FUEL ELEMENT DECOMPOSITION PRODUCTS

G. W. Parker, G. E. Creek, and W. J. Martin
Oak Ridge National Laboratory

INTRODUCTION

In assessing the potential environmental fission product release hazard to the public created by the operation of large nuclear reactors, an increasing fraction of the analysis is being based on data (1) resulting from the examination of accidental releases (2) and from a large number of laboratory experiments (3, 4, 5). A significant uncertainty in the assessment of reactor hazards, however, arises from the lack of data on the production and dispersal of air-borne solid particles during an accident. A large part of such particles, produced as smoke and metal fumes, will presumably be retained in the containment shell provided for most power reactors. Nevertheless, dangerous amounts of these highly radioactive materials are expected to eventually find their way through small defects in the containment shells which generally leak at a rate of 0.1 to 0.5 volume percent per day of the total gas volume in the shell.

One estimate indicates that as much as 20% of the total fission product inventory of a reactor may be involved in an uncontained release cloud which could conceivably lead to radiation casualties from incident radiation as well as from inhaled and ingested gaseous and particulate radioactivity.

The deposition rate or plate-out of released activities, which determines the extent of the area affected by a reactor accident, is a function of various parameters involving weather conditions and the nature of the accident producing the particles. The size and other physical and chemical properties of the released particles have important effects on their deposition rate.

REACTOR FUEL TYPES AND THEIR RELATED HAZARDS

Metallic Uranium – Uranium metal, usually with bonded aluminum or magnesium cladding, constitutes the bulk of the fuel employed in plutonium-producing reactors and in the large British gas-cooled power reactors. The commonest type of failure in reactors employing uranium fuel occurs in the air-cooled, low-temperature graphite piles when a burst slug oxidizes and releases volatile fission products and small air-borne uranium oxide particles. Generally, such releases are not self-propagating and are an inconvenience rather than an extensive hazard. Notable among the accidents

which have occurred in reactors of this type is the incident that resulted in the complete loss of the Windscale No. 1 reactor in October, 1957. Curiously, the accident occurred after the pile had been shut down for an annealing operation. The fuel was inadvertently overheated and ruptured fuel elements ignited, resulting in a further temperature rise to the uranium melting point. A high temperature, estimated to be 1200 to 1300°C, was maintained in a part of the reactor until the fire was finally extinguished by application of a large amount of water. The greatest environmental hazard arose from the release of approximately 20,000 curies of I^{131} to the surrounding English countryside, causing a governmental ban on the sale or consumption of milk and farm products from an area extending to 14 miles from the reactor. Little contribution to the total hazard was made by the escape of solid uranium oxide particles since the stack filters were able to contain essentially all material of this type.

Alloys — Uranium-aluminum and zirconium-enriched uranium fuels require water cooling and thin metal fuel plates to accommodate the high rate of heat production. Aluminum alloys have the added safety factor of a low melting temperature which, to some extent, enables them to serve as a reactor fuse. It is to be expected, therefore, that low fission product releases will usually be associated with a meltdown of fuel materials of this type. On the other hand, it was feared that zirconium, melting at about 1850°C, might possibly initiate a metal-water reaction which would become self-propagating. Much research has been done on this problem and, in general, the data indicate that there is little liklihood of such a reaction becoming self-propagating.

Uranium Oxide, Clad and Dispersed - The most significant use of dispersed UO_2 was as the fuel for the Army Power Package Reactor (APPR). When the UO_2 is dispersed in stainless steel, the resulting fuel has many safety features which recommend it for military applications.

The greatest advance to date in fuel for power reactors resulted from the use of UO_2 in pellets clad with stainless steel or zirconium. High fission densities and fuel burnups of 10,000 to 20,000 MWD/T are among its important advantages. Ceramic fuels are far less likely to fail catastrophically than metallic fuels because there is less possibility of ceramic materials reacting with coolants. The very high melting point of UO_2 (2850°C) gives it a safety advantage. Small cladding ruptures are found to release only small amounts of fission products other than the diffused rare gases stored in the void space. In the event, however, of a serious loss-of-coolant accident, the high rate of heat production would undoubtedly result in the melting of a fraction of the fuel. In that case a large part of the fission products, retained in the fuel at lower temperatures as a result of the high melting point of the UO_2, will be released by volatilization when the fuel assemblies melt.

FISSION PRODUCT RELEASE FROM URANIUM ALLOYS

Melting uranium-aluminum and uranium-zirconium alloys in air and steam have shown a proportional increase in fission product release with an increase in their respective melting temperatures. Little particulate material isproduced under these conditions because the oxides formed are highly refractory. The rare gases, iodine, and cesium are the major fission products released. Ruthenium and tellurium releases are apparently suppressed by alloy formation. Strontium release is also negligible in an oxidizing atmosphere, but a few percent of this element can be released in a reducing or inert atmosphere because strontium is more volatile than strontium oxide. A correlation of fission product release with fuel melting temperature for several fuel types is shown in Figure 1.

FISSION PRODUCT RELEASE FROM METALLIC URANIUM

In some release experiments, metallic uranium was exposed to air at elevated temperatures in a furnace tube with an attached series of filters and traps designed to afford fractionation of the principal volatile products. Most of the solid particles are normally deposited inside the furnace tube. However, the data in Table I show that a large part of the more volatile elements, including tellurium, ruthenium and cesium, were carried out of the furnace to the absolute filters. A heated charcoal trap collected all the vaporized iodine which passed through the absolute filters. Charcoal cooled by liquid nitrogen was used to adsorb the rare gases, xenon and krypton which passed through the hot charcoal trap. Highly irradiated uranium, completely oxidized at high temperatures, released the rare gases almost quantitatively.

Fission Product Release and Particle Production by Highly Self-Heated Uranium – One instance in which particles in the form of uranium oxide smoke were produced by metallic uranium heated in air at 1200°C is illustrated in Table 2. Circumstances leading to this unusual behavior are not well known; however, it is believed to be the result of self-heating induced by an accelerated air flow introduced at the time of melting which caused the uranium specimen to reach a temperature of at least 1800°C. While this type of release is recognized as abnormal it is still informative in relation to the most severe conditions.

Particles collected on the filter, shown in Figure 2, were photographed through an optical microscope. The agglomerates revealed in Figure 3 were in the range of 1-2 microns in diameter. These dimensions are somewhat smaller than those given by Chamberlain (6) for particles produced by oxidation at a lower temperature. Chamberlain stated that only 5 percent of the particles produced under his experimental conditions were less than 5 microns in diameter.

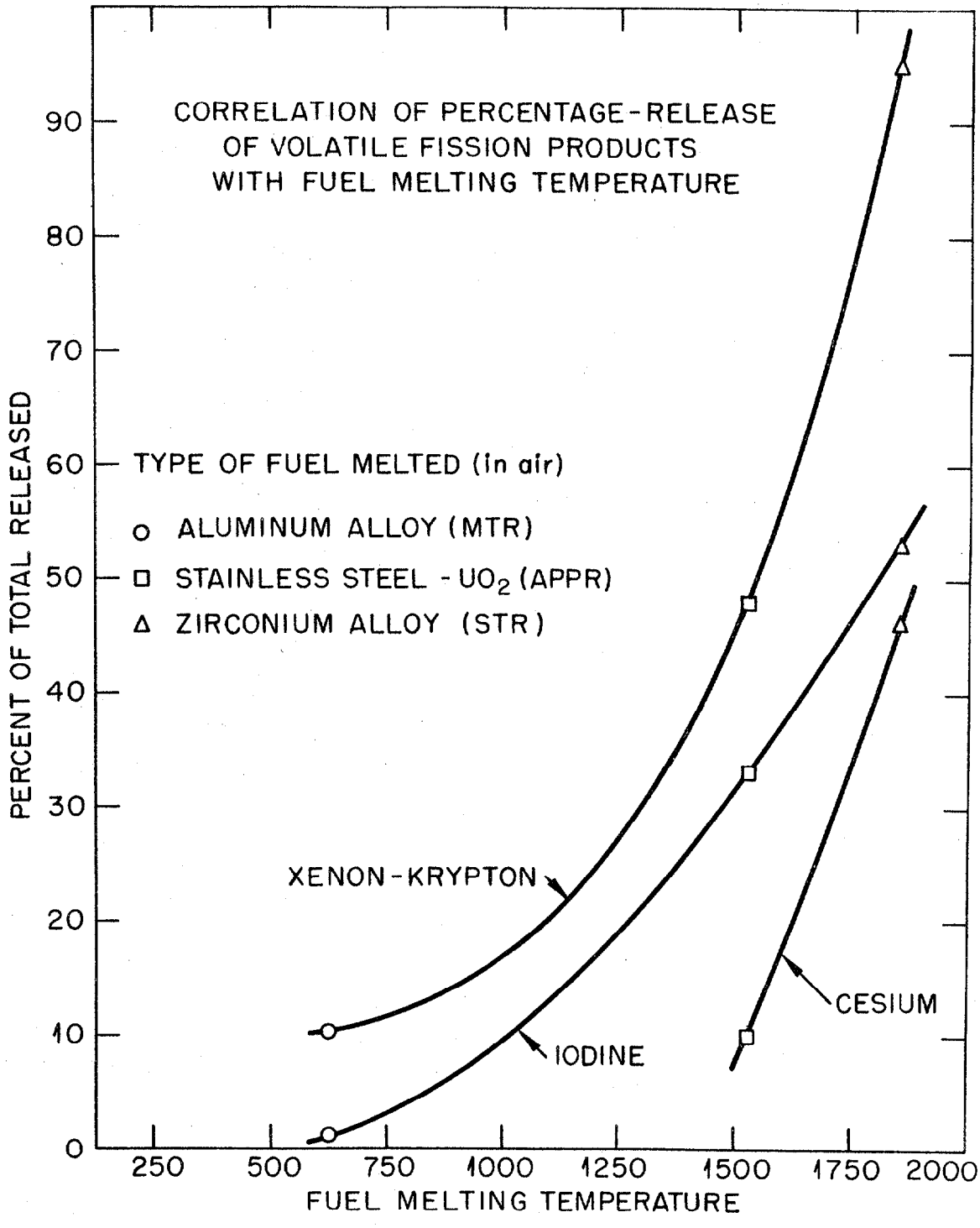
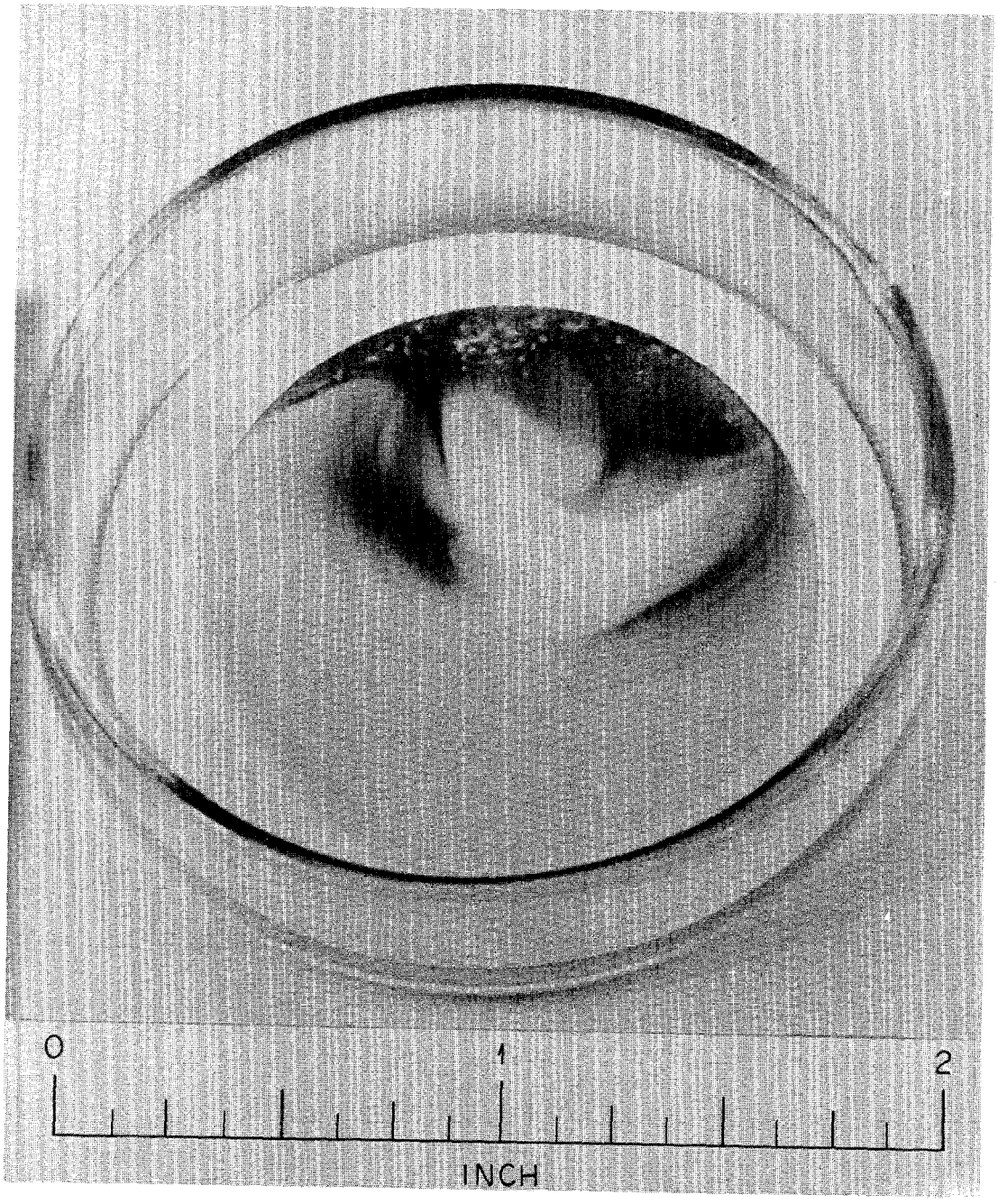
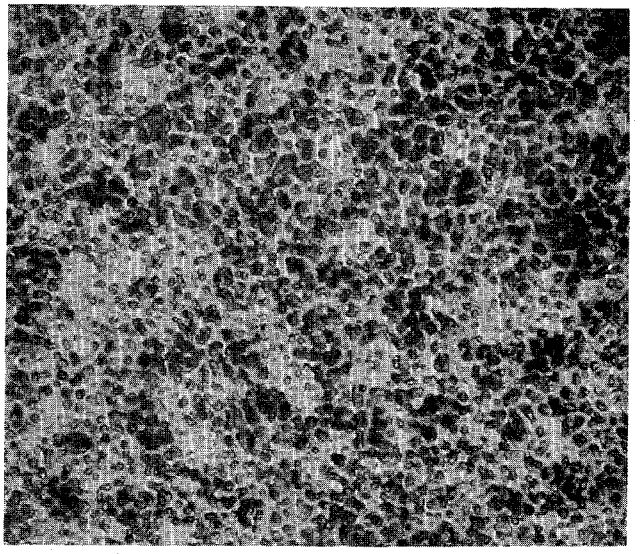

CORRELATION OF PERCENTAGE-RELEASE
OF VOLATILE FISSION PRODUCTS
WITH FUEL MELTING TEMPERATURE
TYPE OF FUEL MELTED (In air)
ALUMINUM ALLOY (MTR)
STAINLESS STEEL - UO2 (APPR)
ZIRCONIUM ALLOY (STR)
XENON-KRYPTON
IODINE
CESIUM
PERCENT OF TOTAL RELEASED
FUEL MELTING TEMPERATURE
0
10
20
30
40
50
60
70
80
90
250
500
750
1000
1250
1500
1750
2000

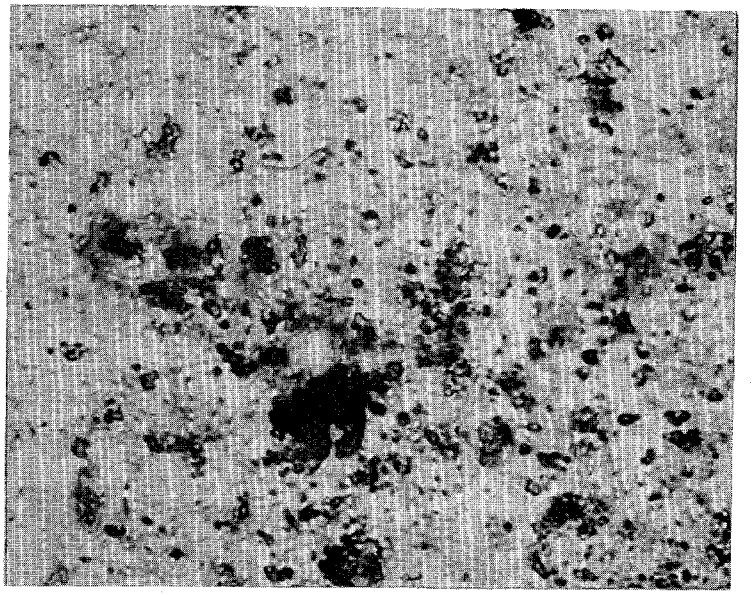
Figure 1



Filter Showing Smoke Like Oxide Particles from Highly Self-Heated Uranium Burned in Air

Figure 2





Appearance of Vaporized Oxide Particles (Above) from Highly Self Heated Burning Uranium (1000X) and (Below) of Ejected Particles from Uranium Melted in Impure Helium (4000X)

Figure 3

TABLE 1

DISTRIBUTION OF ACTIVITY RELEASED BY
OXIDATION OF IRRADIATED URANIUM* IN AIR

Air Flow Rate 600 cc/min

Temp. (°C)	Location of Activity	Percent of Total Activity Released					
		I	Ce	Te	Ru	Zr	Xe-Kr
800	Furnace	2.4	0.001	0.56	24.7	0.035	
	Filters	2.2	-	0.47	47.8	0.010	
	Hot Charcoal	43.4	-	1.85	-	-	
	Total	48.0	0.001	2.88	72.5	0.05	Not Det.
1000	Furnace	3.9	0.0012	55.1	49.6	0.004	
	Filters	4.9	0.0009	24.7	27.7	0.011	
	Hot Charcoal	82.2	-	-	-	-	
	Total	89.4	0.002	79.8	77.3	0.015	97.1
1200	Furnace	25.2	0.0005	40.5	21.2	0.011	
	Filters	1.3	0.0001	55.4	63.8	0.002	
	Hot Charcoal	63.3	-	-	-	-	
	Total	89.8	0.0006	95.9	85.0	0.013	99.2

* 0.2 a/o Burn-up

TABLE 2

FISSION PRODUCTS RELEASED FROM AN IRRADIATED SPECIMEN[a] WHICH REACHED AN ESTIMATED TEMPERATURE OF 1800°C ON HEATING IN AIR AT 1200°C FOR 20 MINUTES

Radioactive Species Released	Per Cent of Total Activity Released	Per Cent of Total Activity Collected on Filter[b]
Gross gamma	0.82	0.19
I	82.3	0.17
Cs	60.3	14.0
Ru	1.8	0.07
Zr	0.04	0.02
Ce	32.7	9.8
Sr	15.8	0.65
Pu	3.5	0.1

[a]Approximately 20 other specimens heated in air at 1200°C reached maximum temperatures much lower than this one which exhibited untypical behavior, possibly due to a high air flow rate.

[b]0.8 μ pore diameter millipore filter.

When metallic uranium was ignited in oxygen at 1200°C, the self-heating effect was even greater than in air and resulted in more vaporized UO_3. Figure 4 shows a collection of particles produced under these conditions and Figure 5 is an electron microscope replication of the very small particles produced. These particles are all below 1 micron in diameter and average about 0.1 microns. For particles of this size, deposition rates are expected to be very low and, in fact, a large fraction of them may be carried away by atmospheric diffusion.

A further illustration of the production of particulate activity from molten uranium is shown in Figure 6. In this case, uranium was melted in purified helium containing sufficient oxygen to form a thin oxide film on the metal surface. Since the film was quite thin it was ejected from the surface by differences in thermal expansion, between metal and oxide which was accentuated on cooling. The path of the particles may be judged from the fact that they are observed only in the right end of the boat (the direction of gas flow). Filter papers in the same photograph represent materials collected at 1150, 1200, and 1250°C. In Figure 3, an optical photograph again shows that the particles are relatively large (in the range 0.1-1.0u).

FISSION PRODUCT RELEASE AND PARTICLE EMISSION FROM UO_2 FUEL MATERIALS

When UO_2 is melted in air, helium, or CO_2, a large fraction of the volatile fission products is likely to be released. In the melting experiments performed to date, the samples have been relatively small to conform to equipment limitations. A direct result of this experimental condition is that the short diffusion paths favored maximum release values. Table 3 summarizes the most important results of these experiments without reference to the relative amounts plated out or carried to the filters. In Table 4, a comparison is made of the effect of furnace-gas-flow rate on the transfer of particles to the filters from the molten UO_2. From this table it can be inferred that, to a considerable extent, the fission products plate-out somewhat in proportion to the deposition of the vaporized uranium oxide. The principal exception to this behavior is seen in the iodine data. In one experiment, at a low rate of air flow, only about 11 percent of the iodine passed through the filter while the remainder collected in the deposited uranium oxide in various places. At a higher air-flow rate, 57 percent of the iodine was carried through the filters to the charcoal retaining traps.

A dispersion of UO_2 in stainless steel (APPR fuel) is intermediate between uranium alloys and UO_2 in melting point of the matrix material and in fission product release. Data obtained with APPR fuel is compared with release data obtained with other fuels in Figure 1.

