HIGH EFFICIENCY FILTER PROGRAM NATIONAL REACTOR TESTING STATION

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Improvement of the high efficiency filter program at the National Reactor Testing Station was begun two years ago when problems which existed with these filters were discussed at the 6th Air Cleaning Conference. A visual inspection program was the first step toward assuring ourselves that out filter installations were giving a high efficiency performance. At the end of the first year of visual inspections, we found that 55% of all filters received on the site were rejected.

Almost a year ago we began sending our filters to Hanford for testing. The visual inspection program was continued to determine if any damage was occurring during shipment from Hanford to the NRTS. Only 4% of the tested filters have not passed the visual test.

It is worth noting at this point, that an incidence of damage to pre-filters is currently being found. While some of this damage may have occurred during shipping, much of it appears to have resulted from imperfections in the media.

A Filter Advisory Committee (FAC) was formed this past spring. This committee is composed of the AEC Industrial Hygiene Engineer, Chief of AEC Health Physics Section, and three members from Phillips Petroleum Company, a health physicist, an engineer, and a Technical Division representative. The Filter Advisory Committee (FAC) was established primarily to review the present usage of mechanical filters for air particulate removal and to set up a program for the in-place testing of high efficiency filters.

It was believed that high efficiency filters were being used at some places where such a high degree of filtration was unnecessary. An investigation into this matter was thought advisable. Furthermore, the experience of the U. S. Navy and other AEC sites indicated that many filter installations were faulty, resulting in loss of the efficiency requirements of the system.

IN PLACE TESTING

The first action of the FAC was the decision to purchase a smoke photometer for in-place testing (Figure 1). DOP generators were constructed from a model loaned to us by the Naval Research Laboratory, Washington, D. C. This program is currently going into operation. Of four filter installations tested so far, efficiencies on 0.9 micron-sized particles ranged from 93.0% to 99.8%.

HIGH EFFICIENCY FILTER USAGE

The necessity of using high efficiency filters in laboratory fume hoods was questioned and studied. As the Chemical Processing Plant (CPP) has the largest number of fume hood installations, a meeting was held between CPP Health and Safety Personnel and the FAC.

FINDINGS OF THE COMMITTEE

1. At CPP many of the fume hood face velocities are below the required standards for the NRTS. A very expensive modification would be necessary to bring these hoods up to standard requirements.

2. Fume hoods are not used for high level, more hazardous radioactive materials. Only microcurie quantities of radioactive materials are used in the hoods.

3. The high efficiency filters with stainless steel frames now used cost from $100 to $130 each. There are fifty-eight fume hoods at CPP using these filters. The filters have a resistance of 1.0" to 1.2" water gage (CPP measurements were as high as 4.0" w g across the filter).

4. Filters are made with an efficiency of 95% on 0.5 to 1.0 micron-sized particles. Resistance across these filters is only 0.2" w g and the cost is approximately $35 to $40 each. Use of the 95% efficient filter instead of the high efficiency filter would increase hood face velocities as high as 35%. In many cases, this would bring the hood face velocities up to standard requirements and avoid costly modifications.

The recommendation made as a result of this study was: All high efficiency filters be removed from all laboratory fume hoods. In those hoods where it is felt that enough particulates could be exhausted to affect outside environs, the 95% efficient filter should be installed.

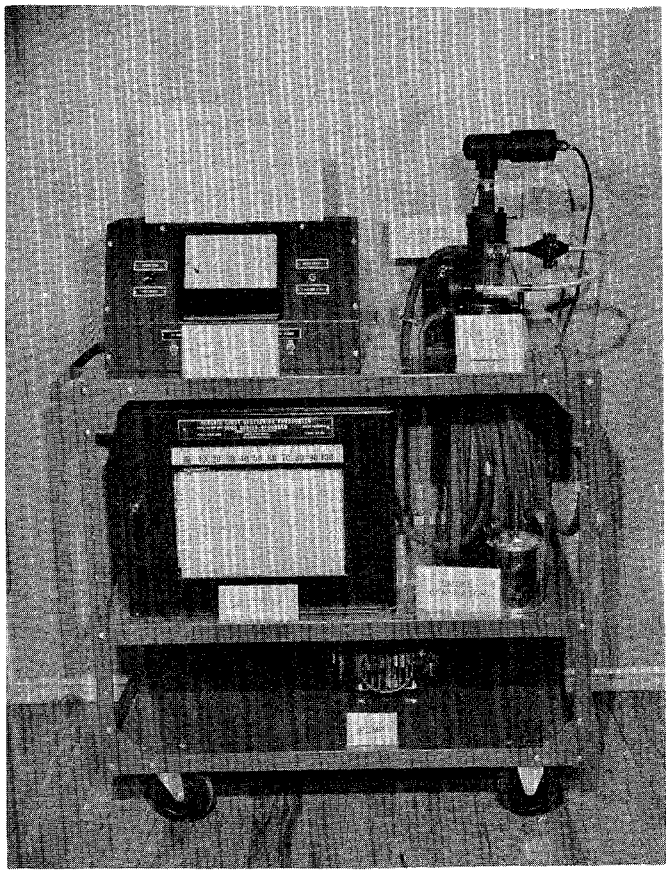

Figure 1 Smoke Photometer and DOP Generator for In-Line Testing

The justifications for a recommendation such as this are:

1. Only microcurie quantities of radioactive materials are used in fume hoods. Higher level materials are handled in glove boxes. Therefore, the 99.95% efficiency on 0.3 micron-sized particles is unnecessary.

2. Danger to the operator from toxic fumes can be serious with poor face velocities. Removal of the high efficiency filters would increase face velocities considerably.

3. Many of the hoods do not handle particulate matter. As these filters do not filter gases, vapors, or fumes, they are completely unnecessary.

4. The leakages found by the Navy on their installations indicate that many of our installations may be inadequate. Furthermore, none of the CPP fume hood filters were inspected or DOP tested at Hanford except those installed in 1961. As 55% of the filters received on site by construction contractors last year were rejected, a fairly high number of leaking filters probably existed in the past at CPP. However, no release of radioactive materials was ever attributed to fume hoods.

5. Use of a 95% efficient filter where necessary would contain almost all material, as particulates over 1.0 micron in size would essentially all be removed from the air stream. Furthermore, these filters could be tested in place to assure this efficiency.

6. A complete filter replacement with the high efficiency type costs about $6,160 at CPP. If all hoods changed to the 95% type (many should not even need these), a total cost would only be about $2,090, or a savings of $4,070 per complete filter change. Frequency of changes would remain about the same, as increased filter resistance is due to room air dust and the 95% filter would catch almost as much as the 99.95% type.

Another item found by the committee was that all high efficiency filters and pre-filters at one facility had stainless steel frames . This was required because of a filter fire occurring there a few years ago in a plywood framed filter. This plywood frame was not fire resistant.

The stainless steel frames cost considerably more than the fire resistant plywood frames and under fire conditions are not as good. While the stainless steel is necessary in some corrosive atmospheres, they were

not necessary in most of the installations at this facility. A change-over to the plywood frames gave us increase d fire protection at a much lower cost.

The FAC has already done much to improve out filter program. We still have many installations to review and will probably find additional corrections to make. The monetary savings and improved protection have proven already the advantages of such a committee.

We believe every installation should be critically examined as to its filtration needs. Filtration of 99.95% efficiency on 0.3 micron-sized particles has become too much of a by-word. These high efficiency filters are too often used indiscriminately for all radioactive particulate filtration when less efficient filters would be more than adequate. As the efficiency increases, so does the cost. It would be well to check each system and provide only necessary filtration.

The first step in correcting the indiscriminate use of these filters might be in using a new terminology for filters other than pre-filter, roughing filter, or high efficiency filter. If filters were listed by efficiency or by some other wording, design engineers, health physicists, etc., might not immediately by-pass "pre" or "roughing" filters as inadequate and always use "high efficiency" filters. After this change in terminology, it would be much easier to stress filter selection based on actual need.

DISCUSSION

SILVERMAN: I would like to take the Chairman's prerogative to comment and then I think we will have time for a question or two from the floor.

I think this type of review is very useful and I think it brought out some very salient points about designation of filters.

We have seen misapplications many times. I can remember being at ANP when someone there decided that if one absolute filter was good, two would be better, and they put in two filters. Thinking like that is usually done in ignorance of the filter theory and performance and can cost a lot of money.

I think Mr. Wehmann has indicated how much money it can be in their few cases at Idaho. I have seen many places where efficiency-absolute type filters will be put in where they were not needed and while we must admit that a 95 percent filter will pass twenty times more than the high efficiency, if it is not above MPC, why put it in?

I would only make one correction. Mr. Wehmann said that these ultra-type filters do not remove gases and vapors but, unfortunately, they do. They get saturated. The old paper type was capable of removing up to a half pound of most mineral acids. The glass paper type does such a good job on alkali and HF that the filter disappears.

With those points, I will turn the floor over to any questions that might have been raised by the paper.

GILBERT: Do you use your photometer on your hoods when you put in these? I assume you are talking about Aerosolve 95. Do you use your photometer on that installation?

WEHMANN: The present plans will be to use the smoke photometer in this situation.

SILVERMAN: The 95 is based on atmospheric dust values, and that won't always give you 95 on DOP. Aersolve units are as based on a NBS atmospheric dust efficiency which turns out to be a different particle size than DOP. It is really a blackness test. I think if you get around 70 on your photometer, that will be pretty good.

ANDERSON: I think they have a DOP test for Aerosolve filters now and the company gives a rating for them.

SILVERMAN: But it is not 95?

ANDERSON: One of the four grades is 90 to 95 percent.

SILVERMAN: On DOP?

ANDERSON: Yes.

SILVERMAN: That is not two-tenths of an inch pressure drop?

ANDERSON: No. It is up closer to an inch pressure drop.

SILVERMAN: Any other questions.

SCHWENDIMAN: I deplore the use of "low-level" without a little more quantitative measure. When you say "low-level," are you talking in terms of five millicuries? Are you talking microcuries? I think it makes a difference, so that we can all understand that "low-level" means in your terminology.

WEHMANN: I will have to excuse myself from answering that question. I was not present the day of this committee meeting but I am sure that it is in the microcurie amounts. We go into our glove boxes for anything more than this.

O'NEILL: I don't know what goes on in some of these laboratories, but are you able to project over a two-year period what levels of activity will be employed? We are in the same predicament, at Argonne, where we have people working with low levels, but feel that we must employ the high efficiency filters regardless because we don't know what they are going to be doing three months or six months from today. If we changed filters when they changed operations, we would be in a predicament.

WEHMANN: This is probably a problem with everybody here. I know at Idaho it is a problem.

SILVERMAN: I think what Mr. O'Neill was pointing to was that without casting any aspersions on anyone here that chemists and physicists are about as unpredictable as any other humans.

THE EVALUATION OF RADIOACTIVE RELEASES FROM CHEMICAL PLANTS

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ABSTRACT

Recent accidental releases of radioactive material at ORNL have resulted in the establishment of building and ventilation design criteria and the requirement for a hazards evaluation for those facilities which contain radioactive material of physiological hazard greater than that equivalent to one gram of Pu^{239}.

A quantitative method for estimating the hazards associated with the maximum credible accident in a radiochemical facility has been developed. The maximum credible accidents in such facilities are chemical or nuclear explosions which disperse radioactive aerosol and gases into ventilation streams which exhaust to the atmosphere. Approximate physical properties of these aerosols and gases have been combined with the efficiency of ventilation cleanup devices and meteorological correlations to evaluate the hazard to the environment.

The method of evaluation has been applied to ORNL radiochemical facilities, which have been modified to meet the new containment criteria, to demonstrate the acceptably low personnel exposure and ground contamination that would result from the maximum credible accident in each facility.

INTRODUCTION

The Bldg. 3019 evaporator explosion and other accidental releases of activity in the fall of 1959 precipitated a review of ORNL radiochemical facilities, the aim of which was to outline building changes that were required to confine the effects of the maximum credible accident to the involved facility. Such building changes were considered advisable to prevent jeopardizing laboratory personnel and other laboratory facilities in the event of such an accident. This review led to the establishment of building and ventilation design criteria, one requirement of which was that secondary building containment would be placed around all process cells which could otherwise leak significant activity directly to the environment in the event of an accident. It was specified that these criteria, along with the necessity of a reactor-type hazards evaluation, would apply to those facilities which contain radioactive material of physiological hazard greater than that equivalent to 1 g of Pu^{239}.

Maximum credible accidents in such radiochemical facilities are chemical or nuclear explosions which disperse radioactive aerosols or gases into ventilation streams which exhaust to the atmosphere. A realistic hazard evaluation must take into consideration the physical properties of the radioactive gas or aerosol that is formed and the efficiency of air cleanup devices for removal of these radioactive materials prior to discharge to the atmosphere.

We have attempted to make such an evaluation of ORNL radiochemical facilities using properties of aerosols and gases that are found in the literature. The studies, in general, have demonstrated the adequacy of secondary containment and present air cleanup devices but have pointed up the necessity for reliability of these devices, particularly filters, and have indicated areas in which further experimental work is required.

THE BUILDING 3019 EVAPORATOR EXPLOSION

The need for adequate primary and secondary containment was acutely demonstrated in the Bldg. 3019 evaporator explosion. A chemical explosion occurred in an evaporator complex that contained approximately 1500 g of Pu as solution, precipitate, and scale and scattered 600 mg of the Pu through a cell door, blown open by the explosion, directly to the environment. Although no personnel were injured or received an intolerable radiation dose during the accident, a portion of ORNL was significantly contaminated. In addition, the operating area of the facility was contaminated by air flow through open pipe chases and other penetrations which communicated through the cell wall.

