THE CONTAINMENT OF FISSION PRODUCTS IN A POWER REACTOR

J. A. Riley
Risley, England

INTRODUCTION

Previous papers have discussed some of the basic mechanisms governing the behaviour of iodine present in gases and have also described investigations into methods of removal from gas streams. The present paper shows how these findings might be applied in general to a typical oxide fuelled gas cooled reactor.

The purpose of the gas treatment facilities in the power reactor postulated for this study is to prevent the uncontrolled discharge of gases containing radioactive isotopes, particularly iodine, to atmosphere either during normal operational procedures or during emergency conditions. Reactor containment may be considered in three stages: a pressure circuit itself, a biological shield which surrounds the pressure circuit and an outer containment structure which includes the space occupied by operating personnel and which needs to be designed in such a manner as to contain all gases which could be released as a result of any failure in the pressure circuit.

DISCHARGES TO BE TREATED

In order to design a comprehensive gas treatment system it is necessary to consider the types of discharge that can occur under normal and emergency conditions. These discharges and the circumstances in which they might contain radioactive material are listed below.

(a) Even with the highest standards of design and construction, some small leakage of coolant gas from the high pressure primary circuit will occur, and if the circuit gases were to be contaminated as a result of defective fuel elements then active gases would leak into the containment space formed by the biological shield. To prevent active gases diffusing into the operating areas, a flow of ventilating air may be passed from the operating spaces into the biological shield and eventually discharged from a stack. The air flow would be continuous and before discharge it would be necessary to treat it in order to maintain fission product releases below a specified level. The fission product removal plant must, therefore, operate continuously under conditions approximating to ambient temperature and pressure.
(b) Periodically it will be necessary, as part of the reactor operating routine, to blow-down the pressure circuit to atmosphere.

For a power producing reactor of 1000 M.W. thermal output, the amount of gas (assumed to be CO₂) in the primary circuit may amount to 100 tons. This means that if the blow-down operation is to be completed in a period of say, 5 hours, then high gas flow-rates will result. The treatment of such a gaseous discharge to remove active species thus requires a plant that, although not required to operate continuously, must be capable of handling large gas volumes at elevated temperatures for short periods.

(c) The reactor primary circuit would need to be protected by a system of relief valves, which under certain conditions such as a sudden pressure rise caused by the rapid failure of a heat exchanger tube, would discharge CO₂ and possibly a mixture of CO₂ and steam. The gas discharge rate may amount to several hundred pounds per second and in order to treat this type of discharge a plant of high through-put is necessary with the additional requirement that it must be immediately available or capable of guaranteed automatic start-up if a relief valve lifts.

(d) In the unlikely event of a rupture of the primary circuit, the reactor would rapidly depressurise and the released gases would enter the main containment structure. This type of incident could be associated with the failure of a large number of fuel element cans or even the burning of parts of the reactor core; in either case a large amount of activity would be liberated. To minimise the leakage of active gases from the containment structure, and also to minimise the deposition of fission products on the surfaces within the containment, these gases should be treated as rapidly as possible. Unfortunately the decontamination factors obtainable with presently available removal processes may not be sufficient to allow direct discharge of the treated gases to atmosphere and it becomes necessary to arrange for the gases to be recycled from the outlet of the treatment plant back to the containment structure so that by repeated passage through the plant they can be decontaminated to the required level for discharge.

The above types of gas discharge to be treated can be divided into two classes, those which are continuous and are associated with low activity levels, and those which are of an intermittent nature and which may contain considerable amounts of activity.

GAS TREATMENT PROCESSES

Of all the fission products which might be released in a major incident, consideration of both the quantities released and their biological significance indicate that radio-iodine is the most important. Consequently in the following paragraphs attention is concentrated on the probable behaviour of iodine.
Iodine in the gases leaking from the primary circuit or liberated into the containment following a major incident will be present in at least two forms; free vapour and vapour adsorbed on particulate matter and condensation nuclei. The gas treatment processes must therefore comprise gas filtration followed by a process for removing iodine vapour. Of the methods for removing iodine vapour from gas streams which have been investigated in the past, the two types most applicable to the present study are absorption by liquids in scrubbing towers, and adsorption on solids, notably activated carbon. Processes based on absorption in liquids require a supply of liquid absorbent, a scrubbing tower and a liquor circulating pump, and although suitable for continuous operation becomes unattractive for intermittent operation and for situations where the gas rate can fluctuate widely, since they either have to be operated unnecessarily during periods when no gas is flowing or have to be capable of starting up automatically without delay and with complete reliability on receipt of some signal. Also, they produce large volumes of active effluent which require subsequent treatment. Since liquid absorption processes have been found to give lower efficiencies than those obtainable with activated carbon beds, and because of their inherent disadvantages they are not considered further in the present study. Provided solid adsorbents do not deteriorate with time, adsorption systems are immediately available in an emergency since they contain no moving machinery and do not require any power supplies. Activated carbon, the most effective adsorbent for iodine vapour, suffers from the disadvantage that it can ignite in oxidising atmospheres at fairly low temperatures and it is advisable to limit the temperature to say 100°C, depending on the carbon.

GAS TREATMENT APPLICABLE TO A TYPICAL REACTOR SYSTEM

To illustrate the application of gas treatment methods to a power reactor, the case of a 1000 M.W. thermal reactor using uranium dioxide fuel canned in stainless steel has been considered. A schematic diagram of the containment and gas treatment systems is shown in Fig. 1. If the fuel rating is 10 M.W./t.e. U and at equilibrium conditions the amount of $^{131}$I in the free space inside the can is taken as 1% of that in the fuel material itself then the $^{131}$I which is readily available for release will amount to approximately 2,000 curies per tonne of fuel in the core.

Leakage from the primary circuit

If the leakage rate from the primary circuit is assumed to be 1/2% per day of the total coolant, and if the maximum incident condition under which the ventilation air treatment plant would be operated was that resulting from the failure of 0.1% of all the cans in the reactor, then the rate of release of $^{131}$I into the ventilation air would be 1 curie/day which would be about ten times the probable permissible daily discharge of $^{131}$I over a long period from a high stack. Thus the ventilation treatment plant should provide a decontamination factor of at least ten.
Figure 1

SCHEMATIC ARRANGEMENT OF GAS TREATMENT PLANTS
The ventilation air flow rate may be as much as 5000 c.f.m. and since the specific activity of $^{131}$I in irradiated fuel may be of the order $10^4$ curies/g. total iodine, the actual mass concentration of iodine in the ventilating air will be only about $5 \times 10^{-4}$ ug/M$^3$. At this low concentration, experimental evidence suggests that to achieve a D.F. of the required order would require the use of "absolute" filters followed by a carbon bed sized to give a gas residence time of about 0.5 sec. In order to reduce the loading of the "absolute" filters they should be preceded by coarse filters capable of recovering particles down to 5 microns. The "absolute" filters would have a removal efficiency of 99.99% down to a particle size of 0.1 micron. A requirement of the ventilation plant is that the pressure drop across it should be kept as small as possible to minimise the ventilation fan power requirements. This means that the depth of the carbon bed must be small but yet must be sufficient to ensure that channelling of the gas does not occur as a result of poor packing of the carbon particles. The size of the plant items are given below:

Prefilter area  
Absolute filter area  
Carbon bed (12 mesh)  
After filter area  
Total pressure drop

The "after filter" is included to remove any carbon particles entrained from the carbon bed. The above carbon bed actually gives a residence time of 1 sec. the bed area being chosen to limit the pressure drop.