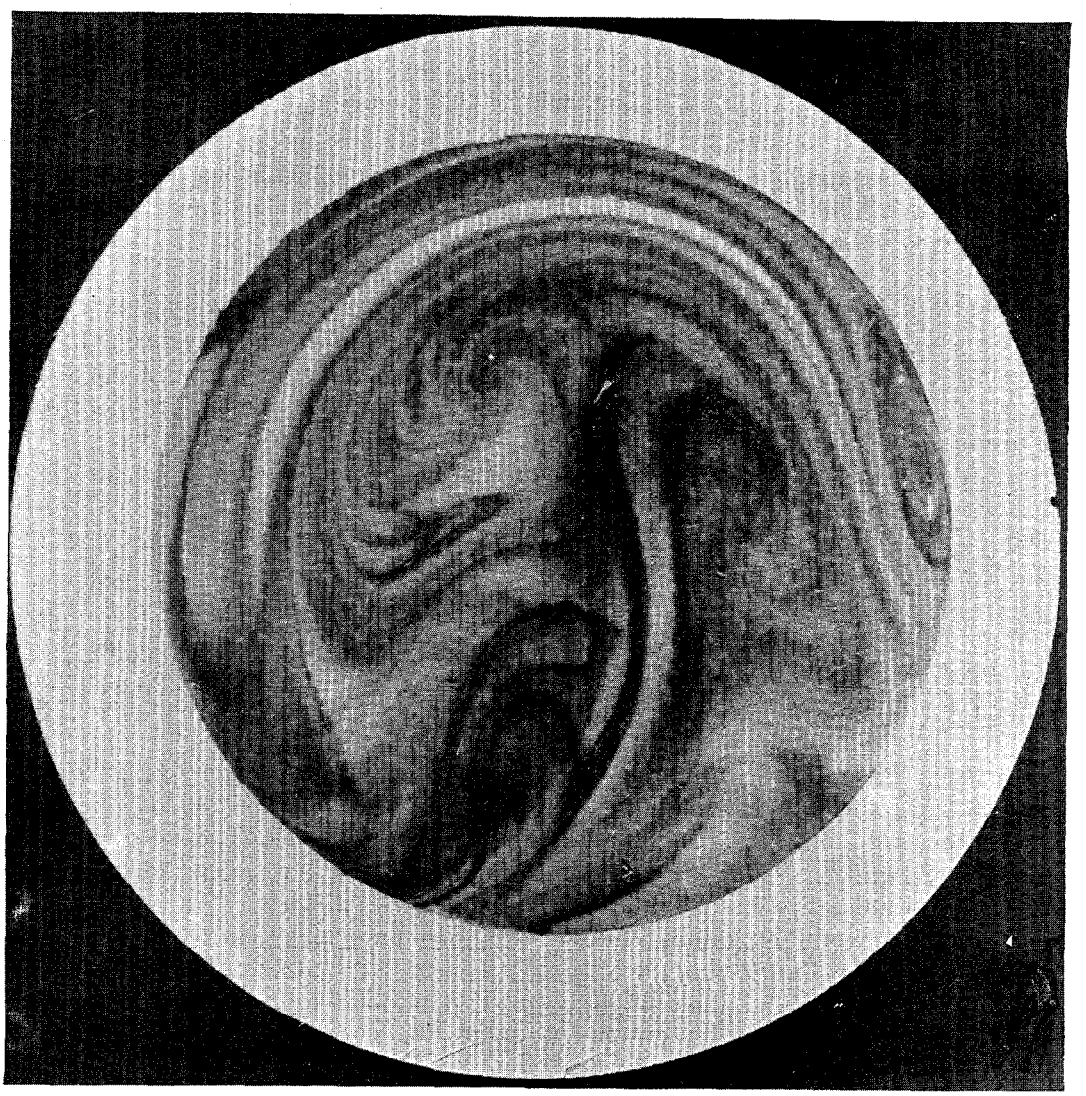


Figure 4

Vaporized Uranium Oxide from Ignition of Metallic Uranium in Oxygen at 1200 °C, 4X

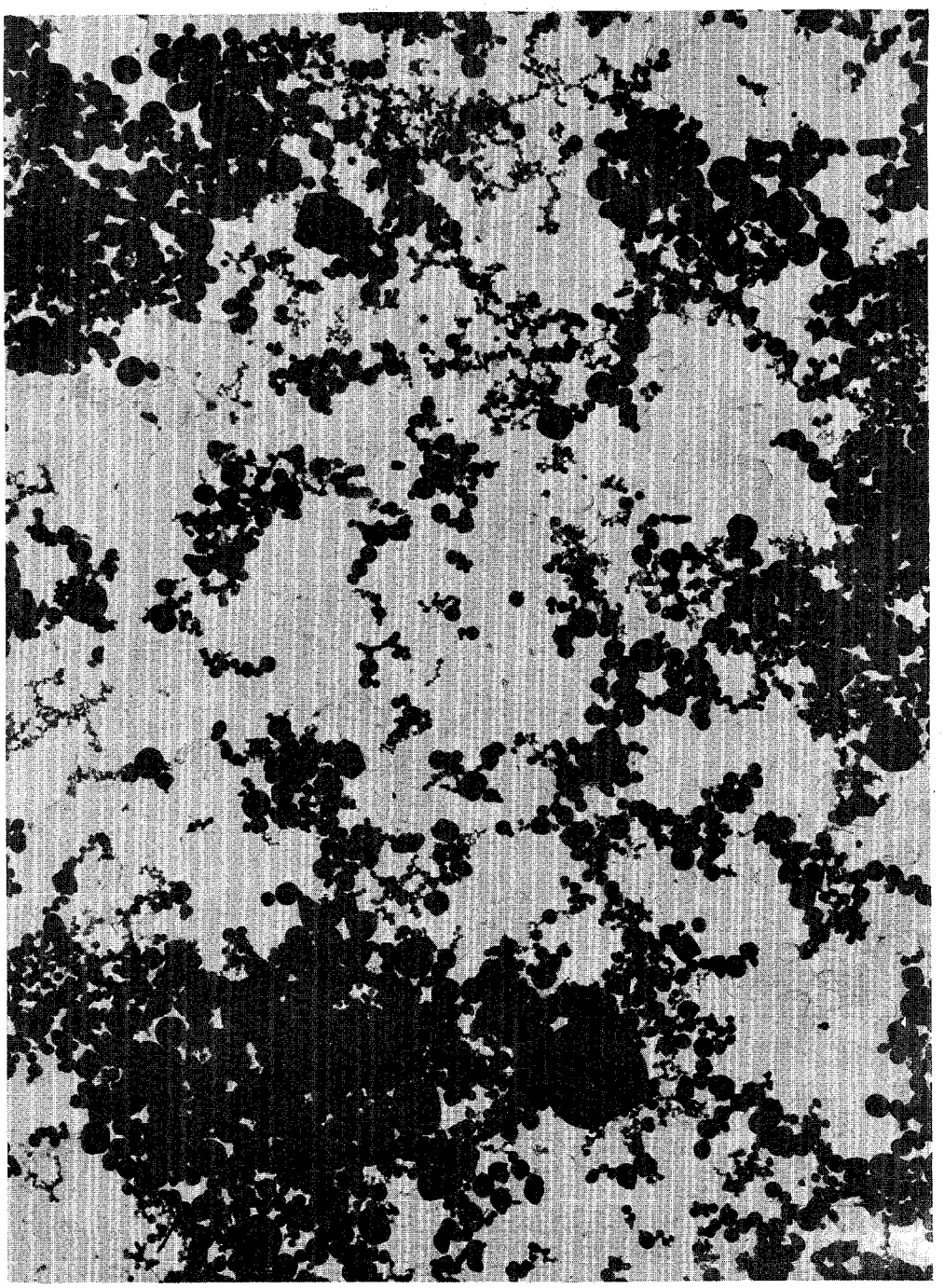


Figure 5

Electron Microscope Photograph at 20,000X of Particles from the Ignition of Uranium in Oxygen

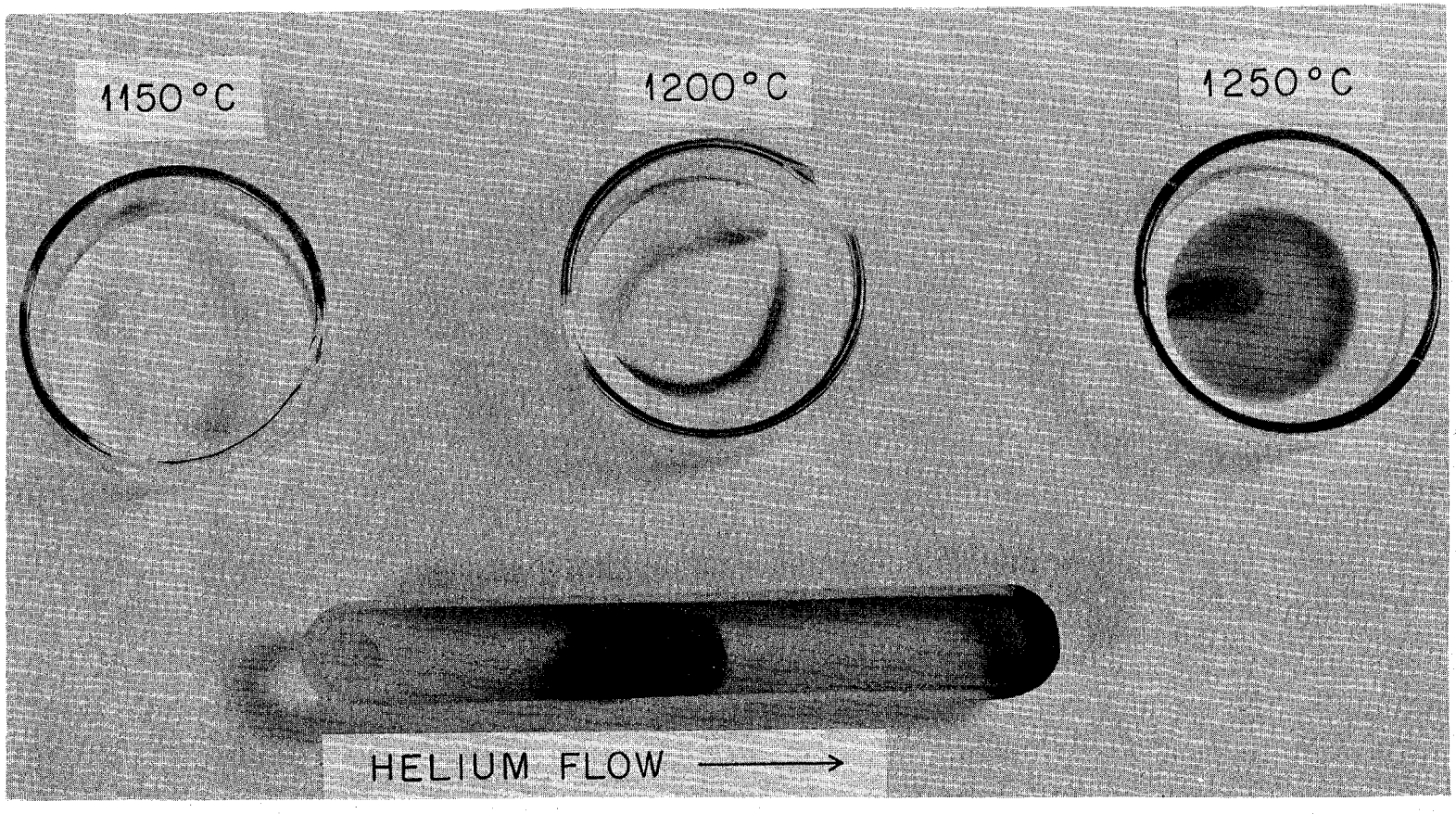


Figure 6

Appearance of Melt Residue, Ceramic Boat, Filter Papers and Particles Collected from Metallic Uranium Melted in Impure Helium

TABLE 3

EFFECT OF IRRADIATION AND ATMOSPHERE ON FISSION PRODUCT RELEASE

RESULTING FROM THE MELTING OF UO_2

Atmosphere	Irradiation Level (MWD/T)	Wt. of Sample (g)	No. Results Averaged	Percentage of Individual Fission Products Released								
				Xe-Kr	I	Te	Cs	Ru	Sr	Ba	Rare Earths	UO_2 Vaporized
Helium (Impure)	Trace	0.22	2	99.5	89.7	92.0	91.3	61.0	2.1	4.6	2.2	
	2800	0.03	3	99.9	92.2	98.2	98.5	90.4	2.1	6.6	5.1	21.2
Air	Trace	0.2	2	98.4	94.9	79.1	37.7	67.7	0.2	0.5	0.5	
	2800	0.04	3	100.0	99.7	93.6	92.5	95.0	0.4	1.8	3.0	
CO_2	Trace	0.2	3	80.6	76.8	71.2	60.9	44.9	0.3	1.1	0.85	14.1
	2800	0.02	3	99.9	98.7	98.6	90.2	74.3	0.5	2.5	2.8	
	11000	0.05	3	99.9	99.9	99.0	96.6	79.1	0.6	2.9	2.3	

EGCR UO_2, with O/U Ratio of 2.04 and Density 95% of Theoretical (Average).

TABLE 4

EFFECT OF FURNACE AIR VELOCITY ON DEPOSITION OF FISSION PRODUCTS RELEASED ON MELTING MODIFIED UO_2 FUEL MATERIAL AT 2650°C

	Per Cent Released and Location of Deposit							
	Furnace Tube		Filters		Charcoal Trap		Total	
Velocity of Furnace Air (lfm)	0.3	5.0	0.3	5.0	0.3	5.0	0.3	5.0
Gross Gamma	10.1	8.8	1.2	6.2	0.4	3.4	11.6	18.4
Rare Gases					77.1	69.5	77.1	69.5
Iodine, γ	62.3	12.1	5.4	9.8	11.3	56.9	79.0	78.8
Tellurium, β^-	66.8	28.6	8.3	43.2			75.1	71.8
Cesium, γ	34.5	18.3	15.6	30.9			50.1	49.2
Ruthenium, γ	63.6	33.0	0.8	24.6			64.4	57.6
Cerium, β^-	0.24	0.17	0.11	0.37			0.35	0.54
Strontium, β^-	0.1	0.05	0.01				0.11	0.05
Barium, β^-	0.55	0.08	0.01	0.07			0.56	0.15
Zirconium, γ	0.02	0.02	0.01	0.01			0.03	0.03
Uranium*	6.4		1.4				9.8	

Air Flow = 200 cc/min = 0.3 lfm or 3000 cc/min = 5 lfm Across Sample.

Heating Time, 90 sec.

* Vaporized as UO_3

Size of Uranium Oxide Particles Produced on Melting UO_2 – In general, the presence of air during the melting of UO_2 is observed to enhance uranium vaporization because UO_3 is more volatile than UO_2. This difference is easily seen in Figure 7 where filter papers obtained in experiments in which melts were conducted in different atmospheres are shown. The ratio of the amount vaporized in air to that in helium is about 10. The atmosphere also affects the size of the vaporized particles; those produced in air are larger than those obtained in a helium or CO_2 atmosphere. The shape of particles and their composition are also affected by the atmosphere. Crystalline U_3O_8 particles shown in Figure 8 were produced in air, while spherical UO_2 particles shown in Figure 9 were produced in helium and the UO_2 particles shown in Figure 10 were produced in CO_2. The size distribution for each case is given in Figure 11.

CHEMICAL STATES OF RELEASED FISSION PRODUCTS

The fission product chemical species shown in Table 5 are believed to be the most probable states of these elements that would be released and deposited under typical reactor accident conditions when reactor fuels are heated to high temperatures in an atmosphere containing some oxygen. The postulated states, although somewhat speculative, are based in part on the observed behavior of the fission products in release experiments, including the effect of atmosphere on volatility, and on the known physical properties of the various chemical species. It is not believed to be worthwhile to consider the probable chemical states of the fission products which would be released in a pure, unoxidizing, atmosphere because it seems unlikely that such conditions would exist during a reactor accident.

REFERENCES

1. Leonard, B.P., Hazards Associated with Fission Product Release, Geneva Conference Paper 428 (1958)
2. Loutit, J.F., Marley, W.G., and Russell, R.S., The Nuclear Reactor Accident at Windscale, in Medical Research Council, The Hazards to Man of Nuclear and Allied Radiations (2nd Report), Appendix H (1960) HMSO.
3. Creek, G. E., Martin, W. J., and Parker, G.W., Experiments on the Release of Fission Products from Molten Reactor Fuels, ORNL-2616, 1959.
4. Hilliard, R.K., Fission Product Release from Uranium Heated In Air, HW-60609 (1960).
5. Parker, G.W., Martin W.J., and Creek, G.E., Fission Product Release from Reactor Grade UO_2 by Oxidation, Diffusion and Melting, ORNL, CF 60-12-14
6. Chamberlain, A.C., Progress Report on Uranium Oxidation and Fission Product Emission, UKAEA Report RSRWP/57 (1958).

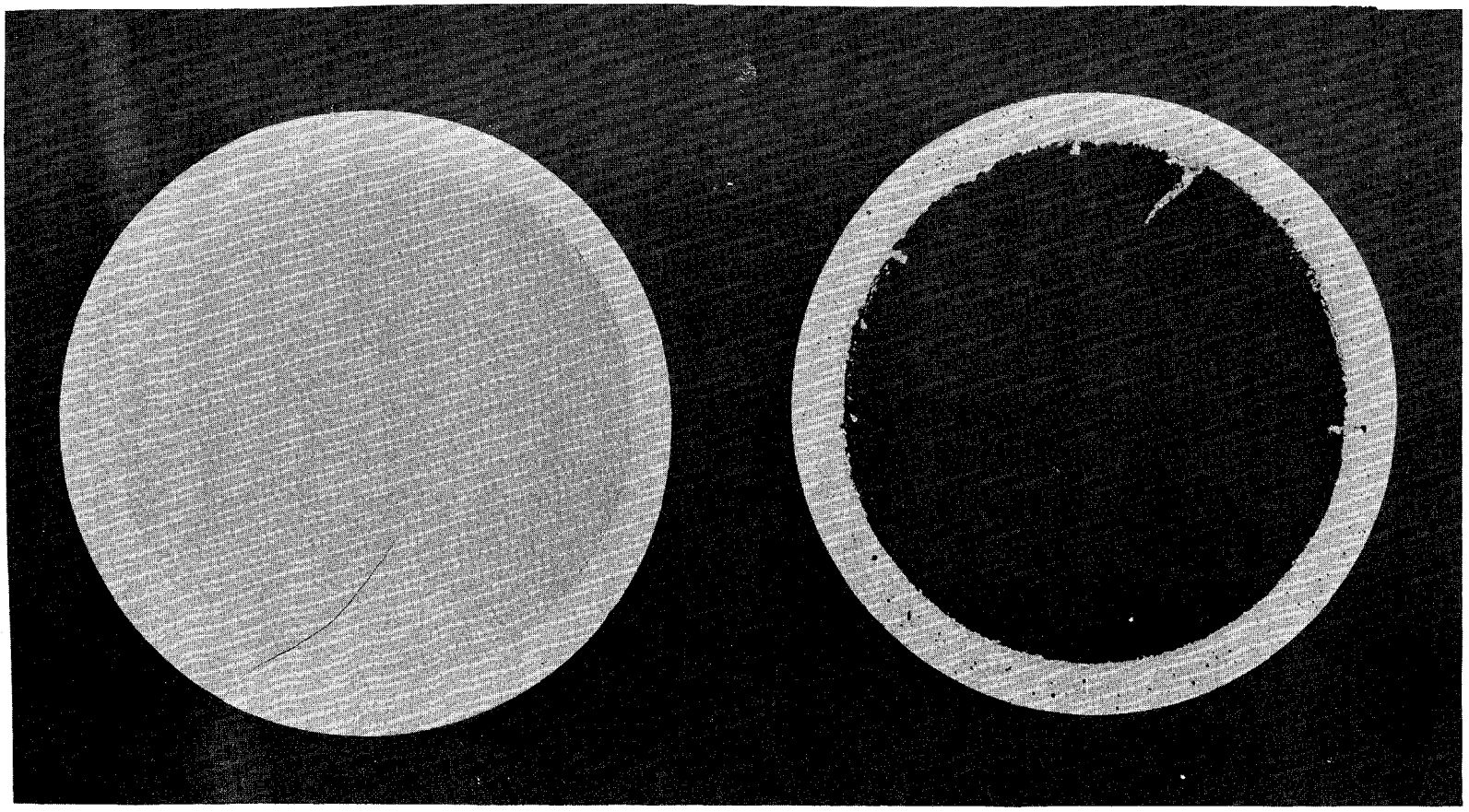
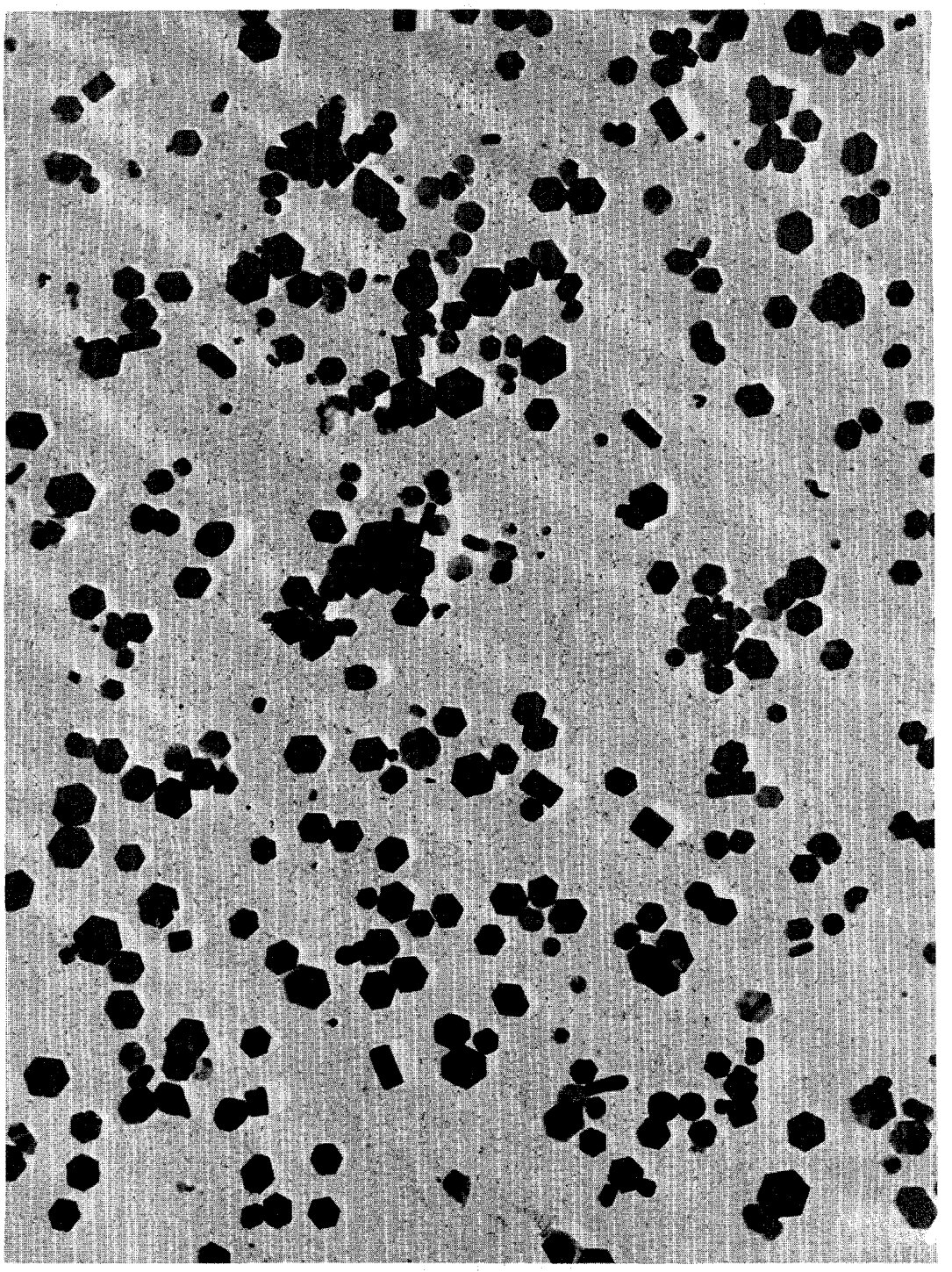


Figure 7

Vaporized Uranium Oxide Collected from Melts of UO_2 in Helium (left) and in Air (right), 2X



Replica of crystalline U_3O_8 particles vaporized from UO_2 melted in air. 40,000X. (Particle diameters 0.02 to 0.2 microns.)