A post-explosion examination of the facility revealed that the loss of plutonium to the environment would have been maintained within acceptable limits if (1) the door had not been blown open, (2) the penetrations through the cell wall had been minimized, and (3) the entire cell bank had been contained within a building. The release of plutonium through the existing cell and vessel ventilation filters was determined to be negligible.

The cell ventilation cleanup system, consisting of pocket-type roughing filters backed up by absolute filters, collected approximately 1.5 g of Pu and there was no measurable contamination on the exhaust side of the absolute filters. Examination of the roughing and absolute filters indicated that the roughing filters contained 98.8% of the plutonium and that the particles collected by the filters had a mass mean particle size of 0.67μ with a standard deviation of 2.3.

CONTAINMENT CRITERIA FOR A PROJECTED RADIOCHEMICAL FACILITY

A schematic diagram of a radiochemical facility which meets the minimum recommended design criteria is shown in Fig. 1. The diagram depicts a typical vessel in a process cell which is completely surrounded by a building. The cell, which constitutes primary containment, is capable of withstanding the blast effects of the maximum credible explosion without rupture and permits only a minimum leakage of radioactive material to the secondary containment shell, the building structure. Other criteria for the process vessels, cells, and buildings are as follows:

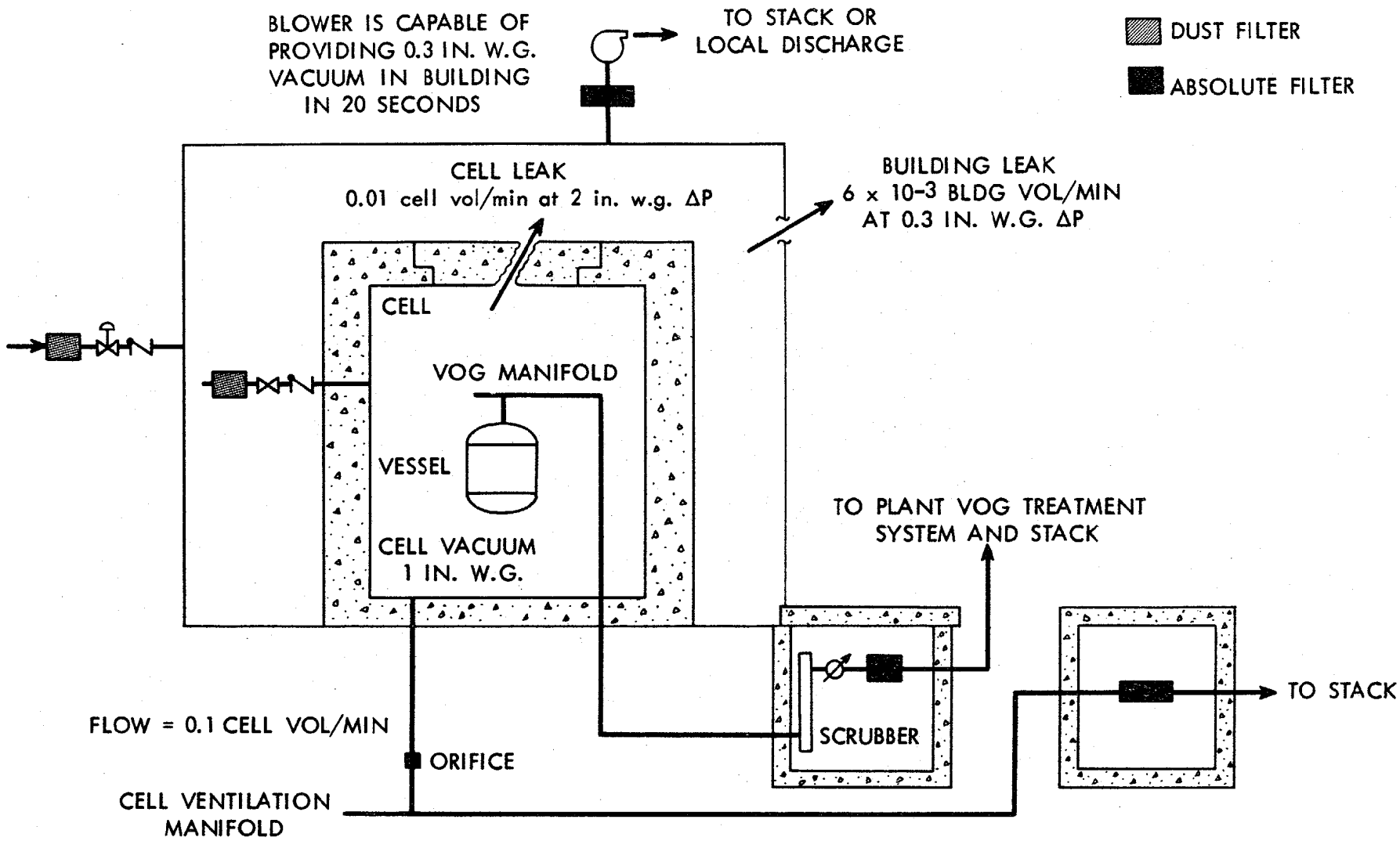
BLOWER IS CAPABLE OF PROVIDING 0.3 IN. W.G. VACUUM IN BUILDING IN 20 SECONDS
TO STACK OR LOCAL DISCHARGE
DUST FILTER
ABSOLUTE FILTER
CELL LEAK
0.01 cell vol/min at 2 in. w.g. ΔP
BUILDING LEAK
6 x 10-3 BLDG VOL/MIN
AT 0.3 IN. W.G. ΔP
CELL
VOG MANIFOLD
VESSEL
CELL VACUUM
1 IN. W.G.
TO PLANT VOG TREATMENT SYSTEM AND STACK
SCRUBBER
TO STACK
FLOW = 0.1 CELL VOL/MIN
ORIFICE
CELL VENTILATION MANIFOLD

Figure 1

Filters are to be located such that they will be protected from the maximum credible explosion.

Process vessels are maintained at a vacuum of at least 2 in. w.g. during normal operation by a VOG system which passes through a local scrubber and filter system as well as plant treatment system before being exhausted at a stack.

A cell is maintained at a vacuum of at least 1 in. w.g. during normal operation. The cell ventilation exhaust capacity is at least equivalent to 1/10 of a cell volume per minute. The air intake to the cell is through a roughing filter and check valve. The cell exhaust passes to a cell ventilation manifold, roughing and absolute filters, and from thence to the stack. The cell is sealed such that the leak rate is less than or equal to 1/100 of a cell volume per minute at 2 in. w.g. differential pressure.

The building is maintained at a few hundredths of an inch w.g. vacuum during normal operation. The intake is through duct filters and check valves. The exhaust is through roughing and absolute filters located at the roof of the building or at the stack. The cell ventilation blower must have sufficient capacity to evacuate the building to 0.3 in. w.g. vacuum in 20 seconds by closing the intake. The building is sealed such that a leak rate of no more than 6×10^{-3} building volumes per minute will occur at a differential pressure of 0.3 in. w.g. This criterion is included to ensure that the building vacuum will be capable of balancing a vacuum of 0.3 in w.g. that could be created on the lee side of a building by a 30-mile-per-hour wind. At ORNL it is pertinent to assume that winds of speed greater than 30 miles per hour are sufficiently rare as to be incredible.

TYPES OF DISPERSIVE ACCIDENTS

The most serious accidents that may credibly occur in large radiochemical facilities are chemical and nuclear explosions which rupture vessels that are filled with radioactive process solutions or solids. It is our current belief that a radiochemical facility can be designed in such a manner that the maximum credible explosion will correspond in gas production and blast effects to that of 3 lbs of TNT. Three pounds of TNT liberates approximately 5700 Btu of energy, generates approximately 100 cu ft of hot gases, and creates a shock wave which has a pressure of approximately 800 lbs per sq ft and an energy of approximately 230 ft-lbs per sq ft at a distance of 15 ft. Thick concrete cells of the type used in ORNL radiochemical facilities can withstand such explosive effects without rupture.

Examples of the types of explosions that may credibly occur in a radiochemical facility of special design and simulate the gas production and/or the blast effects of the reference TNT detonation are the detonation of 10 cu ft of a H_2-air misture, the explosion of several pounds of a nitrated organic material, and a single nuclear burst of the order of 10^{18} fissions. Our studies indicate that the initial and maximum nuclear burst in vessels of the size used at ORNL will be of the order of 10^{18} fissions. A maximum credible accident will occur if the vessel is ruptured during this maximum burst, thus terminating the reaction; the accident would have less serious consequences if the vessel contains the excursion and the reaction recurs with 10^{19} to 10^{20} or more fissions until it is shut down by other means.

EFFECTS OF DISPERSIVE ACCIDENTS

The effects of the maximum chemical explosion are that an aerosol of the radioactive material would be formed in the cell air and a small fraction would reach the environment through the vessel off-gas system, cell ventilation system, and through successive leaks from the cell and from the building. The maximum nuclear burst would disperse new gaseous fission products and an aerosol composed of new nonvolatile fission products and the original radioactive material. Another effect of the maximum nuclear burst is that operating personnel would receive prompt neutron and gamma radiation through the shield. The maximum integrated dose through a 5-ft-thick concrete wall before personnel evacuate the facility would be less than 1 rem, however.

Gaseous Fission Products

The gaseous fission products which could be released in a nuclear excursion are the isotopes of xenon, krypton, bromine, and iodine. It is usually appropriate to assume that 99% of the bromine and iodine are removed in a vessel off-gas system consisting of scrubbers and absolute filters. It has been found that the isotopes with half-lives of the order of 1-10 minutes are controlling in downwind dose calculations. The maximum permissible concentrations of these isotopes are rather large, since they constitute only external radiation hazards; they make up for the higher permissible concentrations, however, because of their greater activity.

Radioactive Aerosols

The aerosol that would be dispersed in cell air by the maximum credible accident would consist of a dispersion of a radioactive solution, solid particles, or smoke. The solution-type aerosol will be emphasized, since more information is available on this type of aerosol and since most of the ORNL facilites are of the wet chemical type. Smokes and dusts may be evaluated using an analogous procedure, provided their properties are known or are assumed.

The physical properties of aerosols are such as to restrict very effectively the escape of radioactive particles to the environment. This is seen commonly in practice, since through the use of appropriate de-entrainment mechanisms the condensate from the evaporation of a radioactive solution may be made to contain only 10^{-4} to 10^{-6} of the activity of the solution. Gravitational settling is often sufficient to restrict an aerosol concentration; we have been able to show this through an approximate correlation of the solution concentration in air or vapor arising from cooling towers, evaporators, and air-sparged vessels. This correlation is shown in Fig. 2.

In order to evaluate the release of aerosols from a cell, we must be able to ascribe removal efficiencies to filters and to cracks in cell walls. For superficial velocities less than approximately 0.15 ft per second it has been found that an aerosol formed by vigorous mixing of a solution with air is metastable and has a concentration in the order of 10 milligrams per cubic meter. This metastable concentration is approximately equivalent to fog, which has a concentration of approximately 10 mg/M^3

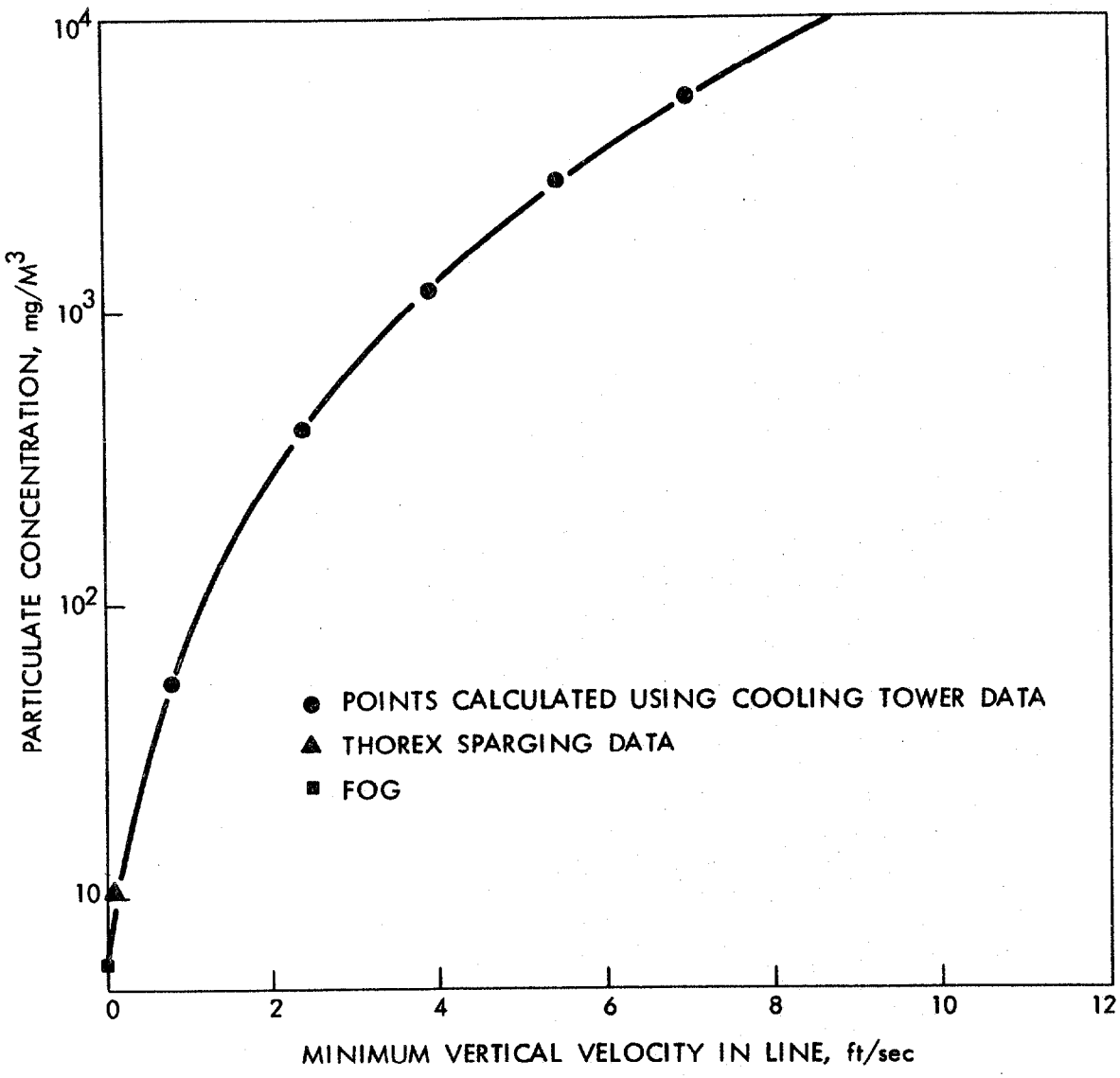
PARTICULATE CONCENTRATION, mg/M3
10^4
10^3
10^2
10
0
2
4
6
8
10
12
MINIMUM VERTICAL VELOCITY IN LINE, ft/sec
POINTS CALCULATED USING COOLING TOWER DATA
THOREX SPARGING DATA
FOG