Gas discharges from these two cases can be treated conveniently in a common plant since they are both intermittent and of fairly short duration. The rate of discharge from the relief valves will actually determine the size of the plant. Postulating again that 0.1% of all the fuel cans have burst, blowdown of the reactor circuit would result in the discharge of 200 curies of $^{131}$I if the gases were not treated. If in this case we assume that the discharge limit on a blowdown, which will occur only occasionally, is 1 curie, then the blowdown plant must provide a D.F. of at least 200. During blow down of the reactor circuit the iodine mass concentration in the gases would be about 0.5 ug/M$^3$ and at this concentration the combined D.F. of a filtration system plus an activated carbon bed with a residence time of 0.5 sec. might be expected to be 1000. The gases leaving the reactor may be at about 300°C and it might be necessary to cool them to 100°C to avoid damage to the carbon bed. A conventional water cooled heat exchanger could be used but it may be more desirable to use a form of heat sink consisting of a bed of stone chippings to which the gas can give up its heat. This system has the advantage that it is immediately available as is the rest of the gas treatment.
plant. The heat sink principle is not useful where a continuous heat removal facility is required or if it was thought likely that it would be necessary to blow down the circuit frequently over a short period of time.

The sizes of the filters and the carbon bed depend on the volumetric flow rate, which for a fixed mass rate depends on the absolute pressure. With the high flow rates occurring during relief valve discharge the pressure drop along the ducting from the treatment plant to the stack may be such that the absolute pressure in the plant is a few atmospheres. Assuming that this absolute pressure is 2 atmospheres the sizes of the plant items are given below for a relief valve discharge rate of 200 lb./sec.

<table>
<thead>
<tr>
<th>Plant Item</th>
<th>Size</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heat sink</td>
<td>300 ft$^2$ in area. 12 ft. thick packed with 1&quot; stone chippings.</td>
</tr>
<tr>
<td>Prefilter area</td>
<td>250 ft$^2$</td>
</tr>
<tr>
<td>Absolute filter area</td>
<td>250 ft$^2$</td>
</tr>
<tr>
<td>Carbon bed</td>
<td>600 ft$^2$ area and 1 ft. thick</td>
</tr>
<tr>
<td>After filter area</td>
<td>250 ft$^2$</td>
</tr>
<tr>
<td>Total pressure drop</td>
<td>60 in. W.G.</td>
</tr>
</tbody>
</table>

Since "absolute" filters are fragile and will only stand a limited pressure drop across them some consideration must be given in a design to the shock wave that will be generated when the relief valves lift.

**Major rupture of the primary circuit**

If in the remote event of a major rupture in the primary circuit say 20% of the fuel element cans fail then some thousands of curies of $^{131}$I could be liberated in the containment space. It is known that iodine rapidly plates out on surfaces and this would rapidly reduce the amount of gas-borne iodine within the containment.

It is necessary, however, to process the containment gases as rapidly as possible because of the leakage from the containment structure being fairly close to ground level where permissible discharge levels are lower than from high stacks. With present knowledge of iodine removal methods it is not possible to provide plants which will give a guaranteed once-through D.F. of several thousand and it becomes necessary to provide means for re-circulating the gases through the plant and back to the containment until it becomes possible to discharge the gases to atmosphere.

It is desirable to process the gases as rapidly as possible to minimise the leakage from the containment structure. A recirculation time of only a few hours is desirable and this means that the plant must have a large
throughput and will require powerful recirculating fans with guaranteed power supplies. The plant itself will consist of a cooler to limit the gas temperature to 100°C followed by filters and a carbon bed as described for the plants above. If it is thought likely that a graphite fire could follow a major incident then the amount of heat to be continuously removed by the gas cooler might necessitate the provision of a water cooled exchanger rather than a simple heat sink.

The size of a plant for a recirculation rate of about 30,000 c.f.m. is given below. The gas cooler in this case is a water cooled finned tube heat exchanger designed to cool the gases from 300°C to less than 100°C on a continuous basis.

<table>
<thead>
<tr>
<th>Equipment</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heat exchanger</td>
<td>finned tubes in duct 6 ft. square</td>
</tr>
<tr>
<td></td>
<td>total length of finned tubing 6000 ft.</td>
</tr>
<tr>
<td></td>
<td>Water requirement 720 gallons/min. at 70°F.</td>
</tr>
<tr>
<td>Prefilter area</td>
<td>120 ft²</td>
</tr>
<tr>
<td>Absolute filter area</td>
<td>120 ft²</td>
</tr>
<tr>
<td>Carbon bed</td>
<td>250 ft² area and 1 ft. deep</td>
</tr>
<tr>
<td>After filter area</td>
<td>120 ft²</td>
</tr>
<tr>
<td>Recirculating fan</td>
<td>Recirculating fan about 250 H.P.</td>
</tr>
</tbody>
</table>

**BEHAVIOR OF FISSION PRODUCTS OTHER THAN IODINE**

The discussion so far has been concerned mainly with iodine. The plants described above would be effective in the removal of other volatile fission products Br, Te and Cs from the gas streams while particulate activity like Sr would tend to be removed on the filters. The noble gases Kr and Xe would not be retained on the carbon beds to any extent, but their biological significance is very small when discharged from high stacks.

**FUTURE DEVELOPMENT**

In the above gas treatment plants, heat removal equipment is necessary to protect the active carbon beds. The development of copper mesh systems, which can operate at elevated temperatures, may present a more attractive method of iodine vapour removal for use with future reactor systems.
CONCLUSIONS

The above discussion gives an outline of how existing techniques for iodine removal from gas streams might be applied to a gas cooled reactor system. Further development work and a more complete understanding of the forms in which iodine can occur following release from fuel elements may enable higher efficiency plants to be designed thereby eliminating the need for recirculating systems and allowing the rapid treatment of all reactor discharges on a once-through basis.
AIR CLEANING IMPLICATIONS IN FALLOUT STUDIES

George T. Anton
U. S. Atomic Energy Commission
Washington, D. C.

ABSTRACT

The relationships between air cleaning and air sampling research connected with fallout studies are indicated. Efforts of more specific interest to the air cleaning community are pointed out. Capability of the AEC, Health and Safety Laboratory, at its new facility for advanced aerosol technology is explained.

Section II is devoted to radioactive particle sizes from early fallout at the Pacific and Nevada nuclear tests and to the stratospheric dribble of fallout from the world-wide reservoir of radioactivity injected above the troposphere. This subject is presented because of the air cleaning implications in Civil Defense but probably more because of the special problems of air cleanliness connected with industrial processing.

SECTION I

Upper Air Sampling and Air Cleaning

In sampling the atmosphere we have most of the problems, study the same parameters and employ the same processes that are so familiar to you in air cleaning. We like (1) large sampling volume flow rates, (2) efficient removal of particulates usually down to some sub-micron size limit - say less than 0.01\textmu m in the stratosphere, (3) knowledge about individual particles and size distribution. Research on filters and filter theory plus the concomitant instrumentation and equipment design are problems very much mutual. We study filters, electrostatic precipitators, molecular sieves combined with cryogenics for whole air sampling, impactors and thermal precipitators. Our principal effort comprises sampling air for particulates and in so doing we often range into parameters that may vary considerably from those in air cleaning. We are more interested in what is removed on a filter than in the air that passes through it. We sometimes spend thousands to obtain a very small sample and then several hundreds of dollars trying to preserve and analyze it.

We part company greatly when we go into reduced pressures at higher altitudes. There the Cunningham slip factor in the particle transport equation become important and air mass sampling rate at the same time reduces.
factorially. In fact all the parameters we usually think about in the sampling equations are changed under conditions of reduced pressures, sometimes by orders of magnitude. All this may be new only to those present who have had no occasion to think about it.