Figure 8

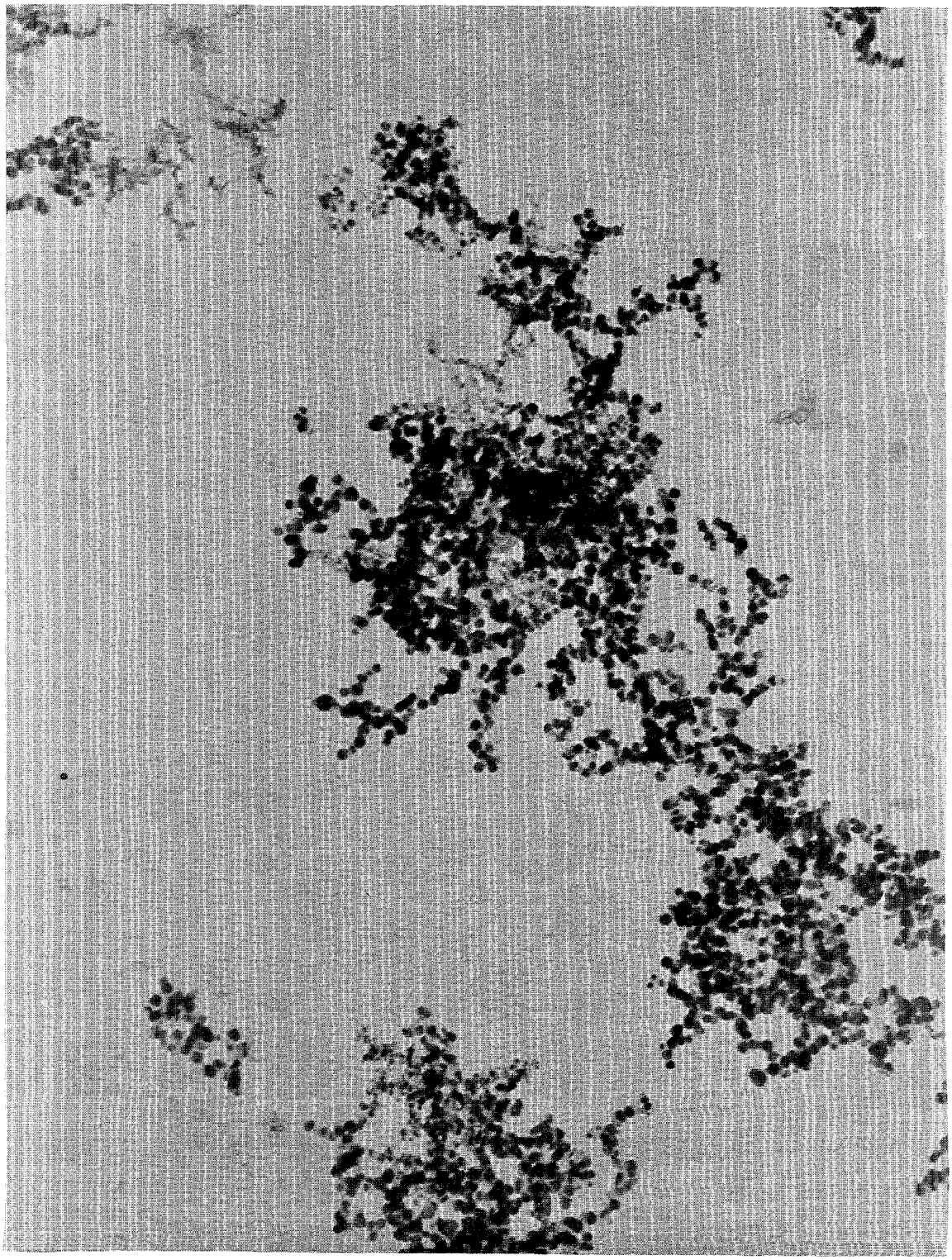


Figure 9

Replica of Spherical UO_2 Particles Vaporized from UO_2 Melted in Helium (69,000X)
(Particle Diameters 0.01 to 0.05 microns)

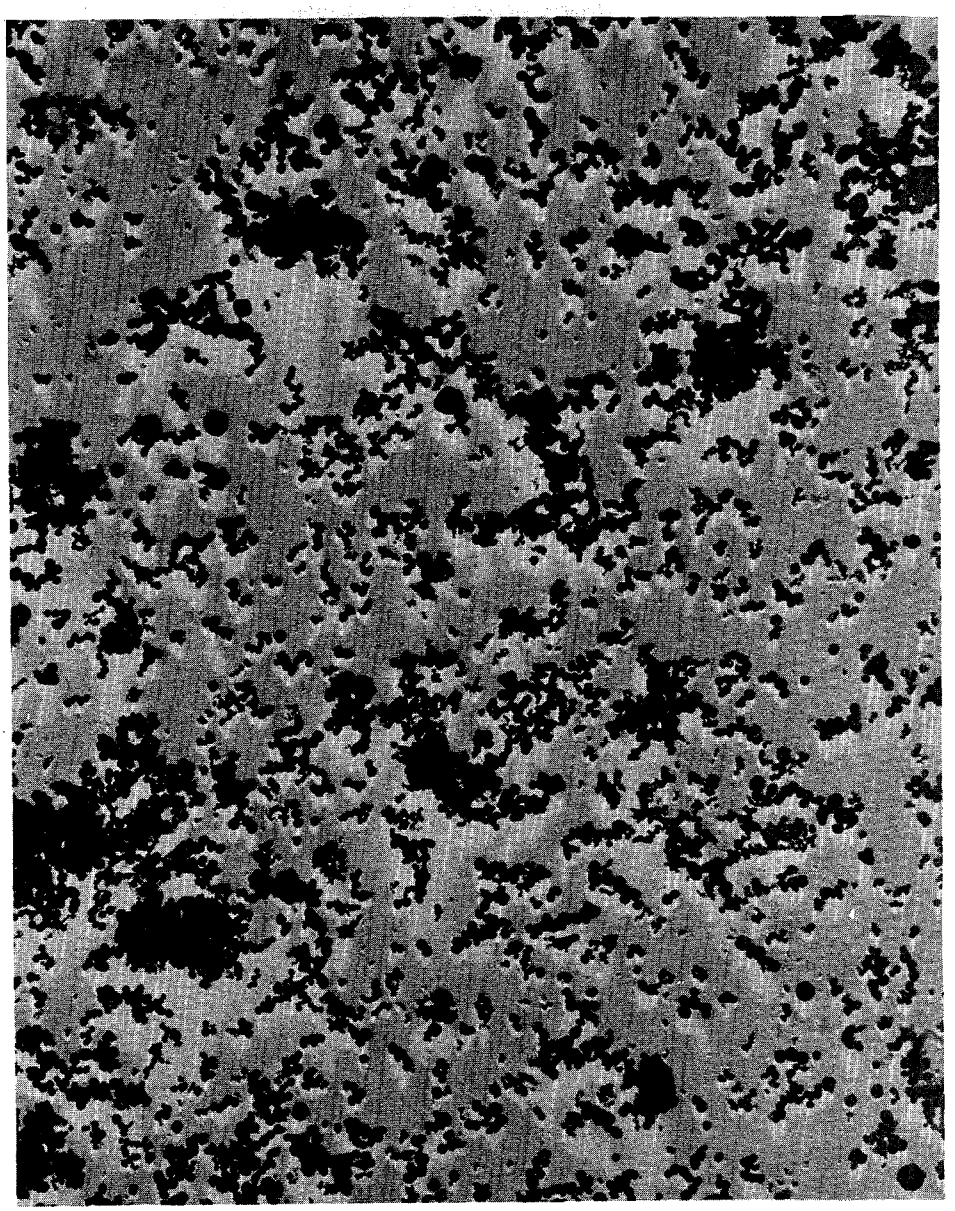
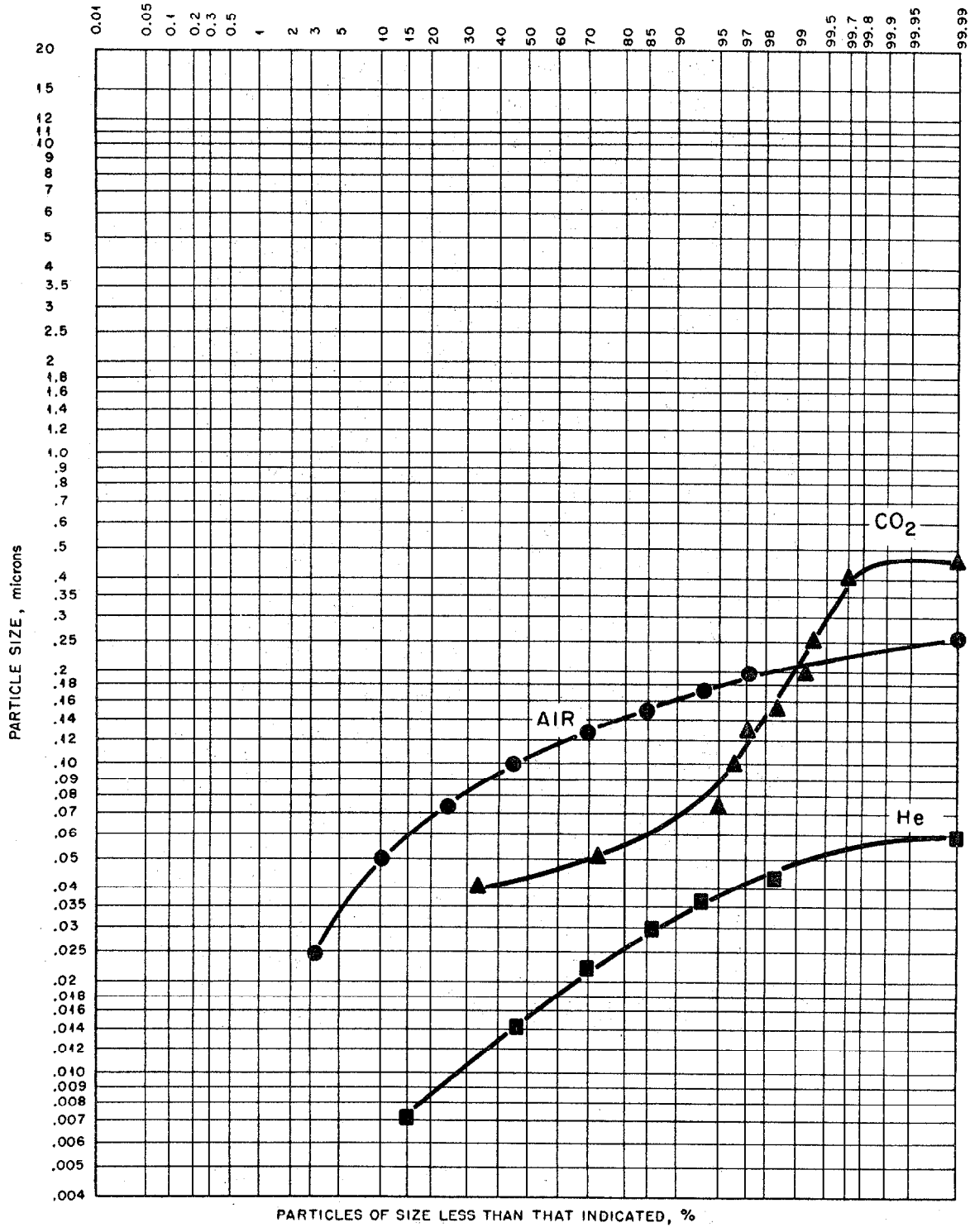


Figure 10

Replica of Spherical UO_2 Particles Vaporized from UO_2 Melted in CO_2 (20,000X)
(Particle Diameters 0.04 to 0.4 microns)



Particle Size Evalution of Oxides Vaporized from Melted UO_2. (Arc-image Furnace).

Figure 11

TABLE 5

PROBABLE CHEMICAL STATES OF RELEASED FISSION PRODUCTS*

Element	Vaporized State	Deposited State
Rare Gases	Kr, Xe	
Iodine	I_2	$I^- \rightleftharpoons I_2$
Tellurium	TeO_2 + Te	TeO_2
Cesium	Cs	Cs_2O
Ruthenium	RuO_4	Ru_2O_3 + Ru
Strontium	Sr	SrO

* This simplified summary is based partly on observed chemical behavior, including changes in volatility with atmosphere, and partly on physical properties. The equilibration of mixed states assumes an atmosphere containing oxygen and some reducing agent on plate-out surfaces.

THE HANFORD REACTOR CONFINEMENT PROGRAM

H. W. Heacock and C. E. Jones
General Electric Company
Hanford Atomic Product Operations

ABSTRACT

A project that provides facilities for confining the fission product release from a nuclear incident has been completed for the eight existing Hanford reactors. Similar facilities are also being constructed into the New Production Reactor. The existing facilities are reviewed and are compared to those planned for NPR. The testing program supporting design and operating experience with the existing system are summarized.

As a result of the tests performed by Arthur D. Little, Incorporated, Cambridge, Massachusetts, activated charcoal was selected as the media for the halogen filters. Roughing filters were deleted from the system based on results of the filter life tests. In-place DOP testing of the absolute filters following their installation insures a high degree of filtration efficiency.

Functional tests of the system that simulate a burning irradiated fuel element have been satisfactory. Such tests have resulted in confinement zone pressures being held 0.3 inch water gage negative with respect to atmospheric pressure. In practice it is anticipated the system will remove 99.9 percent of the particulates, about 95 percent of the halogens, but none of the noble gases.

INTRODUCTION

A project that provides facilities for confining the fission product release from a nuclear incident has been completed for the eight existing Hanford Reactors, and similar facilities are being constructed into the New Production Reactor which is scheduled for completion in 1963. An earlier report (1) presented to the Sixth Air Cleaning Conference in July 1959 described the status of the confinement program relative to the existing production reactors. This report updates the earlier report, summarizes the experience gained in operating the system, and compares the confinement program in the old areas with that in NPR. Since considerable time has elapsed, a brief review of the philosophy and development of the confinement system appears in order.

To begin with, it is important to note that these are both referred to as confinement rather than containment facilities, since in neither of them was the familiar containment

sphere used. Rather, the objective is to control the flow of the ventilation air to definite paths and to exhaust it through the filters before its release from the stack. To achieve this objective two general principles have been used to develop the system: (1) the confinement zone should be maintained at a pressure slightly less than atmospheric, and (2) the exhausting equipment should be designed to provide the highest degree of reliability feasible. This approach is considered necessary because of the inherent massiveness of the Hanford Production Reactors. It is very important to realize that although this concept has been used on the Canadian NPD-2 as well as at Hanford it is an entirely different concept for controlling fission product release from that used for Dresden, PWR, and other recently completed reactors.

In the old reactors the negative pressure concept insures that air leakage is into the confinement zone; thus, all air leaving critical parts of the building will pass through the filtering facilities. The system reliability has been upgraded to insure that the confinement zone is maintained at negative pressure. This is accomplished by (1) a separate power supply to drive the emergency exhausting equipment, (2) automatic controllers for this emergency power supply and for the ventilation dampers, and (3) emergency power-driven pumps to furnish water to the fog spray.

The quantities of air exhausted vary from 85,000 to 150,000 cfm for the various reactor buildings or zones and indicate the magnitude of the project. Although there is nothing unique in the system, several problems were encountered and numerous tests were required in support of design to aid in applying industrial ventilation practices to such a large system. These tests will be defined in more detail later.

Without becoming involved in an extensive discussion of the early Hanford confinement studies, it was concluded that it would be feasible to remove approximately 99.9 percent of the particulates and at least 50 percent of the halogens but none of the noble gases that may be released from minor reactor incidents. In practice it is expected that about 95 percent of the halogens will be removed. Accordingly, the basic features of the confinement facilities for the existing Hanford reactors include:

1. A dense, finely atomized water spray system within the rear face enclosure (fuel discharge location) of each reactor.

2. A filtering facility to remove a high percentage of all particulate matter and most of the halogens from the reactor building exhaust ventilation air.

3. Suitable instrumentation to monitor, record, and where necessary, control critical ventilating equipment to insure passage of the exhaust air through the filter building.

The basic philosophy used for the confinement facilities in the old reactors was also used for the New Production Reactor (NPR). The application of this philosophy, however, required improved filter performance and alteration of the system to permit the potential release of large quantities of steam that would accompany the possible rupture of a high pressure cooling loop. The basic criteria[2] for the system require that:

1. The primary confinement zone surrounds all portions of the nuclear system that contain the high pressure, high temperature radioactive reactor coolant.

2. The portion of the reactor block outside the primary confinement zone is enclosed by an intermediate confinement zone.

3. The primary and intermediate zones are nominally leak tight and are designed to withstand the maximum and minimum potential internal pressures generated in the zone during a nuclear release.

4. The steam release vent covers remain closed during minor incidents to permit building pressure control by the fog spray with subsequent filtration of the building exhaust air. In the event of a primary loop rupture the steam release covers open. After the steam pressure has been reduced and before any fission product release occurs the automatically operated closures, provided on each vent, close to insure filtration of the exhaust air. The steam release vents are arranged and sized to vent either the reactor or heat exchanger buildings. Special ventilation barriers in the wall between the buildings permit this common path yet provide for separate normal ventilation systems.
5. Backup closures, for emergency use, are provided on all primary zone closures. During normal operation these zones will be maintained at sub-atmospheric pressures with the primary zone pressure the more negative.

6. The confinement system, except for backup closures, is automatically actuated when an incident occurs.

DESCRIPTION OF THE SYSTEM

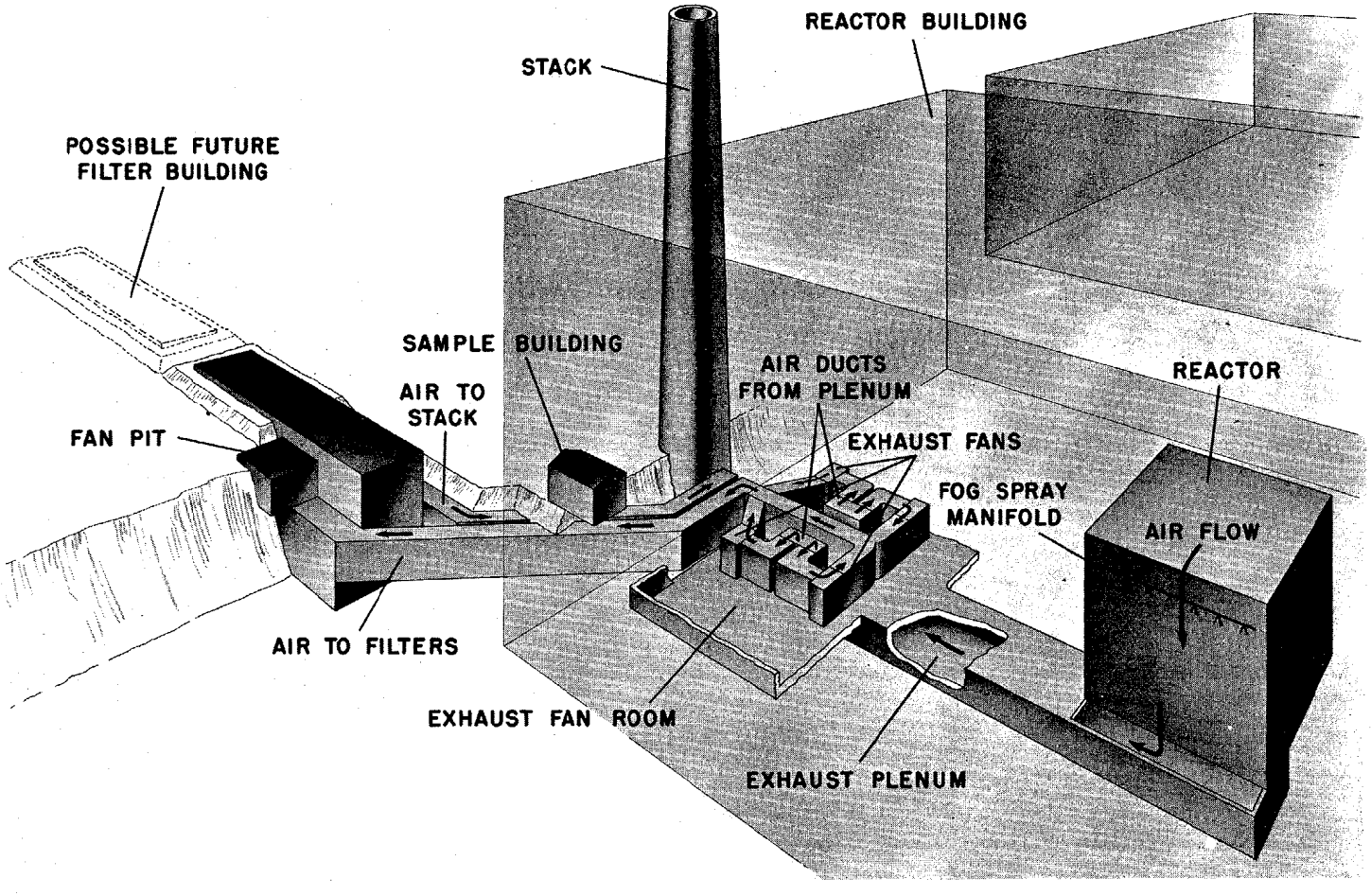
Old Areas

As a portion of the review we should orient ourselves to a typical facility layout. Figure 1 provides orientation of the buildings and graphically illustrates the flow of exhaust air from the confinement zone (areas adjacent to the reactor) of the building. The reactor block is located within the reactor building structure so that a ventilated space exists between the block and the outside walls. During normal operation air is supplied to the ventilated areas by two supply fans. This air passes over the reactor block surfaces and is drawn into a common plenum on the suction side of the exhaust fans. In the past the fans have discharged the air directly to the stack; however, the air is now diverted through the filtering facility prior to its release.