Figure 2

and a particle size of approximately 10 microns. For orientational purposes a 1-in.-per-hr rain with mass mean particle size of 3000 microns has a concentration of 1000 mg/M^3. At ORNL the particle size distribution of the metastable aerosol in a ventilation stream downstream from the source has consistently been found to have the particle size distribution shown in Fig. 3. Another piece of relevant information reported by Garner in Transactions of the Institution of Chemical Engineers is that the weight distribution of particles smaller than 10 to 20 microns will be fairly constant, even if there is gross entrainment of larger droplets. The knowledge that this distribution is fairly constant and constitutes approximately 10 mg/M^3 may be used to estimate the approximate concentration of particles smaller than a given size, even in an air stream which is very concentrated with liquid droplets. Practically, it is possible to assign efficiencies to an absolute filter and calculate the effluent concentration.

The following efficiencies were conservatively assigned to an absolute filter: 100% for particles greater than 5 microns, 99.95% for particles between 5 and 0.3 microns, 95% for particles between 0.3 and 0.1 microns, and 87% for particles less than 0.1 micron. The filter efficiency for particles smaller than 0.1 micron is based on data obtained at Harvard. Applying these efficiencies to the particle size distribution in Fig. 3, the effluent concentration of liquid aerosol from absolute filters is calculated to be 0.14 mg/M^3. Calculations indicate that it is appropriate to assume that the liquid particles in the aerosol have essentially the original solution composition. In many instances it is also appropriate to assume 0.14 mg/M^3 as the filter effluent concentration of heavy element dust. This would indicate a conservatively high penetration of dust even if a large fraction is smaller than 0.1 micron, since it has been observed that heavy element dust exists in relatively stable air at concentrations only in the order of 0.1 to 1 milligram per cubic meter. It must be assumed that filters are only 87% efficient in removing smoke, since smoke particles are predominantly in the range 0.05-0.1 micron.

In evaluating the concentration of aerosols in air which leaks from a cell, it is considered that the design leak rate of a typical cell is equivalent to a flow of 100 cfm through a 5-in. orifice. Cell cracks will not simulate a single orifice but will consist of many small tortuous paths through 5 ft of concrete. The evaporator de-entrainment studies by Walsh and Schlea at SRP indicate that a single right angle impingement of characteristics that we think are indicative of cell cracks will conservatively reduce any liquid aerosol concentration to 10 mg/M^3. Fine heavy element dust would be reduced to the order of 1 milligram per cubic meter and the concentration of smoke in leaked air would probably be no more than approximately 100 milligrams per cubic meter.

METHODS OF EVALUATION

The downwind radiation dose that would be received from the release of radioactive material from a stack or elevated source during unchanging weather conditions may be expressed as the product of the curies released, atmospheric dilution factor, and appropriate conversion factors divided by the mpc_a. This relation is shown in Fig. 4. The mpc_a of a radionuclide may be considered as that concentration of the radionuclide in air which will cause 100 mr of radiation dose in 40 hr of exposure. In the case of radionuclides which are predominantly internal radiation hazards, the builk of the dose does

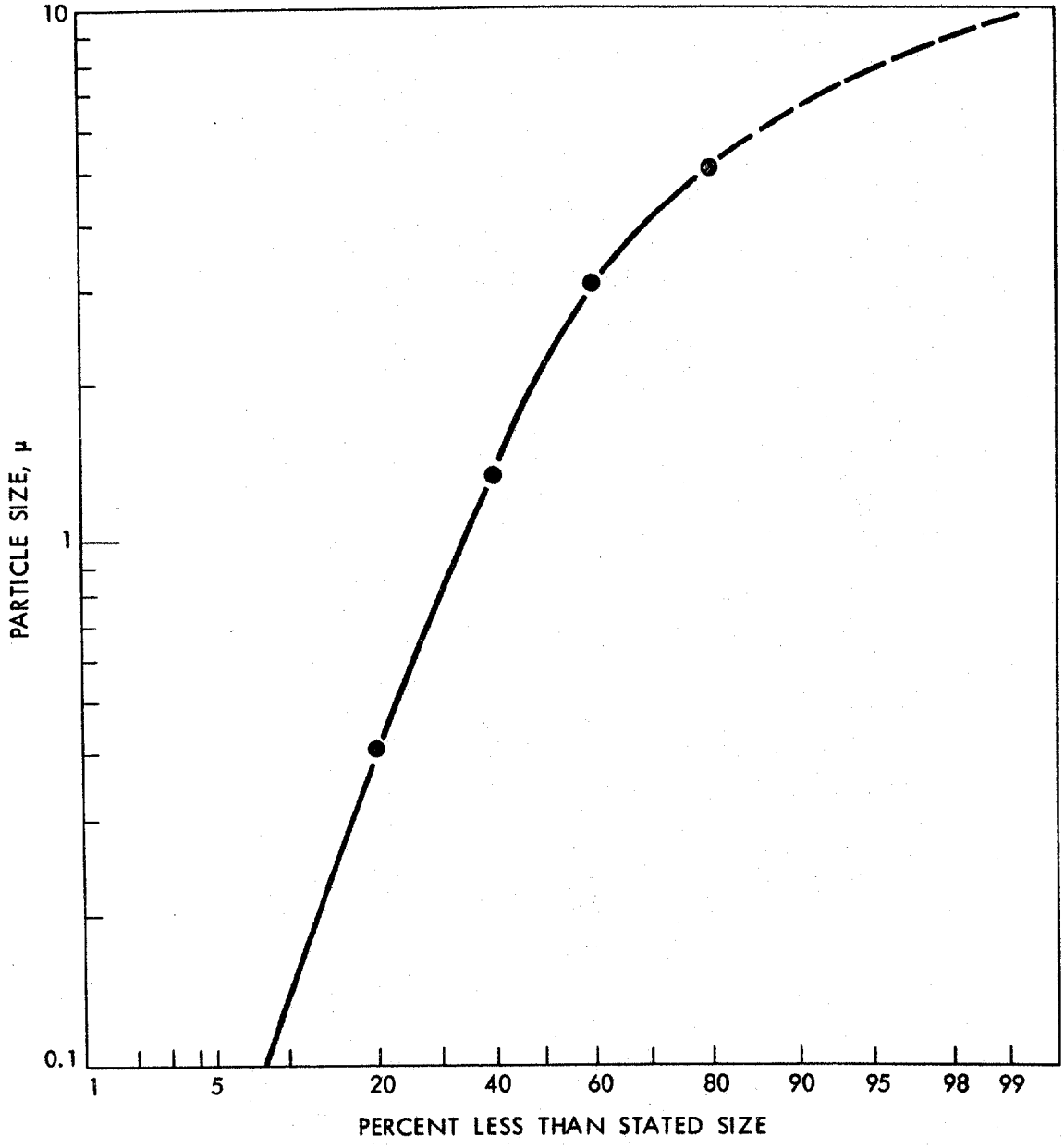
10
1
0.1
PARTICLE SIZE, μ
1
5
20
40
60
80
90
95
98
99
PERCENT LESS THAN STATED SIZE

Figure 3

MAXIMUM AVERAGE GROUND DOSE RESULTING FROM RELEASE OF RADIOACTIVE MATERIAL FROM A STACK

$$\text{DOSE, rem} = D = \frac{Qk\,(0.1)\,(1.44)}{(mpc)\,(1.44 \times 10^5)} = \frac{10^{-6}\,Qk}{(mpc)}$$

Q = QUANTITY OF MATERIAL RELEASED, CURIES

(mpc) = CONCENTRATION, CURIES/M^3, OF RADIONUCLIDE IN AIR THAT CAUSES 0.1 rem OF RADIATION DOSE FOR 40 HOURS OF EXPOSURE (SEE NBS-69)

Figure 4

not occur during the exposure period but is accumulated over a lifetime, due to the presence of the radionuclide in the body. In the downwind exposure calculation we chose to use the so-called maximum average atmospheric dilution factor (Fig. 5), which is a measure of the maximum downwind ground concentration averaged over a period of the order of 1/2 hr and is an approximate measure of the maximum downwind ground concentration averaged over a several-minute period. We chose to evaluate the constant at a conservatively low wind speed of approximately 3 miles per hour, since this is the average ORNL wind speed and since it constitutes approximately the worst case. The plume rise of a stack causes the effective atmospheric dilution to be greater at significantly lower wind speeds, and of course at very high wind speeds the dilution is significantly greater because of the extreme turbulence. We applied this concept to the calculation of the downwind internal and external dose arising from the gaseous fission products and from the aerosol; it implicitly assumes that the aerosol which escapes through an absolute filter is of such a small size that it behaves as a gas and is inhaled and exhaled as a gas. We think it is a fairly good approximation, since the aerosol particles which escape through an absolute filter are generally less than 0.1 micron in size and have negligible settling velocity.

The downwind dose resulting from the release of gaseous fission products or aerosol through the vessel off-gas system is calculated using the relations given in Figs. 5 and 6. In calculating the effects of the gaseous fission products, it is assumed that a sustained or single burst of 10^{18} fissions occurs in the vessel and that the gaseous fission products continuously leave the vessel and are entrained as they are formed. For each gaseous radionuclide the maximum downwind dose is calculated taking into consideration decay of the radionuclide in transit to the ground and the decontamination factor for the radionuclide in the vessel off-gas treatment system. In general, it may be assumed that the decontamination factor for xenon and krypton gases is 1 and that the iodine and bromine isotopes are decontaminated by a factor of 10-100 in the caustic scrubber. The aerosol release is calculated assuming that aerosol is continuously generated in the vessel for a 1-hr period following the accident and is continuously entrained in the air which is normally flowing through the vessel off-gas manifold. It is assumed that the filter effluent contains a concentration of 0.14 milligrams per cubic meter of air which has the original solution composition of radioactive material.

The equations for evaluation of the cell ventilation system release are given in Fig. 7. It is assumed that a burst of 10^{18} fissions occurs which ruptures the process vessel and scatters its contents throughout the cell, terminating the reaction. It also assumes that the gaseous fission products are evenly distributed in the cell and remain mixed. The downwind dose from individual gaseous fission products is calculated taking into consideration decay in the cell and in transit to the ground and decontamination of individual gaseous radionuclides in the treatment system. The downwind aerosol dose is calculated assuming that aerosol is entrained in a volume of air equivalent to one cell volume which passes through the exhaust at the bottom of the cell ventilation manifold. If one wishes to take into account additional generation of aerosol which might occur in the cell ventilation manifold, one would multiply the aerosol downwind dose by the ratio of the air flow rate at the filter to the cell purge rate.

MAXIMUM AVERAGE ATMOSPHERIC DILUTION FACTOR AND DECAY TIME OF RADIONUCLIDES EN ROUTE TO THE GROUND

k = MAX. AVG. DILUTION FACTOR, SEC/M^3 = $\dfrac{2}{\pi e u h^2}$

u = WIND SPEED, M/SEC

h = EFFECTIVE STACK HEIGHT, M

t_D = DECAY TIME, SEC = $t_v + \dfrac{1}{u}\left(\dfrac{h^2}{C^2}\right)^{\frac{1}{2-n}}$

t_v = DECAY TIME EN ROUTE TO STACK

C = ATMOSPHERIC DIFFUSION CONSTANT (SEE AECU-3066)

n = ATMOSPHERIC STABILITY PARAMETER (SEE AECU-3066)

Figure 5

GROUND DOSE FROM VESSEL OFF-GAS SYSTEM RELEASE

A. GASEOUS FISSION PRODUCTS (FROM 10^{18} FISSIONS)

$$D_{AV} = \sum_i \left[\frac{Q_i^o \, e^{-\lambda_i t_D}}{X_i} \right] \frac{10^{-6} \, k}{(mpc)_i}$$

X_i = DECONTAMINATION FACTOR FOR i

λ_i = DECAY CONSTANT FOR i, SEC^{-1}

Q_i^o = CURIES OF i FORMED IN 10^{18} FISSIONS

$$= \frac{(10^{18} \, \lambda_i) \, (\text{FISSION YIELD})}{3.7 \times 10^{10}}$$

B. AEROSOL OF LONG-LIVED RADIOACTIVE SOLUTION

$$D_{BV} = \left[(A_V)(m)(0.14 \text{ mg/M}^3)(4.77 \times 10^{-4})(3.6 \times 10^3) \right] \frac{10^{-6} \, k}{(mpc)}$$

A_V = VOG FLOW RATE, cfm

m = CONC. OF ACTIVITY IN SOLUTION, CURIES/mg

Figure 6

GROUND DOSE FROM CELL VENTILATION RELEASE

A. GASEOUS FISSION PRODUCTS

$$D_{AC} = \sum_i \left[\frac{R Q_i^o \, e^{-\lambda_i t_D}}{(R + \lambda_i) X_i} \right] \frac{10^{-6} k}{(mpc)_i}$$

R = CELL AIR REMOVAL RATE CONSTANT= CELL VOLUMES/SEC

B. AEROSOL OF LONG-LIVED RADIOACTIVE SOLUTION

$$D_{BC} = \left[\frac{V_C \, (0.14) \, m}{35.3} \right] \frac{10^{-6} k}{(mpc)}$$

V_C = CELL VOLUME, cu ft

Figure 7

The effect of a release to the secondary containment shell may be calculated using the equations in Figs. 8 and 9. The volume of cell air which leaks to the secondary containment cell is calculated knowing the cell leak rate at 2 in. of water differential pressure and assuming turbulent flow during the period in which the cell is pressurized. A pseudo dose to personnel in the secondary containment shell may be calculated by assuming that the leaked cell air is uniformly distributed in the volume of the secondary cell and personnel are exposed to this air for 2 minutes before evacuation. The concentration of aerosol in the leaked air is calculated considering impingement which occurs in the tortuous path through the cell wall and the gaseous fission product concentration is that concentration obtained by dispersing all of the gaseous fission products in the volume of the cell.