Direct Flow Sampler (DFS) developed by General Mills, Incorporated (Fig. 1 and 2) employs one square foot of IPC (Institute of Paper Chemistry) paper; a porous, high volume, medium background-count paper. At face velocities above 500 feet per minute the collection efficiency is high and at around 90 K feet it approaches 100% for particle sizes down to small fractions of a micron. We sample at 500-700 feet per minute and at altitudes of 50 thousand feet and above. We are working toward a sampling capability of around 120 thousand feet with the present model. Efficiency would be of around 40% if this same paper and equipment were used at ground level for air cleaning. (See Fig. 4 and 5)

The Electrostatic Precipitator (EP) developed by Del Electronics Corp. (Fig. 3) provides a similar difference in usage and design. Well up in the stratosphere, say 100 to 150 K feet, we believe, thanks to work by Dr. Junge, that we must sample particulates that are predominantly in the .01 to .10 micron diameter range and perhaps even as small as .001 micron at the latter altitude. Both Davies and Cunningham have indicated that the product of the diameter and pressure varies directly and almost linearly with a slip correction factor through three orders of magnitude. Vary pressure or diameter and the correction is proportional to the new product. This means that with increase in altitude or with decrease in particle size the efficiency of collection should increase. In practice it is not so simple. There are many factors some of which, such as ionic winds, may be quite significant. Herein, however, lies its potential over the DFS in the zone around 100 to 150 K feet.

One specific difference in design criteria is the adjustment we must make in voltage. It must be set for the pressure or altitude at which it samples because the breakdown strength of the air reduces with altitude up to about 150 K after which it rises again.

For example, when the product of \( d \times p \) in microns and millimeters respectively has a value of .10 the Cunningham slip factor correction is 1500. If we design for a light, very efficient, high volume (in our language this means 500 to 1000 cfm) sampler for a narrow, specific particle size range then we would expect to lose the particles larger than design size by passage out the end. Our design and research work on the high altitude EP will be of little value in normal problems of air cleaning but the attendant basic research will be useful.

Areas of Related Work

The Fallout Studies Branch pursues a generalized objective of learning how to predict what will happen to radioactivity injected into the atmosphere at any location and time. This innocuous statement has far reaching implications
Figure 1
Ash-Can Sampler - an original model of the stratospheric sampler having a cylindrical filtering surface of 5 sq. ft.
Figure 2

Direct Flow Sampler (DFS) - 1961 Test Model - The DFS has displaced the Ash Can System. Latest modification will remove louvers in front of the filter.
Figure 3

Test Flight preparations showing two Direct Flow Samplers with the Electrostatic Precipitator mounted across them shortly before launch.
I.P.C. FILTER PAPER COLLECTION EFFICIENCY VERSUS VELOCITY FOR VARIOUS SIZES OF SPHERICAL PARTICLES AT STATION PRESSURE
I.P.C. FILTER PAPER COLLECTION EFFICIENCY VERSUS VELOCITY FOR VARIOUS SIZES OF SPHERICAL PARTICLES AT 83,000 FEET ALTITUDE

FIGURE 5
in both scope and complexity. Distinct areas of study may be listed to show this: (1) input information on formation, characterization and abundance of radioactive debris, (2) aerospace characterization of transport, mixing and circulation from the point of injection through the various atmospheric layers until it deposits on the earth which, in turn, characterizes the space and time patterns of deposition, (3) fate of the fallout or distribution with time in man, by direct intake through inhalation and ingestion or through a food chain.

Effort of interest to the air cleaning community includes areas such as (1) aerosols in sizes from near molecular to a few hundred microns diameter, (2) filters and impactors for removal of particulates and particle sizing, (3) air sampling devices for various purposes, (4) sampling methods for a variety of environmental conditions, (5) testing facility for the preceding to intercompare, calibrate and standardize equipment, methods and data.

Work of More Specific Interest

The basic work done at General Mills, Inc. at Minneapolis on filters and impaction and the theoretical studies on particle dynamics at Johns Hopkins University by S. K. Friedlander should be useful to nearly everyone here.

Work on Micrometeorology at the AEC National Laboratories, produce by-product instruments and techniques that some of you might find useful.

Armour Research Foundation has a project under Langer to develop an electrostatic particle size classifier. There appears to be some hope that this one might work.

Dr. Clyde Orr's thermal precipitator work at Georgia Tech. is about complete. It may find its place as a particulate sampler.

The aerosol testing facility at the Health and Safety Laboratory of our New York Operations Office is probably the subject of greatest interest to this community. It is being set up under our colleague Alfred Breslin to do the necessary research and servicing in support of the sampling work being done in our fallout program.

They will have a walk-in sized environmental chamber, filter testing equipment, aerosol and dust generating equipment and chambers, air sampler calibration equipment and particle sizing equipment which includes an electron microscope.

Here I emphasize their capability for generating fairly homogeneous aerosols of various controlled sizes for use in calibrating and comparing filters and samplers. The present lower limit for particle generation is an aerosol that has a count median diameter of .008 microns, a mass median of .025 microns, and a sigma of 1.7. This step promises to be a quantum jump forward in aerosol technology for use in sampler evaluation and standardization techniques. Dilute solutions of uranine dye are sprayed from a Dautreband or
similar aerosol generator. Coarse particles are removed by perforated plates
designed for passage of controlled particle sizes. An ion generator feeds ions
downstream of the control plates to neutralize the charges on the particles
just as the stream passed into a plenum chamber. Aerosol is drawn from the
opposite end of the chamber. Assessment is done using isokinetic millipore
filter samplers and a fluorimeter, sensitive to about $10^{-11}$ gm., to measure
the dissolved uranine from the filter. Electron microscope grids are placed
on the collection surface to collect particle sizing samples.

SECTION II

Particle Sizes and Radiological Defense

There is considerable current interest in particle sizes of radioactive
fallout in connection with Civil Defense. I am including, therefore, some
information that may be useful in air cleaning applicable to this subject.

Early Fallout Particle Sizes

Particle size of fallout decreases with distance from Ground Zero. Also, the size decreases with increase in lateral distance from the line of
maximum radiation intensity or "midline" of the fallout pattern.

The relative amount of radioactivity associated with particles less
than 44 u in diameter increases as the mass of cab and tower materials is
decreased. Since the smaller sizes travel further before deposition the amount
of fallout at greater distances from Ground Zero in this smaller particle size
range increases.

Test devices detonated on 500 and 700-foot towers produced approximately
30% of the fallout radioactivity in sizes less than 44 u diameter within the limits
of 1 mile from Ground Zero out to a distance corresponding to $H + 12$ hr. fallout
time. A device of comparable yield mounted on a 700-foot balloon produced
70% of the activity in particle sizes less than 44 u.

Within the class of particles less than 44 u diameter it was found that
38 to 50% of the radioactivity was associated with particles less than 5 u dia-
meter for tower shots and for balloon-supported detonations the figure was
found to be 51 to 83%. The following table illustrates these figures.
Method of Support | Distance Limits | Radioactivity in Particle Sizes less than 44 μ | Percent of activity within the 44 μ Fraction Associated with Size Range < 5 μ
---|---|---|---
500 to 700 ft. Steel Towers | 1 mile from GZ out to Distance corresponding to H / 12 hr. fallout | 30 % | 38% to 50 %
700-foot Balloon | Same | 70 % | 51% to 83 %

Data not yet published\(^8\) from Operation Plumbbob at Nevada will show that significant percentage contributions of radioactivity by particles less than 5 microns in diameter were observed at virtually all sampling locations for both tower and balloon supported detonations. These data should be scrutinized carefully when they become available.

Another significant finding by the UCLA group, from which most of this information is derived, is that the debris from balloon supported bursts is not only smaller in particle size but is also significantly more soluble in water or 0.1N hydrochloric acid. The implication is that more of the radioactivity from tower shots is incorporated or fused inside insoluble particles when the vaporized and melted tower, perhaps soil and other insoluble materials cool and condense. Another implication is that an air burst of an operational bomb, not having even the cab and instrumentation material mass as employed in a balloon burst, will have even smaller particle sizes and be more soluble than the figures shown for balloon supported detonations. There will be no early fallout, however, from such an air burst.

