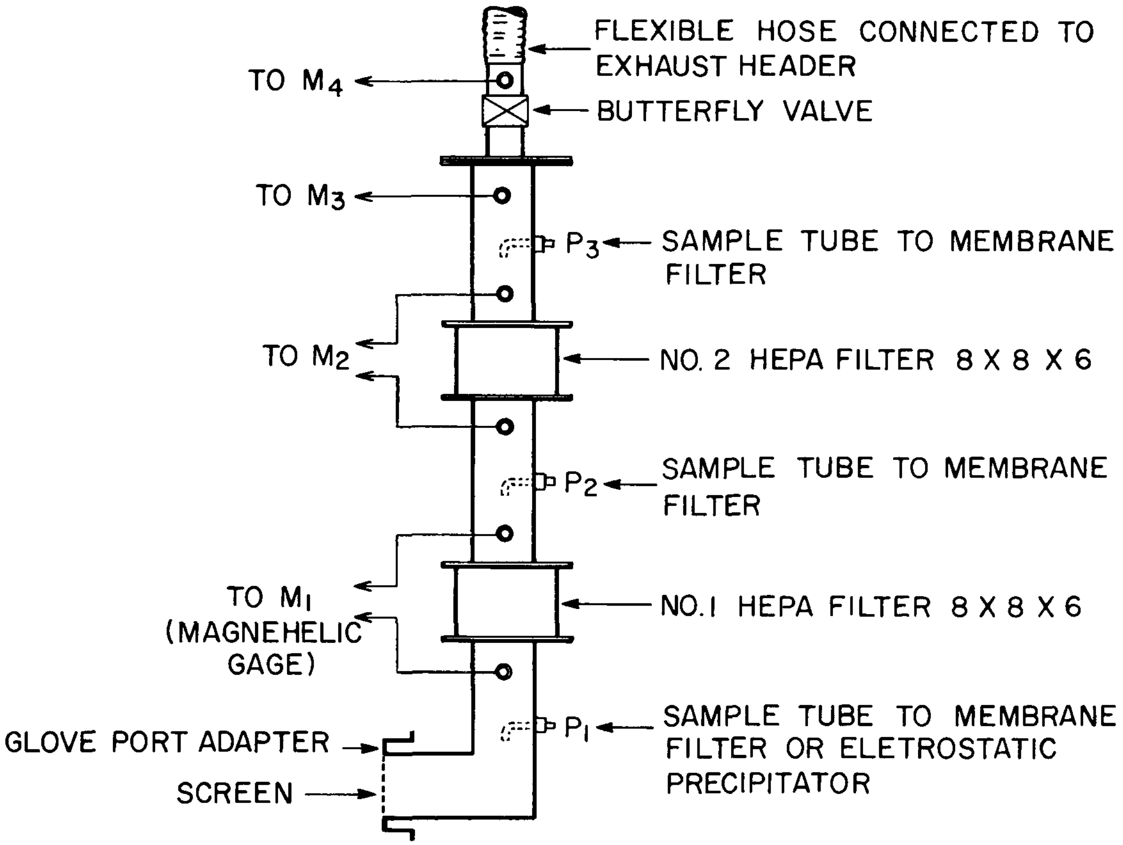

FIGURE 1

Test Duct

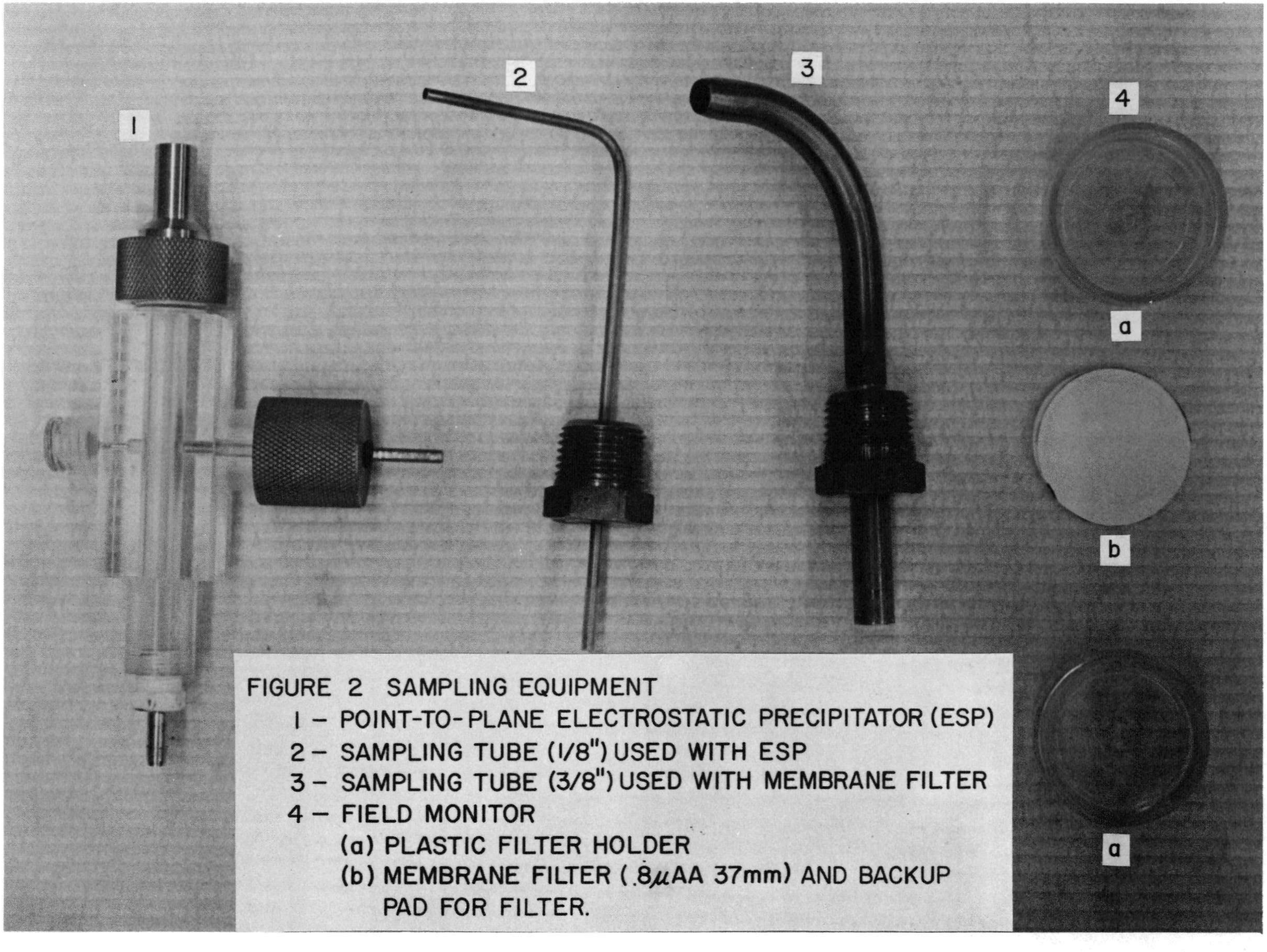

FIGURE 2 SAMPLING EQUIPMENT

1 - POINT-TO-PLANE ELECTROSTATIC PRECIPITATOR (ESP)
2 - SAMPLING TUBE (1/8") USED WITH ESP
3 - SAMPLING TUBE (3/8") USED WITH MEMBRANE FILTER
4 - FIELD MONITOR
  (a) PLASTIC FILTER HOLDER
  (b) MEMBRANE FILTER (.8μAA 37mm) AND BACKUP PAD FOR FILTER.

Chip Burning Tray

Degreased plutonium chips were burned on a coarse stainless steel screen supported on a grate fabricated from perforated sheet metal. The grate was 4 x 6 x 2-inches and positioned in a 12 x 8 x 2-inch stainless steel tray. Chips were ignited by touching them with a hot soldering gun. The photograph in Figure 3 shows a quantity of chips being burned. It is believed the aerosol particles result from the emanation of sparks during the burning process.

CHECK-OUT OF TEST EQUIPMENT

Before tests were run using plutonium, a "cold" check-out of the test duct and associated equipment was made.

Test Duct

The test duct was attached to a vacuum source in a manner than the negative pressure at the outlet of the duct could be set at the value expected when performing experiments with plutonium. By adjusting the butterfly valve a series of readings were taken to relate pressure drop across the filters with the airflow measured by a Gelman Wallac * thermal anemometer. Table 1 and Figure 4 present data on flow compared to pressure drop across the filter.

The Airpure® absolute filters used in the test duct had been tested with dioctyl phthalate (DOP) smoke at the time of manufacture. The label affixed to the No. 1 HEPA filter (See Figure 1) stated DOP penetration of .006% at flow of 50 cubic feet per minute (cfm). The No. 2 HEPA filter tested .004% DOP penetration at 50 cfm.

To insure that these filter units had not been damaged, DOP penetration tests were performed at 50 cfm flow on No. 1 and No. 2 HEPA filters installed in the test duct. DOP smoke, generated by an air set impaction type generator(2), was introduced into the airflow preceding filter No. 1. Smoke concentration was measured before and after passing through the filter using a standard foreward light scattering photometer. This procedure was repeated for filter No. 2. Results of this test are presented in Table II.

Calibration of Sampling Rotameters

The rotameters used in conjunction with the sampling tubes were calibrated for use with (a) Field Monitors and (b) the electrostatic precipitator.

(a) Field Monitor Rotameter.

A Field Monitor loaded with a 37 mm 0.8μ AA Millipore filter was connected to a wet test meter at its inlet and the rotameter being calibrated at its outlet. The rotameter was in turn connected to a vacuum system.

Airflow was measured at different rotameter settings to give the data needed for constructing a calibration curve. This curve was later used in experimental runs to achieve isokinetic sampling of air flowing through the test duct.

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*Gelman Instrument Company, Ann Arbor, Michigan

Airpure® is the trademark of Flanders Filters Inc., Washington, North Carolina.

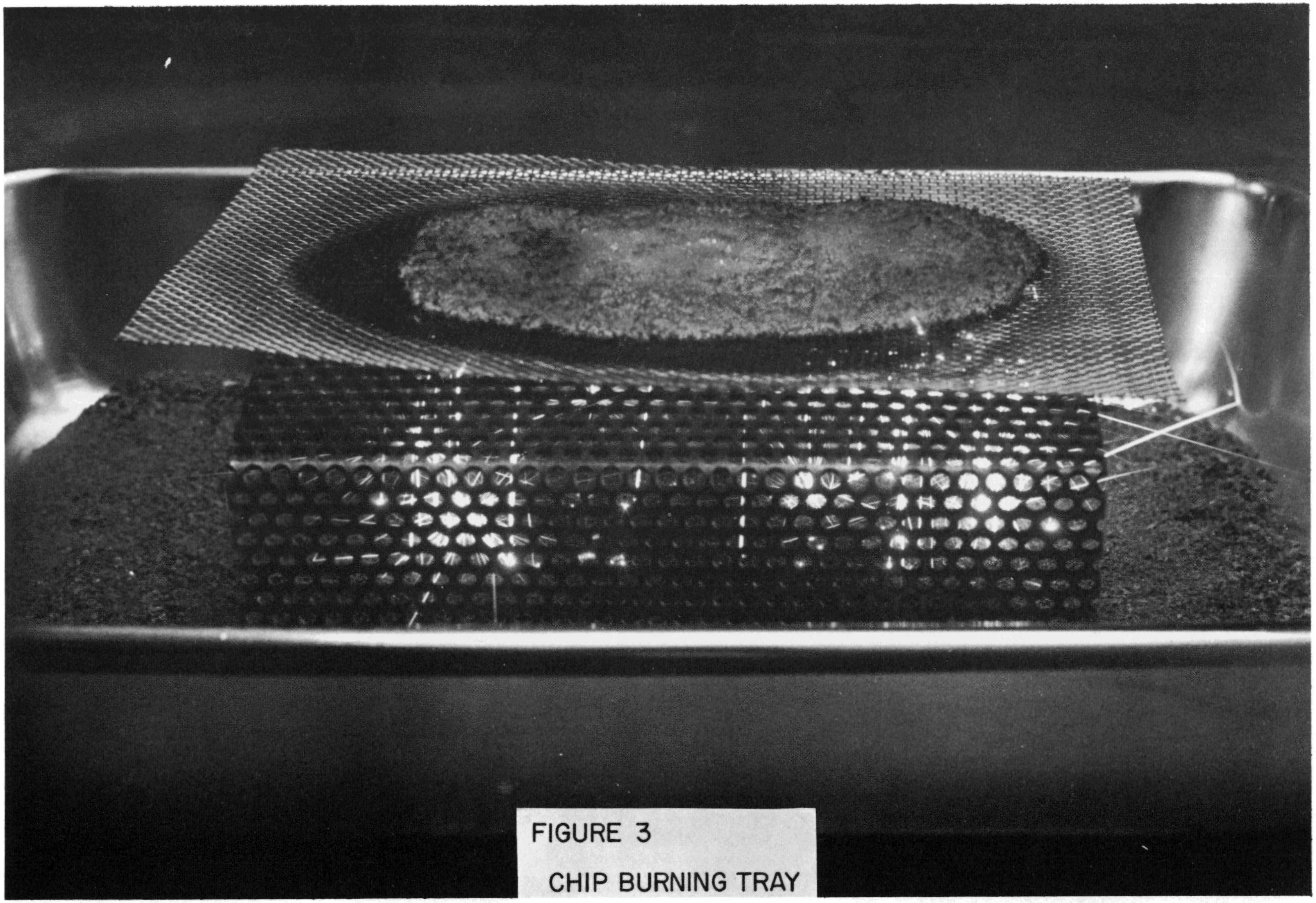

FIGURE 3

CHIP BURNING TRAY

TABLE I

AIRFLOW THRU TEST DUCT AT DIFFERENT VALVE SETTINGS

VALVE POSITION (notch)	MAGNEHELIC GAGE READINGS (Inches of water)				VELOCITY OF AIR (ft/min)	VOLUME OF AIR (cfm)
	M1	M2	M3	M4		
1 (closed)	0	0	0	-3.25	0	0
2	0	0	0	-3.25	0	0
3	.13	.13	- .20	-3.25	50	5.6
4	.32	.34	- .65	-3.25	165	18.5
5	.53	.55	-1.07	-3.25	260	28.9
6	.79	.80	-1.62	-3.25	390	43.3
7	.88	.90	-1.95	-3.25	490(1)	
8	1.00	1.00	-2.00	-3.25	500	55.6

(1) THIS VALUE WAS REJECTED AFTER DATA WERE PLOTTED.

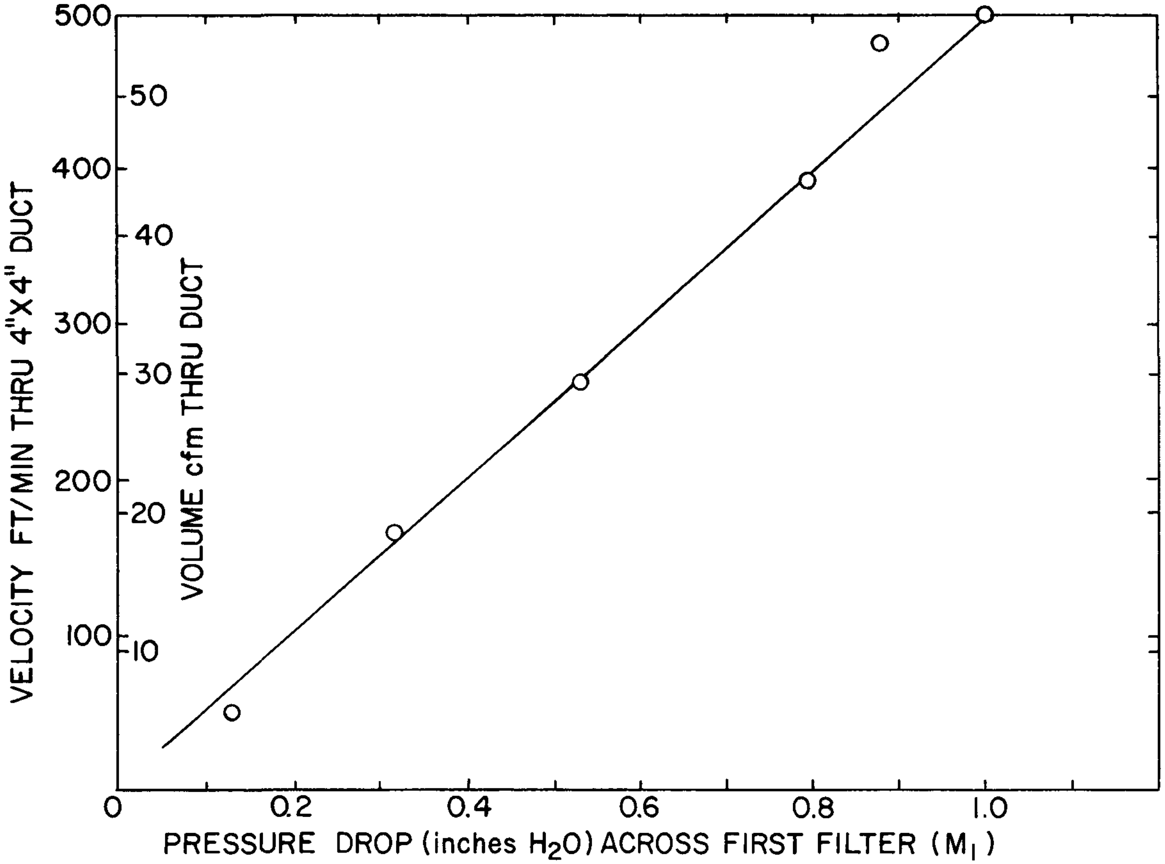

FIGURE 4

PRESSURE DROP ACROSS HEPA
FILTER VS. AIR FLOW

TABLE II

DOP PENETRATION TESTS OF FILTERS IN TEST DUCT

HEPA FILTER	DOP SMOKE CONCENTRATION PRECEDING FILTER (%)	DOP SMOKE CONCENTRATION FOLLOWING FILTER (%)	FILTER EFFICIENCY
1	85 TO 100	.002 TO .003	99.997 %
2	85 TO 100	.002 TO .003	99.997 %

(b) Electrostatic Precipitator Rotameter.

A similar calibration was made of the rotameter used with the electrostatic precipitator. In this case, however, a soap bubble in a 25 ml. buret was used to measure the small volumes of air drawn through the sampler and its 17 mm 0.8μ AA Millipore back-up filter at different rotameter settings.

Testing Using the Electrostatic Precipitator.

A test of the precipitator was made by sampling the fume from burning magnesium ribbon. The electron microscope revealed cubic crystals of MgO had been deposited on the grid. It was concluded the electrostatic precipitator could be used for sampling plutonium aerosol.

EXPERIMENTAL RUNS

After the "cold" testing and rotameter calibrations had been completed, the test duct was installed on a glovebox for the experiments with plutonium. Connections were made to the Magnehelic gages and sample rotameters. The installation is shown in Figure 5.

Filter Penetration.

The first series of experimental runs involved the following steps:

(1) Approximately 100 grams of plutonium machining chips were thoroughly degreased by repeated washings with Freon® TF. The chips were dried using a flow of argon and the weighed quantity placed on the chip burning tray located just below the inlet of the test duct.

(2) The flow of air through the test duct was set at the desired flow by adjustment of the butterfly valve at the outlet of the duct.

(3) Airflow through the Field Monitor filter holders was set at the flow required for isokinetic sampling of air by sampling tubes P1, P2, and P3.

(4) The plutonium chips were ignited by touching them at several points with an electric soldering gun.

(5) Air was sampled throughout the time required for the chips to burn and continued for the period of time required for a change of air equivalent to two volumes of the glovebox air.

(6) The Field Monitors and their contained membrane filters were removed and replaced by new Field Monitors and filters.

(7) Steps (1) through (6) were repeated for a different set of conditions.

Burning tests were made at airflows of 10, 20, 30, 40, and 50 cfm through the test duct. After these tests had been completed the Field Monitors were opened, their filters removed, and subjected to alpha counting. The sample collected at position P1 (before the first HEPA filter) was quite active and a small circle of (4.6 mm diameter) was cut from the filter for determination of the total alpha activity. The filters taken at positions P2 and P3 were very low in

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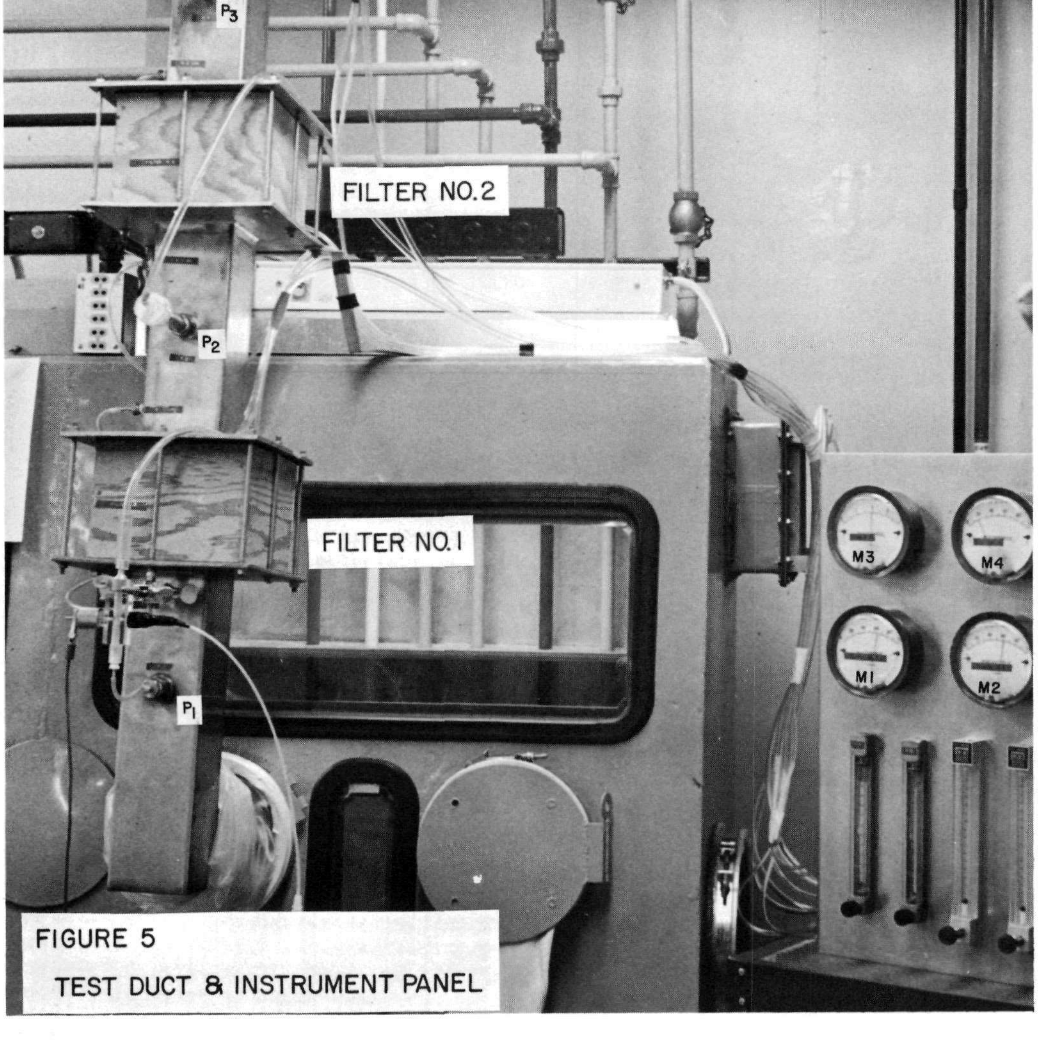

FIGURE 5
TEST DUCT & INSTRUMENT PANEL

activity and a counting period of one hour was used. The alpha activity determined for the three sample positions on each burning experiment are tabulated in Table III.

Particle Size

The second series of experiments was performed to obtain information on the size of the plutonium oxide particles in the fume from the burning chips. In this study the samples were collected using a smaller sampling tube inserted in the test duct at position P1 and connected to the electrostatic precipitator. Sampling periods were varied while plutonium chips were burned with airflow through the test duct held at 30 cfm.

Plutonium oxide particles were deposited directly on copper electron microscope grids which had been prepared by coating with collodion and carbon. Alpha activity was determined on the grid and the backup filter to measure the efficiency of the precipitator. Data on the samples taken are presented in Table IV.

The particle samples collected on the electron microscope grids were examined in the electron microscope. It was observed that the plutonium oxide particles were well formed crystals in the form of cubes, octahedrons, and dodecahedrons. Most of the crystalline particles measure in the range from .01 to .10μ. No separate particles were observed but instead aggregates containing hundreds of individual crystals in branched chains and masses. The aggregates measured from 3 to 15μ. Electron photomicrographs of the particles collected by the electrostatic precipitator are shown in Figures 6, 7, 8, and 9.

It was suspected that collection of particles using the electrostatic precipitator could favor agglomeration of the particles. Sample of particles collected directly on membrane filters at sample location P1 were studied using a replication method. In using this technique films of carbon and platinum were evaporated on the exposed surface of the membrane filter on which plutonium particles had been collected. The cellulosic filter medium was then dissolved away in such a manner as to allow the "coated" particulate matter to deposit on a suitably prepared electron microscope grid.

Electron photomicrographs of particles collected on membrane filter media are shown in Figures 10 and 11. The photomicrographs reveal agglomeration patterns similar to those observed when particles were collected using the electrostatic precipitator. It was therefore concluded that the agglomeration observed did not necessarily result from particle collection by the electrostatic method.

TABLE III

PENETRATION OF HEPA FILTERS BY PLUTONIUM PARTICULATE FROM BURNING Pu CHIPS

AIRFLOW (cfm)	Pu CHIPS BURNED (grams)	TIME OF BURNING (min)	ALPHA ACTIVITY ON MILLIPORE FILTERS (d/m)			PENETRATION THRU FIRST FILTER (%)
			P1	P2	P3	
10	99.0	17	0.5×10^6	1.7	1.3	3×10^{-4}
20	101.0	12	4.1×10^6	0.7	1.3	2×10^{-5}
30	98.0	18	0.1×10^6	0.3	0.7	3×10^{-4}
30	194.0	27	2.4×10^6	2.7	1.0	1×10^{-4}
40	101.5	17	3.8×10^6	0.0	0.0	0
50	100.8	17	7.4×10^6	4.0	0.3	5×10^{-5}

TABLE IV

ELECTROSTATIC PRECIPITATOR SAMPLES

SAMPLE NO.	TIME (min)	Pu CHIPS BURNED(g)	ALPHA ACTIVITY EM GRID (d/m)	ALPHA ACTIVITY BACKUP FILTER	EFFICIENCY OF PRECIPITOR (%)
CONDITION: FLOW THRU TEST DUCT - 30 cfm FLOW THRU ESP-41 ml/minute VOLTAGE-8200 TO 8700 VOLTS AS REQUIRED FOR CORONA DISCHARGE					
1	5.0	28.2	2516	726	78
2	1.0	28.5	1167	642	65
3	10.0	102.0	921	88	91
4	0.5	25.2	413	330	56
5	10.0	74.0	4523	631	88
7	10.0	71.8	3033	713	81
				AVERAGE	76
CONDITION: FLOW THRU DUCT -50 cfm FLOW THRU ESP-68 ml/minute					
8	5.0	110.0	34,000	24,144	58

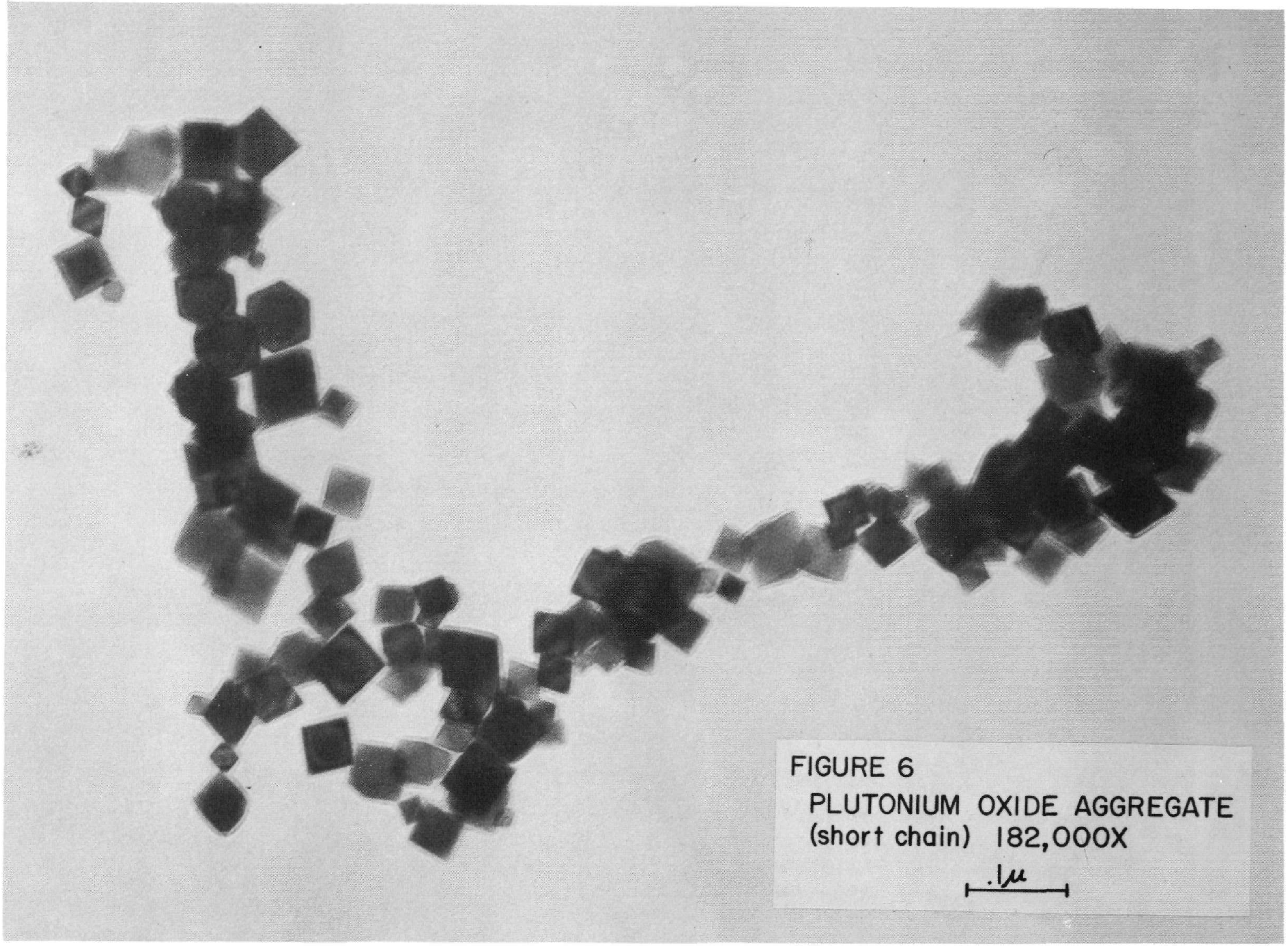

FIGURE 6

PLUTONIUM OXIDE AGGREGATE
(short chain) 182,000X

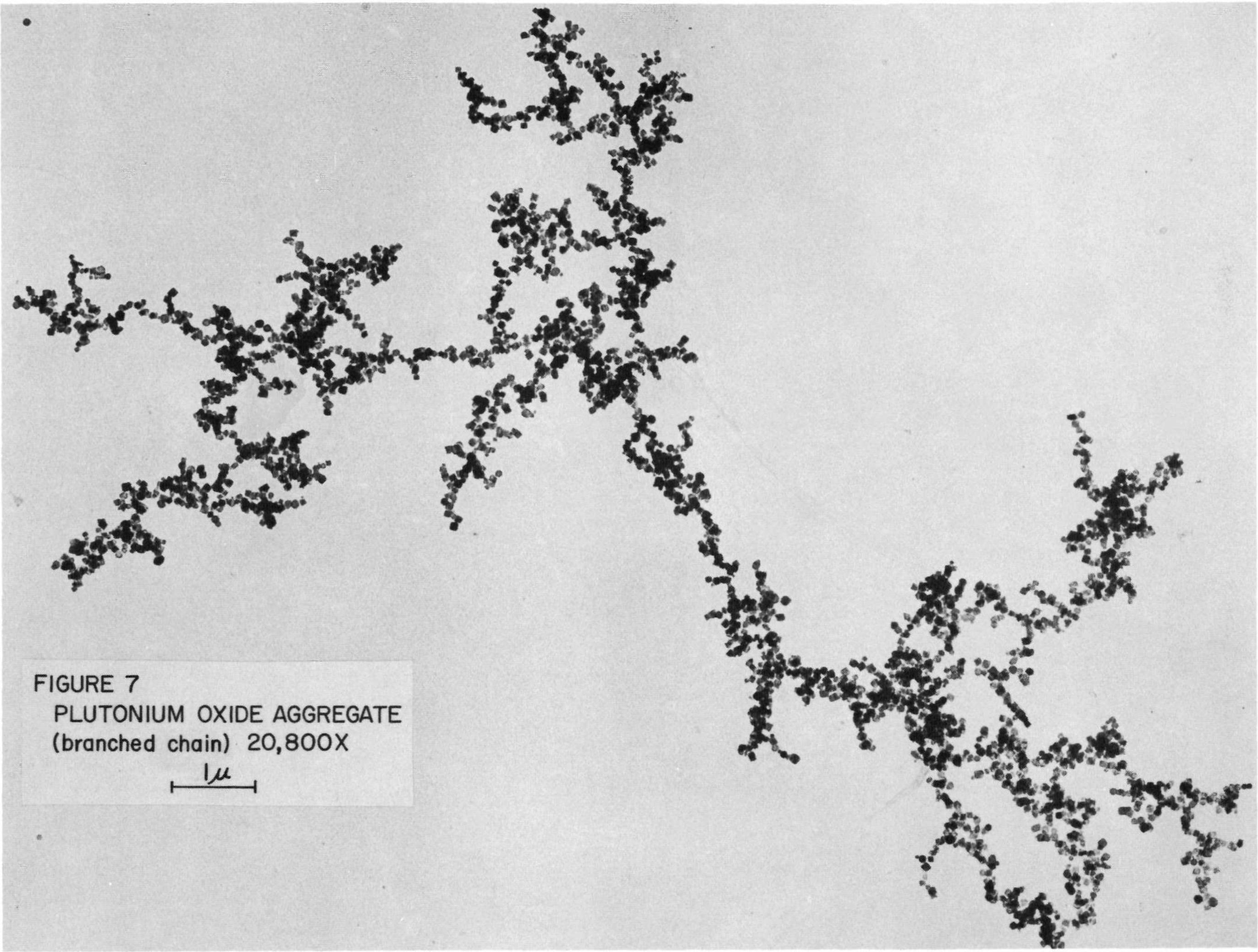

FIGURE 7
PLUTONIUM OXIDE AGGREGATE
(branched chain) 20,800X

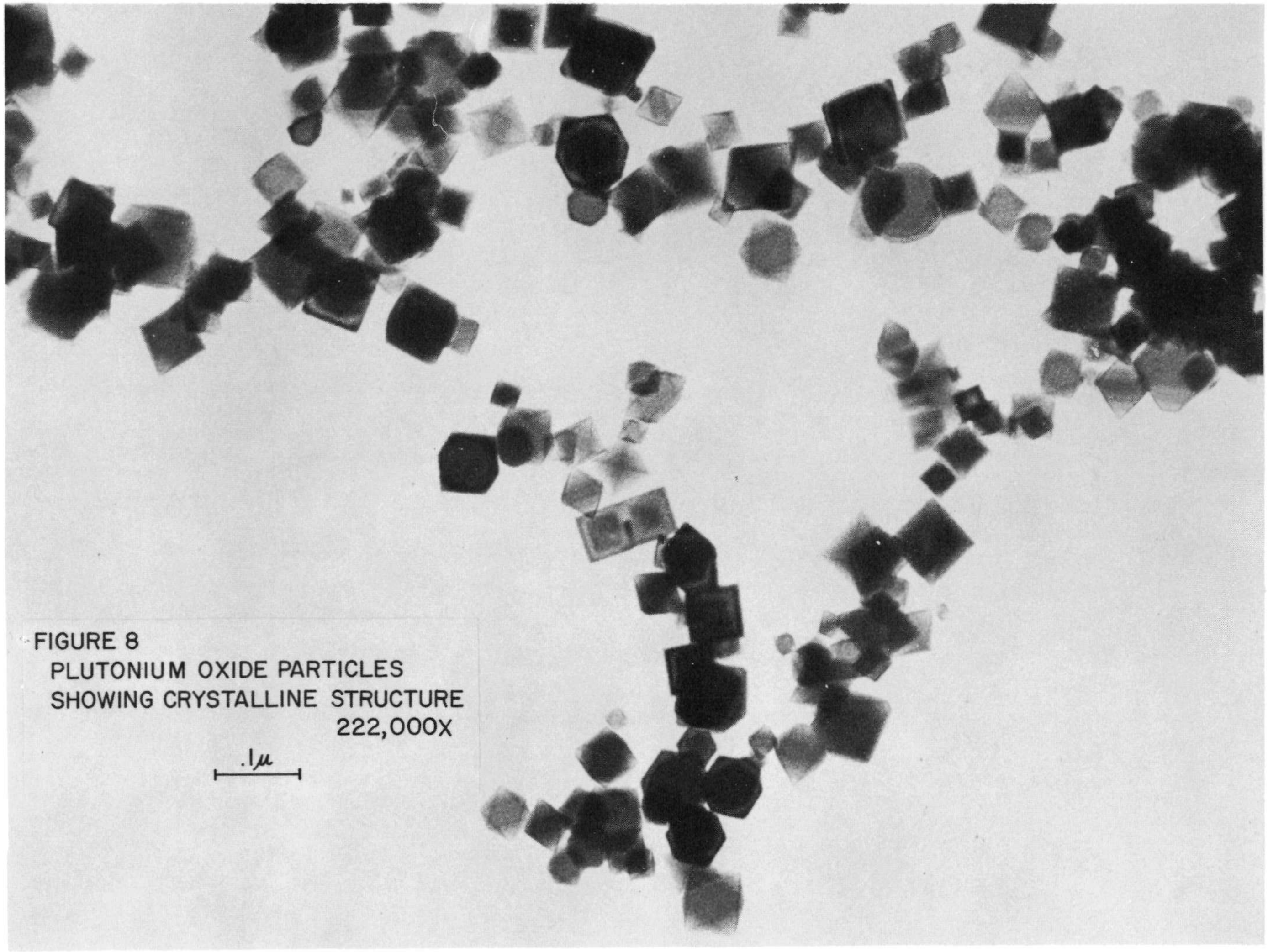

FIGURE 8
PLUTONIUM OXIDE PARTICLES
SHOWING CRYSTALLINE STRUCTURE
222,000X

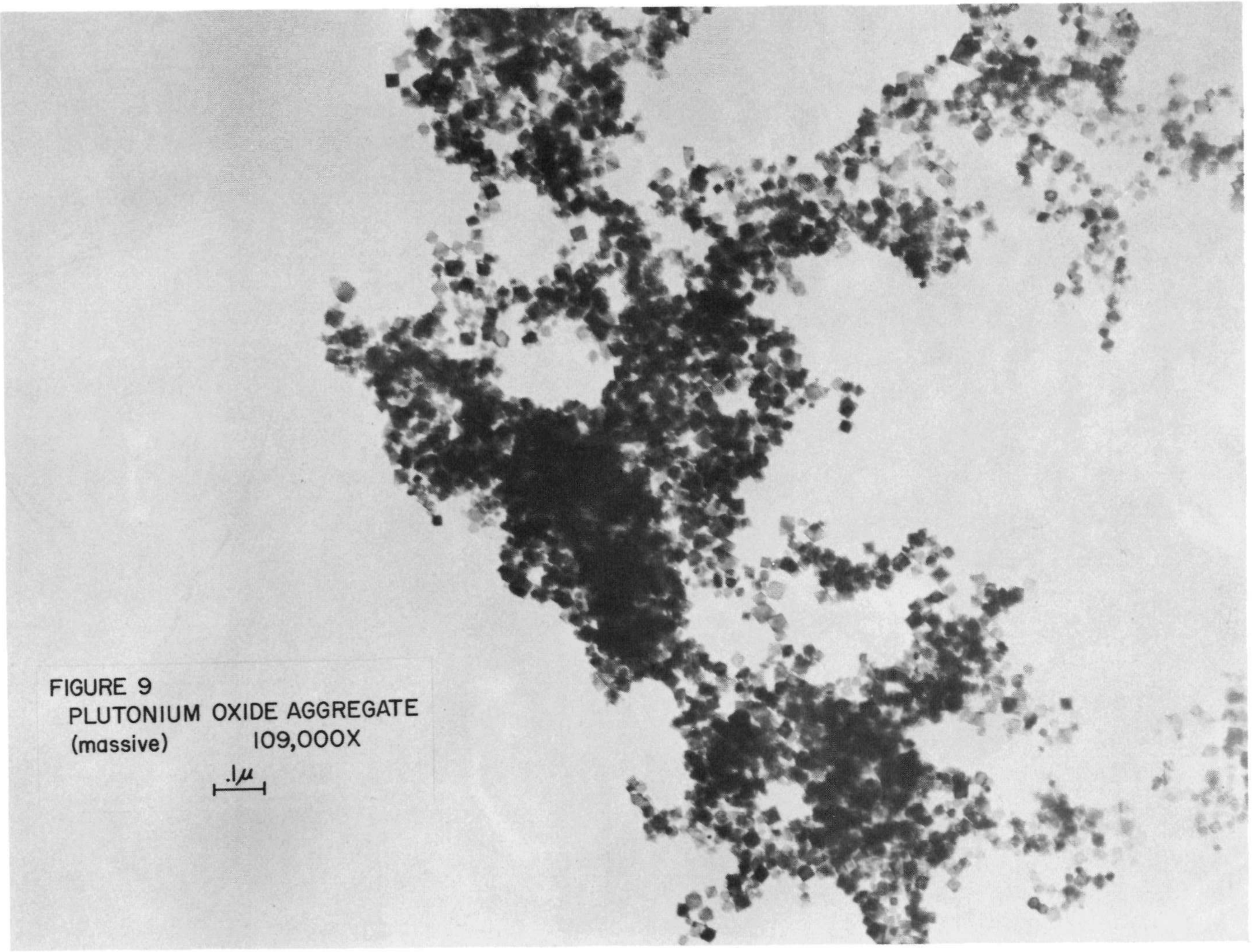

FIGURE 9
PLUTONIUM OXIDE AGGREGATE
(massive) 109,000X

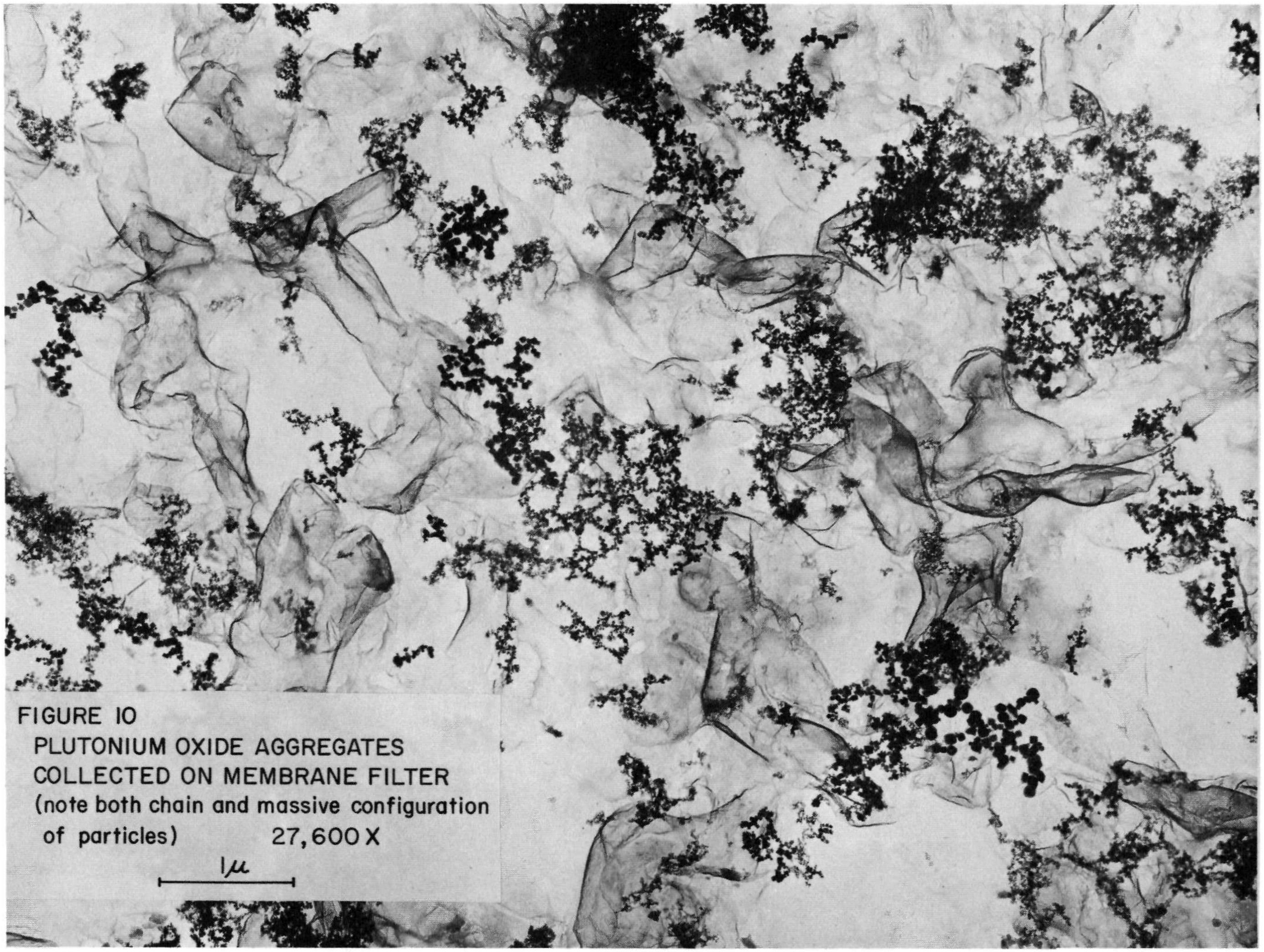

FIGURE 10
PLUTONIUM OXIDE AGGREGATES COLLECTED ON MEMBRANE FILTER (note both chain and massive configuration of particles) 27,600 X

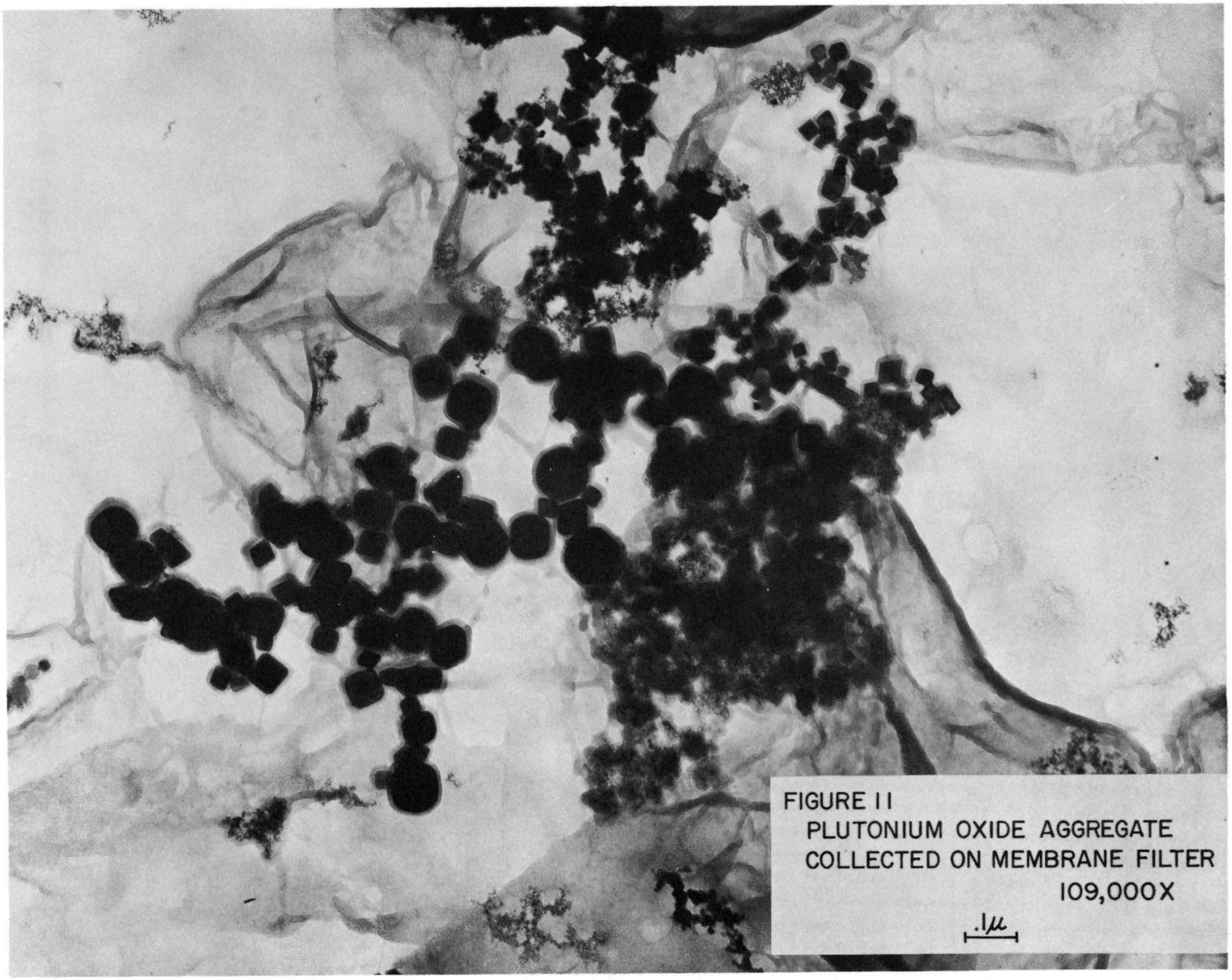

FIGURE 11
PLUTONIUM OXIDE AGGREGATE
COLLECTED ON MEMBRANE FILTER
109,000X

DISCUSSION

DISCUSSION

The primary purpose of the experimental work was to observe the efficiency of a commercial type HEPA filter for filtering plutonium oxide aerosol produced when plutonium metal is burned. The filter was tested at flow velocities of 1 to 5 ft. per minute (10 to 50 cfm). Under the conditions of the test the filter performed exceptionally well. The filter efficiency for this type of plutonium aerosol was greater than for the DOP smoke challenge.

Samples of particles from the plutonium oxide aerosol were collected using techniques based on electrostatic precipitation and membrane filters. Examination of the particles under the electron microscope revealed small plutonium oxide particles (crystals) had agglomerated into chains and masses. The size of the particle agglomerates typically exceeded one micron in size. The high efficiency of the HEPA filter is believed due to the fact that the relatively large agglomerates were readily retained by the filter medium by the mechanism of impaction.

Additional experimentation should be carried out to find a means of generating plutonium particles in a non-agglomerated state with which to challenge HEPA filters. In addition, practical tests should be performed on the effects of chemical attack and loading on the performance of HEPA filters.

REFERENCES

(1) Morrow, P.E.; T.T. Mercer, 1964, A Point-to-Plane Electrostatic Precipitator for Particle Size Sampling. American Industrial Hygiene Association Journal 25.8-14

(2) Echols, W.H.; J.A. Young, Studies of Portable Air Operated Aerosol Generators. U. S. Naval Research Laboratory Report No. 5929, July 26, 1963

SECTION III.

FIRE DETECTION AND SUPPRESSION SYSTEMS

ORGANIZATION AND PLANNING FOR FIRE CONTROL

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April 14, 1971

ORGANIZATION AND PLANNING FOR FIRE CONTROL

Picture in your mind, my concept of a catastrophe about to take place.

It is a dark night in the middle of February, a gale force wind is blowing, 15" of snow has already fallen - drifting shut the main arterial roads of the ABC Nuclear Fuel Processing Company plant. The usual limited graveyard shift is doing their best to maintain production schedules even though a number of their key technicians had not been able to make the 15 mile trek from town.

At the time of the midnight shift change an unnoticed event took place that was later to become the key link in a chain of events that most always preceeds a catastrophe. An explosion-proof light fixture being repaired in a solvent extraction recovery process glove box had not been fully restored to service. The burned out light had been replaced but the explosion-proof globe was not restored to its proper position. The shift change bell had sounded and being a bad night, there was more than the usual eagerness to get on the road home, so the electrician had forgotten about tagging out the unit and had left the explosion-proof globe laying in the glove box.

As might be expected, the solvent extraction recovery process had fallen behind schedule due to the maintenance shut down during the prior shift. The tankage for storage of radioactive materials to be recovered had filled up.

The third shift extraction process supervisor, decided to substitute for the operator who had failed to show up at the plant due to the inclement weather. Working the line was not a normal job function of the supervisor so he made a quick check of the Department Operations Manual to refresh his knowledge of the solvent unit, went through a rather cursory start-up check list and then started things going by turning on the solvent circulation pump. Naturally about this time, the supervisor was paged over the intercom to call the Plant Office, so he left the working area to get to a telephone.

As you might expect in this fictitious story, the Supervisor in his haste to start-up operations, forgot to close a drain valve at the base of one of the exchange columns, so the inevitable occurred. A solvent mixture bearing waste radioactive materials poured from the open valve when the circulation pump was started. Despite the fact the glove box exhaust system was operating, the ventilation rate was not designed to handle such an accidental spill. No drains had been placed in the glove box and in very short time a flammable vapor/air mixture developed.

About this time the supervisor, returning from the phone call, immediately observed the trouble. He actuated the emergency shut down system which controlled the solvent circulation pump and all electrical devices in the glove box. A false sense of relief filled the supervisor and he was quite elated that he had successfully gotten himself out of trouble. The exchange column continued to drain its contents of flammable and radioactive liquid. Shortly thereafter he decided that he needed glove box lighting to enable him to clearly identify the proper valve to close and so that a clean-up program could commence.

He flipped the lighting control switch, and all things blacked out as far as he was concerned.

A tremendous explosion occurred instantaneously with this event knocking the supervisor unconscious.

The open light fixture completed the chain of events needed to make this story a catastrophe. Besides immediately rupturing windows in the glove box and spreading excess amounts of radioactivity to the immediate room area, an ensuing fire soon raged uncontrolled involving the ordinary combustibles left in the room area and also the combustible glove box construction materials.

Perhaps this is a good spot to stop in this fiction episode - you have had the build-up and the big event - but no conclusion. Since I can't ask you to tune in next week, I should perhaps ask you to write your own conclusion by interjection of actual factors that could be brought into the story by making the assumption that this episode took place in your own plant or facility.

To help you in reaching a conclusion I have a few tactful questions to ask.

1. Management attitude:

   Has management clearly and consisely shouldered their responsibilities regarding the overall Safety and Loss Prevention program for the plant, by establishing favorable policies and objectives that are adequately documented, by providing an adequate number of skilled technical personnel in their respective Safety and Loss Prevention disciplines required by the hazards of the plant and by providing adequate financial resources to carry out the policies, objectives and programs established?

   Does Management apply to Safety and Loss Prevention matters the same skills, techniques and talents that have proven successful in finance, production and marketing?

2. Preplanning for Emergencies:

   Has an effective and well documented emergency plan been established for the plant, with responsibility established for its control, development and implementation?

   Are employees trained in proper procedures for calling the Plant Emergency Squad for assistance?

   Is it (emergency plan) clear to all employees and do they individually receive instruction and training so that they may be expected to perform the required response during a given emergency situation, with a high degree of reliability?

   Is the plan designed to be effective at all times; not just during the day shift but at night, over holidays, or during bad weather, strikes or other shutdown periods?

   Are periodic pre-planning surveys made by the Plant's Fire Control Department (at least on a semi-annual basis), documented in such a fashion that they may be used as a part of the "run card" for the particular building involved?

3. Self-Inspection Program:

   Has plant established a preventative maintenance type inspection program for mechanical equipment including fixed fire protection systems and devices? Does this program require periodic testing of such devices by competent personnel on a regular basis? Are mechanical defects discovered by this program, reported immediately, repaired or equipment replaced?

   Are loss prevention surveys made on a frequent basis by the respective disciplines of the plant's Safety and Loss Prevention Department (Fire Protection Engineering, Fire Control, Safety including Industrial,Radiological and Nuclear). Are recommendations submitted as a result of these surveys to the persons responsible for their completion? Is a follow up program in effect to insure a successful program?

   Does your Self-inspection Program involve Senior Management in periodic audit teams?

   Does an orderly procedure exist for the establishment of Special Committees to perform specialized studies, including use of outside experts if necessary?

4. Plant Emergency Squad - Organization and Training:

Depending upon the size and complexity of the plant or facility involved, its remoteness and special problems involving security:

Is an adequate Plant Emergency Squad organized and available for all hours of days, nights and weekends?

Where necessary are specialized talents included such as Health, Physicists, Radiologists, Nuclear Safety experts, and Utilities Department representatives?

Are members of the Plant Emergency Squad thoroughly trained in fire-fighting fundamentals?

Are they thoroughly acquainted with the capabilities and limitations of fixed special extinguishing systems?

Does training include use of pre-planning information developed for the building or department with emphasis on the special hazards involved?

Are adequate emergency equipment and vehicles provided to meet the anticipated needs of the Plant Emergency Squad?

Is the training program designed to develop resourcefulness of individuals by including in its drills simulated emergencies involving personnel injuries and equipment damage?

Is a fire training burning pit area available to allow the development of on-the-job skills in the use of fire fighting equipment, protective clothing and self contained breathing apparatus?

5. Mutual Aid Agreements and Cooperation with Public Fire Departments:

Have agreements been made with nearby public and private fire departments, establishing mutual assistance organization and response plans during possible catastrophe and natural disasters?

Are the fire department officers periodically invited to visit the plant and are they made aware of the plant's hazards and fixed fire protection systems (sprinklers, water supplies, special extinguishing systems, smoke and heat venting facilities)?

Have clear instructions been given to select employees for sounding the fire or emergency alarms and for calling for Mutual Assistance?

Are joint training exercises periodically run to establish efficiency, teamwork and tests of response plans?

Let us hope that your answers to the foregoing questions permit a successful conclusion to my story.

Much more could be said about the need for proper organization and planning for fire control, safety and loss prevention. One could theoretically make a case for saying that virtually every step - every phase - every operation of a Plutonium Handling Facility in some way involves these subjects.

There should be no question about the basic fact that successful organization and planning must incorporate the "cradle to the grave" principle. All new projects must include planning for fire control and other disciplines of safety and loss prevention, during the earliest possible review stage. Once the project is a living reality, assuming that it has gone through many additional review steps before finalizing design and construction, fire protection or prevention hardware and other safety devices should have been provided commensurate with the potential problem involved. Organization and planning must continue throughout the active life of the project, and vigilance must be maintained in the event the unexpected occurs. A lot of time, talent and money must be expended with no guarantee that they will ever get a chance to prove their worth; but can anyone clearly suggest an approach which does not consider these essentials?

Submitted by: Roland I. Spencer, Executive Assistant
Factory Insurance Association
March 30, 1971

THE CATEGORIZATION AND POSTING OF NUCLEAR FACILITIES FOR FIRE PROTECTION PURPOSES AT WADCO

R. M. Gough

Hanford Engineering Development Laboratory
U. S. Atomic Energy Commission

Operated by WADCO Corporation
Richland, Washington 99352

April 14, 1971

U. S. Atomic Energy Commission Contract AT(45-1)-2170

THE CATEGORIZATION AND POSTING OF NUCLEAR FACILITIES FOR FIRE PROTECTION PURPOSES AT WADCO

Over the years the need was realized for more flexible rules governing the use of water for fire fighting purposes in nuclear facilities at Hanford. Out of this need has emerged a system for identifying the risk of causing an uncontrolled nuclear reaction that not only establishes a criteria for fixed fire protection systems in nuclear facilities, but also aids the site fire department in making decisions on the correct procedures to use in combating fires in these facilities.

The Industrial Safety and Nuclear Safety departments of the Hanford contractors along with the Hanford Fire Department participated in creating a plant wide system for posting of these areas. While the identification systems are varied between contractors the symbols adopted have the same meaning to the fire department. This was an essential requirement when setting up a program such as this in a multi-contractor site served by one fire department.

Before the system was developed and placed in effect, several small fires occurred in areas where water was not allowed to be used. Normally, these would have been little more than "nuisance fires" resulting in little or no fire loss. However, since the use of water was prohibited the fires were permitted to reach proportions where the contamination spread cleanup costs resulted in a relatively large loss. Subsequent investigations concluded that water could have been used safely. After these experiences some people began questioning the validity of the blanket policy of no water in nuclear facilities.

After careful studies by fire protection and nuclear safety engineers, a criteria was developed that is consistent with criticality safety which allows the maximum use of water for fixed and manual fire fighting in facilities containing fissile material. It was found that criticality hazards could be lessened or removed entirely by effective engineering. In WADCO nuclear facilities both fire detection and automatic extinguishing systems are recommended in order to gain early and effective control of fires. Automatic sprinkler systems are recommended in all areas except where the nuclear hazard would prohibit the use of water. Storage vaults and certain gloveboxes are examples. In some cases, shields or directional spray nozzles are used to keep water out of these areas. Limited discharge sprinkler systems and provisions for large drainage capacities are methods to be used by fire protection personnel in system design. Sprinkler systems are not always acceptable and consideration is then given to other type extinguishing systems.

The responsibilities for this program is divided among three groups in WADCO. The Nuclear Safety Organization categories facilities into four groups. This categorization is done with the idea of obtaining the least possible restrictions on the use of water consistent with criticality safety. Each building is reviewed and audited periodically to see that their criteria is observed and up to date. Adjustments have been made in inventories and processes in order to reduce restrictions.

The operating management of each facility routinely reviews the building occupant adherence to the requirements of Nuclear Safety. They are responsible for actual posting of areas and notifying Nuclear Safety when inventories, processes, locations or other items change that could effect criticality safety.

The Industrial Safety department maintains building floor plans denoting posted facilities and notifies the fire department of category changes. Industrial Safety occasionally urges measures be taken to allow less restrictive fire fighting categories which allow increased facility fire protection. We have found that operating personnel are quite receptive to making adjustments which result in less restrictive categorization.

For fire-fighting purposes, each laboratory area containing fissile material has been classified into four categories, depending upon the quantity and nature of fissile materials present. These categories are:

> Category A areas have no probability of criticality if water is used to fight fires because fissile materials are present in a relatively small quantity. Fire fighting methods are not restricted in any way by criticality safety.
>
> Category B areas have a very low probability of criticality if water is used to fight fires as the fissile material present is either in a very dilute solution or confined in a geometry such that a criticality cannot occur. Water in any form may be used in any amount.
>
> Category C areas have a moderate probability of criticality if unrestricted use is made of water to fight fires. Fissile material is present in amounts and configurations that could be made critical upon rearrangement and the addition of water. The only forms of water which may be used are high expansion foam, automatic sprinkler systems, or hose fog nozzles.
>
> Category D areas have a high probability of criticality if water is used to fight fires as fissile material is present in configurations that could be made critical upon the addition of water. Water may be used only in the form of high expansion foam.

In order to provide immediate on-the-scene fire fighting instructions, all areas are posted except Category A, with appropriate category signs denoting the criticality concerns for that area. These signs are centered on the door frame immediately above doorways and spaced 1/8 inch from the frame. Firemen are required to feel above the door for this sign if visibility is poor. The raised feature allows the fireman to differentiate between the different shaped signs which denote categorization. Because of pre-fire plans and frequent building tours the firemen have a general idea of what to expect.

> The signs for Category B are: Diamond shape with phosphorescent green background.
>
> The Category C sign is: Equilateral triangle with a phosphorescent red background is used to denote rooms or areas. A square sign with the notation "C HOODS" on a phosphorescent orange background is used to denote gloveboxes, refrigerators or other enclosures within a room.
>
> The Category D sign is: Round sign with a phosphorescent blue background is used to denote rooms or areas. A rectangular sign with the notation "D HOODS" on a phosphorescent yellow background is used to denote gloveboxes, refrigerators or other enclosures within a room.

An example of posting would be the triangle sign and the rectangle. This tells the fireman he can use water fog in the room. Don't direct water at a glovebox. Use high expansion foam or some other extinguishing media other than water in the glovebox. Automatic sprinklers can be installed in this area, however, directional heads or noncombustible shields over glovebox openings may be advisable.

These restrictions in the use of water are designed to minimize the risk of criticality while fighting a fire. They must be observed by the fire fighter. However, there may be a point where a decision must be made to use water in direct conflict with posting. In this case the senior fire officer shall obtain consultation from the facility emergency director. If the facility emergency director is unavailable, the decision to use water against posting restrictions rests with the senior fire officer.

There have been two major end results from the initiation of this system. The first is more confidence on the part of the fire department officers when responding to fires in nuclear facilities at Hanford. The second is the increased fire protection of these areas as the result of the elimination of unnecessary restrictions on manual fire fighting methods and the use of automatic sprinkler systems.

FIRE DETECTION AND SUPPRESSION IN GLOVEBOX SYSTEMS

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April 14, 1971

FIRE DETECTION AND SUPPRESSION IN GLOVE BOX SYSTEMS*

by

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ABSTRACT

An automatic fire-extinguishing system has been designed and tested for protection against non-metallic fires in glove boxes at the Savannah River Laboratory. The extinguishing agent is Halon 1301 (bromotrifluoromethane). A number of glove boxes are presently protected by manually actuated systems pending conversion to automatic systems as operations permit.

---

* The information contained in this article was developed during the course of work under Contract AT(07-2)-1 with the U. S. Atomic Energy Commission.

INTRODUCTION

Studies of fires and explosions in glove boxes showed that preventive measures alone do not provide adequate protection because operating errors and unforeseen situations cause accidents. To establish guides for selecting extinguishing systems for glove boxes, studies of extinguishing agents were made, and detectors were tested. As a result of these studies, an automatic fire-extinguishing system was developed to protect glove boxes at the Savannah River Laboratory.

FIRE PREVENTION AND PROTECTION

Fires or explosions in glove boxes for work with radioisotopes are potentially very serious because of the quantity of highly radioactive materials that could be dispersed by destruction or breach of the protective containment. In an earlier study of fires and explosions and incidents with serious potential that have occurred at atomic energy sites, the modes of initiation and propagation were identified and preventive measures were suggested for each cause. Although preventive measures reduce the potential for fires and explosions and may reduce the consequences of those that do occur, experience shows that preventive measures alone do not ensure adequate protection. Serious fires and explosions have occurred as a result of operating errors and unforeseen incidents. Thus, effective alarm and extinguishing systems are necessary to supplement preventive measures.

It was also apparent from the study that the installation of automatic fire extinguishing systems has been deterred because of uncertainties in selecting appropriate extinguishing agents, adverse experience with detection systems, particularly in the environments encountered in work with radioisotopes, and also the high cost of some systems. Therefore, studies were made of extinguishing agents and tests of various detectors were conducted* to establish guides for selecting fire extinguishing systems for glove boxes.

---

* Tests of detectors were conducted at Fenwal Incorporated, Ashland, Mass.

DETECTORS

Tests of various detectors showed that thermal detectors are very reliable and those that are rate-anticipating are particularly suitable for glove box applications. The tests also showed that for early detection it is preferable to locate a detector in the top near the center of a small box; additional detectors are desirable in larger boxes, particularly above principal pieces of equipment or above areas where a fire might conceivably originate. The detection systems will have supervised circuits whether they are connected to the extinguisher to discharge the Halon 1301 automatically or to provide an alarm only. A schematic diagram of the wiring of the supervisory system is shown in Figure 1.