The sample building, constructed above the ventilation ducts leading to and from the filter building, houses the bulk of the instrumentation associated with the confinement facilities.

Components

As an aid to review and for ease in discussion the system has been divided into four parts. These are the fog spray, filtering facility, ventilation modification and instrumentation.


REACTOR BUILDING
STACK
POSSIBLE FUTURE
FILTER BUILDING
SAMPLE BUILDING
AIR TO
STACK
AIR DUCTS
FROM PLENUM
REACTOR
FAN PIT
EXHAUST FANS
FOG SPRAY
MANIFOLD
AIR FLOW
AIR TO FILTERS
EXHAUST FAN ROOM
EXHAUST PLENUM

Fog Spray

Briefly the fog spray is a finely divided water spray system located within the rear face (fuel discharge) enclosure and is designed for automatic or manual operation. This is shown in more detail on the next slide (Figure 2). Specifically the spray system is designed for the multifold purpose of:

1. Absorbing a portion of the halogen vapors released during any uranium fire.

2. Settling out a portion of the airborne particulate matter released during fuel element fires.

3. Washing down exposed surfaces within the rear face enclosure for removal of contaminated particles.

4. Providing some degree of thermal cooling to exposed fuel elements.

5. Condensing any steam that may be formed to prevent unnecessary pressure build up within this area.

A scintillation detector system continuously monitors the reactor building exhaust air and actuates the spray system whenever the presence of radio-iodine is detected in the exhaust air stream. Operating experience to date has shown the fog spray system to be effective in: (1) reducing spread of contamination, (2) reducing air borne contamination during periods of reactor maintenance and (3) cooling exposed irradiated fuel. During a recent incident that involved the partial burning of an irradiated fuel element there was no detectable spread of contamination to ventilating equipment down stream of the rear face enclosure, although before the installation of the fog spray similar incidents had resulted in noticeably greater radiation levels emanating from this exhausting equipment.

Filtering Facility

As in indicated on the next slide (Figure 3), the filter building is of reinforced concrete construction and is almost entirely below ground. Although the normal radiation activity on the filters should be quite low, the undergound installation was used since, (1) earth is cheap shielding, (2) the building and associated ductwork would cause less hindrance to movement of vehicles and personnel within the area, and (3) abandonment, in place, would be much simpler should this ever become necessary. The blanked duct extensions were provided for this future installation.

The volume of air being filtered various from 85,000 to 150,000 cfm. The total number of filters are housed in the two cells of the filter building. Each cell can be isolated, by means of water seal pits, from the exhaust air flow for filter replacement. Each filter building cell contains provision for three banks of filters in series and each bank is composed of two halves, each of which is a structurally integral unit for ease of filter replacement. Each integral unit consists of an aluminum frame holding from 24 to 40 individual absolute or activated charcoal filters rated at 1000 cfm each.

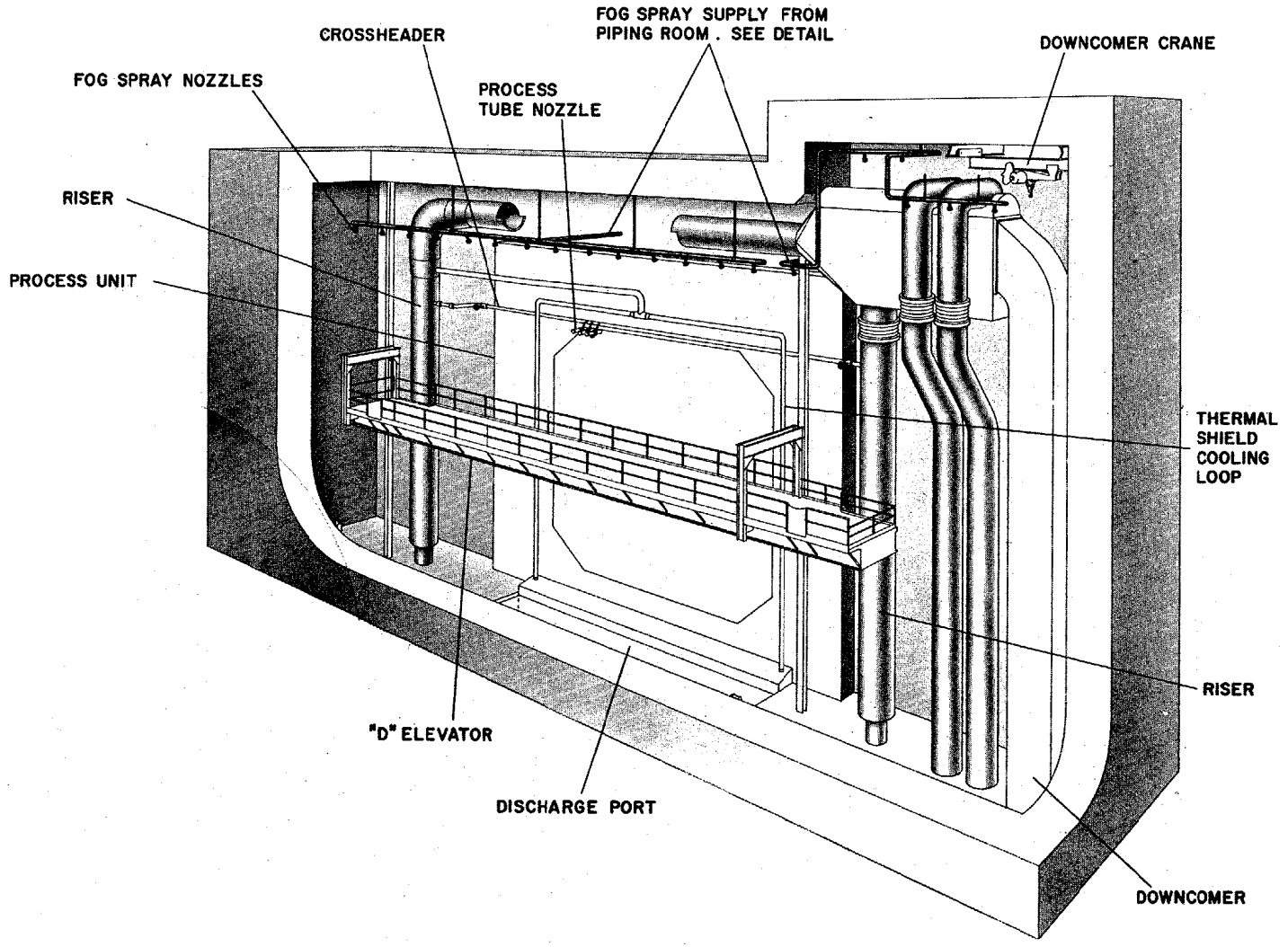


FIGURE 2
FOG SPRAY SYSTEM

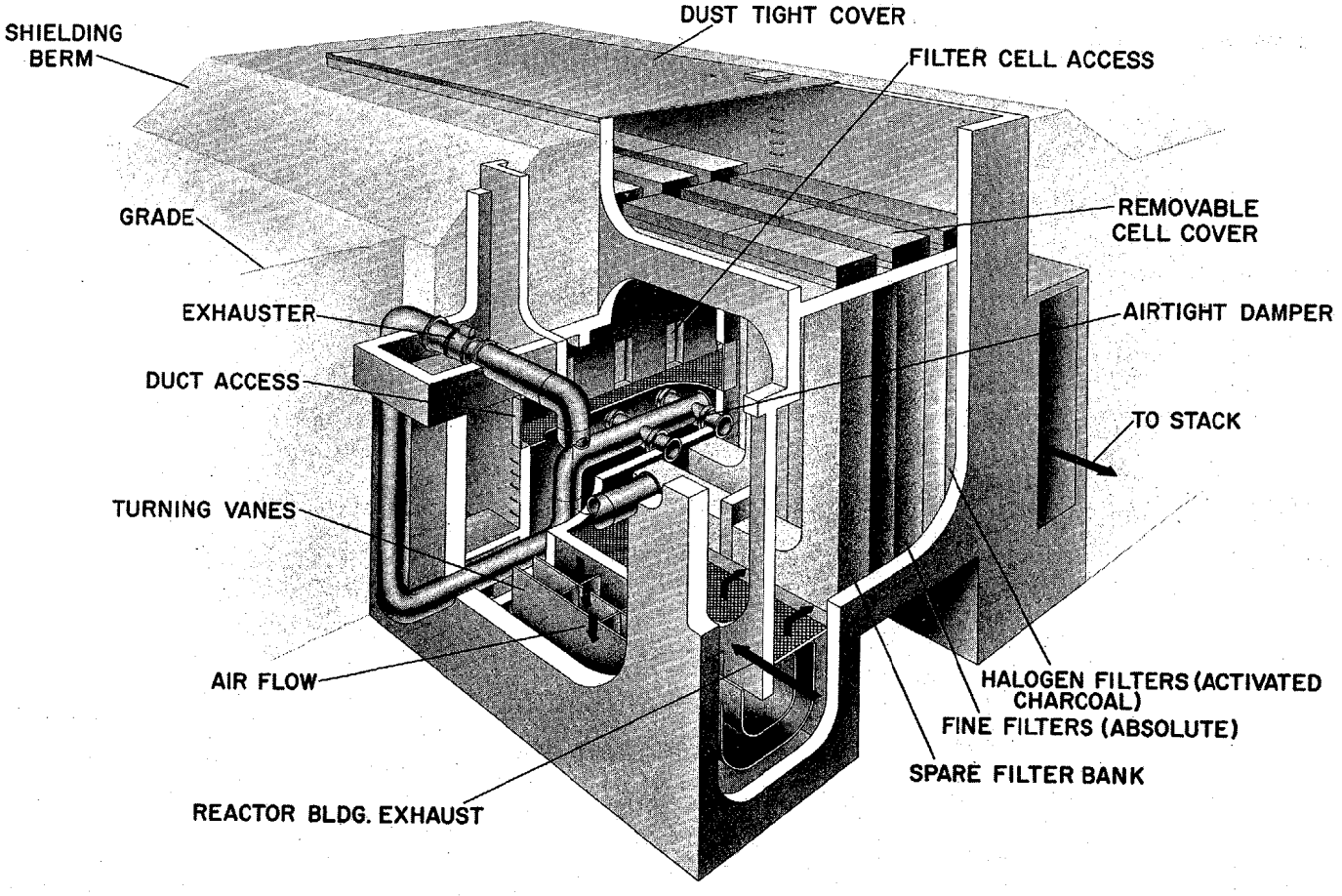


FIGURE 3
FILTER BUILDING

Thus a minimum of 96 individual filters, absolute or charcoal, is required for the smallest building; 160 of each are required for the filter building which has the greatest air flow. The original design, completed before the completion of the filter life tests, proposed three banks of filters in each compartment, namely: roughing filters, fine or "absolute" filters, and halogen collectors.

As a result of these filter life tests, to be presented in more detail by Mr. F. E. Adley[3], it was found that the roughing filters added little to the effectiveness of the system and were nearly as expensive as the absolute filters. With the development of a method for testing in place, a decision was made to install a single bank of absolute filters. These filters are rated for emergency operation at 200°F and a relative humidity of 100 percent for a period of two hours without loss or decrease in filtering efficiency.

The third filter bank constitutes the halogen removal system and consists of filters whose filter media is a bed of activated coconut shell charcoal one inch in depth. This particular component is the result of an extensive testing program conducted by Arthur D. Little, Inc.[4] and will be discussed later. The completed building now houses the absolute filters in the first bank and the activated charcoal filters in the third bank; the second bank is unused.

All the filter banks were designed so that normal filter replacement can be accomplished by a portable crane and without access to the interior of the filter cells. In order to prevent the spread of contaminated particulate matter, an exhauster was provided to create a positive sweep of outside air into the cell when the cell covers are removed for filter replacement. This air is then routed through the filters in service. After isolation of the filter cell from the ventilation air stream, the selected frame of filters will be withdrawn into a plastic bag and transported to the disposal ground for disposal of the dust loaded filters and recovery of the frame for reloading.

While the probability of a serious incident is very low, an incident involving several tubes of fuel elements would no doubt contaminate the filters to the extent that immediate replacement would be impossible. Although the incident itself would not dictate immediate replacement of the filters since they are well shielded, when the pressure drop through the filters is great enough to make continued operation of the ventilation system untenable, the decision must be made either to replace the filters or construct a new filter building. To provide for this later eventuality, the filter building design has included means for making such an extension and space has been reserved for the possible new building.

The next slide (Figure 4) shows an enlarged isometric detail of the filter frame construction. Because the filters must be replaced by semi-remote methods, a continuous inflatable seal has been used around the periphery of both the upstream and downstream faces. Individual filters are mounted in the filter frame in a more or less conventional manner, with compression gaskets to provide the seal for individual units.

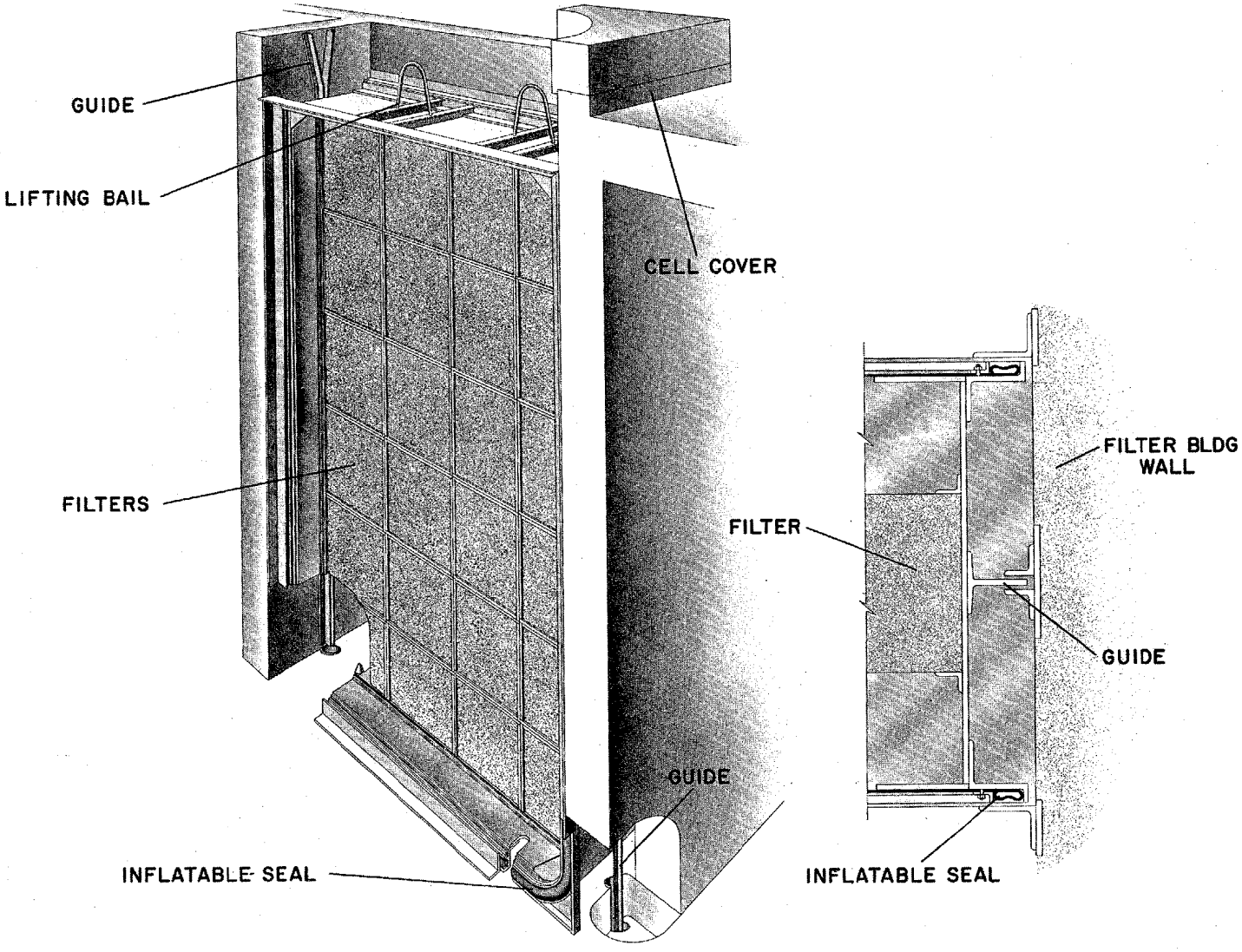


FIGURE 4
TYPICAL FILTER BANK ARRANGEMENT

Ventilation System

Because the confinement facilities were added to existing buildings, the criteria for modifying the existing fans and balancing the system were few. A typical ventilation flow diagram is shown in the next slide (Figure 5). The modifications to the system were as follows:

1. The exhaust fans were upgraded to maintain the same general volume of air flow but at an increased static pressure of approximately six inches water gage caused by addition of the filter facility.

2. The ventilating system was balanced to maintain the confinement zone pressure slightly less than atmospheric pressure.

3. Alarms were provided to warn operating personnel of any significant change in ventilation balance.

4. Dependable emergency power, either steam turbine or diesel engines, was provided to drive the emergency exhaust fans in the event normal power was lost.

5. Instrumentation was provided to automatically shut down the supply fans in the event of a reactor incident to insure that zone pressures are maintained negative with respect to atmospheric pressure.

6. Ventilation barriers were installed to permit ventilation air balance crews to successfully balance the system with the confinement at a negative pressure.