### TABLE 2

<table>
<thead>
<tr>
<th>Support</th>
<th>Particle Size Range (Microns)</th>
<th>Solubility (Percent of ( \gamma ) Activity)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Water</td>
<td>0.1N HCl</td>
</tr>
<tr>
<td>Tower</td>
<td>&gt; 44</td>
<td>&lt; 1</td>
</tr>
<tr>
<td></td>
<td>&lt; 44</td>
<td>&lt; 2</td>
</tr>
<tr>
<td>Balloon</td>
<td>&gt; 44</td>
<td>31</td>
</tr>
<tr>
<td></td>
<td>&lt; 44</td>
<td>14</td>
</tr>
</tbody>
</table>
A few statements and conclusions from WT-1172\textsuperscript{7} contain additional pertinent information:

1) From the abstract, "From careful consideration of numerous pertinent physical and physiological factors and from analysis of field and laboratory investigations, it is evident that there is no apparent situation in nuclear warfare where, during the first few days after the detonation, one could inhale sufficient radioactive material to induce a serious radiation injury to lungs or intestines without simultaneously being subjected to supralethal doses of external beta-gamma radiation."

2) From the Discussion pertaining to insoluble material, "For example, if one were inhaling airborne particles in the 0.1 to 50.0 \textmu{} size range, the 5- to 50-\textmu{} size particles would initially be trapped in the nose and upper respiratory tract. If these particles were insoluble, they would be carried rapidly to the throat, swallowed, and eliminated in the feces within a few days." and later: "animals retain far greater amounts (factors of 10 to 150 times) of radioactive particles in the intestinal tracts than are retained in the respiratory organs following an acute inhalation exposure. Under field conditions, where the greatest amount of radioactivity is found in the large particles, the acute radiation damage from inhalation, if any, is far more likely to involve the intestinal organs than those of the respiration because the bulk of all initially inhaled insoluble material is sooner or later deposited in the intestines, where it remains for longer periods before being eliminated."

The foregoing may be simplified into about two statements regarding the inhalation hazard in early fallout from a nuclear situation:

1) The inhalation hazard is a secondary immediate hazard by 2 to 3 orders of magnitude when compared with the external penetrating radiation exposure hazard, and 2) the preponderance of radioactivity will be associated with particles larger than 1\textmu{} diameter and over 90\% protection down to this size may be obtained simply by breathing thru 16 folds of an ordinary, dry, men's handkerchief during cloud passage. This latter is, of course, fortuitous because if the inhalation hazard were really significant the external exposure would be well above the lethal range. I suppose most people would take the precaution, anyway, for that added measure of protection, but probably more, because the feeling that positive action is being taken would make one feel more protected.

\textbf{Stratospheric Fallout Particle Sizes}

This information is taken principally from the work done for the Atomic Energy Commission by the Armour Research Foundation.\textsuperscript{10} It may be of interest, not as a Civil Defense point of interest, but as a specialized problem in air cleaning. Some examples may be the film manufacturing industry, low background radiochemical and counting laboratories, and similar situations where a tiny bit of contamination is too much. There are situations in laboratories today in which samples of marginally detectable amounts of material
cost in excess of $100,000 to obtain. In some such instances even exposure to the ordinary atmospheric contaminants could ruin chances for a good analysis.

With the background of pointing out extremes of possible conditions let us go to the little data we have at hand.

Equipment used was a roughing cyclone followed by a small glass cyclone which was followed by a 9-inch Millipore filter. A second system was a 6-stage Andersen sampler followed by a 47-mm Millipore filter.

TABLE 3

<table>
<thead>
<tr>
<th>Particle Diameter Microns</th>
<th>Roughing Cyclone</th>
<th>Small Cyclone</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0 - 1.4</td>
<td>11</td>
<td>9</td>
</tr>
<tr>
<td>1.4 - 2.0</td>
<td>23</td>
<td>20</td>
</tr>
<tr>
<td>2.0 - 2.8</td>
<td>48</td>
<td>59</td>
</tr>
<tr>
<td>2.8 - 4.0</td>
<td>63</td>
<td>92</td>
</tr>
<tr>
<td>4.0 - 5.6</td>
<td>74</td>
<td>100</td>
</tr>
<tr>
<td>5.6 - 8.0</td>
<td>88</td>
<td>100</td>
</tr>
</tbody>
</table>

The small cyclone effectively removed particle sizes 2.5 to 5.0 μm.

TABLE 4

<table>
<thead>
<tr>
<th>Impaction Stage</th>
<th>Jet Velocity fps</th>
<th>Particle Size Retained Microns</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3.4</td>
<td>7.0 and larger</td>
</tr>
<tr>
<td>2</td>
<td>4.89</td>
<td>5.5 - 10.0</td>
</tr>
<tr>
<td>3</td>
<td>9.75</td>
<td>3.5 - 5.5</td>
</tr>
<tr>
<td>4</td>
<td>17.31</td>
<td>2.0 - 3.5</td>
</tr>
<tr>
<td>5</td>
<td>41.92</td>
<td>1.0 - 2.0</td>
</tr>
<tr>
<td>6</td>
<td>76.40</td>
<td>0.7 - 1.0</td>
</tr>
</tbody>
</table>

-542-
<table>
<thead>
<tr>
<th>Dates</th>
<th>Roughing Cyclone</th>
<th>Small Cyclone</th>
<th>Millipore Filter</th>
<th>Roughing Cyclone</th>
<th>Small Cyclone</th>
<th>Millipore Filter</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>7/2/59</td>
<td>34</td>
<td>19</td>
<td>47</td>
<td>360</td>
<td>3,990</td>
<td>12,900</td>
<td>6,990</td>
</tr>
<tr>
<td>7/17/59</td>
<td>29</td>
<td>17</td>
<td>54</td>
<td>112</td>
<td>1,321</td>
<td>4,239</td>
<td>2,545</td>
</tr>
<tr>
<td>7/28/59</td>
<td>29</td>
<td>17</td>
<td>54</td>
<td>112</td>
<td>1,321</td>
<td>4,239</td>
<td>2,545</td>
</tr>
<tr>
<td>8/17/59</td>
<td>29</td>
<td>17</td>
<td>54</td>
<td>112</td>
<td>1,321</td>
<td>4,239</td>
<td>2,545</td>
</tr>
<tr>
<td>8/29/59</td>
<td>29</td>
<td>17</td>
<td>54</td>
<td>112</td>
<td>1,321</td>
<td>4,239</td>
<td>2,545</td>
</tr>
<tr>
<td>9/8/59</td>
<td>42</td>
<td>18</td>
<td>40</td>
<td>58</td>
<td>221</td>
<td>1,120</td>
<td>510</td>
</tr>
<tr>
<td>9/11/59</td>
<td>15</td>
<td>8</td>
<td>77</td>
<td>61</td>
<td>320</td>
<td>325</td>
<td>285</td>
</tr>
<tr>
<td>9/30/59</td>
<td>15</td>
<td>8</td>
<td>77</td>
<td>61</td>
<td>320</td>
<td>325</td>
<td>285</td>
</tr>
<tr>
<td>10/7/59</td>
<td>43</td>
<td>21</td>
<td>36</td>
<td>57</td>
<td>582</td>
<td>826</td>
<td>432</td>
</tr>
<tr>
<td>10/20/59</td>
<td>43</td>
<td>21</td>
<td>36</td>
<td>57</td>
<td>582</td>
<td>826</td>
<td>432</td>
</tr>
<tr>
<td>10/27/59</td>
<td>36</td>
<td>18</td>
<td>46</td>
<td>60</td>
<td>426</td>
<td>658</td>
<td>399</td>
</tr>
</tbody>
</table>