EXTINGUISHING AGENTS

Studies of the properties and application of extinguishing agents and subsequent tests to compare carbon dioxide and Halon 1301 (bromotrifluoromethane)* showed that Halon 1301 can replace carbon dioxide to provide faster, more effective extinguishing capability and to eliminate some of the problems associated with carbon dioxide and other agents. Although Halon 1301 will not extinguish metal fires, it can be used to prevent the propagation of metal fires to surrounding materials, particularly to protect absolute filters in exhaust ducts. Only 5 vol % of Halon 1301 is needed for most fires because it reacts chemically with the intermediate products involved in the propagation of flame. The reaction is fast, and the inhibiting reaction starts before the effective concentration is attained. Tests also showed that the increase in volume during discharge and evaporation of the small quantity of agent does not upset the control of atmospheric pressures in cells or boxes, and because of the greater vapor density, Halon 1301 has a more prolonged effect than carbon dioxide in preventing rekindling of a fire. Because normal pipe, tubing, and fittings can be used in Halon 1301 systems, the cost is less than that for carbon dioxide even though the cost of the agent is higher. Carbon dioxide systems require high pressure hardware.

SELECTION OF SYSTEMS

All glove boxes have or will be provided with fittings and piping or hose for connections to extinguishers or extinguishing systems. All laboratories in which radioactive materials are handled now have manually actuated Halon 1301 fire extinguishers. Laboratories where metal fires are possible also have extinguishers or extinguishing agents appropriate for the particular metal or metals.

In general, extinguishers mounted on glove boxes will have electric switch-operated valves for manual or automatic operation. Where the hazard is very low, the squeeze-grip manual extinguishers may be used for economy.

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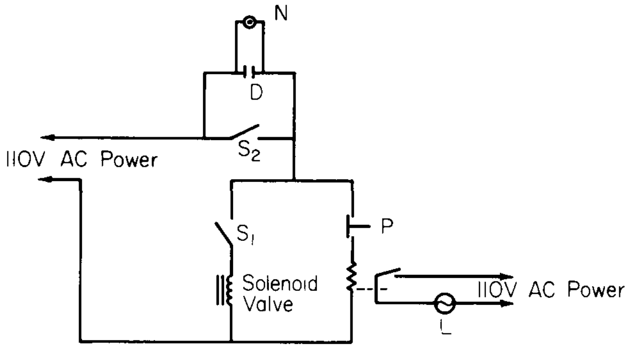
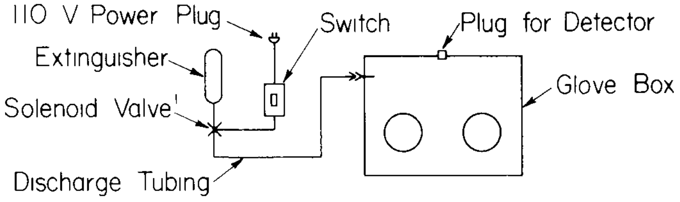
* *Standard on Halogenated Fire Extinguishing Agent Systems (Halon 1301)*, NFPA Bulletin 12A, National Fire Protection Association, Boston, Mass. (1970).



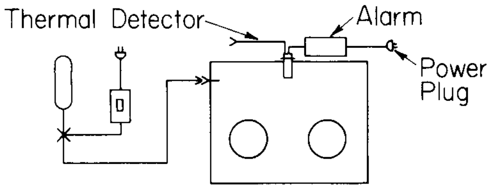
S_1	Detector Test Switch (Disarm Switch)
S_2	Manual Discharge Switch
D	Detector
N	Neon "Supervisory" Lamp
L	Lamp Indicating Detector Fired
P	Push-to-Test Button Switch

Note (1) Solenoid valve for use on small boxes. Squib-actuated valve will be substituted for systems on large boxes and cells.

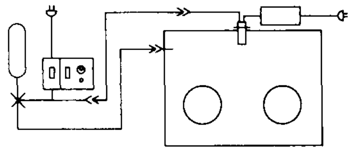
FIG. 1 WIRING OF SUPERVISORY SYSTEM



MANUAL-ELECTRIC SYSTEM



MANUAL-ELECTRIC SYSTEM WITH DETECTOR AND ALARM



AUTOMATIC SYSTEM

(1) The solenoid will be used on small boxes. On shielded cells and large glove boxes a squib-actuated valve will be substituted.

FIG. 2 CONNECTION DIAGRAMS FOR ALTERNATIVE FIRE PROTECTION MEASURES

Immediate supervision is responsible for determining whether or not fire protection is necessary and the degree of protection required. To assist these custodians in determining the degree of fire protection needed, fire protection guidelines were developed that are based on radiological hazards and are consistent with the general laboratory guides for radiological health protection for all operations. The recommended protective measures for four levels of hazard are illustrated in Table I. The connection diagrams for the "manual" or "manual-electric," "manual-electric with detector and alarm," and the "fully automatic" systems are shown in Figure 2. Of course, consideration of the amount and type of combustible materials, sources of ignition, method of handling, operations involved, and monetary value of equipment may, upon evaluation, increase the degree of protection indicated by analysis of the radiological hazards alone.

TABLE I

Guide for Minimum Fire Protection
Based on Radiological Hazard

	Tracer	Low	Medium	High
β, γ body burdens or γ dose rate	<100 body burdens	>100 body burdens	γ dose rate of <100 R/hr at 3 inches	γ dose rate of >100 R/hr at 3 inches
α curies	$<10^7$ d/m ^{244}Cm or equivalent	<1	>1, <500	>500
Extinguisher	Wall-mounted manual nearby	Wall-mounted manual in lab	Box-mounted manual - electric	Automatic system

The manually actuated extinguishers are actuated by pulling a safety pin and squeezing a handle grip that is basically the same as that on conventional small carbon dioxide extinguishers. The manual-electric system is actuated by closing the line switch (110-volt AC) indicated on Figure 2. To add detection and alarm to the manual system, a thermal detector is plugged into a small gasketed hole in the top of a glove box; the associated alarm box is connected to the normal 110-volt AC power (Figure 2).

To convert to fully automatic actuation, the manual switch box will be replaced by a supervisory control box which can then be plugged into the detector alarm unit. A connection is also made from the alarm unit to the building alarm system on all automatic systems and may be on the detection and alarm unit when used with the manual system. The automatic system has a manual discharge switch and a "disarm" switch to prevent discharge of the extinguisher so that either the detector can be tested without discharging the Halon 1301, or the system can be temporarily disarmed when the planned operations might actuate the extinguishing system. The switches are guarded with a snap-up cover (aircraft type) to avoid accidental tripping.

Normally, solenoid valves will be used on cylinders of Halon 1301 for small boxes (20-50 ft^3). The Halon 1301 will be released into a box directly through the bulkhead fitting for the snap-on connector without any nozzle. Squib-actuated valves will be used on cylinders for larger boxes to ensure quick release of Halon 1301 and, hence, rapid attainment of the desired 5-6% concentration of the agent in the box. Draft-tube nozzles will be used on the larger systems to expedite mixing of the agent with air, and flexible tubing will be used to connect the extinguisher to boxes rather than the rigid tubing corrently used on most of the manual systems. The additional cost of the flexible tubing is offset by the convenience and reduced cost of installation.

The squeeze-grip manual extinguishers (2-3/4 lb Halon 1301) presently mounted on selected glove boxes in the laboratory and on some of the small cells will be wall-mounted. These extinguishers will provide protection at low unit cost for laboratories with glove boxes in which the potential hazard is very low and will also provide backup for general fire protection in those laboratories equipped with electrically operated extinguishers whether they are manual or automatic.

The modular system also permits variation in the location and mounting of extinguishers as well as in the selection of the degree of protection, although in all installations the inlet for Halon 1301 into the containment is at the top. For example, extinguishers normally are mounted on the glove boxes (Figure 3), but in special cases, they may be mounted adjacent to boxes or under them (Figure 4) for convenience. There is also freedom in the method of mounting to make them readily accessible but to avoid potentially hazardous interference with normal operations conducted in glove boxes, particularly the transfer of process materials and waste (Figure 5).

TESTS OF AUTOMATIC SYSTEMS

Although tests conducted earlier in a small glove box (20 ft^3)* with squeeze-grip manual extinguishers clearly demonstrated the advantages of Halon 1301 over carbon dioxide for fires in containment systems, a prototype automatic system assembled by Fenwal was tested on the same box (Figure 6). The detector was inserted in the center of the top of the box, and the cylinder was charged with about 3/4 lb of Halon 1301. The times required for the detector to respond to the heat from n-butanol varied from 3 sec with an 81 $in.^2$ surface (9 x 9 in. pan) to about 30 sec for a 7 $in.^2$ area (3-in.-diameter cup). With two loosely wadded paper towels in the pan, the response time was about 25 sec; and with two wadded towels in a one-pint ice cream carton (11 $in.^2$ opening), the response time was 45 sec. There was little variation in response times for different locations of the fires in the box except when the fire was directly under the detector. Upon discharge of the Halon 1301, the time required to extinguish the fires was less than two seconds.

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* Glove box 40 ft wide, 3 ft high, and 20 in. deep.

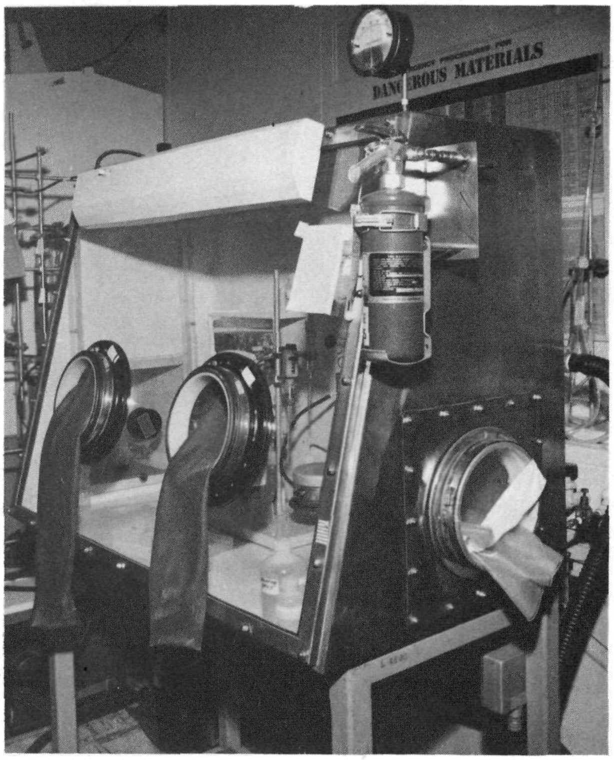

FIG. 3 EXTINGUISHER MOUNTED ON GLOVE BOX

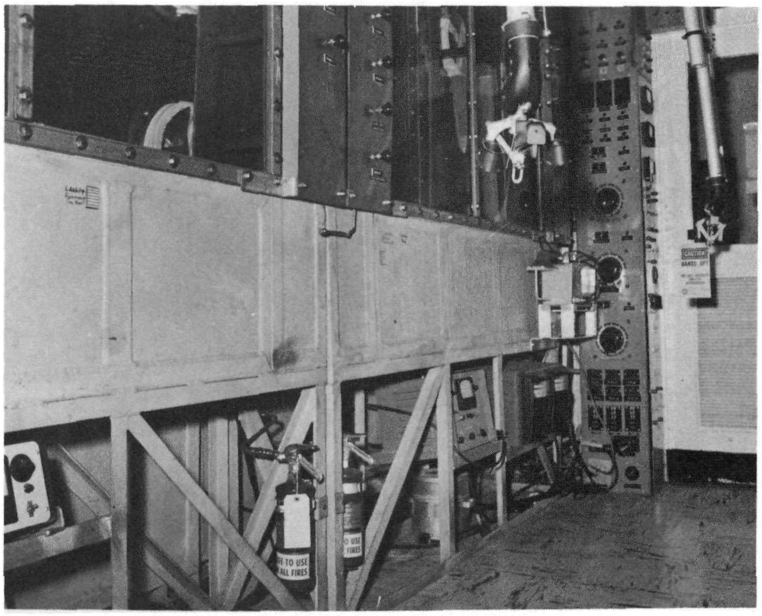

FIG. 4 EXTINGUISHER MOUNTED UNDER GLOVE BOX

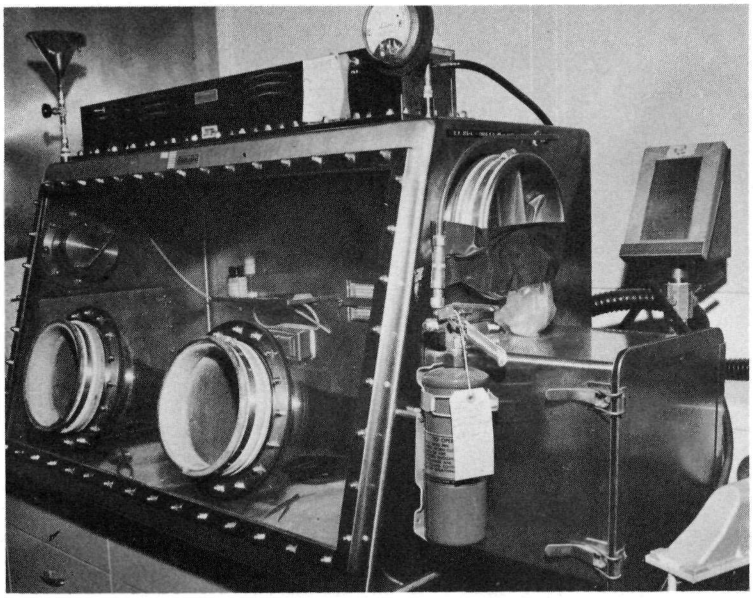

FIG. 5 EXTINGUISHER MOUNTED ON GLOVE BOX WITH FLEXIBLE HOSE

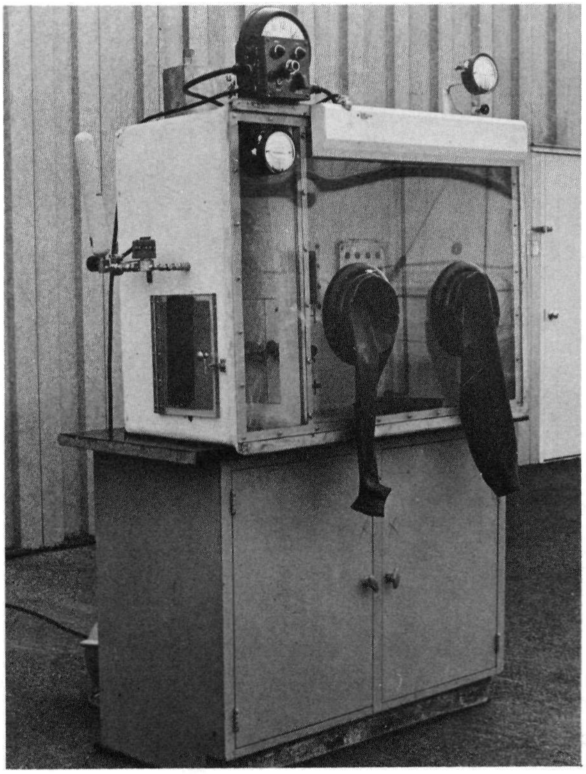

FIG. 6 PROTOTYPE AUTOMATIC EXTINGUISHING SYSTEM

FIRE DETECTION SYSTEMS IN GLOVEBOXES

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April 14, 1971

FIRE DETECTION SYSTEMS

IN

GLOVE BOXES

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Introduction

This paper is presented to discuss the use and application of a new detector system. This system is primarily intended for installation in glove boxes which have a high acid environment, but it also has features which make it desirable where containment of radioactive materials is a necessity. In addition, this paper will also discuss the modifications of a well known detector, which makes it desirable for this same use.

These problems of containment and chemical atmospheres were no doubt the factors evaluated by the Fire Protection and Safety Engineers in the early 60's at the Hanford Project. At that time, they selected a continuous line fire detector for installation in many of their glove boxes. This type of system has an advantage over the spot type, which may have dead areas in its span of coverage.

The unit chosen was the Fenwall Continuous Fire Detection System, which utilizes a tubing containing a eutectic salt mixture. This salt mixture melts at a predetermined elevated temperature and becomes electrically conductive. In this state it can then conduct a current between its central conductor and the outer metallic sheath. Figure 1 shows the sensing element for this system.

An open circuit voltage of 24 volts is impressed on the central conductor and the sheath. When fire or heat fuses the salt, the electrical circuit is completed in a fashion similar to the closing of a set of contacts. The current flow then activates a zone alarm and other alarm functions as desired. An additional desirable feature of the eutectic salt detector system is that it is self-restoring upon resolidification of the salt.

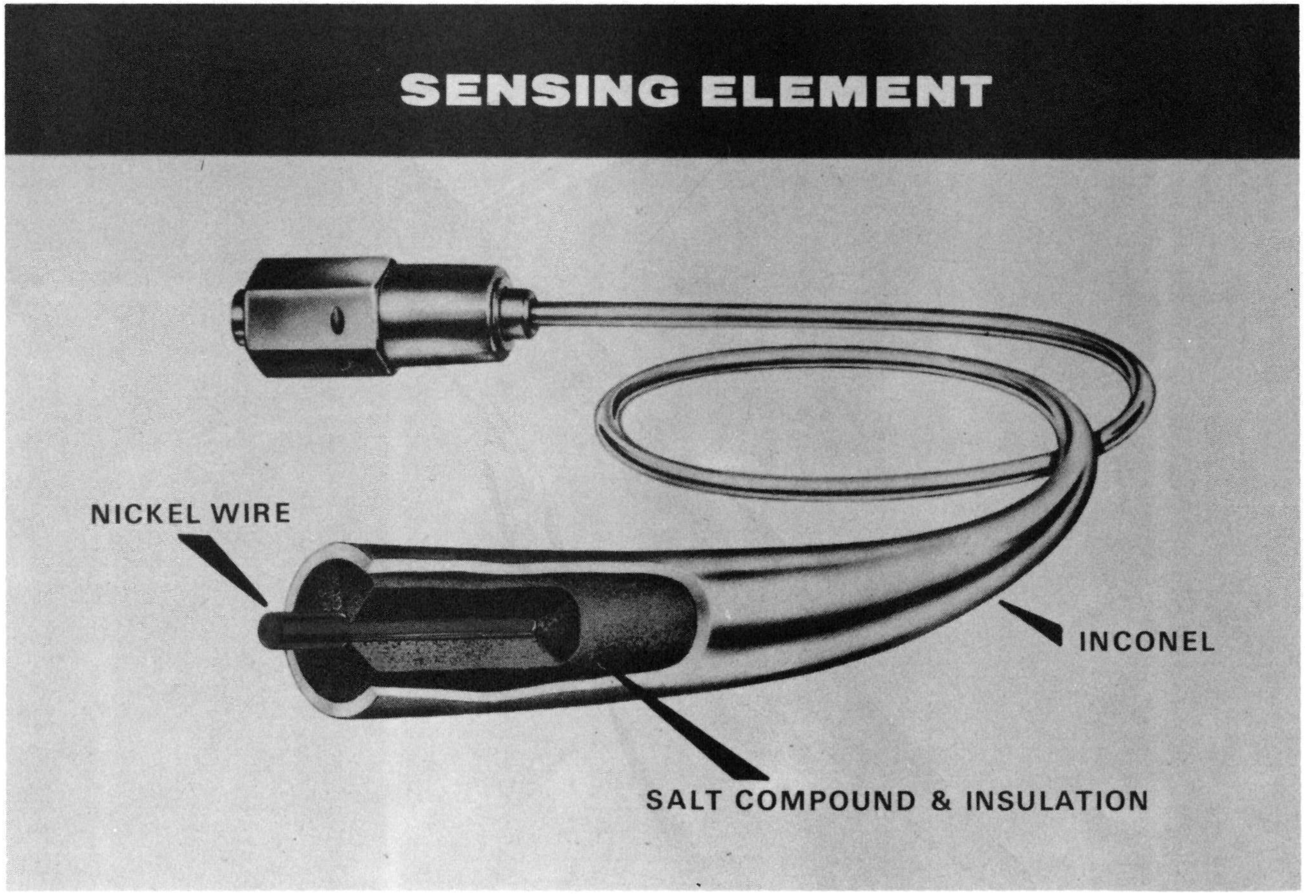

SENSING ELEMENT
NICKEL WIRE
INCONEL
SALT COMPOUND & INSULATION

FIGURE 1

The choice of this system might not be questioned today if it were not for corrosion and its effect on the sheath of this detector. The jacket is Inconel, and Figure 2 shows a small section of one of these systems as it is installed and in service today. Figure 3, however, shows a typical section of a system which has corroded so badly that it is removed from serice. This unit is installed in a glove box used for Pu recovery operations. In an environment such as this, when the concentrations of nitric acid are 35% and more, the Inconel Sheath has a usable operating life of about five years. The failures cause shorting of the detector and it can no longer perform its intended functions.

At the first few signs of these failures, the operating contractor (Atlantic Richfield Hanford Company) requested Vitro/Hanford Engineering Services to investigate and recommend a course of action. We then initiated an in-depth study to determine the most acceptable and economical solution.

Factors Governing Selection of a Fire Detection System for Pu Recovery Glove Boxes

Our study disclosed that stainless steel was an optimum material for satisfactory operating life in the concentrated acid environment of the glove boxes. The boxes themselves are of stainless construction; and, if the detector system could be of the same base material, we would have a high system integrity. At the same time, the operating life would be extended greatly beyond the five years of the Inconel.

A check with Fenwall, however, soon disclosed that the Continuous Strip Detection System was not available with a stainless sheath. Since simple replacement was precluded, we continued our study of the factors pertinent to installation of another type of system. Containment of radioactive material and ease of installation were important additional factors. Added to this, we wished to provide rate of temperature rise detection in addition to fixed temperature detection ability. We also wished to secure automatic resetting of our detectors so that they could be periodically tested in place.

One of the UL Inc. listed devices which met these requirements was the stainless model of the Fenwall Detect-a-Fire. It is a combination fixed temperature and rate compensated detector with several fixed temperature settings available. The brass hex head and soldered plug in the bottom, however, would be affected by the acid environment and these conditions made it unsuitable for our use.

The New Glove Box Detector Systems

A modification of this system soon evolved. It provided for a combination fixed temperature, rate-or-rise type of detection system which was fully compatible with our operating requirements. The system, as developed, used

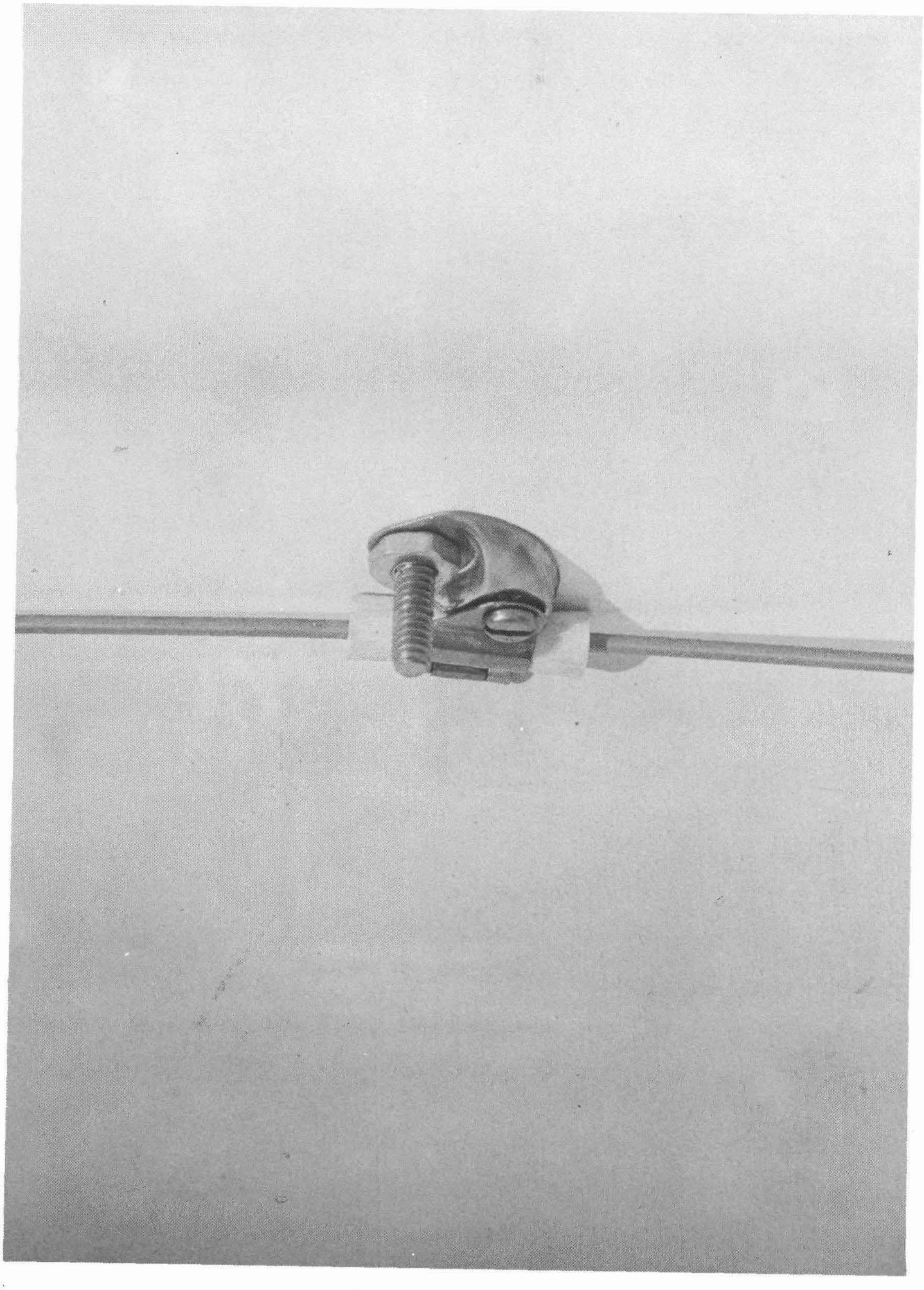


FIGURE 2

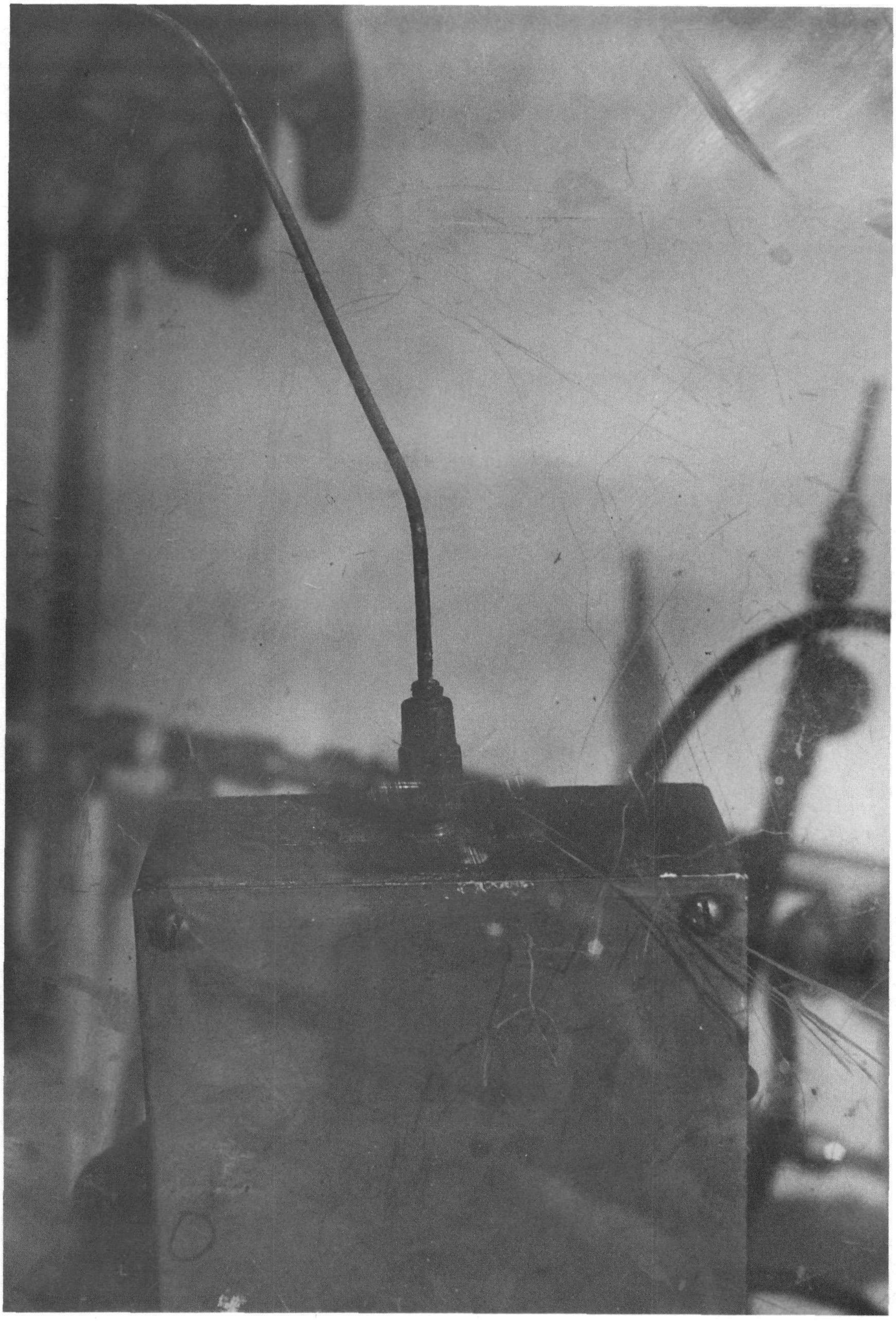


FIGURE 3

a Fenwall Thermoswitch in a stainless steel well. These components are shown in Figure 4. The thermoswitch is Model No. 17021-0 and is adjustable over a range of -100°F to 400°F. Its internal operating mechanism is shown in Figure 5. The initial installations were made with units calibrated to 160°F. The stainless steel sheath is Model No. 11201-0 and is 321 stainless. The current cost of these components is approximately \$35 and compares favorably with the present unit price of \$22 for the UL listed Detect-a-Fire.

The well-type units were installed on 8-ft centers in the longer glove boxes thus giving us a very favorable cost compared to the replacement costs for the continuous strip system. The mounting details for a unit are shown in Figure 6; and, from this, you will note the simplicity and yet the means by which containment is achieved. Figure 7 shows an installation in an operating glove box and is one unit in a line of four. The electrical circuit we are using is that of normally open alarm contacts and end-of-line resistor for complete electrical supervision of the system.

Fire tests were conducted to ascertain the response characteristics of the thermoswitch in a well as compared to a standard Detect-a-Fire. Two series were planned so that both fixed temperature and rate-of-rise performance could be evaluated in a glove box of 6 ft^3 volume.

The results of these tests are shown in Figure 8, which is Test No. 1. Test A was planned with a slow heat build up to simulate a Class A type of fire and thus require the detector to operate at its fixed temperature setting. You will note that the response of the well type unit compares very favorably with the standard unit experiencing a lag in the range of only 10-20%.

In Tests B and C, we prepared tests to determine response characteristics with a faster acting hydrocarbon fire. For these tests, we used a small alcohol pan fire of only .08 ft^2 area, 15cc of fuel, and the same detectors mounted at top center of the box. These two series of tests were identical except that the fire pan was moved from the extreme inlet side to the extreme outlet side. During these tests, we had an air exchange rate of almost four air changes per minute which is a rather stringent requirement.

Test No. 2 was conducted in a larger glove box of 30 ft^3 volume, which is more nearly a typical operating size. The results of these tests are shown in Figure 9. In Tests A and B, we wished to again determine response characteristics with a fast acting hydrocarbon fire. For these tests, we used a fire pan of 0.88 ft^2 with 50cc and 60cc of fuel for each test respectively. The smaller quantity of fuel was used for the fire directly under the detectors, and the larger quantity with the fire pan at the far end of the glove box. In both series, the detectors were directly in front of the vent at one end of the box as it was impossible to mount the units at top center of the box. One of the four glove ports on the box was left open and no forced ventilation was provided. The test was conducted outside, however, and a breeze blowing into the open port affected the response time of Unit No. 1 in the last two tests.

In Tests No. 1 and No. 2, a standard Fenwall Detect-a-Fire with a 160°F setting (Test Unit No. 2) and a Fenwall Thermoswitch, set at 150°F, in a well (Test Unit No. 1) were utilized for testing.

FIGURE 4

PRINCIPLE OF OPERATION

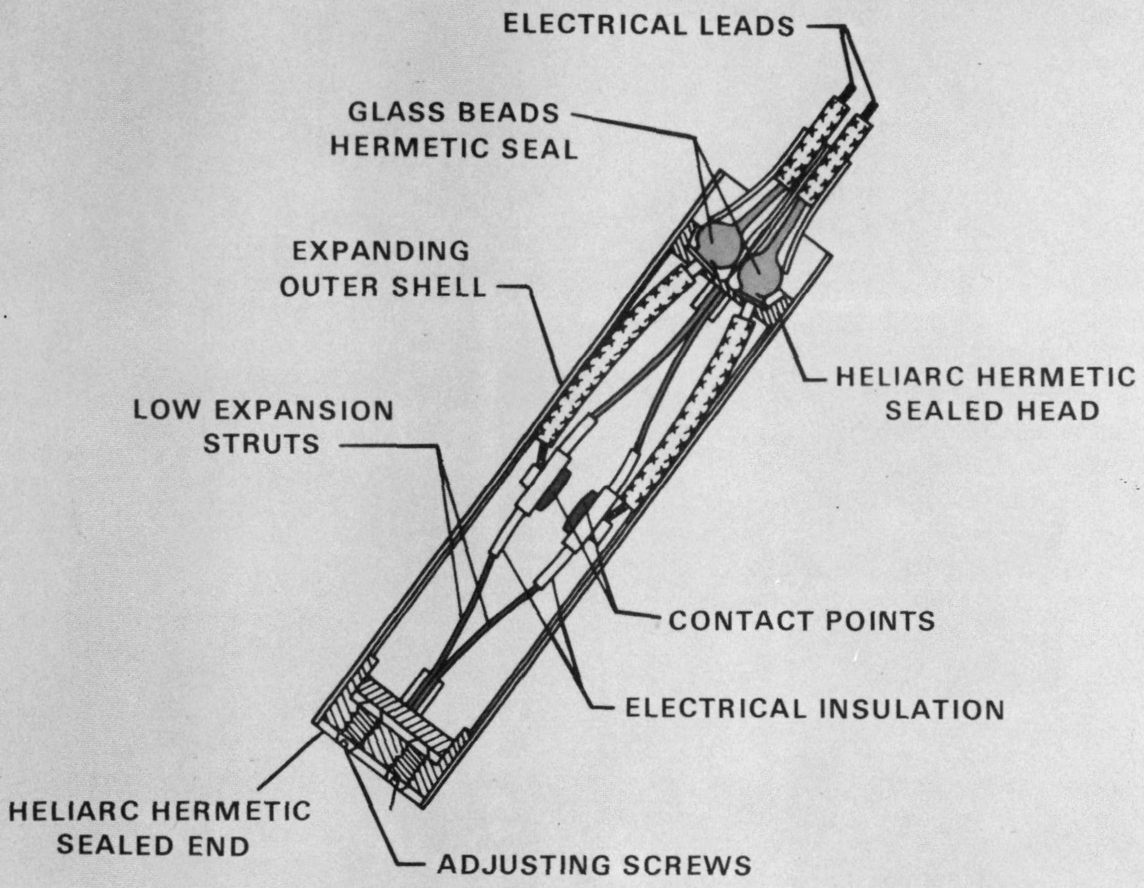


FIGURE 5

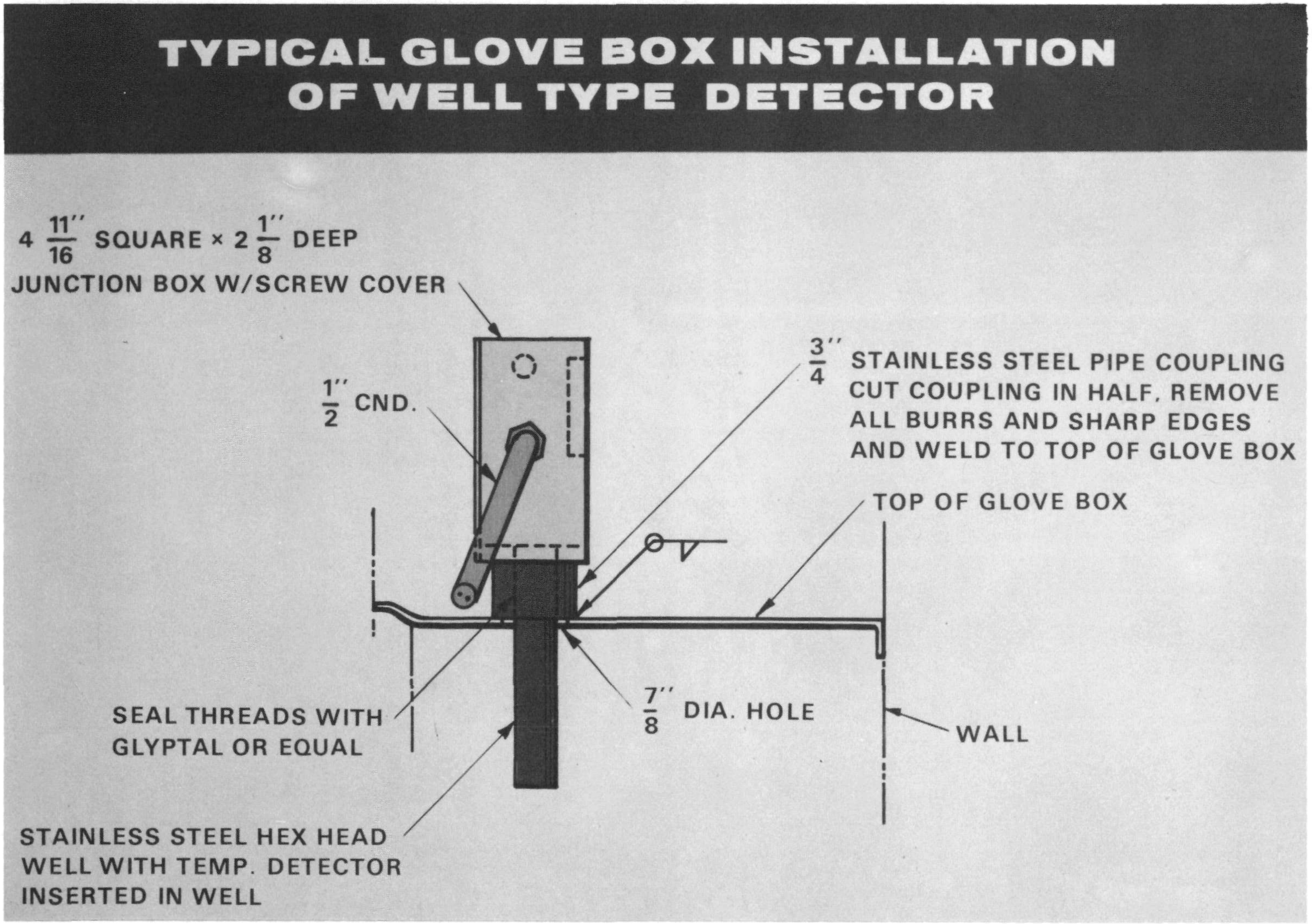

TYPICAL GLOVE BOX INSTALLATION OF WELL TYPE DETECTOR
4 11/16" SQUARE × 2 1/8" DEEP JUNCTION BOX W/SCREW COVER
1/2" CND.
3/4" STAINLESS STEEL PIPE COUPLING CUT COUPLING IN HALF, REMOVE ALL BURRS AND SHARP EDGES AND WELD TO TOP OF GLOVE BOX
TOP OF GLOVE BOX
SEAL THREADS WITH GLYPTAL OR EQUAL
7/8" DIA. HOLE
WALL
STAINLESS STEEL HEX HEAD WELL WITH TEMP. DETECTOR INSERTED IN WELL

FIGURE 6

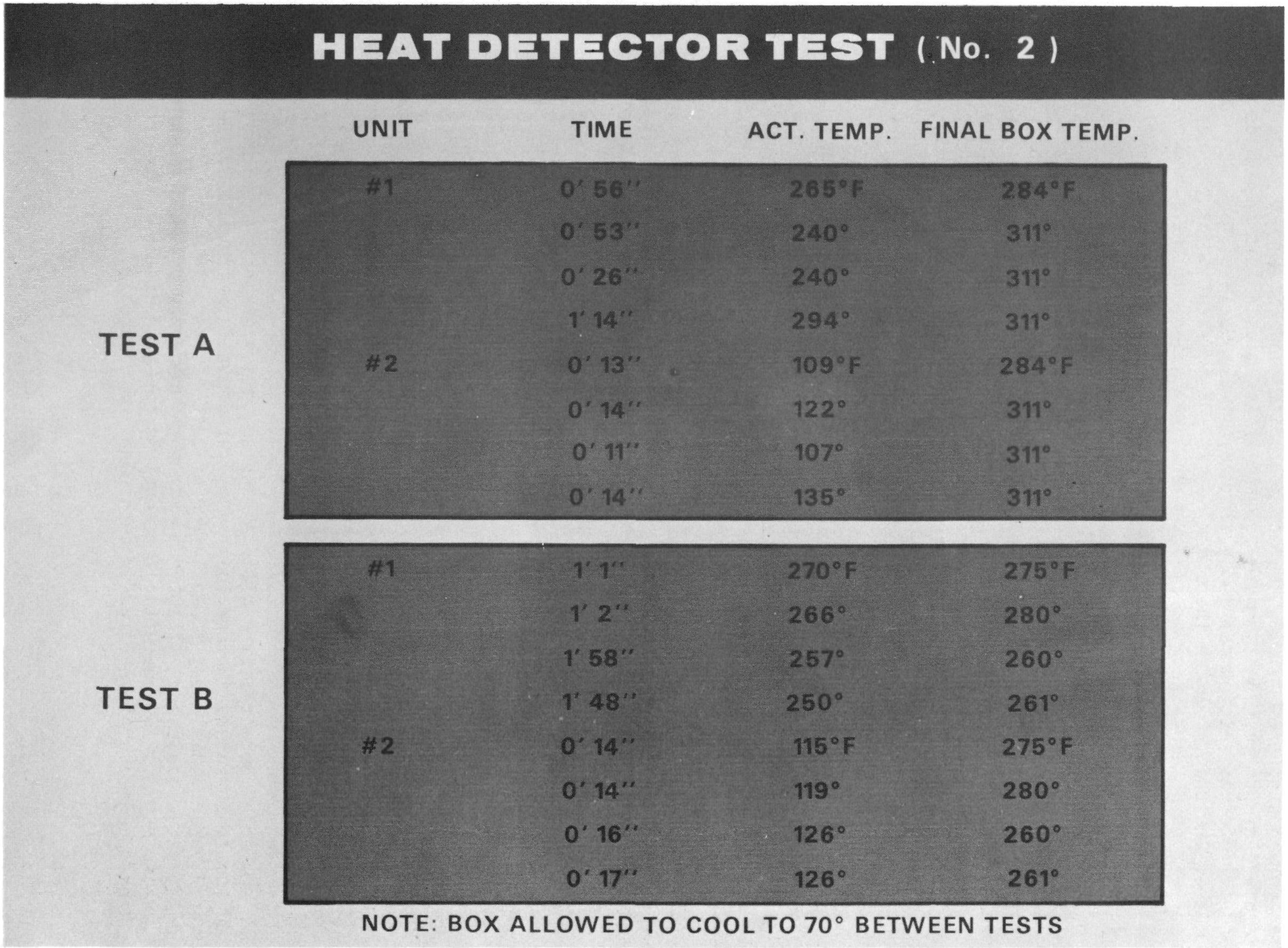
FIGURE 7

HEAT DETECTOR TEST (No. 1)

	UNIT	TIME	ACT. TEMP.	FINAL BOX TEMP.
TEST A	#1	7′ 25″	195°F	
	#2	6′ 45″	182°F	
		BOX SEALED AGAINST LEAKAGE		
	#1	4′ 25″	195°	
		4′ 10″	197°	
	#2	3′ 35″	185°	
		3′ 45″	185°	
TEST B	#1	1′ 50″	210°F	210°
		1′ 55″	205°	210°
	#2	0′ 31″	120°	210°
		0′ 30″	115°	210°
TEST C	#1	1′ 0″	215°F	250°F
		1′ 15″	220°	250°F
	#2	0′ 21″	110°	250°F
		0′ 13″	90°	250°F

NOTE: Box Allowed to Cool to 75°F Prior to Start of Each Test

FIGURE 8



HEAT DETECTOR TEST (No. 2)

	UNIT	TIME	ACT. TEMP.	FINAL BOX TEMP.
TEST A	#1	0′ 56″	265°F	284°F
		0′ 53″	240°	311°
		0′ 26″	240°	311°
		1′ 14″	294°	311°
	#2	0′ 13″	109°F	284°F
		0′ 14″	122°	311°
		0′ 11″	107°	311°
		0′ 14″	135°	311°
TEST B	#1	1′ 1″	270°F	275°F
		1′ 2″	266°	280°
		1′ 58″	257°	260°
		1′ 48″	250°	261°
	#2	0′ 14″	115°F	275°F
		0′ 14″	119°	280°
		0′ 16″	126°	260°
		0′ 17″	126°	261°

NOTE: BOX ALLOWED TO COOL TO 70° BETWEEN TESTS

FIGURE 9

This data shows that the well-type detector performs reliably for either the slow build-up type of fire or the more rapid developing flammable liquid type fire. It has a time lag inherent to the use of the insulating well, but within the necessary limits of maximum temperature in our Pu recovery glove boxes, since the replaced units had a fixed operating temperature of 255°F. The thermoswitch itself is adjustable over a wide range, and any desired setting may be obtained by simulated test and simple adjustment of the unit. It can therefore be set and calibrated for its desired application by lowering its fixed temperature rating and thereby decreasing its operating time.

The second type of detector to discuss is the standard Fenwall Detect-a-Fire in a stainless jacket. Conversations with the manufacturer regarding some of our special requirements persuaded him to "remodel" his standard stainless production model. He has assigned this new unit a model number of 28021-S-140. The only significant changes are that of heliarc welding a cap on the end of the sensing sheath and replacing the brass hex head with a stainless steel unit. These replacements should overcome any objection to its use in high acid environments, which could cause deterioration and degradation of the thermoswitch. Its life expectancy should, therefore, be equal to the assembled well-type unit described earlier and which we have had in service for two years or more.

Figure 10 shows a picture of the standard detector. The new unit, not yet manufactured, will have the same physical size and will perform, we expect, in an identical fashion to this standard run unit. The cost is reported to be in the order of $35. The first lot of 100 will be utilized in a glove box installation in a Pu r&d laboratory managed by WADCO at the Hanford Project. The design for these systems has just been completed by Vitro/ Hanford Engineering Services. In addition, we are currently in design for a second glove box system in another Pu facility operated by this contractor and will utilize this same new detector for that installation.

Summary

The investigation described here served to develop two new devices for inclusion in an already extensive line of fire detection units. Both have the inherent resistivity necessary for installation in high acid environments. We also feel that they have the necessary capability for reliable fire detection under these circumstances, as they have demonstrated in the tests that they will reliably detect both Class A and Class B fires before glove box integrity becomes a factor.

The new model Detect-a-Fire will have an advantage of shorter response time and should be selected where that is of paramount importance. The well-type unit, however, has an added advantage of ease of installation in existing contaminated equipment. Its thermoswitch can be removed, tested, or recalibrated in a clean area without removal of the well itself. Thus it has a higher degree of integrity for contamination control.

FIGURE 10

With a choice of two units, the Fire Protection Engineer can specify the unit or combination of units which will be most desirable for a particular application. Production-type glove boxes, where standard conditions exist, can utilize the new model Detect-a-Fire in a selected temperature range. R&d-type glove boxes, which could be subject to wide temperature ranges, may best utilize the well-type with its features of recalibration while retaining contamination control. Both devices deserve consideration for future special hazard fire detection systems.

FIRE PROTECTION TEST FACILITY FOR CELLS AND CAVES

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April 14, 1971

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ABSTRACT

A fire protection test facility has been constructed by ARHCO at Richland consisting of a simulated hot cell and a filtered exhaust system. The purpose is to study fires and fire protection components and system under conditions similar to those of typical heavily shielded remote or manipulator operated hot cells and in the attached filtered ventilation exhaust systems.

Tests will be performed to establish criteria for effective fire protection systems, components and methods of operation for both the hot cells and their exhaust systems.

INTRODUCTION

The nuclear facilities operated by ARHCO at Richland include five chemical processing plants which use remote process cells. Two of them, the Purex Separation Plant and the B Plant Fission Products Processing Plant are still in operation. Manipulator hot cells are used in one laboratory and will be included in the Waste Encapsulation and Storage Facilities now under construction. There are several other caves and hot cells at the Richland Plant that are operated by other contractors.

At the time the original plants were built, a great deal was yet to be learned about the fire danger, and in fact about the nature of the probable airborne effluents from these processes. With the growing knowledge of the nature of these radioactive effluents, has come the realization that they can be released as a result of a fire, either in the cell or in the ventilation exhaust system.

The Atlantic Richfield Hanford Company is vitally interested in achieving a maximum degree of safety in the facilities under its contract responsibility. We, therefore, are taking whatever action is necessary to provide the optimum fire protection for these facilities. An audit of the existing fire protection equipment and methods showed that improvements could be made in many instances. Steps were taken to improve or replace the fire protection at many places in the plants. In most cases there were familiar protection problems and yielded to standard fire protection methods. Others, notably the heavily shielded and remotely operated process cells, presented a more difficult problem. These cells sometimes contain hundreds of gallons of organic solvent and other combustibles and have a highly radioactive corrosive environment. All of them are equipped with the originally installed manually operated water spray extinguishing systems. Fire detection is provided by some form of temperature detector except at the Purex Plant where flame detectors are used.

We want to provide the best fire protection system for these caves and hot cells that we can devise. Improvements in speed and reliability of fire detection, effectiveness and promptness of extinguishing, and the positive containment of radioactive contamination from a fire are felt to be possible. With this in mind, a program was undertaken to find a fire protection system or systems to include:

1. A fire detector which will detect a fire, without fail, in a highly radioactive corrosive environment, will be easily and routinely checkable for operation, will require little maintenance, and will not produce false alarms under any reasonable conditions.

2. An extinguishing system which will automatically extinguish any fire which might develop in large remote or manipulator cells, will cause a minimum of damage to facilities and equipment, and will create the least problem of waste disposal.

3. A protected filtered exhaust system which will either contain the radioactive products-of-combustion within the cell or create a condition in which the exhaust air can continue to be safely filtered for the duration of the fire.

In addition, we want to look for methods of making our exhaust filtration system safer from fire whether from a fire within the filter system (which might be started by a concentration of radioactivity) or from a fire elsewhere in the exhaust system.

In order to carry out this program, we have erected a test facility near one of the deactivated chemical process buildings.

DESCRIPTION OF FACILITY

Figure 1 shows a general view of the facility. The simulated hot cell is enclosed within the corrugated shell. It is an insulated box which can be varied in height by lowering the cover. The height was chosen to represent the deepest remote process cell at one of the chemical separations plants operated by ARHCO. The cover can be lowered until it simulates the size of any of the several smaller cells.

The cell contains an old tank and a mass of old used pipes to simulate the vessels and remote jumpers of an active remote process cell.

Fires of various types will be set manually at the base of the cell. There are provisions for adding organic solvent while the fire is in progress to simulate the effect of a broken organic pipe or leaking vessel.

PHOTOGRAPH OF TEST FACILITY

FIGURE 1

Perhaps in the future, we may install a glove box within the cell for tests of some aspects of glove box fire protection.

The ventilation duct emerging from the side of the cell is located at about the same height as the exhaust exit of a typical remote process cell. Its size is chosen to represent the exhaust flow rate and velocity from a cell of this size in its normal operating mode with the cell cover blocks in place.

The exhaust ductwork length was chosen to give some opportunity to study the effect of distance on the smoke, products-of-combustion and other exhaust. We expect to find cooling, stratification, agglomeration and settling of smoke particles, for example.

There is an entry point for dilution air into the duct near the exit of the duct from the cell. With this, we expect to learn the effects on the smoke of dilution with the exhaust air from one or several cells parallel to the one containing the fire. We expect that there will be cooling, stratification, and some affect on the quality of the smoke. This will have an affect on the action that will be necessary to clean and cool the gas before it enters the filter.

At the right end of the picture is the air treatment equipment. In place for the first test is a standard sprayed chamber air washer with four banks of sprays, each of which can be operated separately. It is equipped for the initial tests with a wave plate moisture separator and is followed by a moisture separator section that uses the type of demister pads that have been selected by Savannah River Plant for protection of HEPA filters.

The fan is a standard centrifugal model which can be modulated by a variable inlet vane damper to give flow up to 300 percent of that required and pressures up to six inches of water.

As the tests progress, we will be able to insert or remove duct sections or add special sections as we search for combinations that will give us the information that we want.

Figure 2 shows a diagram of the facility as it is set up for the initial tests. Instruments include temperature at nine points, pressure at four points, flows at two points and differential pressures at three points. The fire detectors are located in the cell and in the entrance to the exhaust duct. Smoke and dust measurements are shown both upstream and downstream of the air cleaning equipment. The sensing points of any of these instruments may be moved if it proves to be desirable. Some of the combinations of exhaust components that are contemplated for future tests are shown on Figure 3.

The top diagram shows the air washer used with a segment of fiberglass filter similar to the first layer of the deep bed fiberglass filter in use at Richland. This arrangement can be used to find the affect of water, heat, and smoke on this type of filter. The instrumentation for this arrangement is about the same as for the previous diagram.

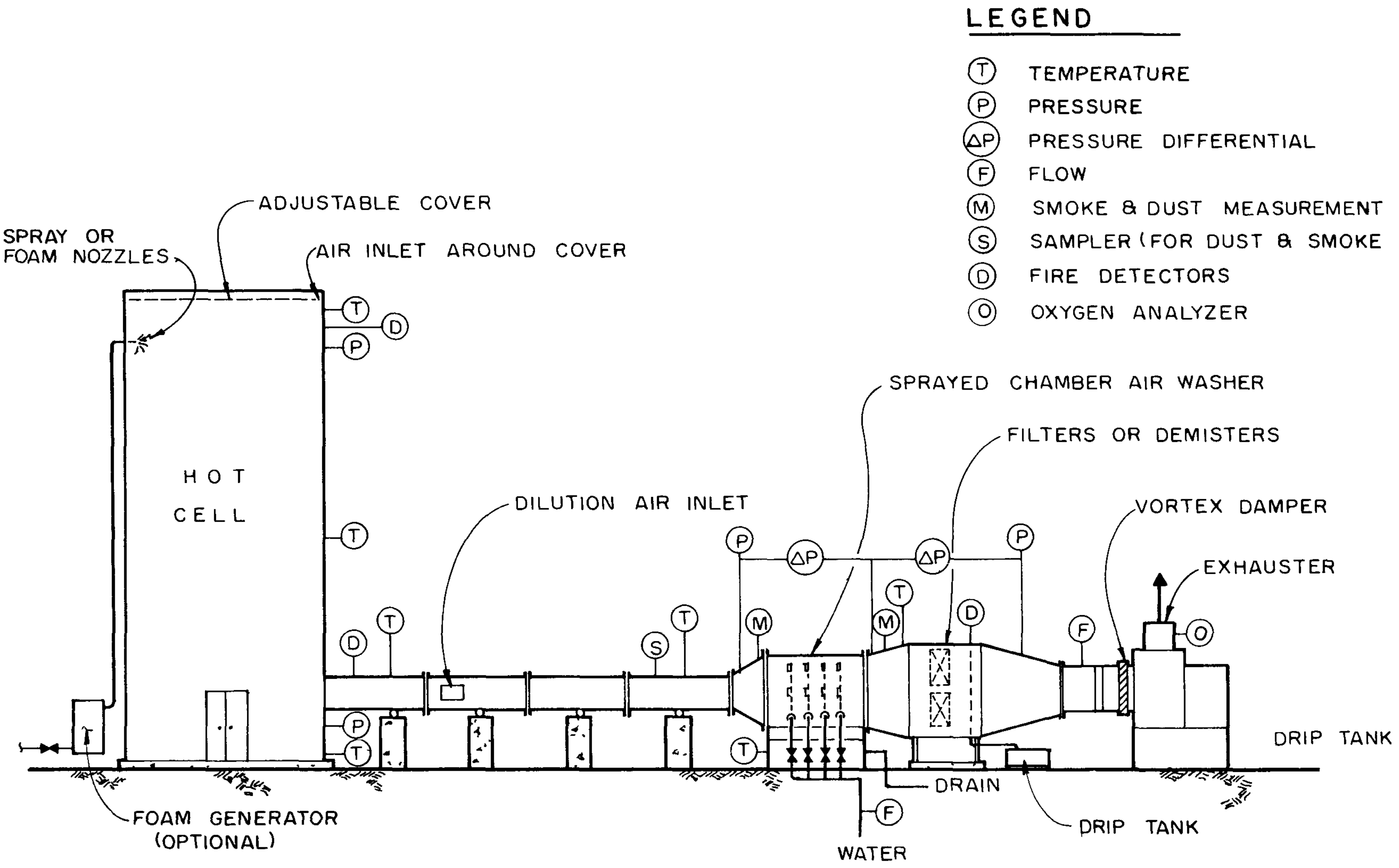
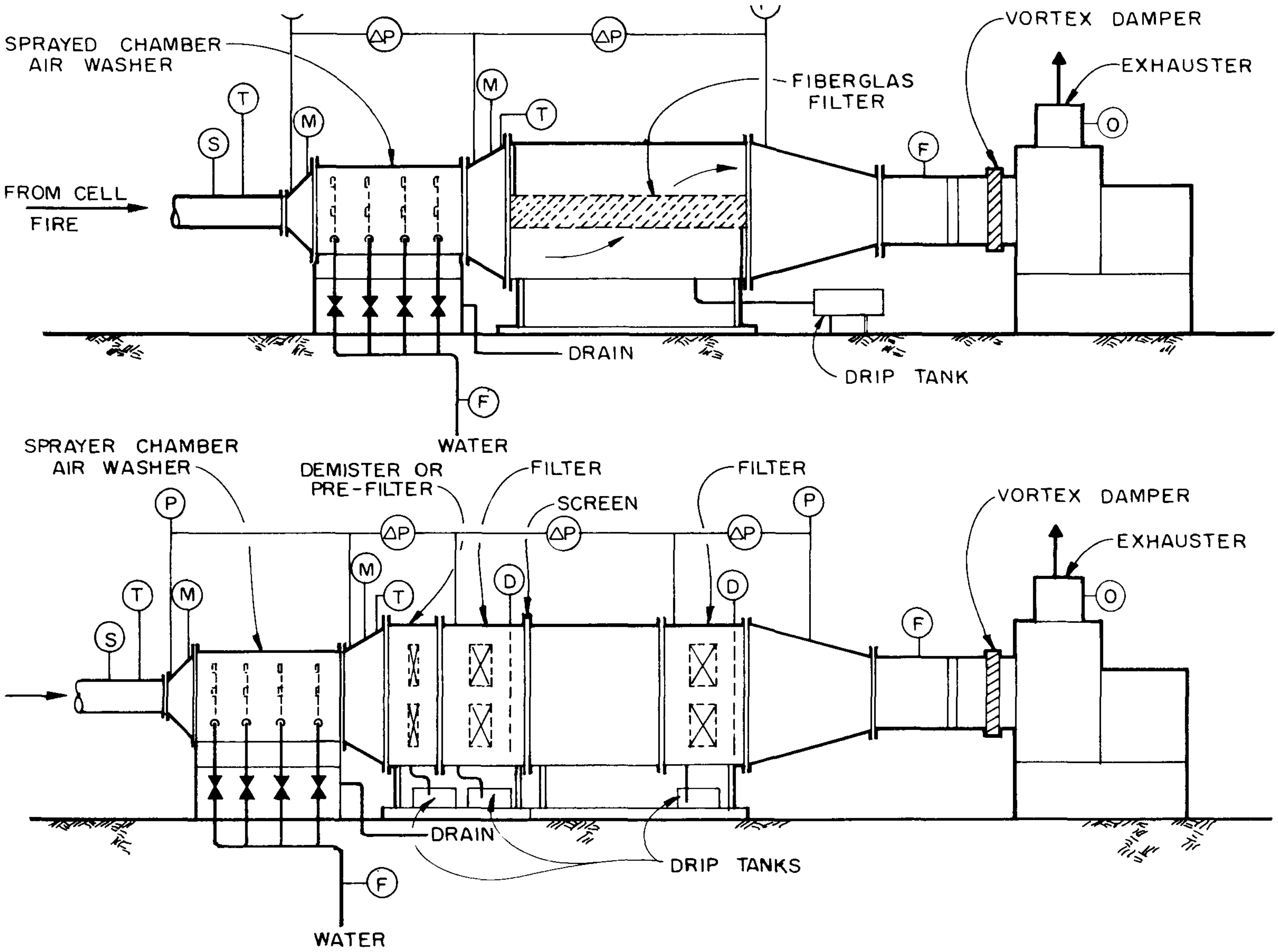


DIAGRAM OF FIRE TEST FACILITY

FIGURE 2



VARIATIONS OF EXHAUST SYSTEM

FIGURE 3

The lower diagram is a variation which includes two banks of filters and a demister after the air washer. The last filter bank consists of HEPA filters. The other may be of another type depending on the purpose of the test. There are many other combinations that may be used especially as we get into the problem of detection and extinguishing of filter fires.

Figure 4 shows the instrument panel which is located in the control building. The instruments are mostly indicating instruments which will be photographed by a sequencing camera while the tests are in progress. The clock will be used to relate the instruments readings to time. Upon completion of a test, additional data can be superimposed onto the panelboard so that all data on a given test can be recorded on a single roll of film.

It is planned to photograph other phases of the tests as well, such as movies of the progress of the fire and of operation of the washer and the discharge of the fan. In addition, instruments are provided for measuring the soot concentration in the gas stream at two points, in order to find the effectiveness of the air cleaning devices. We also have sample ports for collecting samples for studying the characteristics of the soot and smoke. An oxygen analyzer is provided for intermittent measurement of the oxygen content of the exhaust gases. As the testing progresses, we have allowed for adding to or altering the instrumentation if desired.

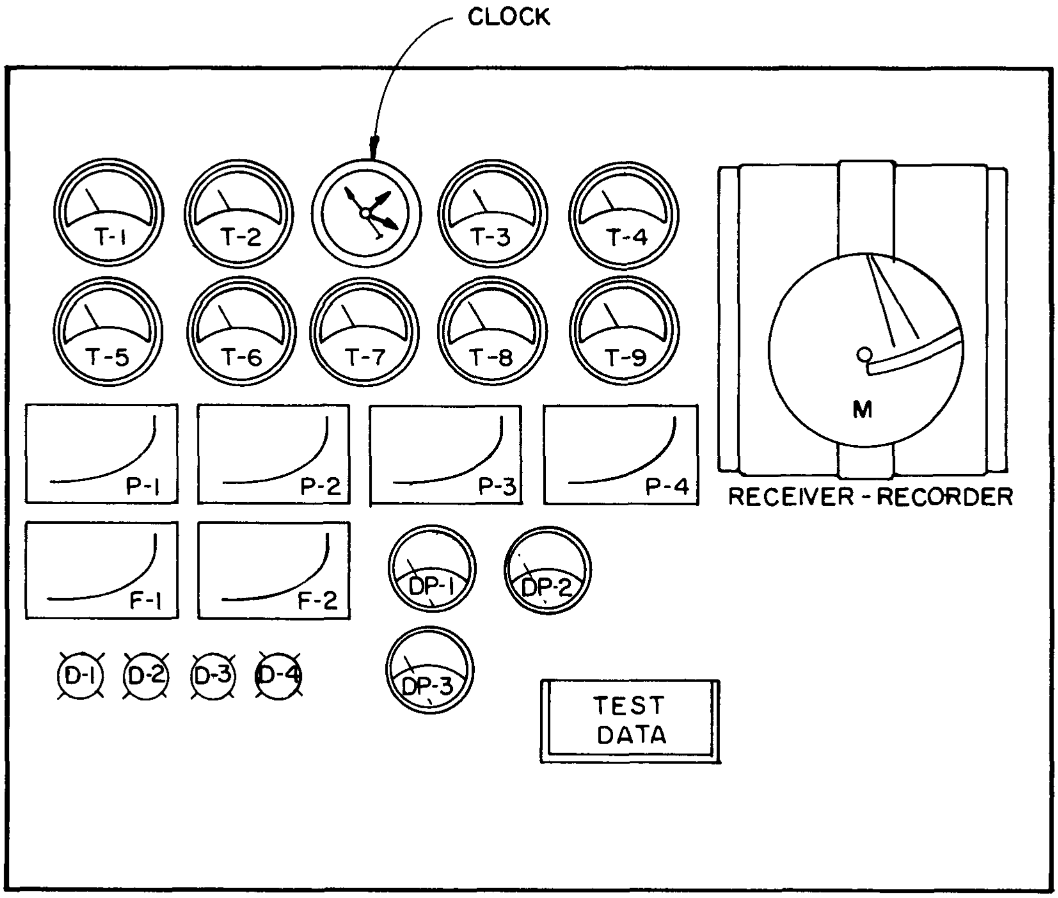
Figure 5 shows a glove box which is set up adjacent to the other facility. It has been used to test hood fires using high expansion foam and recently was set up to demonstrate the effectiveness of Halon on glove box fires. It is available for testing other glove box fire protection equipment, but there are no plans for any such tests at present.

TESTS TO BE PERFORMED

The tests to be performed in this facility fall into three categories:

1. Tests of fire detection and extinguishing equipment under conditions similar to those to be found in typical remote process cells, caves or hot cells. This will include study of the actual fires for temperature, air flow pattern, and smoke quantities and particle characteristics. It will allow us to compare fire detector apparatus, select detector locations and compare extinguishing media and apparatus. One facet of this is to compare extinguishing media for quantity required and compatibility with the process waste system. This is an important consideration since the extinguishing media will become part of this waste.

2. Tests of methods of containing the contaminated smoke and heat generated by a fire. We hope to develop criteria for an automatic system that will be unfailingly available when a fire is detected in a cell, to cool and clean the smoke laden air so that we can continue to operate after a small fire, or safely shut down after a maximum credible fire. This system should require the minimum first cost and upkeep consistent with this performance.