Instrumentation

The next slide (Figure 6) shows an engineering diagram of the confinement facilities and the associated instrumentation. The dashed lines have been added to the diagram to indicate the general location of the system components. In addition to the normal amount of instrumentation associated with measuring system flow, pressure and radioactivity, instrumentation has been included to provide alarms in the control room for the following off-standard conditions:

1. Static Pressure in the confinement zone increasing and approaching atmospheric pressure.

2. Water flowing through the fog spray header.

3. Water pressure supply to the fog spray either low or off.

4. Abnormally high radioactivity (Iodine 131) in the building exhaust air.

5. Failure of one or more monitoring instruments.

6. High radiation activity on either particulate sampler.

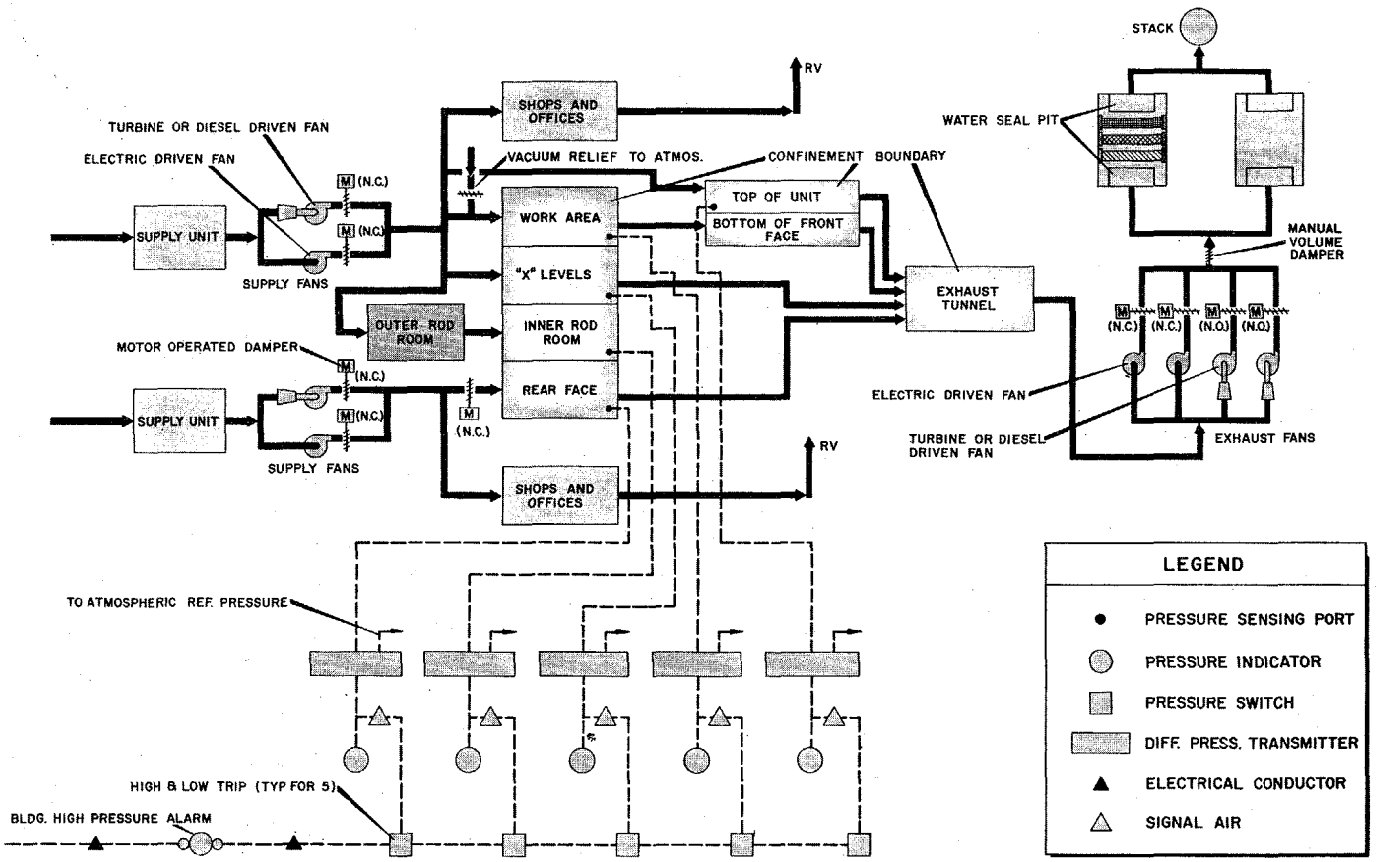


FIGURE 5
TYPICAL FLOW DIAGRAM

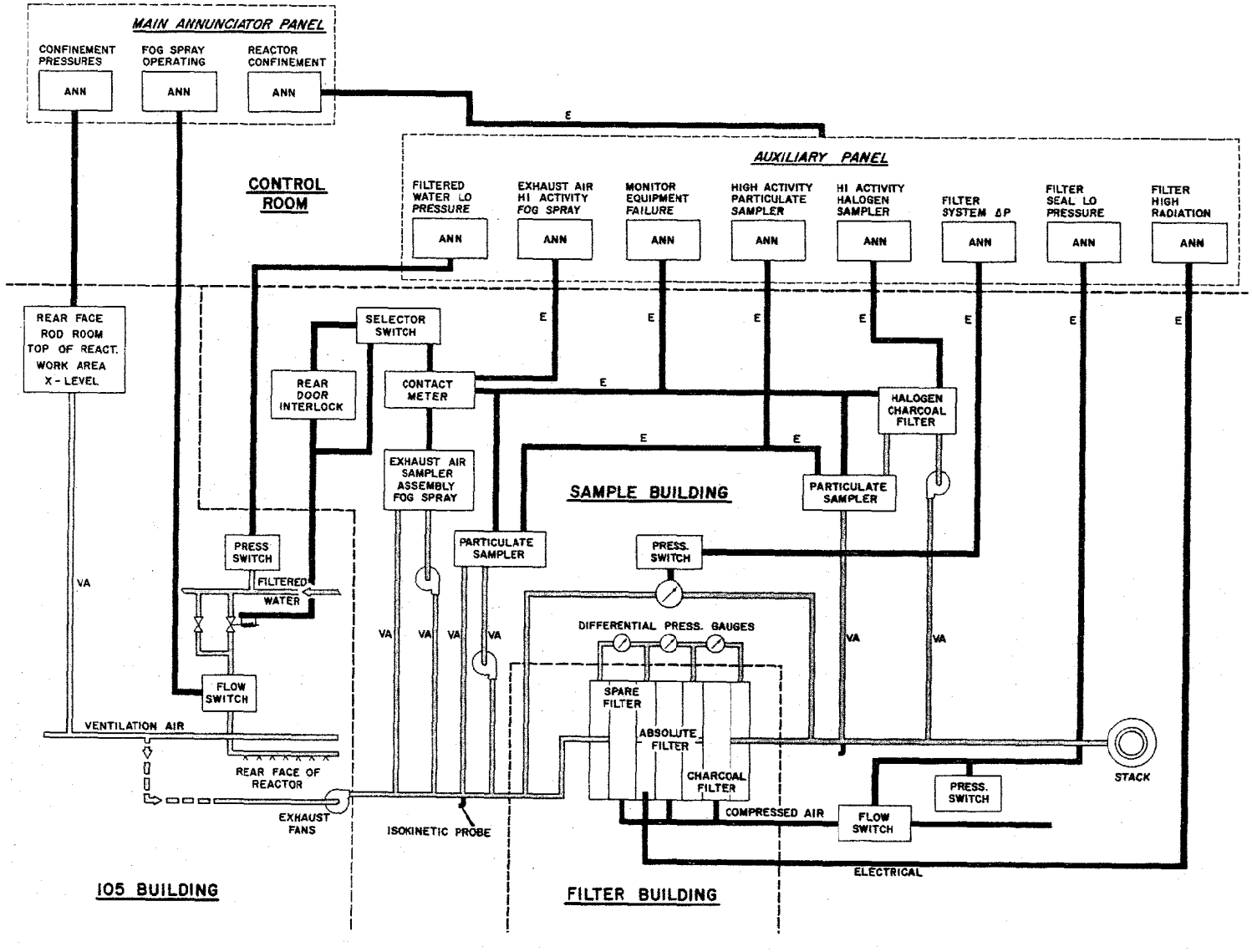


FIGURE 6
ENGINEERING FLOW DIAGRAM

7. High radiation activity on the halogen sampler.

8. High pressure drop across filters.

9. Low air pressure on filter seal.

10. High radiation activity on the filters.

NPR

Ventilation Zones

The NPR reactor building has been divided into five ventilation zones. These are shown on the next slide (Figure 7), and consist of:

1. The primary confinement zone, or zones, that surrounds all portions of the nuclear system containing the high pressure high temperature reactor coolant.

2. The intermediate confinement zone, or zone II, that encloses all portions of the reactor block that are not included in the primary zone.

3. Zone III includes the work areas adjacent to the confinement zones where radioactive material is handled but the potential of a nuclear incident is essentially non-existent.

4. Zone IV includes the shops and office space, is at atmospheric pressure and vents directly to the atmosphere.

5. Zone V includes the control room and has a separate ventilating system.

Zone Control

During normal operation the exhaust from both the primary and intermediate confinement zones of the reactor building is filtered, but the exhaust from the heat exchanger cells and pipe gallery is exhausted to the atmosphere. In the event of a minor incident (one where the pressure buildup remains less than a nominal 2 psig) the fog spray system will be actuated at $\pm$ 1/4 psig to lower the building pressure and the filters will be closed off to prevent damage. When the fog spray reduced the pressure below 1/4 psig the exhuast air will be filtered in the normal manner.

During minor incidents in the heat exchanger building, the exhaust from the heat exchanger cells and pipe gallery is diverted through the reactor building exhaust filters by way of the common wall ventilation barriers. Again the fog spray and filter isolation systems are used if necessary. Schematic examples of these conditions are shown on the next two slides (Figures 8 and 9). These slides also define the path of the exhaust air and steam in the event of a major incident.

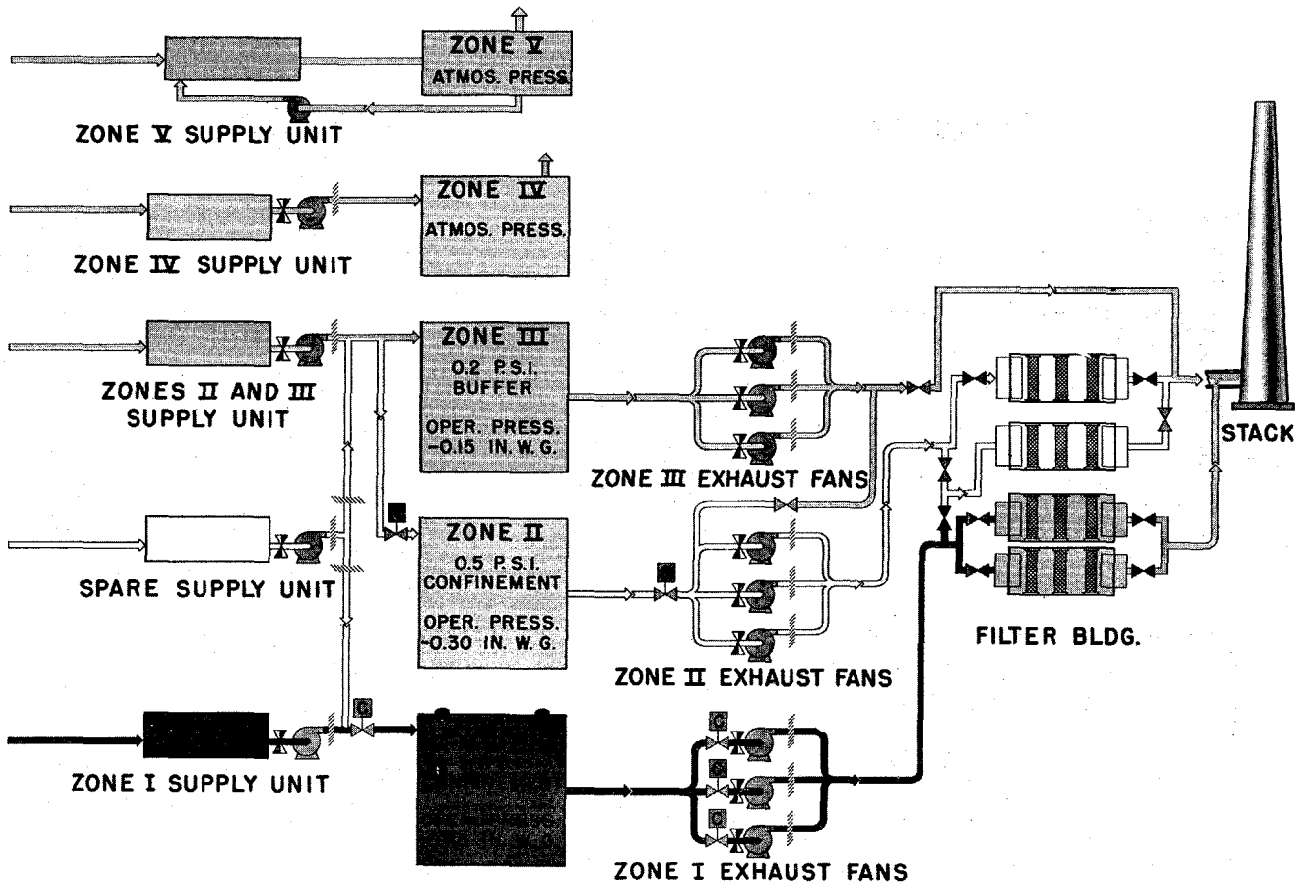


FIGURE 7
NPR VENTILATION FLOW DIAGRAM

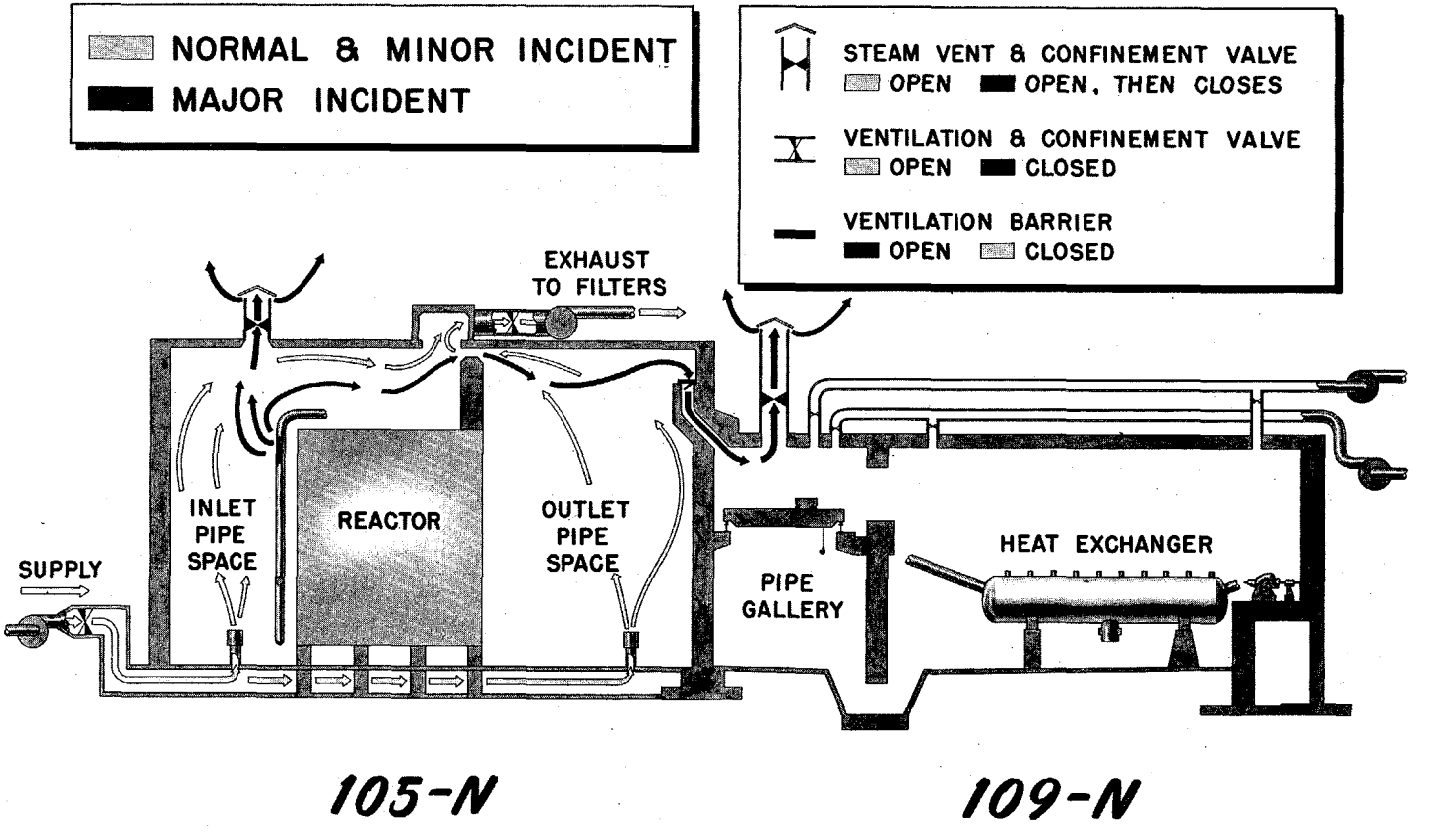


FIGURE 8
AIR & STEAM FLOW - 105-N

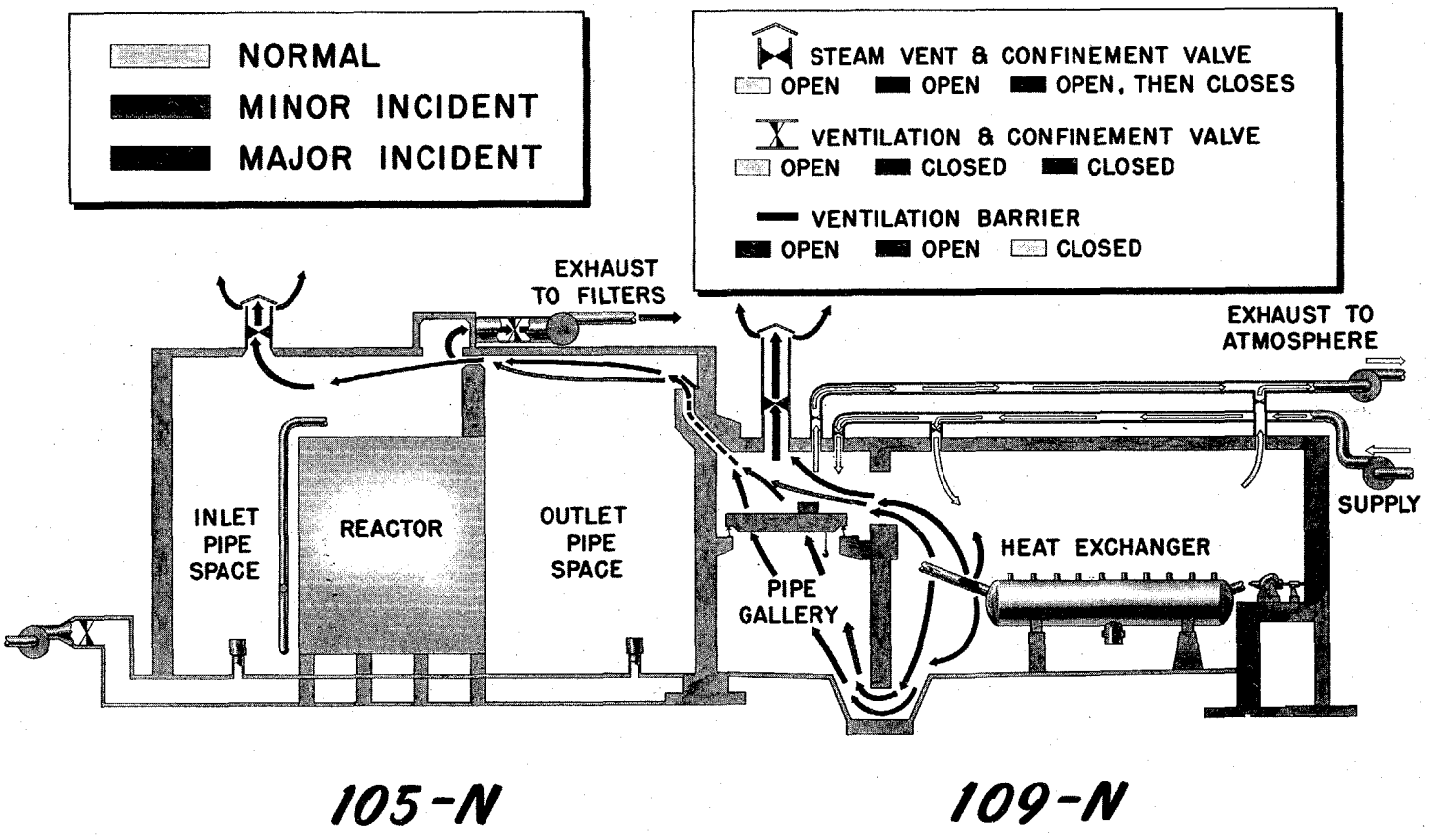


FIGURE 9
AIR & STEAM FLOW - 109-N

A major incident is considered to include the rupture of a major line carrying high pressure high temperature reactor coolant. When the pressure in the zone reaches 2 psig the steam vents, shown in the next slide (Figure 10), open and remain open until the pressure build-up is released. The vents close automatically when the steam release can be controlled by the fog spray. When building pressure drops below 1/4 psig, all exhaust air can be diverted through the reactor building exhaust filters by manually opening the isolation valves. Backup closures are being provided for emergency use in the event the primary closure fails to function.

Difference in Two Systems

Although the same basic philosophy has been used in both NPR and the old reactors, the confinement system for NPR has been complicated by the steam venting requirement. The fact that the NPR system is being constructed as part of a new building has permitted certain refinements to be included which were not feasible in the modified system. One major refinement is the fact that the zone I and II confinement zones in the NPR buildings will be nominally leak tight.

TESTING PROGRAMS

General

The testing program in support of the existing reactor confinement project consisted essentially of design tests or tests of specific components rather than development tests in which a component or system was developed from basic criteria. Tests concerning spacing the rear face fog spray nozzles, exhaust air filter frame seal, and ORNL irradiated uranium burning tests were previously reported by Walker[1]. As mentioned before, Mr. F. E. Adley[3] will present additional information on the filter life and environmental component tests. Other tests of interest were the Halogen Collector Test Program[4], the in-place DOP filter tests, and the absolute filter humidity tests.

Halogen Collector Test Program[4]

This test, performed by the Arthur D. Little, Incorporated, of Cambridge, Massachusetts, was by far the most extensive and costly test program performed in support of this project. This test program was undertaken to provide pilot-plant data on laboratory-proven methods of removing trace quantities of radioactive iodine from an air stream.

Basically the test evaluated the effectiveness of silver plated copper mesh, activated charcoal, molecular sieve and sodium thiosulphate crystals as candidate halogen collectors. The tests were conducted at 70° and 160° F and 70 and 95 percent relative humidity. The iodine injection rates were 0.01, 0.1, and 1.0 ppm. The analytical method was a tracer technique using Iodine 131 added to Iodine 127.