The above table shows that the total beta activity ranged from 285 to 7,000 pc/g of material collected. Each gram of material collected in the small cyclone (mainly particles in the 2.5 to 5-micron range) had 5 to 10 times more total beta activity than a gram of material retained in the roughing cyclone. Further, each gram of material on the Millipore filter was 1 to 5 times more radioactive than a gram collected in the small cyclone.
### TABLE 6

**RADIOACTIVITY ASSOCIATED WITH DRY ATMOSPHERIC PARTICLES CLASSIFIED BY THE ANDERSEN SAMPLER**

<table>
<thead>
<tr>
<th>Dates</th>
<th>Total Beta Activity, pc/100,000 Ft.³</th>
</tr>
</thead>
<tbody>
<tr>
<td>7/15/58</td>
<td>1,315</td>
</tr>
<tr>
<td>10/20/58</td>
<td>1,999</td>
</tr>
<tr>
<td>11/18/58</td>
<td>1,935</td>
</tr>
<tr>
<td>2/13/59</td>
<td>2,000</td>
</tr>
<tr>
<td>6/1/59</td>
<td>1,207</td>
</tr>
<tr>
<td>9/18/59</td>
<td>1,935</td>
</tr>
</tbody>
</table>

*Stages combined during period 1 and 2, 3 and 4, 5 and 6.*

Total beta activity for the Andersen sampler ranged from 1,300 to 2,000 picocuries (pc)/100,000 cubic feet of air processed. Particles in the 1- to 2-μ range and particles below 0.7 μ were main contributors.

For both sampling systems, the majority of the activity was associated with particles collected on the Millipore filter. Sizes on the Millipore were those under about 2 or 2.5 μ for the Cyclone system and under about 0.7 μ for the Andersen Sampler.

**Comparison of Particle Sizes from Pacific and Nevada Tests**

A few but significant factors distinguish particles from Pacific tests from those at Nevada. Among the physical factors that affect the physical and chemical nature of fallout material are: 1) type of detonation (high altitude, tower, surface or near-surface, or sub-surface); 2) type and amount of material engulfed by the fire-ball (type of soil if it is a surface-land burst); and, 3) percent-fission yield and size of the detonation. These factors will form the guide lines regarding such things as solubility in the body and delineation of the hazard from intake by ingestion and inhalation.

It was shown earlier that Nevada particles incorporating radioactivity are less soluble in water or in 0.1N HCl than are particles from Pacific tests. This is explainable from the knowledge that Pacific particles are mostly of coral origin whereas those from Nevada are from soil or form the mass of metals supporting those shots and are largely insoluble silicates.
and metal compounds.

Particle sizes from Pacific tests are larger probably due to agglomeration history of these particulates which are different chemically and are hygroscopic. Fall on the Marshallese at Rongelap Island in 1954 was visible and variously described as a sooty-snowlike material. Several dozen natives received sickness doses of radiation but surveillance since that time has shown that very little body burden was accumulated from either inhalation or ingestion during the 2 days before evacuation.

A report of studies on particles and particle sizes made during Operation Redwing in the Pacific has been examined. It indicates that particle sizes of fallout generally run much larger and that very little if any activity is found in the 0.1- to 5-u size range. It found, also, that about 2 to 10% of the particles did not contain activity despite detonation on the surface. At one station studied it was found that activities associated with a size group followed a normal distribution.

The following tables, taken from two shots, are presented.

### TABLE 7

**Distribution and Activity Characteristics of Particle Sizes**  
(Shot A, Activities at H + 12)

<table>
<thead>
<tr>
<th>Size Group (µ)</th>
<th>Number of Particles</th>
<th>Composite Activity (well c/m)</th>
<th>Minimum</th>
<th>Maximum</th>
<th>Median</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>31-42</td>
<td>8</td>
<td></td>
<td>78</td>
<td>11,354</td>
<td>835</td>
</tr>
<tr>
<td>43-60</td>
<td>20</td>
<td></td>
<td>33</td>
<td>833,600</td>
<td>6,985</td>
</tr>
<tr>
<td>61-84</td>
<td>37</td>
<td></td>
<td>58</td>
<td>459,321</td>
<td>12,213</td>
</tr>
<tr>
<td>85-102</td>
<td>6</td>
<td></td>
<td>4,460</td>
<td>50,608</td>
<td>32,143</td>
</tr>
<tr>
<td>103-120</td>
<td>42</td>
<td></td>
<td>69</td>
<td>525,449</td>
<td>4,112</td>
</tr>
<tr>
<td>121-145</td>
<td>13</td>
<td></td>
<td>19,063</td>
<td>683,362</td>
<td>77,622</td>
</tr>
<tr>
<td>146-170</td>
<td>34</td>
<td></td>
<td>3,686</td>
<td>771,326</td>
<td>113,209</td>
</tr>
<tr>
<td>171-200</td>
<td>24</td>
<td></td>
<td>3,816</td>
<td>1,675,122</td>
<td>166,982</td>
</tr>
<tr>
<td>201-240</td>
<td>27</td>
<td></td>
<td>25,565</td>
<td>1,310,318</td>
<td>168,795</td>
</tr>
<tr>
<td>241-260</td>
<td>25</td>
<td></td>
<td>32,178</td>
<td>726,969</td>
<td>145,494</td>
</tr>
<tr>
<td>261-315</td>
<td>9</td>
<td></td>
<td>53,105</td>
<td>493,500</td>
<td>223,424</td>
</tr>
<tr>
<td>316-382</td>
<td>1</td>
<td></td>
<td>---</td>
<td>---</td>
<td>1,774,146</td>
</tr>
</tbody>
</table>
### TABLE 8

**Distribution and Activity Characteristics of Particle Sizes**
*(Shot B, Activities at H + 300)*

<table>
<thead>
<tr>
<th>Composite Size Group (µ)</th>
<th>Number of Particles</th>
<th>Minimum Activity (well c/m)</th>
<th>Maximum Activity (well c/m)</th>
<th>Median Activity (well c/m)</th>
<th>Group</th>
</tr>
</thead>
<tbody>
<tr>
<td>11-33</td>
<td>5</td>
<td>0</td>
<td>3,222</td>
<td>372</td>
<td>4,209</td>
</tr>
<tr>
<td>34-66</td>
<td>28</td>
<td>0</td>
<td>80,483</td>
<td>1,596</td>
<td>191,972</td>
</tr>
<tr>
<td>67-99</td>
<td>49</td>
<td>0</td>
<td>47,181</td>
<td>7,103</td>
<td>519,360</td>
</tr>
<tr>
<td>100-132</td>
<td>61</td>
<td>0</td>
<td>48,757</td>
<td>15,129</td>
<td>998,547</td>
</tr>
<tr>
<td>133-165</td>
<td>78</td>
<td>4</td>
<td>53,806</td>
<td>17,243</td>
<td>1,564,034</td>
</tr>
<tr>
<td>166-198</td>
<td>46</td>
<td>0</td>
<td>387,697</td>
<td>25,877</td>
<td>1,628,637</td>
</tr>
<tr>
<td>199-231</td>
<td>19</td>
<td>19</td>
<td>99,094</td>
<td>34,435</td>
<td>693,709</td>
</tr>
<tr>
<td>232-264</td>
<td>16</td>
<td>94</td>
<td>136,203</td>
<td>49,444</td>
<td>849,701</td>
</tr>
<tr>
<td>265-297</td>
<td>10</td>
<td>8</td>
<td>122,553</td>
<td>55,708</td>
<td>599,034</td>
</tr>
<tr>
<td>298-330</td>
<td>14</td>
<td>19</td>
<td>155,625</td>
<td>55,282</td>
<td>926,556</td>
</tr>
<tr>
<td>331-363</td>
<td>1</td>
<td>---</td>
<td>---</td>
<td>64,086</td>
<td>64,086</td>
</tr>
<tr>
<td>364-396</td>
<td>2</td>
<td>3,176</td>
<td>138,856</td>
<td>71,016</td>
<td>142,032</td>
</tr>
<tr>
<td>397-429</td>
<td>0</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>430-462</td>
<td>3</td>
<td>1,267</td>
<td>39,308</td>
<td>10,997</td>
<td>51,572</td>
</tr>
<tr>
<td>463-495</td>
<td>0</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>496-528</td>
<td>2</td>
<td>92,688</td>
<td>197,740</td>
<td>145,214</td>
<td>290,428</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>334</strong></td>
<td></td>
<td></td>
<td><strong>8,523,877</strong></td>
<td></td>
</tr>
</tbody>
</table>