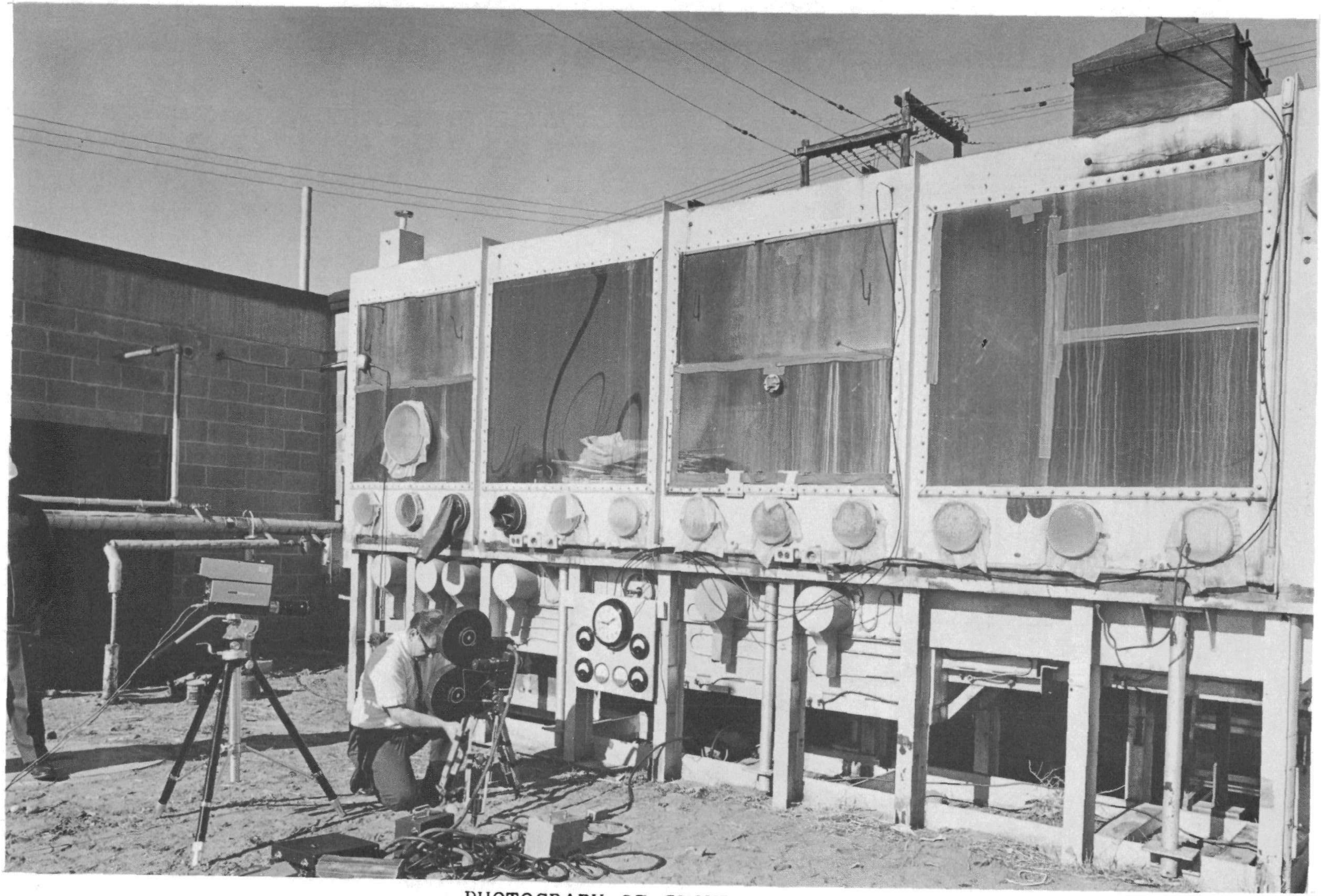


LEGEND

T — TEMPERATURE GAGE
P — PRESSURE GAGE
F — FLOW GAGE
DP— DIFFERENTIAL PRESSURE GAGE
M — SMOKE & DUST MEASUREMENT
D — FIRE DETECTOR INDICATOR

DIAGRAM OF INSTRUMENT PANEL

FIGURE 4



PHOTOGRAPH OF GLOVE BOX

FIGURE 5

3. Tests of filter fire detection and suppression methods. A good deal of work has been done by others on this problem. Since it fits in with the rest of the program, we intend to try a few ideas for detecting and extinguishing fires in filters. We hope to be able to detect and extinguish a filter fire while ventilation flow is continued, without plugging the filters or destroying their effectiveness.

As an overall goal, we want to collect numerical data that will enable us to establish criteria for an integrated fire protection system for the remote process cells, caves and hot cells at Richland. We expect that these data will be useful to designers of hot cells and caves at other locations.

We welcome suggestions from others that are interested in this problem. Perhaps investigation into specific problems that they have in their area can be included in the program.

ENGINEERING FIRE-SUPPRESSION SYSTEMS FOR LARGE VOLUMES USING HALON 1301

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April 14, 1971

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Abstract

Engineering studies of Halon 1301 fire-suppression systems for large volumes (up to 10,000 cu ft) are summarized and the results are presented. Guidelines are given for designing such a system, with particular attention paid to fire detectors, alarms and controls, and the Halon distribution arrangement.

Introduction

This report summarizes engineering studies of prototype fire-suppression systems for equipment test trailers and field assembly buildings, the details of which are reported separately.[1,2] These discussions deal with the protection of environments rather than the interiors of glove boxes. The volumes considered range from 3000 to 10,000 cu ft. However, we believe that extrapolations can reasonably be made to larger volumes. We should note in passing that although Halon 1301 will not extinguish an actively burning plutonium fire, it will extinguish Class A and Class B fires which could ignite plutonium.

Note further that the decision to use Halon 1301 as an extinguishing agent for a particular application should be based on an analysis of the problems involved and the relative cost effectiveness as compared with other extinguishers such as water sprinkler systems or carbon dioxide flooding systems.

System Analysis

As shown in Table I there are four principal elements to be considered in the design of area fire-protection systems. These are detection methods, the choice of the extinguishant, the means of obtaining the power for the system, and the alarm and control methods. In our view, fires are most easily detected by an ionization type of smoke detector coupled with a temperature-compensated rate-of-rise heat detector. The latter is useful in the case of flash fires. The extinguishant can be one of three: water delivered through a sprinkler system, carbon dioxide in a flooding system, or the more recently developed Halon 1301 (or even Halon 1211) in a flooding system.

Work done under the auspices of the U.S. Atomic Energy Commission.

Table I. Elements of area fire-protection systems.

Detection	Smoke, by ionization detectors.
	Heat, by temperature-compensated rate-of-rise heat detectors.
Extinguishant	Water, carbon dioxide, or Halon.
Power	Batteries plus charger, hence 24-V dc system.
Alarm/control	Normal condition (green). Trouble condition (yellow) plus audible whistle Alarm condition (red) plus audible bell. Local and remote signaling. Time delay plus override (optional).

Table II. Comparison of extinguishants required to protect a 40-ft trailer.

Extinguishant	Weight (lb)	Concentration	Toxic?	Cleanup required?	Cost of extinguishant
Water	3300	1 gal/ft^2	No	Yes	\$ 0.15
Carbon dioxide	600	40% of vol	Yes	No	\$100.00
Halon 1301	100	7% of vol	Slightly	No	\$267.00

The power source to operate the detectors and to activate the extinguishers and the alarms is important. To insure reliability of this power, especially in remote areas, we suggest batteries kept charged by means of the main power system. In turn, this restricts the electrical components to the 24-V dc type. The alarm and control system involves visual and audible signals as indicated in Table I. A definite requirement is the provision for both local and remote signaling of the operating conditions. Provision of certain time delays and overrides is optional depending upon the particular hazards and other requirements of the protection.

A number of factors must be considered in the choice of an extinguishing agent. We have listed five in Table II as applied to an off-site 40-ft electronics trailer. To these considerations may be added the questions of hazards or damage involved in the use of the particular extinguishing agent. In the case of water sprinklers, there may be the additional problem of protecting the system from freezing during cold weather.

In our case, Halon 1301 was chosen for its light weight, low toxicity, and freedom from cleanup problems.

System Design

Our work has involved development of Halon extinguishing systems for off-site trailers and field assembly buildings.

Our first trailer design is shown in Fig. 1. It consists of two cylinders of Halon 1301 (one operational and one standby), two ionization smoke detectors mounted on the ceiling, a manual pull box, an alarm panel, a battery

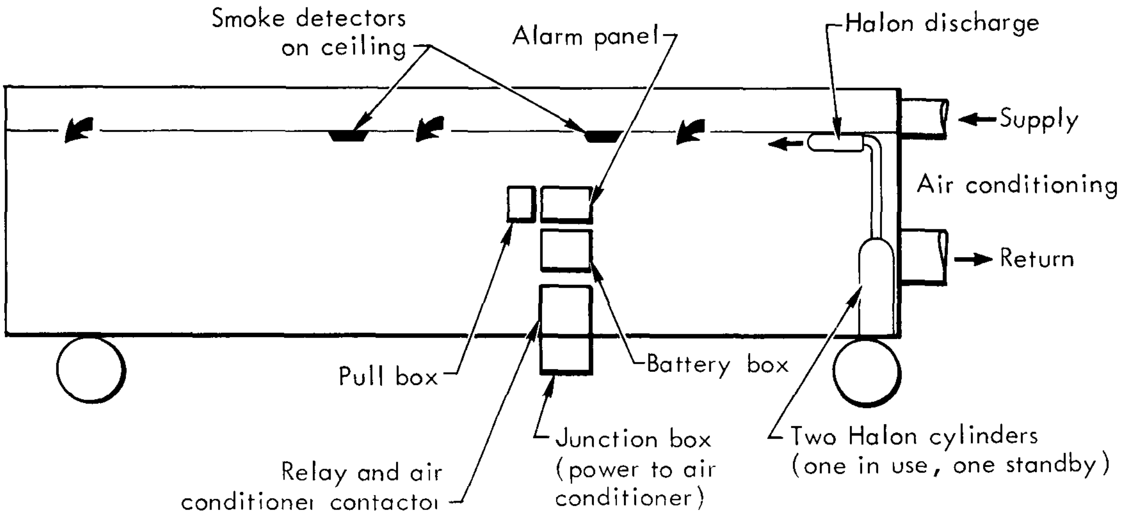


Fig. 1. First LRL Halon 1301 fire-suppression system, for uncompartmented trailer.

and charger box, a relay and air conditioning contactor, and a power junction box connecting the main power source to the externally located air conditioner. The system operates as follows:

1. It is activated either by a signal from one of the smoke detectors or by manually pulling the pull box.

2. When this is done, an alarm bell rings in the trailer, a 30-sec time delay is activated, and a signal is sent through a dial alarm telephone system to a manned control point. At the end of 30 sec, the Halon from one bottle is discharged and a second time delay is activated.

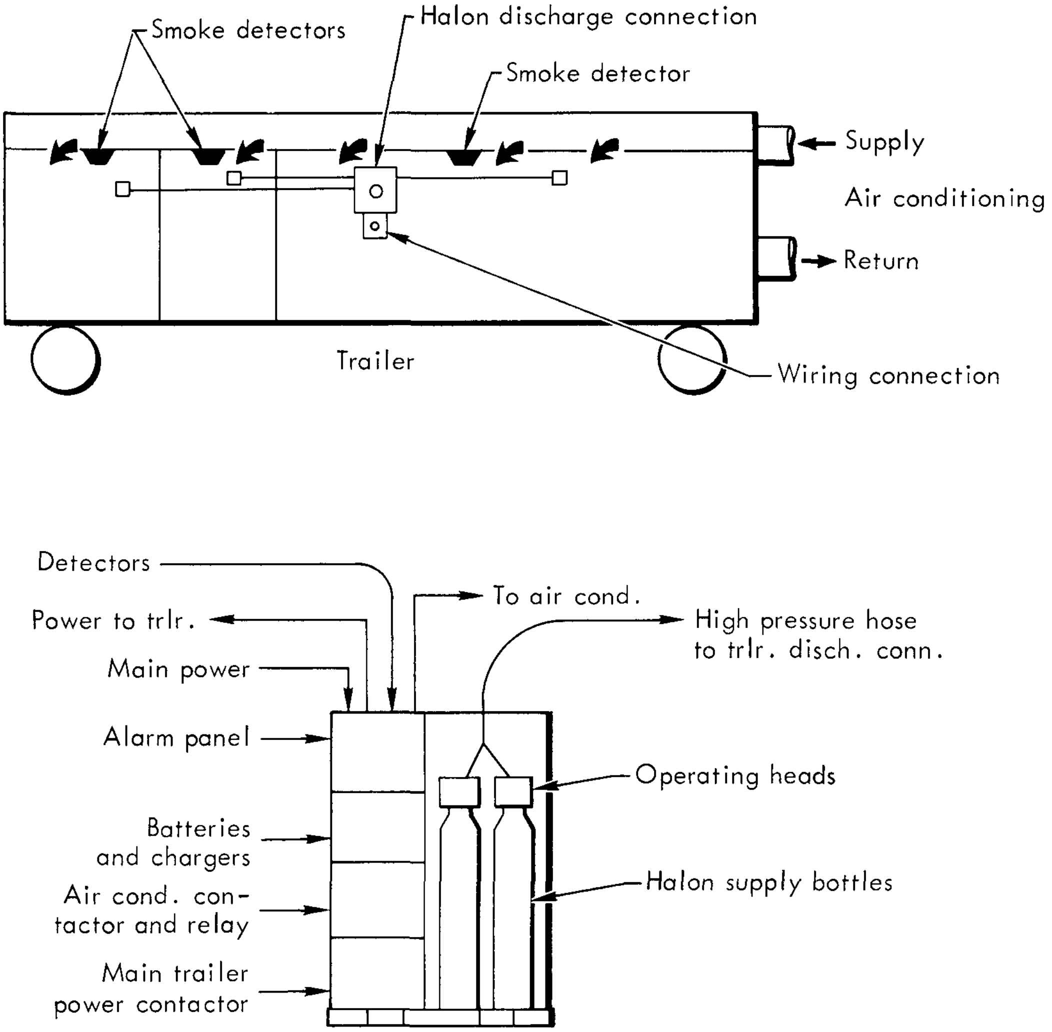
3. 135 sec later, the second time delay closes, operating the relay and the air conditioning contactor to shut off the air conditioning.

4. If the fire is not extinguished, a manual cable located outside the trailer can be pulled to discharge the second bottle of Halon.

5. If the fire is extinguished, the system can be reset and a selective switch moved to put the second bottle into the automatic operation.

We have found through a number of experiments that it is necessary to provide some internal air circulation after the Halon is discharged in order to maintain a reasonably high concentration of Halon near the ceiling. Otherwise the cold, heavier-than-air Halon will tend to concentrate near the floor level. If the fire has started near the ceiling or involved it before the alarm activates the system and the agent is discharged, there is a good possibility that extinction will be incomplete or that reignition will occur.

Figure 2 shows our second generation system as applied to a three-compartment, off-site trailer. It consists of an external fire-protection package containing all the elements previously discussed and provided with a coil of high-pressure tubing and a coil of multiconductor wire which can be plugged into the trailer. The installation within the trailer, therefore, consists merely of the Halon discharge plumbing and the necessary fire-detection system; a local alarm and a pull station can be added as well



Fire Protection Package,
Skid-Mounted (Trailer Optional)

Fig. 2. Second LRL Halon 1301 fire-suppression system, for three-compartment trailer.

if desired. The system provides the option of serving as a field fire-protection package in remote areas or as merely an alarm system connected to fire headquarters when the trailer is located at base and manned fire protection is immediately available.

Figure 3 shows the test setup recently employed in evaluating a Halon system in a cubical field-assembly building 17 ft in each dimension and approximately 5000 cu ft in volume. To study the detector requirements, six clusters of detector/sensors were mounted on the lower faces of the ceiling joists. Each cluster consisted of a heat detector, an ionization smoke detector, and a thermocouple. To evaluate Halon concentration, three sampling tubes were located 1 ft below the ceiling, and three more directly under them 1 ft above floor level. These sampling tubes led to a gas collection and analytical system.

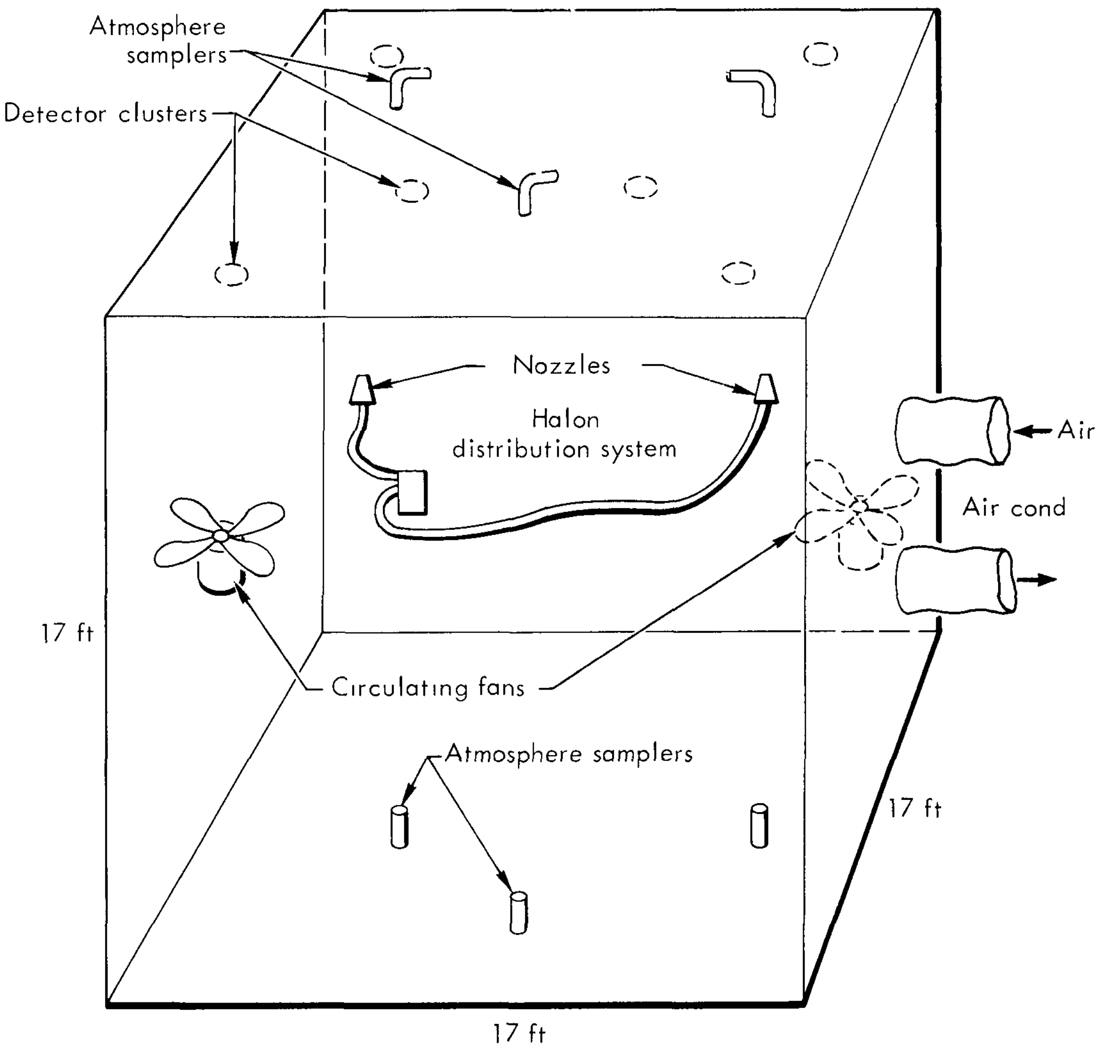


Fig. 3. Test setup for evaluating Halon 1301 fire-suppression system in a cubical building of 5000 cu ft volume.

Test Methods and Results

Distribution of the Halon was examined by using flexible high-pressure hoses which could be located in different positions. These were fitted with Bete nozzles. Air circulation after the Halon discharge was accomplished by using either the air conditioning system in a closed circuit operation or two axial-flow fans located (after some experimentation) as shown in Fig. 3.

Detector types and locations were evaluated by timing the responses of the detectors to smoky or actively flaming fires set in a 30-gal container located approximately in the center of the floor.

We found that the ionization detectors promptly signaled the presence of either kind of fire; their location did not seem to be particularly important. In one or two cases of actively flaming fires, the heat detectors operated first. Three different arrangements of the Halon discharge tubing were employed, both with and without postdischarge circulation. The results are shown in Table III. The most satisfactory arrangement was the one in

Table III. Results of Halon distribution tests in 5000-cu-ft cubical room.

Distribution arrangement	Circulation time after distribution of Halon (min)	Ratio of Halon concentration, bottom/top	
		After 1-2 min	After 5-6 min
Helical	None	6.8	40
Toward upper corners	1.5	2.5	~150
Straight up	1.5	1.2	~80
Straight up	3	1.7	2.5

Table IV. Results of fire extinguishing tests in 5000-cu-ft cubical room.

Fire type	Postdischarge circulation	Detectors reporting fire, and time required	Halon concentration			
			After 2 min		After 5 min	
			Top (%)	B/T[a]	Top (%)	B/T[a]
Smoky	Fans, 3 min	Smoke, 3.5 min Smoke, 4.1 min	5.7	1.3	1.2	4.4
Flaming	Fans, 3 min	Smoke, 0.1 min Smoke, 0.2 min	4.1	1.6	1.7	3.5
Flaming	Air conditioner, 3 min	Smoke, 0.1 min Smoke, 0.3 min	3.6	1.2	1.8	1.6
Flaming	None	Smoke, 0.2 min Smoke, 0.2 min	3.0	1.3	1.8	1.3
Smoky	Fans, 3 min	Smoke, 0.5 min Smoke, 1.1 min	3.7	1.9	4.1	1.5
Flaming	None	Heat, 0.1 min Heat, 0.1 min	3.2	2.2	1.4	4.4

[a] B/T is bottom-to-top ratio of Halon concentration in room.

which the Halon was discharged vertically from a point 3 ft out from each side of the room at approximately midheight and halfway between the front and back. When this was coupled with postdischarge circulation for 3 min, a satisfactory Halon concentration ratio (bottom to top) was obtained.

These findings were confirmed in fire extinguishing tests; the results are summarized in Table IV. In these tests, Halon discharge time was approximately 7 sec; extinguishment was rapid in all cases. As can be seen from the table, a smoky fire with little heat is detected less promptly than an actively flaming one.

The effectiveness of the postdischarge circulation can be judged by the improved concentrations of the Halon near the ceiling and the bottom-to-top concentration ratios.

Conclusions

From our studies briefly described herein, we have reached certain conclusions which are the basis for the system recommendations given in Table V.

Table V. Recommendations based on studies of Halon 1301 fire-suppression systems.

1. Detectors

   Use in pairs, one ionization smoke detector plus one heat detector. Mount slightly below ceiling level, using one pair for each 300 sq ft of ceiling area.

2. Halon cylinder valves

   Electropneumatic type preferred.

3. Halon distribution system
   - a. Nozzles: Open, unrestricted type. Use two per 1000 sq ft of floor area for volumes up to 10,000 cu ft.
   - b. Discharge direction: For square areas, upward from midheight. For rectangular areas, horizontal in long direction.
   - c. Discharge levels: One for ceilings 4 to 20 ft high, two for ceilings 20 to 30 ft high.
   - d. Postdischarge forced circulation: Required for 2 to 4 min.

4. Alarms, controls, operation
   - a. Local and remote normal, trouble, and alarm signals required.
   - b. Normal condition: Visual signal only.
   - c. Trouble and alarm: Visual and audible signals.
   - d. Operation: Alarm on first detector signal, Halon discharge on second signal.
     Disable possible after first smoke or heat detector signal.
     Disable impossible after two heat or two smoke detector signals.
     If disabled, trouble signal should operate visually and audibly both locally and remotely.

We believe that ionization smoke detectors should be used as the primary source of warning or system activation. They can be backed up with temperature-compensated rate-of-rise heat detectors if flash fires are a problem. The detectors should be mounted slightly below the ceiling and free of obstructions, and there should be one detector for about every 300 sq ft of ceiling area.

We believe that the solenoid-activated, pneumatically operated ball valve, first applied to this use in our studies, is an improvement over existing explosive-operated, rupture disk systems for most fire situations. The ball valve, which can also be operated manually, serves double duty: as a shutoff valve during extinguishant transfer and transportation, and as a system operating valve. It can be obtained commercially with the pneumatic operator and solenoid activator at a cost considerably less than that of the rupture disk system, and it is easier to fill; moreover, it is considerably less expensive to use since it requires no replacement parts to put it back into operation after it has been activated.

In regard to distribution of the extinguishant, piping should be smooth, short, and free of sharp bends; the nozzles should be of the open, unrestricted type. In our view, two such discharge points should be employed for every 1000 sq ft of floor or ceiling area for volumes of up to about 10,000 cu ft. For approximately square areas, the discharge should be upward from about midheight; for rectangular areas, the discharge should be a short distance below the ceiling and aimed horizontally in the long direction.

For ceiling heights of 4 to 20 ft, one discharge level should be sufficient. For ceiling heights between 20 and 30 ft, it is probably better to use two discharge levels. In any case, we feel that postdischarge internal circulation is necessary for a period of, say, 2 to 4 min.

With respect to alarms and controls, there should be both local and remote indications of the operating condition. A visual signal is sufficient for normal condition, but for trouble and alarm situations, both visual and audible signals are musts. The operation of the system should be carefully thought out. We show in Table V one set of possibilities which we are employing in certain of our installations.

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SECTION IV.

ENVIRONMENTAL AND SAFETY CONSIDERATIONS

SAFETY CONSIDERATIONS AT THE BABCOCK & WILCOX PLUTONIUM DEVELOPMENT LABORATORY

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April 14, 1971

Safety Considerations at the Babcock & Wilcox Plutonium Development Laboratory

R. V. Carlson, W. K. Goodman

ABSTRACT

Since 1966, Babcock & Wilcox has conducted research and development programs in its Plutonium Development Laboratory (PDL). The PDL was specifically designed to handle plutonium-bearing oxides and solutions in a wide variety of processes, on both bench and pilot-plant scales.

This paper describes the safety organization, the administrative procedures and controls, and several systems that allow maximum flexibility in operations without compromising safety. Two glovebox accessories designed by B&W are also described. The first, a specially designed manometer, protects the gloveboxes from inadvertent overpressurization resulting from a malfunction of the compressed inlet air supply or the outlet filter system. The other, a sphincter-type valve, permits rapid filter changes with minimum risk of spreading contamination.

1. INTRODUCTION

Since early 1966, Babcock & Wilcox has conducted research and development programs in its Plutonium Development Laboratory (PDL), which was built in four stages and completed in 1968. This 20,000-square-foot laboratory has a variety of capabilities, including (1) preparation of PuO_2 by oxalate and hydroxide precipitation and by sol-gel techniques, (2) preparation of UO_2-PuO_2 by coprecipitation, sol-gel, and wet and dry powder blending techniques, (3) a pilot line for fabricating pellets and shards, (4) loading of fuel pins by conventional as well as vipac and sphere-pac techniques, (5) plutonium scrap recovery, and (6) complete analytical services. To provide an adequate, minimum-cost facility for plutonium technology, the equipment, the building, and the administrative procedures were designed to provide maximum flexibility in operations without compromising safety.

Several construction features of the building and a summary of the administrative controls are presented in the following sections. Two glovebox accessories designed by B&W are also described. The first, a specially designed manometer, protects gloveboxes connected to a compressed dry air supply from inadvertent overpressurization, and the second, a sphincter-type valve, permits rapid filter changes with minimum risk of spreading contamination.

2. PDL CONSTRUCTION FEATURES

The final portion of the PDL was completed in 1968. The building lies in a rural area of central Virginia, about six miles east of Lynchburg. An interior view is shown in Figure 1. To minimize the area that could be affected by a fire or a release of contamination, all the rooms are as small as practicable. In addition, the air pressure varies from room to room; rooms housing operations with high potential for releasing contamination have the lowest pressure.

The interior surfaces of the PDL are covered with materials that resist penetration of contamination and facilitate decontamination. Most of the concrete floors are covered with a 1/8-inch layer of Dex-O-Tex Dex-O-Cote, a urethane-based coating. The interior walls are concrete block covered with four coats of Vitra-Tile. Both of these acid-resistant materials bond tightly to concrete and provide a seamless surface that prevents contamination from penetrating the concrete. Since both materials take the shape of the surface being covered, the surface must be smooth prior to their application to facilitate decontamination. The false ceilings in the operating areas are constructed of steel ceiling tile, caulked at the seams, and covered with Vitra-Tile. The resultant sealed barrier prevents contamination from penetrating the service area above the false ceiling.

3. ADMINISTRATIVE CONTROLS

The Lynchburg Research Center (LRC) management has established guidelines and general operating procedures for the PDL based on a conservative approach to safety. However, to cope with the diverse and changing nature of the operations, a safety organization has been delegated the authority to make decisions related to the specific operations being conducted at any given time. This organization is summarized in Figure 2. It operates parallel to and is frequently audited by the technical and administrative management of the LRC.

The Emergency Officer's main duties are to coordinate the responses to an emergency and to assume primary responsibility for making decisions required at that time. In this capacity, he has absolute authority over all personnel. The Isotopes Committee, composed primarily of consultants from other B&W divisions and from outside organizations, advises the Director on matters of safety.

Each LRC facility has a Facility Supervisor who has prime responsibility for operations in that facility. He is responsible for (1) compliance with license conditions, (2) the custody of special nuclear material, (3) establishing the fissile mass limits for a given operation, (4) approving changes in specific operating procedures, and (5) notifying the Director of all matters that constitute a major departure from previously established procedures. The site's Nuclear Safety Officer, Health Physicist, and Nuclear Materials Management Group advise the Facility Supervisors.

The PDL has three Area Supervisors who must ensure that operating personnel are aware of and adhere to current operating procedures. These supervisors report the day-to-day operations to the Facility Supervisor.

Since specific operating procedures are changed frequently, it is necessary to rely on several techniques in addition to personnel training to ensure the safety of the operations. First, free access to the PDL is restricted only to PDL operating personnel; all other LRC personnel must be escorted. Secondly, signs are used extensively to inform operating personnel of current procedures. Thirdly, several independent checks of all operations are made on a periodic basis. Examples of the multi-check system are shown in Figure 3.

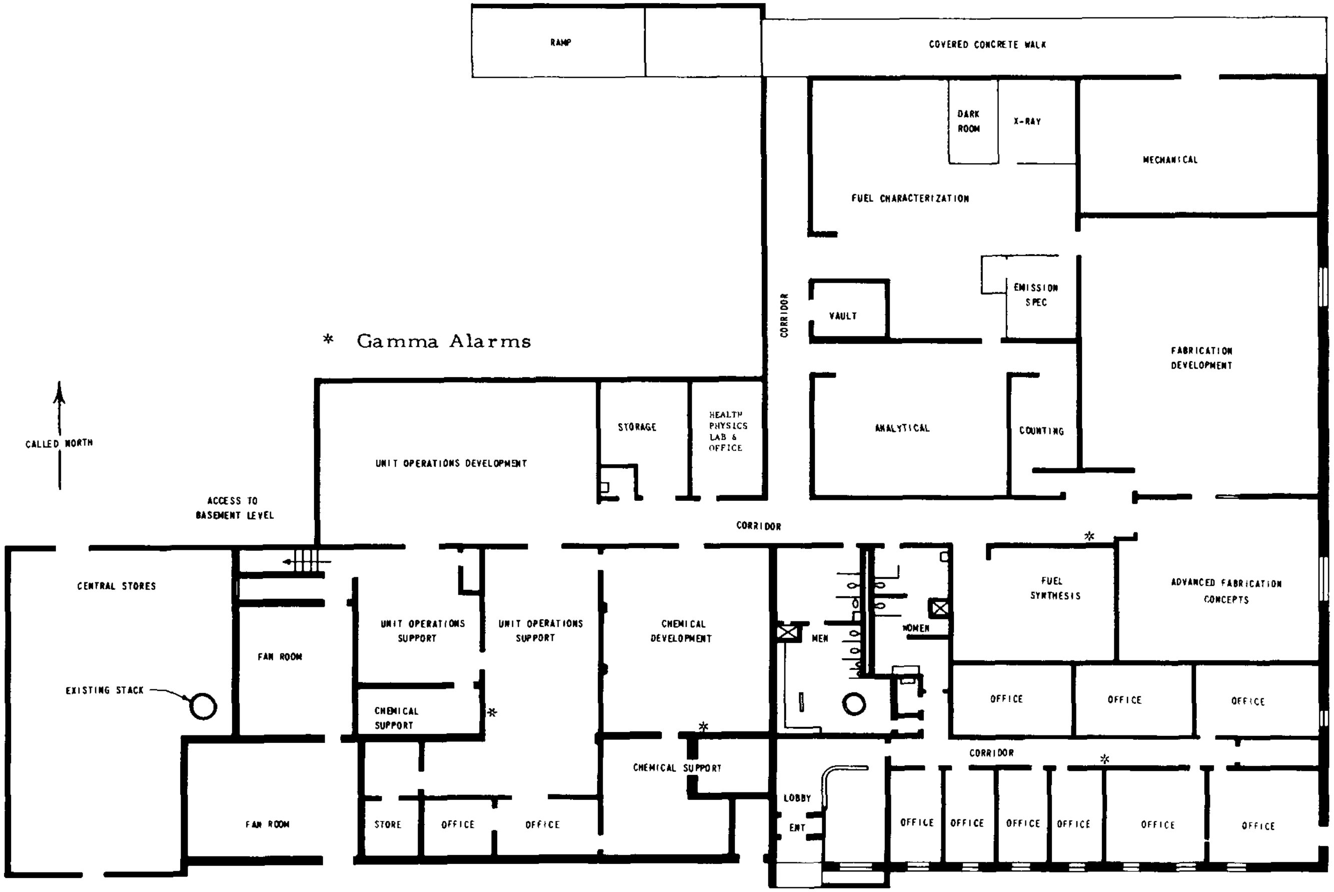

Figure 1. Floor Plan of Plutonium Development Laboratory

Figure 2. Outline of PDL Safety Organization

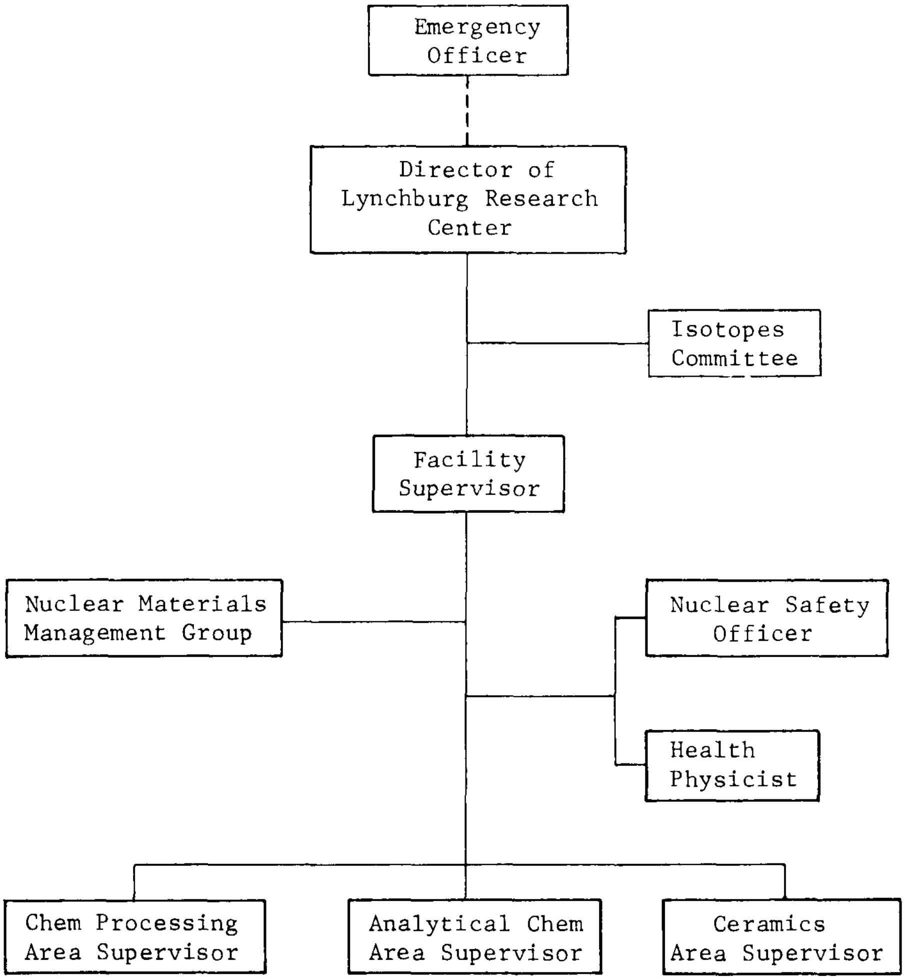

Figure 3. Administrative Control System

	Primary Responsibility	Check	Independent Check	Periodic Over-Checks
Establish Procedures	Engineer	Fac. Supv. Line Mgmt.	Isotopes Comm. Director	
Daily Operations	Technician Engineer	Area Supv. H.P. Group	Fac. Supv. Line Mgmt.	Nuc. Safety Officer
SNM Transfers	Operator	Area Supv. Acctg. Clerk	Fac. Supv. Acctg. Officer	Safeguards Officer Nuc. Safety Officer

4. GLOVEBOX ACCESSORIES

B&W has designed and built special gloveboxes for handling plutonium. A number of these are connected to a supply of compressed dry air to minimize humidity, especially in areas in which oxide powders are handled. If the outlet filter system fails, then the glovebox becomes pressurized; if the inlet filter system fails, then the glovebox is subject to excess negative pressure. Since either of these events could lead to glovebox failure and a release of plutonium, a special manometer was designed to protect the gloveboxes automatically from pressure extremes. Figure 4 is a photograph of the system. This Plexiglas and aluminum system can be filled with any of several liquids. Operation of the system is simple -- at a glovebox-to-room pressure differential which depends on the liquid being used, air will bubble through the liquid to reduce the differential. The manometer is connected to the room side of the inlet absolute filter, so contamination cannot be released if glove box atmosphere is vented to the room because of overpressurization. Since there are no moving mechanical parts, the system should never malfunction.

Another accessory, a sphincter-type valve for changing the filters, is installed on all the B&W-designed gloveboxes. This system, which is constructed of polyvinyl chloride (PVC) and neoprene, is shown in Figure 5. It will accept commercially available, 6-inch-diameter absolute filters. By alternating the PVC and neoprene components, a sphincter valve is formed. In operation, the system contains two absolute filters separated by a PVC sleeve. Air from the glovebox passes through the first filter into the open area of the sleeve and then into the offgas line. The second filter is capped to prevent room air from flowing directly into the offgas line and reducing the negative pressure in the glovebox. When the first filter requires changing, a new sleeve and filter are attached to the second filter. The train of filters is then pushed in so that the defective filter is discharged into the glovebox. The entire filter-changing operation consumes less than 5 minutes, and since no bagging is required, the possibility of releasing contamination during a filter change is essentially eliminated.

5. CONCLUSION

B&W's Plutonium Development Laboratory has operated safely for over 5 years. By dividing the facility into relatively small rooms, minor releases of contamination have been prevented from spreading throughout the facility. As a result of the flexibility allowed by the safety organization in day-to-day operations, specific operating procedures have been tailored to a changing scope of work. A manometer that prevents pressure extremes in gloveboxes and a sphincter-type valve that is used for changing glovebox filters have operated successfully. Both of these systems have increased the safety of operations in the PDL.

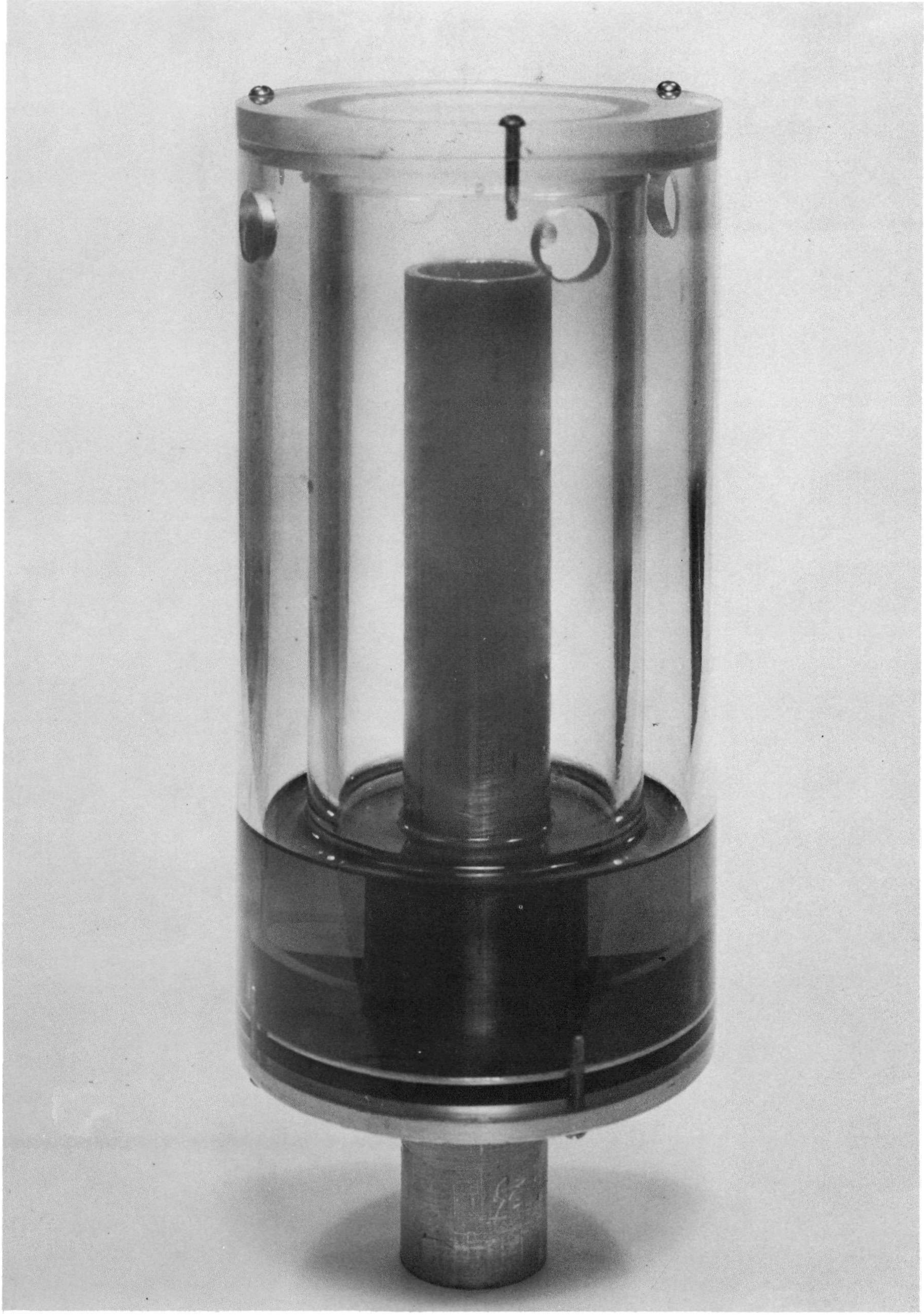

Figure 4. Photograph of Manometer

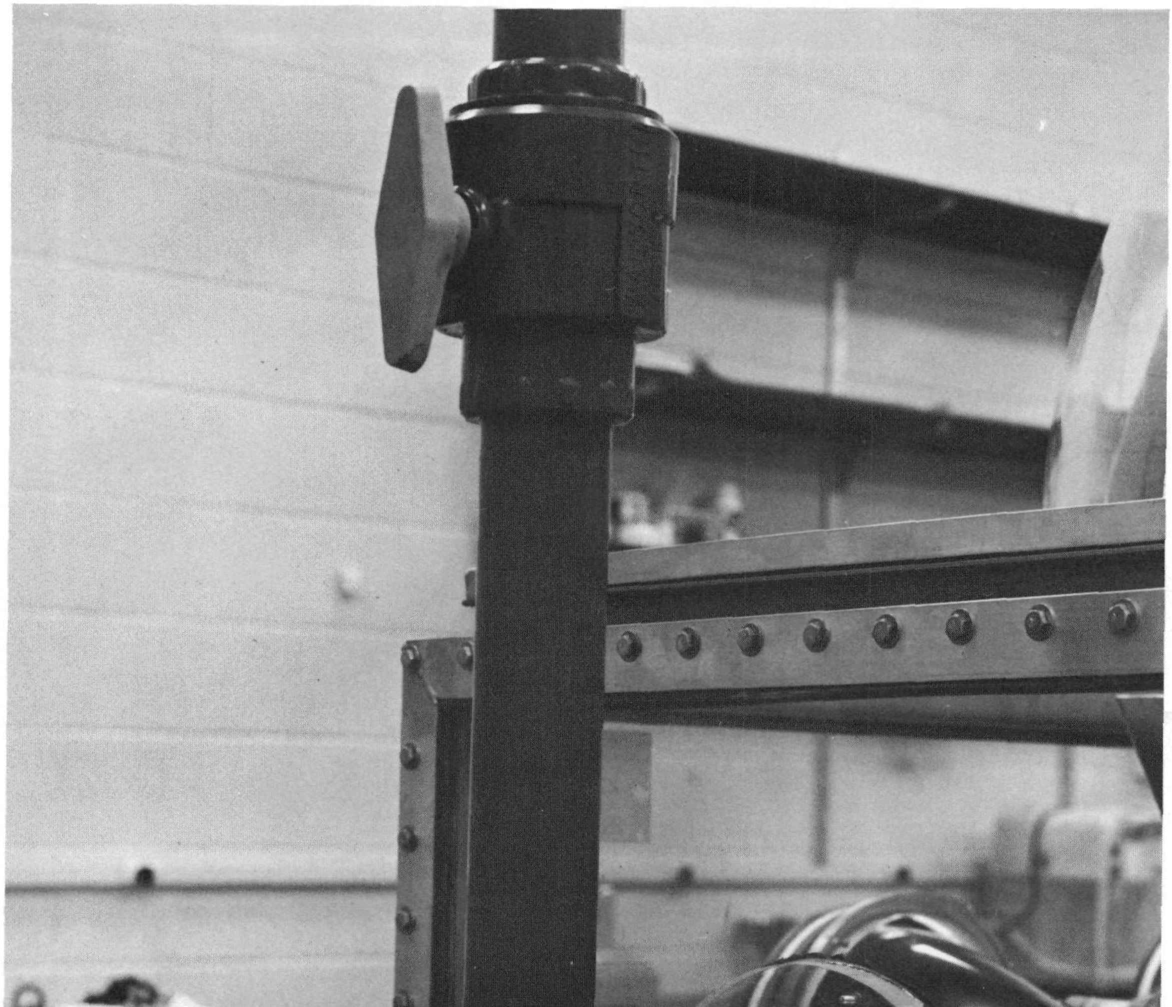

Figure 5. Photograph of Filter Changing System Installed

IN-RESIDENCE HEALTH AND SAFETY SUPPORT IN A PLUTONIUM FACILITY

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April 14, 1971

IN-RESIDENCE HEALTH AND SAFETY SUPPORT IN A PLUTONIUM FACILITY*

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Abstract

Health and Safety Technicians (monitors) provide in-residence safety services to the metallurgical research and engineering effort of the plutonium facility at the Lawrence Radiation Laboratory, Livermore. This paper describes the qualifications of these technicians and typical services rendered by them to the facility scientific personnel, such as, contamination control, waste recovery, monitoring of air sampling equipment, frequent smear and swipe samples, radiation surveys, equipment calibration, out-processing of liquid and solid wastes to control fire and criticality hazard, etc.

The Plutonium Facility

The Lawrence Radiation Laboratory plutonium facility is administered by the Chemistry Department through its Plutonium Metallurgy Section, and is shared by the Plutonium Engineering Group of the Mechanical Engineering Department. The chemists and engineers assigned to the facility are engaged in test operations of Pu-bearing engineering assemblies, the development of unique or improved fabrication techniques, as well as fundamental and applied research in plutonium metallurgy.

The building is divided into three zones: 1) the radioactive materials work area (RMA) or laboratory area, 2) the administrative area (offices), and 3) a large mechanical equipment room. Figure 1 shows the building layout and the location of hazards. An additional increment to this building is planned for FY 1973, to be used by the Plutonium Engineering Group.

Hazards Control Department Support

Although responsibility for operational safety rests with each employee and his supervisor, the Hazards Control Department is responsible for aiding supervision by monitoring hazards, making hazards analyses, assisting in developing standardized safety procedures and information, and organizing emergency response capability. The Department provides safety support, organized as shown in Fig. 2, to the "client" divisions of the Lab through area representatives assigned to the various subject areas of the Lab's work—chemistry, physics, craft shops, biomedicine and so on. A team of in-residence Health and Safety Technicians (monitors) functions under the direction of each area representative. An advantage to this form of organization is that

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*Work performed under the auspices of the U.S. Atomic Energy Commission.

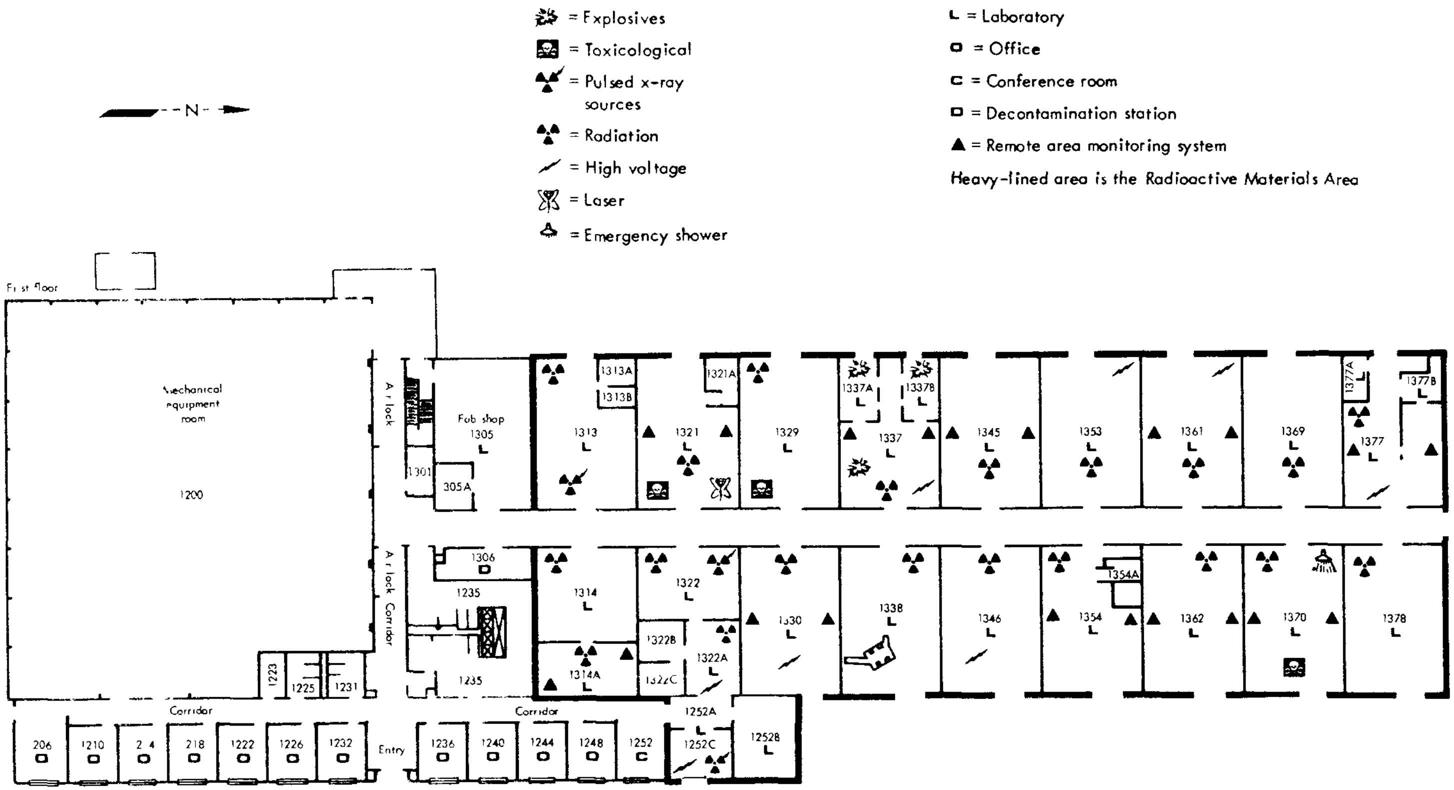

Fig. 1. Building 332 floor plan showing work areas and the location of hazards.

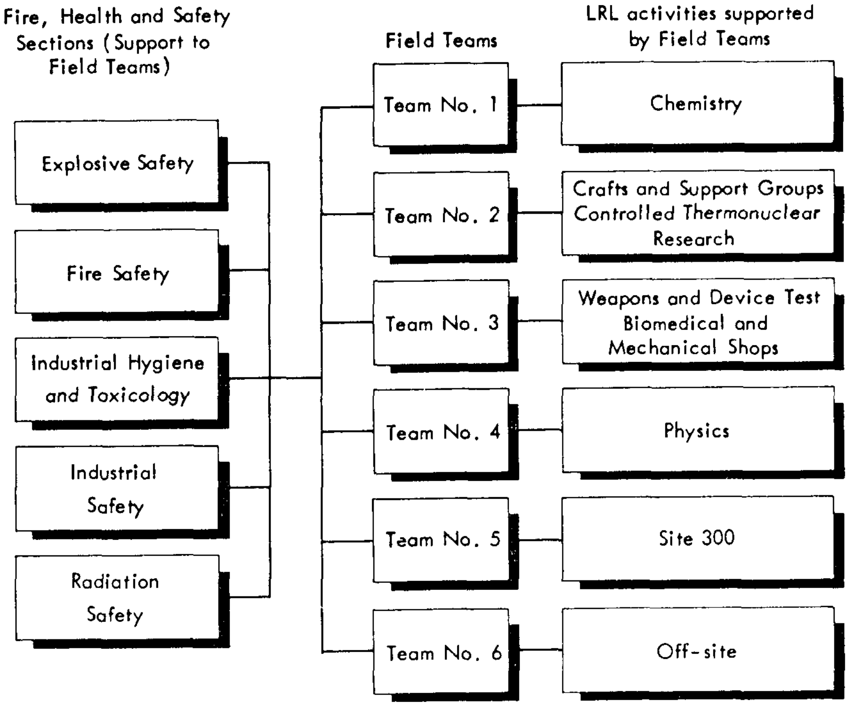

Fig. 2. Organization of Hazards Control Department safety support to "client" activities of the Lab. The sections on the left establish the safety program for their specific discipline and provide any required advisory assistance, such as hazards analyses, to the field teams in the middle. The technicians in the teams are directed by an area representative and support all of the safety programs. The six designated subject areas served by the respective teams are on the right.

an experimenter need only go to one person—the area representative—for his safety support. The area representative then coordinates all required services, such as, industrial safety and hygiene, fire and explosives safety, etc. Although the area team provides all these services, our interest here is primarily with the radiation safety aspects of our plutonium facility.

Health and Safety Technician Qualification

Qualifications for Hazards Control technicians are high. We normally require an associate degree in physical science or technology, or equivalent experience. Many of the technicians have received AA-level education in nearby junior colleges before or during employment. Chabot College in Hayward has a unique and popular program in radiation technology. Professional health physicists of the LRL Hazards Control Department have taught several of the courses offered in that program.

Technician candidates who come from a military (particularly U.S Navy) nuclear speciality are usually well prepared. In addition to their excellent training, they normally come to the Laboratory with three or four years of nuclear power experience.

In addition to the formal education and experience requirements for employment, the Hazards Control Department provides a basic 12-hour course in radiation safety and numerous special topic seminars for new technicians. (See appendix for an outline of the formal facility indoctrination.) Before promotion to supervisory level an advanced seminar[1] covering a wide range of radiation safety topics and taking about one year is required. The academic level of difficulty is about that of sophomore to junior level college work.

So hazards control technicians assigned to our plutonium facility are well experienced and academically prepared. But their training doesn't end here. Through formal training conducted by the Hazards Control Department and the scientific personnel of the facility as well as on-the-job training he eventually achieves unrestricted access to all aspects of the facility operation. At this point he is able to perform monitoring routines with only nominal supervision.

Technician Services

Four Health and Safety Technicians are assigned by the LRL Hazards Control Department to provide in-residence safety support to the plutonium facility. Although assigning in-residence safety technicians to a high hazard facility is not unique, the technicians' qualifications, training and the services rendered by them in the plutonium facility make them especially effective.

The services performed by the technicians are divided between two broad areas. First, they are the eyes and ears of the Hazards Control Department in the building. By this I mean many of the problems investigated, and the resulting improvements in equipment, techniques, etc., made by the professional staff are brought to their attention by the building monitors. For example, the technicians have been instrumental in the development of an ion-exchange waste treatment system to handle radioactive liquids. Another is the technician participation in field testing of a waste barrel gamma scintillation counter described below. These examples illustrate application of experience and training gained through the programs they support.

The second area of service is the day-to-day service to the occupants of the building and to the Hazards Control safety professionals. This service primarily includes contamination control and waste handling and control. Various fire safety, industrial safety and industrial hygiene tasks are also required, but these will not be considered in this paper.

The technician's role in contamination control is varied. They conduct comprehensive instrument calibration and periodic functional check programs. An example of these functional checks is the daily test with a radioactive source of our hand and foot counter shown in Fig. 3. Personnel working in the facility check their hands and feet for radioactive contamination on these instruments; if any contamination exists both an audible alarm and a visual readout are produced. Placards tell the individual to contact the technicians if contamination is detected. The technicians are trained in the use of the decontamination facilities which are maintained in the facility and are in a position to start immediate decontamination.

The resident technicians carefully control all survey instruments. In this connection daily functional checks, identification for calibration, and minor repairs are made.

Another important service is monitoring and servicing the various air sampling systems in the building including the filter paper room air sampling system, the continuous air monitor (CAM) and high volume portable air sampler which is used if radioactive releases or spills are suspected. Malfunctioning or suspicious results are immediately reported to the Health Physicist assigned to the team and facility.

Prefilters for the room exhaust are routinely changed by the technicians. When loading of room or glove box HEPA filters becomes significant, the monitors, under the guidance of the area team's industrial hygienist, replace the filter and assist in a DOP smoke check of the new unit.

Contamination surveys of floors, work areas, equipment and building exits are done frequently. Our technicians use two alpha probes side-by-side in the configuration shown in Fig. 4 to increase the area scanned during floor surveys. Smears are normally counted within hours after being taken by the technicians. A gas flow proportional counter is maintained in the building for this purpose.

A very important service offered by the facility health and safety technicians is assistance in "bag-outs" and "bag-ins" (removing from and inserting items into contaminated glove boxes). Besides helping move objects in or out of the box, the technician performs a complete contamination survey of the area about the box. This technique allows inadvertent releases to be promptly detected and cleaned up.

Another contamination control item performed is monitoring the building waste retention system. Before emptying the tanks, for example, a monitor will withdraw a sample for radiological and chemical analysis.

The technicians also play a vital role in the building criticality-safety program. They monitor criticality limits in glove boxes and assist in the use of criticality-safe containers. In addition, the Remote Area Monitoring system (RAM)— a criticality alarm system—is visually checked daily and calibrated weekly.

The other important area of Hazards Control support in the plutonium facility is waste control and handling. As mentioned earlier, the monitor is present during bag-outs of wastes and surveys the area to detect any material that may have gotten loose. Bagged-out waste is carefully labeled on the plastic bag itself for later identification. These plastic bags are placed in yellow trash cans clearly marked "Radioactive Waste". A lid is provided to reduce the fire hazard. When the cans are full, the contents are transferred to a holdcart, an example of which is shown in Fig. 5. This cart was developed to store the contents of several trash cans of waste material until the material could be checked with a gamma scintillation apparatus. The cart is basically a metal box with a lid (normally closed). Trash so contained is protected from ignition sources, tearing, being stumbled on, etc. Either before putting bagged trash into the container, or as it is being barrelled, the waste is given a rough check by the technician with a GM instrument.

After being placed in a barrel, the waste is evaluated with a gamma scintillation detector in conjunction with a 100 channel pulse height analyzer[2] as shown in Fig. 6. The technicians are trained to operate the equipment and

Fig. 3. Hand and foot counter console with audible alarm and visual readout. Note check sources attached by key chains for convenient use.

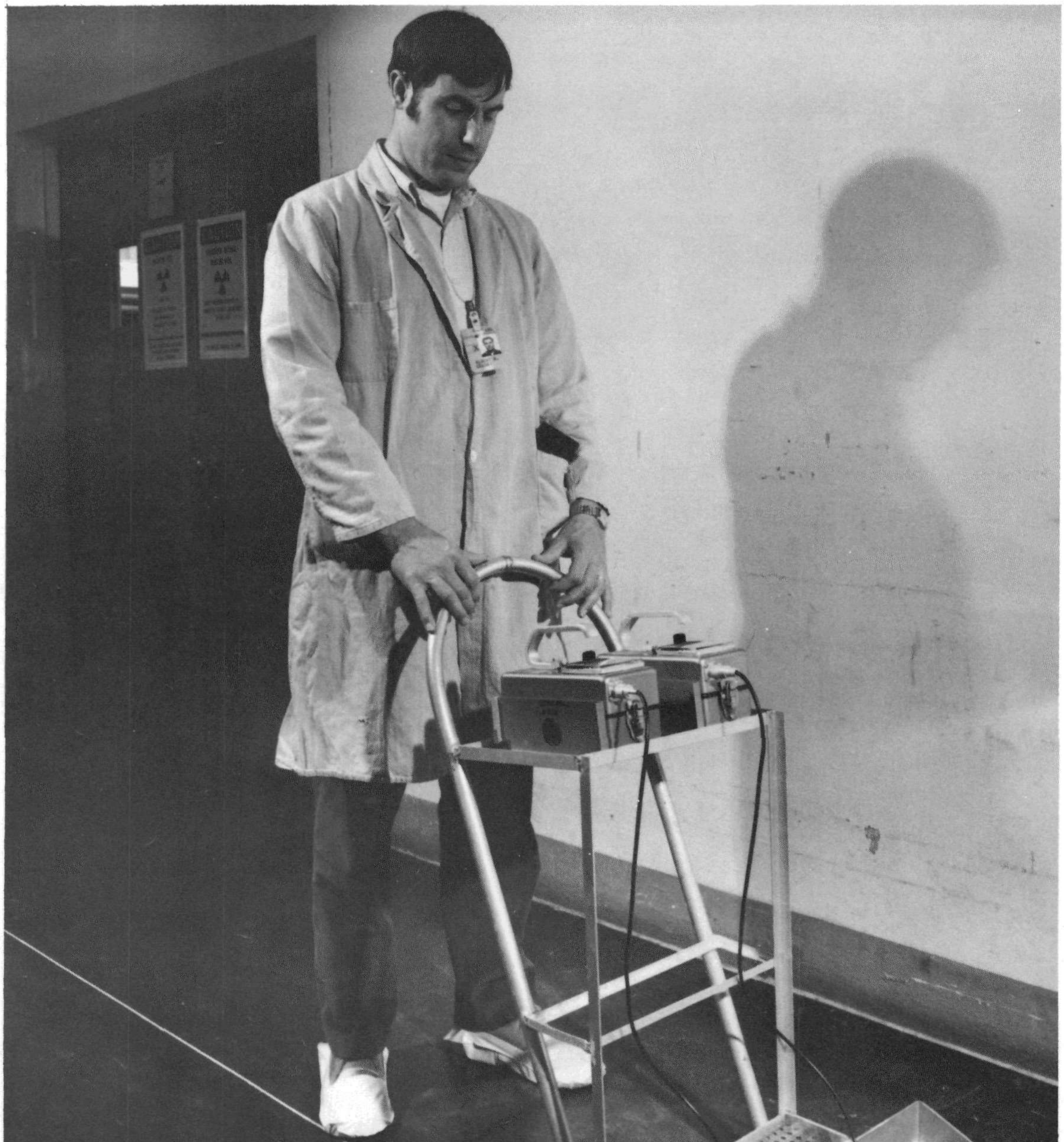

Fig. 4. Cart-mounted alpha survey system.

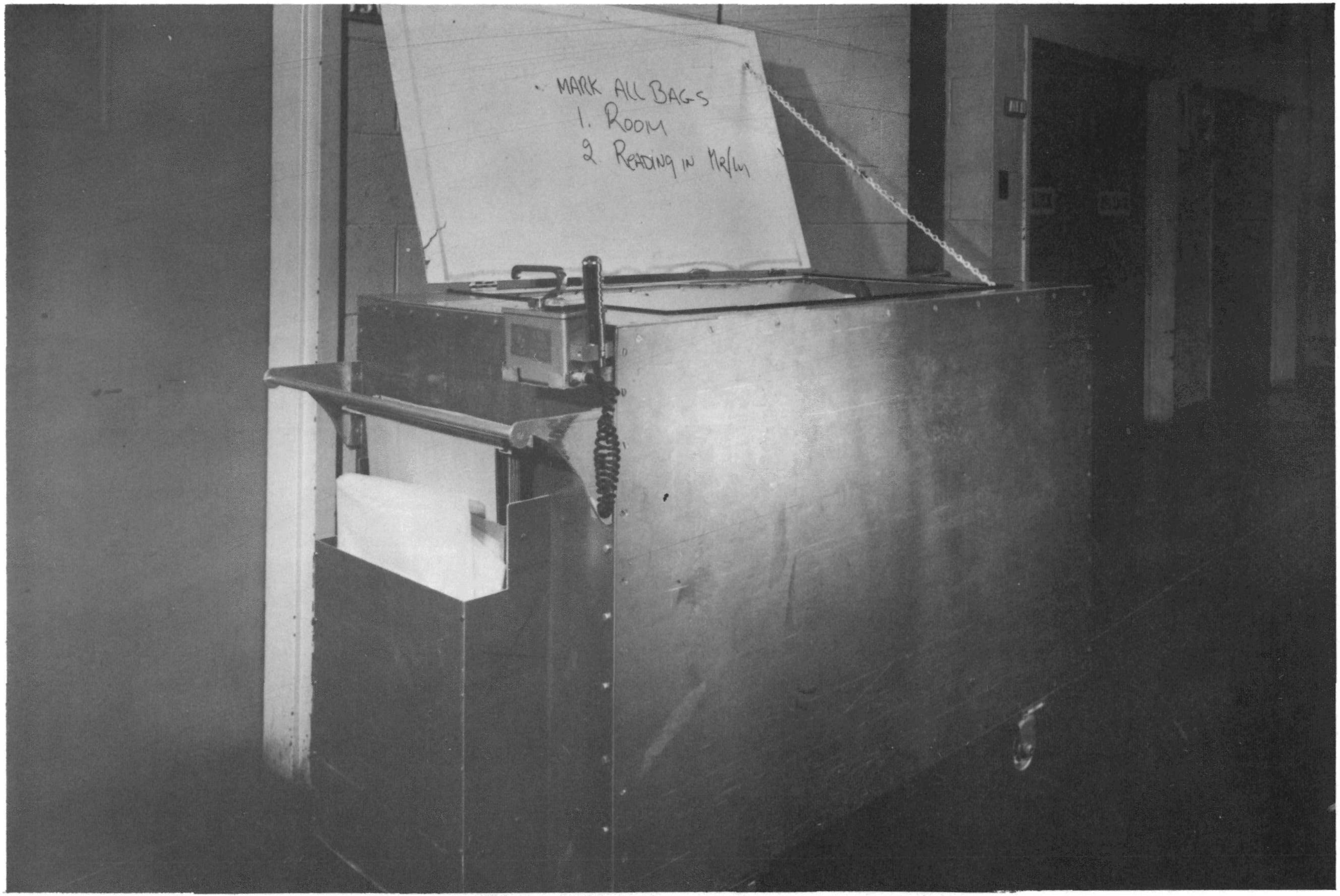

Fig. 5. Mobile radioactive-waste storage container.