Efficiency and loading tests were performed on each candidate halogen collector. Of the units tested only the activated charcoal collectors achieved a high iodine removal efficiency over a sustained period at various operating conditions (shown in the next

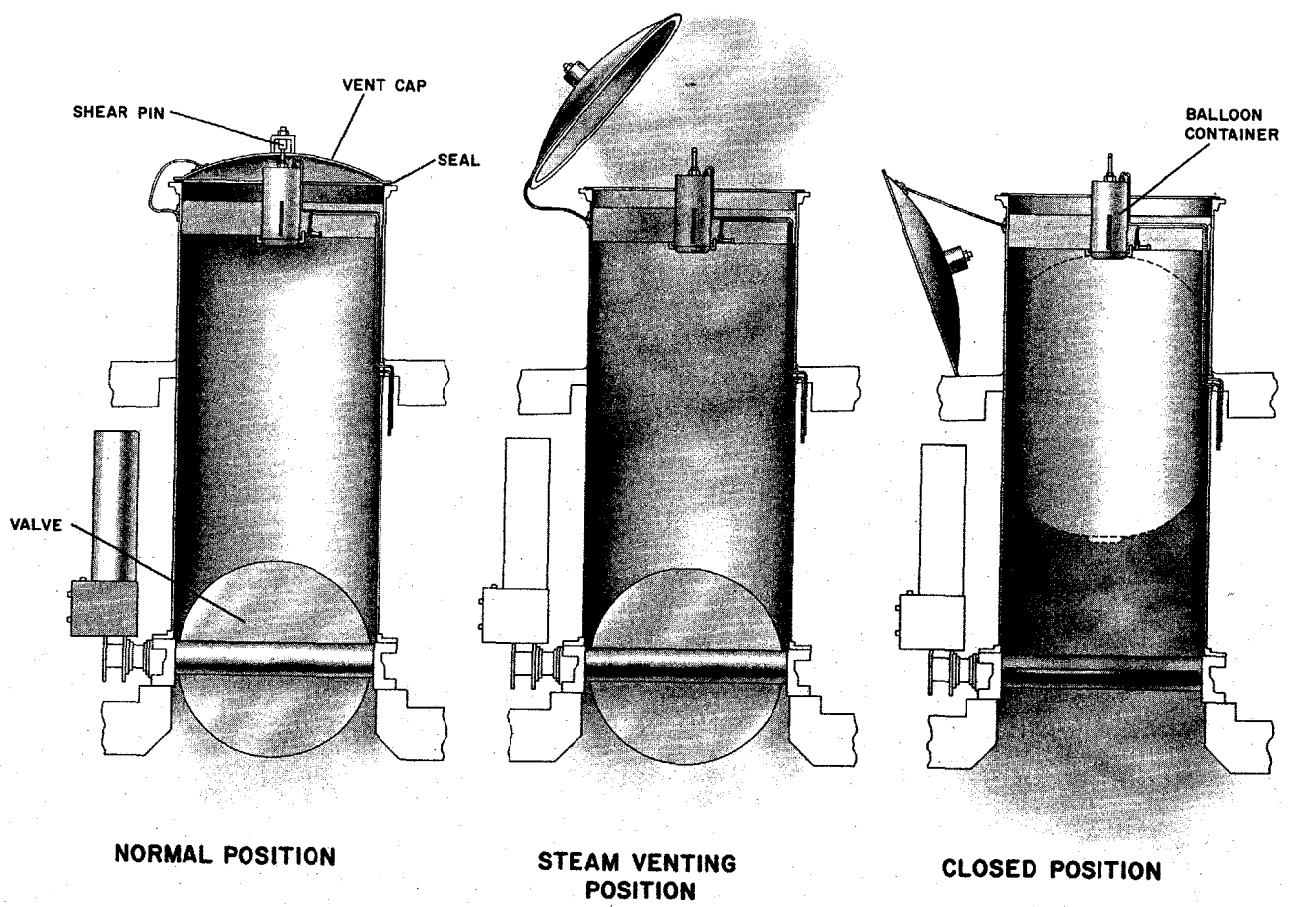


FIGURE 10
STEAM VENT

slide, Figure 11). Because of the capacity of the charcoal, loading tests on these filters were discontinued without evidence of a break-through despite a total iodine loading of 1535 grams per filter. Desorption tests conducted for 45 hours following the loading test gave a total iodine leakage of 6.85 grams or slightly less than 0.5 percent. Although it was planned to investigate filters with both one inch and one-half inch beds of activated charcoal, information received from the filter vendor after the completion of testing revealed that all filters tested contained a one-half inch bed of activated charcoal.

The molecular-sieve collectors attained a high efficiency for a brief period. They began to lose their effectiveness when their great affinity for water produced a break-through. The sodium thiosulfate collectors did not attain a high iodine removal efficiency and failed after a short testing period because of crystal attrition. The silver-plated copper-ribbon bed attained high efficiencies until cumulative iodine loading reached 12 grams for four filters. At this point the iodine removal efficiency began to decline steadily. The second set of silver plated copper beds never attained satisfactory removal efficiencies. This was believed due to the migration of a tar, used in manufacture, onto the mesh during the degreasing operation. The decision was made to use activated charcoal with a one inch bed depth as the halogen collector. The over-all efficiency of this system, as established by the tests conducted by Arthur D. Little, Incorporated, is considered as 90-96 percent although in most instances the efficiency exceeded 95 percent.

In-Place DOP Testing

To insure the high degree of filtration efficiency required of the confinement facilities, the high efficiency filters were tested for leakage and damage following installation. The procedure [5] for checking the integrity of the filter installation was an adaption of a method developed by the Naval Research Laboratory for testing Naval absolute filter installations.

The method consists of generating an aerosol upstream of the filter bank and probing the downstream face of the bank with a particle detector. The portable generator developed by the Navy produced a polydisperse aerosol by the atomization of liquid dioctylphthalate (DOP) with compressed air. The particle size is adjusted by varying the air pressure to the portable generator. A particle detection instrument was also developed by the Navy, however, commercial detection instruments are available and were used in our installation.

The sensitivity of the portable test equipment developed for testing the confinement filters has been demonstrated to be more than adequate for leak detection purposes. In comparison tests with the standard DOP filter testing equipment at Hanford, it was found that the portable equipment could discriminate between filter units which had just failed or passed the efficiency requirements of 99.95 percent removal at 0.3 micron particle size. While the field test was not meant to verify the filtration efficiency of the installed filters, our experience indicates that the test is reliable for detecting the leakage of unfiltered air through the filter.

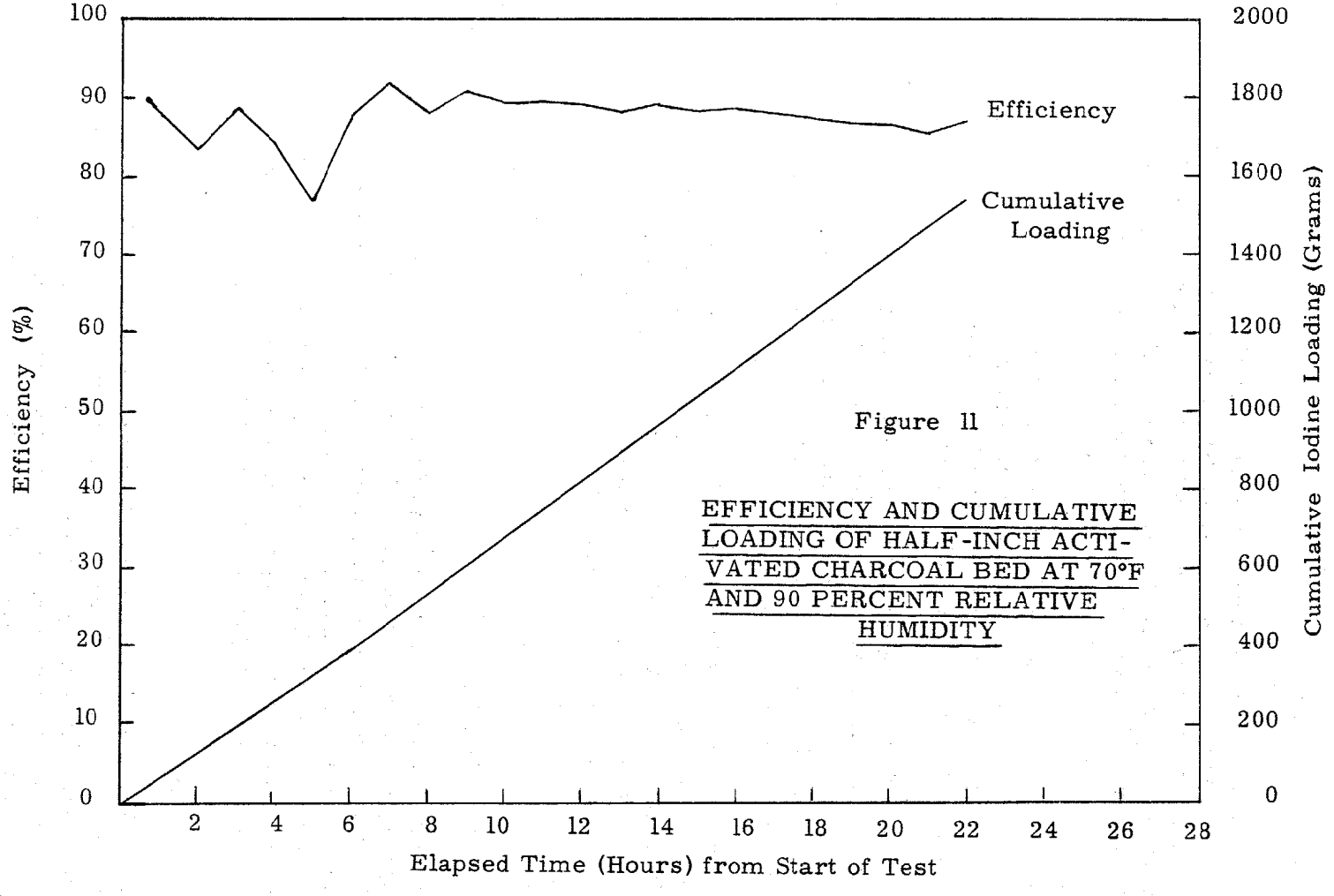


Figure 11

EFFICIENCY AND CUMULATIVE LOADING OF HALF-INCH ACTIVATED CHARCOAL BED AT 70°F AND 90 PERCENT RELATIVE HUMIDITY

SYSTEM OPERATION

General

The initial existing reactor confinement system was placed in operation in November, 1960. Since that time filter installation has been completed at all of the reactors and ventilation balances have been completed in five of the eight reactors. These balances have proven to be tedious due to the magnitude of the system, the low pressure differential that is required between areas within the zone and the fluctuation of building pressures caused by varying wind velocities.

The confinement system has functioned satisfactorily when reacting to a simulated reactor incident during the system acceptance test. Operation of the exhaust fans and automatic shut down of the supply fans produced 0.3 inch water guage negative pressures, as referenced to atmospheric pressure, within the building.

As mentioned earlier the fog spray system was very effective in controlling the spread of contamination from an incident involving the partial burning of an irradiated fuel element. No significant levels of contamination were found on ventilating equipment downstream of the burned element, although earlier incidents had significantly increased the radiation levels of this equipment.

Operating experience has indicated that filter life may exceed expected values. This in general is attributed to the installation of dust baffles upstream of the intakes to the supply fans, operation of the washers on the ventilation air supply the year round, and good housekeeping practices within the building. A special effort has been made by occupants to eliminate or control dust producing activities.

Problem Areas

Building Low Pressure Alarm

Probably the most serious operating problem to arise is the difficulty of maintaining adequate pressure sensing ranges in the confinement zone without continually alarming the system. Since the buildings are large and leakage rates high, it has been practically impossible to maintain the pressure range within the 0.02 to 0.06 inch water gauge negative with respect to the atmospheric reference. Although the system functions well when responding to a simulated incident the problem of eliminating spurious warning signals has not been fully resolved.

Exhaust Air Humidity

Following the installation of absolute filters in one reactor building, certain abnormal maintenance problems caused the exhaust air to reach 100 percent humidity. Although permanent correction will require a period of months, temporary relief was obtained after approximately four months by shutting off the washers on the ventilating air supplied to the building. Visual inspection of the filters after approximately four months service in this high humidity air showed the filters to be wet but in apparently good condition.

Fog Spray Activation

At first it was planned to activate the fog spray system by a signal from a scintillation detector that measured the concentration of 0.36 MEV Gamma due to radioactive iodine in the exhaust air stream. Test results from the first installation were unsatisfactory since the background fluctuations often exceed the calculated high trip limit as much as 100 fold. Secondary radiation caused by irradiated Argon 41 in the exhaust air stream flooded the iodine channel being monitored and made this method of direct measurement useless in the detection of low concentrations of iodine.

The condition was corrected by substitution of a linear dual differential system to identify the presence of Iodine 131 in the exhaust air. Two channels were measured: 0.36 MEV or Iodine 131 and all energies above 1 MEV. The iodine signal is corrected by subtracting from it a portion of the signal indicated by the higher energy channel to compensate for the secondary radiation. The difference in these two channels drives a difference meter that furnishes the signal to activate the fog spray.

Absolute Filter

The fragility of the absolute filters was of some concern early in the project. Specifications were written requiring vendor qualification before placing the final order. Some difficulty was experienced with the filters not meeting the 100 percent humidity requirement in the specification. It was later determined that Hanford's testing method was too severe since water droplets were entrained in the air stream. The final results of the testing on filters for the project were: 1440 accepted for use, 301 rejected for exceeding dimensions, 199 rejected for gaskets, and 27 failed to pass the DOP test.

Filter Frame Seal

Some difficulty has been experienced recently in the failure of vulcanized joints on the inflated neoprene filter frame seal. To date, nine of the 56 seals used in the halogen filter banks have failed. This problem has not been fully resolved.

CONCLUSIONS

Confinement facilities have been provided for the eight old reactors and are being included in the New Production Reactor. Functional tests of the installed equipment have proven compliance with design criteria. During tests simulating a nuclear incident the confinement zone pressure has reduced to 0.3 inch water gauge negative as referenced to atmospheric pressure.

Preliminary data indicate that filter life should exceed expected values if operating conditions remain unchanged. Tests by Arthur D. Little, Incorporated, indicated the superior performance of a bed of activated charcoal as a halogen collector when compared to other dry halogen collecting agents.

The NPR confinement system is designed to control building pressures either through a fog spray or by opening of steam vents. For pressure buildups of 2 psi or less the vents remain closed. When the fog spray reduces the building pressure below 1/4 psig the building may be exhausted through the filters. Pressure buildups greater than 2 psig will be vented to atmosphere via the steam vents prior to fog spray actuation and the routing of all exhaust air through the filters. Several refinements have been designed into the NPR system that were not possible in the modifications for the older reactor areas.

One problem, largely nuisance in nature, has not been fully resolved. It originates in the spurious alarms caused by fluctuating pressures in the confinement zone in the older areas. These alarms require continuous investigation by operating personnel and are normally caused by variations in direction and velocity of area wind.

ACKNOWLEDGEMENTS

The authors wish to express their appreciation for the assistance received in preparing this paper. Those who were especially helpful were: E. L. Etheridge, G. E. Wade, Dr. R. E. Trumble, and Dr. J. E. Wade.

REFERENCES

1. Walker, R. C., July 1959: A review of the Existing Reactor Confinement Program at Hanford; Sixth AEC Air Cleaning Conference at Idaho Falls, Idaho, TID-7593, pp. 365-374.

2. Private communication with G. E. Wade, September, 1961 on NPR confinement system criteria.

3. Adley, F. E., September 1961: Life Loading Tests on Certain Filter Media; Paper presented at Seventh Air Cleaning Conference at Brookhaven National Laboratory, October, 1961.

4. HW-65587 (ALI-54), March 1960, Halogen Collector Test Program; Prepared for General Electric Company, Richland, Washington by Arthur D. Little Incorporated, Cambridge, Massachusetts.

5. Etheridge, E. L., September 1961; Field Leak Testing of HAPO Reactor Confinement Filter Installations; HW-71083, General Electric Company, Richland, Washington, 16 pages, Unclassified.

DISCUSSION

ROBINSON: Have you had any experience with the fog spray causing filter damage or plugging when it is in operation?

HEACOCK: We have not. However, we have had, for over four months, in several of the reactors saturated air going through the filters. We have visually inspected them and found they were wet. In fact, there was water in puddles on the floor, but we could detect no breakthrough of the filters under the saturated flow conditions.

RECENT AIR CLEANING DEVELOPMENTS AT ARGONNE NATIONAL LABORATORY

C. L. Cheever
Argonne National Laboratory

INTRODUCTION

This paper reviews studies we have made on filtration and characterization of plutonium combustion fume. The possible release of plutonium to the environment is, of course, a concern wherever dangerous amounts of plutonium are handled. Because of the severe health hazard, it is common practice to use AEC high efficiency filters for exhaust air cleaning in plutonium facilities.

We have conducted small-scale experiments with the objective of (1) determining the percent of fume penetration through AEC high efficiency filter media, (2) characterizing plutonium combustion fume, and (3) determining the percent of plutonium which becomes airborne and reaches the filter media.

It was recognized that our tests were made under a particular set of conditions which would not prevail during an accidental plutonium fire. However, we hoped to obtain information which would be useful for consideration of plutonium hazards.

AEC FILTER PENETRATION TESTS

The tests were conducted in a plutonium system glove box. The box atmosphere was filtered room air, and moisture content ranged from 30 to 70 grains per pound of dry air. Figure 1 shows our first test assembly located inside this box. 1-5/8" I.D. copper tubing was used as the air duct. Fiberboard flanges, bonded to the tubing with epoxy resin, held the high efficiency filter media. A nichrome wire coil connected to a rheostat was used to ignite one to two gram amounts of plutonium. Plutonium was in the form of a slab, cylinder, or small turnings. High efficiency filter media was placed over the inlet to remove dust particles in the box atmosphere. We chose an air flow rate of 5 fpm as this is the rated flow through the media in high efficiency filters. A new test assembly was used for each run.

Subsequent assemblies were built with the burn section placed at a lower level than the filter section to prevent possible spattering of plutonium onto the first filter. This also reduced the chance of getting plutonium burn residue onto the first filter. In addition, a plastic pouch containing tweezers and glass jars

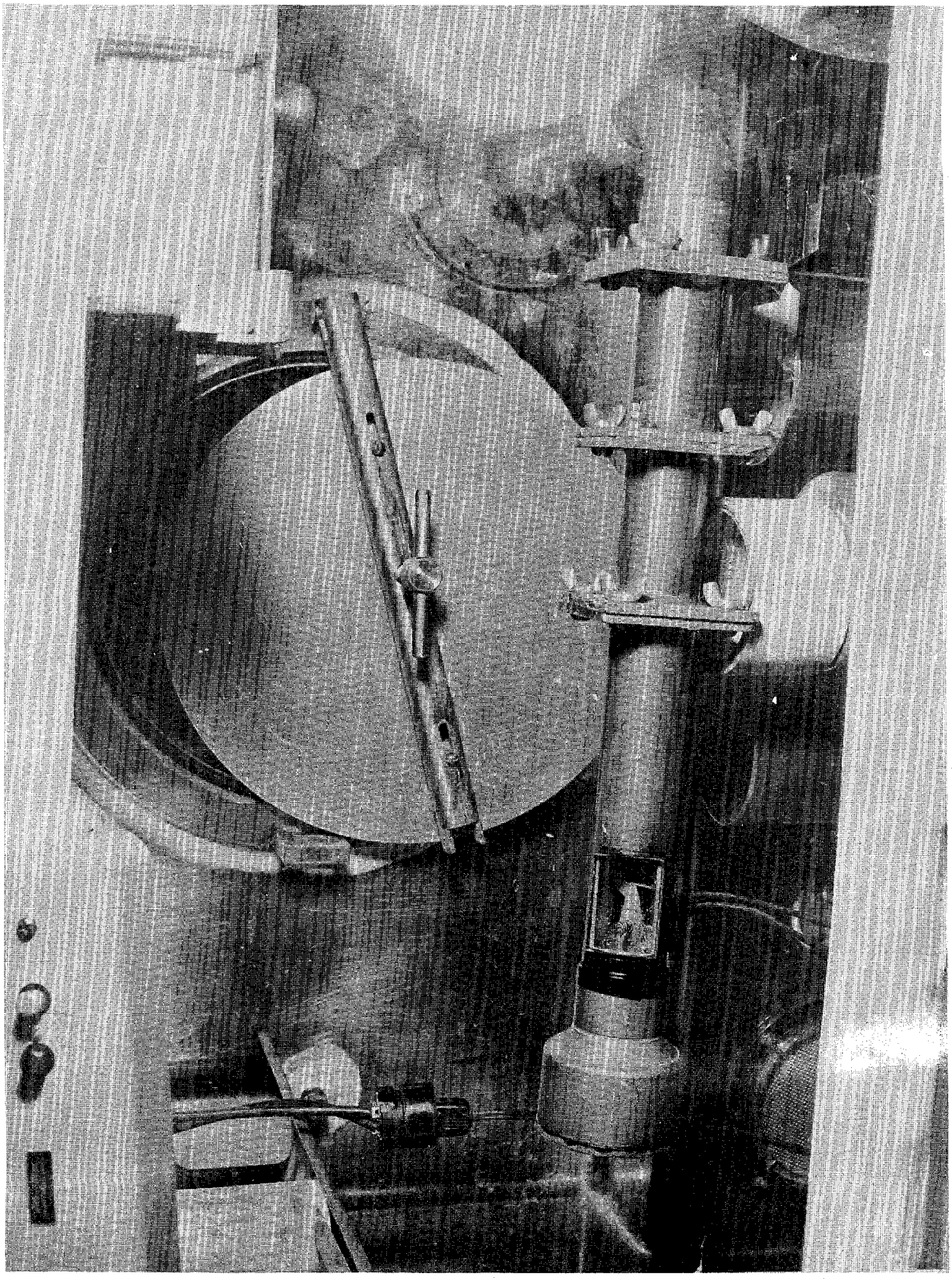


Figure 1

was placed over the filter section. From three to seven filters were placed in series in this section. At the completion of the test, filters were removed from the flanges and placed in the jars within the plastic pouch, starting with the downstream filter and working back. The samples were pouched out of the glove box and placed in an exhaust hood where they were relocated into clean glass jars. They were then counted by the Background Radiation Group after the filter media had been dissolved in a mixture of hot concentrated hydrofluoric and nitric acids and plated out on stainless steel planchets.

The results of four test runs indicated:

1. Average penetration of the first filter was about 0.5%. Penetration ranged from approximately 0.005 to 0.9%. In all tests, the activity on the second filter in series was less than 0.6% of the activity on the first filter.

2. An average of 0.00055% of the plutonium that was burned was collected on the filters. The range was from 0.00008% to 0.0012%.