Although activity varied considerably with size groups and shapes one can say that activity varies as the $D^{3+}$ (dia.). Again, this confirms that little activity is on the small (0.1-5-µ) size range for Pacific surface bursts.

It is important to note that no similar surface bursts were measured at Nevada where all measurements indicated were tower and balloon detonations some of which may be classed as near-surface bursts.

**CONCLUSIONS**

In conclusion, the following chart summarizes particle sizes for fallout. It is fairly up to date and probably realistic based on some but not all of the available data. I think I can say without fear of serious opposing opinion that more research and data are needed. I do know that some data obtained have not yet been completely analyzed and published. Some data from times long past, I fear, may never be analyzed.
<table>
<thead>
<tr>
<th>Location</th>
<th>Condition of Firing</th>
<th>Description of Measurement Limits</th>
<th>Radioactivity and Particle Sizes</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nevada</td>
<td>500-700 ft towers</td>
<td>1 mile from G.Z. to H + 12 hr fallout distance</td>
<td>30% on particles less than 44 µ sol. in 0.1 N HCl, 14 to 36%</td>
<td>From &lt; 44 µ class the fraction of activity on &lt; 5 µ particle is 38 to 50%</td>
</tr>
<tr>
<td>Nevada</td>
<td>700 foot balloon</td>
<td>Same as above</td>
<td>70% on particles less than 44 µ sol. in 0.1 N HCl, &gt; 60%</td>
<td>From &lt; 44 µ class the fraction of activity on &lt; 5 µ particles is 51 to 83%</td>
</tr>
<tr>
<td>Pacific</td>
<td>Surface (coral and water)</td>
<td>Many miles from Ground zero class: Early fallout</td>
<td>Little if any activity in &lt; 5- µ rays</td>
<td>Activity varies with D &lt;(µ) of particle. (dia.) Activity on 2 shots was about 10³ d/m or less, on 25 µ particles</td>
</tr>
<tr>
<td>Stratospheric</td>
<td>All material in strat. from all sources</td>
<td>Surface air particulate measurements were taken on stratospheric dribble. Residence time has averaged from several months to a few years. Class: Delayed fallout</td>
<td>From Cyclone System Activity in 2.5 to 5µ particles are 5 to 10 times those above about 5 µ. Activity on particles &lt; 2.5 µ size was 1 to 5 times the activity on the 2.5 to 5 µ size range.</td>
<td>Activity in total samples ranged from 178 pc/100,000 ft³ in Nov. 59, to 8973 in Dec. 1958</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>From Andersen Sampler About 90% or more activity is in &lt; 5.5µ particle range. Particles less than 0.07 to 1.0 µ have from 26 to 63 % of the total activity.</td>
<td>Only 3 sample available, each sampling continuous for about 3 months in 1958 and 1959</td>
</tr>
</tbody>
</table>

**SUMMARY - PARTICLE SIZES FOR FALLOUT**
REFERENCES


5. Langer, G., Electrostatic Classification of Submicron Airborne Particles, Armour Research Foundation.


PERFORMANCE OF HIGH EFFICIENCY PARTICULATE FILTERS SUBJECTED TO STEAM-AIR MIXTURES

J. B. Hays
Savannah River Laboratory
E. I. duPont de Nemours & Co.

ABSTRACT

Five high efficiency particulate air filters from two different manufacturers were exposed to steam-air mixtures with entrained liquid moisture for periods of 10 days. Four of the filters survived the test without obvious failure and had final DOP penetrations of less than 1% at the conclusion of the 10-day exposure. One filter, however, was severely damaged as a result of this exposure.

INTRODUCTION

In major accidents accompanying the operation of water-moderated-and-cooled reactors, the release of particulate and gaseous activity would probably be accompanied by the release of steam. In a reactor partial containment system designed to reduce the spread of radioactive material by the utilization of high efficiency particulate filters, this release of steam would represent severe service requirements for the filters. The accident could result in wet steam, steam-air mixtures with entrained liquid moisture, or only high humidity air passing through the filters. The first two conditions, wet steam and steam-air mixtures with entrained liquid moisture, represent the most severe service requirements for the filters. This presentation describes the evaluation of commercially available, high efficiency filters for these conditions.

Service Requirements

The service requirements for the filters in a typical partial containment system are as follows:

1. **Rapid Steam Release.** The filters should be capable of withstanding a steam flow of 7000 cfm per filter for 10 to 30 seconds, and 2000 cfm of steam for as long as 5 minutes.

2. **Steam-Air Mixtures.** The filters should be capable of passing steam-air mixtures for several months. Figure 1 depicts a typical steam-air mixture.
Curve A shows the mass ratio of steam to air as a function of time and Curves B and C show the amount of condensate carried with the mixture for two different ventilation inlet air temperatures as a function of time. During the first few minutes, the flow is essentially all steam. The mass ratio of steam to air decreases rapidly, and after 2 hours the mixture is 87% by volume air. As depicted by Curve B, for an inlet air temperature of 500°F, the entrained condensate rate in the steam-air mixture is at least 0.6 lb per minute per filter for as long as 90 hours. This steam-air mixture was calculated assuming that the rated pressure drop across the filter does not change with time. If the pressure drop changes with time, the quantity of air mixing with the steam is reduced because of the characteristics of the exhaust ventilation system. The quantity of steam as a function of time is fixed by the reactor accident.

Bench-Scale Tests

Prior to exposing full-size filters to the above described conditions, tests were conducted on small samples of media taken from high efficiency, fire-resistant filters of three different manufacturers. These samples were exposed to a wet steam flow equivalent to 2000 cfm through a full-size filter that was rated at 1000 cfm of air at a pressure drop of 1 inch of water. Samples that were not treated for water repellency blinded very rapidly upon exposure to this steam flow; for example, the pressure drop increased to 29 inches of water within 5 minutes, and the sample ruptured. Samples of media from filters that had been treated for water repellency by Manufacturers "A" and "B" had pressure drops of only 1.8 inches of water after 5 minutes of exposure to the 2000 cfm steam flow. The water-repellent medium from Manufacturer "C" blinded very rapidly and ruptured when exposed to this steam flow. The results of these tests showed that the water repellency of the filter medium was a very important characteristic.

Additional tests on samples of water-repellent media from Manufacturers "A" and "B" showed that these media could withstand dry steam flows equivalent to 10,000 cfm through a full-size filter for 15 minutes without rupturing. The data below were typical of these media.