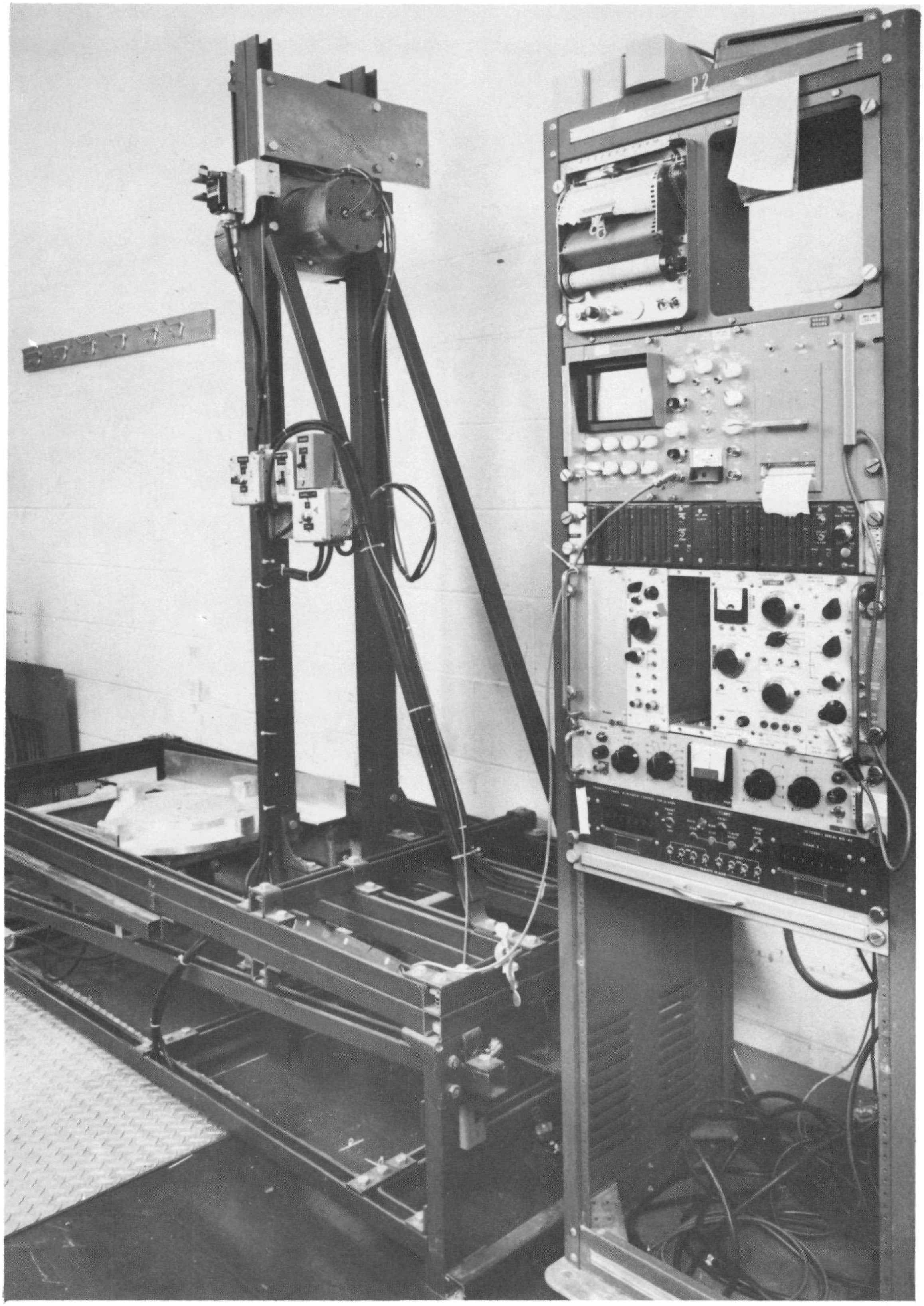

Fig. 6. Mechanized waste barrel counter with gamma scintillation detector and 100-channel pulse height analyzer.

assist in the evaluation of the data. If unusual quantities of Pu are detected the contents of the barrel are investigated. When the source or sources of activity are found and recovered, the identification marked on each bag allows pinpointing of the room and glove box generating the waste. Following the counting, the technician moves the barrels outside to a holding area for pick-up by the Disposal Section of the Hazards Control Department.

In summary, through well qualified and trained Hazards Control technicians assigned to the plutonium facility, comprehensive services can be offered to both the Hazards Control Department and the scientific staff of the plutonium facility. These services include monitoring compliance with safety procedures, providing contamination control assistance, and handling and monitoring wastes.

APPENDIX

OUTLINE OF SAFETY INDOCTRINATION FOR PLUTONIUM FACILITY

I. Introduction
  A. Purpose of Indoctrination
  B. Scope of Presentation
II. Operation of the Building
  A. Rules Governing Exclusion Areas Access
  B. Building Organization
    1. Administration of the Building Operation
    2. Support Groups
  C. Building Areas and Zones
  D. Communications Within the Building
  E. Building Ventilation System
  F. Glove Box Atmosphere Controls
III. Mechanical Engineering Facility/Plutonium Metallurgy Facility
  A. Scope of the Facility
  B. Pu Engineering/Pu Metallurgy Group Responsibilities
    1. Assure work conforms with established procedures
    2. May delegate secondary responsibilities
    3. Initiate all C and M work
  C. Pu Engineering/Pu Metallurgy Authority
    1. Authority to stop a hazardous operation
    2. Disposing of equipment (contaminated or uncontaminated)
    3. S. S. material accountability
  D. Responsibilities of Personnel Using Facilities
    1. Responsible to supervisor in building
    2. Transferring fissionable or contaminated material
    3. Operators may not adjust atmosphere controls
IV. Hazards Control
  A. Responsibilities and Functions
    1. To help building personnel conduct work safely
    2. Field Team concept
      a. Field Rep coordinates discipline support
      b. Field team for Building 332, including Monitor support
      c. Examples of support
    3. Stopping a hazardous operation
    4. Examples of services provided
  B. Special Hazards of Plutonium
    1. Radioactive properties

        2. Biological hazards
            a. Internal and external
            b. Comparison with other hazardous materials
            c. Modes of entry into body
            d. Protective measures
        3. Pyrophoricity
        4. Criticality

C. Monitoring Support and Rules for Safety
    1. Routine service operations
        a. Direct monitoring of material handling
        b. Air sampling
        c. Floor, desks, etc., monitoring
        d. Special protective material carts
    2. Building safety rules
        a. Things to do and not do while in building
        b. Working with hazardous materials
        c. Criticality safety controls
        d. Emergency procedures

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2. J. F. Tinney et al., "Drum Scanner for the Measurement of ^{239}Pu Contaminated Waste," Hazards Control Progress Report No. 35, Lawrence Radiation Laboratory, Livermore, Rept. UCRL-50007-69-3 (1969).

BATTELLE'S RESEARCH PLUTONIUM FACILITY ENVIRONMENTAL CONTROL AND SAFETY

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April 14, 1971

BATTELLE'S RESEARCH PLUTONIUM FACILITY ENVIRONMENTAL CONTROL AND SAFETY

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ABSTRACT

Research and development studies of plutonium-bearing materials are conducted in the Battelle-Columbus Plutonium Facility located at West Jefferson, Ohio. The facility provides nearly 7500 sq ft of laboratory space and is operated with a philosophy for total material containment which provides safe conditions for both laboratory personnel and the surrounding environment. Through the use of numerous safety systems and close supervision of personnel activities, research is conducted in an efficient manner. Education and training is provided by frequent seminars and lectures as well as by on-the-job supervision by qualified personnel. Alarm systems and established procedures are the main safeguards in preventing serious personnel injuries and in attending to emergency situations when they exist within the facility.

INTRODUCTION

The Battelle-Columbus Plutonium Facility is located at West Jefferson, Ohio approximately 17 miles west of Columbus, on a 1000-acre tract of land developed for nuclear science technology. This facility is one of a complex of four permanent nuclear facilities including a 2-Mw swimming pool research reactor, a hot-cell facility with maximum capacity of 10 million curies, and a critical assembly building. The Plutonium Facility is utilized for research and development programs for both industrial and government agencies with operations being conducted under License SNM-7 and as an Exempt Facility under AEC Contract W-7405-eng-92. The laboratory was specifically designed for conducting plutonium fuel studies and has an integrated capability ranging from basic and applied research through development. Program activities have touched on almost all phases of plutonium research including the synthesis of fuel compounds, property determinations, product characterization, fuel-element fabrication and reprocessing. Essentially, the laboratory has the flexibility of satisfying varied sponsor requirements and can be altered to fulfill almost any need.

FACILITY AND CONTROL SYSTEMS

Building Construction

The Battelle-Columbus Plutonium Facility is located in a separate building especially constructed for plutonium handling and houses no other activities. The building layout given in Figure 1 depicts the construction in three separate sections, as necessitated by increasing demand for new capabilities. The initial building, measuring approximately 72 by 24 ft was constructed in 1960 and a 53 by 24-ft addition was added in 1964, providing a total laboratory floor space of about 3050 sq ft. These two sections were constructed of

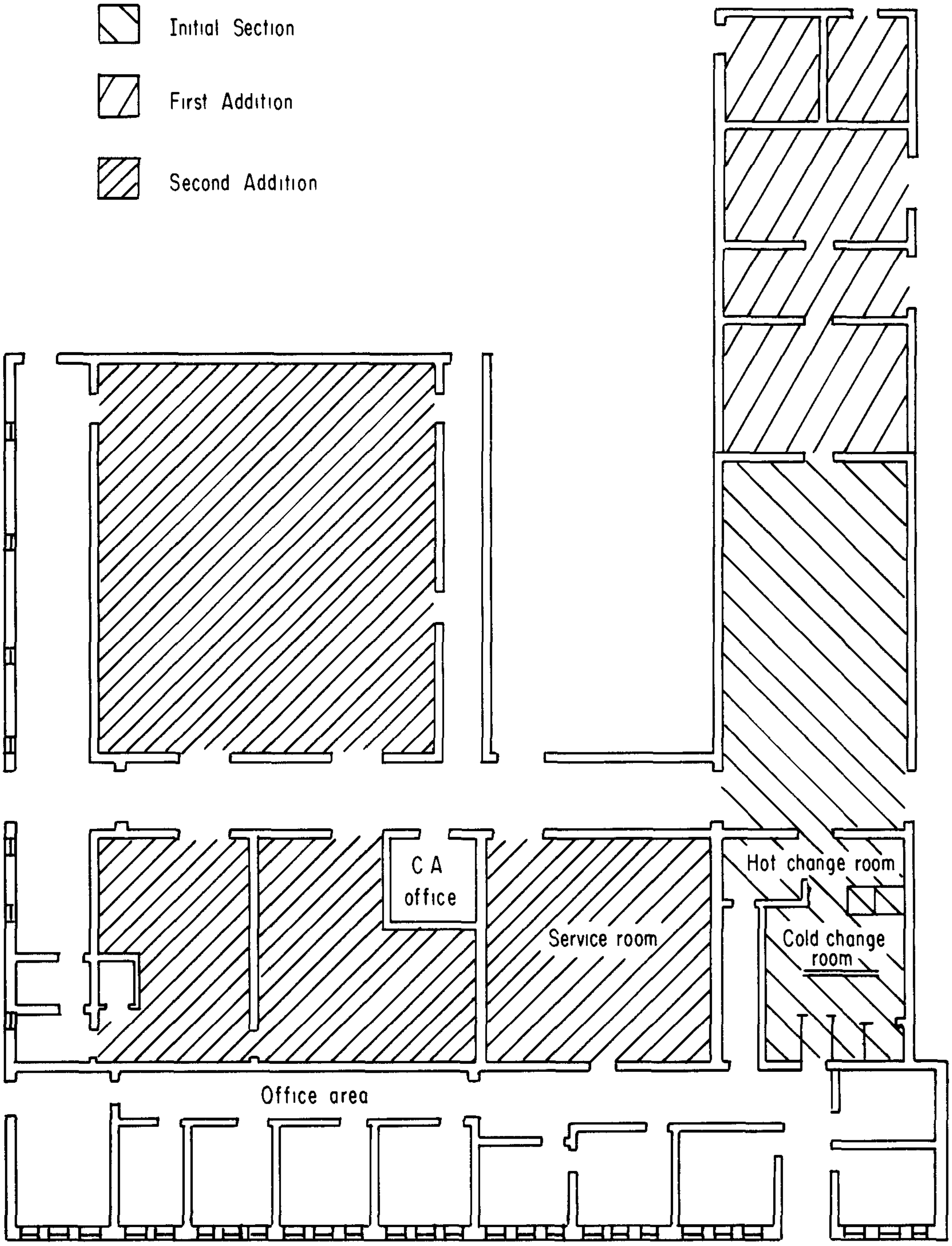

FIGURE 1. BATTELLE-COLUMBUS PLUTONIUM LABORATORY

double metallic siding positioned on a concrete slab, with glass wool being used as an insulation material. A third section of the laboratory, added in 1967, increased the laboratory floor space to about 7160 sq ft. This addition is of cement block construction on a concrete slab. The roof over this lab area is 16 ft high with a 12-ft dropped ceiling, thereby providing a 4-ft ceiling space for utilities and services. To facilitate cleanup of the areas, the walls and floors were sealed and filled prior to application of an epoxy paint surface.

Ventilation and Pressure Regulation

Makeup air for the laboratory is introduced from the atmosphere and passed through an electrostatic filter and a precondtioner where the temperature is raised or lowered to 55 F. During the summer, this preconditioned air is passed over cooling coils to further lower the temperature before it enters the laboratory. During the winter or heating season, the air is passed over heating elements located in the ceiling light fixtures before it enters the lab. The air in the lab is changed a minimum of 13 times per hour and flows from around the ceiling light fixtures to the floor, which the exhaust air is picked up at 31 locations in the lab (see Figure 2). Each exhaust system is equipped with a prefilter, a high-efficiency fire-resistant filter, a pressure-regulating damper, a pressure-indicating manometer, and a pitot tube. Thus, the volume of air being exhausted at any location can be adjusted and maintained with a fair degree of confidence. A much smaller amount of air is exhausted from the laboratory through open-front hoods and air-atmosphere glove boxes.

Because the laboratory was constructed in separate sections, the blowers that drive the exhaust are situated at three different locations. The exhaust for the initial section is provided by three 1000-cfm blowers located on the roof near the front of that section. Three additional 1000-cfm blowers were provided for the first building addition. Each of these three-blower systems was designed with common manifolds so that failure of any one blower would not result in a loss of laboratory exhaust. The second building addition was designed with two 7200-cfm blowers centrally located on the roof. This exhaust system is operated with only one blower in use at a time, the second blower being maintained on a standby basis to automatically actuate when the first blower fails or is purposely shut off. To insure against a loss of laboratory exhaust in the event of power failure, all eight blowers are wired into an emergency power supply.

In order to control any accidental release of contamination in the laboratory, the entire building has been pressure regulated to prevent the spread of particulate matter from one room to another or to the surrounding atmosphere. As shown in Figure 3, the hallways and hot-change rooms are maintained at a pressure greater than that of any of the laboratory rooms, and the building itself, with the exception of the office area, is maintained at a negative pressure to the outside atmosphere. The laboratory room in which Pu^{238} activities are conducted is maintained as the most negative pressure area since Pu^{238} represents a somewhat greater radioactivity hazard than Pu^{239} because of its corrosive attack of gloves and plastic pouches, and its somewhat lower MPC. Atmospheric pressures in the laboratory are normally controlled from minus 0.05 in. of water in the hallways to minus 0.20 in. of water in the Pu^{238} room. This regulation assures contamination control and permits only gentle air currents from area to area as doors are opened and closed.

Glove-Box Atmosphere Control

Three separate types of glove-box atmospheres are in service in the laboratory in accordance with the particular operations conducted. These include

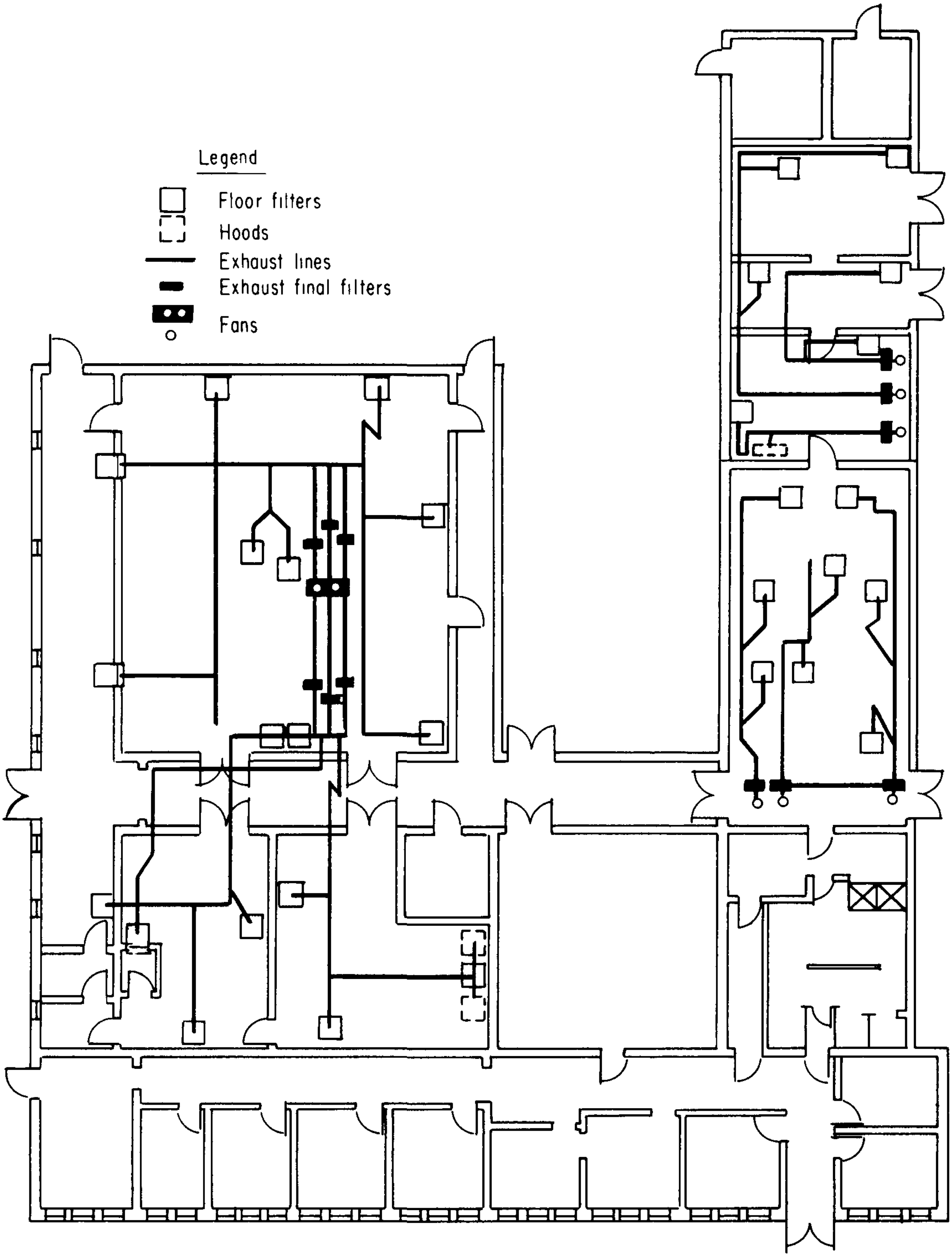

FIGURE 2. PLUTONIUM LABORATORY EXHAUST SYSTEM

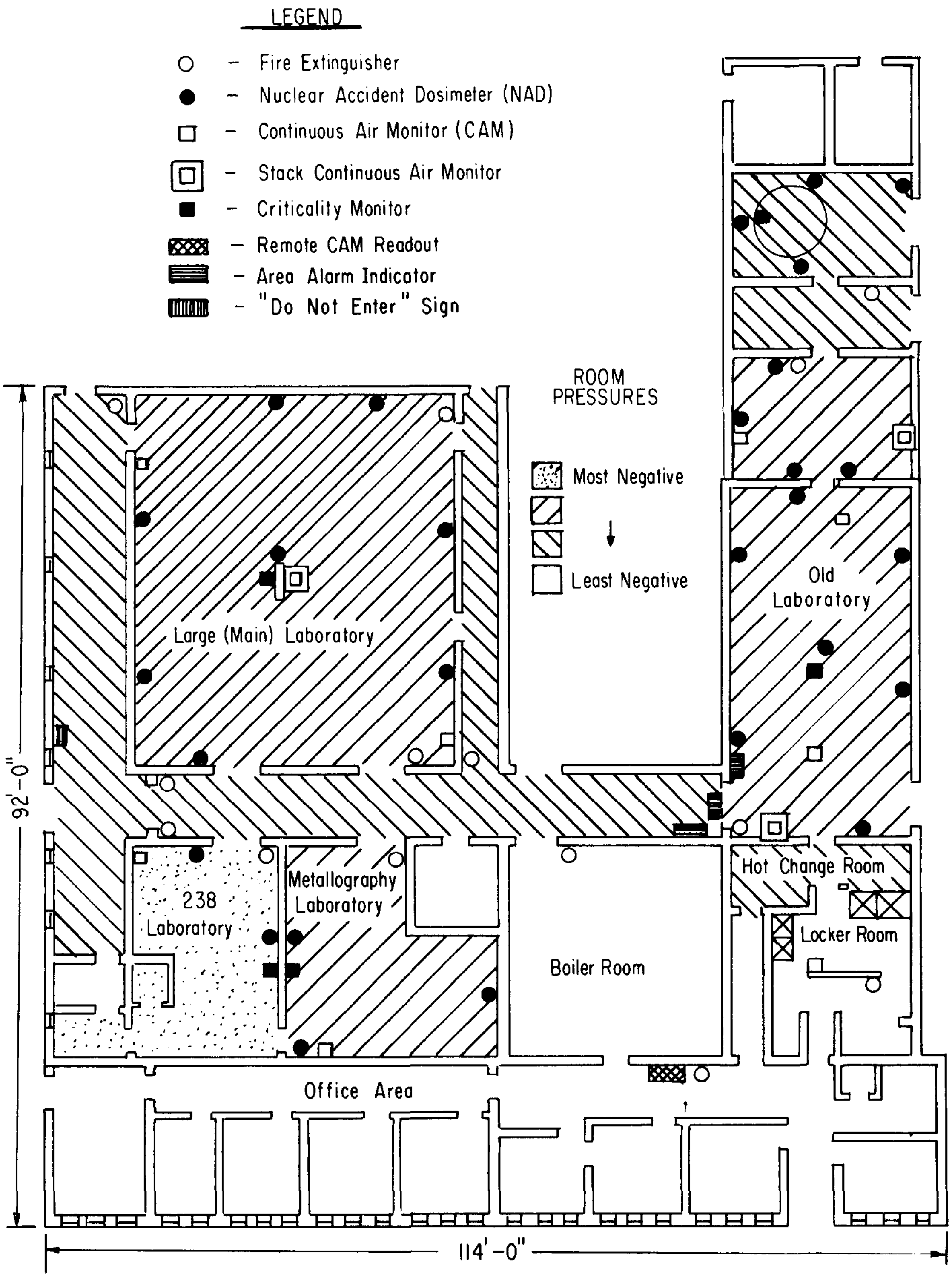

FIGURE 3. PLUTONIUM LABORATORY CONTROL AND ALARM SYSTEMS

the following: (1) once-through dry-nitrogen atmosphere for conventional operations, (2) high-velocity-air atmosphere used mainly for chemical and decontamination operations, and (3) vacuum and high-purity inert-gas atmospheres for work with highly reactive materials. Of the 43 glove-box units in the laboratory, 37 of them are normally maintained under the dry-nitrogen atmosphere, with the remaining six boxes and three open-front hoods being utilized with high- or low-velocity air. Five of the nitrogen-atmosphere boxes can be evacuated and utilized for specialized operations. By employing the once-through dry-nitrogen system of maintaining an inert atmosphere, it is believed that the potential for fire hazards is greatly reduced and the contamination of plutonium-containing material from oxidation is more easily prevented. Nitrogen for the laboratory is supplied from a liquid-nitrogen tank with a capacity of 481,000 cu ft; this tank feeds an atmospheric evaporator for conversion to gas. The resulting high-purity nitrogen is supplied to the glove-box control systems at a pressure of 20 psi. The flow of gas through a glove box is regulated by a pressure exhaust valve and a flow meter on the gas inlet as shown in Figure 4. A sensing line transmits the pressure in the box to the exhaust valve, which regulates the exhaust flow. In like manner, a line from the box to a pressure-sensitive switch is used to regulate the inlet gas through a solenoid valve. A separate magnehelic gage senses the box pressure and it utilized for adjustments and as an indication of the general condition of the control system. The pressure in the boxes is maintained from a lower limit of 0.5 to an upper limit of 1.5 in. of water negative to the room atmosphere. The nitrogen exhaust from each box is filtered at the box outlet and is dumped into the exhaust ducts that also remove air from the rooms.

One of the features that is presently being built into the glove-box system is an oxygen-monitoring system, which will measure the oxygen content in each box. Once this system has been completed, an alarm system and readout panel will be installed to enable immediate location of boxes with atmospheres exceeding some given limit of oxygen. This upper limit of oxygen will probably be set at a level which will not support combustion. The oxygen-monitoring system will act as a means of alerting laboratory personnel to the existence of a hazardous condition within a box. This system will also serve to alert personnel to the initial stages of a deteriorating situation and provide adequate time for maintenance prior to degeneration to a serious condition.

Shielding

Since the greatest portion of activities conducted in the laboratory involves Pu^{239}, the amount of shielding needed for the resultant alpha emission is minimal. However, several research programs have been conducted using Pu^{238}, which also emits gamma and neutron radiation as well as the alpha. To cope with this problem, one room in the laboratory has been equipped with glove boxes that have been shielded with both neutron and gamma absorbers. A framework was attached to each box on which 4-in.-thick slabs of boronated polyethylene were tailored to the glove-box contour. This shielding reduced the neutron exposure by about 50 percent. Although greater neutron shielding would be desirable for handling kilogram quantities of material, the thickness required would impose severe restrictions on glove-box operations conducted therein. To reduce the gamma radiation emitted from Pu^{238}, a quarter inch of lead was placed over the outside surface of the boronated polyethylene. This shielding reduced the gamma exposure by about 50 percent. The weight and construction of the lead sheet limited the amount of gamma shielding that could be used. Protection for the eyes was provided by overlaying each window with a 4-in.-thick slab of optical-grade Plexiglas. This shielding serves as a barrier to only the neutron radiation, reducing the exposure by about 30 percent.

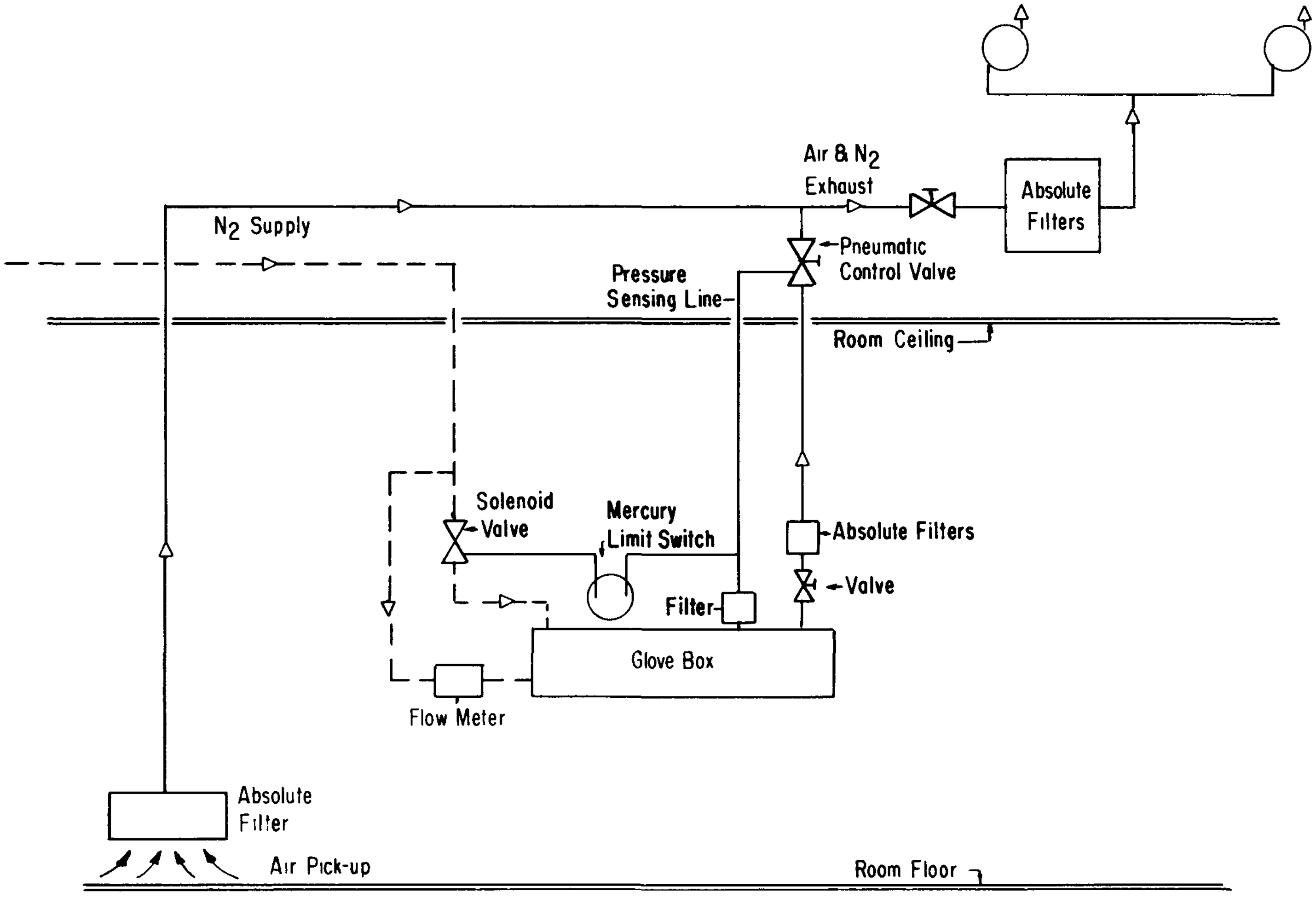

FIGURE 4. GLOVE BOX NITROGEN SUPPLY AND EXHAUST SYSTEM

SAFETY CONTROLS

In operating and maintaining a safe plutonium facility, it is vital that reliable procedures be developed, sound training be presented to the employees, and each of the facility systems be utilized in the most knowledgeable manner. The safety controls include not only those designed and installed in the facility, but also those that involve the behavior of personnel in the daily routine activities as well as emergency situations as they occur within the laboratory.

Laboratory Ventilation System

As previously described, air in the laboratory is introduced at the ceiling level, drawn to a floor pickup station equipped with absolute filters, and drawn through the exhaust duct by eight exhaust fans. The glove-box atmospheres, primarily nitrogen, are likewise exhausted through in-line absolute filters and drawn into the same duct as that used to remove air from the laboratory. The exnaust gas is then drawn through final absolute filters prior to being passed into the outside atmosphere. This system provides for double filtration of all gases emitted from the facility. All of the absolute filters used in the room exhaust ducts are equipped with pressure manometers to indicate when the resistance to air flow has increased to the point where a filter change is warranted. The system is somewhat self-balancing in that the cleaner filters draw slightly more air, which causes them to collect more dust particles and in turn reduce the air flow to the amount passing through other filters.

The absolute filters used throughout the Plutonium Facility are purchased from the Cambridge Filter Corporation and are flow checked for integrity prior to receipt at Battelle. Although present filters were not DOP tested upon installation, action is presently being taken to include this is our operations as another means of assuring total containment of contamination within the facility.

Liquid Effluent Disposal

The facility is equipped with two 6000-gal-capacity holding tanks to accumulate liquid effluent that is suspect for contamination. The "hot" drain system, which includes all laboratory floor drains, a hot sink for the disposal of cleaning water, and a hot-personnel shower, dump directly into these tanks. All other laboratory drains, which exist as covered standpipes and the normal-personnel showers, are fed into a "cold" drain system that is dumped into the normal facility sewage system.

Present operating procedures dictate that the water in the holding tanks be agitated by internal mixers prior to sampling for contamination. If this radioassay shows the contamination to be less than AEC regulated MPC levels, the effluent is dumped directly into the normal drainage paths. If contamination is found to be above permissible guidelines, the liquid is diluted to acceptable levels prior to being discharged or it is removed to the Battelle-Columbus Hot-Cell Facility for concentration by evaporation. However, with the trend toward greater pollution control, thought is being directed toward removal of any highly contaminated liquid effluent from the tanks by a government approved organization for ultimate disposal.

Small-capacity water recirculation systems which combine a pump, reservoir and heat exchanger are used to isolate water that passes into the glove boxes from the house supply.

Nuclear Materials Control

Control over nuclear materials at the laboratory is maintained as an integral part of the overall Battelle-Columbus Nuclear Materials Control System. Personnel functioning under this system maintain records of the location and amount of fissionable material involved in both government and industry sponsored programs. Members of the Nuclear Materials Staff exercise jurisdiction over all incoming shipments and off-site removals. Routine plutonium scrap is prepared for shipment in accordance with the existing AEC regulations. Plutonium scrap generated from programs utilizing license material is properly segregated in order to facilitate billing of losses and recovery charges to the appropriate sponsor. All plutonium not directly involved in processing is stored in a vault within the Critical Assembly Laboratory, which is located approximately 100 yards from the Plutonium Facility. The vault is equipped with radiation detection monitors and a criticality alarm system. The vault custodian, a member of the Nuclear Materials staff, controls access to the vault and is directly responsible for the receipt and removal of fissionable material.

In like manner the Plutonium Facility is directly responsible for all fissionable material received at the laboratory, processed, and subsequently transferred to the vault. For the purpose of criticality safety, a custodian maintains all internal inventory records and to assure control, perpetual inventory sheets are maintained for each glove box. These inventory sheets are used to record the location of all nuclear materials in the laboratory at all times. The glove-box sheets, which identify composition (with the amounts of various fissionable isotopes) and project, are the sole responsibility of the custodian and are maintained in the Accountability Office within the laboratory area. A periodic internal audit of the glove-box sheets is performed by the vault custodian to check the accuracy of the recorded figures. At the end of each month, the laboratory custodian submits a detailed inventory balance to the Nuclear Materials Central Accounting Office. Separate and distinct forms are utilized for AEC contracts and license material. At the Nuclear Materials Control Accounting Office the report is checked for accuracy and verified with the permanent control ledgers.

As a further means of maintaining control over movement of fissionable materials within the facility, the accountability system has been incorporated into a computer program. Although the system is still in a test stage, it is used in bookkeeping, material control, inventory, specimen location, and other routines designed to assist in nuclear materials management.

Laboratory Alarm Systems

Three distinct alarm systems have been installed in the facility to alert personnel of the existence of an emergency situation. These systems are the criticality detection system, airborne-contamination detection system, and glove-box-fire detection system. To detect and alarm for a criticality incident producing 300 rem per hr at 1 ft occurring anywhere in the laboratory, five gamma monitors are provided at central stations, shown in Figure 3. These monitors are set to alarm at 10 mr per hr and have a response time of 500 msec. Upon excitation, the gamma alarm actuates an audible emergency warning system consisting of howler horns located throughout the facility.

The detection of airborne contamination is accomplished through the use of seven continuous air monitors (CAM) located strategically throughout the lab. The instruments are adapted to an alarm system triggered to activate upon detection of alpha radiation slightly above background levels. The alarm is an audible continuous bell ring which is readily recognized and very unlike the criticality alarm. To assist in an emergency situation involving airborne contamination in the laboratory, each CAM is read out on a continuous recording strip chart located in the office area of the laboratory. This read-out provides instantaneous location of plutonium release and assists in immediate recovery action. The spread of airborne contamination in the laboratory is prevented by the atmospheric pressure control provided by the ventilation system. An intercom system with stations throughout the facility is also available for use during emergencies.

The third alarm system in the laboratory is the fire detection device attached within each glove box, which activates an alarm in a control room at the office area. The fire detectors are incorporated into an ADT alarm system set to activate with an audible clanging bell sound when a temperature of 180 F is reached in a glove-box atmosphere.

The three alarm systems are checked at regular intervals to insure their integrity. The criticality and air-monitor alarms are tested on a weekly basis by Battelle personnel and the fire alarms are checked monthly by ADT. All alarm systems in the laboratory are wired to the emergency power supply system which is automatically activated within 15 sec of an electrical power failure. Emergency power is provided by a natural-gas-powered 100-kva generator with an automatic starter located in the mechanical room of the laboratory.

Other detection systems incorporated in the laboratory are: (1) continuous air monitors on each ventilation stack which trip a warning light and continuously read out at the office area station, (2) warning lights in the control room of the office area that indicate when each liquid holding tank is full. Both of these systems are important in maintaining safe operations but are not adapted to audible alarms since they are not vital in safeguarding laboratory personnel. Aside from these systems, wall-mounted nuclear accident dosimeters are positioned throughout the laboratory to provide data that could be vital in determining the severity of an incident. Also, hand-and-foot monitors are stationed strategically for personnel use so as to permit the detection and isolation of contamination as close as possible to the source. Although the glove-box oxygen-monitoring system has not yet been completed, it will eventually be tied into a local audible alarm to indicate the need for immediate action.

Emergency Procedures

Since the Plutonium Facility is located about 17 miles from the main laboratories of Battelle-Columbus, the site must be self-sufficient in handling emergency situations. To aid in these needs, an Emergency Control Center has been established in the Critical Assembly Laboratory nearby. This center is equipped with radiation detection monitors, forced-air respiratory equipment, first aid needs, and telephone service. In the event of personnel injury, an emergency squad can be summoned from the small town of West Jefferson, Ohio, which is only about 2 miles away. In the case of a major fire, the West Jefferson Fire Department can supply the necessary equipment.

In the event of an emergency, the Laboratory Operating Supervisor or his deputy is in charge of emergency procedures. He is assisted by either the laboratory or site health physicist. If the emergency is of a serious nature, a distress call would be made to the Battelle-Columbus Laboratories for emergency assistance. This act would mobilize the Emergency Control Group for immediate assistance and notify standby units of the particular situation. A local hospital has been fully equipped and is staffed for handling radiation accident victims and is maintained on a standby basis.

Laboratory personnel have been trained in emergency procedures and rely upon the "buddy-system" during evacuation. In the event of a laboratory evacuation all personnel are trained to assemble upwind. The plan calls for a check of those present in order to determine whether someone may be trapped or injured within the facility. At this point emergency procedures and recovery action are instituted. All personnel have had training in first aid and the use of respiratory equipment.

Health Physics and Safety Monitoring

Basic policy and safety procedures at the laboratory are reviewed and evaluated by the Heavy Element and Nuclear Safety Subcommittees of the Radiological Safety Committee. The Heavy Element Subcommittee concerns itself with the handling procedures involved in plutonium work and the necessary monitoring devices required to safeguard all laboratory personnel. The Nuclear Safety Subcommittee involves itself primarily with the handling of all special nuclear materials (SNM) and hazardous situations, such as criticality, that could result from inadequate control. Current standard operating procedures allow a maximum of 300 grams of SNM in glove boxes where water is present and 1000 grams of SNM in glove boxes where no water is present. These limits may be raised either permanently or for short periods provided the existence of adequate safeguards is insured to the Committee.

Smear checks of the laboratory, as well as the changing of air samples, is conducted on a weekly basis unless otherwise warranted. All operations which involve breaching of containment are presented to the operating supervisor and health physicist prior to initiation to insure that procedures for containment of contamination are well formulated. Activities which require the use of respiratory equipment are attended by the health physicist. Records are maintained of all laboratory radiation monitoring and any personnel exposure. These are reviewed by the Battelle-Columbus Safety Office and AEC inspectors.

Aside from the regulation surveys of the Radiological Safety Committee and the daily radiation safety activities of the facility health physicist, the Battelle-Columbus Safety Office conducts a monthly audit of equipment and working conditions in the laboratory. A review of this audit is sent to the Operating Supervisor for comment and subsequent action. An intense safety audit is conducted about once a year which involves all facets of the operation and all systems of the laboratory.

Education and Training

Although a rigid schedule for education of personnel has not been adopted, seminars, training films, and lectures are provided on a frequent basis. Upon initiation of employment at the Battelle-Columbus Plutonium Facility, each employee is acquainted with the general operating conditions by the Operating Supervisor and is presented with the facts of radiation safety and control by the facility health physicist. Each new employee is directed in his daily activities by experienced laboratory personnel until he is capable of working on his own. Throughout this period the employee is gradually acquainted with the potential hazards of handling plutonium, and this training is supplemented by the safety and health physics seminars. The overall effort of employee education is greatly enhanced by the assignment of most technicians to project engineers on specific phases of research programs. Since program objectives generally necessitate sound technician performance, a close working relationship is usually adopted between the technician and engineer; a new employee's activities are thereby closely supervised and controlled. Aside from this type of training, many of the technicians attend Battelle-sponsored formal education programs. These may vary from technical subjects presented at a college level to industrial subjects and skills such as welding and refrigeration presented by vocational schools and commercial groups. Several correspondence courses have been taken by technicians on subjects, such as electronics, which are directly related to their daily laboratory activities. Each employee has been given a notebook of operating procedures with which he must become acquainted and maintain in an updated condition. When new procedures are established, a meeting is generally held and all personnel are informed of each change in an operation and the necessity for it.

In order that a record may be kept of each employee's training advances, all seminars and training sessions are documented and kept on file. Although a more rigorous educational and training program may be beneficial, program schedules often limit the numbers of hours that may be economically devoted to this cause. In general, the personal concern of each technician will many times offset much of the training needed to bring the individual to a safe working level.

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ADMINISTRATIVE PROCEDURES AND CONTROLS IN PLUTONIUM PRODUCTION AREAS

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Rocky Flats Division
Golden, Colorado 80401

April 14, 1971

U. S. Atomic Energy Commission Contract AT(29-1)-1106

ADMINISTRATIVE PROCEDURES AND CONTROLS IN PLUTONIUM PRODUCTION AREAS

W. H. Hauschildt
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Rocky Flats Division

The Dow Chemical Company has been about the business of handling and processing plutonium as a contractor for the Atomic Energy Commission since May of 1953. When it was awarded its first and subsequent contracts for the operation of the Rocky Flats Plant, Dow was well aware of the hazards of operating an industrial plant. Dow was also cognizant of the additional hazards involved in operating a plutonium processing facility. It was The Dow Chemical Company philosophy then as well as now that the plant would be operated with maximum safety, security, and efficiency. With the exception of the fire on May 11, 1969, Dow has fulfilled these goals quite well with respect to the Rocky Flats Plant operation.

During the past 18 years operating personnel have changed, administrators and managers have changed. Capacity, capability, technology and product have changed. The only constant, one might say, has been the plutonium, but even that has changed. Change, inherent with all industry, with all things, is a concern to managers in their efforts to provide continuity with the past, present, and future in the area of procedures and controls. The May 11 fire at Rocky Flats made us painfully aware of the need to control continuity in procedures for the handling and storing of plutonium in process areas.

Immediately following the fire we reviewed in depth our management philosophy and operational concepts to determine how we could apply our most successful management techniques to this area of concern. Our outstanding industrial safety performance led us to pursue in depth our multi-faceted program responsible for this achievement. We searched for ideas which could be adapted to plutonium handling and storage in process areas.

We determined that the Standard Safety Practice Manual, just one aspect of the total safety program, offered an excellent foundation for the administration and control of handling and storing techniques in plutonium process areas. The manual, a compilation of safe operating techniques based on proven experience at this and other Dow plants was issued and amended to provide uniform practices throughout the plant on key industrial operations

The Table of Contents gives some idea of its scope. The introduction states the manual to be:

1. A compilation of current general standard safety practices for the Rocky Flats Division.
2. It provides information that will prevent injury to personnel and damage to property.

3. It directs itself toward those practices which have plant wide application or which are related to interfaces between departments.

I have used this summary to establish the direction we took in the area of plutonium handling and storage.

Prior to the fire, the specific procedures for handling, storing and even processing within product specifications had been the responsibility of the managers of each functional plutonium processing group. The procedure at process interfaces for handling, storing, and processing were worked out by managers of the interfacing groups and changed as required to meet the needs of these specific groups. In addition, the managers of plutonium process areas obtain guidance from the Health Physics Group, the Nuclear Safety Group and the Safety and Loss Prevention Group.

We realized after reviewing past procedures and practices that the administration and control of plutonium in process areas of a multi-functional and multi-building facility must have a uniform guideline or code like the Standard Safety Practice Manual. We also determined that a change in guidelines must be reviewed by higher management who were in a better position to determine the impact of such a change on the entire plutonium processing complex. In addition this guideline, of necessity, must be complimented with guidance supplied by Nuclear Safety, Health Physics, and Safety and Loss Prevention.

The standardization of plutonium handling and storage would seem a rather simple task, but because of the past practice of each group determining its own methods, a built-in resistance was established so that their particular procedure was "the only way to fly".

The task of establishing standard procedures was initiated by requesting all groups to reduce to writing (if not already in writing) their methods of handling and storing plutonium. From this information a series of detailed flow sheets were drawn up and a number of common denominators were established:

1. Chemical and physical forms of the plutonium
2. Location of the particular processing phase such as in-line, external to the line, within the building or inter building.
3. Definitions of the types and time periods of handling and storage

This information was then codified for each physical or chemical form of plutonium involved - the type of container, type of packaging and other information necessary to the handling and storage operation was recorded. In many cases some early recommendations were tried and tested to determine the feasibility of operating within more restricted guidelines. Surprisingly enough we found that in many cases we could operate with tighter controls and more restrictive procedures.

Once the handling and storage procedures were accepted by all groups, certain deficiencies in the process areas which could not meet the established criteria were exposed. Action was then initiated via maintenance or capital construction to correct substandard conditions with the inherent deviations in the procedures until the conditions could be corrected. In no case was a deviation allowed to continue that presented high potential for fire, contamination or other conditions.

The establishment of standard procedures does not guarantee safety in plutonium operations however it does make all personnel now and in the future, who handle plutonium aware that a set of standards and guidelines do exist, they were established by operating people and must be followed until acceptable alternate methods can be tested and proven.

No matter how restrictive they may appear there was reasoning behind the selection of these methods and that present personnel had a piece of the action in determining the best and safest way to process plutonium.

Our future success at Rocky Flats in plutonium areas depends a great deal on how well we utilize these guidelines in the plutonium process areas.

SAFETY CONSIDERATIONS IN THE NEW ROCKY FLATS PLUTONIUM RECOVERY PLANT

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April 14, 1971

U. S. Atomic Energy Commission Contract AT(29-1)-1106

SAFETY CONSIDERATIONS IN THE NEW ROCKY FLATS PLUTONIUM RECOVERY PLANT *

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ABSTRACT

A new chemical recovery facility will incorporate improved health, fire, and safety requirements as determined by the Atomic Energy Commission and The Dow Chemical Company. An architect-engineer will study four construction alternatives. Of the proposed alternatives, Dow feels only a new building complex can meet these improved standards. These standards require lower exposure, reduced fire potential, and reduced contamination.

INTRODUCTION

The new complex will have 50,000 to 60,000 square feet of process area on the upper level and an equivalent area in the lower level for heating and ventilation and plutonium storage. Storage is integral to the building.

GENERAL

Building: The process areas are divided into modules. Each module varies in size but averages 50 feet by 75 feet. Each has a minimum of two walls adjacent to a hallway. Modules will have two to four hour fire walls, supplied air drops, continuous air monitors, gamma detectors, and sprinkler system throughout the building. Alarms will annunciate in the module, the main building zone panel, and other appropriate plant locations. The building will meet or exceed all fire, safety and health requirements.

An incinerator and barrel storage will be part of the building, but will have separate H&V and contamination zone. An enclosed dock will be provided so that trucks can be loaded or unloaded inside the building, then the material moved through more air locks for unpackaging.

Storage: Storage will be a stacker-retriever system with computer control and a continuous inventory. Storage will be inert and positive in pressure to the glove box lines. The storage areas will be connected to the modules and glove box lines. As a result, the storage vault is "hot." Solids are stored, inventoried, and transferred in this system. The stackers are the only moving equipment inside storage and can be

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*Work performed under the auspices of the U. S. Atomic Energy Commission Contract AT(29-1)-1106.

withdrawn to a maintenance area without personnel entrance into the vault. The stacker will handle a pallet that has four shielded storage containers on it. Each pallet container is sealed and the material in the container is inside another sealed can. The retriever will also transfer and store equipment using pallets without storage containers.

The system will essentially eliminate all bag outs and reduce in-line generated waste significantly.

Modules: Generally, each module contains one unit operation. The modules will be separated by maximum possible fire-rated walls or hallways. Most modules will have control rooms for operation control and monitoring which will reduce exposure by reducing operator time at the glove boxes.

Glove Boxes: Glove boxes will be double walled--an inside and outside wall with water in the annulus. Windows will be standard design and have an additional shielding window. Box will be of all welded construction with flanges only at conveyor junction. These flanges will be protected from vapors and liquids by ventilation flow, doors, and slopes. Some lines will have two operating sides and will be separated internally by a shielding wall. Downdraft air locks will be provided on each line for adding cold material. Equipment will be sized so that it can be moved directly to a maintenance line through the retriever without requiring bag outs.

All lines will have heat detectors, and some high fire potential lines will be inert.

Heating and Ventilation: Glove box exhaust lines will vent to the lower level into the plenums. All plenums, fans, and refrigeration system will be on this level. Booster plenums will be increased in number and obligated to various modules with the ability to divert from one booster to another. The incinerator and barrel storage area will have a separate H&V system.

H&V will be further zoned within a module. The glove boxes will be set on a continuous beam and partitions will extend from the ceiling to the top of the boxes. This will form a contamination control zone. Air will be supplied from the top and exhausted at the bottom.

Nuclear Safety: Equipment will be designed critically safe or be shielded by two feet of concrete if not critically safe. Ring-filled tanks will be placed in two-foot thick concrete enclosures. Criticality drains will gravity feed to tanks in the lower level.

Piping and Tanks: Tanks and piping outside the glove boxes will be enclosed in secondary protective enclosures that have separate exhaust system. These enclosures will also have Continuous Air Monitors for contamination detection. Only welded line is permitted in piping runs outside the glove boxes or tank enclosures.

PROBLEMS ASSOCIATED
WITH PLUTONIUM-238 PROCESSING

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April 14, 1971

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PROBLEMS ASSOCIATED WITH PLUTONIUM-238 PROCESSING

INTRODUCTION

The continued interest in plutonium-238 as a power source for aerospace and medical applications, as well as other potential uses, indicates an increased volume of laboratory studies of this isotope--and eventual production-scale processing.

Laboratory work with plutonium-238 over the past two years at this laboratory has revealed several characteristics of plutonium-238 which differ from those of plutonium-239, and require different operating techniques and safety precautions. It is the purpose of this report to document these characteristics; and, where possible, emphasize operating methods which have been found effective.

This report covers only the processing performed by the Atlantic Richfield Hanford Company, Richland, Washington--conversion of plutonium nitrate solution to oxide via oxalate precipitation and calcination. Dissolution of the target element, preliminary separation, and decontamination were performed by Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Washington.

SUMMARY

The high specific alpha activity of plutonium-238 causes several unusual problems in handling and processing this isotope. The problems result from the self-heating effect, the generation of gases by radiolysis of nitric acid solutions, and the neutron field produced through (α, n) reactions.

However, plutonium-238 can be handled safely provided the proper techniques are used and the necessary precautions are taken.

DISCUSSION

GENERAL

The unusual problems in working with plutonium-238 are due to the following properties of the isotope:

- High alpha activity--almost 300 times that of plutonium-239.
- Spontaneous heat generation--0.56 thermal watts per gram of plutonium-238.
- Gas generation in nitric acid solutions of ^{238}Pu, identified as N_2, O_2, H_2, and CO--or CO + NO.(1)

RADIATION EFFECTS

An unusual phenomenon noted is the slow response of ionization detector alpha survey instruments to ^{238}Pu alpha. On occasion a survey performed slowly enough to detect ^{239}Pu contamination has given very little, if any, instrument response. However, when the probe was held over the suspected spot for 5 to 10 seconds, contamination of up to 20,000 d/m was revealed. No explanation for this has been found, but it has been noted by both laboratory technicians and radiation monitoring personnel. Possibly another type of survey instrument would eliminate this problem.

The high alpha activity results in significant neutron yields through (α, n) reactions with low Z elements,(2) present either as impurities or in chemical combination with the plutonium. While no neutron problem was encountered in handling plutonium nitrate solutions, upon conversion to PuO_2 in approximately 100-gram batches, a significant neutron field resulted that was tolerable with moderate shielding. Preparation of other plutonium-238 compounds should be preceded by a careful consideration of possible neutron emission. Plutonium fluoride is particularly active, as shown in Table I.

TABLE I(2)

NEUTRON ACTIVITIES FROM LIGHT
ELEMENT COMPOUNDS CONTAINING
1 g OF 81% ENRICHED ^{238}Pu

Compound	Neutrons/sec
PuN	0
PuO_2 (normal O)	11,300
^{16}O 99.76%	
^{17}O 0.24%	
^{18}O 0.20%	
PuO_2 (^{18}O reduced to 0.002%)	113
PuF_4	3×10^6
Pu_2S_3	700

Some gamma radiation is present in the ^{238}Pu feed solutions, principally from the various plutonium isotopes present. This low-energy gamma is easily shielded. Higher energy radiation is also present from ^{233}Pa (decay daughter of ^{237}Np impurity) and from the decay daughters of ^{236}Pu--mainly ^{212}Pb and ^{208}Tl. In the material processed at this laboratory, the content of these impurities was not great enough to cause a problem due to the short time since separation from the target material.

HEATING EFFECTS

The self-generated heat of plutonium-238 results in some interesting phenomena. Nitric acid solutions of this isotope, in open containers, concentrate quite rapidly. If allowed to go to dryness, the plutonium nitrate will even undergo autocalcination to the oxide. A filter cake of plutonium oxalate will convert to oxide by its own heat.

The heating effect precludes use of ordinary bag-out procedures for ^{238}Pu oxide containers. In a matter of minutes, the heat softens or melts the plastic bag and destroys the integrity of the seal. In this laboratory, the plutonium oxide was loaded in Savannah River-type shipping containers, shown in Figure 1. The operation was performed in the following sequence:

1. The oxide was weighed and loaded into the primary container in a glove box and the container sealed by torquing the end caps to 75 lb-ft.
2. This container was transferred to the glove box airlock and placed directly into a clean secondary container. The secondary container cap was screwed in place, finger-tight.
3. The assembly was placed in a plastic bag and transferred to an open-faced hood.
4. The plastic bag was removed, and the cap tightened with a wrench.
5. The outside of the container was carefully surveyed, and decontaminated as necessary.

Although this practice worked, it is makeshift at best, and requires extreme care and precise timing. A coordinated design of shipping container and loadout facility would be needed for routine production.

Hood gloves fail rapidly in plutonium-238 work, especially where oxide powder is handled. It is postulated that either fine particles soon penetrate the gloves as a result of their own heat, or that the glove material is destroyed by the alpha radiation. Hood gloves must be checked regularly and often for deterioration and failure.

A related consideration is the possibility of fires. Other sites have reported fires spontaneously ignited by ^{238}Pu heat. We have avoided this by maintaining high housekeeping standards--particularly in preventing the accumulation of rags and other combustibles. In addition, the use of flammable materials was avoided except for the plastic tubing which was used in the transfer lines. This was rinsed after each use and was replaced frequently to avoid its prolonged contact with significant amounts of plutonium. All other materials used in the processing equipment (shielding, etc.) were non-combustible.

GASSING

Generation of gas bubbles is visible in nitric acid solutions of plutonium-238 at concentrations of about 5 g/liter or more. At higher concentrations, this amounts to effervescence. The bursting of these bubbles at the surface of the solution creates an airborne mist. There is evidence to indicate that this mist undergoes self-calcination to a very finely divided plutonium oxide dust which can cause widespread contamination.

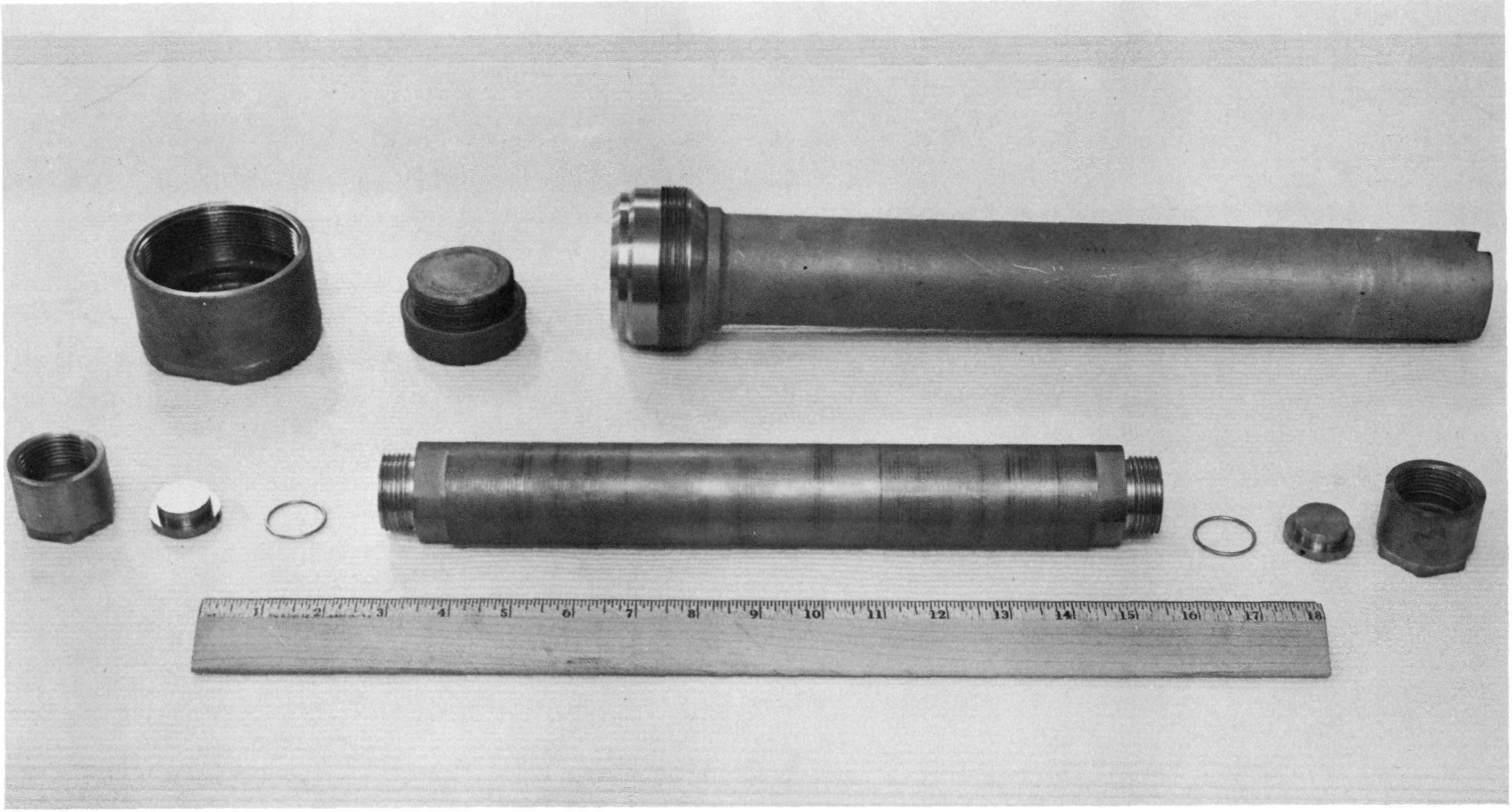

FIGURE 1

$^{238}PuO_2$ SHIPPING CONTAINERS

Because of this misting effect, a plutonium-238 solution cannot be opened to the atmosphere except in a glovebox. A temporary plastic enclosure ("greenhouse") with a slight negative pressure created by an exhaust connection to a glovebox is not adequate to prevent the spread of contamination.

A special shipping can adaptor (Drawing H-2-26288) was fabricated to facilitate loading and unloading of the plutonium-238 nitrate feed solution without opening the cans directly to the atmosphere. This adaptor is shown in Figure 2. This was later modified by the addition of a 1/2-inch ball valve to the dip leg connection with the outer valve connection sealed with a pipe plug. The unloading operation was carried out in the following steps:

1. A plastic sleeve was installed on a glove port of the ^{238}Pu hood, and the outer end of the sleeve was sealed to a short length of 3/4-inch plastic tubing.
2. With the ball valve closed, the pipe plug was removed and the 3/4-inch tube quickly sealed to the valve.
3. The valve was opened and a 1/4-inch plastic tube was threaded from the hood through the larger tube and valve to the bottom of the dip leg.
4. The solution was transferred by vacuum to the hood.
5. The same tube was used to add a flush solution by gravity to the can. The flush was then removed by vacuum, and the 1/4-inch tube was drawn back into the hood.
6. The ball valve was closed. The 3/4-inch tube was disconnected from the valve and sealed, and the plug was replaced in the valve connection.

One man probably could perform these operations with no contamination spread. However, our laboratory operating procedures, as well as prudence, require that two operating personnel must be present in the work area during a potentially hazardous manipulation.

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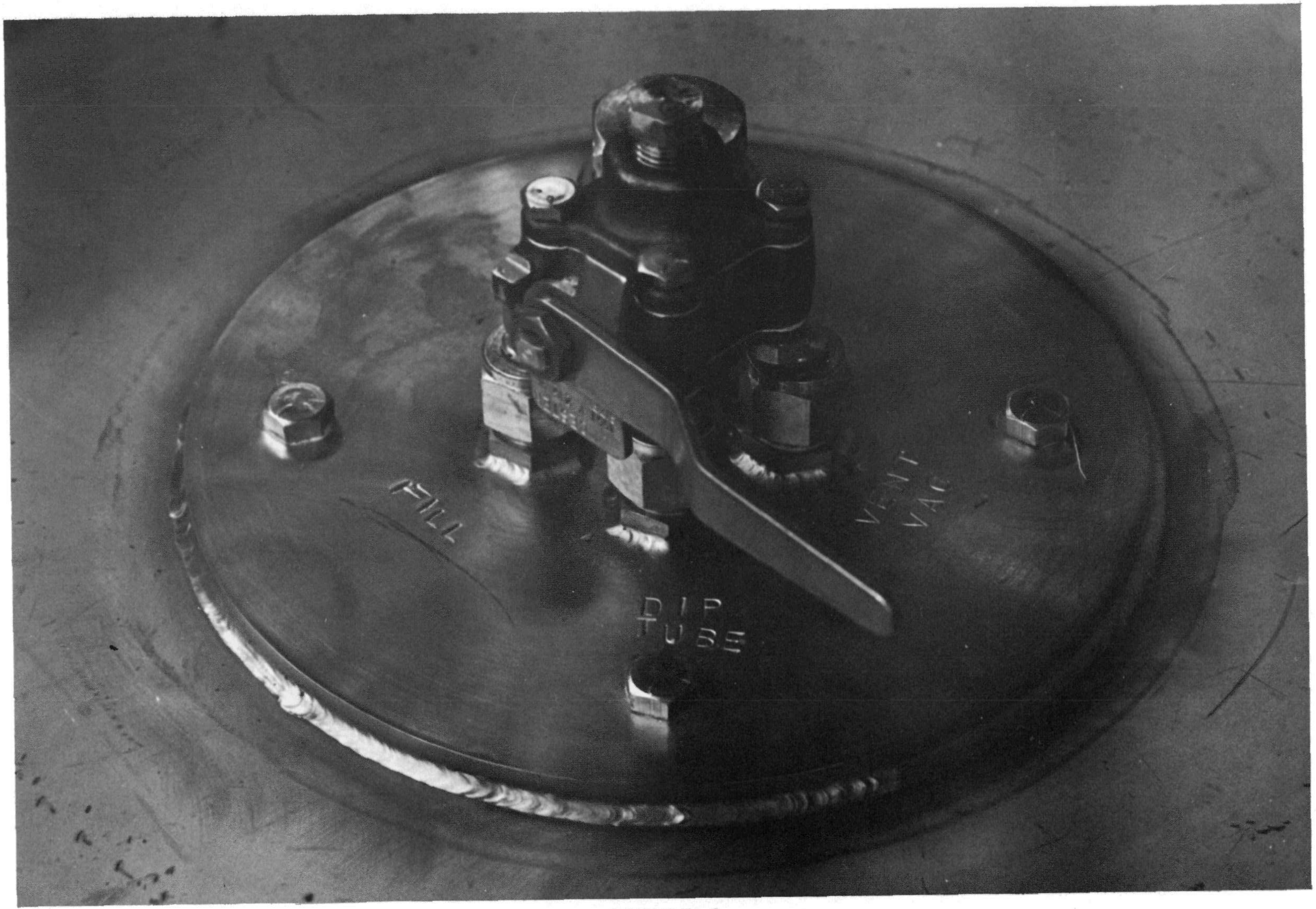

FIGURE 2

SHIPPING CAN ADAPTOR FOR $^{238}Pu(NO_3)$ SOLUTION

MATERIALS FOR SHIELDING NUCLEAR RADIATIONS FROM PLUTONIUM PROCESSING

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April 14, 1971

MATERIALS FOR SHIELDING
NUCLEAR RADIATIONS FROM PLUTONIUM PROCESSING

by

Wm. Cornelius Hall[1] and J. Merriam Peterson[2]

"Operators of nuclear facilities should be aware that chemical explosions and fire are a credible and likely occurrence in the facility. Therefore, design of the facility must provide the maximum degree of fire prevention based on operating needs."[3] Another hazard is nuclear radiation. Organic materials have proved useful in nuclear radiation shielding but are unfortunately combustible and on one occasion were the fuel for an uncontrollable fire. Thus new materials which are efficient nuclear radiation shields and are non-combustible should prove most welcome. Indeed such materials should add significantly to the safety of plutonium handling.

Neutrons and gamma photons are the principal radiation hazards of plutonium processing. Hydrogen with an atomic number of 1 (Z1) is more useful than all other elements for attenuation of neutrons. Because a hydrogen atom has approximately the same mass as a neutron, it degrades the energy of a neutron by elastic scatter. Complete conservation of both energy and momentum occurs by elastic scatter, with all lost projectile energy and momentum transferred to the target atom without excitation. Helium (Z2) is four times more massive than a hydrogen atom and lithium (Z3) is seven times more massive than a hydrogen atom. As one goes up the periodic table with increasingly more massive atoms, neutron moderation by elastic scatter becomes a proportionately smaller probability and eventually becomes negligible. When a neutron hits a massive atom such as lead (Z82), inelastic scatter occurs. Momentum is conserved by inelastic scatter, but kinetic energy is not. Some of the latter is converted to excitation energy of the target nucleus. A neutron of lower energy is then emitted. The energy differential is later emitted as gamma rays. High hydrogen content on an atomic basis is thus desired for efficient shielding of neutrons.

Although hydrogen is the most efficient moderator of neutrons, it is not the most efficient element for neutron capture. Indeed there are about a dozen elements which are more efficient for this purpose. Boron is one of these. Boron has the added advantage of emitting a capture gamma of only half a million electron volts, as compared to the several million electron volt capture gamma emitted by hydrogen. Thus it is desirable to have a high boron content with the high hydrogen content for the most efficient neutron attenuation.

Plutonium handling usually involves glove-box use, and thus minimal shield thickness is an objective. Ideal attenuation materials such as decaborane with boron-10 enrichment or lithium hydride with lithium-6 enrichment involve both hazards and high cost. Concrete, a good structural material of low cost without hazard problems, does not attenuate well enough to satisfy the need for a relatively thin shield. Plastics and cellulose, with high hydrogen contents and reasonable costs, unfortunately have not been fire safe.

Recently some non-combustible, hydrogenous materials have been developed. These are:

1. CHEMTREE 20-S with a density of about 1.5 g/cc and about 2.8 x 10^{22} hydrogen atoms per cc content. At high temperatures CHEMTREE 20-S will release water as steam and become friable. CHEMTREE 20-S is a formable material of cost considerably below that of materials previously used for this application.

2. CHEMTREE 1-5-6-20, which is available as a dry powder that requires only the addition of water as a setting agent. With a density of about 1.4 g/cc and about 4.2 x 10^{22} hydrogen atoms per cc. It is relatively low priced in large volume. CHEMTREE 1-5-6-20 has the advantage of a boron content of about 0.23 x 10^{22} atoms per cc. With a hydrogen-atom concentration over 90% that of cellulose, and a boron content to capture thermalized neutrons, its attenuation efficiency appears to exceed that of cellulose without the fire hazard of cellulose.

Part of the hydrogen content of CHEMTREE 1-5-6-20 is in the form of an organic compound, which undergoes thermal decomposition at prolonged temperatures above approximately 125°C. However, the continuous phase that provides the structural strength to the shield is inorganic and is stable at significantly higher temperatures. In this intermediate temperature range above the decomposition temperature of the organic compound, the continuous non-combustible, inorganic phase will isolate the combustible organic phase and prevent the propagation of fire through the shielding mass. The shield will remain self supporting, retaining shape and dimensions, and will not add significantly fuel to fire, even though emitted gases from the decomposing organic phase will support small flames on the exterior materials surfaces.