The efficiency of the first filter in series was calculated from activity collected on the subsequent filters, with the assumption that penetration of the last filter was not significant. J. J. Fitzgerald and C. G. Detwiler (1) had found the maximum penetration of AEC #1 filter media by potassium permanganate particles (0.01 u) was approximately 10% at a flow rate of 2 cm per second. We anticipated that we would be able to determine the collection efficiency of the secondary filters. However, efficiencies indicated for these filters were not consistent and further tests are needed. The percent of burned plutonium which reaches the filters will vary with conditions such as the physical state and quantity of plutonium, the amount of deposition on surrounding surfaces, the air velocity, etc.

Argonne's Chemical Engineering Division is now conducting tests to determine the efficiency of various filter media for collection of fume from the controlled release of plutonium hexafluoride.

ELECTRON MICROSCOPE STUDIES

We have also taken samples of plutonium combustion fume for electron microscope observation. The sampling technique was electrostatic precipitation of fume directly onto electron microscope grids. An 11,000 volt potential was applied across a 1" gap between a hypodermic needle and a small brass rod holding a grid. Grids had a 100 to 500 A carbon layer vaporized onto a collodion membrane. The carbon provided a conductive surface and physically stabilized the membrane.

The fume source was the effluent from plutonium burn rate studies directed by J. Glenn Schnizlein, Jr. of our Chemical Engineering Division. One-half gram plutonium ribbons, 6.3 cm long, were burned in dry air. The grid sample con-

tained about 19,000 disintegrations per minute alpha activity. Photographs of collected fume are seen in Figures 2, 3, and 4. These photographs were taken by Dr. Raymond Hart of our Metallurgy Division with a Siemans electron microscope. Magnifications were 10,000, 20,000, and 80,000 times, respectively. The average size of the individual particles in these agglomerates is 0.016 microns. They range from about 0.004 to 0.03 microns. Three hundred particles were sized. At high magnifications, discreet particles are identified as cubes. This indicates condensation from the vapor phase.

Electron microscope selected area diffraction patterns were taken of the fume agglomerates. The diffraction rings are seen in Figure #5. The patterns corresponded to information on plutonium dioxide taken from ASTM, X-ray card number 6-0360. A face centered cubic structure with cell size of 5.386 angstroms is reported for PuO_2. Additional electron microscope photographs of fume agglomerates from these tests are shown in Figures 6, 7, and 8. Literally hundreds or thousands of individual particles have formed flume clumps several microns across.

Non-radioactive fumes were collected on AEC high efficiency filter media to indicate the manner in which the fume particles are retained. Collection of zinc oxide fume is shown in Figure #9, and collection of magnesium oxide fume in Figures 10 and 11. Air containing the combustion fumes was drawn through the filter media at 5 feet per minute. A thin layer of fibers was pulled off and boundary areas of this layer were photographed. Otherwise, the filter media is opaque to the electron beam.

Figure #12 shows a thin section of the clean media that was used for fume penetration tests. Fibers from about 0.05 to 3.0 microns in diameter are seen here. The media was taken from a filter which had a penetration rating of 0.01% against 0.3 micron D.O.P. test aerosol.

SUMMARY

We have reported the results to date of our plutonium combustion fume studies. These studies have indicated the nature of plutonium combustion fumes under specified conditions. Individual particles with an average size of 0.016 microns were observed in fume agglomerates, some of which contained thousands of particles.

Plutonium dioxide fume penetration of the first high efficiency filter media in series ranged from 0.005 to 0.9%.