<table>
<thead>
<tr>
<th>Steam Flow, cfm</th>
<th>Pressure Drop, in. H₂O</th>
</tr>
</thead>
<tbody>
<tr>
<td>1,100</td>
<td>1.5</td>
</tr>
<tr>
<td>4,200</td>
<td>2.9</td>
</tr>
<tr>
<td>6,400</td>
<td>5.0</td>
</tr>
<tr>
<td>8,400</td>
<td>5.5</td>
</tr>
<tr>
<td>9,300</td>
<td>7.0</td>
</tr>
<tr>
<td>10,500</td>
<td>7.5</td>
</tr>
</tbody>
</table>
Additional tests were also run on samples of water-repellent media to determine the effects of long-term exposure to dry steam flow. These tests were conducted at a flow equivalent to 1000 cfm through a full-size filter. The results of these tests are shown below.

### TABLE II

**Effects of Long-Term Exposure to a Steam Flow of 1000 cfm**

<table>
<thead>
<tr>
<th>Samples from Manufacturer &quot;A&quot;</th>
<th>Time</th>
<th>Pressure Drop, in. H₂O</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>24 hr</td>
<td>4.0</td>
</tr>
<tr>
<td></td>
<td>54 hr</td>
<td>17.8, ruptured</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Samples from Manufacturer &quot;B&quot;</th>
<th>Time</th>
<th>Pressure Drop, in. H₂O</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>60 hr</td>
<td>7.0, did not rupture</td>
</tr>
</tbody>
</table>

As shown in this table, both media passed 1000 cfm of steam for 24 hours without rupturing. However, the sample from Manufacturer "A" ruptured after 54 hours of exposure. The pressure drop at the time of rupture was 17.8 inches of water.

The results of these tests on samples of media from commercially available high efficiency filters showed that the water repellency of the media was the controlling characteristic for a filter exposed to steam.

**Tests of Full-Size Filters**

The filters used in this test program were the 1000-cfm, fire-resistant model supplied by Manufacturers "A" and "B" that utilized aluminum separators and cadmium-plated steel frames. Each filter was rated for a nominal flow of 1000 cfm at a pressure drop of 1 inch of water. In the tests of full-size filters, the filters were exposed to the previously described service requirements; for example, a steam flow of 7000 cfm for 30 seconds, 2000 cfm for 5 minutes then steam-air mixtures for 10 days. As previously mentioned the steam flow as a function of time is dictated by the reactor accident, and the quantity of air in the steam-air mixture depends upon the pressure drop across the filter and the characteristics of the exhaust system. The steam-air mixture properties shown in Figure 1 represent the case where the pressure drop across the filter does not increase with time. In the tests of full-size filters, the pressure drop across the filter increased with time, and the air flow was throttled to maintain a pressure drop across the filter of 4 to 6 inches of water. Therefore, the mass ratios of steam to air obtained during the tests were higher than those shown in Figure 1. The mass ratios of steam to air obtained during the tests are shown in Figure 2. The calculated ratios for the zero blinded case are shown in Figure 2, Curve C.
PROPERTIES OF TYPICAL STEAM-AIR MIXTURE

Figure 1

Steam - initially saturated
Air - initially dry

Initial Air Temperature

90°F

50°F

90°F

Lb water (vapor & liquid) per lb air

Lb liquid (min/ft²) (filter)

Time, hr
MASS RATIOS OF STEAM TO AIR OBTAINED DURING TESTS OF FULL-SIZE FILTERS

Figure 2

[Graph showing mass ratios of steam to air for Mfg. "A", Mfg. "B", and Curve C (Calculated).]
The discontinuity in the pass ratio curve for the filters from Manufacturer "A" represents the period when the filter was blinded by liquid to such an extent that there was no air flow. As shown in this figure, the filter of Manufacturer "A" blinded more rapidly than that of Manufacturer "B" and, therefore, the resultant mass ratio of steam to air for the filters from Manufacturer "A" was always greater than that for Manufacturer "B".

As mentioned above, the air flow was adjusted to maintain a fixed pressure drop across the filters during the tests. If in a particular containment system the maximum pressure drop across the particulate filters were limited to 4 inches of water, then the volumetric flows as a function of time for the filters from Manufacturer "A" and "B" would be as shown in Figure 3. Up to an exposure time of 1 hour, the filters from both manufacturers blinded at about the same rate. After 1 hour, the difference in blinding rate between the two manufacturers' filters becomes significant. The flow through the filters from Manufacturer "A" dropped below 1000 cfm between 1.3 and 2.5 hours after the start of the test. The flow through the filters from Manufacturer "B" did not decrease below 1000 cfm until 3.0 to 3.7 hours after the start of the tests. The difference between the filters was even more significant after about one day of exposure to the steam-air mixtures; for example, at this time, the filters from Manufacturer "A" passed only 130 cfm at a 4-inch pressure drop, while the filters from Manufacturer "B" passed 630-770 cfm at the same pressure drop. After about 9 days, the filters from both manufacturers were almost completely blinded; for example, the flows through the filters from manufacturer "A" and "B" were 80 and 130 cfm, respectively.

To date, five filters have been exposed to the above described tests. The following table shows the extent of damage incurred by the filters as a result of exposure to these test conditions.

-554-
TABLE III

Condition of Filters after Steam-Air Mixture Tests

<table>
<thead>
<tr>
<th>Filters from Manufacturer &quot;A&quot;</th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>No. 1</td>
<td>No. 2</td>
<td>No. 3</td>
</tr>
<tr>
<td>Duration of test</td>
<td>10 days</td>
<td>10 days</td>
<td>10 days</td>
</tr>
<tr>
<td>Observed condition of filter</td>
<td>One visible pin-hole in fold on downstream side</td>
<td>Hairline crack in folds on downstream side</td>
<td>Eight large, horizontal tears on downstream side</td>
</tr>
<tr>
<td>% DOP penetration</td>
<td>0.37</td>
<td>0.60</td>
<td>No test greater than 1 %</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Filters from Manufacturer &quot;B&quot;</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>No. 1</td>
<td>No. 2</td>
</tr>
<tr>
<td>Duration of test</td>
<td>5 days</td>
<td>10 days</td>
</tr>
<tr>
<td>Observed condition of filter</td>
<td>No visible damage</td>
<td>No visible damage</td>
</tr>
<tr>
<td>% DOP penetration</td>
<td>0.06</td>
<td></td>
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</table>

SUMMARY

The filters from Manufacturer "B" appear to offer the greatest potential as candidates for use in containment systems in which predicted service requirements include exposure to steam-air mixtures. As shown in Figure 3, significant differences in the rate of blinding by liquid water were observed among supposedly identical filters from both manufacturers.

In addition, the filters from both manufacturers were deficient in that the total volumetric flow through the filters was only about 0.1 of the normal rated flow at a pressure drop of 4 inches of water after 10 days of exposure to the steam-air mixtures. Until the factors affecting the blinding of water-repellent filters by liquid water are better understood, specifications or acceptance tests cannot be prepared to insure the user that water-repellent filters will not completely blind and thus rupture.
VOLUMETRIC FLOW THROUGH FULL-SIZE FILTERS
NORMALIZED FOR 4-INCH WATER PRESSURE DROP

Figure 3

Volumetric Flow at 4" w.g., cfm

Elapsed Time, hr
WASTE PROCESSING OFF-GAS SCRUBBER STUDIES

Y. Takashima, W. T. Kyritsis, R. Dennis, and L. Silverman
Harvard School of Public Health

ABSTRACT

Volume reduction of highly radioactive aqueous aluminum nitrate waste by evaporation and calcination to stable solid aluminum oxide in pot calciners can produce gas-borne particulates. A simple containment scrubber concept is introduced utilizing a commercial liquid-jet fume scrubber at the inlet of a closed vessel for condensation of vapor and removal of particulates by continued water droplet impingement. This paper presents results for the jet-scrubber unit on open cycle (once-through) removal of fly ash and iron oxide aerosols, nitric acid, gas and mist, iodine vapor, and oxides of nitrogen and sulfur gases. Preliminary results are also given for the closed system pot calciner-water jet containment scrubber concept during reduction of simulated aluminum nitrate waste solution to calcined aluminum oxide.