3. A new and improved version of CHEMTREE 1-5-20 has a density of about 1.6 g/cc, and an elemental density of approximately 3.8 x 10^{22} hydrogen atoms/cc and 1.7 x 10^{22} boron atoms/cc. It is a flowable powder which forms a workable mortar when mixed with water, and sets into a self supporting structure. With no carbonaceous content, it is stable to approximately 140°C. Even at substantially higher temperatures, none of the products of decomposition will burn nor support combustion.

Non-combustible CHEMTREE 1-5-20, with somewhat lower hydrogen content than that in the previously used combustible material, does have the advantage of a relatively high boron content for thermal neutron capture. It also is cost competitive. Therefore, CHEMTREE 1-5-20 appears to be the most appropriate choice of material for this application involving both nuclear radiation shielding and fire safety.

These new Chemtree Materials appear to be the most appropriate for shielding the neutron radiations of plutonium processing, with due consideration for fire safety, cost, weight, and thickness of the material. Special consideration should be given to the fact that these materials will remain in place and continue to shield regardless of exposure to high temperatures. The first two will, of course, have become less efficient, because thermal decomposition of certain organic components will lower the hydrogen atoms available; but certainly this is far preferable to the use of a material that under the similar conditions will and has gone up in smoke.

The aforementioned Chemtree products are only examples of more than one hundred nuclear radiation shielding materials developed to meet special biological and instrumentation situations that cover the wide spectrum of radiation fluxes encountered in the nuclear field. In this continuing program, initiated in 1961, several additional products are presently in various stages of development.

Attenuation of gamma radiation will partly be accomplished by the mass of the neutron shielding; a thickness of lead, or steel will complete this shielding.

In summary, the most efficient attenuation materials for neutrons from plutonium processing have both hazards and high cost. The less costly materials initially have not proved fire safe, and therefore they are ultimately more costly. Material of comparable efficiency and cost, with good fire retardant properties would seem the most sensible choice. Accordingly, the most nearly ideal materials appear to be the new Chemtree Materials having high attenuation efficiency, competitive cost and non-combustible characteristics. Indeed it appears that no other single step can add so much to the safety of plutonium handling as can the substitution of the New Improved CHEMTREE 1-5-20 for the previously used combustible materials.

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DESIGN FEATURES OF MOUND LABORATORY'S MEDICAL DECONTAMINATION FACILITY

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April 14, 1971

DESIGN FEATURES OF MOUND LABORATORY'S MEDICAL DECONTAMINATION FACILITY

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Mound Laboratory is operated by Monsanto Research Corporation for the Atomic Energy Commission, and is located in Miamisburg, Ohio.

Many of the programs at the Laboratory involve work with ^{238}Pu in the fabrication of heat sources. Some of these sources are: Apollo Lunar, Cardiac Pacemakers, Heart Pump, Life Support, Pioneer, SNAP, Swimsuit Heater, and Transit. Increasing program demands resulted in more personnel handling larger amounts of plutonium. There was, therefore, a need for upgrading our capability of caring for employes who were injured while working with radioactive material.

Originally, minor injuries were monitored only with alpha survey meters such as the Eberline PAC-1SA. These meters gave a good indication of surface contamination but could not detect any radioactive material under the skin. An Eberline Plutonium Wound Monitor was purchased in 1962 to provide monitoring of injuries which break the skin. This instrument is adjusted to detect only the 17 keV gamma energy of plutonium, which greatly reduces the instrument background. The probe has interchangeable collimators to aid in locating the plutonium more exactly.

No special facilities were available for injured radiation workers and minor injuries were monitored either in the health physics counting room or in the medical facility after preliminary monitoring and decontamination. This procedure was adequate until 1967 when a glove box explosion occurred, resulting in minor injuries and high level contamination to several employes. Treatment of the injuries caused contamination in the medical facility and disrupted activities until decontamination was

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*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Atomic Energy Commission under Contract No. AT-33-1-GEN-53.

completed. Previous incidents had caused contamination but were not as difficult to control. It became apparent that a separate facility was required to isolate and treat injured contaminated employes.

The new Medical Decontamination facility was constructed to provide maximum medical and Health Physics care for injured radiation workers. Many of the design features were based on our experiences in working with radioactive materials and monitoring of injuries involving contamination. These features are intended to control radioactive contamination and to prevent its release to uncontrolled areas. The new facility is located next to the medical facility which provides quick response by medical personnel, and is also adjacent to the whole body counting facility. The services of both of these facilities are available if required.

All emergency cases, whether from a radiation area or not, are brought to the south entrance of the Medical Decontamination building. Opening the door activates an alarm signal which alerts medical personnel. This signal can only be silenced from the medical area. The only other entry is directly from the medical area through an air lock.

The ventilation is provided by an air conditioning unit located on the roof of the building. Exhaust air from each room passes through a roughing filter and an absolute filter to remove airborne contamination. Pressure differentials of 0.15, 0.10, and 0.05 in. of water are maintained, with the entry air lock being most negative, to prevent spreading airborne contamination. An air sampling probe is installed in the exhaust duct on the roof to monitor the effluent air if required.

A water collection system consisting of two 400-gallon tanks collects all waste water. The full tank alarm and control panel for the pumps is located in the south entry room. This water can be sampled to determine the proper method of disposal, and can be pumped directly to the sanitary sewers if not contaminated. Contaminated liquids can be collected in metal drums and sent to the liquid waste disposal area for processing. Absorbent material can also be used to soak up the liquid so that it can be disposed of as a solid.

A decontamination table equipped with running water and a drain is available for highly contaminated persons with serious injuries. It is perforated to provide positive drainage. Walk-through showers are provided for persons with minor injuries requiring decontamination before treatment. A foot bath adjoins one of the showers. Scrub sinks are provided with knee operated controls to eliminate handling faucets with contaminated hands. All water is collected in the holding tanks.

All room lighting is provided by recessed, sealed fluorescent units, which aid in ease of decontamination when required. These lights provide 100 foot candles at table top height. Battery operated emergency lights are provided in each room.

The floor is tile for ease of decontamination and replacement if needed. The floor of the central room is electrically grounded for safe handling of oxygen.

Enough equipment and supplies are maintained in the area to handle any anticipated emergency. Additional materials are quickly available if needed. The following medical and health physics equipment and supplies are kept in the area at all times:

Oxygen
Ultrasonic nebulizer
Air monitors
Survey meters
Proportional counter
Scintillation alpha counter
Wound monitors; Rm-15 and WM-1
Vacuum cleaner with absolute filter
Complete medical and health physics supplies adequate for about 4 hours

In the event of an injury in a radiation area the following procedure is initiated:

- The emergency brigade and fire fighters with ambulance proceed to the area and give first aid at the scene.
- A contamination survey is made at the scene by area Health Physics personnel. Medical and Health Physics personnel are informed of the extent of the injury and levels of contamination by telephone or by radio from the emergency vehicle.
- Time permitting, monitoring equipment is checked and made ready for use before arrival of the injured person.
- The ambulance brings the injured person to the emergency entrance.
- For serious injures, the person is taken directly to the decontamination table for treatment.
- For minor injuries, the person is surveyed and contaminated clothing is removed. The person then proceeds to the shower.
- He is then resurveyed and proceeds to the treatment and examination room if not contaminated.
- His injuries are monitored with a wound monitor to detect internal contamination if any.

- The more seriously injured person is treated and decontaminated on the decontamination table. The portable wound monitor can be brought to the table for use in monitoring his injuries.
- Any individual who requires a recovery period is taken to an adjoining area for isolation and recovery.
- For major surgery or injury requiring hospitalization, the person is decontaminated to as low a level as possible and taken to Wright-Patterson Air Force Base hospital.

One of the more interesting cases in the past involves an employe who received puncture injuries to the hand on three different occasions while working in a glove box. This case is mentioned to show the successful results which are possible with the wound monitor.

One of the three occasions resulted in external skin contamination only with no internal deposit of radioactive material. The skin was decontaminated with no difficulty.

On one of the other two occasions, internal contamination was detected by the wound monitor and the material was excised by the medical doctor. From wound monitoring data, it was estimated that 0.35 μCi of plutonium was present in the wound. An assay of the excised tissue indicated 0.25 μCi of material. Follow-up bio-assay samples indicated no uptake of radioactive material.

On the other occasion an estimate of 0.04 μCi in the wound was based on data from the wound monitor. Assay of this excised tissue indicated 0.021 μCi. Bio-assay samples again indicated no uptake of radioactive material. The first excision incident occurred in 1966 and this employe is still working with plutonium at Mound. Continuing bio-assay samples indicate no uptake to this time.

The difference between estimated and assay amounts of material in the wound can be attributed to a more accurate assay counting method, and the presence of skin contamination near the injury.

In conclusion, we are confident of our ability to handle anticipated emergency situations with the new facility. We sincerely hope that it is never needed for its intended purpose.

ROCKY FLATS ENVIRONMENTAL RESEARCH

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April 14, 1971

U. S. Atomic Energy Commission Contract AT(29-1)-1106

ROCKY FLATS ENVIRONMENTAL RESEARCH

The Environental Research Group at Rocky Flats is a new section of the Chemistry R/D Organization. Although the group is new, most of its activities are not. The reports of Symposium speakers Domning, Johnson, Lindsay, Woodard, and Ziegler concern work which had been done in other sections of the Chemistry Group. The work is now continuing in Environmental Research.

A major concern of the group is to ensure that Rocky Flats operations have the least possible adverse effect upon the environment. The term environment is here used broadly to include not only the environment external to the plant buildings, but also to that within the buildings as it relates to employee safety and to the proper containment and management of toxic materials. Plutonium is the material of primary concern but various chemical hazards are also under study.

Table I shows one major category of the group's work--"Environmental Protection":

TABLE I

ENVIRONMENTAL RESEARCH PROGRAM

I. ENVIRONMENTAL PROTECTION

Waste Process Development
Material Testing and Evaluation
Systems Tests
Engineering and Production Support

Waste Process Development includes all projects aimed at reducing the quantity and radioactive (plutonium) content of wastes leaving Rocky Flats. Reuse and recycling of chemicals and of water are important objectives. Mr. Ziegler will detail some of these projects tomorrow.

Material Testing and Evaluation has become a very important and continuing activity. Mr. Lindsay has reported some of the work in this area. Fire safety is only one aspect of this program. Protective coatings for contamination and corrosion control are being tested. Radioactive waste encapsulation and packaging materials are under development. New materials are tested and evaluated for possible use at Rocky Flats.

Systems Tests are performed on large scale mockups of production or production support systems. Mr. Domning has discussed several filter plenum design considerations. He has tested many materials and components used in gloveboxes for fire safety under closely simulated production situations. Many tests have been performed on HEPA filters and filter plenums under fire conditions. Such tests are extremely valuable "proof" tests for materials which have been screened in our Materials Testing Laboratory.

Engineering and Production Support is a miscellaneous category of endeavors. It includes engineering studies and review of design and criteria for peripheral systems of production facilities. The experience obtained in glovebox and filter plenum fire tests is utilized for this purpose. The engineering studies of Mr. Johnson on the glovebox inerting project have provided knowhow which is useful in solving other glovebox atmosphere problems--for example the removal of carbon tetrachloride vapors from the gloveline inert atmosphere.

Production support is also provided when specific requests are made by Production for help or guidance in problem areas which may not be of environmental significance. Examples are determining the cause of difficulties in the plutonium fluoride reduction process and learning how to protect the production waste incinerator from its own corrosive combustion by-products.

Part of the task of protecting our environment involves characterizing it so that we can detect, measure, and interpret changes which may occur. A second major category of projects in our program is "Environmental Measurements." Table II shows the nature of this work.

TABLE II

ENVIRONMENTAL RESEARCH PROGRAM

II. ENVIRONMENTAL MEASUREMENTS

Environmental Plutonium

Detection and Measurement
Transport Mechanisms
Fixation

Chemical Emissions

Environmental Plutonium is a world-wide reality. Rocky Flats has contributed to the local environmental inventory of plutonium. We maintain that this poses no threat to life or health, but we are determined that continued Rocky Flats operations shall not add significantly to the present level of environmental plutonium. The program suggested by this outline is intended to improve our understanding of past events, to define accurately the present situation, and to provide guidance for avoiding future problems.

Very little sampling and analysis of soils in this area had been undertaken before the May 11, 1969 fire at Rocky Flats. Soon after the fire, Dr. Martell, of the Colorado Committee for Environmental Information, collected and analyzed soil samples from the Rocky Flats environs and called attention to the plutonium detected in the soil. Since then, the AEC Health and Safety Laboratory and the

Colorado Department of Health have taken and analyzed many soil samples. The Rocky Flats Health Physics group, itself, has analyzed over 1100 samples. All results are in generally good agreement. They do show that, indeed, small amounts of plutonium have escaped from Rocky Flats.

The Environmental Research group does not plan to duplicate this work but it is preparing to evaluate soil sampling techniques and to study the behavior of plutonium in soil. Techniques for analysis of samples with very low plutonium levels are tedious and complicated. An attempt will be made to develop more rapid procedures for survey and screening purposes.

Work described in an earlier paper by Mr. Woodard on performance of HEPA filters for fume from burning plutonium is being continued. It is being extended to plutonium particulates from simulated and actual production operations. Particle size distributions and physical and chemical characteristics of the particles generated in various operations will be studied. This will also be done for air filter samples collected in the production buildings and from the external environment. Because the activity and number of particles obtained in environmental samples will be very small, sensitivity-enhancing techniques, such as autoradiography and the induced fission-track etch method, will be used.

Transport Mechanisms for radioactive particles are being investigated so that the effects of wind, water, and weathering upon the distribution of environmental plutonium may be understood, anticipated, and possibly controlled. A program to evaluate soil stabilization techniques is underway. Chemical stabilizers and grasses are being evaluated as possible short term solutions to the problem of preventing migration of radioactive contaminants. Dr. Miner and Mr. Hagan of the Chemistry R/D group have performed preliminary studies on migration of plutonium and strontium in soil-water systems. A proposal for a more elaborate study in cooperation with the Idaho Nuclear Company is being made.

Fixation refers to the means--chemical or physical--by which plutonium is retained in soil. The mechanisms by which plutonium is fixed to soil particles or migrates among them depends on whether the plutonium is in a particulate form or a soluble form. Interactions between soil particles and plutonium particles will be studied by laboratory modeling techniques and by analysis of soil fractions. Interactions between plutonium ions and soil particles involve other phenomena which will be studied by modeling experiments and by laboratory studies of soil samples.

Chemical Emissions characterize most industrial plants. Rocky Flats has only minor problems in this area. Nevertheless, attention is being given to reducing those emissions which do occur to minimum practicable levels. Vapors of carbon tetrachloride and other chlorinated solvents are emitted from glovebox air exhaust systems. Sulfur dioxide and oxides of nitrogen are released as a result of chemical operations.

The Environmental Research group is preparing to analyze stack emissions by means of mass spectroscopy and by use of an infrared interferometer spectrometer. It is hoped that continuous monitoring and data processing can be arranged. The time/concentration emission profiles will be used to assess present conditions and to evaluate future corrective actions.

MEASUREMENT OF PLUTONIUM IN SOIL AROUND THE NEVADA TEST SITE

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ABSTRACT

Experiments conducted at the Atomic Energy Commission's Nevada Test Site between 1951 and 1963, using plutonium in both critical and sub-critical configurations, have resulted in distribution of plutonium beyond the boundaries of the Test Site. The Southwestern Radiological Health Laboratory of the Environmental Protection Agency is conducting a survey to assess the distribution and concentration of plutonium in the off-site environment.

Special sampling methods were devised since desert soil is too coarse and dry for auger and "cookie cutter" sampling techniques. Soil sample analyses are performed by a dissolution, ion exchange, electrodeposition procedure followed by alpha spectroscopy. Plutonium has been detected in four locations around the Nevada Test Site. These locations correspond to fall-out areas previously identified for the various test series. Plutonium concentrations in the top 3 cm of soil were 10 to 100 times greater than the concentration in soils from areas not subject to contamination by these series.

MEASUREMENT OF PLUTONIUM IN SOIL AROUND THE NEVADA TEST SITE

by

Wayne Bliss and Leslie Dunn

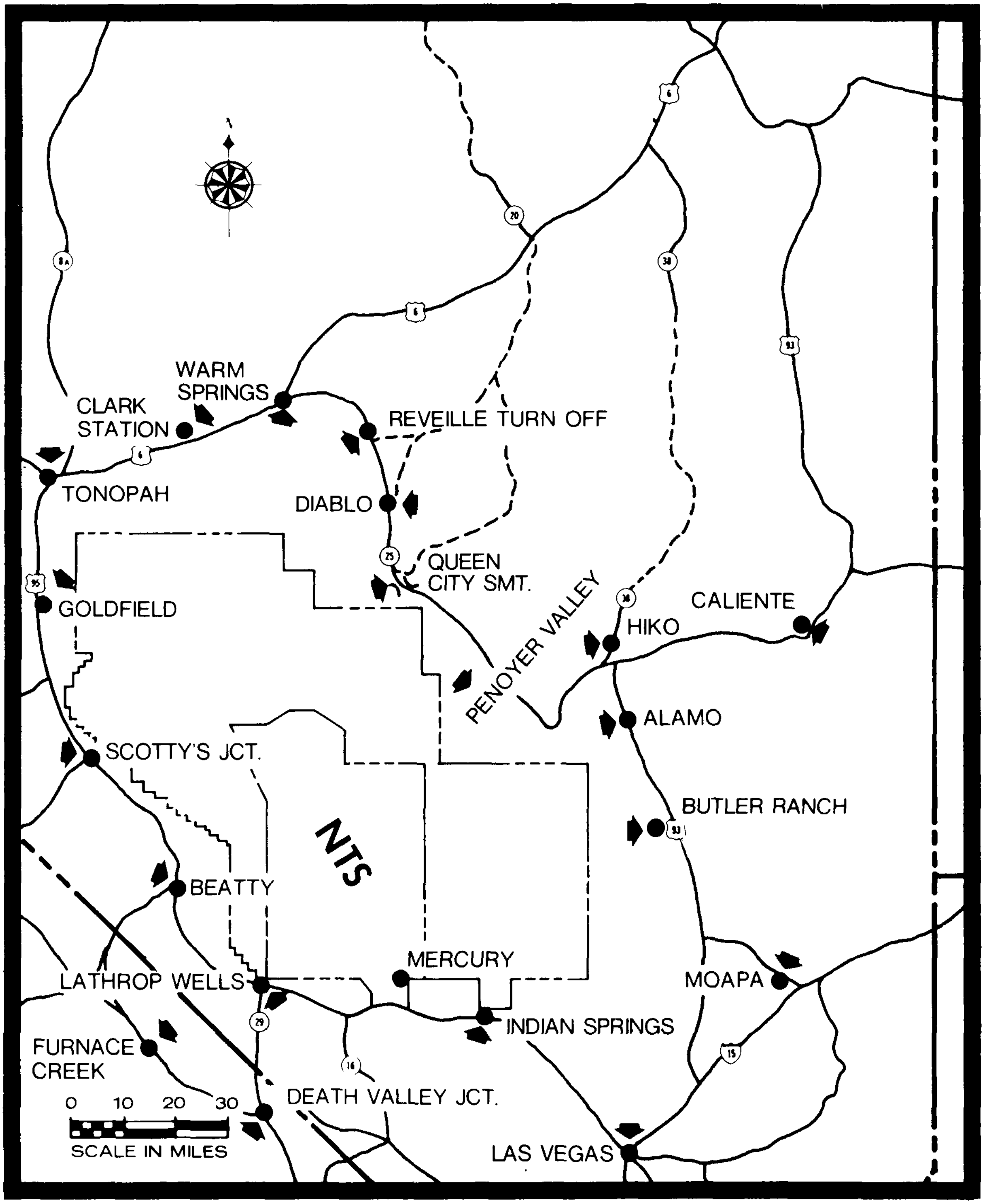
Nuclear experiments conducted by the U.S. Atomic Energy Commission at the Nevada Test Site between 1951 and 1963 using plutonium in both critical and sub-critical configurations have resulted in distribution of plutonium beyond the test site boundaries. These experiments were generally of three types. There were accidental ventings of underground explosions which contributed little, if any, to off-site plutonium deposition. There were also atmospheric detonations of full scale nuclear explosives, such as the Plumbbob series of 1957. A high percentage of the plutonium used in such devices would escape unfissioned.(1) These experiments may not have contributed largely to local off-site deposition. The third type, and probably the principal contributor to current plutonium in the close-in off-NTS area, were the so-called one-point or safety detonations. These tests were to test the effects which would result should the high explosive component of a device be accidently detonated.

As part of its responsibility for radiation monitoring around the Nevada Test Site, the Southwestern Radiological Health Laboratory (SWRHL) has been conducting a soil sampling program to determine off-site plutonium levels. The main objective of the study is to define the current plutonium distribution around the Nevada Test Site, determine if it is migrating by natural phenomena, and determine if man has been or may be subject to plutonium exposure. Should there be any health hazard, it will be shown by the study results. Concurrent with this off-site study, more detailed and complex studies are being conducted on the Nevada Test Site to evaluate soil to man routes and any related hazards. Studies of resuspension, air sampling, plant and animal sampling, and particle analysis, shall be done following this distribution survey. Procedures and results for the early phase of the off-site soil sampling study are presented in this paper.

The Atomic Energy Commission's Nevada Test Site (NTS) lies approximately sixty-five miles northwest of Las Vegas, Nevada in the Great Basin area. The soil on and around the Nevada Test Site is primarily of volcanic origin. The valleys are composed of gently to moderately sloping alluvial fans and terraces. The soil is of coarse texture with low organic content and low water holding characteristics. The mountains are steep to very steep and composed of sedimentary, metamorphic, and igneous stone.(2)

This soil survey was begun at twenty populated locations around the Nevada Test Site and two unpopulated locations (See Figure 1). These locations are both inside and outside the fission product fallout patterns defined for the test series above. Baker, California and Kingman, Arizona were selected as background stations. Initial soil samples were collected from profiles to determine vertical disposition of plutonium. Two profile samples were collected in the vicinity of each location, usually three to five miles apart. Profiles of 23 cm deep and 200 cm square were sampled with layers divided at 1, 3, 7, and 15 cm.

Since desert soil is too dry and too coarse to use "cookie cutter" or auger sampling methods, the samples were collected by a "pit technique". A pit was dug as deep as necessary to accommodate the maximum sampling depth plus some working room. One face of this hole was left vertical. From this face was trowelled or scooped the desired thickness and area layers. A fixed size scoop works well. After the scoop is inserted, its mouth may be covered with a broad



PLUTONIUM SOIL SAMPLING SITES

FIGURE 1

knife to fix the sampled area or volume. Also, it is convenient to slide a flat plate under the inserted scoop to prevent mixing any material which falls into the sampled area with the subsequent sample. After the layer is removed, surrounding material may be cleared away to prevent backfall and so on which may hinder sampling the lower layers.

Area sampling is done with a scoop technique. Not less than ten scoops totalling more than one square foot are used to composite one sample. As above, the scoop is designed for a fixed sample depth and area. To date, this has been five centimeters by 100 square centimeters. Based on the profile results, it appears that five centimeters deep will be sufficient for most cases.

All samples are prepared for analysis in a similar fashion. The sample is first screened and subdivided. There is general agreement that plutonium will reside in some fine fraction of the soil. There is not agreement of what fraction to eliminate. Some analysts discard the material more coarse than 200-mesh; some discard material more coarse than 25-mesh.(3) The SWRHL procedure uses 10-mesh as a dividing point. The more coarse material is gently ground in a mortar to break up the clods and screened. The fine fraction is divided by a riffling apparatus to provide aliquots for analysis. An aliquot sufficiently small to be handled in a 100 cc bottle (approx. 100 gm) is chosen for plutonium analysis and another aliquot of about 400 cc (about 500 grams) is selected for gamma spectroscopy.

The small aliquot is dried at 110°C, ground and mixed. One-gram aliquots are ignited at 700°C and dissolved in a Teflon beaker by digestion with nitric and hydrofluoric acids. Nitrate, fluoride, and silica are removed by evaporation to dryness followed by repeated evaporations in the presence of hydrochloric acid. Plutonium is absorbed from a 9M hydrochloric acid solution of the residue on a column of AG 1 x 2 anionic exchange resin. Co-adsorbed iron is removed from the resin with 7.2M nitric acid after which the plutonium is reductively eluted from the resin with 1.2M hydrochloric acid containing 0.6% hydrogen peroxide. The separated plutonium is electrodeposited from 1M ammonium sulfate media onto stainless steel planchets. The activity of the plutonium is determined by alpha spectroscopy using plutonium-236 as an internal reference standard.(4,5)

The 400 cc aliquot is counted forty minutes on a 4 x 4 inch NaI(Tl) crystal coupled to a 400 channel pulse height analyzer. The taped spectrum is analyzed by a matrix solution for ^{181}W, ^{226}Ra, ^{232}Th, ^{137}Cs, ^{40}K, ^{54}Mn, ^{106}Ru and ^{95}Zr.

Gamma scan results show nothing extraordinary for the locations sampled for this survey.

Typical results which have been found for plutonium are shown in Table 1. The values shown are computed from the concentration in pCi/gram at ±25% at the two-sigma confidence level. These results are preliminary and subject to minor modifications as procedures are refined or repeated analyses are performed.

Plutonium was detected in only the top three centimeters in most cases and the profile pairs agreed to within a factor of three in most cases. Three area samples were collected at Lathrop Wells to evaluate the variance within a group of cores and between locations but unfortunately they were collected at one of the disagreeing cases. Another sample was collected from a cultivated field in which plutonium was found however no plutonium was found in the barley growing there. No data correlations have yet been made between these data and data generated during the test periods when plutonium was known to have been released. It is noteworthy that Lathrop Wells was in or near the fallout pattern of many of the Hardtack, Phase II experiments and Butler Ranch lay in the fallout pattern of the Smoky Event of Operation Plumbbob. No analyses capable of defining

Table 1: Measurement of Plutonium-239 in Soils Around the Nevada Test Site

Location (see Figure 1)		Pu-239 (mCi/km^2)
Penoyer Valley	1 mi E of Co Line	130 ± 6.0
	6 mi E of Co Line	6.7 ± 1.5
Queen City Summit	profile at summit	19 ± 1.0
	(1000 cm^2 surface scraping)	22 ± 1.9
Highway 25/Reveille Turnoff		5.7 ± 0.71
Lathrop Wells	2 mi E	6.1 ± 0.94
	2 mi W	0.3 ± 0.2
	2 mi E surface	17 ± 3.2
	2 mi E surface	2.6 ± 1.3
	2 mi W surface	0
Alamo	4.2 mi N	2.1 ± 0.64
	3.4 mi S	1.9 ± 0.45
Beatty	2.4 mi N	3.8 ± 0.88
	6.6 mi S	3.0 ± 0.84
Tonopah	1.5 mi S	0.5 ± 0.2
	3.9 mi NW	1.1 ± 0.52
Warm Springs	2.6 mi E	1.3 ± 1.0
	4.5 mi W	3.0 ± 1.1
Moapa	4 mi NW	1.9 ± 0.92
	7 mi NW	0.6 ± 0.4
Diablo	2.4 mi N	7.8 ± 1.8
	2 mi S	8.2 ± 1.3
Goldfield	3.4 mi S	4.3 ± 0.79
	3.8 mi N	2.5 ± 0.58
Butler Ranch	1.9 mi S	1.4 ± 0.89
	2.3 mi N	22 ± 2.4
Caliente	4 mi N	1.1 ± 0.64
	4.2 mi S	0.8 ± 0.3
Indian Springs	1.5 mi E	0.9 ± 0.5
	3.7 mi W	1.5 ± 0.39
Furnace Creek, California	1.3 mi S of Inn	0.4 ± 0.1
	0.6 mi N of Ranch	0.6 ± 0.3
Scotty's Jct.	2.3 mi S	4.0 ± 1.3
	2.1 mi N	1.2 ± 0.39
Clark Station	1 mi W	1.9 ± 0.61
	2 mi E	14 ± 2.6 *

Location (see Figure 1)		Pu-239 (mCi/km^2)
Hiko	3.6 mi N	1.6 ± 0.54
	1.5 mi S	0.8 ± 0.4
Kingman, Arizona	0.6 mi E	0.7 ± 0.4
	1.6 mi W	1.0 ± 0.5
Baker, California	1 mi N of Airport	0
	6 mi N of Airport	0.2 ± 0.2
Death Valley Junction, California	1.4 mi S	4.0 ± 0.63
	2.1 mi N	0.5 ± 0.2
Las Vegas	3 mi W	1.8 ± 0.70
	5 mi SW	0.5 ± 0.2

*This result is under question. Another sample will be analyzed.

specific origins of the plutonium have yet been attempted. The locations sampled in this survey which showed plutonium do coincide with fission product fallout patterns defined for the above mentioned test series.

This preliminary information shows there is detectable plutonium-239 in the areas around the Nevada Test Site and point out four general areas for further study. These areas are Lathrop Wells, Goldfield to Scotty's Junction, Penoyer Valley to Reveille Turnoff, and Butler Ranch. The highest deposition of plutonium-239 is northeast of the Nevada Test Site with the second highest deposition being southwest as defined by this survey. Values range from 130 mCi/km^2 to background. Sampling will now be expanded in these four areas to define distribution patterns as they now exist.

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USE OF AERIAL SURVEYS FOR DETERMINING PLUTONIUM CONCENTRATION

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April 15, 1971

Use of Aerial Surveys for Determining Plutonium Contamination

I. Introduction

When high spatial resolution is not critical, radiation survey rates can be improved by installing the detecting instrument on a fast moving airborne platform. This paper describes a Pu aerial survey using an array of NaI detectors that are set up to sense the 60 keV γ-ray from ^{241}Am, a decay product of ^{241}Pu. The detecting system was mounted inside an Air Force helicopter and flown over known concentrations at the NTS. The detectors used in this experiment are normally carried in a Beechcraft Twin Bonanza, which is maintained by the AEC under the ARMS program and used to survey reactor sites for high energy γ-ray emitters. Therefore, the system has not been optimized for the present application. However, concentrations similar to the criteria used for a clean-up level at Las Palamaries have been detected.

II. Detection System

In order to reduce low energy air-scattered background (from naturally occurring nuclides), the 4" x 4" NaI crystals were wrapped with 1/16" Pb to cover all except the downward face. Directional and energy response of a crystal so prepared is similar to that from a thin crystal. However, the low energy end of the thick crystal spectrum may still contain a larger contribution from Compton scattering of high energy γ's than would be present in the thin crystal spectrum.

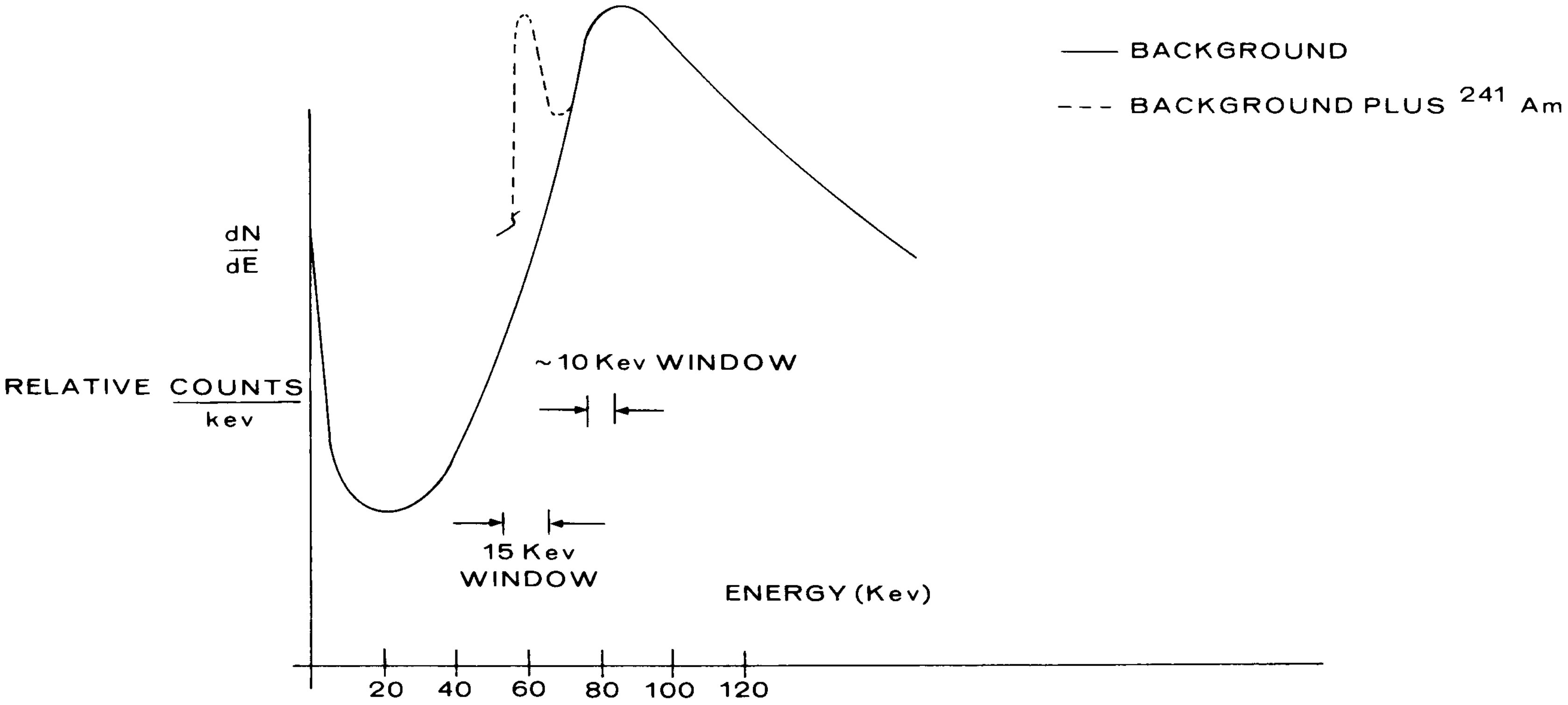
Figure 1 shows a plot of an ^{241}Am spectrum superimposed on a typical background spectrum at 100 feet. Surveys are conducted by monitoring two pulse height windows whose end points are delineated by bars in the figure. The first of these windows is adjusted to count pulses within about ± one standard deviation on each side of the 60 keV peak; the second window, centered on 80 keV, is adjusted so that the accepted count rate matches that from the 60 keV window when no ^{241}Am is present.

Figure 2 shows a block diagram of the electronics and recording system. The output of the array of 14 crystals is summed before being sorted by the two single channel pulse height analyzers. The pulses accepted by the windows are converted to an analog signal, subtracted, and recorded on a strip chart. The subtraction technique partially compensates for variations in natural background levels that can falsely indicate the presence of Pu.

III. Data and Results

This system has been used to survey some known concentrations of Pu at the NTS. Figure 3 shows the difference between the signals from the two windows as recorded by the strip chart during a pass near a safety shot blast-center at Pu Valley. Scales for both count rate and concentration of 5 year old weapon grade Pu are shown. Count rate is converted to

FIG. 1 TYPICAL BACKGROUND SPECTRUM SHOWING ^{241}Am (PHOTOPEAK ONLY)



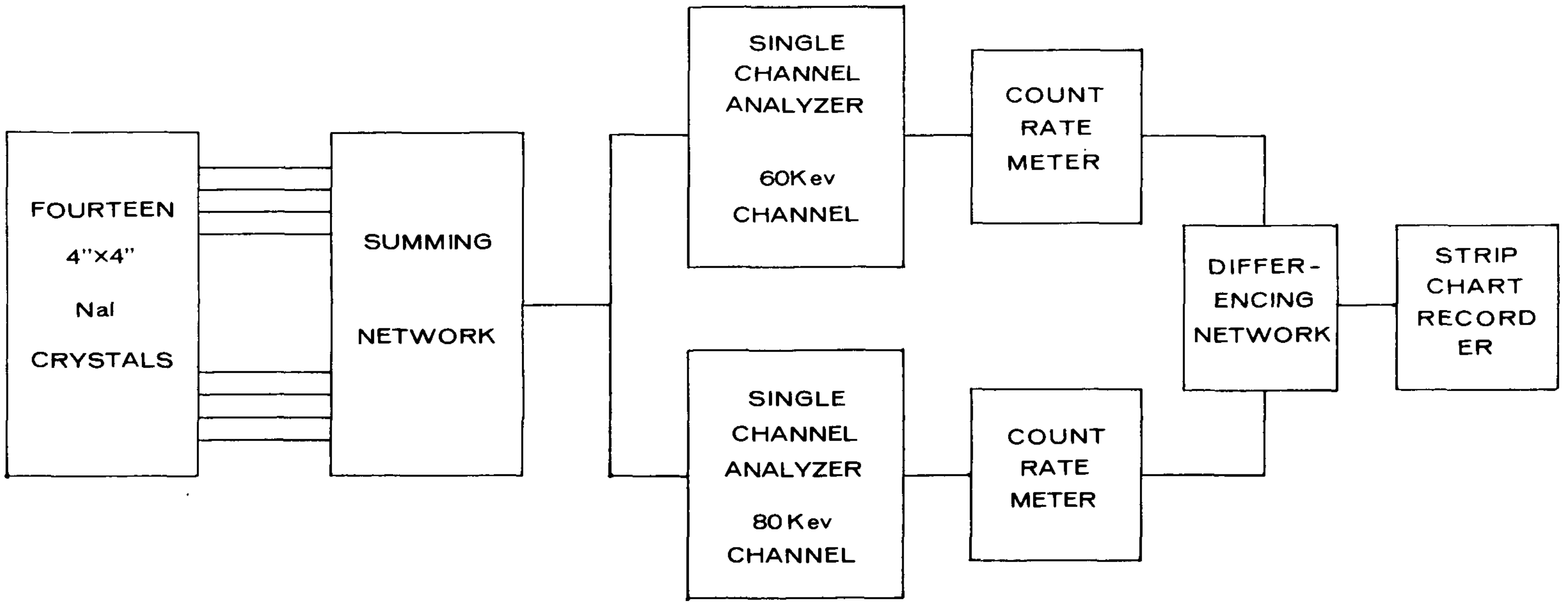


FIG. 2 BLOCK DIAGRAM OF ELECTRONICS

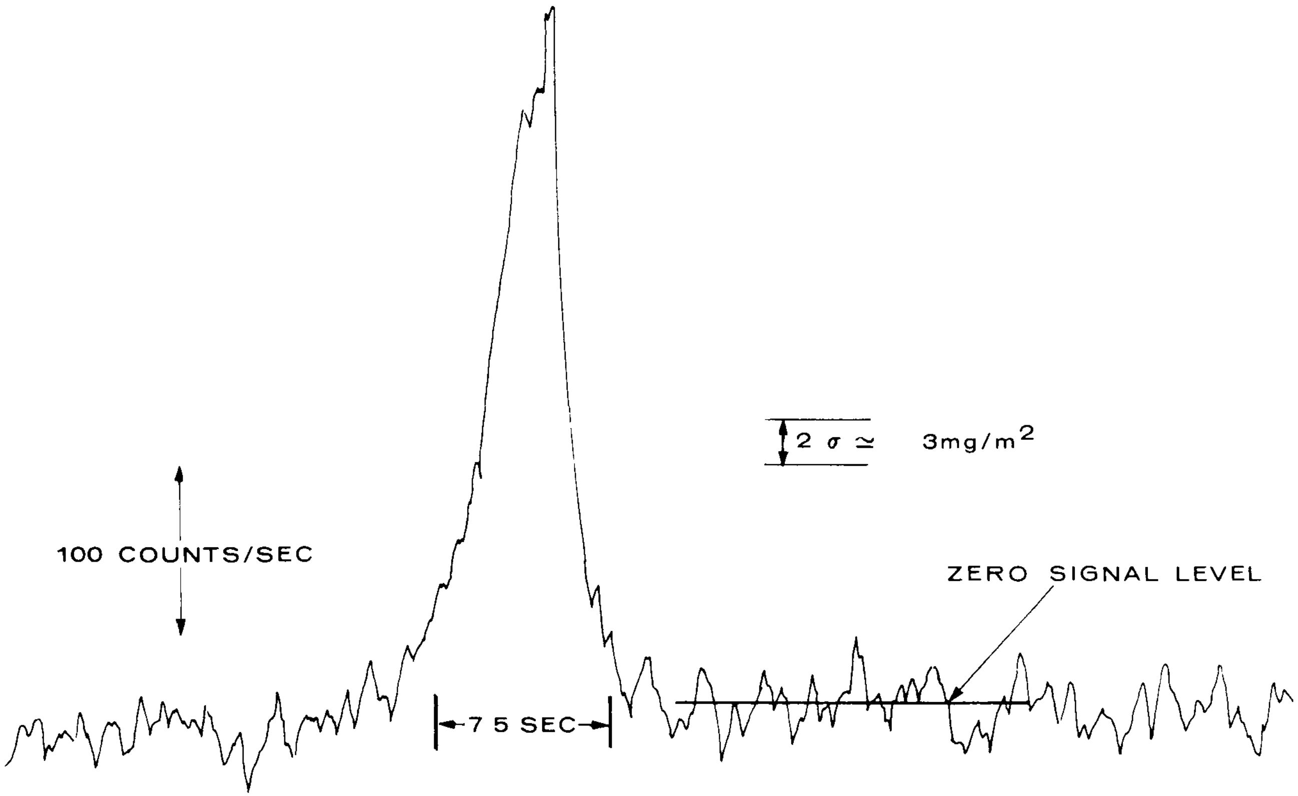


FIG. 3 STRIP CHART SIGNAL

concentration with the following equation:

$$R_d = F \frac{(\rho\lambda_m)}{2} A^{eff} E_3 \left(\frac{h}{\lambda}\right)$$

where R_d = count rate from photons originating from a uniform distribution of Pu in the soil.

ρ = density of mixture of Pu isotopes in soil.

λ_m = mean free path of 60 keV photons in soil.

A^{eff} = end surface area of crystals effective in producing counts in 60 keV window as determined with an ^{241}Am source placed outside the aircraft.

$E_3\left(\frac{h}{\lambda}\right)$ = exponential integral of the third kind (1).

F = rate of origin of 60 keV photons per unit mass of weapons grade Pu.

$F = 0.4 \times 10^{10} \frac{\text{photon}}{\text{K}_g \text{ sec}}$ for ~ 5 year old weapon grade Pu.

h = survey altitude.

λ = mean free path of 60 keV photon in air.

The solid horizontal line shown in Figure 3 represents the true zero level for the differenced signals. Negative signals lasting longer than expected on the basis of statistical errors can be caused by inaccuracies in the background subtraction method. Any phenomenon that causes a variation in spectrum shape can create this zero shift. Variations in altitude and concentrations of other radionuclides may contribute to the problem. Other background subtraction techniques are being investigated.

Deviations from the zero level of + 2 times the standard deviation of the differenced background signal is assumed to represent the minimum readable signal. The standard deviation (2) of the differenced background signal is given by:

$$\sigma = \sqrt{\frac{CR_B}{RC}}$$

where CR_B = background count rate in each window.

RC = count rate meter integration time constant.

The time constant (RC = 2 seconds) is made equal to the survey altitude (100 feet) divided by half the aircraft ground speed (V/2 = 50 feet/second). This selection is believed to represent a reasonable compromise between statistical fluctuations in the background level and spatial resolution of Pu concentration in the ground. The two-second time constant couples with the measured background count rate to give errors corresponding to a minimum readable signal of $\rho\lambda_m = \frac{0.3 \text{ mg}}{\text{m}^2}$.

Spatial resolution (FWHM) perpendicular to the flight direction is equal to about twice the survey altitude (2 h = 200 feet) for $\frac{h}{\lambda} \leq 1.5$. Resolution along the direction of flight is degraded by the integration time constant of the count rate meter and is about 400 feet.

The contour map shown in Figure 4 was constructed from a series of strip chart records taken while flying lines spaced 400 feet apart over Pu Valley. These measurements could be in error by as much as a factor of two. However, levels measured from the air do not disagree significantly with those measured on the ground. Some disagreement is to be expected due to the differences in spatial resolution. Advantages of the aerial survey method over a ground survey method are 1) rate at which large areas can be covered and 2) lack of contamination of survey crew.

It is interesting to note that the neck in the distribution of the level of contamination between 0.3 mg/m^2 and 0.9 mg/m^2 corresponds roughly to a break in a ridge of hills running north and south. This spill-over into the next valley was not evident from ground surveys. Such a low level of contamination is sensitive to accurate determinations of the background level at the point of interest. The aerial technique does make a continuous attempt at background compensation, but this very compensation technique may make the result sensitive to altitude variations or to the presence of hills. Therefore, the shape of this distribution may or may not be real.

Work is continuing in an attempt to devise an optimum method for background subtraction without sacrificing sensitivity. The trade-offs that exist between survey time, spatial resolution and sensitivity are being examined. Some improvement will result when the detectors are mounted outside a twin-engine helicopter, which can safely be flown slower at lower altitudes.

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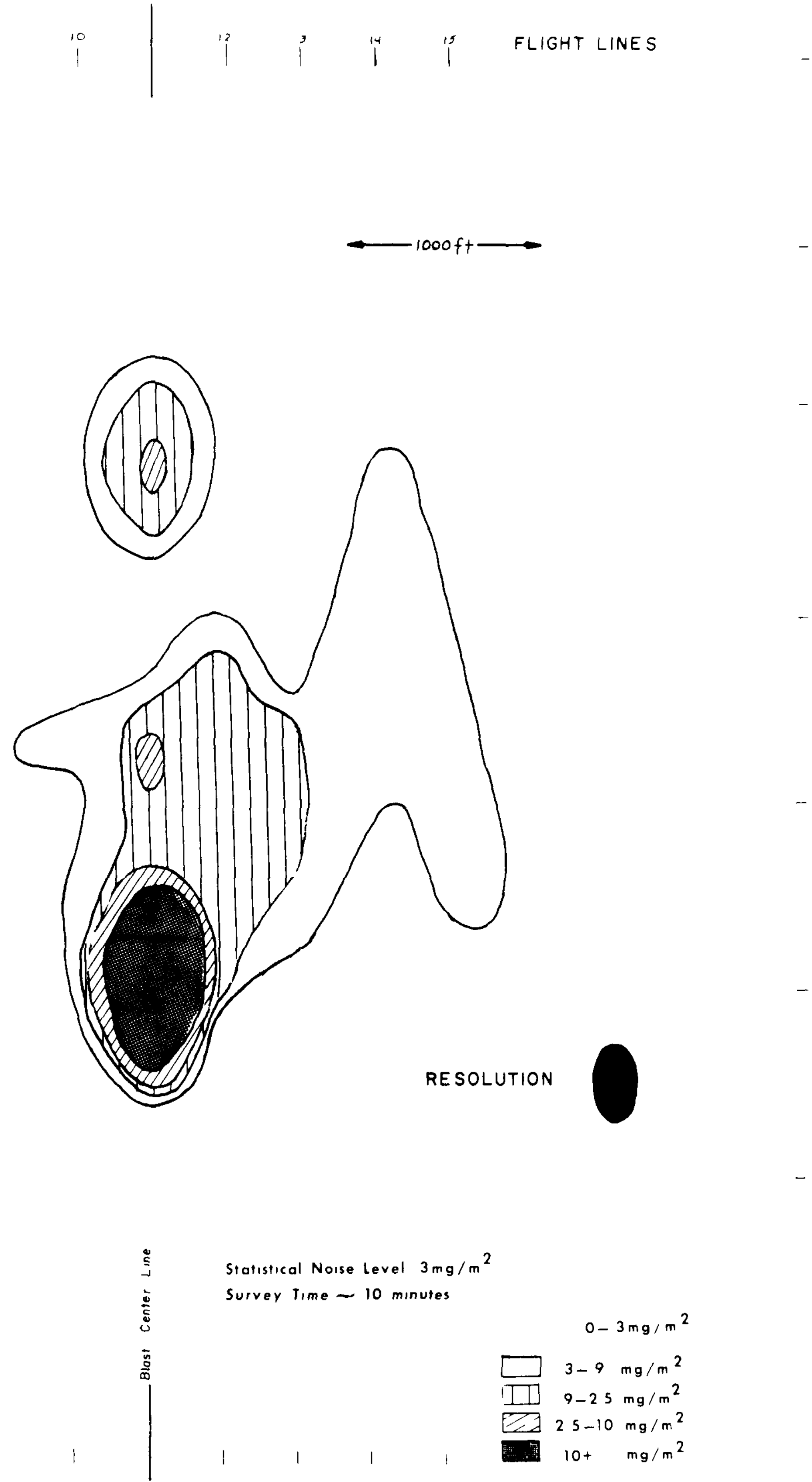

FIG. 4 WEAPON GRADE Pu CONCENTRATION AT Pu VALLEY

ESTIMATING NATURAL TERRESTRIAL BACKGROUND IN THE 17- AND 60-keV ENERGY REGIONS FROM THE COMPTON CONTINUUM PEAK

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April 15, 1971

ESTIMATING NATURAL TERRESTRIAL BACKGROUND IN THE 17- AND 60-keV ENERGY REGIONS FROM THE COMPTON CONTINUUM PEAK*

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ABSTRACT

A 1-yr study of terrestrial natural gamma background in the energy region of 10-100 keV showed that, whereas counting rates varied with geographical location, the percentage of the total activity within any given 10-keV interval was reproducible within about 5%. As a result, the 10-100 keV natural background gamma spectrum can be calculated from a counting rate measurement within this energy range. This technique, using thin NaI(Tl) crystals, is proposed for estimating natural background, which interferes in the detection of low-level plutonium surface contamination. Here detection is based on measurement of either the 17-keV x-ray band or the 60-keV gamma from ^{241}Am, the latter normally present through beta decay of ^{241}Pu. The 80-keV Compton continuum peak is used for calculating background contribution in these two energy regions, since it provides the best counting statistics and is above the plutonium contamination energies. Through the use of a pulse-rate meter having three switch-selected, high-voltage supplies, which permits detection of three separate energies, field tests demonstrate that natural background in the 17- and 60-keV regions can be estimated to within ±10%.

INTRODUCTION

Thin NaI(Tl) detectors are often used in plutonium surface contamination surveys; either the 17-keV x-ray band or the 60-keV gamma from ^{241}Am is measured. The 60-keV gamma is normally present in isotopic plutonium mixtures through beta decay of the ^{241}Pu content. Surveys must be made in the presence of the natural terrestrial background, which varies with the condition of the soil and the geographical location. As a result, plutonium detection sensitivity will vary inversely with this terrestrial background. Background interference poses no problem in locating highly contaminated areas, but peripheral boundary zones between low-level contamination and natural background radiation cannot be established unless we have a means of estimating the contribution of background to the observed counting rates.

We have recently developed a field method for estimating terrestrial background, basing our method on the result of a 1-yr study of low-energy terrestrial radiation in the energy region between 10 and 100 keV.[1]

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*Work performed under the auspices of the U.S. Atomic Energy Commission.

Initially, the study was undertaken as part of our Emergency Planning Program, with the specific purpose of determining natural backgrounds at selected locations surrounding the Livermore Laboratory. The measurements showed that counting rates varied with geographical location, but the percentage of the total activity within any 10-keV interval was reproducible to within about ±5%. Consequently, we can calculate the 10-100 keV natural background gamma spectrum from a counting-rate measurement within that energy range.

This paper describes these measurements and the technique used to estimate the terrestrial background in the field.

EXPERIMENTAL

Measurements were made with our FIDLER detector,[2-5] which is an acronym for Field Instrument for the Detection of Low-Energy Radiation. The FIDLER is a 5-in.-diam by 1/16-in.-thick NaI(Tl) crystal optically coupled by a quartz-light pipe to a selected RCA 8055 photomultiplier tube.[*] The entrance window is 10-mil beryllium, and the entire assembly is housed in a 5/32-in.-thick stainless steel can. The FIDLER is normally used with a battery-powered pulse-rate meter capable of being operated as a single-channel analyzer. By proper adjustment of applied voltage, either 17- or 60-keV energies may be detected. Figure 1 is a photograph of the FIDLER and pulse-rate meter as they are used for field surveys.

In our study we wanted to obtain background counting rates at 10-keV intervals, so we substituted a multichannel analyzer for the portable meter. Figure 2 shows the equipment as it was used for the measurements.

The FIDLER was suspended 12 in. above the ground from an aluminum A-frame. Output pulses from the detector were accumulated on the multichannel pulse-height analyzer for 10-min counting periods. The analyzer, printer, and associated electronics were mounted in a small van with electric power provided by a portable gasoline-motor generator. Eighty feet of cable permitted location of the detector in areas inaccessible to the vehicle. A 10-μCi ^{241}Am source was used before each measurement to calibrate the energy scale of the analyzer and to check the system response.

Measurements were made once each month from October 1969 through September 1970 at the 10 sites shown in Fig. 3. These survey sites were on public rights-of-way and were selected by appearance to represent undisturbed, native soil.

Following the data acquisition, we summed the counting rates in each 10-keV interval to obtain the integral FIDLER response between 10 and 100 keV. Then we calculated the percent of this total in each 10-keV interval.

UNIFORMITY OF SPECTRAL ENERGY DISTRIBUTION

Typical of the results obtained are those that were collected in October 1969 and are presented in Table 1. Although the counting rates varied by almost a factor of 2 depending on location, the percentage of the total activity within a given 10-keV energy interval was markedly reproducible. The

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[*] Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U. S. Atomic Energy Commission to the exclusion of others that may be suitable.

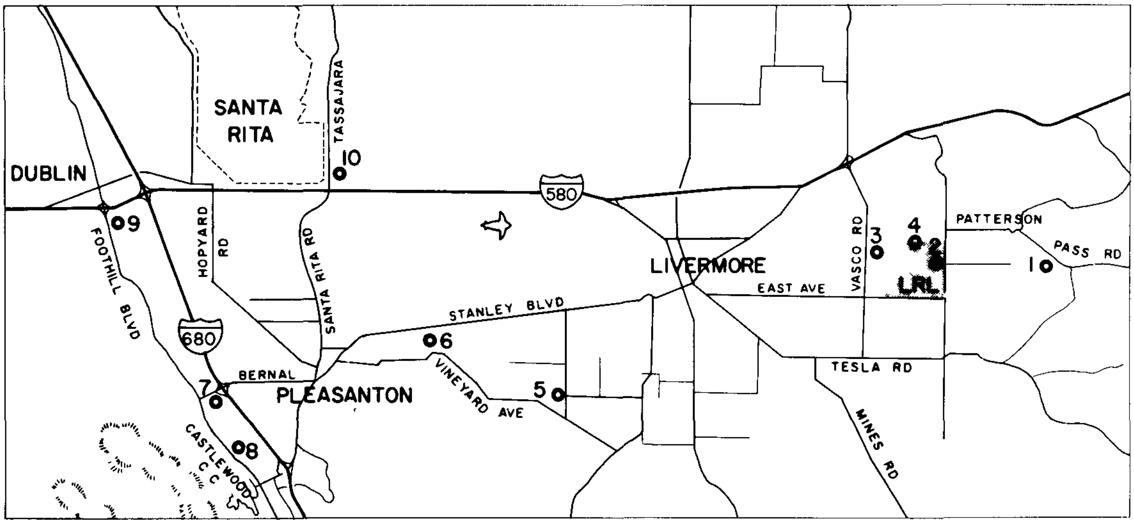

Fig. 3. Locations of natural background radiation measuring sites in the Livermore Valley.

Table 1. October 1969 survey, FIDLER response to environmental radiation in the Livermore Valley.

	Energy Interval (keV)																		
	10-20		20-30		30-40		40-50		50-60		60-70		70-80		80-90		90-100		
Site No.	cpm	% total	cpm	% total	cpm	% total	cpm	% total	cpm	% total	cpm	% total	cpm	% total	cpm	% total	cpm	% total	Total cpm
1	187	4.12	196	4.32	252	5.55	354	7.80	560	12.33	761	16.76	841	18.63	756	16.65	628	13.83	4535
2	216	4.38	223	4.52	268	5.43	374	7.58	575	11.66	800	16.22	920	18.65	842	17.07	715	14.49	4933
3	248	4.16	269	4.48	329	5.52	449	7.53	637	10.68	952	15.97	1124	18.85	1059	17.76	897	15.05	5964
4	228	3.97	267	4.65	322	5.60	456	7.93	727	12.65	958	16.67	1053	18.32	959	16.68	778	13.54	5748
5	184	4.20	203	4.63	251	5.73	350	7.98	522	11.91	730	16.65	798	18.20	734	16.74	612	13.96	4384
6	205	4.02	227	4.45	278	5.45	370	7.26	586	11.49	830	16.28	946	18.56	895	17.56	761	14.93	5093
7	206	4.04	228	4.47	275	5.39	381	7.46	587	11.50	818	16.03	950	18.61	895	17.54	764	14.97	5104
8	211	3.84	244	4.44	298	5.42	418	7.60	657	11.95	895	16.28	1030	18.73	956	17.38	790	14.37	5499
9	249	3.99	276	4.43	363	5.82	485	7.78	760	12.19	1041	16.70	1156	18.55	1030	16.56	871	13.97	6233
10	212	3.82	243	4.38	294	5.30	425	7.66	660	11.89	931	16.77	1030	18.56	958	17.26	797	14.32	5550
Average	214	4.05	238	4.48	293	5.52	406	7.66	627	11.87	872	16.43	985	18.57	909	17.12	761	14.35	5305

February 1970 counting rates were about 20% lower than those observed during summer and early fall. We attribute this decrease to the fact that February was our wettest month, and during that period soil moisture clogged the natural soil capillaries through which radon gas diffuses to the surface. During normal transit, radon daughters are deposited on and near the surface. These measurements illustrate their contribution to the low-energy background.

Table 2 is a 12-month summary in which data for each energy interval represent the average of all 10 stations throughout the year's observation. This table shows that the variation in the percentage of the total activity within all but the lowest energy interval was less than 5%.

Table 2. Twelve-month summary of spectral distribution of natural gamma background.

Energy interval, keV	10-20	20-30	30-40	40-50	50-60	60-70	70-80	80-90	90-100
Percent	4.47 ± 0.34	4.48 ± 0.19	5.50 ± 0.24	7.59 ± 0.34	11.38 ± 0.32	15.57 ± 0.26	18.00 ± 0.41	17.59 ± 0.44	15.46 ± 0.57
Percent deviation from mean	7.60	4.24	4.36	4.48	2.81	1.67	2.83	2.50	3.67

Figure 4 is a plot of the FIDLER counting rate vs. energy, which graphically represents the spectral distribution that we observed. The maximum counting rate at about 80 keV is the characteristic scattering peak of the Compton continuum. The Compton continuum is a result of differential scattering of the gamma rays from uranium, thorium, and potassium homogeneously distributed in the soil. Below 100 keV, the energy distribution within the spectrum does not appear to be materially affected by the relative abundance of these naturally occurring radioisotopes.

The data in Table 2 suggest that the 10-100 keV spectrum can be reproduced from a counting rate measurement anywhere within this energy range.

Fig. 1. FIDLER detector and pulse-rate meter used for plutonium surface-contamination surveys.

Fig. 2. Field assembly for low-energy terrestrial background spectral measurements using the FIDLER detector.

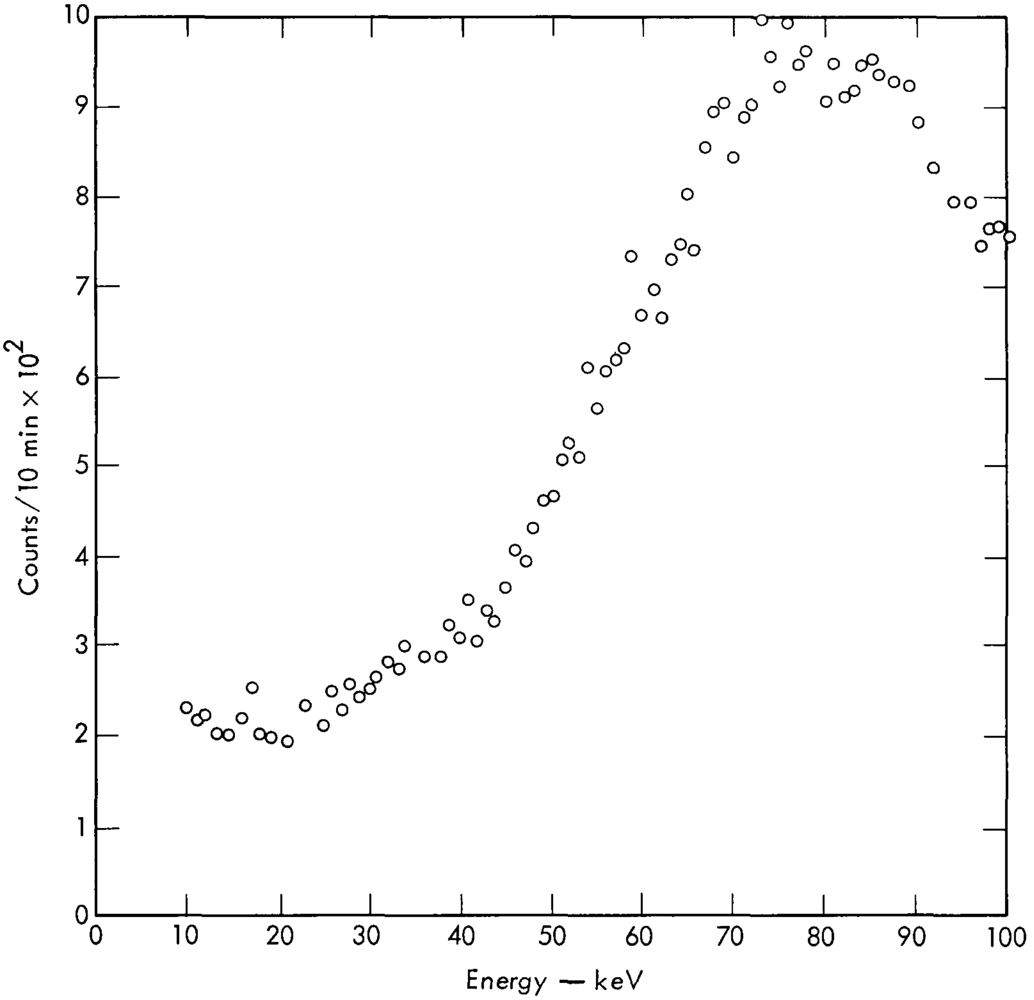

Fig. 4. Response of the FIDLER detector to natural background radiation.

However, a counting rate measurement made in the Compton peak is desirable because it provides the best statistics and is above the plutonium energies of interest. Table 3 shows the accuracy with which counting rates in the 50-70 keV region can be calculated from counting rates made in the 70-80 keV interval. The 50-70 keV interval was selected because this "window width" gives the best $(\text{signal})^2$-to-background value for detecting the 60-keV gammas from ^{241}Am. The calculations are based on the observation that between 10 and 100 keV, the percentage of activity in the 50-70 and 70-80 keV bands are 27 and 18%, respectively. The natural background counting rate in the 50-70 keV interval is, therefore, 27/18 of the observed counting rate in the 70-80 keV interval.

Table 3. Comparison of calculated and observed natural background counting rates.

		Counting rate, cpm		
		In 70-80 keV	In 50-70 keV interval	
Month	Location	interval	Calculated	Observed
October 1969	3	1125	1690	1625
October 1969	5	800	1200	1250
October 1969	9	950	1425	1400
February 1970	3	1050	1575	1615
February 1970	5	600	900	975
February 1970	9	825	1240	1280

To use this approach for field survey work, we have employed a commercial battery-operated pulse rate meter that has three switch-selected, high-voltage supplies. This permits presetting the discriminator window to accept three separate energies detected by the FIDLER. Window width is adjustable from 0 to twice the input sensitivity of the lower discriminator. For our application, we selected a ratio of 3/2. Proper adjustment of the high voltages provides window widths equivalent to 12-18, 48-72, and 72-108 keV. An ^{241}Am source is used for setting the voltages for the two lower energies. While the 48-72 keV interval is near optimum for detecting the 60-keV gamma from ^{241}Am,[6] the 12-18 keV interval is a little too narrow for optimum detection of the 17-keV x-ray band. In calibration, however, the signal from the 17 keV from the ^{241}Am is centered in the window. As a result, the counting response to background and plutonium is reduced proportionately in the narrower window. The voltage setting for detecting the Compton peak is determined by centering the 93-keV gamma emitted from a ^{238}U source in the energy window.

With the instrument thus calibrated, we then read background counting rates at several of the locations within the Livermore Valley. From the 12-month study, we found that counting rates in the 12-18 and 48-72 keV intervals were 6 and 60%, respectively, of the counting rate in the 72-108 keV interval.

Table 4 shows a comparison between the observed and calculated data. If we were monitoring an area suspected of containing low-level plutonium contamination, the natural terrestrial background in the 17- and 60-keV energy regions would be calculated from the counting rate in the Compton peak and subtracted from the observed reading to obtain the net counting rate due to plutonium.

Table 4 demonstrates that we should be able to estimate the terrestrial background to within about ±10%, using the proposed technique. Laboratory

Table 4. Comparison of observed and calculated natural background counting rates using the three-channel pulse-rate meter.

Location	Counts per minute				
	Observed			Calculated from HV-3	
	HV-1, 12-18 keV	HV-2, 48-72 keV	HV-3, 72-108 keV	HV-1	HV-2
1	200	1600	2900	174	1740
2	130	1100	2100	126	1260
3	150	1550	2450	147	1470
4	145	1600	2500	150	1500
5	150	1250	2550	153	1530
6	160	1500	2700	162	1620
7	145	1200	2500	150	1500
8	160	1700	2500	150	1500
9	130	1250	2000	120	1200

calibration[5] shows that for the 17-keV x-ray band, the FIDLER has a plutonium detection sensitivity of about 30 cpm/μg/M^2. To relate the 60-keV americium reading to plutonium requires that we know the americium content of the plutonium. Our method has no effect on the area source detection sensitivity, because this sensitivity is determined by the geometry and intrinsic efficiency of the detector. What it does do is give more confidence to the minimum detectable area source calculation. This term is a function of the background as shown by

$$\text{MDAS} = \frac{3.3}{S_A}\sqrt{\frac{B}{2RC}},$$

where:

MDAS = minimum detectable area source (μg/M^2)

3.3 = number of standard deviations above background counting rate for detectability at 95% confidence level

S_A = area source detection sensitivity (cpm/μg/M^2)

B = background counting rate (cpm)

RC = count-rate-meter time constant (min).

The background is normally not known and is usually established by measurements outside the contaminated area. Establishing the background by this method is frequently more difficult than it might seem. In the first place, we have found natural background differences of nearly a factor of 2 within areas having similar soil appearances. In the case of areas adjacent to these contaminated by plutonium, we cannot be sure that such variations do not represent isolated patches of contamination. Accordingly, when we use a 400-cpm background estimate in the 17-keV band based on a reading outside the area in question, we must recognize that the true value might be as low as 200 cpm or as high as 600 cpm. With the proposed method, we can estimate this background in the same area where we are monitoring, with an accuracy of about ±10% rather than ±50%.

The technique we have described should be useful for more accurately establishing the boundary between contaminated and uncontaminated areas. With the exception of the need for interchanging the pulse-rate meters, no additional equipment is required for making these field estimates.

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ENVIRONMENTAL SAMPLING
(A Critical Look at Common Procedures)

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April 15, 1971

ENVIRONMENTAL SAMPLING
(A critical look at common procedures)

Although no specific reference is made to Plutonium in the title of this paper, the procedures described apply to it as well as to other items of interest found in our environment.

Proper sampling procedures can be described as "objective", ruling out investigator bias. However, environmental sampling is at best "subjective". The reason for this statement is that the item to be sampled, the environment, is indeed a complex entity. Compromise is prevalent and the results therefore are usually less than ideal. During the discussion contained in this paper, the problem of subjectivity, the resultant of compromise, will be discussed in its relationship to the various techniques reviewed.

Samples taken in the environment may be used for various purposes. Background and source of contamination identifications, concentration of contamination and total amount of contamination studies, and hazard "potential" and hazard analysis (including comparisons with existing standards, regulations or guidelines) are some of the major uses of such samples.