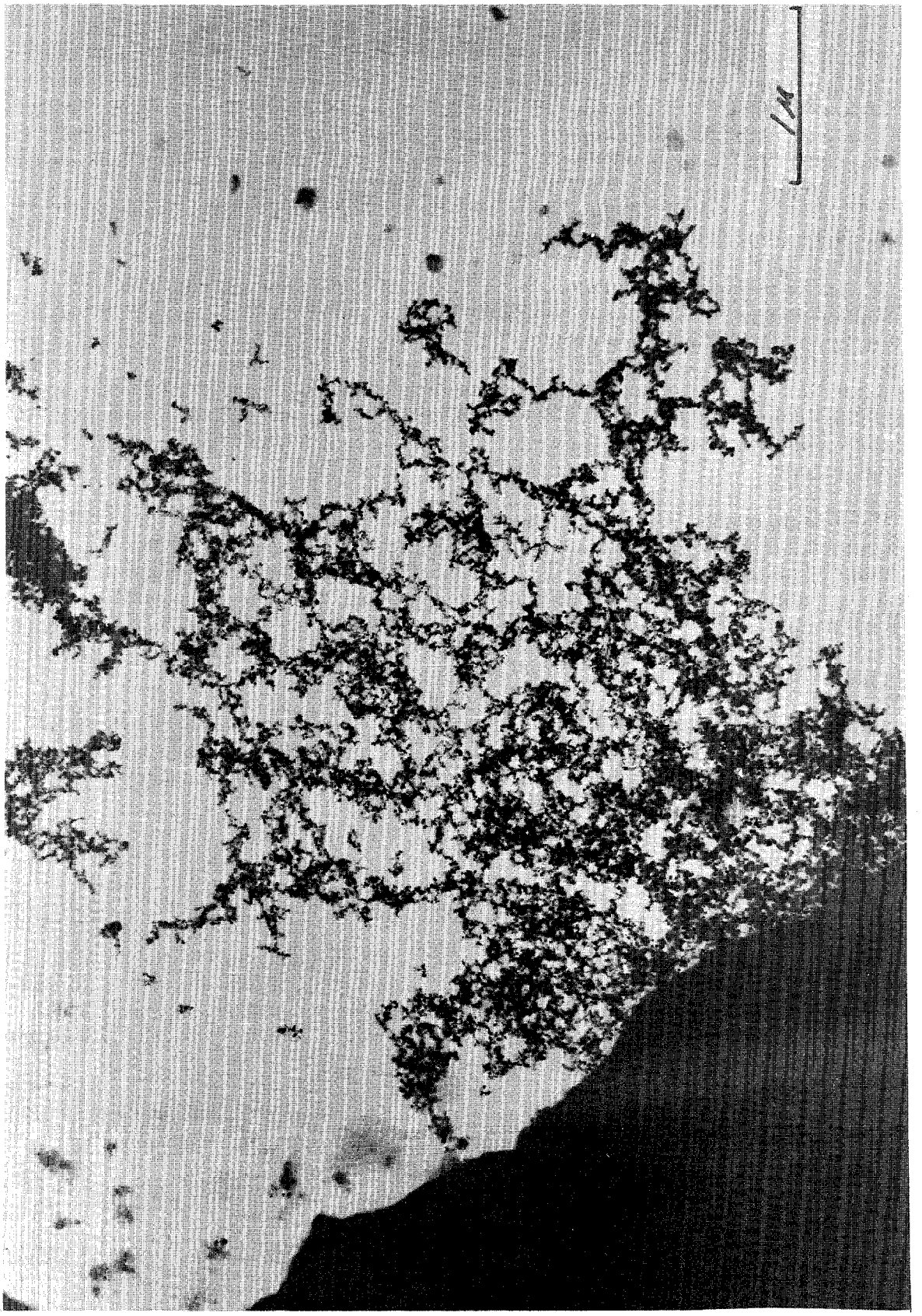


Figure 2

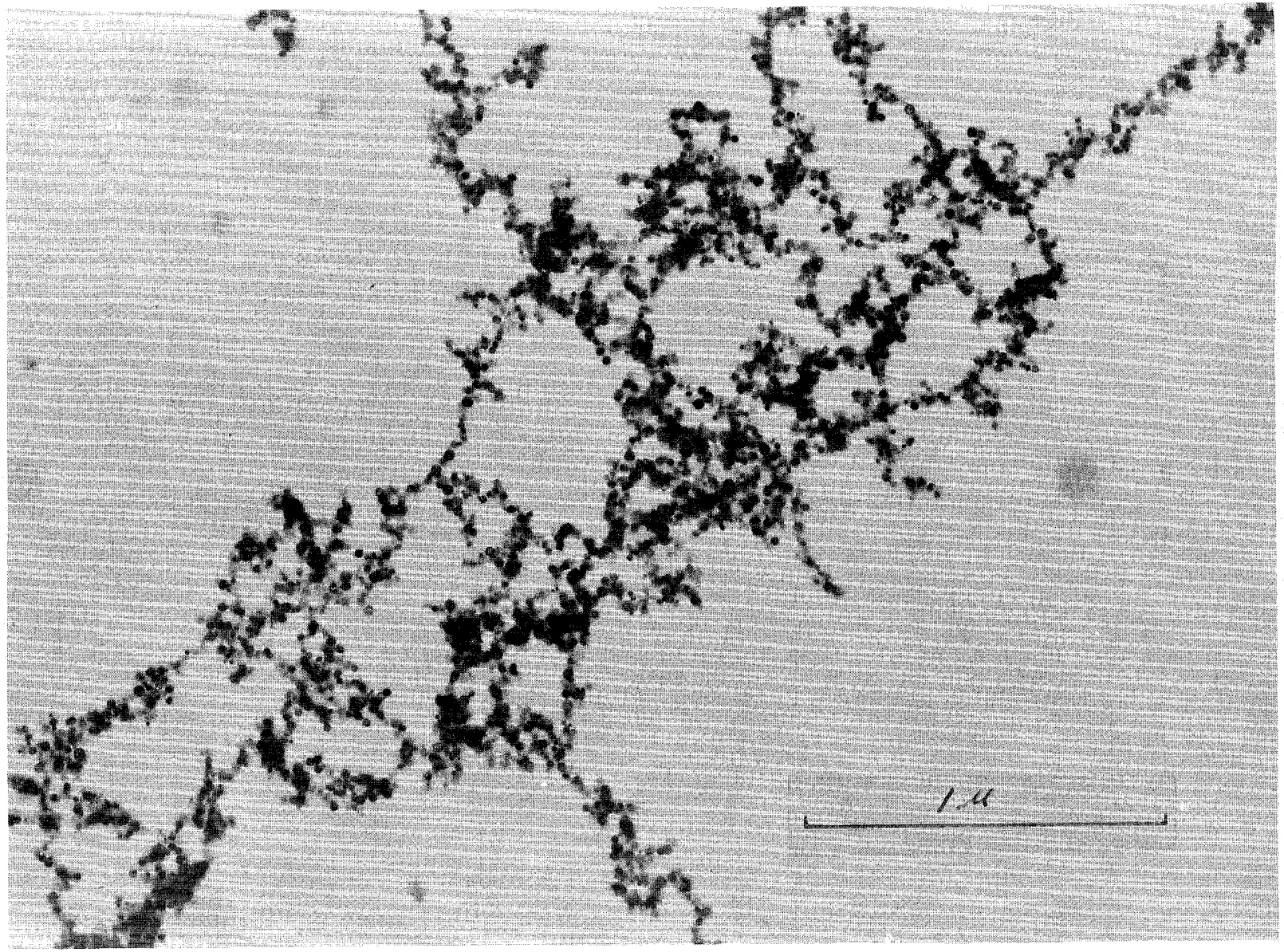


Figure 3

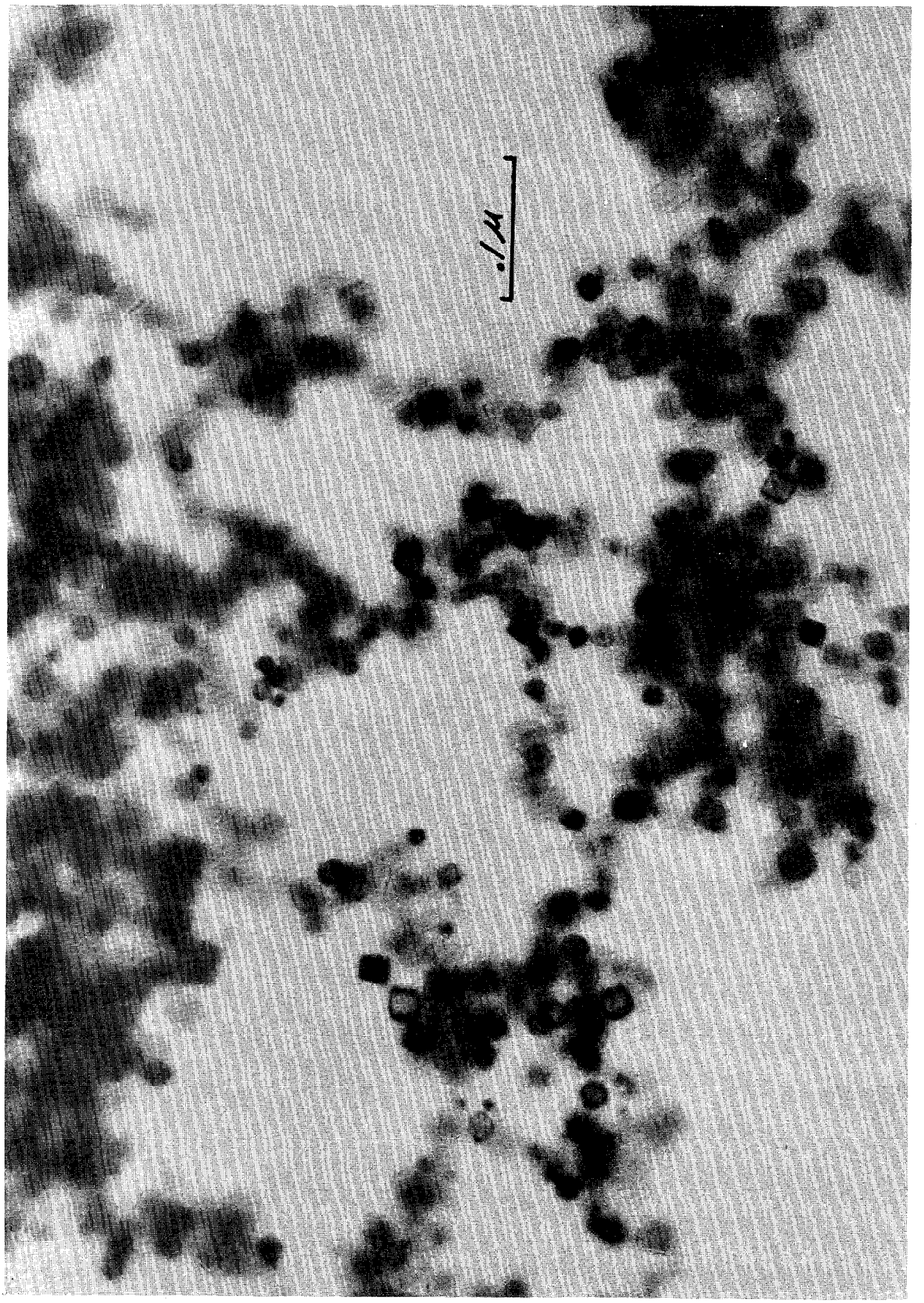


Figure 4

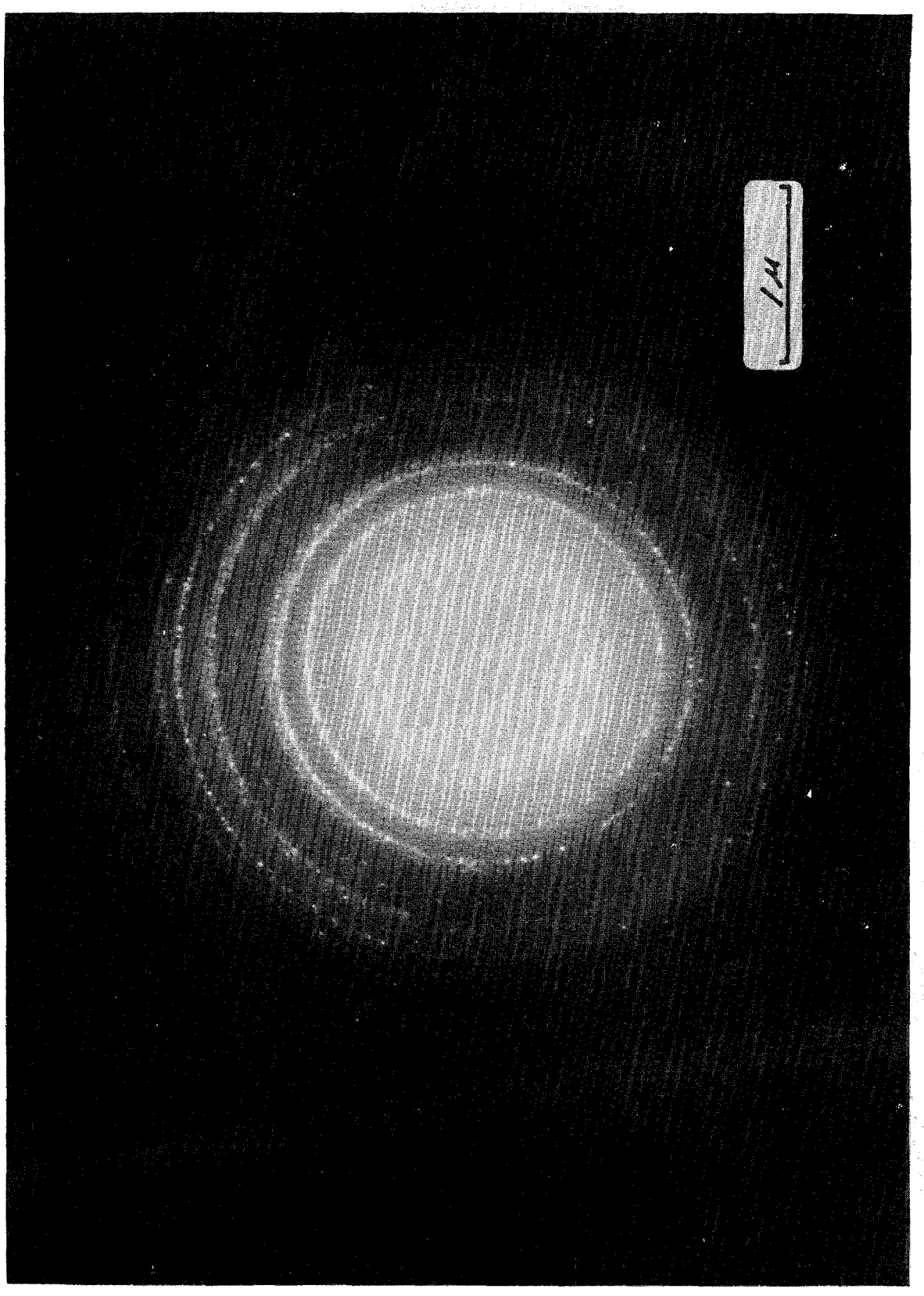


Figure 5

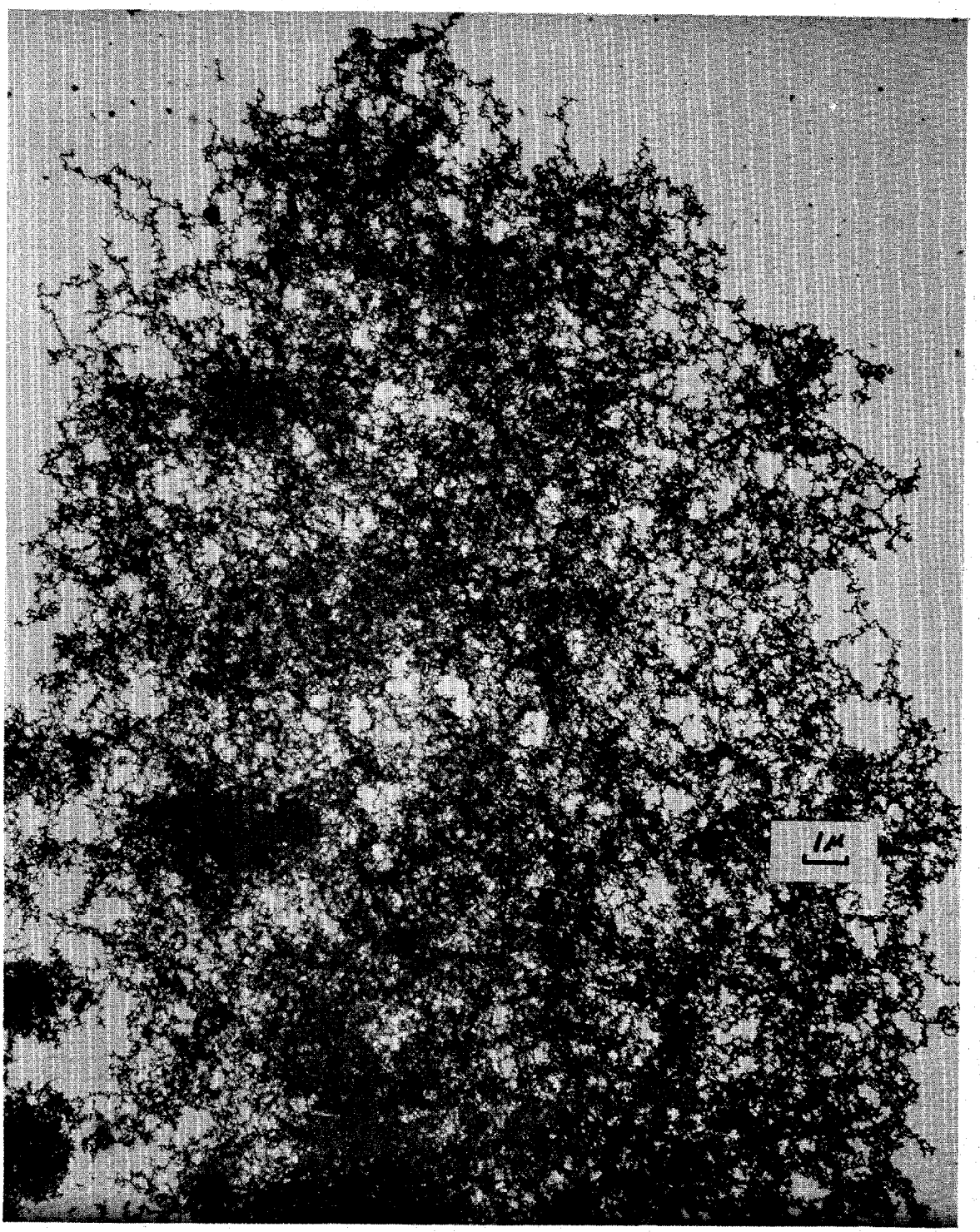


Figure 6

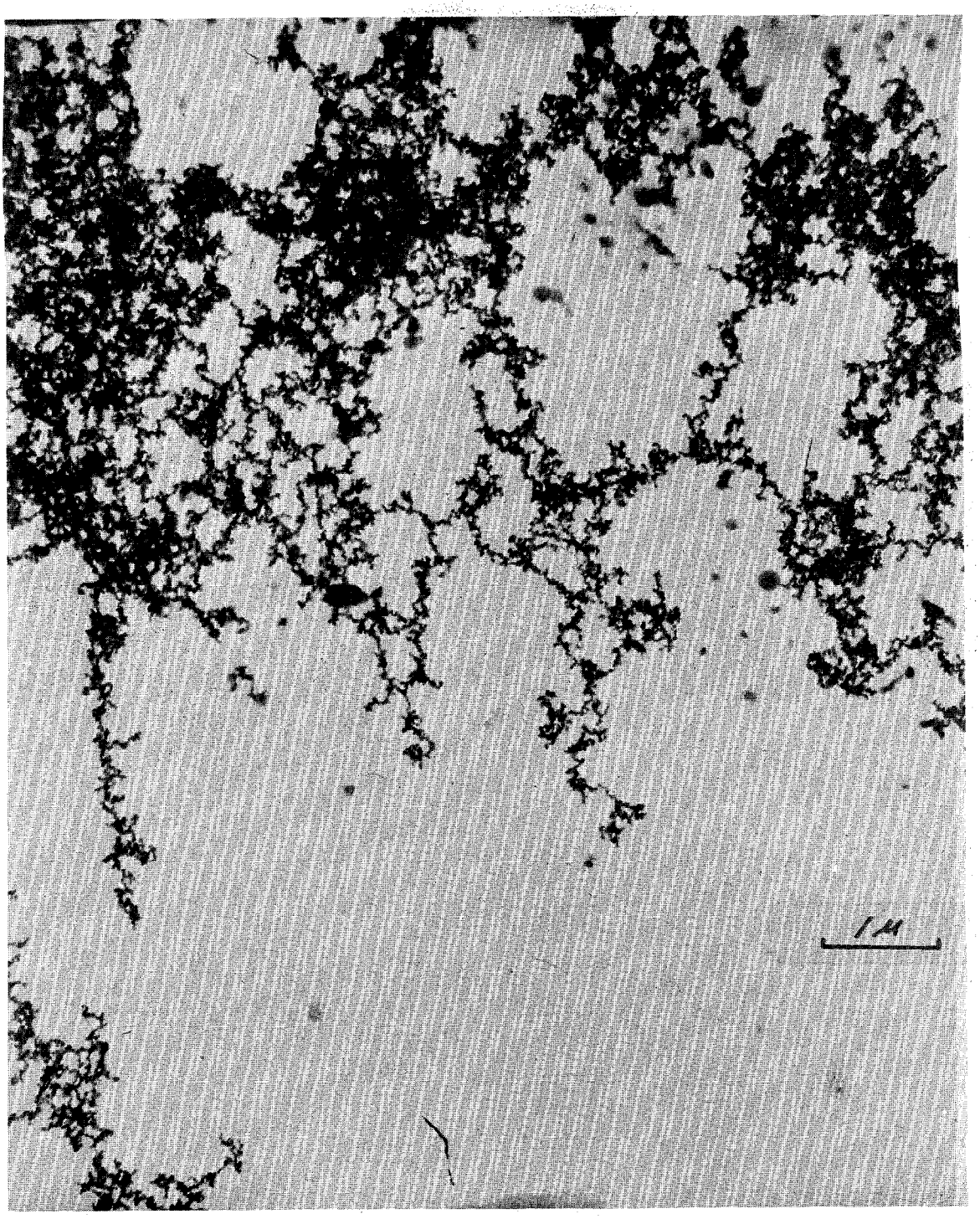


Figure 7

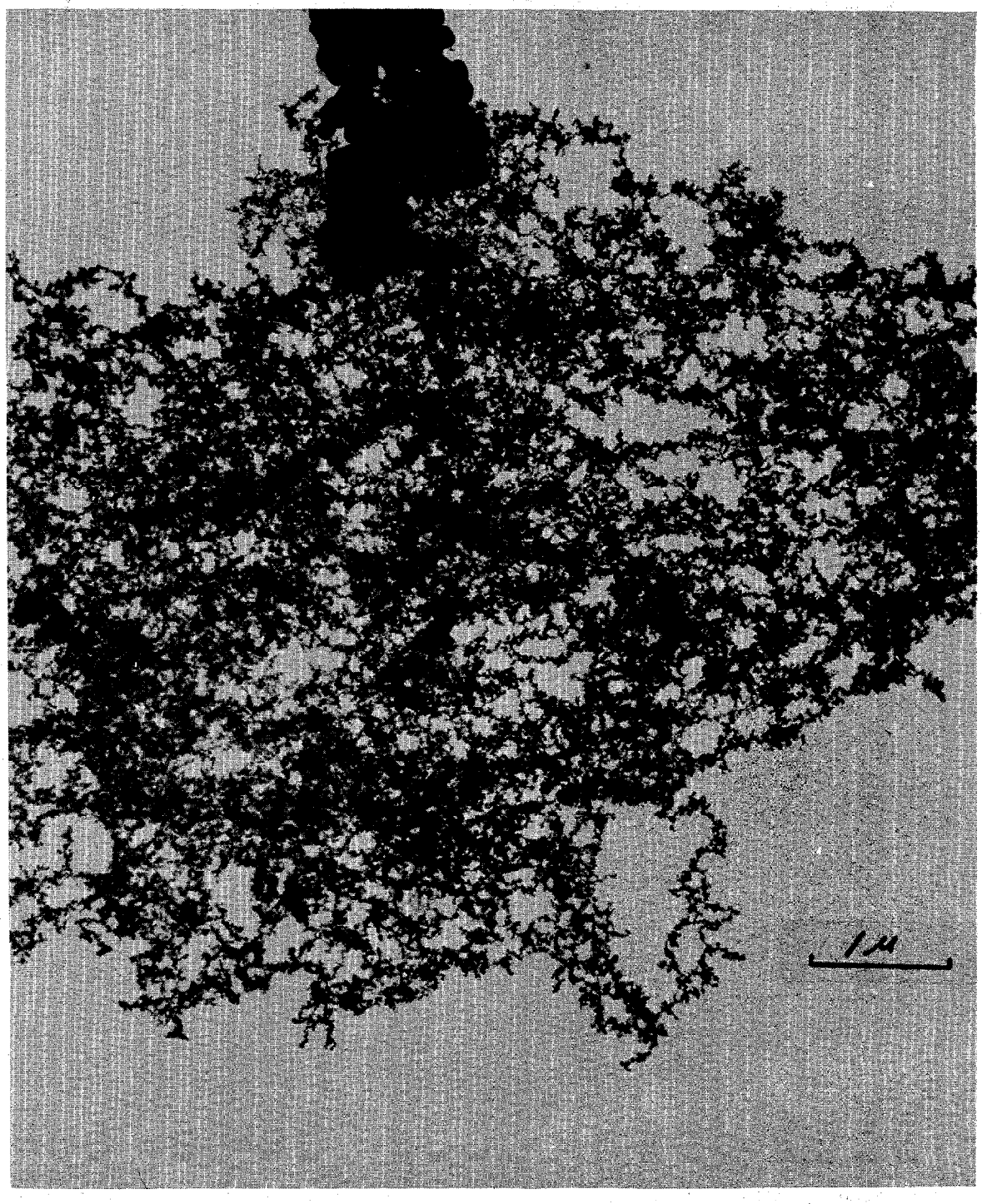


Figure 8

Figure 9

Figure 10

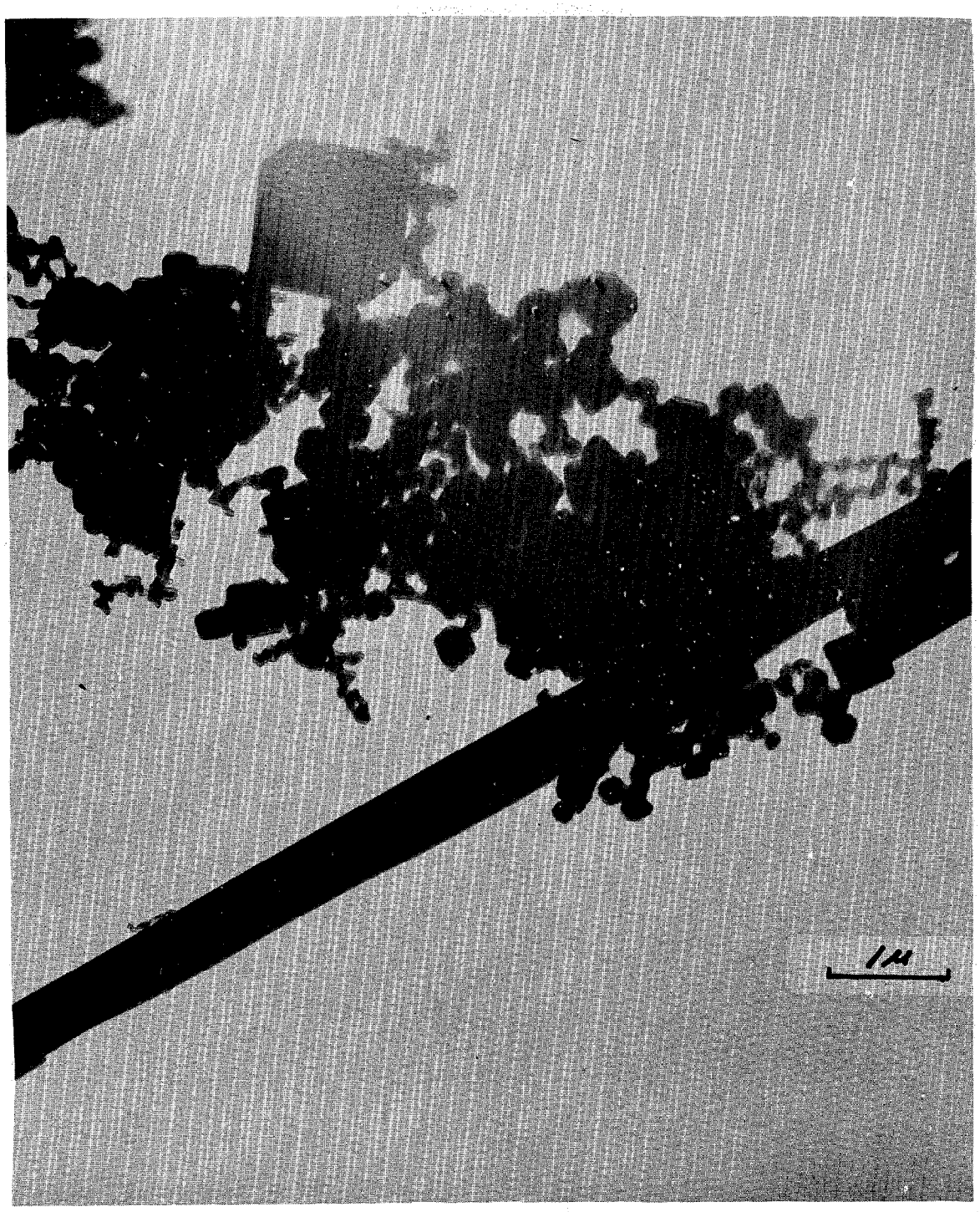


Figure 11

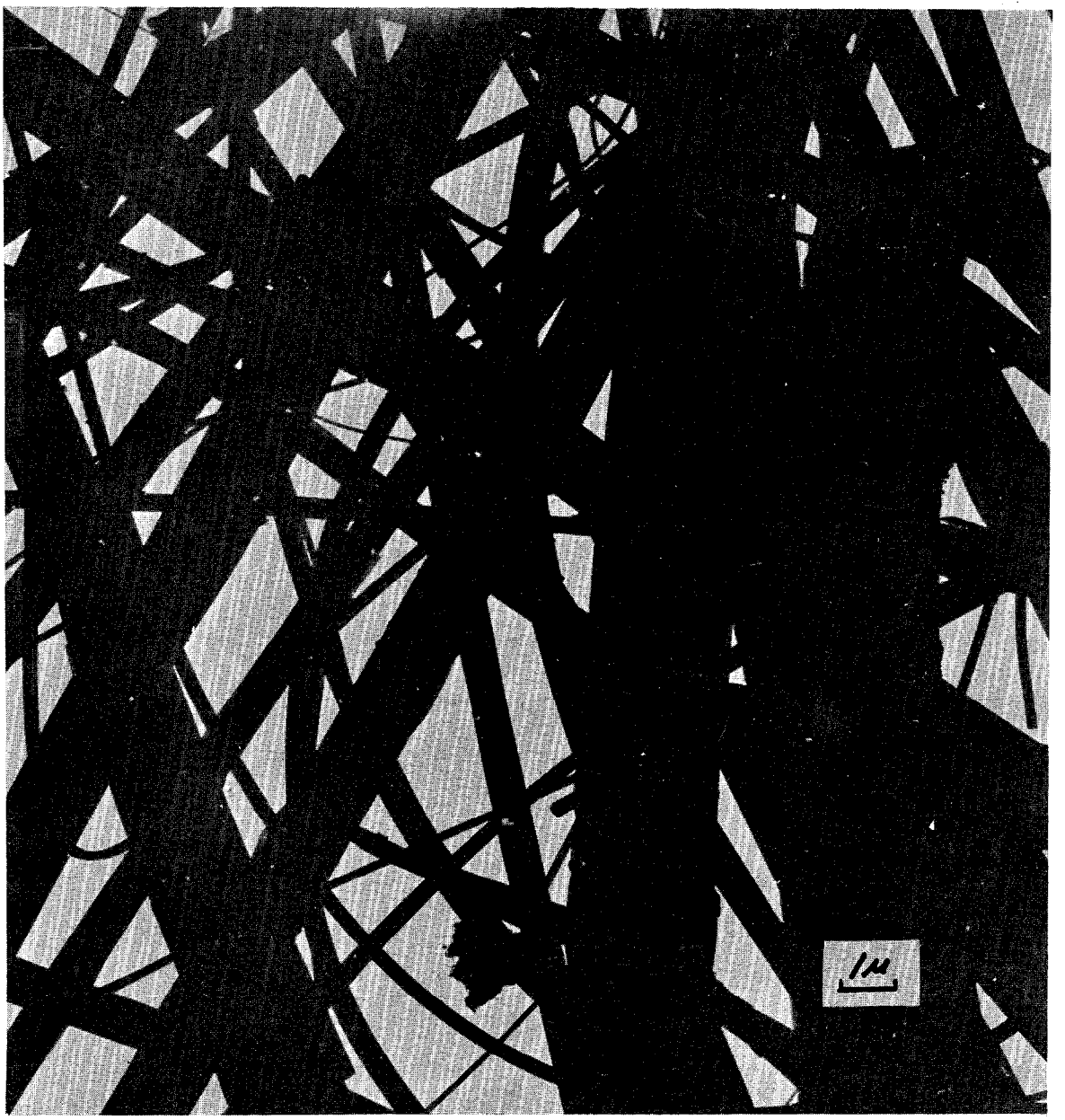


Figure 12

ACKNOWLEDGEMENT

Dr. Jacob Sedlet, Robert L. Mundis, Gilbert C. King, Howard V. Rhude and Odell T. Minick of Argonne National Laboratory contributed to various phases of these studies.

(1) Collection Efficiency of Air Cleaning and Air Sampling Filter Media in the Particle Size Range of 0.005 to 0.1 micron. Fourth Air Cleaning Conference, 1956.

REMARKS

GEMMELL: I think you will all agree with me that probably no group in the United States has done more to further the cause of air cleaning than Dr. Silverman and his people at Harvard. They have gone about the job scientifically and at the same time have performed a great many engineering studies in certain particular phases of air cleaning.

Those of us who have had air cleaning problems have always felt free to call on him and his people for help. Advice and suggestions were always prompt and approximate. Most of us look upon this group as the final backstop for air cleaning problems. It is traditional that at least one half day of this conference be devoted to the studies that have been carried on during the past year at Harvard.

I am sure many of you will recall some of the old timers like Mel First, Dick Dennis and Charles Billings. There are two new faces this year and we are looking forward to hearing from them. It is a real pleasure for me to introduce Dr. Leslie Silverman, who is in charge of the air cleaning program at Harvard. This is a free wheeling gang, so the job of chairman is easy! You just open the gates and away they go.

SILVERMAN: I certainly appreciate the accolades that you passed out. I think we feel that air and gas cleaning problems within the Commission have continued to grow with the growth of the atomic energy program.

During this first discussion I will describe briefly the purpose and intent of the Harvard air cleaning contract with the Commission. The specific activities in each case will best be described by my colleagues who are going to present papers on various aspects of our program.

The Harvard air cleaning contract with the Commission is now in its thirteenth year. It is an R & D contract for air and gas cleaning studies related to nuclear energy processes. We have looked at as many problems as the Commission has tried to throw at us. Once in a while we throw them back because we think they are foul balls but, nevertheless, we try to give a reasonable opinion for doing so in such cases. We are charged with the obligation of collating and collaborating on data from various sites. You have seen some of this done at this meeting in regard to work that has been done at Oak Ridge, following up some of our preliminary work, as well as work at other sites.

We also have the obligation of providing training and education for AEC contractor personnel or people that the Commission designates. This conference is a representation of that way of getting information to those who can use it. Perhaps every four or five years we run a two-day training and education meeting. The last one was in Boston in 1957, and the first one was in

1951, during these we provide a two-day review of air cleaning fundamentals and aerosol behavior.

We also are charged with the obligation, on occasion, and the occasions haven't been many recently, but they still leave a few unsolved problems -- of acting as an impartial evaluator for the Commission. The Commission, like all Government agencies, is confronted by inventors and manufacturers who have the latest word on air and gas cleaning. Without carrying their ball too far, these are all revolutionary. They are going to replace everything that has been proposed before. The Commission says, "It might be a lot easier if we let Harvard evaluate this, both on paper and later in physical entity if it has all the merit that you attribute to it." I would say that the mortality as far as manufacturers' inventions go has been pretty high. Nevertheless, we have performed several evaluations of new devices and reported them in AEC documents which are easily available.

Lastly, we serve as consultants on general air cleaning problems, as has been indicated. If you have some project on which we might be helpful, and request assistance through Mr. Belter's office in the DRD Sanitary Engineering and Enviornmental Branch, we can be of service.

Our chief interest, however, is our research and development program. That has fallen into several categories dealing with both particulate and gas removal. Some of the approaches used in our R & D originated at Harvard and some are created by problems arising in the field.

We have two obligations that relate to developing information on economics for the Commission. The first one will be reported by Dr. First. We hope this is the last year of our economic survey, which has been in progress for three years. This study is developing figures on air cleaning costs at the various sites. Some of you may have the notion that they may be quite variable. We can certainly confirm that, but some idea of how air cleaning costs compare in the nuclear industry with the non-nuclear industry is necessary to give an idea whether or not any handicap exists.

Certainly the information that Mr. Wehmann presented earlier indicates that in some places you put in better cleaning than required, conversely in other instances it may not be enough. We have also prepared a handbook on air cleaning for the Commission. By the end of the year I hope we will have the completed second edition.

We have been working on an incinerator and air cleaning units for the Commission for small operations, laboratories, hospitals and isotope users. This device is now ready for field trials. We are collaborating with the Army Chemical Center on a scaleup of this device since they are in need of one for their nuclear defense laboratories. We propose to perform an evaluation in the coming year on a prototype.

We have also been working on a scrubber project - as it relates to the containment problems - for the liquid waste disposal problem, that has already been mentioned. The problem of aerosols created by liquid waste reduction has been considered for some time. We are trying to develop a closed circuit device

that would be inexpensive and easy to operate. This process if successful would result in a material transfer of gaseous and liquid operations to a situation where there would be no real secondary waste-handling problem since the final product is a solid.

We have also been asked by the Commission to help on the design of equipment for rare gas removal. Some of our work and a lot of the work at Oak Ridge that Dr. Adams did not discuss is in relation to the adsorption removal of rare gases. Oak Ridge information was presented at the last air cleaning conference and has been very helpful in the design of radioactive gas containment for reactors such as the SL-1.

As you know, the SL-1 was a prototype of a proposed Antarctic reactor. I am not prepared to answer for the Army Reactors Branch as to what will be built. They are still faced with a problem of designing a package for gas containment which will be suitable. The Army found that their design engineers required a package for gas cleaning that was one-third of the total reactor package. They thought that this was too large a gas cleaning system in relation to the reactor size. We are working on that project at the present time. We do not plan to report on it today, but most of the basic data have already been gathered. It is a question now of adaptation to our concept of a gas cleanup system.

The diffusion board and foam projects I will talk about later and therefore I will skip them now.

I will mention the iodine project which Mr. Dennis is going to review and I have already given a little preview of the inplace filter test method.

We have at Harvard two other research contracts which help support, and complement some of the AEC research activities, as well as a little bit of Harvard. Our department has a contract with the American Iron and Steel Institute which has been in operation now for almost six years. This work pertains to cleaning high temperature gases such as those evolved from steelmaking processes. Professor Billings is going to talk about one aspect of this, the filter shock wave cleaning studies, a continuation of what he presented at Idaho. Mr. Robert Gussman is going to describe the condensation device we have used for trying to measure numbers of particulates in relation to the agglomeration studies we have under way.

The other project I want to mention that is AEC oriented is related to filler respirators and the type of protection they afford the wearer. The uranine method we described was applied to respirator filter and fit tests. This is actually used for inplace respirator face piece fit evaluations. This work has been published in Biology and Medicine reports. The project is still continuing and represents an aspect of air and gas cleaning that reaches the smallest possible unit operation of protecting a single individual.

That is all I intended to say at this time. I don't think I can dignify this brief discussion by asking for any questions. I think my colleagues can use the time for question periods quite effectively.