The release of activity from the secondary containment shell is by two mechanisms; the normal ventilation flow through the absolute filter and the building leakage which occurs if there is a significent wind to create a lee vacuum on the building. The downwind ground concentration for individual gaseous fission products and the aerosol is calculated using the equations in Fig. 10. The downwind dose is the sum of the dose which occurs from the leak from the building during the 20-second period which is required to evacuate the building to 0.3 in. w.g. vacuum and the release through the building ventilation system. For the gaseous fission products, appropriate corrections are made for decay inside the building and in transit through the building ventilation system.

In addition to the dose calculations, we calculated the downwind ground contamination that will occur from fallout of the radioactive particulate matter using equations given in AECU-3066 and the nomograms in ORO-176. The particle size of solution particles released from the secondary containment shell leak was assumed to be approximately 10 microns and the particle size released through the filter ventilation system was assumed commensurate with the filter efficiencies. The results were expressed as the distance downwind from the source to which the ground is contaminated to the hazard and required decontamination level. The hazardous level for beta-gamma contamination was considered to be that concentration in curies per square meter which would give a reading of 2-1/2 mr per hr above ground as determined by a GM survey meter with an open window. For alpha materials the hazardous ground concentration in curies per square meter was considered to be the arithmetic product of 250,000 times the mpc air for 40 hr of exposure.

RESULTS AND CONCLUSIONS

Using the methods that have been described, we were able to show to our satisfaction that the effects of what we considered to be the maximum credible accident in ORNL radiochemical facilities, which have been revised to meet the containment criteria, result in acceptable personnel exposure and downwind ground contamination. In our large wet-chemical facilities, such as Bldg. 3019, it was calculated that operating personnel or Laboratory personnel downwind from the facility could receive no more than a few multiples of the weekly permissible dose and that the ground downwind from the facilities would not be contaminated beyond 10% of the maximum permissible ground level.

VOLUME OF RADIOACTIVE AIR RELEASED TO BUILDING (SECONDARY CONTAINMENT SHELL)

V_L = LEAKAGE VOLUME, cu ft $= \frac{1}{W_L} \int_0^t P^{1/2}\, dt$

t = ELAPSED TIME WHILE CELL IS ABOVE ATMOSPHERIC PRESSURE

P = PRESSURE RELATIVE TO ATMOSPHERIC, in. H_2O

W_L = RESISTANCE, (in. $H_2O)^{1/2}$ / (cu ft/SEC)

FOR A FACILITY IN WHICH ORNL CONTAINMENT CRITERIA ARE MET:

$$V_L \leqq (0.1\, V_C) \left[\frac{V_{EX}}{V_C} - \frac{1}{407}\right] \left[\frac{407\, V_{EX}}{4\, V_C} - \frac{1}{4}\right]^{1/2}$$

V_C = CELL VOLUME, cu ft

V_{EX} = EXPLOSION VOLUME, cu ft

Figure 8

PSEUDO TWO-MINUTE DOSE TO PERSONNEL BEFORE EVACUATION OF THE SECONDARY CONTAINMENT SHELL

A. GASEOUS FISSION PRODUCTS

$$D_{AP} = \sum_i Q_i^o \frac{V_L}{V_C} \left(\frac{1 - e^{-120\lambda_i}}{\lambda_i} \right) \frac{(35.3)(10^{-6})}{V_B (mpc)_i}$$

V_B = SECONDARY CONTAINMENT SHELL VOLUME, cu ft

B. AEROSOL OF LONG-LIVED RADIOACTIVE SOLUTION

$$D_{BP} = \left[(10 \text{ mg/M}^3)(m)(V_L) \right] \frac{(120)(10^{-6})}{V_B (mpc)}$$

Figure 9

GROUND DOSE FROM SECONDARY CONTAINMENT SHELL RELEASE

A. GASEOUS FISSION PRODUCTS

$$D_{AS} = \sum_i Q_i^o \frac{V_L (10^{-6})}{V_C (mpc)_i} \left[\frac{R_L k_L \left(1 - e^{-(R_L + \lambda_i)20}\right)}{R_L + \lambda_i} + \frac{R_B k_B e^{-\lambda_i t_D}}{(R_B + \lambda_i)} \right]$$

R_L = LEAK RATE CONSTANT, SEC. CONT. VOLS./SEC = $\frac{6 \times 10^{-3}}{60}$

R_B = VENTILATION RATE CONSTANT, SEC. CONT. VOLS./SEC $\cong \frac{0.1}{60}$

k_L = ATMOSPHERIC DILUTION FACTOR FOR LEAK, SEC/M^3 (u = 30 mph)

k_B = ATMOSPHERIC DILUTION FACTOR FOR BUILDING VENTILATION, SEC/M^3

B. AEROSOL OF LONG-LIVED RADIOACTIVE SOLUTION

$$D_{BS} = \frac{(10)\,(m)\,(V_L)\,(10^{-6})}{(35.3)\,(mpc)} \left[k_L R_L (20) + k_B \left(\frac{0.14}{10}\right) \right]$$

Figure 10

One significant conclusion has been that, even if the filter effluent concentration which we have assumed is conservative by a factor of 100, the controlling dose downwind from a facility is that due to the release through the filtered vessel and cell ventilation systems rather than from the release through leaks in the cell and building. This suggests that the use of a filter with better particulate removal efficiencies than those which we assumed could conceivably justify the location of a secondarily contained radiochemical facility in an uncontrolled, populated area.

It is our hope that these containment criteria and methods of evaluation will stimulate investigation, particularly into the properties of aerosols and efficiency of air cleanup devices. The availability of better hazards evaluation data and cleanup devices will permit more public assurance and more realistic containment and siting criteria for radiochemical plants. It will possibly also permit a more realistic assessment of the safety of industrial plants in which nonradioactive but physiologically hazardous chemicals are handled.

DISCUSSION

SILVERMAN: We all hope the valves on the ventilation cells work better than that slide projector.

NICHOLS: We intend to routinely test and maintain critical valves to assure that they have a high probability for functioning as intended in the event of an accident. In some instances we use duplicate sets of valves, as well as radiation monitors and blowers, to provide the required probability of operation. In one instance we chose to maintain a building under emergency conditions (i.e., 0.3 in. w.g. vacuum) during all operations in order to place less reliance on rapidly acting emergency devices.

The need for a high probability of containment system operation may be evaluated, using the methods that were discussed, by superimposing a valve, instrument, or blower failure on the maximum credible accident.

YOUNG: In regard to the estimate of leakage which you reported in terms of 0.01 of a volume per minute; is this based upon calculation or measurement?

NICHOLS: From the building?

YOUNG: From the cell. You had two leakage rates.

NICHOLS: We chose to specify that process cells would be sealed such that there would be less than 0.01 cell volume per minute leakage rate at 2 in. w.g. differential pressure, since this constitutes adequate containment and can be realized in practice. Using gaskets, Fiberglas, caulking compounds, and could be handled satisfactorily and built into the system, and it has been demonstrated.

SILVERMAN: One of the problems in all these calculations are the so-called -- I am not a great fan of the maximum credible accident concept, since those accidents considered incredible today may be very credible tomorrow perhaps due to the use of such things as a different solvent than the one used at the time of the hazard evaluation. How do you correct for this in your operations now?

NICHOLS: We specify that for every new chemical flowsheet and equipment arrangement that is installed within a process cell there must be a complete hazards evaluation, which considers all of the credible accidents that are

associated with that flowsheet and arrangement, and approval by an ORNL safety committee and Laboratory management. Subsequent minor modifications to a flowsheet or equipment arrangement are also evaluated for safety, written up, and presented to a division level Radiation Safety Officer who may choose to approve the revision or refer the revision to the ORNL safety committee.

In general, we feel that a radiochemical plant of a given size has a characteristic maximum credible accident that is determined by a limited volume organic-air, nitrated organic, hydrogen-air, or nuclear explosion. Within the scope of existing technology, minor modifications to a process usually do not materially alter the magnitude of the maximum dispersive accident.

SILVERMAN: The Building 3019 accident was certainly very credible.

NICHOLS: Yes, it had lower energy release and blast effects and dispersed less activity than would our concept of the maximum credible accident in the facility. The effects of the 3019 accident would have been maintained well within acceptable limits if the building at the time of the accident had met our present containment criteria.

REMOVAL OF IODINE FROM GAS STREAMS

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ABSTRACT

Contamination of the atmosphere by radioactive isotopes of iodine constitutes a serious biological hazard and, for this reason, provisions should be made at reactors and other nuclear installations to prevent such releases in the event of an accident. The efficiency of activated charcoal, silver, and copper surfaces for adsorption of iodine vapor has been studied under various conditions and in several gas systems. Iodine vapor is removed from air at 25°C by activated charcoal with efficiencies as high as 99.999+%. Copper and silver-plated copper ribbon exhibit efficiencies up to 98-99%. In steam-air systems over the temperature range of 75 to 118°C activated charcoal has an average iodine removal efficiency of 99.9% for moderate gas velocities. In helium at a temperature of 325°C, the various charcoals exhibit iodine retention efficiencies varying from 16 to 99.99+%. A special impregnated charcoal, Whetlerite ASC, exhibits efficiencies of 99.99+% for iodine vapor up to temperatures of 550°C in helium.

Problems associated with the removal, containment, and disposal of gaseous fission products present in dynamic gas systems are becoming more demanding as the nuclear industry expands. A considerable portion of the radioactive by-products of uranium fission appears as iodine, krypton, and xenon, which by virtue of their volatility are difficult to retain in many types of fuel elements. This behavior has a marked influence on the design, location, and operation of nuclear reactors and fuel reprocessing plants and in the design and operation of in-pile experiments. Either by accident or by design, large amounts of gaseous isotopes can be present in circulating gas, off-gas, or ventilation systems of nuclear installations. Discharge of these isotopes, especially those of iodine, into the atmosphere adjacent to the installation can produce serious biological hazards if provisions are not made to reduce the concentration of the radioactive isotopes to safe or acceptable concentrations before release of the carrier gas to the atmosphere. To date this study has been directed toward specific solutions of problems concerning the removal and containment of iodine from gas streams according to the needs of several reactor development programs.

REMOVAL OF IODINE FROM AIR STREAMS AT 25°C

This initial study was conducted to determine a feasible method for iodine vapor adsorption to be used in the emergency ventilation system of a building housing a 5-Mw

swimming pool reactor (1). In the event of a reactor accident involving fuel melting, building air will be diverted through the emergency exhaust system which will contain provisions for the removal and containment of iodine vapor. After survey of the methods previously utilized for iodine removal, it was concluded that a system employing a solid adsorbent would be more easily adapted to the requirements of the emergency ventilation system.

A laboratory study of the more promising materials was conducted utilizing the system diagrammed in Fig. 1. Wherever possible, the system was constructed of glass to minimize adsorption of the iodine vapor by the walls. A typical experiment involved the following operations. Elemental iodine crystals (I-127 containing radioactive I-131) were contained in the U-tube, and a portion of the air supply was routed through the U-tube to sweep iodine vapor into the main air stream. The time required for introduction of all the iodine vapor into the adsorber column was approximately 15 min, and the average iodine concentration during this time was 0.18 mg of I-127 and 54 μc of I-131 per cubic foot of air. Air flow through the system was then continued for 24 hr. Iodine vapor escaping from the adsorber column was collected downstream by the combination of a plug of carbon wool fibers, a CWS-6 absolute filter, and an electrostatic precipitator. After completion of the 24-hr. test, the apparatus was disassembled and the distribution of iodine radioactivity in the adsorber column was determined by scanning with a sodium iodide scintillation crystal viewing through a small slit in a lead shield. The over-all efficiency of the system was then determined by radiochemical assay of the entire system from Point A to Point B in Fig. 1. By comparing the amount of iodine residing in the adsorber column with the total amount found, an adsorption efficiency was determined. It was realized that the accuracy of this method for determing iodine adsorption efficiency depends upon the premise that all iodine passing through the adsorber was collected and that none was allowed to escape. In calculating the efficiency, all downstream samples (i. e., CWS-6 filter papers, aluminum liner from electrostatic precipitator, etc.) in which radioiodine could not be detected were assumed to contain an amount of I-131 equal to the limit of detection of the radiochemical method of assay. Therefore, the iodine adsorption efficiency calculated is less than the true efficiency and represents a lower limit.