Backgrounds must be established before any of the other investigations could be of any definitive value, except where the magnitude of environmental insult would preclude the need for such information. Other studies may be independent of one another and it would matter not if they are in the same compartment of the environment. When comparing or utilizing another investigator's results, extreme care must be exercised, as these results, regardless of analytical accuracy and precision, may have severe qualifying sampling procedure limitations which may restrict their usefulness in your application.

The major compartments of the environment are usually taken to be water, soil and air. And in that order, let us now discuss the problems of subjectivity relative to each.

The need for water sampling can simply be identified because of effluents from areas of interest. Comparisons can be of value. However, even if proper laboratory analysis is provided, individual sample results may be severely biased by the sampling procedures used. Streams containing settleable solids must be sampled so that the result is meaningful. Sampling extremes in water flow, i.e. rapids or ponds, may introduce bias, along with the distance traveled by the stream before sampling. Additionally, the change in water flow pattern within the stream channel may produce "seasonal" variations which are actually the result of sampling technique.

Dissolved materials do not suffer from "mixing" problems as compared with suspended material, except when sampling for effluent effects on the receiving stream. However, if the chemical characteristics of the stream change, precipitation and/or redissolution may present problems. This situation is prevalent at confluences of water courses.

Large impoundments of water present particular problems. It is well known that large bodies of water are stratified and that there are situations in which the body of water will invert (thermal conditions). Another problem associated is the use of powered recreational boats which are used primarily on weekends and holidays. How, when, where and possibly why are the questions to be answered in regard to a sampling program.

In our surveillance of the U.S.A.E.C.'s Rocky Flats Plant, we are involved with Great Western Reservoir used by the City of Broomfield for their municipal raw water storage. Two (2) % of the water received by this impoundment is from the Walnut Creek drainage, which is the receiving stream for the liquid effluents (both sanitary and industrial) of the Rocky Flats Plant. Our present sampling procedure at this reservoir does not involve stratification, inversion identification or mixing by surface craft. Because of the health hazard potential associated, the sampling is done at a raw water line at the water treatment plant. Depending on the level of raw water storage and water quality, the water is taken from various controlled depths at the reservoir's intake tower. Sample collection at the filter plant for both treated and untreated water indicate the level of decontamination effected. In a study, done by the water quality group of the Environmental Protection Agency (EPA), as yet unpublished, a decontamination effect is demonstrated by normal complete water treatment, in this particular situation, corroborating our findings.

So far in my discussion of the topic, subjectivity has been associated only with improper techniques. However, what about some "proper" sampling procedures? Which is preferable, grab or continuous sampling? Grab samples may miss some peaks or valleys in a time or volume-oriented program but the continuous sampler will only provide average concentrations. Of course, the analytical continuous sampler does not so suffer, but the number of different analyses provided is usually limited.

Another question, how many samples and/or sampling stations are sufficient for a proper environmental sampling program? In a day and age when all data is statistically reviewed, the number of observations is an important consideration. In the situation where the level of environmental insult has been established and the findings indicate that a program change on sample procurement would be "logical", at what sample frequency and at what contamination level are step-ups and cutbacks to be initiated? Would reduction of the respective MPC's by orders of magnitude or the FRC Ranges be applicable? So far the program looks pretty subjective.

How about another type of decision. Are total or gross alpha concentrations an adequate indicator of hazard "potential" in a plutonium or uranium situation? Numbers satisfy the statistician, but laboratory capabilities for routine specific analyses may be less than ideal. It has been our experience that total or gross alpha concentrations are good indicators and that the use of this procedure, in our case, enabled us to investigate Plutonium concentrations in water using a very limited number of specific radionuclide analyses. The use of total or gross alpha concentrations on individual samples with specific Plutonium analyses done on elevated alpha concentration samples and composites is indicated as the best procedure, in our circumstance.

Let us change our media of environment and discussion to soil. The definition of soil is questionable in its application. At a meeting of investigators on Plutonium in soil analysis, a discussion pursued the definition by particle or sieve size. Even with this over-simplified definition, disagreement ensued as to which was proper -- 10 or 55 mesh. In some areas of our country, normal top-soil (smallest dimension) could easily pass through the

55-mesh sieve; while in the arid areas, "soil" particles may be lacking as the sand structure is greater than that allowed by a 10-mesh sieve. "Soil" must be defined locally (and hopefully, objectively).

Soil sampling is a very subjective procedure. How should you take a soil sample? Coring, depth distribution, surface or hazard oriented, etc.? Coring is an easy procedure provided you do not wish information on depth deposition or hazard potential. Coring smears the side of the sample and the primary hazard associated with Plutonium in soil is surface and air suspension or entrainment oriented. Of course, coring is limited to areas having a finely divided and workable soil structure.

Spatial depth investigations are tedious procedures. Extreme care must be exercised to prohibit zones of interest cross-contaminations. While this procedure can provide the most information per sample excavated, problems follow. How many samples will it take to adequately identify the information sought? The use of a relatively small number of samples in conjunction with meteorological data to extrapolate zones of contamination and total release to the environment is questionable. Let's face it, the same results, in relative numbers, could be obtained through the use of the meteorological data and geomorphological information for the locale. Additionally, the area immediately surrounding the individual sample locations (excavations), and in high wind velocity and frequency areas, for several meters or further, will not be representative of the environment for an indefinite period of time, which poses problems should resampling be required.

Where will the sample be taken so that the resultant total analysis be representative of the environment? Selection of high and low suspect areas is relatively easy where wind patterns are established. Sampler bias is difficult to rule out.

In our opinion, the soil sampling procedure adopted by the Colorado Department of Health for its soil sampling of the environment around the U.S. A.E.C.'s Rocky Flats Plant provides the most information for hazard "potential" evaluation, in this particular situation. While total release information so derived could be misleading, this number would be more meaningful in a hazard investigation than total release data. Using the top 1/8" of loose undisturbed soil for small sample aliquots, no smearing of zones or denormalizing of the environment is effected. This allows for resampling without bias due to a previous sampling effort. Sample collection is easy and the number collected can be significant and objective. Organization of the surface areas into zones without specific meteorological data removes a bias. Of course, our opinion may be considered subjective; however, with all the various staff inputs to the resultant procedure, the program was relatively objective.

Actual hazard evaluation cannot be done by soil analysis. Soil sampling can best be described as an environment impact procedure involved with a potential hazard. The use of air resuspension factors derived under conditions not directly related may be extremely misleading. Surface soil structure, contamination distribution (spatial and particle size), soil chemistry, surface contours and wind velocity and direction are <u>some</u> of the many variables that must be considered.

Because, in many instances the inhalation of radioactive materials presents the greatest hazard, one of the best methods of sampling for environmental hazard is air sampling. Here again, the question of grab vs. continuous sampling appears. Location of samplers to provide for all situations proves to be a problem and then what about the sufficiency of the number of samples? What all do you analyze for? Should a portion of the sample be saved for future reference?

Long term averages using low volume samples tell nothing about the extremes or extent of duration of that extreme. High volume samplers which collect samples only during a prescribed cycle fall into this same category. High volume samples collected continuously over a relatively short period of time provide the best information; however, maintenance and sample analysis can become problems.

At the present time the Colorado Department of Health has 16 high-volume air sampling stations distributed throughout the Denver metropolitan area for its Rocky Flats Plant off-site surveillance effort. Additionally, 5 stations are located along the eastern security fence at the plant site. Four of the on-site stations are strategically located around the Plutonium contaminated oil drum storage area and run continuously with the filter media exchanged every 48 hours (100 m^3/hr).

All filter pads (8" x 10" fiber glass web) are normally stored for 1 week to allow for the radioactive decay of natural airborne radioactivity. The sample pads are then counted for both total long-lived alpha and total long-lived beta concentrations. Plutonium analysis is done on a monthly composite for each individual on-site station. Quarterly composites are used for selected off-site locations. Individual Plutonium analysis is performed on all samples showing abnormal total long-lived alpha concentrations. Recent results confirm the Department's instrument calibrations and assumptions that total long-lived alpha concentrations do indeed provide an adequate evaluation of potential hazard related to plutonium.

Summary:

In this brief discussion on environmental sampling, the subjectivity of commonly used procedures has been reviewed. Although this review is not as thorough as may be deemed desirable, it does raise questions as to procedures that are subjective but the results used, hopefully, objectively.

The poorest trained personnel are usually delegated the responsibility for sample collection and the comparison is, unfortunately, always with existing standards.

KNOW the limitations of your surveillance systems and the resultant data.

Comprehensive knowledge with positive thinking regarding the use of pertinent data brings subjective environmental sampling into the realm of objectivity.

AN APPROACH TO PLUTONIUM SURFACE CONTAMINATION LEVELS

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AN APPROACH TO PLUTONIUM SURFACE CONTAMINATION LEVELS*

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In the control of radioactive contamination on surfaces, it is necessary to have some type of indication as to when remedial action is necessary. General guides[1] have been available for some time, usually as based on the dose to the skin or resuspension with subsequent inhalation. As a part of a program of reviewing such guides, several mechanisms for radiation exposure to people or intake of the material were investigated to indicate the differences which could occur as a result of the energy and type of radiation, metabolic properties of the isotope, and method of possible intake.[2] As will be seen, many of the mechanisms proposed are speculative and little real data are available to provide quantitative conclusions which can be rigidly applied to all situations. In fact, it soon became apparent that it would be difficult to define and model all possible situations of interest. The following discussion is based upon this work with emphasis on plutonium.

The basis for the derivations was taken to be the possible radiation dose to people or the intake of the isotope, thus eliminating such considerations as interference by cross-contamination with experimental work or analyses. The values finally derived are called "decision levels", since they indicate a situation where additional investigation is required, taking into account the particular situation involved as compared to the assumptions made in deriving the levels. They are not limits, since they do not provide any kind of a breaking point between safety and harm. They could be called guides, but this term has been appropriated in regulations and is rapidly approaching the connotation of a limit. These levels can be used to define the required sensitivity for routine monitoring procedures and to indicate when a situation is generally within control. They are, however, derived on generally conservative grounds so that exceeding them should be a signal for investigation by professional people who are thoroughly

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familiar with the assumptions made and who can apply reasoned judgment to the particular situation to assess possible actions. For this reason, they are not amenable to inclusion as limits in regulations.

GENERAL BASIS

For areas where direct monitoring of the potential exposure is available, we have rejected secondary calculations of the mechanism monitored as inappropriate. For example, we do not consider resuspension as an adequate basis for such limits in work areas where a more direct measure is available from routine air monitoring. Values of the resuspension constant are so variable that it is difficult to generalize to all situations without being subject to serious errors. This is not to say that such measurements may not be valuable in a specific situation where conditions are relatively uniform and an empirical relation can be derived for this situation. Similarily, we do not consider in these levels the external radiation dose from the surface in areas where routine survey measurements are made, since the direct measurements are much more accurate and indicative of possible exposure to people than calculated values from assumed distributions. We do, however, consider such mechanisms in areas where monitoring is not carried out, such as in the case of the transfer to the home.

Radiation dose limitations are taken from the latest report of the NCRP.[3] For consideration of the intake to the body through inhalation, ingestion, or skin absorption, the MPC's as derived by the NCRP[4] and the ICRP[5] have been used to indicate the permissible intake with the constants for uptake and translocation as given by the ICRP. In each case, the MPC listed was revised to conform to the recommended organ doses in the recent NCRP recommendations.

Since the main purpose of the decision levels is to permit assessment of required instrument sensitivity and to provide a basis for further investigation, it was assumed, for purposes of calculation, that the contamination is present for the full time appropriate to the particular level. Thus, for the worker, the appropriate period was taken as the time at work or 40 hours per week and 50 weeks per year, and for the public, 168 hours per week and 52 weeks per year. Similarly, where the MPC's provided the basis, the intake of food and water was as recommended by the NCRP and the ICRP. In many of the cases considered, a more realistic situation is the occasional sporadic contamination incident. While similar levels can be derived for this case, these are not included here because of space limitations.

DIRECT LEVELS

The direct levels are considered to be those where the individual is exposed to radiation or can take the material into the body by reason of its position on his person or clothing. For the alpha emitters, external radiation to the skin from material on the skin or clothing is normally considered to be unimportant because of the short range of the alpha particles and the existence of a protective layer of dead cells on the skin surface. The possibility of an alpha emitter penetrating into this dead layer to a depth where the alpha particles can reach the sensitive layer has been postulated.[6] Calculations based on several assumed distributions of the radiation dose to the sensitive cells have been made, with the conclusion that dose rates of the order of one to several hundred rems per hour could occur if such penetration is significant. It does not seem to be possible to estimate any level based upon such considerations, however, without being unduly pessimistic in some cases and unduly optimistic in others. It is noted that in most cases of plutonium contamination, the material is readily removed by simple washing so that it seems unlikely that any great penetration has occurred. In some cases of gross contamination, extensive decontamination techniques are used, with probable removal of at least a portion of the stratum corneum, so that penetration of the material to a depth where the alpha particle can reach the sensitive cells is, at least, possible. It is suggested that a limit based on skin dose is probably not necessary, but it would be a good practice to maintain records of the extent and location of significant areas of skin contamination on individuals to show the frequency of such gross contamination and to provide an exposure record in the unlikely event that a reaction does occur at this location in later years. The beta emitter in the plutonium family, ^{241}Pu, has only a 0.02 MeV beta particle with low gamma emission so that the situation with regard to skin dose is very similar to that of the alpha emitters.

Possible mechanisms for relatively direct transfer to the respiratory tract are: inhalation while rubbing the nose with a contaminated hand or sleeve; wearing contaminated clothing while working; or changing contaminated clothing (particularly if the garment is pulled off over the head). The data available on such mechanisms are primarily from the wearing of contaminated clothing in industrial situations and do not consider such mechanisms in the public of playing with the family pet or the many other methods of stirring up local dust. Brunskill[7] studied the air concentrations resulting from the changing of coveralls lightly contaminated with plutonium with breathing zone samplers. His results can be interpreted, assuming that the act of changing coveralls requires about ten minutes and the individual breathes at the standard man rate of ten cubic meters in eight hours, as the inhalation of material from about 1 1/4 cm^2 of the coveralls during the act of changing or the inhalation from about 8 cm^2 per hour. Butterworth and Donoghue[8] studied the breathing zone concentrations from coveralls contaminated during normal wear in a uranium processing plant during periods of carrying boxes, walking, and filing a section of metal rod. On the average, the concentrations corresponded to the inhalation of contamination on 0.5 cm^2 per hour for cotton coveralls and about 0.3 cm^2 per hour for 50% terylene-50% cotton, with a strong indication that the fraction released decreased with increasing contamination level. From these data we assumed that the inhalation of the material on one cm^2 of the clothing or skin would be a reasonable basis for the decision level.

Another possibility for intake is direct ingestion from the hands or from items such as foodstuffs, utensils, etc. There are little data upon which to base an estimate of these mechanisms and it is arbitrarily assumed that the loose contamination on one square centimeter of the body or clothing can be ingested per hour. This mechanism would seem to be a function of the portion of the body or clothing contaminated but, in view of the lack of definitive data, an arbitrary value is imposed for all areas. The limit per unit area is then estimated from the MPC's in water with the lower of the two values for "soluble" and "insoluble" materials in the NCRP tables used.

For absorption through the skin, one can consider two bases for expressing the rate; the permeability, and the fraction transferred per minute. Tregear[9] points out that the permeability is primarily applicable when there is a sustained pool of material on one side of the barrier represented by the skin while the fraction passing per unit time is probably more applicable when there is a surface deposit. Studies of plutonium in 0.1 N nitric acid on rat skin[10-14] can be interpreted to indicate a penetration rate of about 5×10^{-5} percent per minute. Higher transmission was noted with higher normality acid, but in these cases the skin was badly damaged. The penetration from a slightly acid solution of tributyl phosphate and carbon tetrachloride was about five times greater with some indication that decontamination efforts could have increased the penetration rate even more. Some Russian work with young pigs[15] indicated that the average rate from a plutonium citrate solution was about 4×10^{-5}% per min over a six day period with an initial rate of about 1×10^{-4}% per min over the first day. Watters and Johnson[16] report that a $PuCl_3$-DMSO (dimethyl sulfoxide) solution applied to the pad of a rat's foot absorbed to such an extent that the circulating blood contained 0.26% of the applied plutonium five minutes after the application. Several cases of human exposure have been reported, but in most cases it is difficult to obtain any kind of a rate of penetration because of decontamination efforts which changes the deposited quantity drastically over the period of exposure. Langham[17] reports data on the palm of the hand which give a rate of less than 7×10^{-6}% per min. From a pragmatic viewpoint, it would be possible to claim that experience has indicated that the penetration of plutonium cannot be important, since people have been contaminated for many decades without coming up with any significant body burden. However, when the sensitivity of the urinalysis method for estimating body burden with long term chronic uptake is considered, it can only be stated that the intake must have been something less than 10 to 20% of the maximum permissible intake rate required to produce a body burden in 50 years. These experiments can give only order of magnitude answers for our problem, since it is known that the penetration of the skin of various animals varies and can be influenced by the solvent or carrier with which the penetrant is associated. Thus, absorption can be affected by the external conditions to which the contaminated skin is exposed with the possible influence of creams, lotions, soaps, etc. all uncertain. For this type of exposure, it is the total quantity of material which is on the skin surface which is of importance and not the quantity per unit area. In order to derive a decision level which can be measured by instruments which basically measure the amount of contamination on a given area, it is necessary to know the total area contaminated as well as the contamination level. The decision level is given in units of µCi on the body. Within the accuracy of the estimate, it is possible to consider an area of 10,000 cm^2 (average man is 1.85 m^2) contaminated, so that dividing the quantity in µCi by 10 will give a decision level in nCi/cm^2. In estimating the quantity which could be maintained continuously on the skin, it is assumed that the isotope taken up by the blood after absorption behaves the same as if it were taken up from the GI Tract. The maximum permissible intake rate to the blood is then calculated from the ICRP values of the MPC's for soluble isotopes in water using the constants for uptake as given by the ICRP and correcting, where necessary, for the changes in maximum permissible dose from the recent NCRP Report. The rate of absorption through the skin was taken as 10^{-5}% per minute for the plutonium isotopes. Note that this rate is considerably lower than has been measured for other materials and should be used only for plutonium.

Values of the decision levels for each of these mechanisms and for workers or public are given in Table I for the isotopes of plutonium.

TABLE I

DIRECT LEVELS

	Worker		Public	
	^{239}Pu*	^{241}Pu	^{239}Pu*	^{241}Pu
Direct inhalation nCi/cm^2	0.003	0.1	$2x10^{-5}$	0.0008
Direct ingestion nCi/cm^2	10	1000	0.2	6
Skin Absorption µCi	0.07	5	$8x10^{-4}$	0.03

* Also ^{238}Pu, ^{240}Pu, ^{242}Pu

TRANSFER OF CONTAMINATION

In addition to exposure to the individual carrying the contamination, it is possible to have transfer of the radioactive material to the person or environs of another individual with resulting exposure to or intake by the second party. The approach is to evaluate several simple situations which are not intended to depict reality, but are, at least partially, responsive to the type of situation which could occur, and which provide some basis for judgment as to the possible radiation doses which could result. The significant parameter is the total quantity of material transferred per unit time and not the amount per unit area. Again, the values will be expressed as μCi/day with the understanding that levels which exceed one tenth of these as nCi/cm^2 should be investigated to see if the material can be transferred and if the area covered is as great as one m^2.

For resuspension in the home, it was assumed that 30% of the material brought into the home per day was transferred to the area and that this material remained in a resuspendable form with a half-life of one week. Here we beg the question of "loose" versus "fixed" contamination on the clothing or body because there is no test which will enable us to define which is in a particular form for the conditions to be encountered. The degree of resuspension depends upon many variables including the chemical and physical form of the contaminant, the place in the home of the ultimate transfer, the degree and rate of possible fixation, the ventilation patterns and the degree of movement in the area. The amount of resuspension under given conditions would seem to be more a function of the amount of material in the area than the quantity per unit area. Because of the differing ventilation rates in the home as compared to most industrial areas where the measurements of resuspension have been made, the appropriate parameter should be the fraction resuspended per unit time rather than the more commonly used ratio of the air concentration to the surface concentration. From examination of data available in the literature,[18-21] we have chosen the values given in Table II as reasonably, but not overly conservative.

TABLE II

RESUSPENSION FACTORS

Vigorous activity in the area. Includes cleaning or children at active play or running	5×10^{-3} hr^{-1}
Active. Normal traffic in the room. Children at normal play	10^{-3} hr^{-1}
Moderate. Low traffic with reading, watching TV and occasional movement	10^{-4} hr^{-1}
Quiet. No movement. Room unoccupied or inhabitants sleeping	10^{-6} hr^{-1}

In two situations involving a medium size house and a single room, the average resuspension under the given assumed conditions were then given as in Table III.

TABLE III

AVERAGE RESUSPENSION

	House	Room
Floor area	1500 ft^2	300 ft^2
Ceiling height	8 ft	8 ft
Ventilation rate	2 changes/hr	2 changes/hr
Condition	Time	
Vigorous	1 hr/da	1 hr/da
Active	5 hr/da	2 hr/da
Moderate	6 hr/da	4 hr/da
Quiet	12 hr/da	5 hr/da
Not occupied	-	12 hr/da
Average Factor	5×10^{-4} hr^{-1}	3×10^{-4} hr^{-1}

Using an assumed rate of air change in the house, and an average resuspension factor of 4×10^{-4} per hour, the limitation on quantity which can be brought in per day is then based upon keeping the average air concentration below the MPC for the public.

Note that these values represent only one situation with conservative parameters chosen for this situation. There are possibilities of other, perhaps more limiting situations, such as movement to a small apartment or an automobile.

The second mechanism considered in possible movement of material from the work area is the transfer of the contamination to the skin and clothing of other people followed by direct inhalation, direct ingestion, or skin absorption. Again, it was assumed that 30% of the material brought in was transferred and remained with a half-life of one week. For purposes of estimating the area, it was assumed that this material was transferred to a floor area of 1500 ft^2 and the contamination on the skin and clothing of the people living there was the same as the floor. Under these conditions, the quantity which can be brought in per day can be obtained from the assumed rate of removal and the values given in Table I.

Limitations on the quantity which can be on the body of individuals leaving work as based on these considerations are given in Table IV.

TABLE IV

LEVELS BASED ON TRANSFER TO HOME

μCi/day by worker

	^{239}Pu*	^{241}Pu
Resuspension	0.01	0.5
Direct Inhalation	0.01	0.6
Direct Ingestion	100	4000
Skin Absorption	0.03	1

* Also ^{238}Pu, ^{240}Pu, ^{242}Pu

OTHER SURFACES

An important question concerns the release of equipment or materials which have been used in atomic energy work for unrestricted use by the public. Many of the basic considerations discussed on the control of movement to the home are also pertinent here; however there are additional variables which must be considered. For example, the quantity of material moved is widely variable with no real basis for even an arbitrary choice of the total surface area. The channel of disposal and possible further transfer to another party is unknown in the general case. The use of the materials is unspecific after disposal. For example, it may end up in a hobby shop where it can be subject to grinding, polishing, welding, etc. The frequency of transfer to a given disposal channel is, in general, unknown. It would be possible to derive decision levels for a given situation where the above factors can be defined by using these factors in a derivation similar to the above. Ideally, one would base controls on the total quantity of radioacive materials on the items disposed of per day or per week through a given channel of disposal, but this requires better controls and management than is now available or, perhaps than can be economically provided. It should be noted that one can make little differentiation between "loose" and "fixed" contamination in such disposal operations unless the use to which the item is to be put is clearly known, since many operations can result in the conversion of the "fixed" to the "loose" material.

For work areas where radioactive materials are handled many techniques are available for the control of contamination and transfer to the worker, of which control of surface contamination is only one. Any limitations on surface contamination in the work place should be tailored to all of these techniques. Thus, in areas of controlled access where protective clothing is worn, there is no reason to control surface contamination to the same level as in clean areas where monitoring is infrequent.

CONCLUSIONS

We have presented a very brief summary of one attempt to obtain surface contamination levels as based on possible radiation doses to people. The models used are crude and require refinement both in detail and in study of the individual factors involved. They do however, indicate the possibility of more refined study of the possible impact of contamination and they open the door to consideration of very valuable studies of an applied health physics nature which will greatly expand understanding of the possible problems of control. In particular, it is emphasised that the numerical values obtained should be used only as guides to the professional in investigating particular cases and in establishing limits of sensitivity for routine monitoring.

The prospect for deriving general contamination limits for use in release of materials and equipment seems discouraging at the moment because of the lack of definition of the many places these materials can go, the uses to which they are put, and the quantities so moved. It is possible, however, that detailed study of these factors, along with the transfer coefficients, could lead to acceptable models for many situations which could lead to the development of acceptable standards.

This discussion is a very brief account of work done in this area. A more detailed review of these concepts and application to other isotopes is given in reference 2, now in preparation.

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OPERATION OF A TRANSURANIC STORAGE AREA AT NRTS

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Introduction

Prior to October 1970, all NRTS and Rocky Flats generated waste was buried in pits and trenches in an 88-acre waste burial ground. It became apparent in the early part of 1970 that the existing usable area inside the present burial grounds was rapidly becoming exhausted. This was due mostly to the influx of fire waste from the Dow Chemical's Rocky Flats Plant. The first six months of calendar year 1970, Rocky Flats shipped to the NRTS burial grounds over 225,000 cubic feet of plutonium contaminated waste. A study by Idaho Nuclear Corporation Construction Engineering Division indicated the present burial grounds would be exhausted by 1972 or 1973 if the present rate of waste burial continued.

Recent critical reports by the Federal Water Pollution Control Administration, the National Academy of Science, and various state agencies have expressed concern for the present burial practices used at the NRTS. These reports have voiced objections to many practices but have offered very few, if any, specific solutions. The major objection was the burial of plutonium contaminated waste over the Snake River Plain aquifer.

Because of the likelihood of all transuranics eventually being stored in the Lyons, Kansas salt mines, a joint effort by AEC-ID and Idaho Nuclear Corporation has been made to change and upgrade NRTS transuranic storage methods. Using certain criteria, mainly short-term storage, retrievability, and economics, the Improved Temporary Storage Area No. 1 was constructed adjacent to the NRTS burial grounds for the temporary storage of transuranics. In utilizing this above-grade storage area outside the boundaries of the present burial grounds for transuranics, the life expectancy of the burial grounds has been increased by several years. This report describes the methods used in the operation of the Improved Temporary Storage Area No. 1 (ITSA No. 1) for transuranic waste.

Description of Facility

The Improved Temporary Storage Area No. 1 is a paved pad 150 feet by 400 feet surrounded on three sides by a 12-foot high soil berm. The pad is located on the side of a small hill so as to provide good moisture drainage off the storage area. The pad was designed to drain all moisture to the center of the pad and then to the bottom end where there is a liquid sampling station. ITSA No. 1 is contiguous to the present burial grounds and is in an area that could be easily expanded if necessary. ITSA No. 1 is used exclusively for the temporary storage of transuranics of which better than 99% is received from the Dow Chemical, Rocky Flats, Colorado, Plant. At present, all transuranic wastes are stored in either plywood boxes or metal barrels. Any package readily retrievable as a contamination-free package, within an interim period of 20 years, is defined as a 20-year integrity container. However, the method of storage will depend on whether a container will fit this criteria. The method and operation of ITSA No. 1 will accomplish easy retrievability and will keep waste containers relatively moisture free.

Operation

Dow Chemical's plutonium contaminated waste is received by rail in either piggy-back carriers or ATMX cars. The rail cars are received into the Central Facilities Area where the waste is then trucked approximately 7 miles to the storage area. Waste contained in barrels is then removed from the piggy-backs by means of a fork lift fitted with a special platform. Boxed waste is removed by a means of a crane with a box-lifting device.

The wooden boxes are stacked around the outside perimeter of the paved pad to a height of approximately 12 feet in an interlocking arrangement. The storage area is then divided down the center of the 150-foot dimension into two identical storage areas by two separate rows of the Rocky Flats' wooden boxes. These boxes are set 18-24 inches apart at the base, and stacked to a height of approximately 12 feet. The separation of the boxes is made at the center line of the pad where the water can run off between the boxes. The center row of boxes and barrels are stacked to a height of approximately 18 feet in such a manner as to round off the top of the stack to prevent moisture runoff after the final grading.

The storage area was divided into two identical storage areas so as to permit any barrel weighing over 500 pounds to be placed in the bottom four rows. Barrels weighing less than 500 pounds can then be stacked on top of the heavy barrels. This practice is done in order to maintain the integrity of the barrels. Storage can then be accomplished simultaneously in both areas to allow expeditious handling of extra heavy barrels without resorting to double handling.

Out of necessity, containers and packages must be limited to size, weight, and radiation. Barrels or barrel-type containers must be limited to 1,000 pounds. Packages less than 10 tons with two dimensions less than 10 feet and third dimensions less than 20 feet can routinely be handled. Odd shaped packages would require special lifting equipment. For personnel protection, the radiation emitting from the containers is limited to 200 mr/hr at the surface of the container. Packages that exceed any of the limitations require advance approval by the burial ground supervisor. All nonradioactive scraps, such as metal bands, trailer liners, cardboard, blotting paper, etc., are removed from the pad and taken to a nonradioactive waste disposal area after being surveyed by Health Physics. Wastes are not stored on any torn or cracked pavement that will allow plutonium leaching or leaking through the pavement anomalies into the soil below the pad.

When it is feasible to cover the stored waste, it is first covered with 5/8 inches CC exterior grade plywood and then covered with at least a 4-mil polyethylene sheet. The plastic extends all the way to the ground on both sides of the pad with 3 feet of overlap between successive rows. The final covering is 18 inches of soil utilizing bulldozers and earth movers. After the waste is covered, it is then planted with a protective vegetation. Before and after seeding, temporary measures are taken to prevent soil erosion by covering the soil with a straw mulch or with burlap.

The location of burials is made by means of grid coordinates painted on the paved pad. Monuments, which are made out of galvanized sign post material 8 feet long, are attached to the wooden boxes which circumscribe the storage area and are placed in such a way that a minimum of 3 feet of the marker protrudes above the final grade. The monuments are placed at the four corners of

ITSA No. 1 and at the ends of each grid coordinate line. Brass markers are installed on the top six inches of the monuments and identify the area as the ITSA No. 1 plus the grid coordinate. The four corner monuments will also designate the date the pad was opened and the date the pad was closed. This system of grid coordinates will, in the future, enable the relocation of buried waste.

Since the pad is constructed of asphalt pavement, cleated vehicles are not allowed on the area except in emergency situations. Any pavement anomalies on the surface are repaired as soon as possible.

Sampling

Dirt and air samples are taken routinely by the burial ground health physicist. Water samples are taken whenever water runoff accumulates in the liquid sampler. When water samples indicate activity, a search is immediately made at the face of the waste stack to determine if any container is leaking. If no leaky container can be found, then steps are taken to preclude moisture from the face of the stack, for example, covering the face with polyethylene plastic.

Records and Reports

Whenever transuranic waste is to be stored on the pad, six copies of a form, ID-125, accompany each waste shipment. One of the most important aspects of these forms is the disposal data, such as where on the pad this particular shipment is stored. The burial ground supervisor initiates or receives copies of all work requests associated with the operation of the Improved Temporary Storage Area No. 1.

At the beginning of each month the General Accounting Section of the Controllers Branch of Idaho Nuclear Corporation furnishes the burial ground supervisor with an itemized statement of ITSA No. 1 charges for the preceding month.

The charges for off-site storage are determined by the Idaho Nuclear Corporation General Accounting Section of the Controllers Branch. These data are included in a semi-annual report to the AEC-ID Nuclear Technology Division. The report indicates the costs for labor, burden, materials, and equipment usage. It also indicates cost per cubic foot for storing transuranics on the pad and the total volume of waste stored in the preceding six-month period.

PROCESSING OF PLUTONIUM-CONTAMINATED LIQUID WASTES AT LOS ALAMOS

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1. INTRODUCTION

The Los Alamos Scientific Laboratory (LASL) is located in a relatively remote area in the Jemez Mountains in north central New Mexico. Population of the county of Los Alamos is about 16,000 and the nearest town, Espanola, with a population of 5,000, is about 20 miles distant. Rainfall in the area is about 16 inches per year and runoff drains to the Rio Grande which is at least five miles from the main laboratory site. Groundwater which supplies potable water to Los Alamos is obtained from strata over 1,000 feet below the elevation of the Community Center.

A large part of the laboratory effort is devoted to nuclear weapons research and development, but an increasing amount of the work load is associated with peaceful uses of atomic energy. Principal projects in the latter group include controlled fusion, isotope production, the meson physics accelerator, uses of lasers and the space program.

Liquid wastes of the laboratory are categorized as sanitary, storm and industrial and separate systems have been established to manage each type. This paper will discuss a number of aspects of the control of those liquid industrial wastes which are contaminated by plutonium. References (1) and (2) provide some information on this subject and more complete data on the overall radioactive waste management program.

2. WASTE SOURCES AND ACTIVITY LEVELS

Plutonium-contaminated liquid wastes are processed at two widely separated facilities. One of these, at Technical Area 50 (TA-50), receives wastes from several technical areas on the south side of Los Alamos Canyon. The wastes are effluents from radiochemistry laboratories, general chemistry laboratories, accelerator facilities, research reactors, shops and plating rooms. Gross alpha activity level of the waste is in the range of 1×10^{-5} to 1×10^{-3} µCi/ml. Almost all of the alpha activity is plutonium with 80 to 90% being the 238 isotope. The high concentration of ^{238}Pu is a development of recent origin.

The wastes arrive at the treatment facility through a special sewer system installed for the purpose. Where necessary, the wastes are neutralized at the source to permit use of standard materials of construction, cast iron for force mains and clay tile for gravity lines. A schematic of the system is shown in Figure 1.

The other waste management plant(3,4) is located at a technical area, TA-21, where research and development programs involving plutonium are a major part of the work. Plutonium fuel materials are synthesized and their properties are studied; plutonium metal, alloys and compounds are produced and fabricated; plutonium-239 is recovered and recycled; and refined plutonium-238 is prepared and evaluated for its use as a power source.(5)

Radioactivity levels and types of waste solutions at the TA-21 plant necessitate use of a variety of management methods. The main waste stream, largely cleanup and decontamination solutions, is delivered to the treatment plant via a separate sewer line of cast-iron construction. Plutonium activity level is usually less than 1×10^{-3} µCi/ml with the 239 isotope representing about 80% of the total.

Other waste streams at TA-21 are delivered through stainless steel lines, in stainless steel tank trailers or in smaller plastic or metal containers. The wastes are usually of a much higher level of plutonium activity than the sewered wastes and include solutions of a chemical nature not compatible with the chemical treatment methods employed for the main waste stream, e.g. alcohol and vacuum pump oil. The waste delivered in the tank trailers is a highly acid solution containing milligram per liter quantities of americium-241 and plutonium-239.

Approximate volumes of the wastes processed during the years 1966 through 1970 are shown in Table I.

TABLE I

QUANTITIES OF PU-CONTAMINATED LIQUID WASTES, LOS ALAMOS

Year	TA-50, Liters of Waste	TA-21 Liters of Waste			
		Main Stream	Am-Pu	Alcohol	Oil
1966	53,250,000	10,182,000	84,500	424	19
1967	59,678,000	13,292,000	87,070	186	-
1968	60,286,000	11,591,000	65,970	212	6
1969	54,480,000	13,292,000	71,170	424	23
1970	54,796,000	10,889,000	71,600	466	536

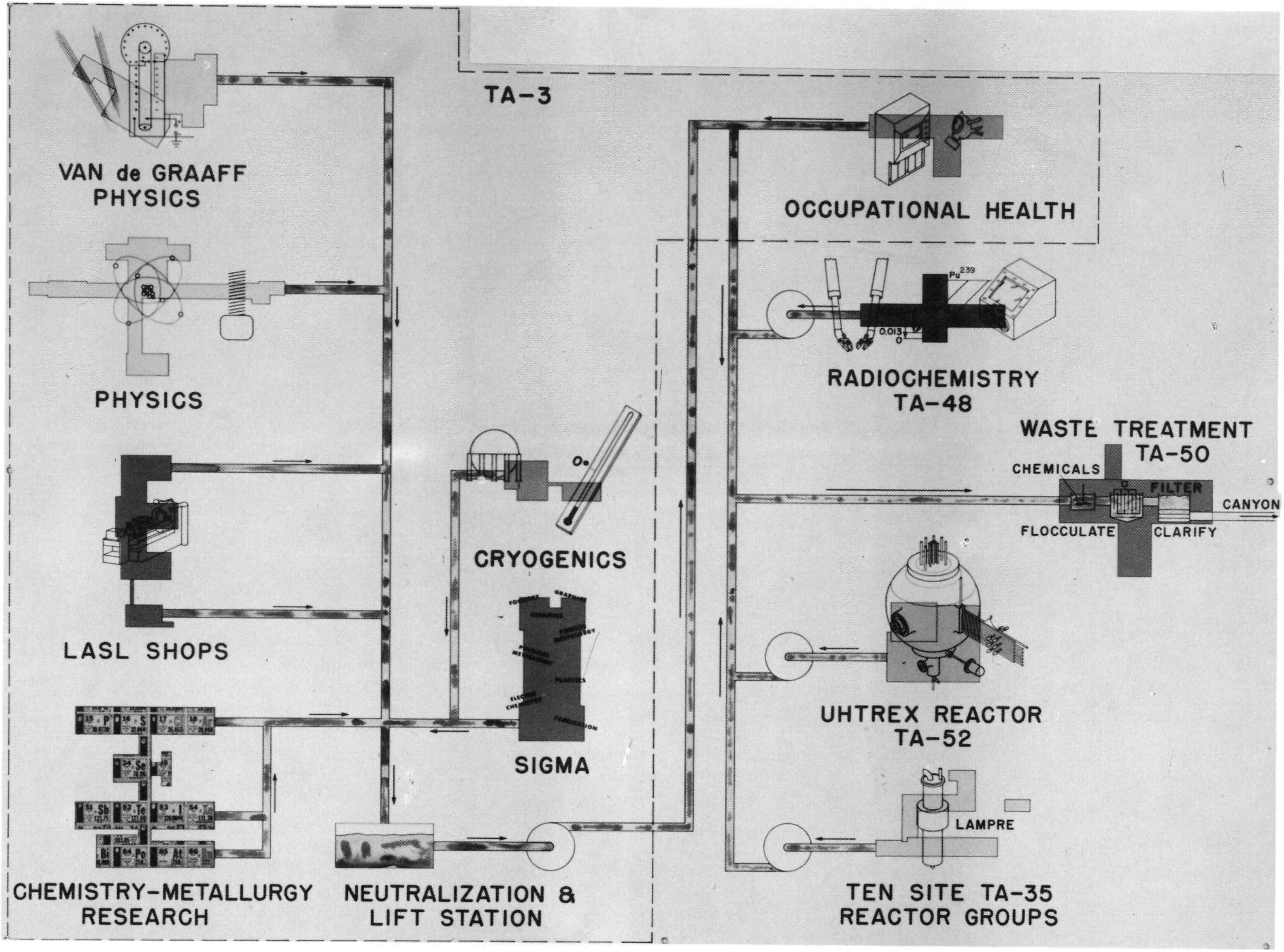

Figure 1. Facilities Served by the TA-50 Waste Management Plant

3. WASTE MANATEMENT METHODS

3.1 TA-50 Facility

The plutonium-contaminated liquid wastes received at this plant through the special sewer system are collected in concrete storage tanks with a total capacity of 387,500 liters. The tanks are mixer-equipped to prevent accumulation of solids; their capacity is adequate to permit 8 hr/day, 5 day/wk operation.

The chemical treatment portion of the plant very efficiently concentrates the alpha active material in a ferric hydroxide sludge. The adsorption-absorption mechanism is most effective at a pH $\geq$ 11 and coagulant aids assist floc growth and sedimentation characteristics. Hydrated lime and ferric sulphate are the principal chemicals added. Details of the 950 l/min operation have been reported in (1) and (2).

The ferric hydroxide sludge is accumulated in a storage tank where it can be settled and decanted to a solids concentration of about 5%; it is then dewatered by vacuum filtration to a solids concentration of 35-40%. This material has the appearance of a damp adobe clay. The vacuum filter is precoated with diatomaceous earth which effectively restricts the passage of particulate activity. Filtrate is recycled to raw waste storage.

Table II provides data on sludge volumes produced and alpha activity contained therein over the last several years.

TABLE II

TREATMENT PLANT OPERATION, TA-50

	Av. α Activity μCi/ml		Vol. of Dewatered Sludge,	Sludge Activity Equiv. ^{239}Pu*	
Year	Raw Waste	Treated Waste	Liters	Ci	g
1966	1.01×10^{-5}	7.4×10^{-8}	61,800	0.33	5.32
1967	1.95×10^{-5}	1.3×10^{-7}	92,300	0.86	13.95
1968	2.41×10^{-5}	8.5×10^{-8}	124,000	1.29	20.96
1969	2.90×10^{-5}	2.5×10^{-7}	175,000	1.52	24.57
1970	4.08×10^{-5}	1.6×10^{-7}	141,400	2.05	33.15

*Equiv. ^{239}Pu is defined as the amount of ^{239}Pu equivalent to the gross α activity of the waste.

Dewatered sludge from the vacuum filter is collected in 200 liter steel drums. Filled, sealed drums are monitored for external contamination and transported to controlled burial areas within Los Alamos County.

Figure 2 is a schematic illustration of the TA-50 plant.

3.2 TA-21 Facility

3.2.1 Main Stream Wastes

Operation of the treatment plant at TA-21 is very similar to that of the TA-50 facility but the TA-21 plant has a capacity for processing wastes at 475 l/min; it is shown schematically in Fig. 3. This plant was placed in operation in late 1967. Just prior to completion, the vacuum filter arrangement for dewatering the ferric hydroxide sludge was dropped from the project. Plans were changed to permit mixing the sludge with other types of wastes and cement in a pug mill operation which is described in the next section.

Table III again illustrates the high efficiency of ferric hydroxide precipitation for the removal of alpha activity.

TABLE III

TREATMENT PLANT OPERATION, DP-257

	Influent Waste Activity		Av. α Activity	
Year	Ci	^{239}Pu, g	Influent μCi/ml	Effluent μCi/ml
1966	0.67	11	6.54×10^{-5}	9.28×10^{-8}
1967	3.71	60	2.79×10^{-4}	2.27×10^{-7}
1968	4.06	66	3.50×10^{-4}	4.42×10^{-7}
1969	3.29	53	2.48×10^{-4}	2.34×10^{-7}
1970	1.08	18	9.92×10^{-5}	1.37×10^{-7}

3.2.2 Americium-Plutonium Solutions

Highly acidic wastes containing mg/l of americium-241 and plutonium-239 have been received in 1000 liter tank trailer lots since 1959.(4) Until installation of the pug mill system at the new plant, these wastes were neutralized with sodium hydroxide; 75 liter amounts of the neutralized solution were transferred by vacuum to 200 liter steel drums containing cement and vermiculite. The drums were tumbled, permitted to set and sent to burial. A major disadvantage of the system was that only 50% of the drum capacity was used.

In the pug mill system which was put into operation in May of 1968, neutralized americium wastes plus other wastes such as plant sludge and neutralized batch solutions are pumped continuously from the holding tank into the pug mill. One of the batch solutions, a strip waste, is the raffinate from a 7% TBP (tributyl phosphate) extraction of non-irradiated enriched uranium

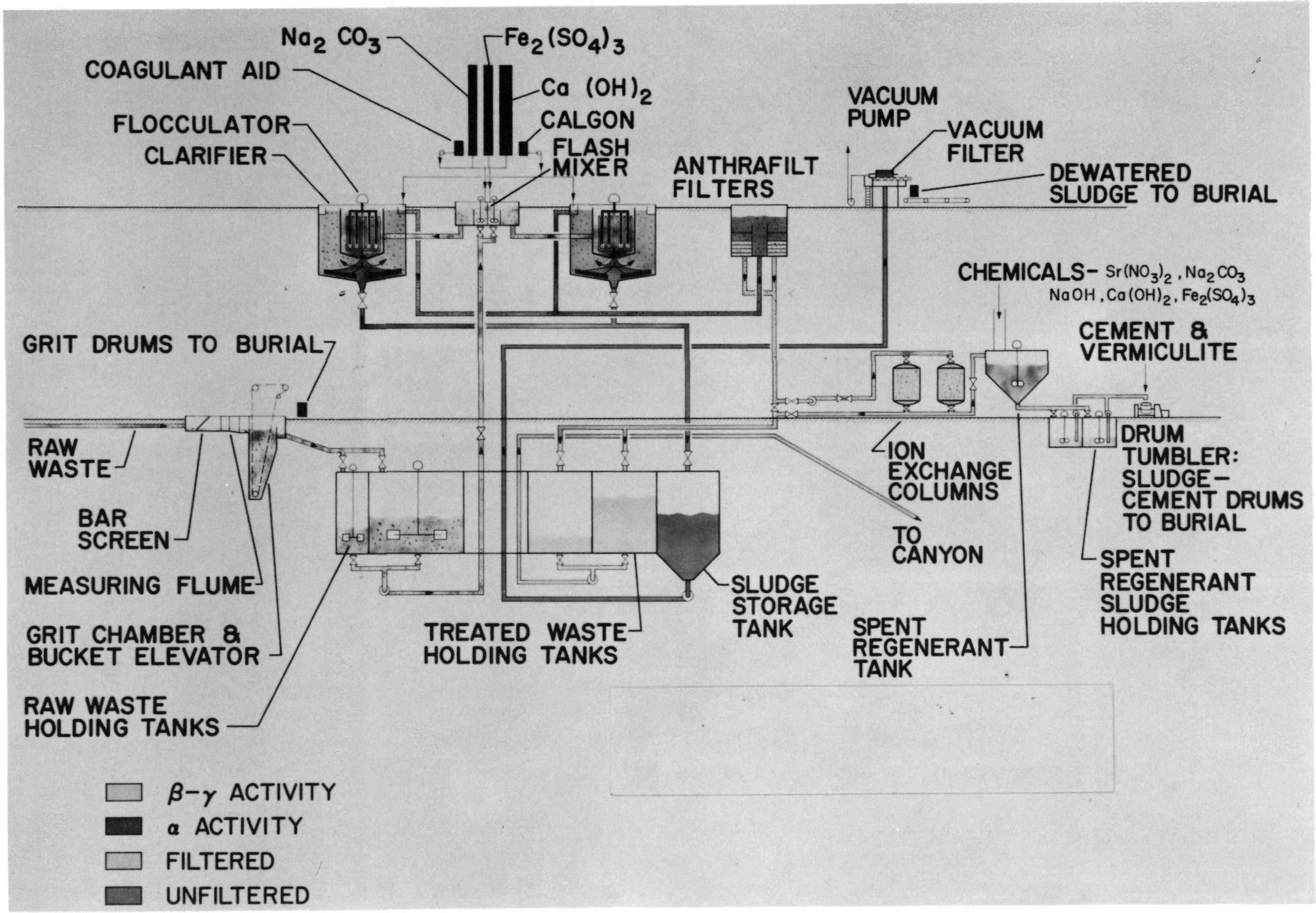

Figure 2. Schematic Diagram of the Waste Treatment Process, TA-50

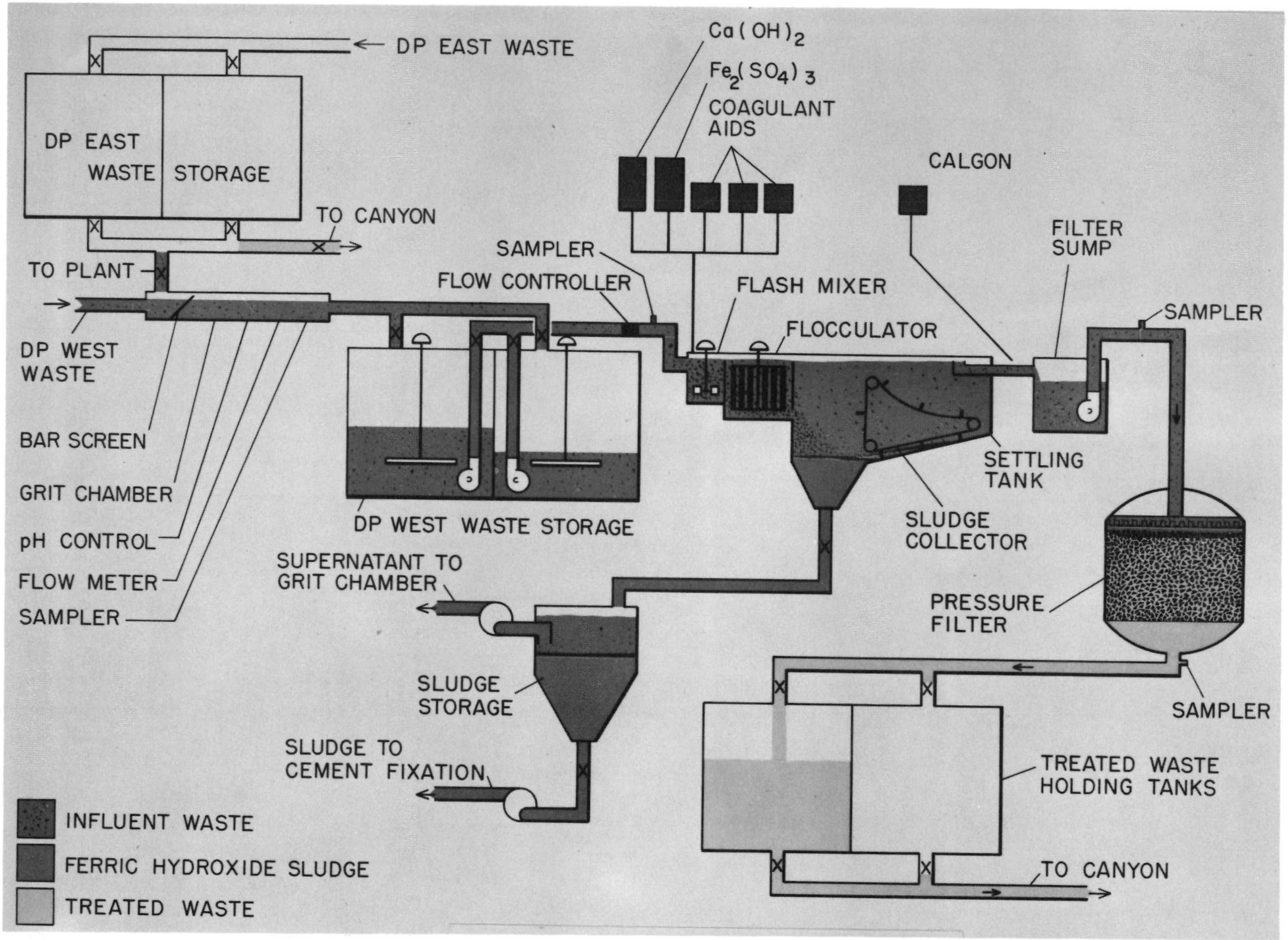

Figure 3. Schematic Diagram of the Waste Treatment Process, TA-21

(93% ^{235}U) solution. Another of the batch wastes is a potassium hydroxide solution resulting from neutralization of off-gases from HF scrubbers in a crucible dissolving operation.

Cement is added through a screw feeder from a silo and the paste is thoroughly mixed before discharging to a pump. The paste is pumped to 2.4 meter diameter pits which vary in depth from five to twenty meters. The system is shown schematically in Fig. 4, a neutralization tank is shown in Fig. 5 and the pug mill is shown in Fig. 6. The pits are in a tuff formation which has a capability for retention of plutonium,(6) but thin gravel strata in several of the borings have led to a practice of spraying the interior surfaces of the pits with asphalt prior to filling with paste to prevent translocation of the waste. Figure 7 shows one of the pits and Table IV provides some operating data from recent runs.

Samples are collected during operation for determination of leachability and compressive strength. The leach samples are collected in 75 ml glass bottles and the compressive strength samples are collected in standard 15 cm diameter by 30 cm high fibreboard tubes used in the construction industry. The leach samples are permitted to set, then removed from their containers and placed in water. The water is changed on a regular basis and the cumulative activity leached from the sample is determined. Excellent retention of alpha radioactivity by the cement paste is indicated by records which show less than 1% of total alpha activity leached from samples after ten years.

Table IV shows the wide variation in compressive strength of the cement paste samples. After collection, they are air cured for 48 hours, then cured under water for 28 days. They are broken in a standard testing apparatus after coating the ends to provide parallel flat surfaces.

The strength of the material is dependent on the water-cement ratio and the kinds of salts in the waste. The operator can produce a strong, dense paste by controlling the proportions of the various types of wastes in the feed solution; for example, an excessive amount of plant sludge in the feed will result in a paste which is difficult to pump and which will have a low compressive strength.

The relationship between leachability and compressive strength is not perfectly clear. Leach tests have been underway for about ten years, but the compression testing program is only a few years old. A preliminary correlation suggests, as suspected, that the stronger pastes are also the more leach-resistant.

Through February 1971, sixteen pits with a total volume of 960 cubic meters have been filled. The pits are filled to within one-half meter of the ground surface, then capped with concrete.

Due to the recent requirement of retrievability of wastes containing transuranium elements, plans are being drawn to pump the cement paste to 200 liter steel drums which can be stored safely for extended periods.

The ^{241}Am waste represents over 90% (radioactivity basis) of the total transuranic wastes produced at Los Alamos. Plans are

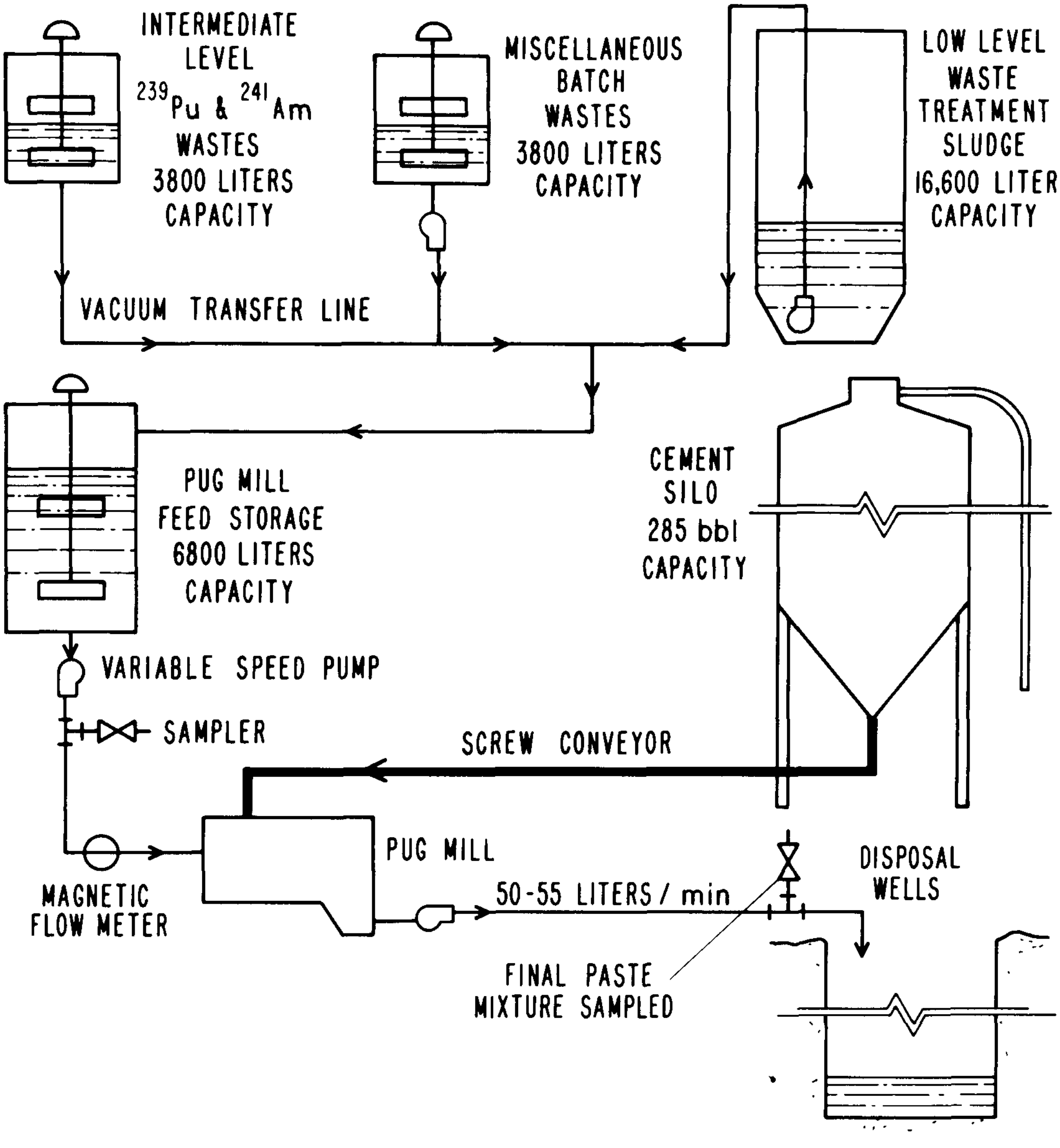

Figure 4. Schematic Diagram of the Pug Mill System, TA-21

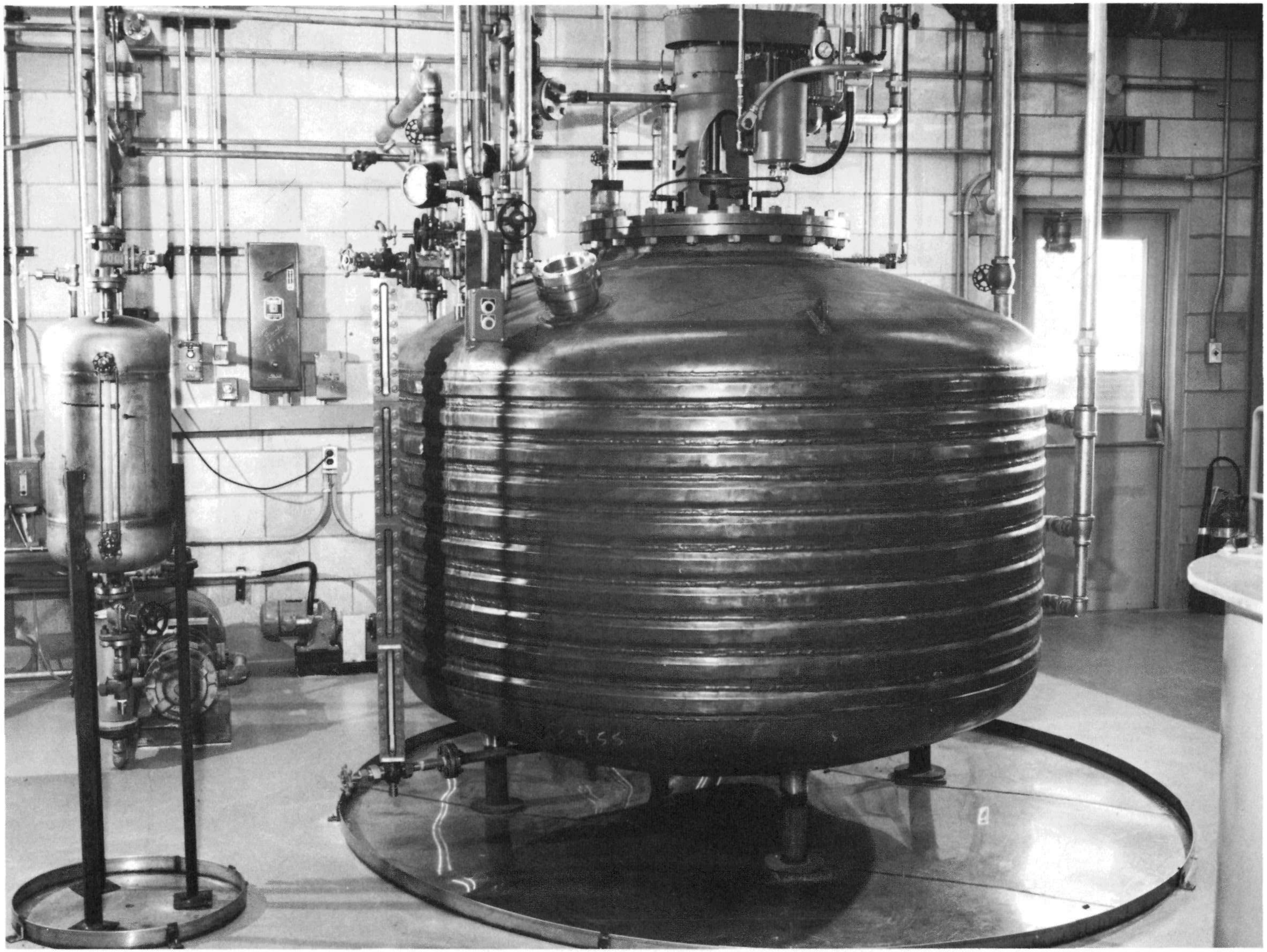

Figure 5. Water-Cooled Waste Neutralization Tank, TA-21

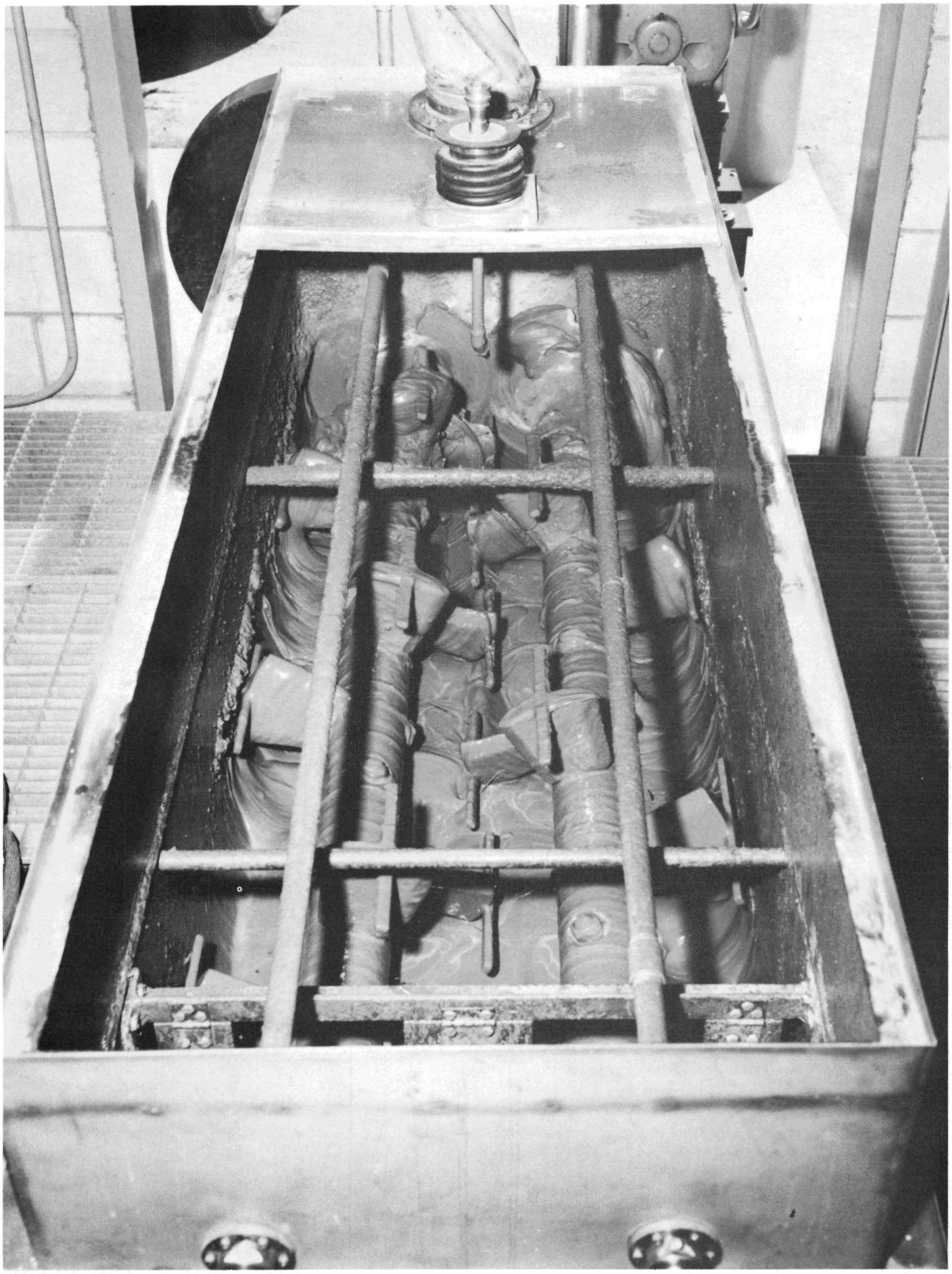

Figure 6. Interior View of the Pug Mill During Operation

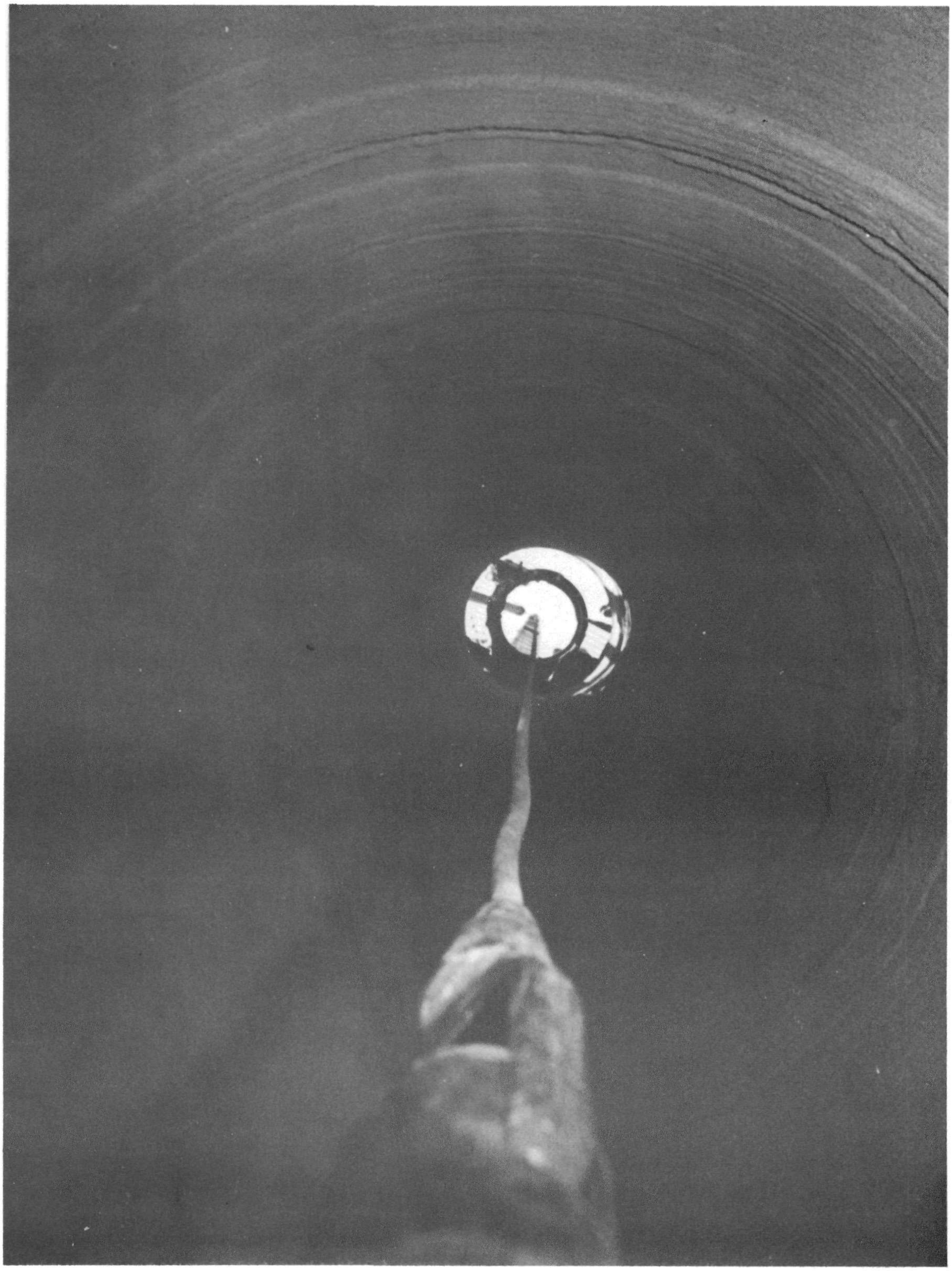

Figure 7. Interior View of a Pit for Radioactive Cement Paste

TABLE IV

PUG MILL OPERATING DATA FOR SEVERAL RUNS IN 1970

	Feed Composition, Liters							Radioactivity of Feed, μCi/ml			
Run No.	Neut. Am.	KOH-F Waste	Treated Strip	Plant Sludge	Total	Total Cement Paste Liters	Oper. Time, Min.	Gross α	Gross β	Gross γ	Av. Cyl. Comp. Str. kg/cm^{2*}
1	3,930	1,980	-	-	5,910	9,420	208	2.38	2.54	0.08	137
2	2,750	5,910	-	1,330	9,990	16,180	355	0.91	1.05	0.04	228
3	1,680	1,420	1,520	1,380	6,000	10,960	187	0.36	0.28	0.01	112
4	2,350	-	2,910	-	5,260	8,820	202	2.28	1.87	0.02	150
5	-	2,790	-	2,220	5,010	9,840	165	0.02	0.02	0.001	117
6	2,960	-	-	2,040	5,000	8,630	175	2.77	2.72	0.02	175

*1 kg/cm^2 = 14.22 psi.

underway to recover this isotope; the resultant weak nitric acid waste will be evaporated and the acid recovered. The heel of the evaporator will be neutralized and fixed in cement. The ^{241}Am and ^{239}Pu will be stored in small volume containers for reuse or burial.

3.2.3 Alcohol-Plutonium Wastes

Forty liter volumes of alcohol containing gram quantities of plutonium-239 are received at irregular intervals for disposal. Due to the volatility of the solution and the level of radioactivity, extraordinary care is essential during processing. A 200 liter drum containing about 128 kg of Portland cement, 4 kg of ground, exfoliated vermiculite, 50 liters of water, and several pieces of brick (to aid in mixing) is strapped into a drum tumbler. The alcohol waste is transferred by vacuum applied to the drum; the drum is tumbled for fifteen minutes; the paste is allowed to set for a few hours and the drum is taken to solid waste burial.

The quantity of this waste is indicated in Table I. No airborne alpha contamination above tolerance has been detected during this operation and leach tests of the paste indicate excellent retention of the plutonium.

3.2.4 Contaminated Vacuum Pump Oil

Vacuum pump oil contaminated with plutonium-238 is delivered for processing at the TA-21 waste treatment plant. The concentration of alpha radioactivity ranges from background levels to 5.4 μCi/ml. It was decided to use incineration for volume reduction and encasement of the residue in cement paste for fixation of the radioactivity.

The incinerator is a salamander of the type commonly used for heating in the construction industry. The salamander is installed in a hood arrangement through which combustion products and large volumes of air are drawn. The air cools the exhaust sufficiently within a duct distance of about six meters so that the gas mixture can be passed safely through a 60 cm square HEPA filter. The filter exhaust is monitored; results have been very satisfactory, averaging about 6 d/m-m^3 (2.7×10^{-6} μCi/m^3) alpha with a one-time maximum of 120 d/m-m^3 (54×10^{-6} μCi/m^3).

Kerosene is mixed with the oil in the ratio of one part kerosene to one part oil to aid combustion. When the burner shows signs of wear or activity has accumulated in the residue to the gram level, the entire salamander unit is placed in one of the pug mill pits and covered with the cement paste.

3.3 Costs

The overall cost of treating liquid radioactive and toxic chemical wastes at both plants in Los Alamos is about $6.60 per cubic meter. This figure is based on the total operating cost of the group and includes the cost of a certain amount of research. It does not include depreciation, however.

In transferring to the pug mill continuous method of managing plant sludge and americium wastes at DP-257, operating costs for processing these particular wastes were decreased 75%.

Placing the paste into 200 liter steel drums rather than the pits, however, will reduce this savings because cost of drums is more than pit cost per unit volume. In addition, a cost of transportation of the filled drums will also be incurred.

4. SAFETY CONSIDERATIONS, RADIOLOGICAL

4.1 Personnel

For normal plant procedures, personnel wear a complete set of protective clothing including underclothing and shoes. Rubber gloves are used to handle anything suspected of being contaminated with radioactivity, canvas and plastic booties are available and outer jackets and boots are provided. For special operations, all plant personnel are equipped with personally fitted full face masks. Full protective suits with independent air supply are also available and during special operations, health physics surveyors are present. Before such an operation is undertaken, an approved operating procedure is on file.

Personnel in waste management wear film badges and dosimeters during working hours. Special operations require nose swipes upon completion to determine whether radioactivity was inhaled.

Urine samples are collected on a regular basis and analyzed for the principal alpha emitters with which personnel might come into contact.

4.2 Plant

All operations areas are checked daily by health physics surveyors who indicate in their reports the areas or articles that are found to be contaminated and the levels of activity. Janitors, plant personnel, and if necessary, personnel of the decontamination section clean the areas to a no-swipe condition. Contaminated areas are painted.

Air samplers are operated in all areas where radioactivity may be released. Air sampler filter papers are changed daily and analyzed. Concentration of alpha radioactivity above 4 d/m-m^3 of air must be investigated by the group involved and steps are taken to correct the situation.

4.3 Operations

Where transfer of radioactive solutions is necessary, a fail-safe vacuum system is used. Vents are equipped with HEPA filters and gauges are used to verify the integrity of the systems.

In the pug mill system, air is withdrawn from the mill during operation to maintain an air pressure in the covered unit which is slightly less than atmospheric. The air withdrawn is evacuated through a HEPA filter.

Though accumulation of critical amounts of plutonium or uranium in any of the many sumps of the waste management systems is highly improbable, steps are taken to assure that sludge accumulations are minor. Many sumps are mixer-equipped and solids cannot accumulate; for the others, inspection and cleaning occur on a regular basis.

In the pug mill system, the pits receiving paste or wash water and the pits awaiting capping are covered with removable steel lids until they have been capped. Air sampling in the area has not indicated problems due to airborne contamination.

Work done under the auspices of the U. S. Atomic Energy Commission.

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FIGURES

WASTE REDUCTION STUDIES

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April 15, 1971

U. S. Atomic Energy Commission Contract AT(29-1)-1106

WASTE REDUCTION STUDIES

D. L. Ziegler
The Dow Chemical Company

The waste process development efforts at Rocky Flats are concerned with three major plant objectives:

1. To decrease the amount or bulk volume of radioactive waste generated in the operation of the plant.

2. To decrease the amount of plutonium and americium contained in the wastes.

3. To convert the radioactive waste into forms which will provide for maximum isolation of radionuclides from man's biological environment.

Sometimes the best way to reduce the volume of waste is to look at the place in the process where the waste is being generated and make process changes which will result in elimination or reduction in amount being generated. Many process improvements have been proposed and are being implemented at Rocky Flats which should decrease the amount of waste being generated. These process modifications are outside the scope of this report which will be confined to a discussion of process development work concerned with existing plant waste streams.

The radioactive waste from the Rocky Flats plant can be divided into four general categories:

1. Large, obsolete or worn-out process equipment. This includes items such as used lathes, gloveboxes, and miscellaneous building materials. In general, these items are decontaminated as far as practical before they are removed from operation.

2. Solid waste from operation of the plutonium recovery process. This includes residues generated in the fabrication operations at Rocky Flats which have been subjected to the aqueous plutonium recovery process. Additional waste materials are generated in the operation of the recovery process.

3. Liquid waste streams. This category includes aqueous waste from the plutonium recovery process and lathe coolant waste from the fabrication operations. In addition, some low-level aqueous wastes are generated from other operations such as the laundries. In general, the liquid wastes are processed in the waste treatment facility which converts essentially all of the radioactivity to a solid form.

4. Nonline-generated solid wastes. This category contains items such as surgeons gloves, paper, and plastic which are generated in the process areas outside of the glovebox lines. In general, this waste contains an insignificant amount of contamination and falls within the limits of the low specific activity definition.

Essentially all of the radionuclides lost from the plant are in the first three categories of waste and the present waste development activities are confined to the liquid and solid waste streams from the plutonium recovery process. One of the most attractive methods of reducing radioactive waste is to recover materials for recycle or reuse. This technique is the basis of development efforts for the liquid waste reductions. These processes utilize distillation, ion exchange, and precipitation. Other unit operations such as electrodialysis, ion flotation, ultrafiltration and solvent extraction are being evaluated.

Figure 1 is a schematic flow diagram of the proposed aqueous waste recovery process. In this process the dilute nitric acid streams, distillate, and americium ion column effluent would be subjected directly to distillation, where the acid would be concentrated as the bottom product and water would be removed as an overhead stream. These two streams contain a minor amount of impurities which would be returned with the nitric acid. The plutonium contained in these streams would be recovered by the distillation operation because it would remain in the bottoms product nitric acid stream.

The ion column effluent is a 7 $\underline{N}$ nitric acid waste stream and contains a large quantity of dissolved solids. Because of the dissolved solids, it cannot be recovered directly by distillation. By the use of a low-level ion exchange, the plutonium can be separated from the impurities. The eluate, a low normality nitric stream containing the recovered plutonium, would go to distillation where the plutonium and acid would be recovered in the bottom product for reuse. The effluent containing the dissolved solids would be subjected to an evaporation step where a portion of the acid can be taken overhead and recovered in the distillation column. The amount of acid which can be recovered by evaporation of this stream is limited by salting out of the dissolved solids in the bottoms product which will go to waste treatment. The recovery of nitric acid can be increased by taking water or dilute acid from the top of the distillation column and recycling it through the evaporator. This will decrease the acid normality in the bottoms and increase the amount of acid going to distillation. The use of this recycle technique will necessitate increasing the size of both the evaporator and distillation column.

An alternative of using a fluid bed waste calciner in place of the evaporator is being evaluated in a test program with Idaho Nuclear Corporation. In this operation, all the aqueous portion would be taken overhead and the impurities would come out as calcined solids. One of the big problems with this alternative will be recovery of the acid from the off-gas stream because of the air used for fluidization. Another alternative being evaluated is the use of a wiped film evaporator where the solids could be removed as a thick slurry.

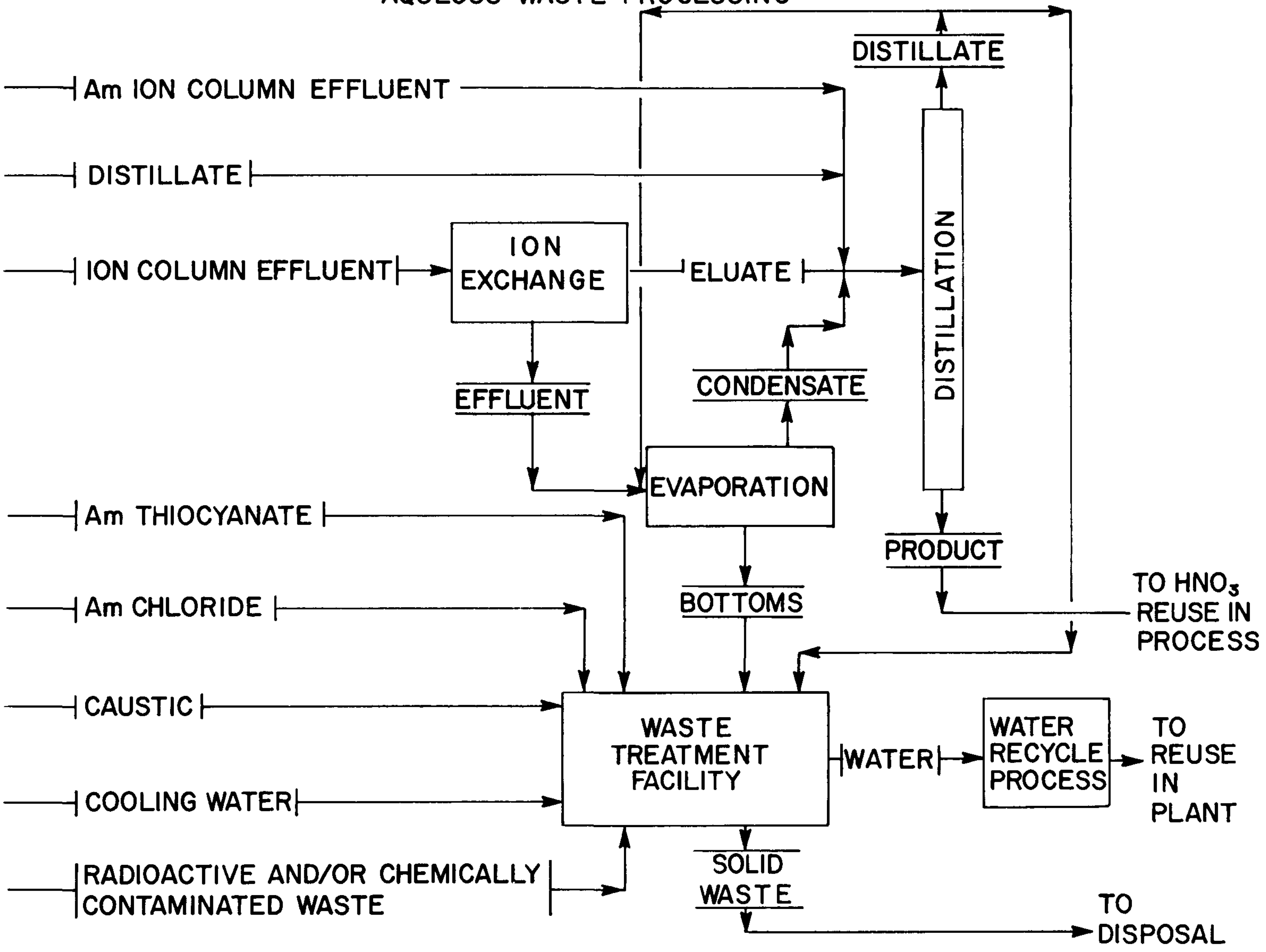
AQUEOUS WASTE PROCESSING

FIGURE 1

The use of the acid recycle process just discussed will aid both in reduction of the amount of waste and reduction in the amount of plutonium being discarded. As long as the plutonium is confined to a reusable stream, it will not leave the plant as radioactive waste. The major waste volume reduction is a result of the decreased amount of caustic which has to be added in waste treatment operation to neutralize the acid streams. At the present time the caustic which is added eventually leaves the process as sodium nitrate evaporator salts.

When the liquid wastes are converted to solids in waste treatment, this waste category is a greater volume of waste than the solid waste discarded from the plutonium recovery process. However, the solid waste category contains more plutonium. This solid waste is composed of materials such as graphite, reduction slag, box gloves, incinerator ash, scrap metal, and other miscellaneous residues which have been subjected to nitric acid leaching in the process for removal of plutonium. Each of these materials have to be considered separately to find improved methods for recovering additional plutonium. For example, a solution of nitric acid-aluminum nitrate has been found effective in removing additional plutonium from the leached reduction slag. The aluminum tends to complex the fluoride ion in solution, thereby yielding the calcium fluoride slag more soluble in this solution than is obtained with just nitric acid. Partial dissolution of the slag tends to increase the amount of plutonium which will be put in solution.

By contrast, the incinerator ash is more soluble in nitric acid with fluoride ion than it is in a solution of hydrochloric acid with stannous ion and fluoride ion. Yet, the hydrochloric acid solution appears to be more effective in recovering the plutonium than is the nitric acid system. This hydrochloric leach system has been shown effective in decreasing the plutonium concentration in non-leached ash to the present discard limits, but only moderately effective in decreasing the concentration in order of magnitude below the present discard limits. Fluorination treatment prior to leaching and fusion methods have also been evaluated. The fusion treatments were not very effective but the fluorination warrants further study. Because of the difficulty of recovering plutonium from incinerator ash, pyrolysis is being evaluated as an alternative to incineration. In pyrolysis, the combustible material would not be subjected to as high a temperature as those encountered in incineration. These lower temperatures should prevent or minimize the formation of refractory plutonium oxide which is difficult to dissolve.

These leaching processes are designed to reduce the plutonium content of the waste but would not result in a waste volume reduction. In fact, the additional processing would tend to increase the volume of waste. The process being considered for graphite waste is designed to reduce the plutonium losses and should essentially eliminate this material as a waste. Presently the plutonium is recovered from graphite by a nitric acid leaching process. The proposed process for graphite is combustion. Both a fluid bed reactor and a static bed reactor are being evaluated for this application. Again the recovery of plutonium will depend on being able to combust the graphite at a temperature which will not yield the refractory-type plutonium oxide. Due to the better temperature control obtained in a fluid bed reactor, combustion has been accomplished at reasonable rates at operating temperatures below 900°C. This process is also being evaluated in a test program with Idaho Nuclear Corporation.

Just as some processes being developed would result in decreasing plutonium losses without reducing waste volume, others are being developed which would reduce the amount of volume of radioactive waste without recovering any additional plutonium. An example is the waste vitrification process being developed. This process accomplished the waste volume reduction through increasing the waste bulk density. The waste streams which could be converted to a glass are the two waste sludges produced in the aqueous waste decontamination process in waste treatment, and leached residues such as insulation, glass, incinerator ash and reduction slag. For this process the waste materials would be calcined, mixed with a small amount of boron and sodium, and vitrified at about 1000°C. At the present time the waste vitrification process is being evaluated using a continuous laboratory vitrification furnace which utilizes the electrical resistance of the melt as the heat source. The power source is an arc welder with one electrode inserted in each end of a ceramic-lined inconel crucible. A pilot-scale test is being made in a project with Battelle Northwest Laboratory.

For Rocky Flats waste, the borosilicate-type glass can be produced using 20 wt % or less additives based on the product weight. More additives would be required to convert our waste to a phosphate-type glass. In addition to the volume reduction which would be obtained by this process, the waste would be converted to a form which would maximize the isolation of the radionuclides from man's biological environment.

Other processes which are under consideration for waste volume reduction at Rocky Flats are: decontamination of obsolete process equipment for use in other operations or other AEC facilities, and compaction or incineration of the combustible portion of nonline-generated waste. If incineration of this waste is implemented, the ash could be combined with other solid wastes and incorporated in the vitrification process. This process scheme would maximize the waste volume reduction and would maximize the isolation of the small amount of radioactivity contained in the nonline-generated solid waste.

EVALUATION OF RESPIRATOR PERFORMANCE BY DOP MAN TEST

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EVALUATION OF RESPIRATOR PERFORMANCE BY DOP MAN TESTS*

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ABSTRACT

Evaluation of respirator performance by quantitative respirator-man tests during simulated working conditions is a new project at LASL, and is supported by the Director of Regulations of the U. S. Atomic Energy Commission. Respirator performance data on twelve types of particulate respirators, approved by the U. S. Bureau of Mines for protection against up to 10 or 100 times the maximum permissible concentration of radioactive and highly toxic contaminants, are presented. Overall respirator performance is evaluated by continuously sampling air from inside the facepiece and recording the dioctyl phthalate (DOP) aerosol penetration, while the subject performs specified exercises in a DOP aerosol chamber. The results indicate that quantitative tests of the fit of the respirator facepiece on the ultimate wearer provide an accurate and meaningful protection factor for that particular person-respirator combination. Comparison of tested respirator facepieces shows a large difference in sizes from different manufacturers, although only one size of each approved device is available. Comfort ratings, which were found to correlate with performance, were recorded.

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*Work performed under the auspices of the U. S. Atomic Energy Commission.

The average DOP penetration ranged from less than 0.01 to 20.0%, depending on the size and shape of the test subject's face and the type of respirator tested. All our test subjects were able to obtain an average penetration of less than 10% with one of the five half-mask respirators evaluated and less than 1.0% with two of the full-face masks.

INTRODUCTION

Evaluation of overall respirator performance by quantitative dioctyl phthalate (DOP) testing of men wearing respirators in simulated working conditions is a new project at the Los Alamos Scientific Laboratory (LASL), and is supported by the Director of Regulations of the U. S. Atomic Energy Commission (AEC). The results of qualitative facepiece leakage tests using isoamyl acetate or an irritant fume have been reported by the LASL Industrial Hygiene Group[1-3] and other AEC Contractors.[4-6] However, methods for evaluating overall respirator performance by quantitative tests on the wearer were developed only in the last decade. In the U. S., the first method using a uranine aerosol was developed at the Harvard School of Public Health by Silverman[7,8] with support by the AEC Division of Biology and Medicine. In 1965, the U. S. Bureau of Mines (B of M), in their Approval Schedule 21B,[9] adopted quantitative uranine and DOP respirator-man test systems for approval testing of particulate respirators for protection against airborne radioactive particulates. Since then, they have used the DOP man test system only to evaluate the overall performance of a particulate respirator worn by six men of widely varying facial size and shape for protection against airborne concentrations of highly toxic particulates up to 10, 100, or 1000 times the maximum permissible concentration (MPC) or threshold limit value (TLV). Although they do not specify the type of facepiece for these various levels of respiratory protection, in practice, they have approved[10] five half-mask respirators for a protection level of 10 x MPC or TLV (protection factor = 10) and seven full-face masks for a protection level of 100 x MPC or TLV (protection factor = 100).

The term protection factor (PF) has been used by the U. S. AEC[11] and is defined as the ratio of contaminant concentration in the ambient air to contaminant concentration in air inside the respirator facepiece. One purpose of our current respirator studies program is to develop quantitative man test systems to evaluate the overall performance of all types of respirators. This paper discusses only the results of the performance evaluation of high-efficiency particulate respirators by DOP man tests.

The LASL DOP Man Test System

The LASL DOP respirator-man test system has three major components: a model Q-127 thermal DOP generator producing a mondisperse 0.3-µm liquid aerosol, a man test chamber, and an analyzing system consisting of a forward light scattering photometer and a recorder. This test system, shown in Fig. 1, is essentially a duplication of the B of M DOP aerosol-man test system used to test approved respirators for protection against radioactive particulates.

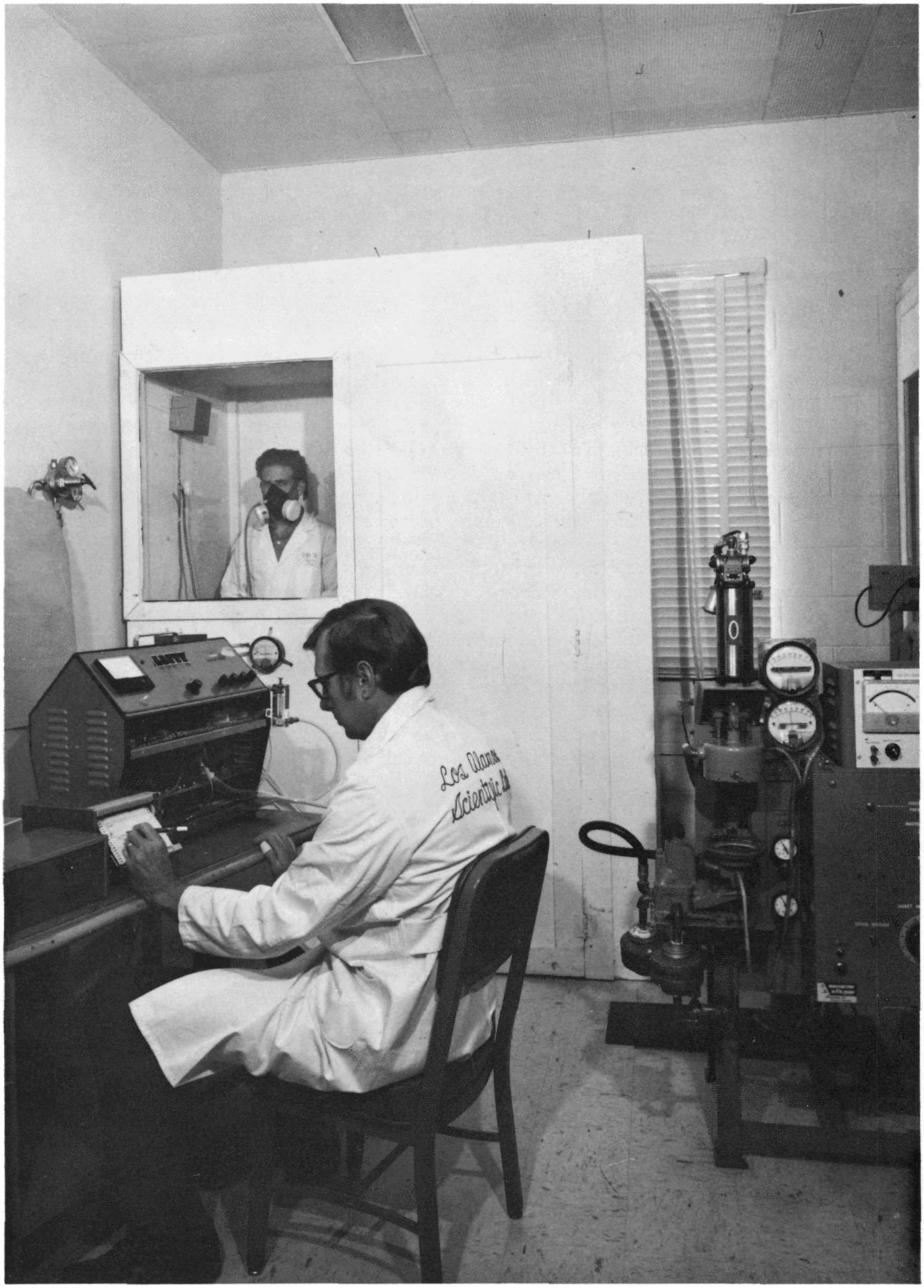

Fig. 1. LASL Quantitative DOP Aerosol Respirator-Man Test System.

The LASL quantitative DOP man test chamber shown in Fig. 1 consists of two 3 by 3 by 8 ft high chambers with an interconnecting door. One chamber, supplied with the challenge aerosol at approximately 100 liters/min in a concentration of 70 to 100 mg/m^3, is used for the tests. The other, swept with room air, is used as an air lock to prevent DOP contamination of the surrounding area. Both the challenge atmosphere in the test chamber and the atmosphere within the facepiece cavity are tested.

The analyzing system is a forward light scattering photometer with four ranges: 100, 10, 1, and 0.1% of full scale. A constant sampling rate of 8 liters/min, specified by the B of M for their DOP man test system, has been provided.

DOP Respirator-Man Test Procedure

Each respirator was modified by inserting a sample probe through the facepiece at a point where it would not distort the facepiece fit. The probe was placed below the nose and near the mouth to detect leaks into the breathing zone of either nose or mouth breathers. Before entering the test chamber, the subject donned the respirator and tested the fit. When wearing a half-mask respirator, the subject tested the fit by either the positive or negative pressure method. Subjects wearing full-face particulate respirators were tested with an irritant fume before they entered the chamber. If a leak was detected, the DOP man test was not run. While in the chamber, each test subject performed a work schedule of exercises while air samples were taken continuously from inside his facepiece at a flow rate of 8.0 liter/min. The exercises consisted of normal and deep breathing while sedentary, turning the head from left to right and up and down, normal smiling, frowning, reciting the alphabet, reading, running in place, and a final sedentary period. Each exercise was continued until we could record the average DOP penetration for that particular exercise. (≈ 0.5 to 2 min). This permitted us to determine the maximum leakage during any exercise and to calculate an average DOP penetration or leakage during the entire test.

DOP Man Test Results With Half-Mask Particulate Respirators

We have evaluated the overall performance of five types of half-mask particulate respirators approved by the B of M for protection against up to 10 x MPC of airborne radioactive particulates. For this approval, Schedule 21B specifies that during DOP man tests the maximum penetration during a single test shall not exceed 1.0% and the average penetration during three tests shall not exceed 0.5%. To meet this requirement, the B of M found it necessary to eliminate certain exercises such as talking and facial movements. The B of M recognizes that this specification is unrealistic because an approval for protection against up to 10 x MPC of airborne radioactive particulates implies an overall respirator efficiency of 90%. All of our DOP man tests on subjects wearing half-mask respirators were made while the subject performed all of the exercises specified above. These exercises are also specified by the B of M for approval testing of respirators for protection against up to 100 x and 1000 x MPC or TLV.

The results of our DOP man tests on B of M approved high-efficiency particulate half-mask respirators are shown in Table I. The per cent of test subjects with maximum DOP penetration of > 1 and > 10% during the specified

TABLE I

Results of DOP Man Tests on Approved[a] High-Efficiency Half-Mask Respirators

Type of Respirator	Number Subjects Tested	Comfort Rating[b] 1 to 5	% of Subjects with Maximum DOP Penetration Listed	
			> 1.0%[c]	>10.0%[d]
MSA Custom Comfo 21B-90	50	2.2	25	2
Willson 1009 21B-94	28	3.5	57	7
A.O. 6057 21B-98	31	2.8	48	10
Acme Duo-Seal 21B-100	43	2.2	14	2
Welsh 7580 21B-105	31	2.7	13	0

[a] Approved by U. S. Bureau of Mines (B of M) for protection against up to 10 x MPC of radionuclide particulates.

[b] Subject makes comfort rating after 20-min DOP man test. The rating is an average for all subjects The rating system is 1-Very Comfortable, 2-Comfortable (could wear 2 to 4 hr), 3-Barely Comfortable, 4-Uncomfortable, and 5-Intolerable.

[c] B of M Schedule 21B specifies 1.0% maximum DOP penetration of respirator during deep and shallow breathing, turning head, and running place for approval of respirator for protection to 10 x MPC. All facial movements prohibited for 10 x MPC approval.

[d] Proposed Schedule 21C specifies 10.0% maximum DOP penetration during all chamber exercises for approval of respirator for protection up to 10 x MPC, same as 100 x and 1000 x respirators. These exercises are normal and deep breathing, normal smiling, cough, turning head from side to side, reciting alphabet, talking or reading, running in place, and frowning.

exercises is tabulated. The subjects with maximum penetration of > 1% performed only the exercises currently specified for approval in Schedule 21B which prohibits all facial movements and specifies a maximum DOP penetration of 1.0% on a single test. The subjects with a maximum DOP penetration of > 10% performed all of the eight basic exercises including facial movements. A B of M proposal, Schedule 21C, specifies a maximum DOP penetration of 10.0% during a single test involving all of the specified exercises for approval up to 10 x MPC. Table I also lists comfort ratings made by the test subjects after the 10-min DOP man test was completed. The rating given is an average for all subjects. The higher the rating, from 1 to 5, the less comfortable the respirator. Note that all of the test subjects wearing the half-mask respirators in the DOP man test chamber had been previously trained and fitted in the qualitative isoamyl acetate fitting chamber. The variation in number of test subjects for each respirator reflects the number of men who were able to obtain a qualitative fit with each respirator. Previous reports[1-6] on qualitative fitting tests on half-mask respirators indicate a wide variation in the per cent of men able to obtain a satisfactory qualitative facepiece fit using either isoamyl acetate or an irritant fume test. Each approved respirator is available in only one size. The largest and smallest were reported to fit 40 to 60% of the men tested. 75 to 80% of the test subjects were able to obtain a satisfactory qualitative fit with the best of the medium-size respirators. Note that our quantitative DOP aerosol respirator man test was made only on men who could obtain a satisfactory qualitative fit.

The variation in facepiece dimensions of approved half-mask respirators is illustrated in Fig. 2. Facepiece length and width are plotted relative to the Air Force small, medium, and large oxygen masks which were manufactured after extensive anthropometric facial measurements of thousands of Air Force personnel. We believe that a more realistic method of expressing the overall efficiency of a half-mask respirator is by the average or integrated leakage during all of the scheduled exercises while the test subject is in the man test chamber. Table II lists the per cent of subjects for whom the average penetration was between the values shown. In addition, the protection factor for each of these average DOP leakages is shown. The PF listed was based on an integrated leakage over the 10-min man test, not on the B of M Schedule 21B. For example, a PF of 10 was listed for an average DOP penetration of > 5.0 and $\leq 10.0\%$. This implies an overall efficiency of 90%. Except during a test that was terminated when the penetration exceeded 1% during initial sedentary normal breathing, all of the men had a protection factor of 10 or greater. In fact, with three of the respirators tested, most of the test subjects had a protection factor of 100 or more. We believe that these results prove that quantitative facepiece fit tests that evaluate the fit of the respirator on the ultimate wearer provide an accurate and meaningful protection factor for that particular person-respirator combination.

Results of DOP Man Tests On Full-Face Particulate Respirators

We have evaluated the overall performance of seven types of full-face particulate respirators approved by the B of M for protection against up to 100 x MPC of airborne radioactive particulates. In addition, we have evaluated the U. S. Army M9A1 full-face mask commonly used by several AEC Contractors. The three available sizes were used to obtain the best possible fit. After donning the full-face mask and before entering the DOP man test chamber, each man was subjected to an irritant fume leak test to make

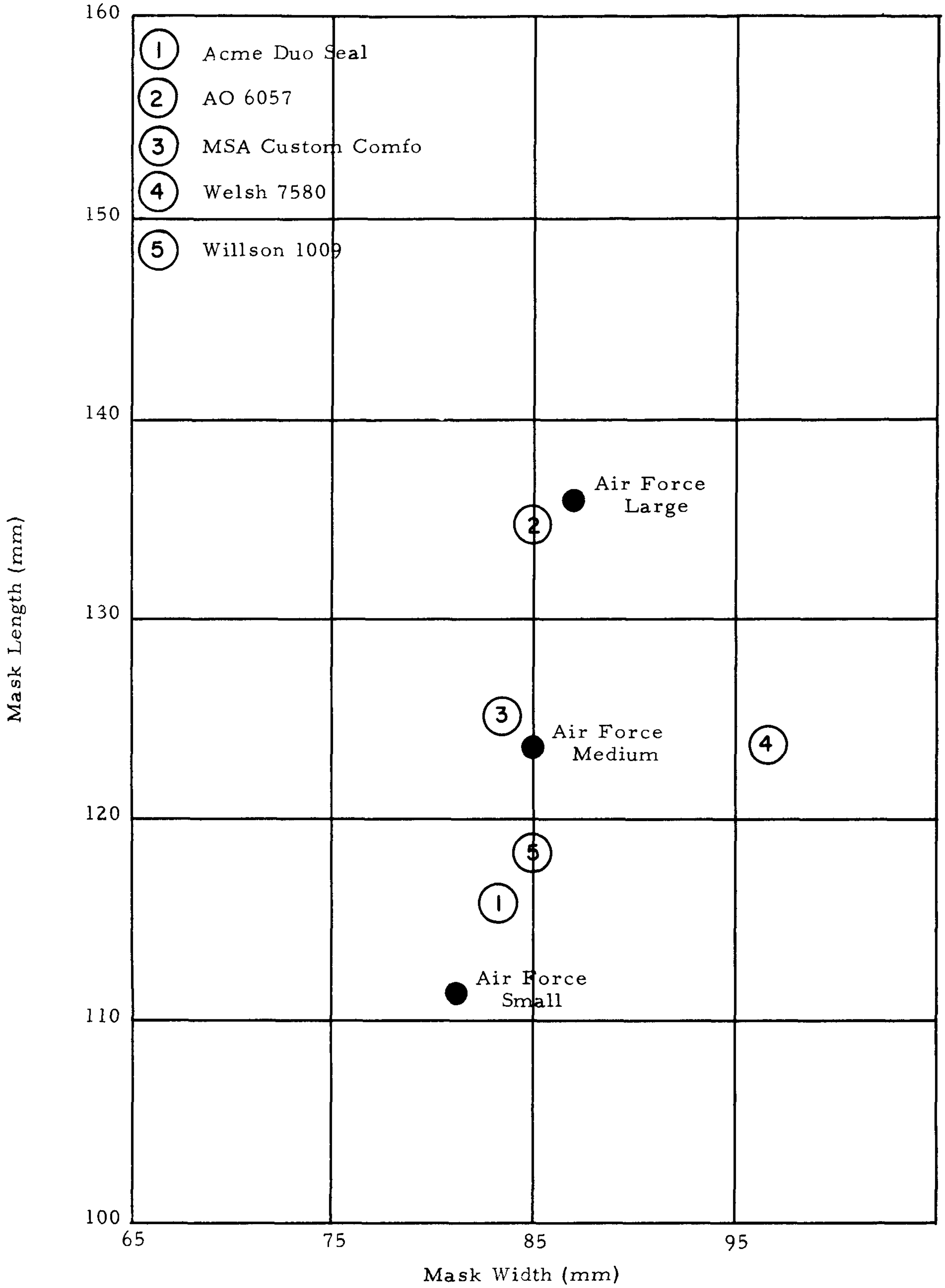

Fig. 2. Half-Mask Facepiece Dimensions vs Three Sizes of Air Force Oxygen Masks.

TABLE II

Results of Quantitative DOP Man Tests on Subjects Wearing Half Mask Particulate Respirators

Leakage Average[a] DOP Pen. %	Protection Factor PF[b]	% of subjects for whom penetration was between values shown				
		Acme 8201R	Amer. Opt. 6057	MSA Custom Comfo	Welsh 7580	Willson 1009
≤ 0.1	1000	19.2	33.3	35.7	34.9	9.4
>0.1 - ≤0.5	500	42.3	11.1	32.1	44.2	15.6
> 0.5 - ≤1.0	100	21.2	8.3	7.1	9.3	21.8
> 1.0 - ≤2.0	50	13.5	16.7	10.7	7.0	15.6
> 2.0 - ≤5.0	20	0.0	11.1	7.1	2.3	12.5
>5.0 - ≤ 10.0	10	1.9	13.9	0.0	2.3	15.6
> 10.0 - ≤ 20.0	5	0.0	0.0	0.0	0.0	0.0
TT[c] > 1.0% N.B.	--	1.9	5.6	7.1	0.0	9.4
No. Men Tested		52	36	56	43	32

[a] Average DOP penetration when subject performed specified exercises in man test chamber. The exercises (based on B of M Schedule 21B) include being sedentary, turning head side-to-side and up and down, talking, frowning, and running in place.

[b] PF- The protection factor was based on the average DOP penetration. For example, a PF of 10 is listed for an average DOP penetration of > 5.0 and ≤ 10.0%. The PF is based on an integrated leakage over the 10-min man test, not on B of M Schedule 21B.

[c] TT- When the DOP penetration exceeded 1.0% during initial sedentary period, the test was terminated.

sure that he had a proper face seal. The subject was not tested in the DOP man test chamber if he could not obtain an adequate qualitative irritant fume facepiece-to-face seal. For full-face particulate respirators approved for protection against up to 100 x MPC of airborne radioactive particulates, which implies an overall efficiency of 99.0%, we believe that a realistic measure of the overall performance is the average DOP penetration during all of the specified exercises in the man test chamber. Table III shows the per cent of subjects with an average penetration of > 1%, or penetration of > 1% where the test was terminated, for each type of full-face mask tested. The comfort ratings on all but two of the full-face respirators are also shown. A more meaningful and useful evaluation of respirators performance is shown in Table IV which lists the per cent of test subjects for whom the average DOP penetration was within the range of values shown. Protection factors are listed for each range. For example, a PF of 100 is listed for an average DOP penetration of > 0.5 and ≤ 1.0%. The PF is based on integrated leakage during the 10-min man test, not on the B of M Schedule 21B. It is evident that most test subjects obtained a protection factor of 1000 or greater based on this system. We believe these data also provide an accurate and meaningful protection factor for a particular person-respirator combination.

In January 1971, we demonstrated the usefulness of the quantitative DOP man test by using it for routine training and performance evaluation of plutonium workmen each wearing the full-facepiece respirator issued to him personally. Because all of the men working in the plutonium laboratories have been re-fitted every 2 years by the qualitative isoamyl acetate chamber method, we conducted the DOP man test without using the irritant-fume fitting test before the man entered the man test chamber. The men were instructed to put on the mask and adjust it as routinely worn during their work. The results of these tests are shown in Table V. We were surprised to find that approximately 6% of the men had a protection factor as low as 10 when tested wearing their currently issued full-face respirator. This is to say that the average DOP penetration was > 1 but < 10%. This situation was corrected by issuing a different type of mask in three cases. In the fourth case, the subject could not obtain a seal while frowning. This was traced to slippage of the silicone rubber head band. Further investigation revealed that in a significant number of masks of this particular type the head bands not only tended to slip but were very susceptible to tearing once a small nick had been made in the material. As a result, we are replacing all silicone rubber head bands with neoprene. This seems to have solved both the head-band slippage and the breakage problems. The last column of Table V shows the final tabulation after the head bands were replaced and all the men had masks that provided a protection factor of 100 or higher. These protection factors were tabulated on the basis of the average DOP penetration. For example, if the average penetration was < 1.0%, the subject had a protection factor of 100.

Compact Quantitative Aerosol Respirator Man Test Systems

Compact quantitative NaCl aerosol man test systems have been described by Hounam[12] in the UK and White[13] in Canada. These units have been in routine use since 1965 at UK Atomic Energy Establishments and at the Chalk River Nuclear Laboratories in Canada to quantitatively measure the protection provided by each particular respirator-person combination. For example, if the maximum Na Cl penetration during specified exercises does not exceed 1.0%, White reports that the individual is considered to have a

TABLE III

Per Cent of Subjects with Average DOP Penetration of >1.0% with Full-Face Particulate Respirators

Manufacturer and Type of Mask	No. of Subjects	Comfort[a]	% of Subjects with Ave. DOP Pen. >1.0%[b]
Acme Full Vision	55	1.9	0
MSA Clearvue	88	1.9	6
MSA Ultravue	84	--	0
Welsh 7680S	108	1.9	3
Willson RFMW	26	3.3	27
Willson TFMW	26	2.8	7
Scott Scottoramic	34	--	9
U. S. Army M9A1	55	2.3	13

[a] 1 - Very comfortable
2 - Comfortable
3 - Barely comfortable
4 - Uncomfortable
5 - Intolerable

[b] Includes tests terminated when DOP penetration exceeded 1.0% during sendentary normal breathing.

TABLE IV

Results of Quantitative DOP Man Tests on Subjects Wearing Full Face Particulate Respirators

Leakage or Average[a] % of DOP Pen.	Protection Factor PF[b]	% of subjects for whom penetration was between values shown							
		Acme	MSA			Willson		Scott	
		Full Vision	Clear-vue	Ultra-vue	Welsh 76805	RFMW 809	FTMW 809	Scotto-ramic	USA M9A1
≤ 0.01	10,000	19.6	51.2	77.4	37.5	6.3	27.3	5.9	10.7
>0.01 - ≤0.05	2,000	25.0	11.9	17.9	32.7	25.0	33.3	23.5	25.0
> 0.05 - ≤0.10	1,000	17.9	4.8	4.7	9.6	6.3	0.0	2.9	12.5
> 0.10 - ≤0.50	200	32.1	21.4	0.0	10.6	31.3	12.1	41.2	30.4
> 0.50 - ≤ 1.0	100	0.0	7.1	0.0	6.7	0.0	3.0	17.6	7.1
> 1.0 - ≤2.0	50	0.0	0.0	0.0	1.0	3.1	3.0	2.9	1.8
TT[c], >1.0	< 100	5.4	3.6	0.0	1.9	28.1	21.2	5.9	12.5
No. Men Tested		56	84	84	104	32	33	34	56

[a] Average DOP penetration when subject performed specified exercises in man test chamber. The exercises (based on B of M Schedule 21B) include sedentary periods, turning head side-to-side and up and down, talking, frowning, and running in place.

[b] PF- The protection factor was based on the average DOP penetration. For example, a PF of 100 is listed for an average DOP penetration of > 0.5 and ≤ 1.0%. The PF is based on an integrated leakage over the 10-min man test, not on B of M Schedule 21B.

[c] TT- When the DOP penetration exceeded 1.0% during the initial sedentary period, the test was terminated.

TABLE V

Results of Quantitative DOP Fitting Tests On Plutonium Workers' Personal-Issue, Full Face Particulate Masks

Total No. of Subjects Tested - 65

Leakage or Average % DOP Pen.	Protection Factor PF	% of subjects for whom penetration was between values shown	
		Initial Test[a]	Final Test[b]
≤ 0.01	10,000	38	38
> 0.01 to ≤ 0.1	1,000	31	42
> 0.1 to ≤ 1.0	100	25	20
> 1.0	10	6	0

[a] The initial test was made on each subject's currently issued mask.

[b] Tabulation after training and fitting with best fitting and most comfortable mask.

satisfactory fit. This fitting test method was adopted to ensure the high degree of respiratory protection required against airborne plutonium aerosols and other highly toxic radioactive particulates.

We suggest that a compact, economical, quantitative DOP aerosol man test system should be used by each AEC contractor to evaluate the fit of the respirator on the ultimate wearer. This would provide an accurate and meaningful protection factor for each particular person-respirator combination. We have demonstrated the feasibility of this system by using a single-jet polydisperse DOP air generator (one quart), a simple plastic hood within which the subject stands, and a portable photometer. The DOP aerosol surrounding the man in the plastic hood is the challenge atmosphere which has a mass median diameter of 1.1 μm and a sigma of 1.75. Comparative tests using our laboratory test setup (Fig. 1) with 0.3-μm monodisperse DOP indicate that the penetrations measured using the compact polydisperse DOP man test are 15% to 25% lower. We consider this an acceptable man test considering its economy and compactness.

We have also developed a quantitative NaCl respirator man test system that can be adapted to make it portable from one laboratory area to another. This system will be reported in a LASL Document later this year.

A quantitative Freon-12 respirator man test system has been in routine use by the Hanford Environmental Health Foundation since January 1970. According to Adley,[14] this system enables them to measure facepiece leakages of < 1.0%.

Conclusions

1. We believe that evaluation of overall respirator performance by quantitative DOP aerosol respirator man tests provides an accurate and meaningful evaluation of the particular person-respirator combination.

2. Our evaluation of half-mask particulate respirators indicates that more than one facepiece size is needed to provide adequate protection for all individuals tested. We believe that comfort ratings on half-mask respirators, and to some extent full-face respirators, provide a meaningful measure of worker acceptance.

3. Our DOP man tests demonstrate that most test subjects were able to obtain a high degree of protection from both full-face and half-mask respirators. Also people with certain face sizes and shapes cannot obtain a satisfactory fit and good performance without testing several of the commercially available respirators.

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PLUTONIUM MOVEMENT IN AQUATIC SYSTEMS: A REVIEW

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PLUTONIUM MOVEMENT IN AQUATIC SYSTEMS: A REVIEW

Although there is much information concerning plutonium, very little is available on plutonium movement in aquatic systems. Two marine studies, specific to plutonium movement and distribution, will be reviewed, as well as the current study now being conducted at Colorado State University on the Rocky Flats environs. The two studies are Pillai, Smith and Folsoms' "Plutonium in the Marine Environment" published in Nature, Aug. 1964, and Aarkrog's "Radioecological Investigations of Plutonium in an Arctic Marine Environment" published in Health Physics, Jan. 1971. (1) (2)

Pillai, Smith, and Folsum's immediate aim was to show that ^{239}Pu from fallout could actually be measured in the marine environment. Before this study, no results were available on the levels of plutonium present in the marine environment. The article summarizes preliminary findings concerning the plutonium content in sea water, and the concentration factors for this element in certain selected organisms. Blank samples containing all chemical reagents as well as plankton samples obtained in 1950 prior to extensive nuclear testing were measured for ^{239}Pu to provide background information. The sea water sample with the lowest activity contained three times background activity. All water analyses were made on surface samples and, therefore, do not represent distribution of plutonium in deep waters. It was found that marine plants have large concentration factors while the lower concentration factors in the fish samples indicated that possible discrimination against plutonium could take place at higher trophic levels. The concentration factors given were taken as the ratio of the sample activity to that of the average coastal water activity sampled near the southern California coast. In the Pacific sea water, and in the surface sea water near the California coast they found 0. 35 - 0. 39 fCi/l. Bottom sediment showed a median value of 0. 1 pCi/gm. They found concentration factors of 250 - 500 for bottom feeding animals, 2600 for mixed zooplankton, and 1600 for phytoplankton. The fish had a concentration factor of approximately 3. It was generally concluded that plutonium is concentrated in the marine environment. However, this was merely a beginning at establishing concentration and distribution patterns in an aquatic system.

Aarkrog, on the other hand, was concerned with samples of the human food chain at Thule, Greenland, after the crash of a B52 airplane on the ice in Bylot Sound. Upon impact with the ice the chemical explosive in the four unarmed nuclear weapons detonated releasing plutonium into the environment. The total amount of plutonium in Bylot Sound was estimated to be approximately 0. 1 Ci or 1 - 2 gm. Therefore, it was expected that the accident increased the plutonium burden at Thule by about two orders of magnitude, provided the main part of the contamination was confined to Bylot Sound. Because of the increased plutonium levels in bottom sediments, the highest levels in animals were found in the bottom feeders The plutonium from the accident was mainly present as insoluble plutonium oxide particles In most cases the animals present were contaminated, but no concentration factors were able to be determined The concentration in sea water increased by a factor of two over the fallout background in sea water which was estimated to be 2 fCi/l No significant differences between plutonium levels in surface and bottom waters were detectable. No significant amounts of particulate activity were noted, except in a few cases, and these were attributed to particles stirred up during the bottom sampling

In sea bottom sediments the increase was one order of magnitude above fallout background and beneath the impact area, two orders of magnitude. Sedimentation experiments on melted ice cores showed that 85 - 95% of the debris and associated plutonium sank immediately in water and only 1% was suspended as fine particles in the water phase Most of the activity that had not been removed by decontamination of the area was bound on the sea bottom. It was evident that the measurements of the bottom sediments could not account for the total amount of plutonium supposed to have been released to the marine environment of Thule. This was due to various factors, including the possibility that some of the plutonium might have attached itself to pieces of debris not included in the sampling. Bivalves, which are bottom feeders, showed increased levels up to a distance of 15 Km. from the crash site. The median levels in bivalves were one order of magnitude greater than fallout background and in the crash area they were three orders greater. Bivalves were estimated to contain 5% of the estimated deposition in the sea bottom sediments Aarkrog suggested that bivalves are good indicators of plutonium in the marine environment. Bottom feeders showed unevenly distributed activity due to plutonium being in particulate form. Crustacea showed concentration factors of about 750 as compared to 3500 for bivalves. Analytical error due to low activity samples and particulate contamination made the concentration factors somewhat uncertain The shells of the crustacea showed higher levels than did the flesh probably due to surface contamination from bottom sediment Zooplankton showed no plutonium concentrations that could not be ascribed to fallout. Large marine plants also showed no levels above background, while various bottom animals showed values 1000 times greater than background. The highest levels in fish (470 pCi/kg) were found in a bottom fish, the Greenland halibut. Larger animals showed low plutonium levels, and also plutonium was not detected with certainty in human urine samples.

Since marine organisms have been shown to concentrate plutonium, it seems likely that fresh water organisms should concentrate it to an even higher degree. This would be predicted from comparisons of fallout fission product concentrations observed in marine and fresh water systems.

Because very little information is known concerning plutonium movement in aquatic systems, especially fresh water systems, a study of the Rocky Flats environment should answer many questions as to the distribution patterns, concentration processes, mobilization processes, and also possible food pathways of plutonium to humans. Qualitative as well as quantitative studies will be done. These will include measurements, taken seasonally, of such parameters as flow rates, volumes and depths of the monitoring ponds, pH, temperature, dissolved oxygen, and conductivity. Aquatic plants and animal life in the three main water courses of importance will be examined. This study will include species identification and abundance, and also a description of the water use past and present. Animal, plant, water, and sediment samples will be analyzed for plutonium. Initially these measurements will be taken in the 6 monitoring ponds and as far downstream as possible. Eventually samples will be taken from off site reservoirs. Aquarium studies will be designed to quantify any concentration process found in the initial sampling. Long term studies of the action of bacteria on plutonium will be made to establish if any mobilization processes do occur. This will be done by culturing sediment obtained from the holding ponds.

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SOLUBILITY OF PLUTONIUM IN SOIL/WATER ENVIRONMENTS

A Theoretical Study

by

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INTRODUCTION

Production of plutonium is increasing and will continue to increase in the future because of its importance as a source of nuclear energy. Pu-239, of the 15 known isotopes of plutonium, is the isotope of greatest importance. It is readily fissionable, has a relatively long half-life of 24,360 years, and can be produced in amounts large enough to be practical as a source of nuclear energy. As the production of plutonium increases, the problems of environmental waste storage and accidental release to the environment will continue to require close attention to prevent health hazards from inhalation of airborne plutonium as well as ingestion of contaminated food and water.

An important aspect of the behavior of plutonium in the environment, particularly its distribution and migration in soil and water, is the behavior of its dissolved species. The chemistry of aqueous plutonium is highly complex. Plutonium exists in at least four oxidation states. Plutoniums tend to combine with many other ions to form very complicated systems. Most studies have been conducted in the laboratory under conditions differing from those found in the environment. The question immediately arises, how can the behavior of dissolved plutonium be predicted and evaluated for such complicated systems?

The purpose of this study is to present an approach to these problems. More specifically, an evaluation of certain questions will be presented. For example:

(1) What parameters are necessary to evaluate the concentration of dissolved plutonium?

(2) What chemical form of plutonium controls the concentration of dissolved plutonium?

(3) Which dissolved species predominate under different specified conditions?

(4) What is the total concentration of dissolved plutonium under these specified conditions?

(5) Which anions, normally found in the environment, may complex with plutonium to significantly affect the concentration of dissolved plutonium?

APPROACH TO THE PROBLEM

Based on theoretical considerations and laboratory information, a large number of reactions can be reasonably expected to occur in the environment. If the assumption that equilibrium or near equilibrium can be reached with these reactions, then only a few will predominate and significantly affect the concentration of dissolved plutonium. The problem is to determine which reaction(s) predominate in the system under consideration. This will be accomplished by the use of phase relations and stability field diagrams. These relationships are dependent on those parameters which affect the dissolved plutonium.

Plutonium is known to exist in at least four oxidation states. The oxidation state refers to the valence state of plutonium, which is usually written as Pu^{+3}, Pu^{+4}, Pu_2^{+}, or PuO_2^{+2}. The intensity of the oxidizing or reducing conditions in a solution affects the relative proportion of the various species in solution. This intensity is represented by its reduction-oxidation potential, Eh. Eh is expressed in volts and the saturated calomel electrode is used as the reference electrode.

Aqueous or dissolved plutonium ions exist in a hydrated form, that is, surrounded by water molecules and should properly be written, for example, as $Pu(H_2O)^{+3}$. In this report the notation Pu^{+3} will be used. Aqueous plutonium readily complexes with hydroxyl ions in solution to form hydroxides, such as $Pu(OH)_4$. Therefore, the pH of a solution also affects the relative proportion of the plutonium species.

The phase relationships between the various plutonium species are a function of Eh and pH when other ions are present in insignificant concentrations or absent completely. In this case, stability diagrams can be expressed in terms of these two variables. Stability field diagrams indicate which species is the most stable and which is dominant in known specified conditions. Plutonium solids which control or fix the concentration of dissolved plutonium can also be determined. These diagrams are constructed using standard free energies of formation or equilibrium constants. The data in Table I were used to construct the stability diagram given in this study. These data were either obtained or calculated from equilibrium data found in Cleveland (1), Latimer (2), Coleman (3), Gelman et al., (4), and Seaborg and Katz (5). Due to the nature and complexity of plutonium, variations in the data were found. The data used in the construction of the diagrams were based on their experimental validity and on their relation to or consistency with other data. In order to keep the diagrams internally consistent some of the data in Table I differ from the literature values. However, these variations are still within the determination error of the literature values.

Stability diagrams have been used extensively by geochemists in recent years and details of their construction and interpretation can be found in Garrels & Christ (6) and Hem (7). However, a brief example of the construction and interpretation of a diagram for the aqueous plutonium system is given below.

The reaction of Pu^{+3} with water to form the oxide, PuO_2, can be written as follows:

$$Pu^{+3}_{(aq)} + 2\ H_2O = PuO_{2\,(c)} + 4H^{+} + e \qquad (1)$$

where the symbol (aq) means that substance preceding it is in aqueous solution, (1) refers to the liquid state, (c) to the crystalline or solid state, and e indicates an electron.

Table I: Standard free energies of formation ΔF^{o}, of plutonium species at 25°C.

Species	State	ΔF_f^{o} (k.cal/mole)
Pu (metal)	solid	0.0
Pu^{+3}	aqueous	-140.5
Pu^{+4}	aqueous	-118.8
$PuO_2{}^{+}$	aqueous	-204.9
$PuO_2{}^{+2}$	aqueous	-183.5
$Pu(OH)_3$	solid	-280.2
$Pu(OH)_4$	solid	-340.0
PuO_2OH	solid	-248.3
$PuO_2\ (OH)_2$	solid	-281.9
$PuOH^{+2}$	aqueous	-187.4
$PuOH^{+3}$	aqueous	-174.0
PuO_2OH^{+}	aqueous	-233.0
$PuO_2\ (OH)$	aqueous	-325.4
PuO_2	solid	-240.4
α-Pu_2O_3	solid	-412.0
β-Pu_2O_3	solid	-390.0

The net change in free energy for the reaction (equation (1)) can be obtained from the standard free energies of formation of the chemicals involved in the reaction.

$$\Delta F_R^o = \Delta F_{fPuO_{(s)}} + 4\Delta F_{fH^+_{(aq)}} - \Delta F_{fPu^{+3}_{(aq)}} - 2\Delta F_{fH_2O_{(1)}} \qquad (2)$$

where ΔF_R^o is the net change in the free energy of the reaction and $\Delta F_{fPuO_2(s)}$ $\Delta F_{fH^+_{(aq)}}$, etc., refer to the standard free energies of formation of the respective substances. These standard free energies of formation are the changes in free energy that occur when one formula weight of the compound is formed from the elements at a temperature of 25°C and a pressure of one atmosphere.

The free energy of the reaction, ΔF_R^o, is related to the equilibrium constant, K_{eq}, and the standard potential, E^o, as follows:

$$\Delta F_R^o = -RT \ln K_{eq} \qquad (3)$$

$$E^o = \frac{\Delta F_R^o}{nf} \qquad (4)$$

where R is the universal gas constant; T, the absolute temperature; n, the number of electrons, f, the Faraday constant, 23.06 Kcal/volt.
Equation (3) can be written as follows for a temperature of 25°C.

$$\Delta F_R^o = -1.364 \log K_{eq} \qquad (5)$$

The oxidation potential, Eh, can then be determined from equation (6) or (7).

$$Eh = E^o - \frac{RT}{nf} \ln K_{eq} \qquad (6)$$

$$Eh = E^o - \frac{.0592}{nf} \log K_{eq} \qquad (7)$$

The equilibrium constant, K_{eq}, for equation (1) can be determined from equation (8).

$$K_{eq} = \frac{[PuO_2]\ [H^+]^4}{[Pu^{+3}]\ [H_2O]^2} \qquad (8)$$

where [] refer to the chemical activity of the substance. The activity of water, $[H_2O]$, is taken as unity.
Therefore, equation (7) can be written

$$Eh = E^o - \frac{.0592}{nf} \log \frac{[PuO_2]\ [H^+]^4}{[Pu^{+3}]} \qquad (9)$$

and

$$Eh = E^o - \frac{0.0597}{nf} (4) \log [H^+] - \frac{.0592}{nf} \log \frac{[PuO_2]}{[Pu^{+3}]} \qquad (10)$$

The equilibrium line for equation (1) represents that line where PuO_2 is equal to Pu^{+3}. This is the line between the species PuO_2 and Pu^{+3} in Figure 1. In this case equation (10) reduces to

$$Eh = E^o - \frac{.0592}{nf} (4) \log [H^+] \qquad (11)$$

or

$$Eh = E^o + \frac{.0592}{nf} (4)\ pH. \qquad (12)$$

From the data listed in Table I equation (12) becomes

$$Eh = -\frac{4(.0592)}{23.06}\ pH$$

Therefore, Eh can be plotted as a function of pH as given in Figure 1.

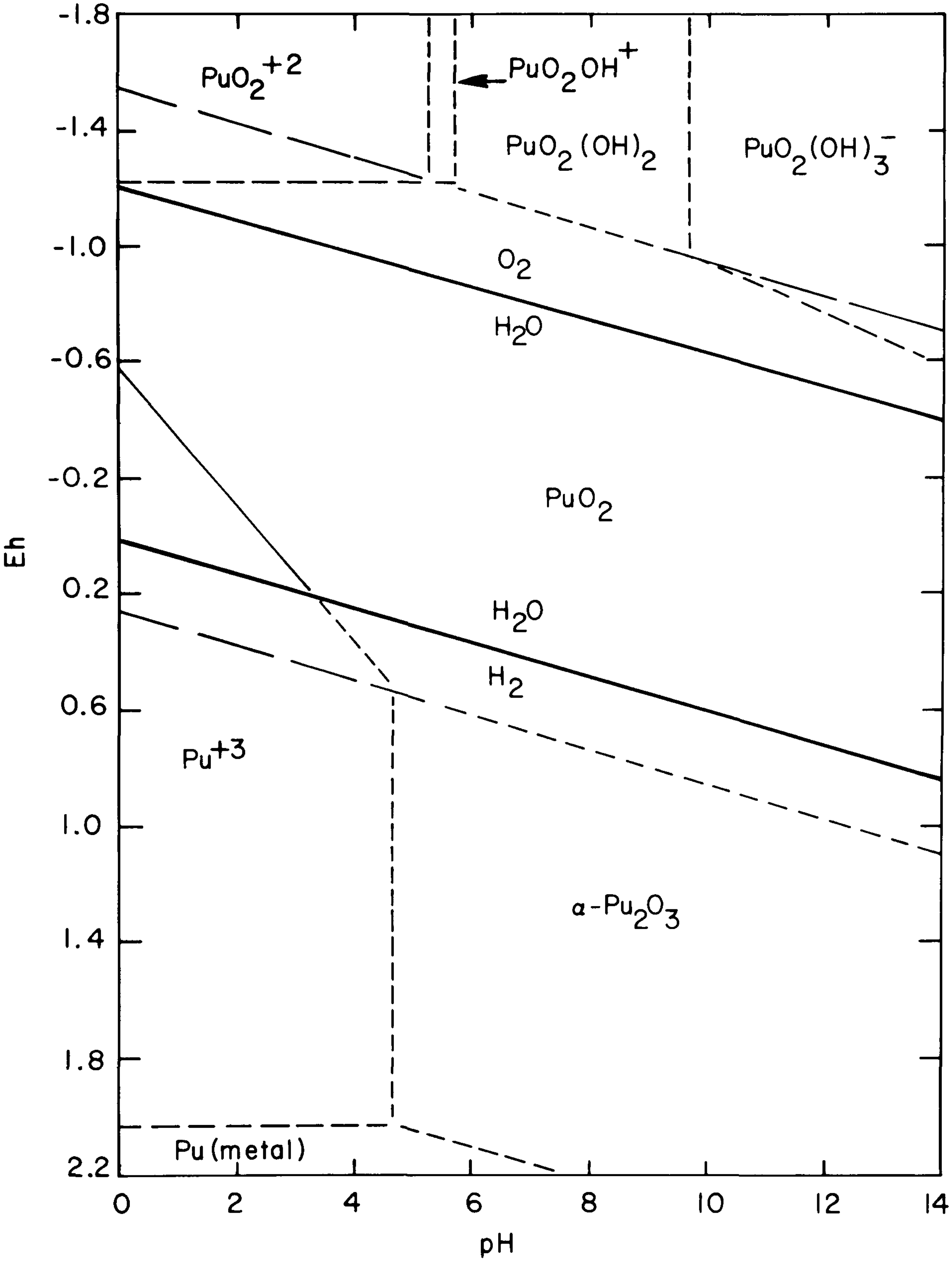

Figure 1: Stability relations of PuO_2 in equilibrium with water at 25°C and one atmosphere total pressure.

Above the upper sloping heavy line water is oxidized and below the lower sloping heavy line water is reduced. The short dashed lines outside these two lines indicate a region of metastability due to the instability of water. The long dashed lines indicate which of the solid compounds can exist under equilibrium conditions with the assumption that water would be stable. Natural soil/water environments are seldom more oxidized than -0.5 volts or more reduced than 0.3 volts. The pH conditions normally range from about 4 to 9.

Even though stability field diagrams are valuable aids or tools in the study of aqueous chemical systems, they have limitations. For example, these diagrams are based on theoretical considerations and on equilibrium conditions. These diagrams relate to environmental conditions only in so far as the species considered in the diagram are the same as those that can exist in the environment and only in so far as chemical equilibrium, or near equilibrium, exists in the environment. Chemical equilibrium of the dissolved species with plutonium solids implies that the actual or potential amounts of available plutonium is greater than the amount soluble in the volume of water under consideration. Stability diagrams do not contribute to a knowledge of the rates of chemical reactions.

These diagrams have their greatest value in relating good field measurements to theoretical considerations. However, in absence of good field measurements, they do contribute to an understanding of the behavior of the chemical species of a particular system. Further discussion of the value and limitation of stability field diagrams may be found in Hem (7).

The stability diagrams indicate that for the species considered (Table I) and for conditions of equilibrium the oxide solid, PuO_2, is the dominant solid and also the solid which controls the concentration of dissolved plutonium. For example, if an excess of two solid compounds PuO_2 and $Pu(OH)_3$ are placed into an aqueous environment initially both compounds will dissolve. Eventually an equilibrium will be reached between the solution and the stable solid PuO_2 and plutonium (PuO_2) will cease to dissolve. However, for the unstable phase, $Pu(OH)_3$, the solution is still undersaturated with respect to soluble plutonium. Therefore, $Pu(OH)_3$ will continue to dissolve. This creates a supersaturated condition for the stable solid, PuO_2. Dissolved plutonium then reacts to form the stable PuO_2 in order for the solution to remain at equilibrium with the stable solid. This process continues until the unstable $Pu(OH)_3$ solid dissolves completely. In this manner the concentration of dissolved plutonium is controlled or fixed by the stable compound, PuO_2.

There is some question as to the ability of dissolved plutonium to maintain an equilibrium with the solid, PuO_2, in an aqueous environment at ambient temperatures. If for example, PuO_2 is formed at a rate much slower than the rate of dissolution of $Pu(OH)_3$, the solution will become supersaturated with respect to the oxide. The dissolved plutonium may reach an equilibrium concentration with respect to $Pu(OH)_4$, a metastable compound. PuO_2, an oxide, is formed at high temperatures (700-1000°C) whereas $Pu(OH)_4$, an hydroxide polymer, forms rapidly at ambient temperatures (Cleveland, (1)). If the rate of formation of $Pu(OH)_4$ is comparable to the dissolution of $Pu(OH)_3$, then the metastable solid, $Pu(OH)_4$ will control the concentration of dissolved plutonium under these conditions. For this reason another diagram was constructed without considering PuO_2 (Figure 2). Figure 3 was constructed in which the oxides, α-Pu_2O_3 and β-Pu_2O_3, were not considered.