The major portion of this study was centered on charcoal since this material has been shown to have a very high efficiency for iodine vapor adsorption under various conditions.

For proper design of an iodine adsorber the effect of air velocity and adsorbent particle size on the adsorption process must be known. The effect of superficial air velocity (volumetric air flow divided by cross-sectional area of adsorbent container) on iodine adsorption was studied at velocities of 82, 170, and 275 fpm through adsorbers containing 6-8 mesh charcoal. Very little difference was observed. The depth of penetration into the charcoal mass and the over-all efficiency of the system were almost equal for the three air velocities. The size of the charcoal particles does not affect the adsorption efficiency. Study of 2-4, 4-6, and 6-8 mesh charcoal (Columbia SXC) at a superficial air velocity of 170 fpm yielded efficiencies of 99.63, 99.89, and 99.99+%, respectively. A typical iodine distribution for an adsorber 8 in. deep, containing 6-8 mesh, Columbia SXC charcoal, and operated at a linear air velocity of 170 fpm is given in Fig. 2. For optimum performance an iodine adsorber should contain charcoal of

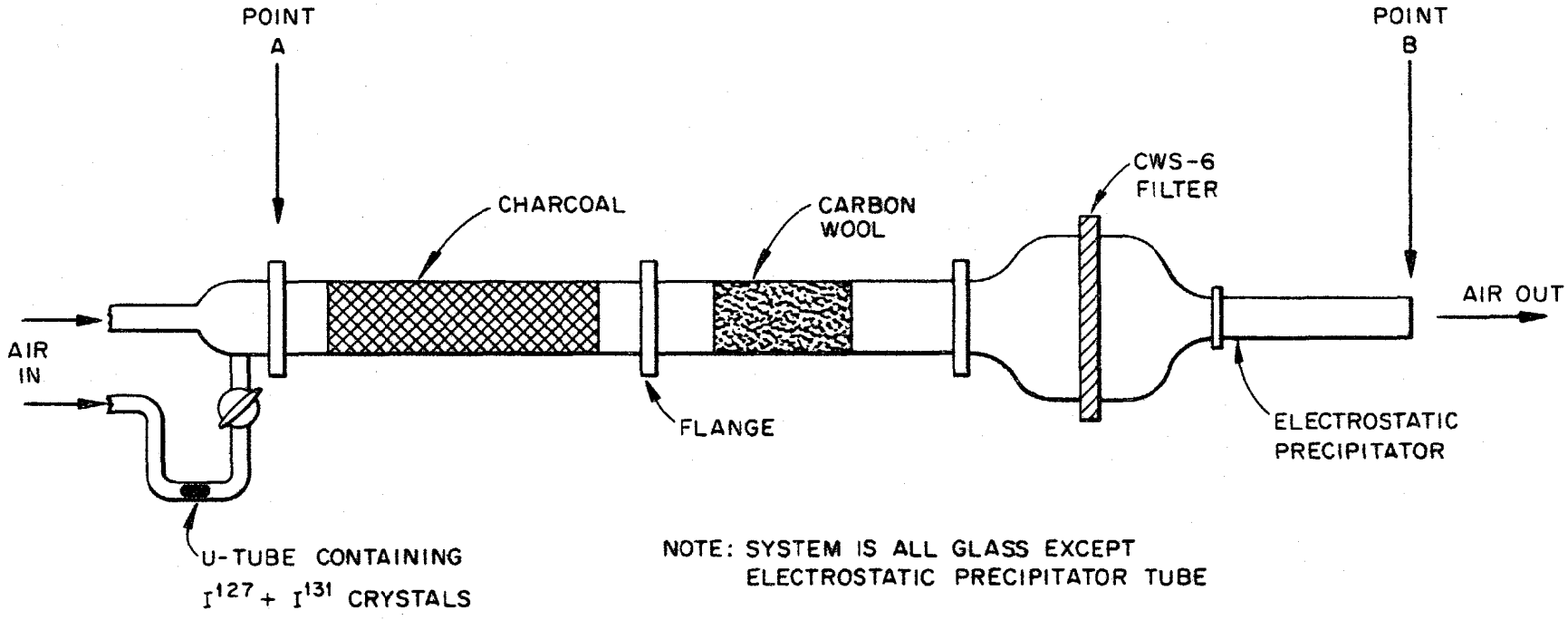
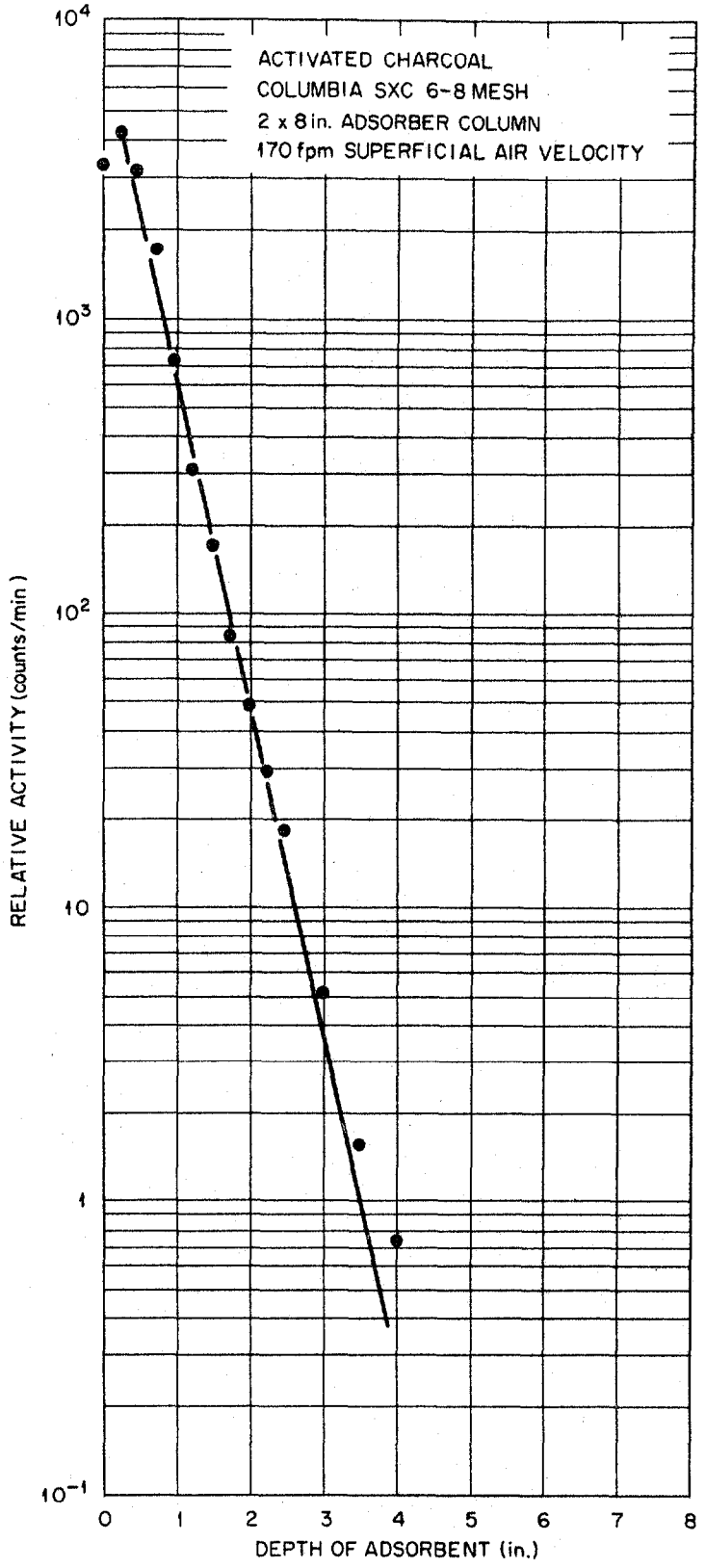


Figure 1 Experimental System.



Distribution of I^{131} Radioactivity in Charcoal Column.

Figure 2

particle size 6-8 mesh, or smaller, and even though little effect of air velocity was noted over the range studied, the air velocity should be kept low because of pressure drop and prevention of mechanical damage to the system.

Early in the study it was observed that dust particles in the air sweep might be responsible for transport of iodine through an adsorber. Iodine adsorbed on a particle of dust would not be available for reaction at the charcoal surface. In addition, fine particles of charcoal containing iodine might be carried from the adsorber by the air sweep. For these reasons the absolute filter (CWS-6) and the electrostatic precipitator were included in the experimental system in addition to the carbon wool. In one run, the CWS-6 filter was placed immediately downstream from the charcoal mass, and particularly "dusty" charcoal was used. A significant amount of dust and iodine activity was found on the filter, and a detectable trace was found on the inner surface of the electrostatic precipitator. In all later runs the dust was removed from the charcoal before iodine injection by introducing air at a flow rate greater than that to be used in the experiment, and the dust problem was greatly reduced.

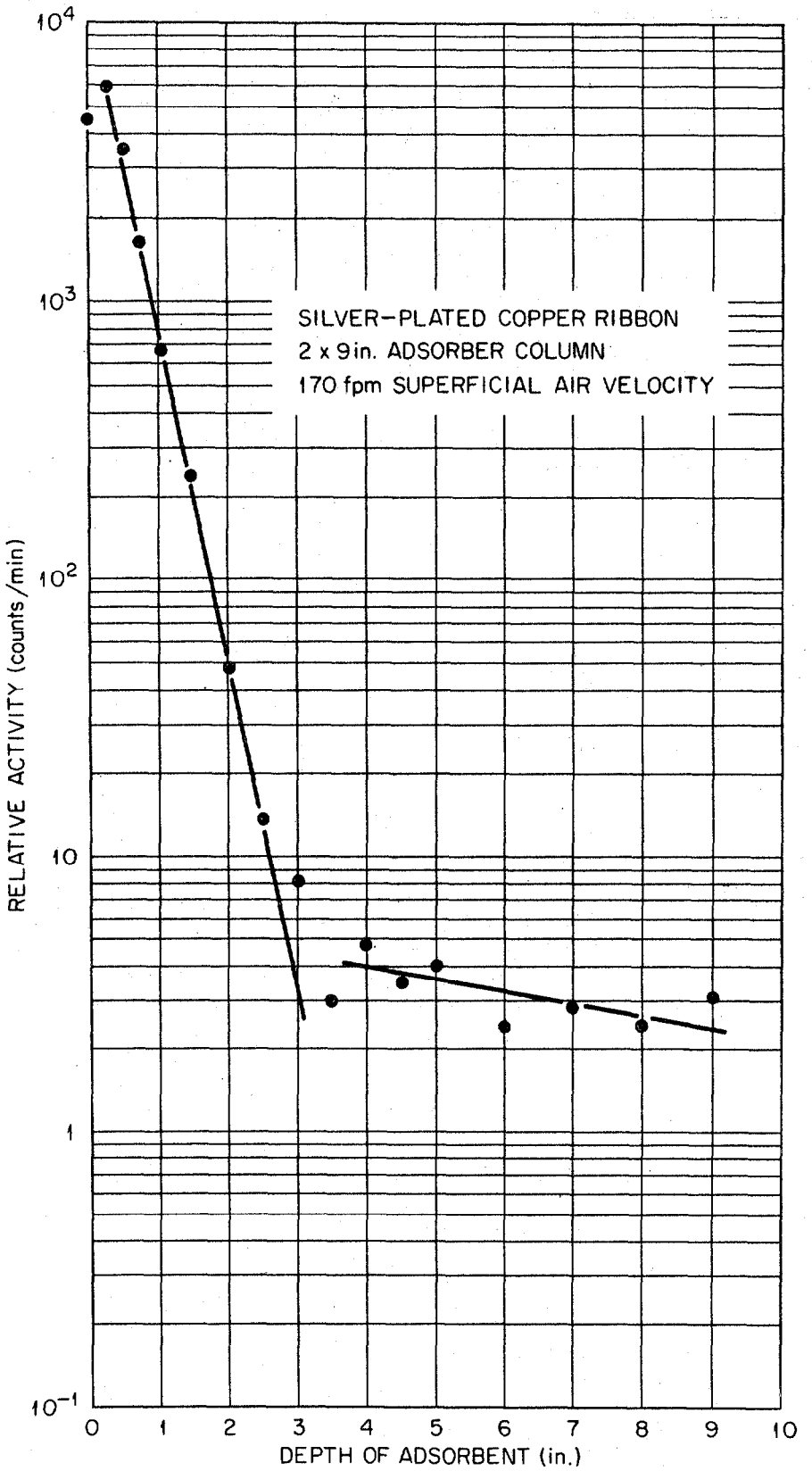
Most of the tests were operated for a 24-hr. period, and during this time no downstream transport of iodine could be detected; the major portion of the iodine was concentrated at the inlet of the adsorber. One test was operated for 70 hr after injection of the iodine, and no movement could be detected. Once adsorption takes place, the iodine is firmly held on the charcoal surface.

Charcoal would not be expected to lose iodine adsorption efficiency upon exposure to clean air; however, to check this point, one test was made on charcoal that had been exposed to the laboratory atmosphere for several weeks and then exposed to air flow for 350 hr prior to iodine vapor injection. An efficiency of 99.99+% was obtained, and no detectable effect on iodine vapor adsorption was noted.