As a result of evaluating these diagrams, the conclusion is reached that for values of Eh and pH generally found in the environment, PuO_2 should control the concentration of dissolved plutonium. However, the metastable $Pu(OH)_4$ solid,

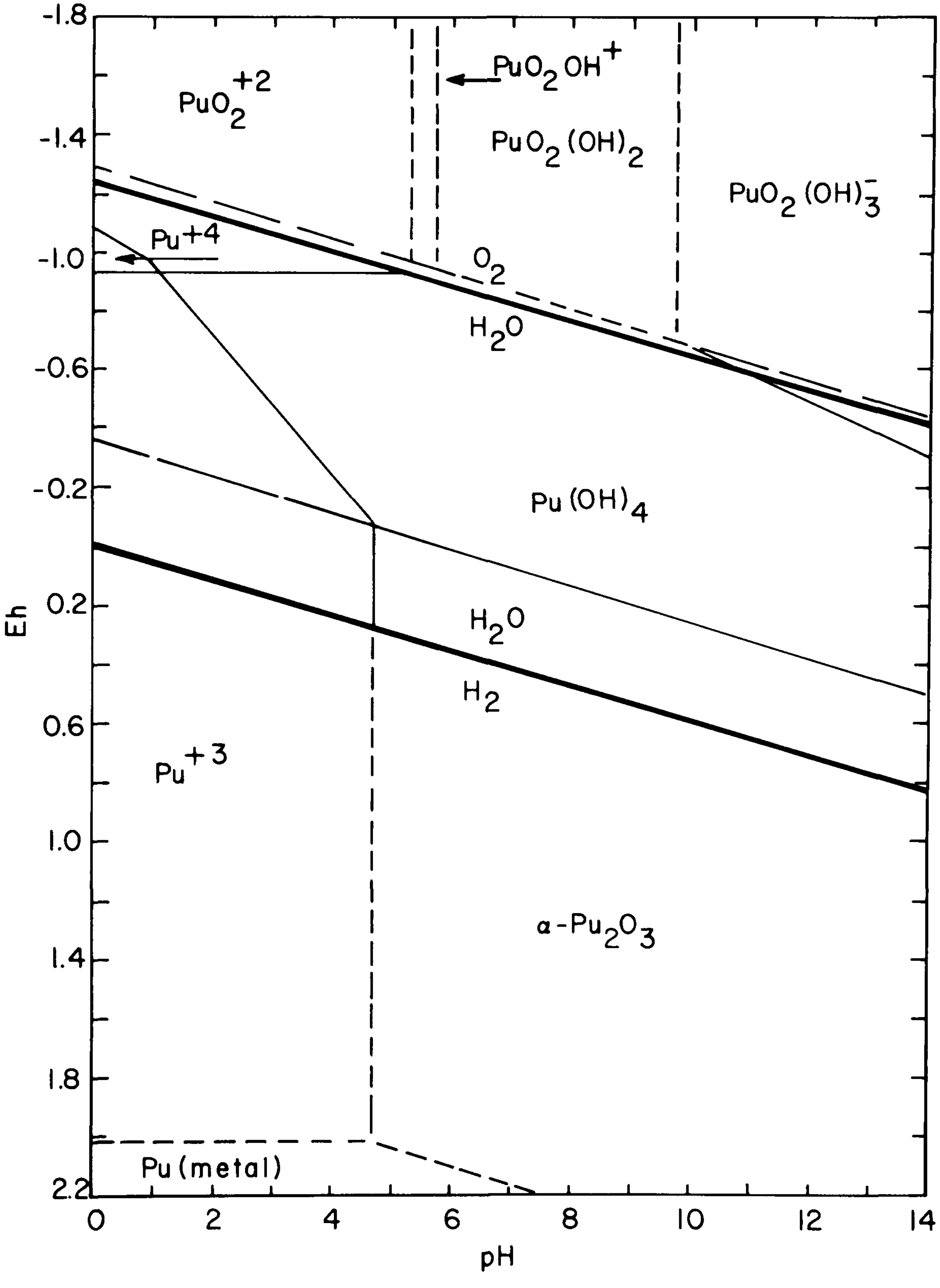

Figure 2: Stability relations of $Pu(OH)_4$ and Pu_2O_3 in equilibrium with water at 25°C and one atmosphere total pressure.

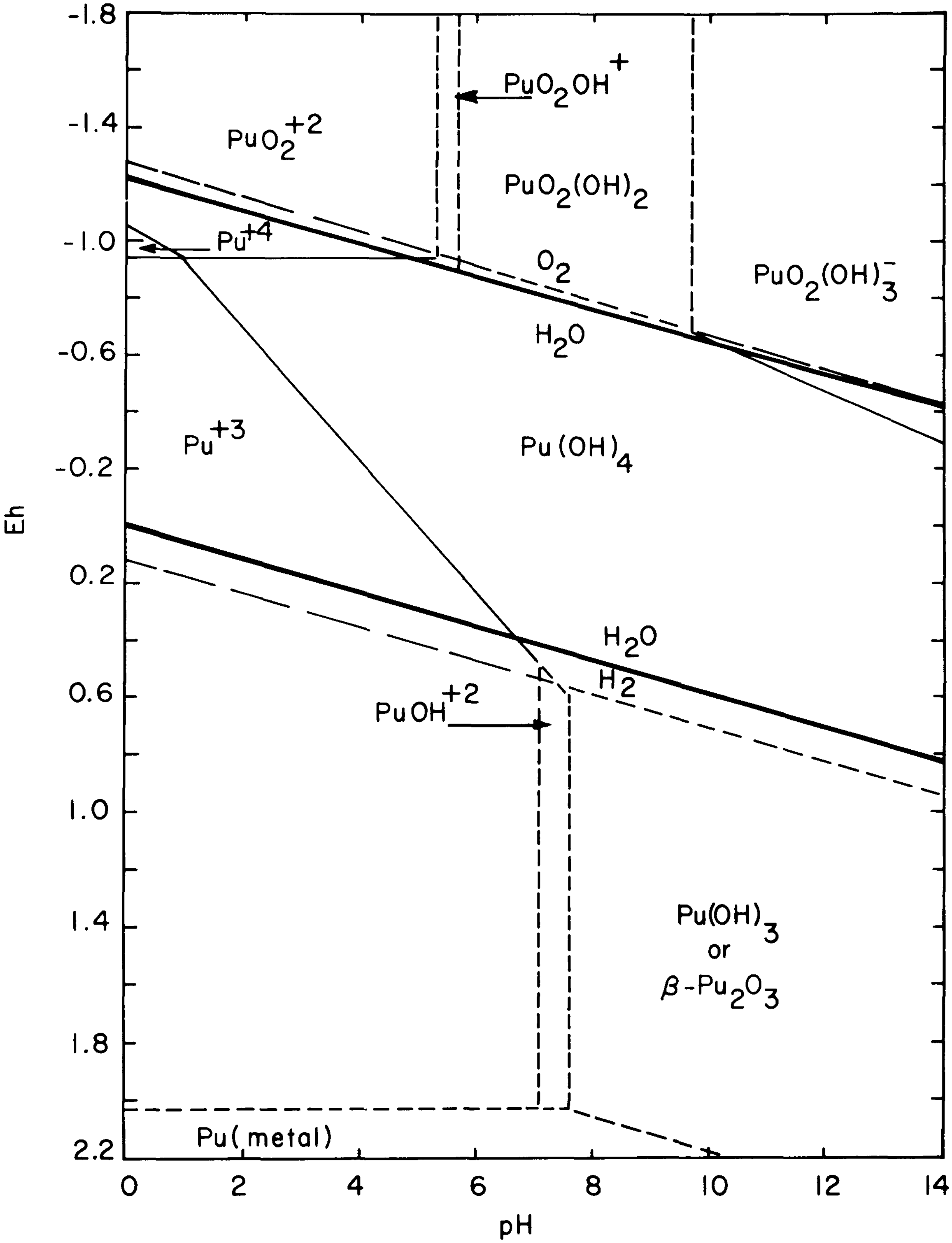

Figure 3: Stability relations of $Pu(OH)_4$ in equilibrium with water at 25°C and one atmosphere total pressure.

because of reaction rates, may control the soluble plutonium in soil/water environments. However, solubility studies of the oxides for environmental Eh and pH values and temperature have not been attempted to the author's knowledge. Such studies would contribute much to the understanding of the dissolution chemistry of plutonium in soil/water environments.

CONCENTRATIONS OF DISSOLVED PLUTONIUM

In the previous section the predominant species and those which probably control the dissolved plutonium were evaluated. However, the relevant questions of the relative proportion of the various dissolved species as well as the total maximum concentration attainable still remains.

Equilibrium relations can be written between the different aqueous species in order to determine the relative distribution of the various species. The predominant aqueous dissolved species can also be represented by a stability diagram in which only aqueous species are considered (Figure 4).

The concentration of any species can be calculated from its solubility relationship with PuO_2. It is assumed that PuO_2 controls the solubilities. The sum of all these species is equal to the total concentration of aqueous plutonium. The following equations were used in making calculations.

$$Pu^{+4}_{(aq)} + e = Pu^{+3}_{(aq)} \tag{14}$$

$$Pu^{+4}_{(aq)} + 2H_2O = PuO_2{}^{+}_{(aq)} + 4H^{+}_{(aq)} + e \tag{15}$$

$$Pu^{+4}_{(aq)} + 2H_2O = PuO_2{}^{+2}_{(aq)} + 4H^{+}_{(aq)} + 2e \tag{16}$$

$$PuOH^{+2}_{(aq)} + H^{+}_{(aq)} = Pu^{+4}_{(aq)} + H_2O + e \tag{17}$$

$$Pu^{+4}_{(aq)} + H_2O = PuOH^{+3}_{(aq)} + H^{+}_{(aq)} \tag{18}$$

$$Pu^{+4}_{(aq)} + 3H_2O = PuO_2OH^{+}_{(aq)} + 5H^{+}_{(aq)} + 2e \tag{19}$$

$$Pu^{+4}_{(aq)} + 5H_2O = PuO_2(OH)^{-}_{3(aq)} + 7H^{+} + 2e \tag{20}$$

$$Pu^{+4}_{(aq)} + 2H_2O = PuO_2 + 4H^{+}_{(aq)} \tag{21}$$

$$C_{Pu} \text{ total} = \sum_{i=1}^{n} (C_{Pu} \text{ species})_i$$

$$= C_{Pu^{+3}(aq)} + C_{PuO_2{}^{+}}\ C_{PuO_2{}^{+2}(aq)} + C_{PuOH^{+2}(aq)} + C_{PuOH^{+3}(aq)} + C_{PuO_2OH^{+}(aq)} + C_{PuO_2(OH)^{-}_{3}(aq)} + C_{Pu^{+4}(aq)}$$

where C represents concentration in moles/liter. The chemical activity of the various aqueous Pu species (shown in equations (14) through (20) can be evaluated in terms of the activity of the Pu^{+4} aqueous species. This can be accomplished with the aid of equations (2) through (8) and the free energy of formation data in Table I. The activity of the Pu^{+4} species can be determined from free energy data and equation (21). For the purposes of this study chemical concentration (moles/liter) is assumed to be the same as the chemical activity. More refined calculations can be made to better evaluate concentration with respect to activity if desired (Hem, 8). The results of the calculation of total concentration of dissolved plutonium as a function of Eh and pH, are given in Figure 5 where the dissolved plutonium is in equilibrium with PuO_2.

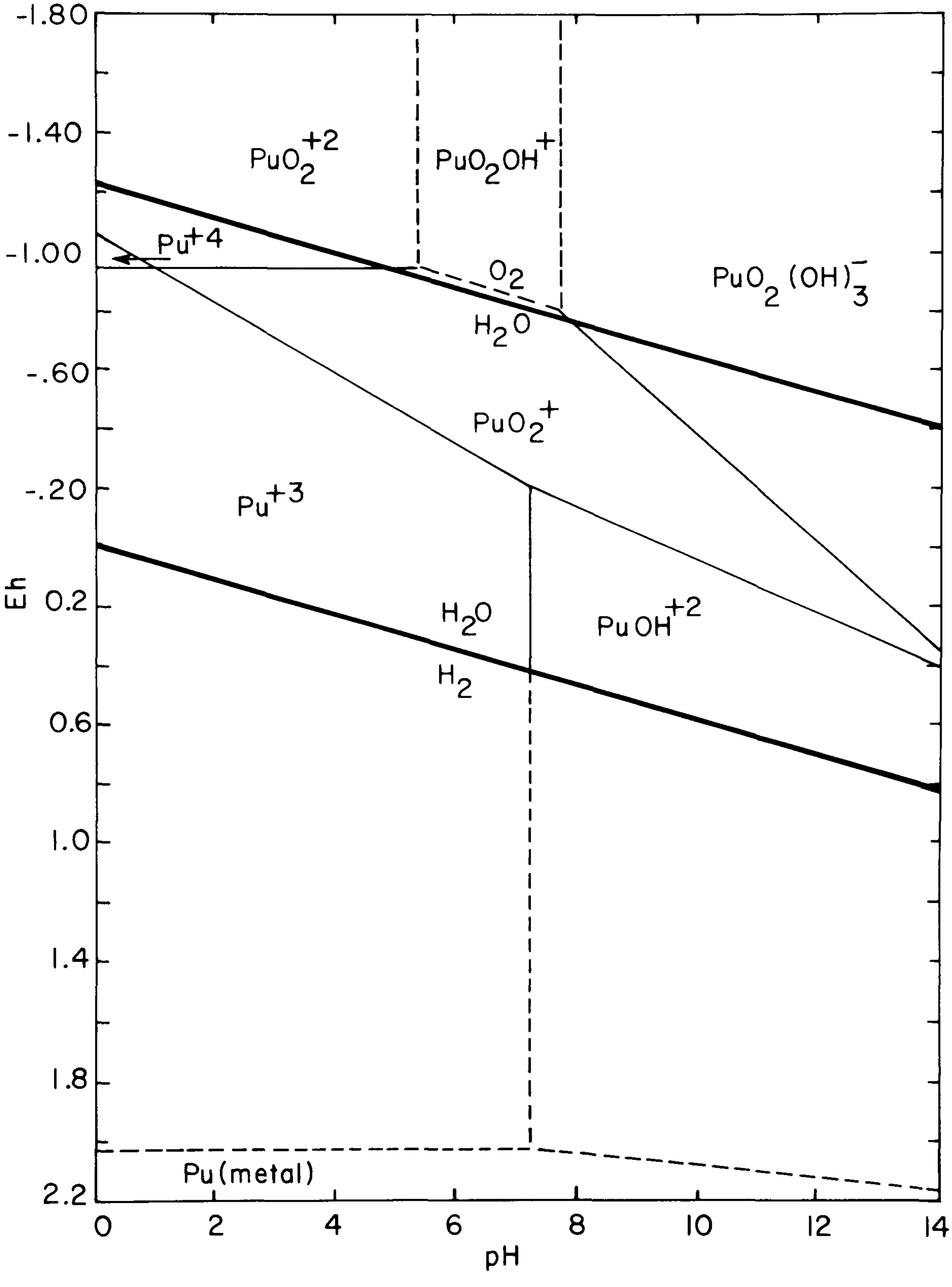

Figure 4: Stability relations of dissolved plutonium species in water at 25°C and one atmosphere total pressure.

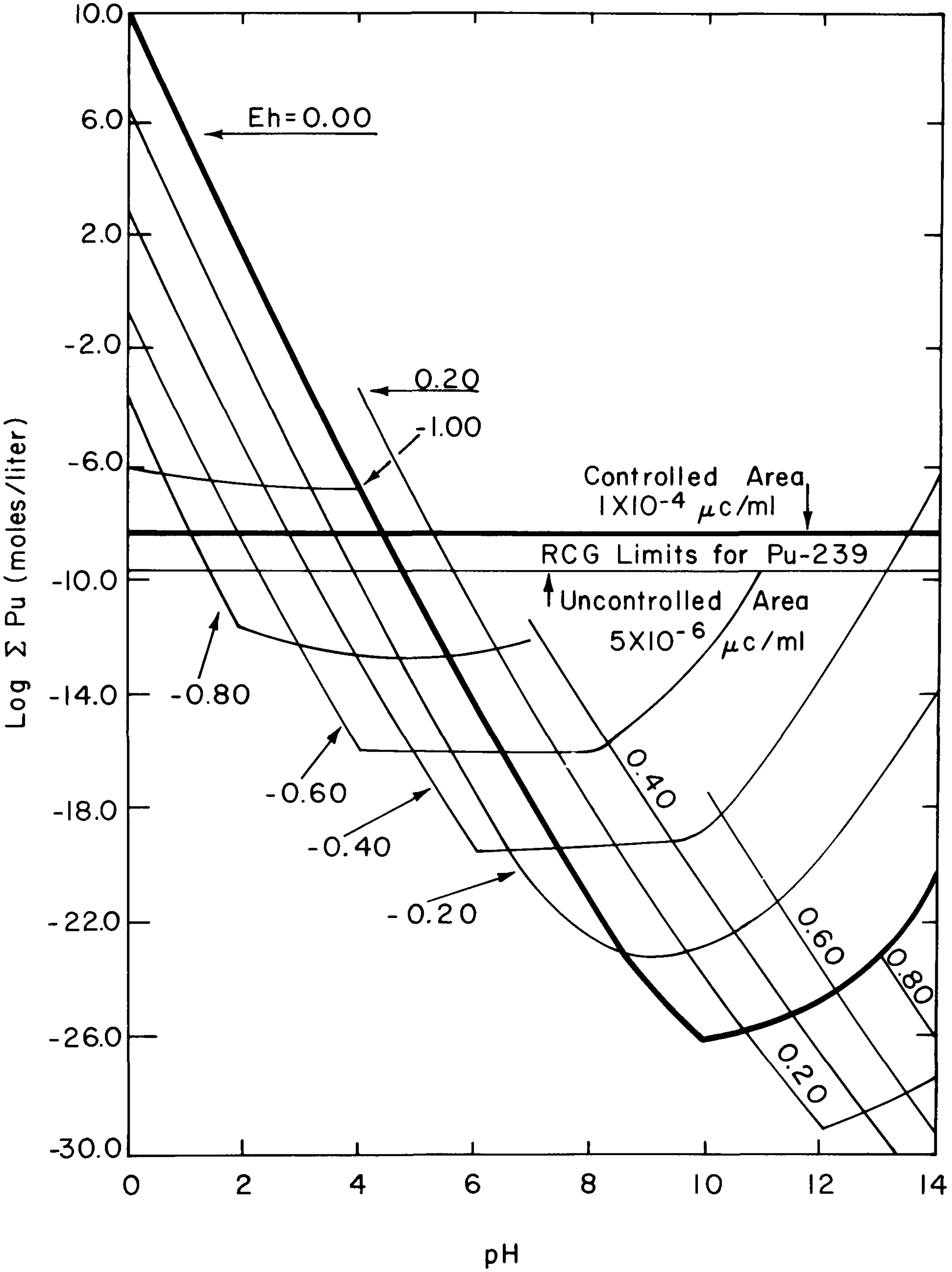

Figure 5: Total dissolved plutonium as a function of Eh, pH and PuO_2 solubility at 25°C and one atmosphere total pressure.

Figures 6 and 7 indicate total dissolved plutonium under conditions for which figures 3 and 4 were constructed, respectively. When the total concentration is limited by PuO_2 (Figure 1) dissolved plutonium in contact with the solid is below the RCG (Radiation Concentration Guide) for pH and Eh values normally found in the environment. The RCG value of plutonium-239 for controlled areas is 7.1 x 10^{-9} moles/liter (1 x 10^{-4} μCi/ml) and for uncontrolled areas or drinking water tolerance, the value is 3.5 x 10^{-10} moles/liter (5 x 10^{-6} μCi/ml). When $Pu(OH)_4$ limits dissolved plutonium, the total concentration may or may not exceed RCG limits over the ranges of Eh and pH values normally found in the environment. Values for Eh and pH need to be specified in order to evaluate whether RCG limits will be exceeded. Pu_2O_3 limits the total dissolved plutonium to lower concentrations for reduced conditions only (more positive than 0.00).

COMPLEXES OF PLUTONIUM

In addition to undergoing hydrolysis reactions, plutonium reacts with anions to form ion pairs (complexes), thus increasing the concentration of dissolved plutonium. Cleveland (1) in reviewing plutonium complexes gives the relative complexing ability of the various soluble plutonium species as

$$Pu^{+4} > Pu^{+3} > PuO_2^{+2} > PuO_2^{+}.$$

The relative complexing tendancy of monovalent anions with Pu^{+4} is given as

$$F^{-} > NO_3^{-} > ClO_4^{-}.$$

For the common divalent anions the ability to form complexes with Pu^{+4} is

$$CO_3^{-2} > SO_3^{-}\ C_2O_4^{-2} > SO_4^{-2}.$$

The divalent anions are complexed more strongly than are the monovalent anions. Other anions may complex with plutonium, for example, phosphate and peroxide anions and organic anions containing nitrogen, phosphorous or carbonyl groups. Some organic complexes such as EDTA, ethylenediamine tetraacetic acid, are also very stable (strongly complexed).

Moskvin and Gelman (9) list the relative tendancy of anion groups to form complexes with Pu^{+4} as

$$CO_3^{-2} > \gamma^{-4} > C_2O_4^{-2}.$$

where γ^{-4} denotes an anion of EDTA. To determine the contribution of the various complexes to the total concentration of dissolved plutonium, the concentrations of the various species of plutonium and the anions are needed in addition to the stability constants for the complexes. Although Pu^{+4} tends to complex most strongly with the various anions in solutions, the concentration is much lower than other plutonium species such as Pu^{+3} and PuO_2^{+2}, under environmental conditions, (see Figure 4). Therefore, a Pu^{+3} complex could possibly contribute more to the dissolved plutonium than a complex of Pu^{+4}, and consideration must also be given to the fact that more than one species of the anion may be present in solution. For example, EDTA (γ) exists in form as γ^{-4}, γ^{-3}, γ^{-3}, γ^{-2} and γ^{-}. Therefore the concentration of the anion species must either be calculated or determined.

Moskvin and Gelman (9) determined the stability constant for the $PuCO_3^{+2}$ complex as $10^{47.2}$. This value was based on a solubility product of $10^{-56.7}$ for $Pu(OH)_4$. In this present study $10^{-51.9}$ was used for the $Pu(OH)_4$ solubility product. After correcting for the difference in the solubility product the value of $10^{36.3}$ was obtained for the stability constant of $PuCO_3^{+2}$. It should be mentioned that the stability constant was experimentally determined in a solution having ionic strength near ten. It is doubtful that this value is

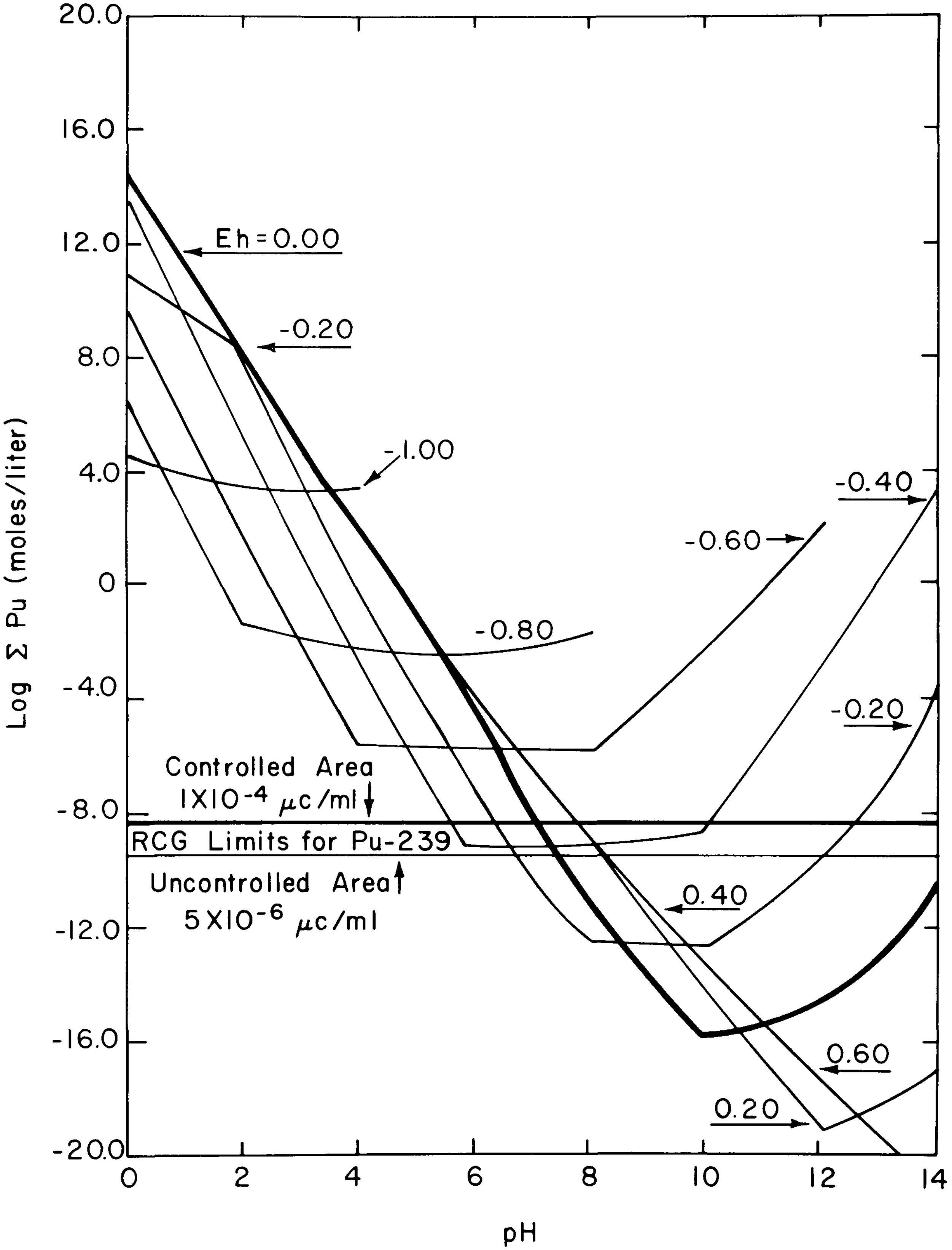

Figure 6: Total dissolved plutonium as a function of Eh, pH and $Pu(OH)_4$, Pu_2O_3 solubilities at 25°C and one atmosphere total pressure.

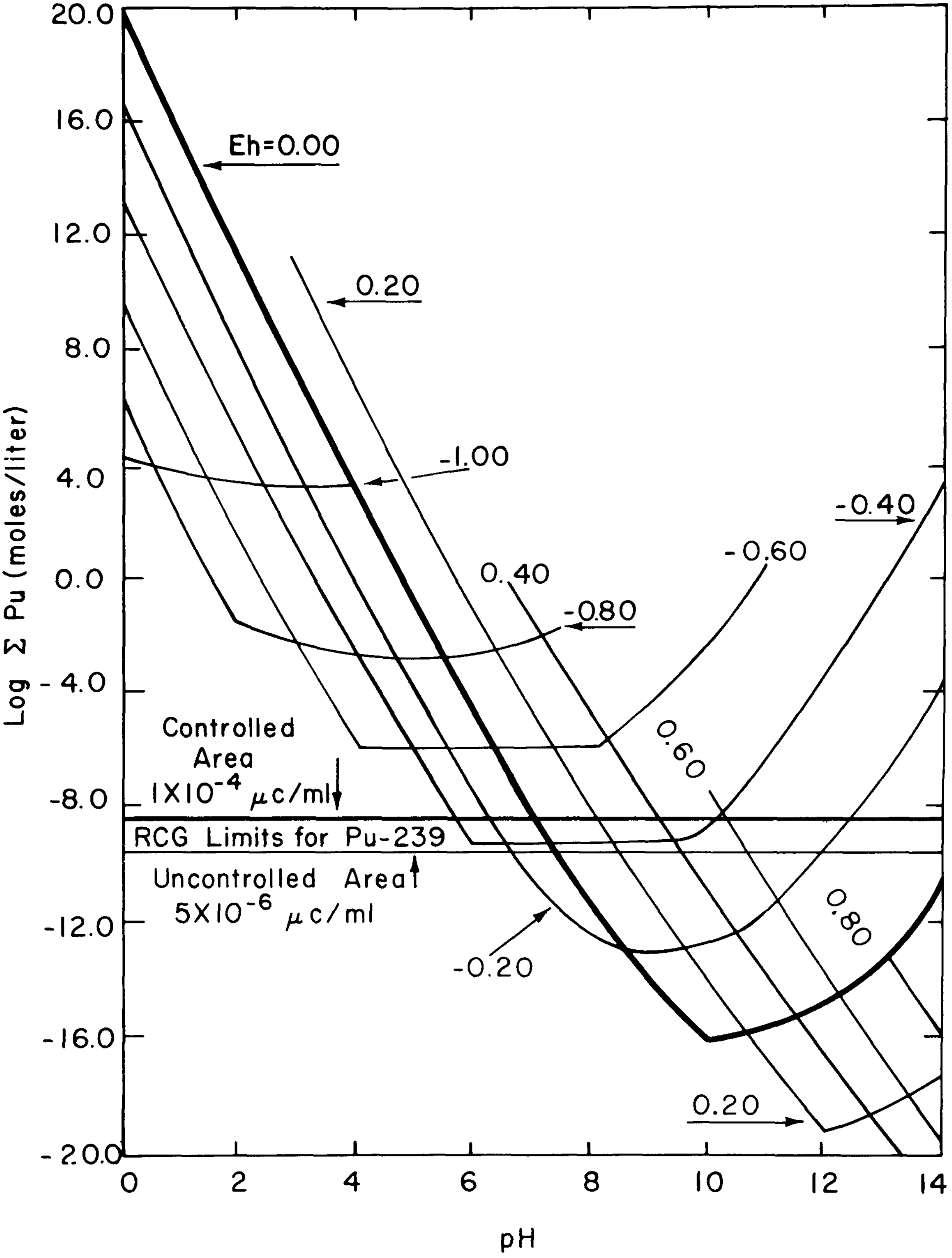

Figure 7: Total dissolved plutonium as a function of Eh, pH, and $Pu(OH)_4$ solubility at 25°C and one atmosphere total pressure.

applicable to conditions in the environment; however, an example of the effect of a complex on the total solubility of dissolved plutonium is given in Figure 8 using the above constant for the complex. The concentration of CO_3^{-2} is evaluated on the basis of an atmospheric partial pressure for CO_2 of $10^{-3.5}$. The concentration of CO_3^{-2} is pH dependent on the assumption that the soil/water environment is in equilibrium with the atmosphere.

Figure 8 indicates that the $PuCO_3^{+2}$ complex masks the effect of the oxidation potentials except for pH values greater than 10. Also the total concentration exceeds RCG limits for controlled areas at pH values lower than 8. If dissolved plutonium is controlled by $Pu(OH)_4$ then the total concentration would be approximately 10 orders of magnitude more than that shown in Figure 8. Differences in Eh would not affect the total concentration over the entire pH range. Such concentrations are never observed in nature to the author's knowledge. This observation could be the result of a number of factors, such as (1) non-equilibrium conditions (2) a more stable compound, such as PuO_2, controls the dissolved plutonium, and (3) the stability under environmental conditions would be helpful in determining the relationships between theoretical predictions, laboratory data obtained under specific conditions, and the actual solubility under environmental conditions.

SUMMARY AND CONCLUSIONS

The phase relations of the various plutonium species, both solid and aqueous, are evaluated as a function of hydrogen ion concentration (pH) and oxidation-reduction potential (Eh) for conditions normally found in soil/water environments. The evaluation is based on theoretical considerations and laboratory data obtained from the literature. Concentrations of dissolved plutonium are calculated from the evaluated phase relations. The significance of plutonium complexes on the concentration of dissolved plutonium is discussed and the effect of plutonium carbonate on dissolved plutonium evaluated.

PuO_2 was found to be the most stable phase and therefore, under equilibrium conditions should control or limit the concentration of dissolved plutonium. However, if the rate of reaction between PuO_2 and the solution in contact with it is extremely slow with respect to the other solid species of plutonium then the metastable phases $Pu(OH)_4$ and Pu_2O_3 would limit the concentration of dissolved plutonium. Pu_2O_3 is the metastable phase under reducing conditions and $Pu(OH)_4$ the metastable phase under oxidizing conditions. Consideration is given to the assumption that the rate of reaction between Pu_2O_3 and the solution is also extremely slow. Under these conditions $Pu(OH)_4$ is the solid that will limit the concentration of dissolved plutonium for conditions found in normal soil/water environments. The above results are based on the assumption that the amount of available plutonium is greater than the amount soluble in the volume of water under consideration.

The predominant dissolved plutonium species is dependent on specific environmental conditions. For example, the predominant species is Pu^{+3} for conditions in the region bounded by $-0.60 \leq Eh \leq 0.20$ at pH 4.0 to $-0.20 \leq Eh \leq 0.40$ at pH 7.0. The predominant species is the hydrolyzed form, $PuOH^{+2}$ for conditions in the region bounded by $-0.20 \leq Eh \leq 0.40$ at pH 7.0 to $0.10 \leq Eh \leq 0.60$ at pH 10. PuO_2^{+} exists as the predominant species for conditions in the region bounded by $-0.90 \leq Eh \leq 0.60$ at pH 4.0 to $-0.40 \leq Eh \leq 0.10$ at pH 10. The species $PuO_2(OH)_3^-$ could exist as the major species at high pH values (8 to 10) for oxidized conditions (Eh = -0.7 to -0.40). However, these conditions do not exist normally in the environment.

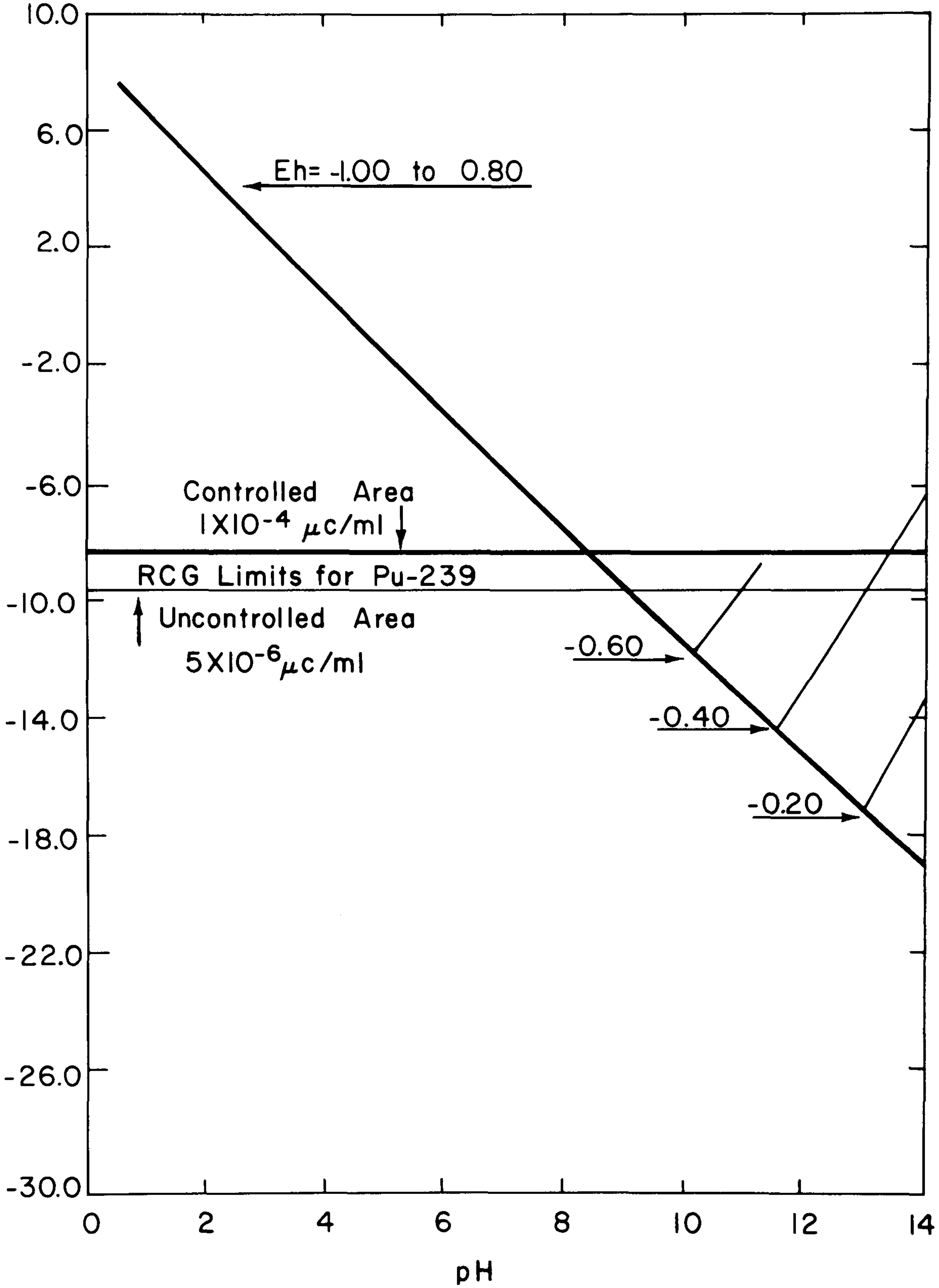

Figure 8: Total dissolved plutonium as a function of Eh, pH, PuO_2 solubility, and a CO_2 partial pressure of $10^{-3.5}$ at 25°C and one atmosphere total pressure.

Calculated concentrations of dissolved plutonium limited by or in equilibrium with PuO_2 are lower than Radiation Concentration Guide (RCG) values for pH and Eh values normally found in the environment. If dissolved plutonium is limited by Pu_2O_3 and/or $Pu(OH)_4$, then the concentrations are below RCG limits for values greater than -.45 and for values of pH between 7 and 10. For essentially all other Eh and pH values normally found in the environment, the concentration tends to be higher than RCG values.

A review of literature data indicates that Pu^{+4} complexes more strongly with other ions in solution than do the other dissolved plutonium species. Also, divalent anions tend to complex more strongly with Pu^{+4} than do the monovalent anions. Carbonate ($CO_3{}^{-2}$) and organic anions tend to form strong complexes with plutonium. Due to the concentration of these anions in soil/water environments $CO_3{}^{-2}$ would increase the solubility of plutonium most significantly. Assuming that the soil/water environment is in equilibrium with the atmosphere having a CO_2 content of $10^{-3.5}$ then the dissolved plutonium is independent of the Eh values normally found in the environment. For pH values above 8 the concentration is less than RCG values. At pH values lower than 8 the concentration exceed RCG values.

If $Pu(OH)_4$ limits the concentration of dissolved plutonium, then the concentration would be approximately 10 orders of magnitude higher. Such concentrations do not normally exist in the environment. This discrepancy may be due to: (1) non-eqilibrium conditions, (2) PuO_2, not $Pu(OH)_4$, limits the concentration, and/or (3) error in the stability constant of the carbonate complex. The value for carbonate stability constant was obtained at an ionic strength of about 10, which is not representative of normal environmental conditions. It is therefore, doubtful that the value for the stability constant can be applied to soil/water environment.

The conclusions drawn from this study are based on data obtained under conditions not normally found in the environment. Therefore, laboratory data obtained under environmental conditions and actual field measurements are needed to substantiate the above findings. However, these results do indicate the parameters that may affect the solubility of plutonium in soil/water environments and also indicate an approach which can be used in evaluating the behavior of plutonium in the environment.

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ADMINISTRATIVE PROCEDURES FOR CRITICALITY PREVENTION IN PLUTONIUM OPERATIONS

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ADMINISTRATIVE PROCEDURES FOR CRITICALITY PREVENTION IN PLUTONIUM OPERATIONS

INTRODUCTION

The current Hanford chemical separation facilities include a Purex type solvent extraction plant for nuclear fuels reprocessing, a radioactive waste processing plant, a uranium calcination plant (UO_3), a plutonium metal fabrication and scrap recovery plant, and associated laboratory facilities. These plants require criticality control in varying degrees depending upon the enrichment of the uranium and the amount of plutonium or other fissile materials that are processed.

This paper describes the administrative procedures used by the Atlantic Richfield Hanford Company (ARHCO) to prevent nuclear criticality during the processing of plutonium. It covers criticality prevention policies and operating instructions, technical criteria, criteria for specifications and auditing, unusual conditions, other controls that aid in criticality prevention, and built-in safety factors.

REQUIREMENTS AND POLICY

The Atomic Energy Commission (AEC) policy and requirements for non-reactor nuclear criticality safety are defined in AEC Manual 0530(1) and RL Chapter 05R4.(2) The policy requires that the "processing, transportation, storage, and use of fissile materials and the design, construction, and modification of nuclear facilities shall be performed in a manner that will reduce to the lowest practical level the probability and consequences of nuclear incidents."(2)

The ARHCO criticality prevention policy(3) applied to all activities involving fissile materials is stated as "ARHCO shall exercise control such that the probability of a criticality incident is held at the lowest practical level. Where practicable, the design of manufacturing and laboratory facilities and equipment handling fissile materials will include geometric limitations to minimize the probability of a criticality incident. In addition, there will be a criticality prevention system based on written specifications and implemented by written administrative procedures. The specifications will establish limits so that no single credible equipment failure or human error can cause a criticality incident."

CRITICALITY PREVENTION METHODS

Criticality prevention is normally accomplished by imposing limits on the size or shape of equipment, on the quantity or concentration of fissile material or both. The limits normally increase the complexity of the process and thereby increase the processing cost. Every reasonable precaution is taken to prevent criticality within processing facilities at costs that are reasonable relative to the risk involved.

In specifying limits on dimensions, concentrations, or masses, all credible conditions must be considered. The worst foreseeable combination of plutonium density, diluent composition and distribution, neutron reflection and interaction must be assumed. Some conditions may be assumed to be incredible if specifically excluded by technical or design considerations.

Geometrically favorable equipment(4) is subcritical by virtue of neutron leakage under the worst foreseeable process conditions. To be "geometrically favorable," the diameter of a cylinder is limited to a maximum value which results in a neutron multiplication factor (K_{eff}) no greater than 0.98 under the worst foreseeable conditions. Other configurations have lower limits for the maximum K_{eff}. An annular vessel may be considered to be a special form of a slab when due consideration is given to interaction across the inner area of the unit. Reliance on geometrically favorable equipment, sized for plutonium solutions, requires the specific exclusion of the accumulation of solids to concentrations beyond the design basis. Geometrically favorable equipment is used as much as possible in plutonium finishing operations. Solution systems lend themselves quite readily to this control.

If reliance is to be placed on a fixed poison incorporated in a structure, the minimum quantity and dispersion are specified so as to reduce the K_{eff} of the system to 0.98 for the worst foreseeable system to be contained. Periodic inspection (at least once per year) is specified to verify the quantity and location of the poison in the structure. Fixed poisons receive very limited use in ARHCO plutonium operations.

When it is not practical to prevent criticality by using favorable geometries or fixed poisons, reliance must be on mass and concentration limitations or soluble poisons. The process conditions controllable by operating personnel are limited to insure that neutron loss by leakage or absorption will prevent criticality even though any single credible error or omission has been committed. When practical, instruments and/or mechanical devices shall be provided to assist operating personnel to measure and control the process conditions within prescribed limits.

ADMINISTRATIVE CONTROLS

General requirements and limit values have been established at ARHCO for the administrative control of chemical processing plants. These controls include mass, concentration, and array limits and the use of soluble poisons.

The basic requirement in criticality safety is that the K_{eff} must always be less than one. The upper limits for K_{eff} are based on the degree of confidence in the accuracy of the factor determination for the worst foreseeable conditions. The degree of confidence is based on the experimental data available for similar systems. These are:

1. If reliable experimental data exist for closely similar systems and adequate calculation techniques exist for relatively small extrapolation, the limit is 0.98.

2. If limited experimental data exist for a similar system and relatively large but reasonable extrapolations are necessary, the upper limit is 0.95.

3. If no applicable experimental data is available such that calculations must be based on theory derived from experimental data, the upper limit is 0.90.

For control by mass limits, the quantity of plutonium in a given location is limited to an amount less than half that required to sustain a nuclear reaction under any credible conditions of geometry, moderation, and reflection.

For control by solution concentration limits, the concentration of plutonium dissolved or dispersed in another medium is limited such that neutron absorption in the diluent prevents criticality. The concentration of plutonium in solution shall not be greater than 50 percent of the minimum critical concentration in that vessel. In addition, there is specified for the vessel a mass limit such that a specified K_{eff} is not exceeded under the worst conditions attainable by the inadvertent concentration of plutonium, e.g., precipitation, evaporation, etc.

For control by array concentration limits, the dispersal in space of discrete accumulations of plutonium is controlled with respect to geometry and distance. A proper array insures that nuclear reactivity of any single subcritical unit is not significantly increased by the interaction of neutrons with adjacent units.

For control by soluble poisons, neutron absorbing materials are put in solution with the plutonium in sufficient concentration to prevent criticality under all foreseeable process conditions. In the Plutonium Finishing Section, soluble poisons are not used as primary controls because of the lack of massive shielding and possible loss of poison.

There is a difference in the "number of errors" criteria based on the presence or lack of massive shielding for personnel. Most of ARHCO's plutonium processing facilities are contact maintained and lack massive shielding and require three independent errors for criticality.

Additional Safety Factors

Many constituents of product and particularly scrap are neutron poisons or absorbers to varying degrees. However, because it cannot be assured that these poisons or absorbers are always present, or to what degree, they are seldom taken into account when establishing limits for criticality prevention. The solvent extraction scrap recovery process is a nitrate system and nitrogen has a poisoning effect; plutonium-240 has a poisoning effect in thermal systems. The chlorine in the solvent extraction diluent, CCl_4, has a poisoning effect and uranium-238 is a neutron absorber. These and other poisons or absorbers provide some additional safety margin.

Gamma and neutron radiations are used to determine the quantity of plutonium in packages, particularly for wastes. The calibrations of these package counters are based on essentially clean plutonium sources. If the scrap plutonium should contain some small amounts of fission products, the tendency is for the package counter to count more gamma and/or neutrons than are due to the presence of plutonium, indicating more plutonium than there actually is. This factor works in the direction of increased safety margin.

CRITERIA FOR SPECIFICATIONS

Processing specifications, operating standards, operating procedures, and criticality prevention specifications are four separate written instruments with which operating personnel must be thoroughly familiar. Efforts are made to simplify the controls to minimize the complexity of routine operations. Qualities of good criticality prevention specifications are considered to be the following:

1. Easy to understand,

2. Easy to comply with,

3. As brief as possible,

4. As few restrictions as possible,

5. Consistent limits, and

6. Easy to audit.

Compliance with specifications is more apt to be assured when specifications are brief, with minimal and consistent limits, and when work is the least restricted.

Procedure for Producing Approved Specifications

The need for an approved criticality prevention specification is usually identified by the operation concerned or the associated Operations Support Engineering (OSE) group. The engineering group (OSE) formulates, drafts, reviews, and issues the specification; and criticality specialists [Research and Development (R & D)] review and approve the technical content. The engineering department managers (OSE and R & D) approve and the operations manager accepts the specification. Any revisions require the same review and approvals.

Administration of Specifications

Safe work practices in industry are the responsibility of line management. This precept applies equally to criticality safety in plutonium processing where management supplies the proper facilities, equipment, and personnel training.

Necessary standard operating procedures are based on process specifications and standards, criticality prevention specifications, and standard safety rules. The criticality prevention program is then carried to the process operator through the "Standard Operating Procedure," proper posting, and training.

Auditing for adherence to procedures and specifications is accomplished in several ways. The day to day audits by line management and process engineers indicate the effectiveness of the program and point out areas where retraining is needed or procedure changes are necessary. Quarterly audits by experts from outside the Manufacturing Department assess the criticality prevention performance. An annual audit by non-ARHCO criticality experts also provides an in-depth review of policy and procedures.

A facilities change notice system for equipment and facilities changes provides a review for possible effects on criticality prevention before the work is done.

DETECTION OF UNUSUAL CONDITIONS

In a fuels reprocessing plant, unusual conditions are considered as those other than standard conditions of operation. However, in a scrap recovery operation, there are so many variables in the feed that unusual conditions might be considered normal. Equipment wears or otherwise deteriorates, small amounts of some contaminants affect dissolution rates or upset solvent extraction or ion exchange columns. Plutonium may build up from incomplete dissolution, settling out as finely divided solids in packed columns and still tanks, or by liquid entrainment in vapor or gaseous streams.

In order to prevent criticality, it is necessary to detect these unusual conditions before accumulations are excessive. The most reliable method to detect accumulations is visual inspection, and this can be accomplished in some batch dissolvers. The next most reliable method for detecting plutonium build-up is by radiation detection, and this method is extensively applied using both fixed and portable instruments. Perhaps the least reliable method is by plutonium accountability, especially in a scrap recovery plant where the accounting is rather difficult because of input uncertainties. Periodic flushing is required when the material unaccounted for exceeds a set amount.

An example of an unusual condition was the detection of a plutonium build-up in two nongeometrically favorable waste tanks. The two tanks were equipped with single fixed neutron monitors located under the middle of the tank bottoms; either the monitors did not detect the build-ups or they were so gradual that the reading was improperly interpreted. The plutonium accumulation only slightly exceeded the specification limits (400 grams) when detected on a routine 6-month survey with a portable neutron detection instrument. The facility using the two tanks was shut down immediately. The incident was investigated and agitation and detection were improved after the solids were removed and before the tanks were used again.

Package counting is used to aid in accountability control and is the only method available for most plutonium wastes. Plutonium-bearing scrap shipped to ARHCO for storage and/or processing from many other AEC sites necessitated improvement in intersite accountability. The NaI counter initially developed as a portable counter was adapted to measure a single level of gamma energy from a package. Further refinements included incorporation of a rotating table into the counting system to provide a more complete analysis of a package. This new plant application of the NaI gamma counter took the place of the early neutron counter. A new neutron package counter is being added to the gamma counter as no one system is the answer to all problems.

A lithium drifted germanium [Ge(Li)] detector with a multichannel analyzer is available to aid in problem solving by virtue of its fine discrimination for gamma energies. By use of the Ge(Li) detector, various isotopic content levels or sources can be identified. Several isotopes have been detected where no indication had been given in the description of the material received.

Other Controls Which Aid in Criticality Prevention

There are other controls exercised in the handling of plutonium that contribute to criticality prevention in their application. The extremely toxic nature and high value of plutonium are reasons for careful handling. Strenuous practices for the control, containment, and accountability have aided the criticality prevention program. The control of contamination spreads reveals the degree of adherence to procedures. Material balances of processes are mandatory at frequent intervals for precious metals control.

There is hardly a processing or handling operation that doesn't produce some kind of scrap or waste. Keeping the scrap and waste picked up and out of the way is an integral part of the job. Good housekeeping and elimination of fire hazards contribute to good criticality prevention practices. It is a necessary part of the total plutonium handling environment.

The relatively large inventories of product and scrap which have to be stored make it necessary to maintain a good inventory system so specific scrap and product items may be withdrawn from storage at the proper time for processing. The variations in scrap matrix, plutonium isotopic content, presence of contaminants, density, and heterogeneity result in many categories.

Requirements for security and safeguards against diversion cause storage facilities to be kept under lock and key so that all movements to and from storage are under close supervisory scrutiny. This arrangement keeps access limited to authorized and trained personnel.

Since all these administrative controls must be exercised, it is important that the maximum use is made of each as they contribute to criticality prevention.

CONCLUSION

Increasing amounts of plutonium are being handled by an increasing number of facilities throughout the country. Increased amounts and varieties of plutonium materials at ARHCO have served to indicate new problem areas that can be encountered by others. Scrap materials are of particular concern for all aspects of plutonium handling, as well as for criticality prevention. To effectively administer a plutonium processing facility, criticality prevention must be integrated into the facilities, procedures, and training.

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SAFETY CRITERIA FOR THE DESIGN OF FACILITIES PROCESSING PLUTONIUM

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April 16, 1971

SAFETY CRITERIA FOR THE DESIGN OF FACILITIES PROCESSING PLUTONIUM

The subject of my talk today is "Safety Criteria For The Design Of Facilities Processing Plutonium." You may well ask why at this point in time do we need to issue such criteria? I believe the answer to that question is that it is a matter of history --- the time has now arrived.

The earliest plutonium processing facilities were built during and immediately after World War II. At that time even the ordinary industrial codes for the protection of facilities had been suspended or modified due to wartime shortages. The DP West site at Los Alamos is an example of a design originating at that time, and it certainly does not meet present day ideas for the design of a plutonium handling facility. In the early 1950's the AEC developed the concept of protected construction. Class I protected construction is a high quality construction designed to withstand a certain degree of enemy attack. The technology of nuclear weapons soon overtook the ideas behind this type of construction, and since it was costly, it was essentially abandoned after a few years. The facilities built of this type of construction are principally the Savannah River Plant, some here at Rocky Flats, and the AEC's Germantown Headquarters. It must be admitted that this produces a superior type of construction and does have many of the attributes found in the present-day concept of a confinement facility. When this concept was abandoned, the follow-on facilities were built primarily to the industrial-type building codes. The AEC has always specified in its health and safety codes and standards that these are our minimum standards of construction. The principle behind this concept is that the AEC should build to the standards used by the better type of industry. This type of construction is seen here at Rocky Flats in the 776-777 building and others.

In the early 1960's an incident occurred at ORNL which caused the release of a small amount of plutonium. Although the material was confined to the site, the incident caused ORNL management to review their design situation and develop what is called the "double barrier concept." That is, there will be two barriers in the path of any possible release of highly hazardous materials from the facility. In general, these two barriers include tandem high efficiency particulate air filters.

In the middle 1960's it was decided that the laboratory operations at Mound did not have the capacity for the production of quantities of plutonium-238 that were desired from that facility. In establishing the design for a new plutonium-238 facility, the Albuquerque Office developed the concept that the building should have a reasonable degree of engineered accident resistance. The thought was that the building should be able to remain standing in the event of an internal accident --- such as a fire or explosion --- and thus be able to continue to confine any radioactivity. This became the Albuquerque standard when the construction began on the new weapons facilities --- such as Building 707 here at Rocky Flats.

In 1969 we had the fire here in Building 776-777 at Rocky Flats. It was found that a building constructed to the industrial building codes was not too bad a structure if the filters remained intact and the ventilating system was able to continue operating, and if the incident could be brought under control before the structure failed. While the building held up and the ventilating system did succeed in controlling any releases from that fire, it was recognized that the potential for release of plutonium to the environment did exist, and even more so, it was recognized that very high losses, both dollar-wise and programmatic-wise, could be sustained in an accident involving an ordinary industrial-type building such as this.

With this background we can see that it is timely to consider the criteria that should go into the design of a plutonium processing plant.

OBJECTIVES

In developing criteria it is necessary to first consider the basic objectives that the AEC hopes to achieve for its own operations. These objectives are:

1. Public Safety - This involves environmental control during normal operations and the prevention of accidental conditions which can produce adverse effects on the public.

2. Employee Safety - For this we wish to control overexposures to hazardous materials during normal operations, and we wish to provide for life safety under accident conditions.

3. Program Continuity - For this we wish to make provisions to limit adverse programmatic impact due to accidents.

4. Property Loss - Here we want to limit the financial loss due to property damage accidents.

CONDITIONS TO BE CONSIDERED

There are two types of conditions which must be considered in making the evaluation as to whether the facility design meets the desired objectives. We must first consider the normal operations of the plant. Under this condition it is necessary to be assured that the normal effluents from the plant do not impinge adversely on the environment. I am sure all of you are aware of the environmental emphasis now being given to new facilities. One of the problems with a plant that handles long-lived radionuclides is the matter of very, very low levels of day-in and day-out releases which may pass through filters or other paths to the environment. Although these cannot generally be measured in terms of air concentration, after many years of operation small quantities of the material can be detected outside of the plant. This has happened at Rocky Flats and at other places as we have heard described at sessions during the last few days. It is a problem that we cannot dismiss. It would be nice to set zero as a design objective for such releases. Perhaps this is not an obtainable goal, but it certainly is something that we need to keep in mind.

In addition to protecting the environment we must also consider the exposure of the employees in the plant to the day-to-day working conditions. In general, protection against plant hazards is achieved by compliance with the codes and standards which have been developed by the various standard-making groups such as ANSI, the American Conference of Governmental Industrial Hygienists, the National Fire Protection Association, and many others. Compliance with these codes will assure proper guarding of machines, proper lighting, proper ventilation to protect against exposure to toxic materials, and so forth. Of particular concern in plutonium facilities is exposure to ionizing radiation. As a design objective, it was suggested that a radiation level not exceeding .25 mr per hour should be the goal. We are sure that this is not obtainable in all locations; however, in developing the design, those locations which will not achieve this level should be examined and identified so that any necessary administrative control can be put into effect. In addition to the whole-body exposure to ionizing radiation, the workers who handle plutonium will also have a higher exposure to the forearms and fingers. An exposure limited to 30 rem per year has been recommended. (NCRP has recently reduced the limits for exposure of the forearms to 30 rem per year and raised finger limits to 75 rem per year). This will mean that where workers are continuously handling plutonium and where the gamma and neutron levels are appreciable, consideration of remote operations may be necessary.

Secondly, accident conditions must be considered in reviewing the design objectives. These accident conditions are of two types. There are those that may be plant associated such as fire, explosion, nuclear criticality, mechanical failure, etc., and there are natural phenomena such as earthquake, tornadoes, floods, wind, temperature extremes, etc. Under the accident conditions we need to assure ourselves that the objectives stated earlier have been met.

CURRENT STATUS OF CRITERIA DEVELOPMENT

I have attached the first draft of the safety criteria that was prepared by the Division of Operational Safety. This was submitted to the Albuquerque Office for particular application to a proposed new Plutonium Recovery Facility at Rocky Flats and replacement of DP West at Los Alamos. Albuquerque and its contractors have reviewed this document and submitted their comments to Headquarters where they are now undergoing more thorough review to develop a second draft. We would expect to distribute our second draft to all of the Field Offices for review and comment. Finally, we would expect the Division of Construction to take these criteria and incorporate them, with additional details and design specifications, in AEC Manual Chapter 6301 "Design Criteria Manual."

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Attachment:
Safety Criteria (Draft)

DRAFT
Division of Operational Safety
December 9, 1970

ITEMS TO BE CONSIDERED IN THE DEVELOPMENT OF GENERAL SAFETY CRITERIA FOR THE DESIGN OF FACILITIES PROCESSING PLUTONIUM

For Application to DP West and Rocky Flats Plutonium Recovery Facilities to Supplement the Guidance Now Provided in the 0500 and 6000 Manual Series

I. SITE STUDIES

Site studies (see also MC 6203) necessary to provide the technical bases (normal and accident) for location, engineering design, and operation of the facility should consider the following:

- Location
  - a. Population distribution (isolation from populated areas).
  - b. Other facilities and operations (isolation from external hazards--fire, explosions, etc.).
- Meteorology (for wind loads and diffusion parameters).
- Hydrology (ground water levels, flooding, etc.).
- Geology and soil characteristics (regional and local geology, etc.).
- Seismology.

II. PLANT FEATURES

A. Structural Design

General

Structural design, loading combinations, and construction of critical safety and fire protection features should, as a minimum, be in accordance with the latest edition of pertinent nationally recognized codes. Where local codes or regulations are more stringent than the nationally recognized codes, they should be followed.

Wind and Storm Resistance

The structure should be designed to withstand, with a margin of safety, wind velocities greater than the maximum observed at the site (reference ANSI Standard A-58.1).

The effects of tornadoes should be considered in design. In the event of a tornado, noncritical parts of the facility may be damaged; but design should assure that control and confinement of radioactive material is maintained.

Seismic Design Resistance

The National Oceanic and Atmospheric Administration should be consulted for seismic design parameters. The seismic design of the following items should be established on a dynamic rather than a static basis:

- Main structures.
- Fire protection systems.
- Ventilation systems.
- Confinement piping and equipment.
- Critical utilities, instrumentation, monitors, and alarms.

Other Natural Phenomena

Design loads and considerations for other natural phenomena (snow, flooding, etc.) should provide a conservative margin of safety greater than the maximum historical levels recorded for the site.

Fire Resistance

The structure should be designed with sufficient fire resistance, including a margin of safety, so that it will remain standing and continue to act as a confinement (or containment) structure during a credible fire that can be postulated for the facility.

Noncombustible and heat resistant materials should be used for construction wherever practicable for the facility, equipment, glove boxes, and conveyor lines.

Explosion - Internal Pressurization

To establish the explosion and/or pressurization design, an analysis of all credible explosion and pressurization events must be made. The analysis should consider the process, the size of equipment, and the amounts of chemicals and/or gases that could be involved.

B. Plant Layout

Design of the layout of the facility, including partitions, should cover:

- Process flow.
- Contamination control (localize effects).
- Fire control (localize effects).
- Compartmentalization.
- Personnel exits (NFPA 101).
- Airlocks to assist in ventilation control.

C. Water - Leakage Collection

Collection systems should be provided for water runoff from sprinklers and process leakage. Nuclear safety, confinement, and retrievability of liquids should be considered in the design of collection systems.

D. Maintenance

Provisions should be made for maintenance on contaminated items. These provisions should include special areas for hot maintenance and contamination control.

E. Decontamination

Design should provide for ease of decontamination, both for routine maintenance and access and for the overall facility in the event of an internal accident or decommissioning.

No new facility should be built unless plans are made for eventual decontamination and dismantling; design should consider this eventuality.

F. Provisions for Additional Safety Features

Design should permit future additions of safety features, such as complete inerting, without major structural changes.

III. EFFLUENT CONTROL AND MONITORING

A. Effluent Standard

In accordance with the General Manager's memorandum of November 17, 1970, radioactive effluents shall be as low as economically and technically practical. For new facilities, design should consider total confinement to the maximum extent practicable. Operation should not result in a "buildup" of radioactive material in the environment around the plant.

B. Effluent Monitoring

All effluents must be monitored to determine and record activity releases under normal and accident conditions. Backup and fail-safe* sampling and instrumentation systems should be used. To achieve optimum accuracy and reliability, the design should be consistent with state of the art technology.

Automatic alarms should be provided to alert operators of unexpected increases in activity levels of effluents.

C. Environmental Monitoring

A preoperational environmental monitoring program should be conducted to determine "background levels" of radioactivity in the environment. Appropriate plans and specifications for equipment to be used in postoperational environmental monitoring program should be established during the design phase of the program. The design of such equipment should be consistent with state of the art technology.

IV. VENTILATION SYSTEMS

A. General

Ventilation systems should be designed to confine radioactive material under normal, abnormal, and accident conditions, to provide sufficient dilution, and to lower radioactive discharges to the maximum extent

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*Operable in all emergency conditions.

practicable. Ventilation airflow should be from nonradioactive zones to moderately radioactive zones to highly radioactive zones.

B. Process - Building Ventilation

Separate process and building ventilation systems may be required to maintain radioactive effluents as low as practicable. Closed recycle ventilation systems for process operations should be considered.

C. Design of Ventilation Systems Should Assure:

- Continuity of operation, i.e., standby blowers and sources of emergency power.
- Acceptable cleanup (filtration, scrubbers, etc.) of ventilation exhaust. (Exhaust recycle where necessary.)
- Capability for *in situ* periodic testing and surveillance of exhaust cleanup provisions (filters, scrubbers, etc.).
- Means for necessary replacement of the exhaust cleanup provisions (filters, etc.).
- Means to prevent or minimize ventilation flow reversals.
- Alarms to alert people of flow reversals and of other malfunctions.
- Means for *in situ* testing of monitor and control systems.
- Sufficient flow capacity and/or controls to assure confinement if gloves rupture, process cell is opened, etc.
- Means for facilitating manual fire control actions.
- Means for maintaining confinement under credible accident conditions (sand filters, coolers, etc.).

V. RADIATION PROTECTION

A. Shielding Design

Personnel exposures must be maintained as low as practicable; levels less than one-tenth of the permissible dose limits given in AEC Manual Chapter 0524 are desirable. For shielding design purposes, the following are upper limit reference exposure rates for dose to the whole body:

- Radiation dose rates in routinely occupied areas (occupied greater than 2 hours per day) should be less than 0.25 mrem/hr.
- Radiation dose rates in nonroutinely occupied work locations (occupied less than 2 hours per day) should be maintained as low as practicable. Dose rates not in excess of 2.5 mrem/hr are desirable.
- Credible accidents should be considered in shielding design along with evacuation times and other safety factors to assure that personnel exposures during accidents are minimized to the extent practicable.

B. Air Monitoring Systems

Sensitive automatic alarming devices (CAMs) should be installed at work areas where airborne contamination can be expected. The system should be fail-safe* and contain an easily visible radioactivity readout and flow rate indicator and should be designed such as to provide a meaningful relationship between the monitoring alarm setpoint and pertinent exposure guides.

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*Operable in all emergency conditions.

C. Radiation Monitoring Systems

Design should provide a fail-safe area radiation alarm instrumentation to alert plant personnel of unexpected increases in radiation exposure levels.

D. Criticality Alarm Systems

Design of criticality alarm systems should meet, as a minimum, ANSI Standard N2.3-1967 Immediate Evacuation Signal.

E. Nuclear Accident Dosimeter

Design should include nuclear accident dosimeters. Performance parameters and placement should be consistent with the requirements of MC 0545.

F. Separation of Radiation Protection and Effluent Monitoring Systems from Process Control Systems

Protection and monitoring systems should be separated from control instrumentation systems to the extent that failure or removal from service of any control system, component, or channel leaves the protective and monitoring systems operable.

G. Provisions for Decontamination of Personnel

Design should provide provisions for decontamination of personnel and health protection activities reasonably close to sources of contamination. Provisions should also be provided for highly contaminated emergency cases.

VI. CONTAINMENT AND CONFINEMENT FEATURES

A. Glove Boxes and Conveyor Lines

Design should provide:

- Automatic fire suppression.

- Manual supplemental firefighting capabilities where fire potential exists.
- Inert atmosphere where flammable material is processed or where process atmosphere is recycled.
- Noncombustible or fire resistant coatings, shielding, and materials of construction.
- Breaks or partitions to localize fire and contamination effects to discrete areas.
- Fire and heat detectors and alarms.
- Engineered safety features to minimize the consequences of potential explosions.
- Decontamination ease; rough surfaces, square corners, cracks or crevices, and absorbent materials should be avoided.
- Remoting should be considered where it is anticipated that exposures to hands and forearms would approach 30 and 15 rem/yr., respectively.

The information in TID 16020 should be considered in glove box design.

B. Structures .

- Internal rooms and compartments should be designed to provide a secondary confinement and/or containment barrier.
- The main external structure should be designed to provide a final confinement and/or containment barrier.

VII. FIRE PROTECTION

- Automatic fire suppression systems required in fire areas where vital operations are conducted or the maximum probable loss is more than $1,000,000.

- Fire walls to limit fire areas to not more than those specified in AEC Appendix 6301.
- Limit fire areas so that maximum credible loss (assuming suppression systems fail and no manual firefighting actions) is not more than $25,000,000.

VIII. PROCESSES AND WASTE HANDLING

Design of process and waste handling systems should provide:

- Isolation between incompatible chemicals, materials, and processes.
- Safe relief for pressurized systems.
- Nuclear safety control.
- Capability to be maintained properly and safely.
- Materials of construction compatible with process chemicals to minimize corrosion.
- Ease of decontamination.
- Capability for safe shutdown if abnormalities occur.
- Process instrumentation, control indicators, and alarms to monitor and maintain control over the process and detect and alarm for abnormal and accident conditions.
- Limitation on explosive or flammable chemicals.

IX. STORAGE FACILITIES

Criteria for plutonium storage facilities are given in the General Manager's memorandum of July 7, 1970.

X. NUCLEAR CRITICALITY

Design of nuclear criticality control provisions, including equipment and procedures, should meet, as a minimum, the requirements of MC 0530. Design should further assure that fissile material will not be displaced to form a potential critical mass in the event of an internal or external accident.

XI. UTILITIES (ELECTRICITY, WATER, STEAM, COMPRESSED AIR, FUEL, ETC.)

A. General

Design capacity of each utility should consider service demands for normal, abnormal, and accident conditions.

B. Fire Protection

Water supply must be adequate for credible fire (at least 4 hours) and must be reliable (two sources).

C. Emergency Utilities

Alternate (emergency) sources of utilities should be provided to maintain with adequate capacity the continuity of critical services that cannot be interrupted.

Reliable emergency power should be provided to critical instruments, controls, and other necessary emergency utilities provided for process equipment and other requirements in sufficient capacity and sufficiently rapidly (automatic) to allow, at least, operations to be shut down safely and maintained safely in the shutdown conditions with all essential safety features functioning.

XII. EMERGENCY PLANNING

Emergency requirements should be considered early in design to assure that facility features provide for ease of evacuation and other emergency requirements (reference MC 0544).

XIII. QUALITY ASSURANCE PROGRAM

A quality assurance program should be established, covering the life of the project from conception through operation, to assure that structures, components, systems, and facilities are designed, developed, manufactured, constructed, operated, and maintained in compliance with established engineering criteria.

RDT Standard RDT F2-2T provides guidance on the essential elements of a quality assurance program.

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