Several tests were conducted on silver-plated copper ribbon, a material studied extensively at the Harvard University Air Cleaning Laboratory (3). The ribbon was woven into a mesh configuration and coated with silver equal to 5% by weight. The runs were conducted at a face velocity of 170 fpm using a column packed with the mesh compressed to a density of 25 lb/ft3. The iodine adsorption efficiency was found to be 92%, with an iodine distribution in the adsorber as shown in Fig. 3. A break is noted in the distribution curve at a depth of approximately 4 in., indicating that the efficiency beyond this depth is much lower than at the entrance. This effect has also been observed in work at Harvard. The other two tests were run under the same conditions as before except that an absolute filter was added to the air supply. The adsorption efficiency was increased to 98 and 99%, indicating that particulate matter in the air supply may be responsible for transport of iodine through the silver-copper ribbon column.

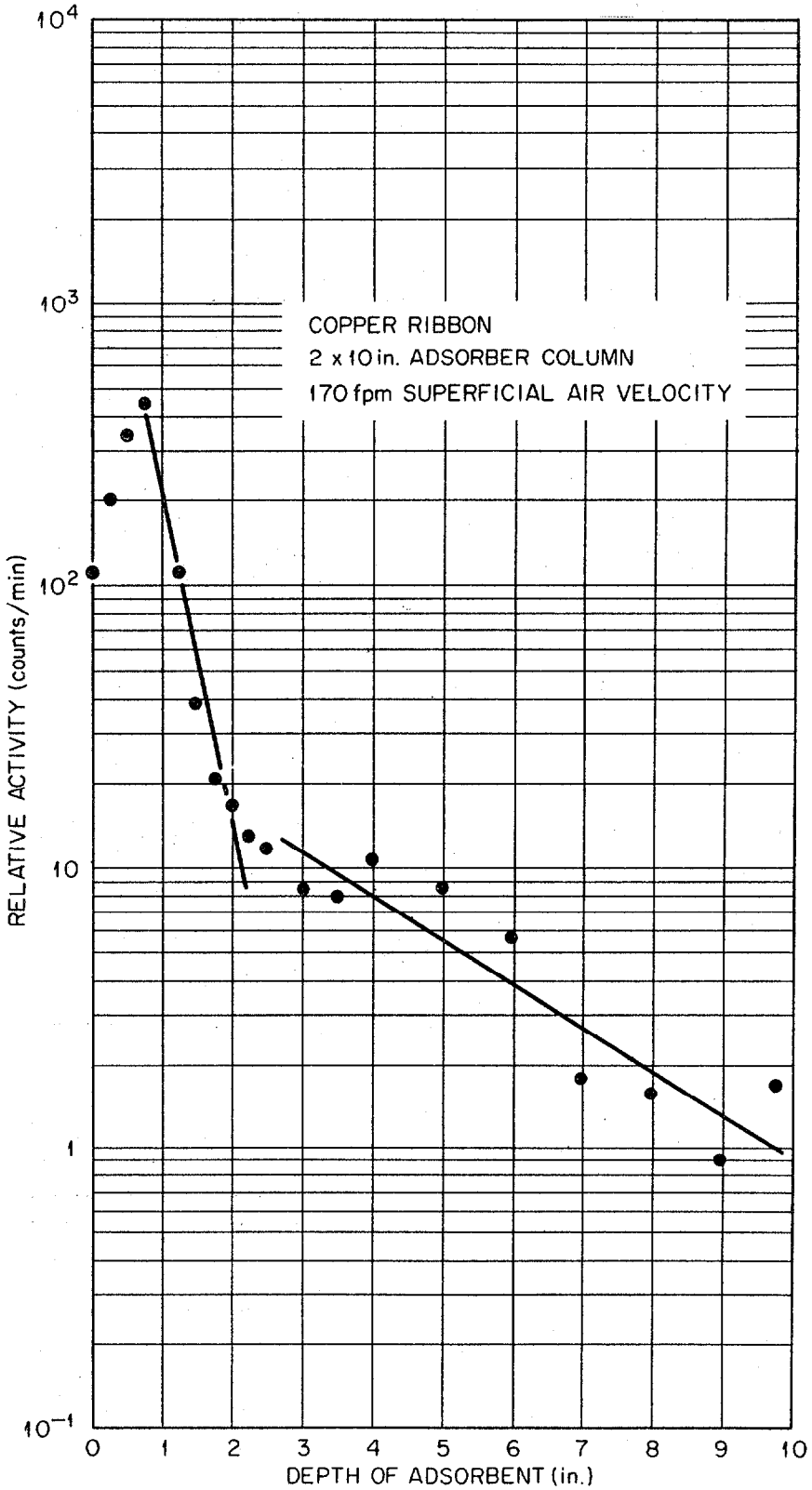
One test employing new copper ribbon was made under conditions similar to those of the silver-copper tests. An iodine efficiency of 98.5% was obtained with an iodine distribution in the test column as shown in Fig. 4.

The choice of using activated charcoal or silver-copper ribbon for iodine adsorption will depend upon the application. Each material is subject to shortcomings under various conditions.



Distribution of I^{131} Radioactivity in Silver-Plated Copper Ribbon Column.

Figure 3



Distribution of I^{131} Radioactivity in Copper Ribbon Column.

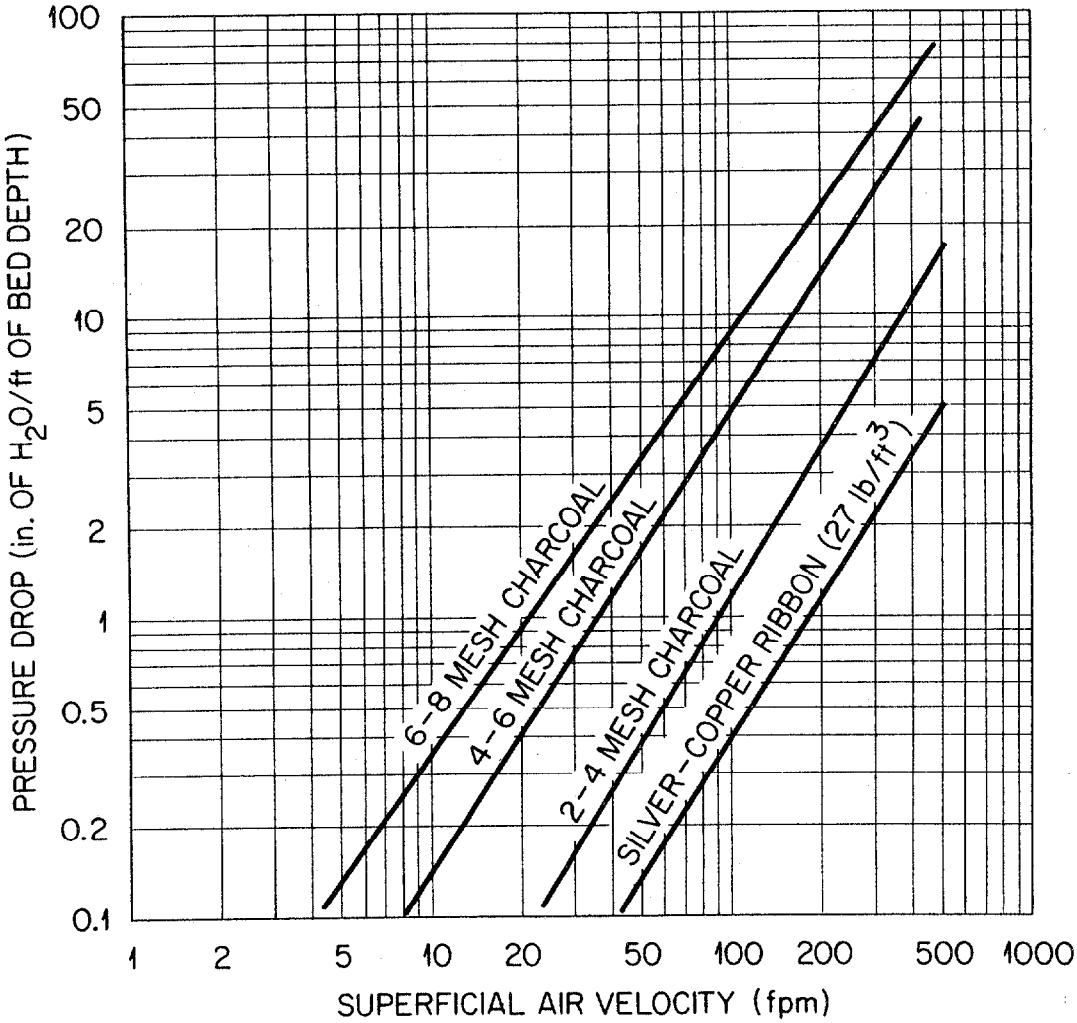
Figure 4

Activated charcoal is a more efficient adsorbent, but the pressure drop through the charcoal mass is significant in some applications. Silver-copper ribbon, while being somewhat less efficient, exhibits an almost insignificant pressure drop. Fig. 5 displays pressure drop as a function of superficial air velocity for several mesh sizes of charcoal and for silver-copper ribbon at a packing density of 27 lb/ft3.

Upon comparison of the distribution curves of iodine adsorbed by charcoal and silver-copper ribbon a distinct difference is noted. The iodine distribution line for charcoal does not "break", indicating that the adsorption efficiency is constant over the length of the adsorber. The distribution curve for silver-copper ribbon breaks and changes slope at a depth of approximately 4 in. This effect could result from a different mechanism of adsorption becoming predominat at this depth and is thought to indicate that iodine vapor is the species being adsorbed at the entrance to the adsorber while the species giving rise to the break is due to iodine adsorbed on dust particles which are penetrating the adsorber. Efficient prefiltration of the air supply reduced this effect somewhat.

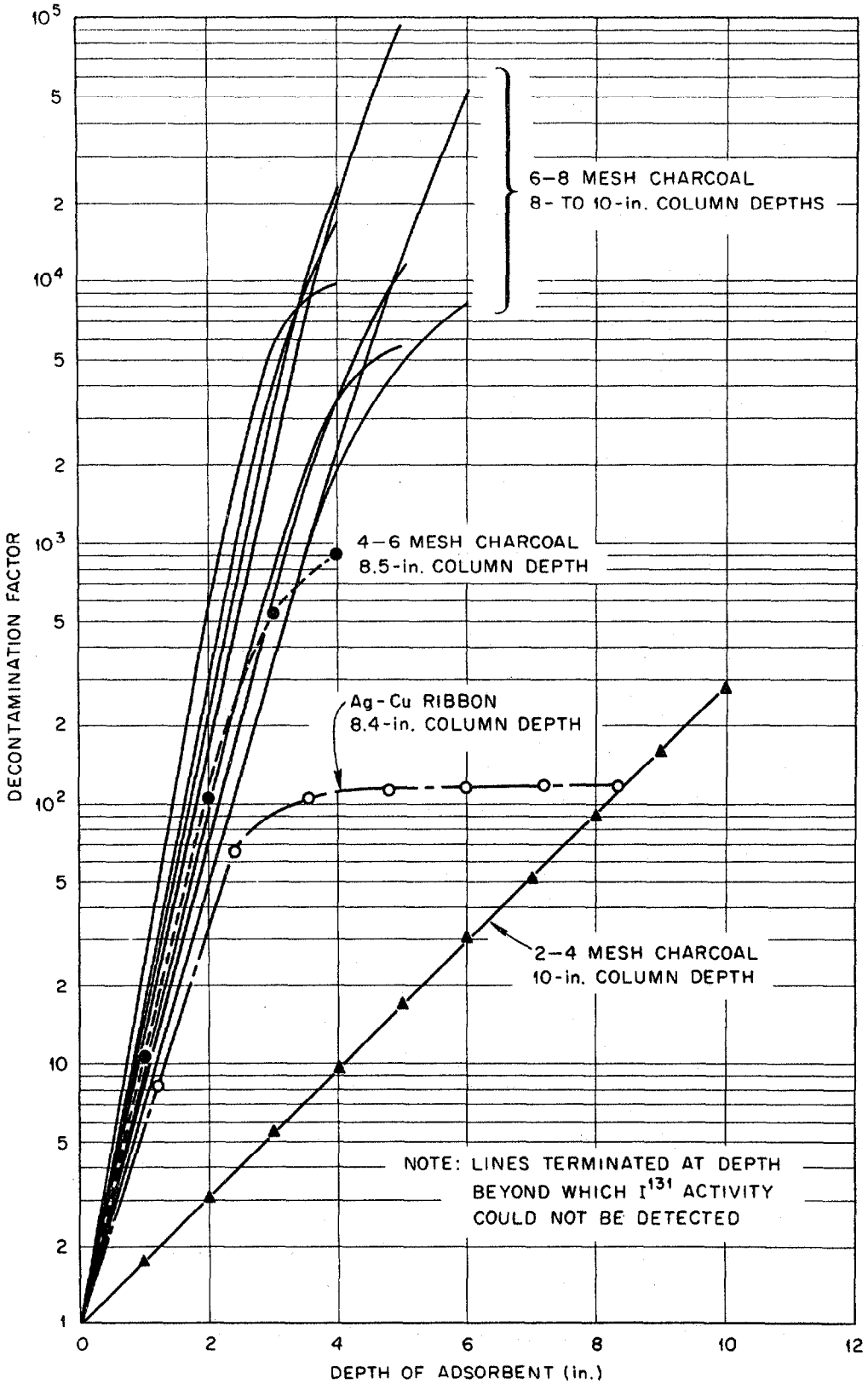
The application of charcoal for iodine adsorption may be limited somewhat by its tendency toward rapid oxidation at high temperatures in an oxidizing atmosphere. Heating of the charcoal can occur from both the beta decay of adsorbed radioiodine and the heat content of the gas containing the iodine vapor. For this reason, the use of charcoal in systems where the temperature is high ($>150^{o}C$) and oxidizing gases are present should be viewed cautiously, and some means of cooling the gas stream or charcoal applied. Silver-copper ribbon gains in efficiency when the temperature increases but reaches a maximum near $300^{o}C$, and at higher temperatures the collected iodine is released. One factor to be noted is that once exposed to high temperatures the silver-copper ribbon is no longer effective at room temperature.

It seems that the choice of using charcoal or silver-copper ribbon will depend mainly upon the decontamination efficiency desired in the air cleanup and upon the pressure drop that can be tolerated. On the basis of decontamination efficiency alone, 6-8 mesh charcoal is superior to the other adsorbents tested. In Fig. 6 the various mesh sizes of charcoal and silver-copper ribbon are compared, based upon decontamination factor as a function of depth of adsorbent. The multitude of lines for 6-8 mesh charcoal represent tests under the various conditions of superficial air velocity, moisture content, and duration of air sweep. The decontamination factor is defined as the amount of iodine found in the adsorber column divided by the amount of iodine which passed through. In an attempt to relate pressure drop, superficial air velocity, and decontamination efficiency, a plot was made of the decontamination factor as a function of pressure drop divided by superficial air velocity. This information is contained in Fig. 7. The curve for 6-8 mesh charcoal represents one interpretation for consolidating the data presented in Fig. 6 for this adsorbent. Although these curves are based on limited data, some tentative conclusions can be drawn regarding the relative merits of the silver-copper ribbon and several mesh sizes of charcoal for application at air velocities in the range 150 to 200 fpm. From the standpoint of pressure drop, silver-copper ribbon is superior to charcoal for decontamination factors up to 100. Between decontamination factors of 100 to 300, 2-4 or 4-6 mesh is more suitable; from 300 to 1000, 4-6 is the choice; above 1000, 6-8 mesh is preferable.



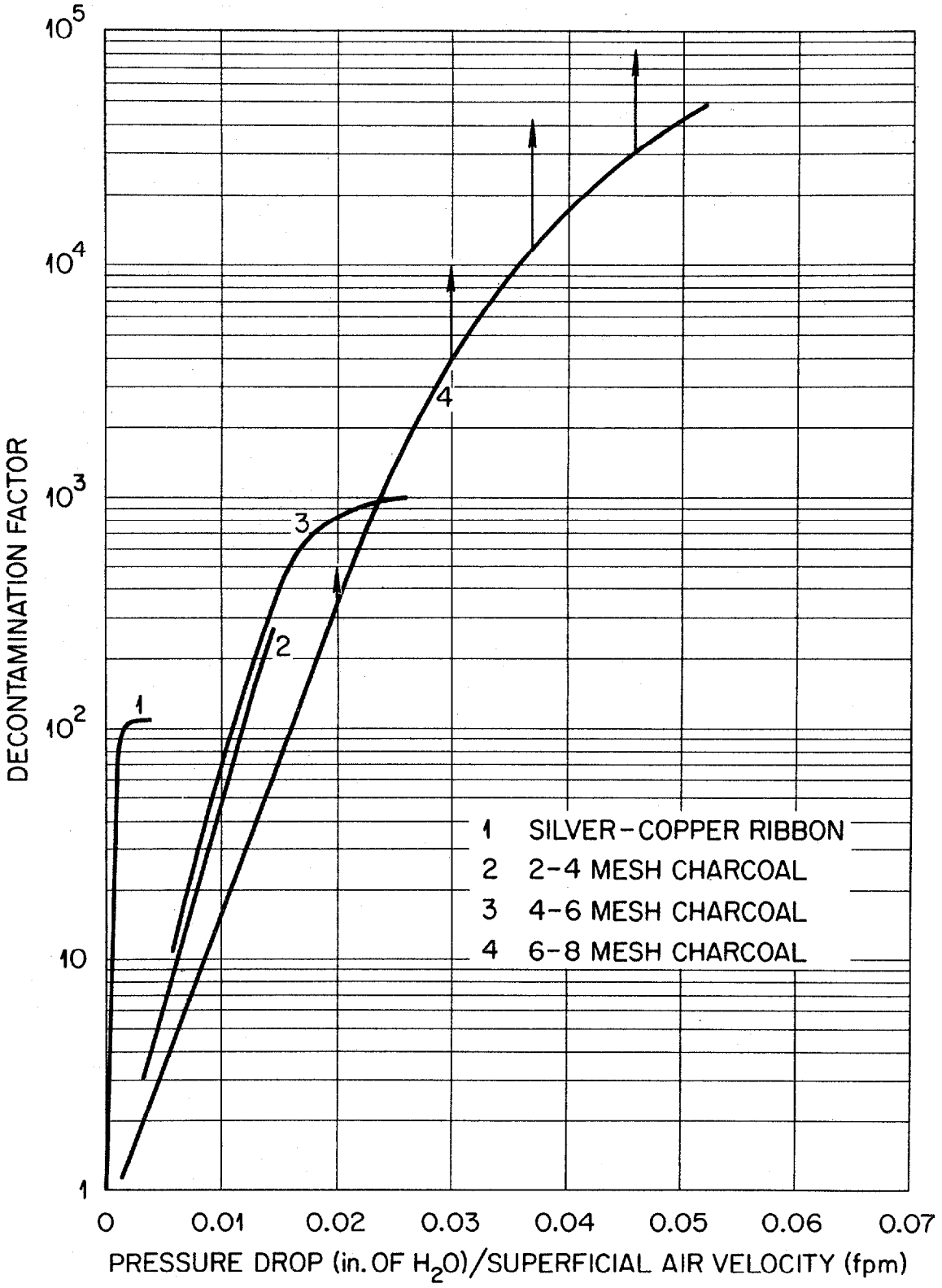
Pressure Drop vs Superficial Air Velocity.

Figure 5



Decontamination Factor as a Function of Adsorbent Depth.

Figure 6



Decontamination Factors as a Function of Pressure Drop and Superficial Air Velocity.

Figure 7

REMOVAL OF IODINE FROM AIR-STREAM MIXTURES

This program was initiated to extend the previous study to include the removal of iodine vapor from a gas stream consisting of steam and air at temperatures in the 80-100°C (2). Such conditions would exist following an accident involving any reactor depending upon water for cooling, shielding, neutron moderation, or heat transfer. Specifically, the experimental conditions were chosen to simulate the following accident involving a pressurized water reactor. The accident is postulated to occur as the result of loss of coolant with a resulting melting of the reactor core. Escape of fission products from the containment vessel could occur through leaks caused by the brief pressure surge. From a hazards standpoint, it appears necessary to include an iodine removal system within the containment vessel to reduce the leakage of iodine vapor into the atmosphere. The leakage of contaminated gases from the containment vessel would occur until the pressure differential between the atmosphere and the interior of the containment vessel becomes zero. This condition is assumed to occur approximately one hour after core meltdown.

The laboratory study was conducted utilizing the experimental system diagrammed in Fig. 8. The system was constructed from glass pipe with external heating provided by flexible heating tapes to a point just downstream of the experimental charcoal trap. Thermocouples were provided to monitor temperatures at various points. One thermocouple was located, internally, immediately downstream from the charcoal trap to monitor the temperature of the steam-air mixture. Wall temperatures of the glass pipe were maintained equal to or, in some cases, slightly greater than (1-2°C) the temperature of the steam-air mixture. Four steam condensate drains were provided to collect the condensate during a test run. Metered air and steam were mixed and supplied to the experimental apparatus through a heated metal system. Some difficulty was encountered in supplying a small measured amount of steam. This difficulty was overcome by using an insulated and internally heated gas rotameter to serve as an approximate measure of the steam injection rate during the test. At the end of the test a final calculation of the steam injection rate was made based on the temperature history and the amount of condensate collected.

The activated charcoal test adsorber was designed to simulate a small section of a commercial charcoal canister* previously tested at room temperature. One canister was destroyed and the charcoal, 8-14 mesh, and the perforated metal cylinders were used in construction of the experimental test adsorber. A bed depth of 0.75 inches was used for all tests.

A typical experiment involved the following operations. Elemental iodine crystals (I-127 containing radioactive I-131) were contained in the U-tube, and a small portion of the air supply was routed through the U-tube to sweep iodine vapor into the main stream. Iodine vapor escaping from the adsorber under test was collected downstream either by the CWS-6 filter; in the condensate drained periodically from the system; on

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* Dorex H-42, available from Connor Engineering Corporation, Danbury, Conn. Similar canisters are available from Charles E. Manning Company, Pittsburgh, Pa.

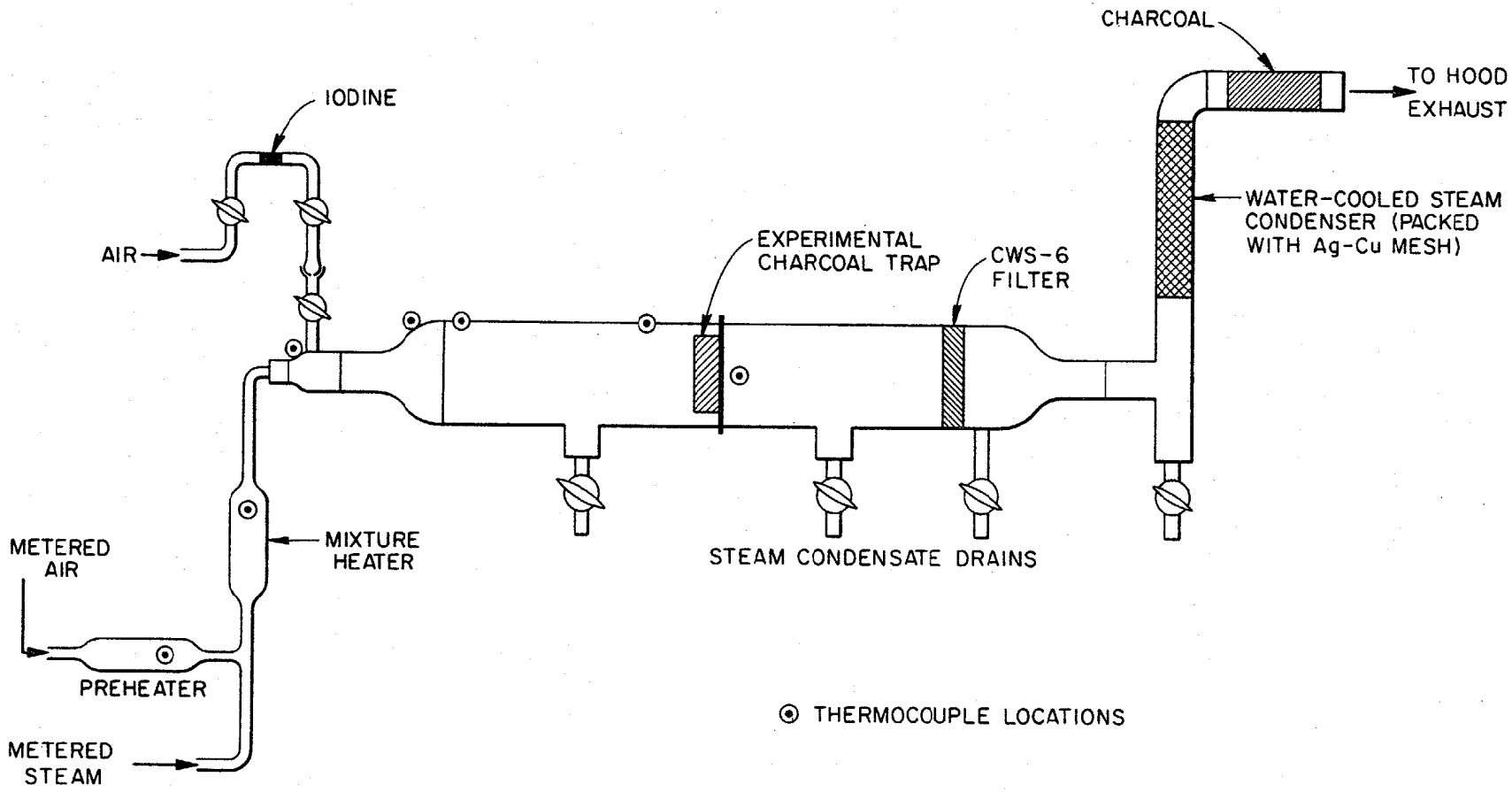


Figure 8 Experimental System for Steam-Air Experiments.

the silver-copper packing around the water-cooled condenser; or in the final room temperature charcoal adsorber. After completion of the test, the apparatus was cooled, completely drained, and disassembled. The iodine adsorption efficiency of the adsorber under test was then determined by radiochemical assay of all components of the system from the entrance face of the charcoal adsorber to the exhaust from the system. By comparing the amount of iodine residing in the test adsorber with the total amount found, including steam condensate, an iodine adsorption efficiency was obtained.

Six experimental runs were made at gas velocities ranging from 23.9 to 74.9 ft/min over the temperature range of 75 to 118°C and an average iodine removal efficiency of 99.9% was obtained. One test was performed at a temperature of 105°C and with gas velocity of 290 ft/min. To obtain a measure of the effect of water vapor on the iodine adsorption efficiency, two tests were made at approximately 100°C with air sweep alone. The results of these tests are contained in Table I.

Comparing the average iodine adsorption efficiencies under the two sets of conditions (99.9 vs 99.95%) it appears that the presence of water vapor does have a slight deleterious effect on the iodine adsorption efficiency of charcoal. The effect could occur by the blocking action of water vapor adsorbed on the charcoal surface thus preventing the iodine vapor from efficient and rapid contact with the charcoal surface. This effect coupled with the short bed depth of 0.75 inches would allow some iodine molecules to pass through the charcoal mass without contact with a charcoal surface.

The effect of superficial gas velocity through the charcoal mass on iodine adsorption efficiency may be observed by comparing run 7 with runs 1-6. Over the velocity range, 23.9 to 74.9 ft/min, a very small effect, if any, was noted. At a gas velocity of 290.8 ft/min the iodine adsorption efficiency dropped significantly. By operation of an iodine adsorber at a high gas velocity, less charcoal would be required for a given volume flow rate. However, as evidenced by these data, the resulting loss in iodine adsorption efficiency at higher gas velocities could not be tolerated in many reactor applications.

It is of interest to compare the iodine adsorption efficiency at 100°C with air sweep to a previous test of a full size canister at 25°C with air sweep. At 100°C the efficiencies were 99.93 and 99.98%; at 25°C the efficiency was 99.99%. The increased temperature lowered the iodine adsorption efficiency slightly.

Based upon the results of these tests it appears that activated charcoal exhibits a high efficiency for iodine vapor removal from air-steam mixtures for periods up to 2.75 hours with gas velocities less than 75 ft/min.

REMOVAL OF IODINE FROM HELIUM AT ELEVATED TEMPERATURES

In support of the Experimental Gas-Cooled Reactor Program (EGCR loops) a study of the removal of iodine vapor from high temperature helium streams was performed. Experiments were conducted by injecting iodine vapor (containing I-131) into a test system for a short period of time; then helium flow was continued for a longer period. During operation of the experiment a measure of the release of iodine from the experimental charcoal bed was obtained by monitoring the accumulation of I-131 radioactivity

TABLE 1

EFFICIENCY OF REMOVAL OF IODINE FROM STEAM-AIR MIXTURES ON CHARCOAL FROM DOREX H-42 CANISTERS

Run	Steam, % Sat.	Temperature of Charcoal (°C)	Gas Velocity (fpm)	Duration of Test (hrs.)	Iodine Removal Efficiency (%)
1	96.3	75	26.5	2.75	99.80
2	101.9	87	30.1	1.0	99.94
3	98.9	96	23.9	2.0	99.93
4	56.4	118	74.9	2.0	99.91
5	82.9	102	36.8	2.0	99.86
6	73.2	118	47.2	2.0	99.91
7	98.7	105	290.8	2.5	99.54
8	No steam	103	50.3	2.0	99.93
9	No steam	98	49.6	2.0	99.98

TABLE 2

RETENTION OF IODINE BY VARIOUS MATERIALS IN HELIUM AT ELEVATED TEMPERATURES

Adsorbent Material	Temperature (°C)	Bed Depth (in.)	Gas Velocity (fpm)	Duration of Test (hr.)	Iodine Retention (%)
Pittsburgh BPL Charcoal, 8-14 mesh	320	0.375	13.6-110.8	144	16.0
Columbia G Charcoal 8-14 mesh	320	0.25	13.6-110.8	166	94.7
Pittsburgh PCB Charcoal, 6-16 mesh	325	0.25	14.9-117.4	168	97.6
Pittsburgh BPL Charcoal, 12-30 mesh	325	0.25	13.6-75.6	168	30.2
Whetlerite ASC* Charcoal 12-30 mesh	325	0.25	13.4-44.1	216	99.99+
Silver-plated Copper mesh, 10 wt. % Ag	320	0.33	13.2-72.5	193	95.7

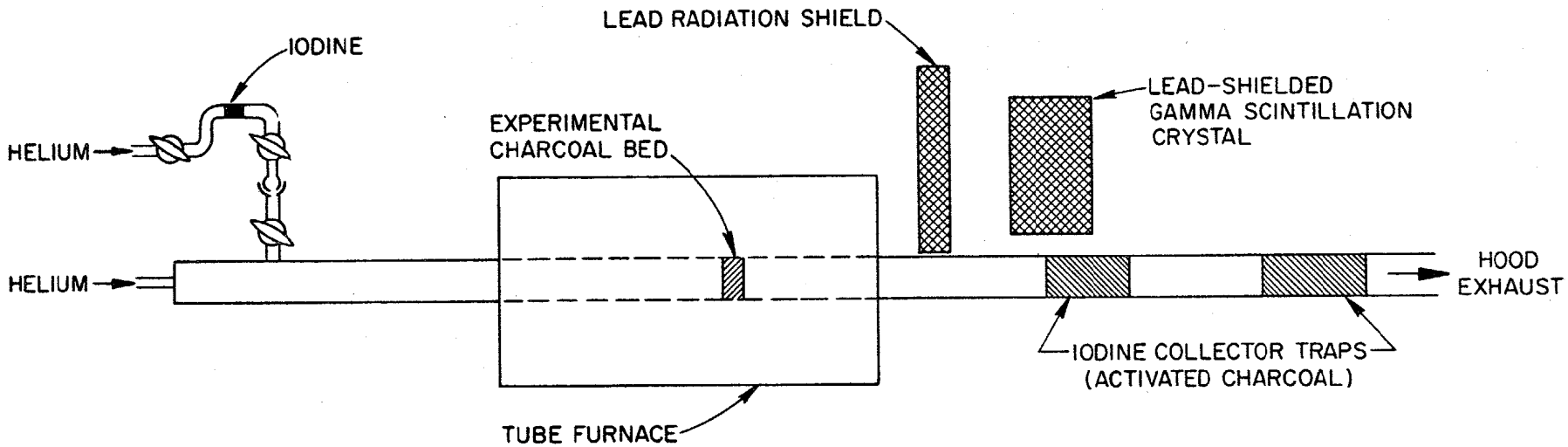
*This material is made by impregnating Pittsburgh BPL charcoal with salts of silver, copper, and chromium.

at the front of a downstream charcoal collector bed. In later tests the release of iodine from the experimental bed was also monitored by continuous scanning of the iodine distribution within the bed. Upon termination of the experiment, an I-131 assay was obtained beginning at the experimental bed and including all downstream components. With this information, an iodine efficiency figure is obtained for the test bed. Fig. 9 illustrates the experimental apparatus.

Initial test results indicated that activated charcoal removes iodine vapor from helium streams with high efficiency at temperatures as high as 430°C for periods up to 3 hours, but that continued helium flow will slowly remove the iodine contained in the charcoal bed. Additional study revealed that a wide variation in iodine removal and retention from high temperature helium exists among the various types of charcoal. A method of investigation was initiated to serve as a screening test of various materials for iodine retention and at the same time to obtain data for a more detailed study of the mechanisms of iodine release from various materials. To this end, iodine retention tests were performed using test adsorber beds approximately 0.25 inches deep to give a rapid indication of the relative value of the adsorbent material. Table 2 lists the pertinent test conditions and results obtained during this screening study. One type of charcoal, Whetlerite ASC, is noted to exhibit markedly superior iodine adsorption and retention characteristics. Whetlerite charcoal is prepared by impregnating Pittsburgh BPL charcoal with salts of copper, silver, and chromium. It is designed for use in gas masks for protection against war gases and is available in commercial quantities. The apparent value of the metal salts for iodine retention may be assessed by comparing the 30.2% iodine retention of Pittsburgh BPL, 12-30 mesh, to the 99.99+% iodine retention by Whetlerite, ASC, 12-30 mesh, both at 320°C.

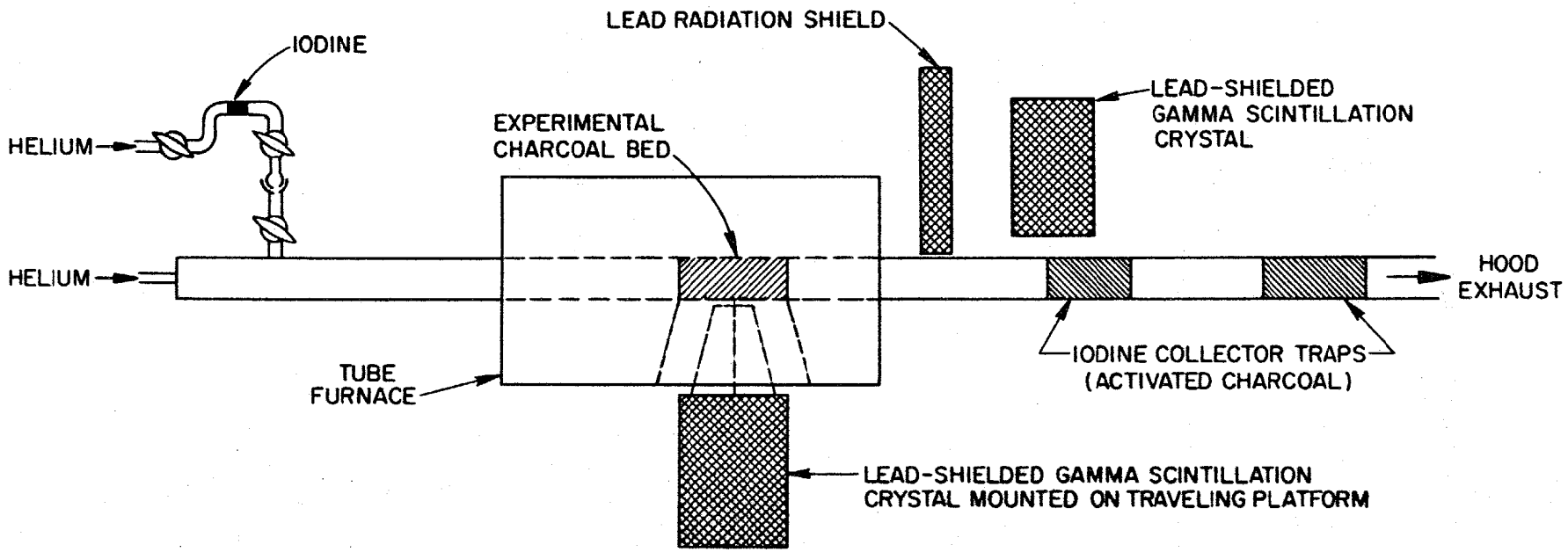
The balance of the program has been directed toward a study of the migration of iodine through relatively deep beds (~4 inches) of Whetlerite charcoal as a function of temperature. Fig. 10 indicates the experimental apparatus used. The distribution of iodine radioactivity in the test bed was monitored continuously by the gamma scintillation crystal mounted on the traveling platform. Iodine released from the test bed was accumulated on the downstream charcoal beds and also monitored by a gamma scintillation detector. Table 3 contains the results of this study. At temperatures up to 550°C the movement of iodine was hardly detectable. At 650°C the iodine was more mobile but the 4.25 inch deep bed of Whetlerite charcoal delayed the iodine penetration for ~143 hours (6 days).

Based upon the results of this laboratory study, Whetlerite charcoal appears well suited for iodine vapor removal in high temperature helium systems.



Experimental System for Helium Experiments.

Figure 9



Experimental System for Helium Experiments.

Figure 10

TABLE 3

RETENTION OF IODINE-131 BY WHETLERITE CHARCOAL IN HIGH TEMPERATURE HELIUM SYSTEMS

Temperature °C	Temperature °F	Helium Velocity (ft/min)	Duration of Test (hrs.)	Total Migration of Iodine (in.)	Retention of Iodine (%)
325	617	38.8	468	0.06	99.99+
425	797	53.0	897	0.07	99.99+
550	1022	53.7	602	0.15	99.99+
650	1202	60.2	220	>4.25*	86.73

*A small portion of the iodine content emerged from 4.25 in. trap at ~ 143 hours.

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1. R. E. Adams and W. E. Browning, Jr., Removal of Radioiodine from Air Streams by Activated Charcoal, ORNL-2872, March 17, 1960.

2. R. E. Adams and W. E. Browning, Jr., Removal of Radioiodine from Air-Steam Mixtures, ORNL-CF-60-11-39, November 14, 1960.

3. L. Silverman, R. Dennis, E. Kirstal, and F. Stein, Iodine Collection Studies, p. 322-343, TID-7593, October, 1960.

DISCUSSION

SILVERMAN: Thank you very much, Mr. Martin. On such short notice you did an excellent job. You did say that we should refer our questions to George Parker and George Creek, but I think, since you are intimately associated with the work, you might try to answer questions.

CHEEVER: How did you prepare the millipore filter samples for electron microscope observation?

MARTIN: This I don't know. We have submitted a sample of the millipore filter to the analytical department and they did that.

LOYSEN: You showed one slide near the end that had crystalline particles and then you had what looked like a lot of little ones in the background. Are these actual particles?

MARTIN: The particles themselves were very definite. This must have been the filter paper you were seeing. The particles appeared to be of a fairly good size, as I recall.

SILVERMAN: His last slide showed the comparison and it showed that the iodine is in the deposited state and I said that must have been the case.

MARTIN: There is a possibility, too, that there are trace amounts of oxygen with the UO_2.

SILVERMAN: This is right. You have stated that you vaporize UO_2 at these high temperatures.

MARTIN: The only time we feel we vaporize UO_2 is in the helium atmosphere. In the air atmosphere we collect the particles as U_3O_8. In CO_2, less than 2 percent of them are UO_2 and the remainder are U_3O_8. In the helium atmosphere, the particles were UO_2.

CRAIG: In your experiments did you make any attempt to find out, in the absence of the fuel element whether or not you had any background concentration of aerosol; if so, do you feel that adsorption or condensation on the aerosols already present in your system might have an influence?

MARTIN: We did not. We felt all along that we do get condensation on possible dust particles, because in, for instance, the uranium oxidation experiments we usually sucked room air through and there is a possibility that there could be dust particles. One more thing I might say. I have a list of several ORNL papers that do have more data on this, if anybody is interested.

SILVERMAN: Those of you who were at the Sixth Conference at Idaho remember we had a classified session on the confinement program at Hanford. We are fortunate today in having Mr. Heacock from Hanford to discuss the status of the confinement program, which, I believe ought to be finished by now.