INTRODUCTION

Proposed use of nuclear power requires an increasing study of the parallel problem of gaseous and liquid waste disposal. Aluminum, zirconium, and other nitrate solutions containing highly radioactive fission products and diluent materials arising from the separation of uranium fuel from alloying or cladding materials, must be placed in a safe non-dispersive form. Many proposed methods for liquid waste volume reduction and fixation of radioactive fission products and inert diluents in stable solid media, for subsequent burial, are described in AEC reports (1) (2). Calcination has been studied with fluidized beds, spray dryers, rotary kilns, and in batch pots. Each of these processes, we believe, presents some airborne particulate contamination problems. In reviewing the various processes for fixing radioactivity in solids, it is evident that even under the most favorable conditions a certain fraction of the waste escapes fixation. The residual material remaining in the gaseous phase requires special treatment by appropriate air and gas cleaning techniques before discharge to the atmosphere.

The four methods of waste calcination may be classified, in terms of their gas cleaning requirements, as high or low gas volume systems (based upon the ratio of gas flow per gallon of waste processed).

For gas or air cleaning, minimum particulate or gaseous concentrations are desirable. It is possible that the gas cleaning system may be a decisive
factor in the selection of a waste treatment process. However, at present our considerations do not exclude any of the calcination techniques mentioned above. If the solid waste resulting from the calcination process meets the required specifications in regard to physical characteristics, a serious attempt should be made to select appropriate gas cleaning. The only exclusion to be considered is the case where cost for adequate cleaning renders the entire operation economically impractical.

With respect to the Phillips, ICPP calciner (1), relatively high gas volumes and dust loadings are inherent with the fluidizing process. Selection of air as the fluidizing medium was considered the best choice for their system since steam would lead to nitric acid formation. Carbon monoxide and other inert mixtures were also proposed but eliminated on a basis of cost. Although venting of nitric acid gases or oxides of nitrogen to the atmosphere would be undesirable in many locations, it has been considered geographically acceptable at Idaho.

According to Wheeler (1), high efficiency particulate removal was attained with a small Venturi scrubber used in pilot studies. Based upon his calculations, exit solid loadings from the absolute type filters in the range of 0.00005 grains per cu. ft. would represent satisfactory decontamination for strontium-90. It should be emphasized that an absolute type filter effluent should be 100 to 1000 times lower than his reported values provided that the filter is not defective. Actual solid concentrations on a number basis might be several million particles per cu. ft. with a 0.00005 grain per cu. ft. loading according to Hatch (1). If radioactivity were preferentially distributed upon the smaller particles decontamination estimates, based upon overall weight collection efficiency, might not be reliable.

A common feature of the low gas flow calcination methods is the condensation and recovery of nitric acid. Based solely upon small laboratory and pilot installations where acid recovery is considered an integral part of the over-all calcination process (pot calcination, rotary kiln or spraying drying) it appears that high particulate removal accompanies the nitric acid condensation recovery and fractionation process. Perona (2) indicated that the vapor leaving the ORNL nitric acid recovery column was reasonably particulate free.

The fact that a membrane filter used by Hatch (1) as a sampler on the final off-gas for the BNL rotary kiln, showed negligible plugging suggested that very low particulate concentrations existed in the gases discharging from their fractionating and condensing units. In order to minimize gas volumes, steam was used as the carrier gas and joints and bearing sleeves were specially constructed. Because of steam and absence of non-condensable gases, it would appear that cleaning can be more effective and perhaps less expensive. However, in scaling to the size of the Phillips calciner, the low volume systems may present problems not apparent at this time.
In our opinion the merits of the calcination processes must be defined on the basis of type of solid waste produced and the over-all costs of the treatment process. Data so far indicate that no one waste concentration technique can have universal application. Simple pot calcination appears to offer significant advantages in first cost of equipment and in ease with which the evolved gases and particulates may be contained and removed.

The combined calcining-scrubber process is illustrated in Figure 1 which shows the schematic arrangement of our test apparatus. A simple pot calciner using self-sintering decay heat from the nuclear residues, and probably with added external heat, evaporates and concentrates a solution of aluminum nitrate waste. During heating, first steam evolves followed by oxides of nitrogen. These gases pass to a liquid jet scrubber mounted on a closed vessel where vapor condensation takes place. Most particulates from the calciner will be impacted by the jet. The jet scrubber need not achieve high particulate collection efficiency on the first contact since repeated contact of spray droplets and dust particles will occur in the "non-flow" system, eventually washing out all the particles. Non-condensable gases will be retained in a simple gas holder, since it is unlikely that their volume will be large. They can be vented later, after monitoring through a low-volume gas recovery system (e.g. adsorption bed).

**EXPERIMENTAL PROCEDURE**

**General**

A Schutte-Koerting fume scrubber (SK No. 4010-3 inch) was selected for study, since it combines the functions of primary gas mover, steam and vapor condenser, and gas cleaner. Since the manufacturer's information and the technical literature did not contain quantitative data on the performance of the scrubber for fine particulates, vapors, or gases, a preliminary study was undertaken to evaluate its open-circuit performance. It was anticipated that it might not differ radically from other comparable wet scrubbing devices (with the same collecting forces being applied).

This study included tests of air-moving efficiency of the device, and particle, vapor and gas removal efficiency. Fly ash, iron oxide fume; nitric acid vapor, iodine vapor; and nitrogen dioxide and sulfur dioxide were selected as typical contaminants. Tests were conducted at several liquid and air flow rates, with water and sodium hydroxide solutions as scrubbing liquids. Preliminary studies were also made on the calciner-containment scrubber system with a simulated aluminum nitrate waste, to determine the relative quantities of off-gas, (oxides of nitrogen, i.e., NO, NO₂, N₂O₄), and particulate carry over.
FIG. 1 - CALCINATION FLOW DIAGRAM
Open-Cycle Tests

Equipment Arrangement

The equipment arrangement for open-cycle tests is shown schematically in Figure 2. Fly ash aerosols entered a 30 gallon retention tank which permitted (a) separation of coarse particles and (b) simulated the mode of entry of particles evolving from evaporation or calcining processes. A 3 inch Schutte-Koerting (SK No. 4010-3) fume scrubber mounted on the top of a 55 gallon drum aspirated the test gas stream from the retention drum. Major particle and vapor removal took place within the jet droplet stream. Spray liquids (water or caustic solution) were recycled by an external pumping system which permitted a broad range of nozzle pressures (up to 100 psig).

The 55 gallon droplet separation drum provided the reservoir capacity for the spray liquid. During most tests the liquid level was approximately 21 in. from the top of the drum. Droplet carry over was reduced by means of a dividing baffle extending from the top of the drum to within 3 inches of the liquid surface. The effluent gas passed through a mist eliminator section containing Metex screening to minimize moisture problems during downstream aerosol sampling.

Provisions were made to collect aerosol samples up- and downstream of the scrubber. A Stairmand disk (3) served as a flow metering and gas mixing device in the inlet pipe. Gas and liquid temperature and static pressures were determined throughout the test circuit. Following completion of fly ash tests, the 30 gallon elution drum was removed. Remaining test aerosols or gases were introduced directly to the ejector through a straight run, 5 feet long, of 3 inch diameter pipe.

Test Aerosols and Gases

Table 1 summarizes the methods of aerosol generation, sampling, and analysis used in this study. All particulate samples were collected isokinetically for representative operating periods with Whatman thimbles or 1106 B all-glass paper. Membrane filters were used for particle size studies. Passage of the 16 u MMD fly ash aerosol through the 30 gallon drum reduced its mean size by count and weight to 0.48 and 5.5 micron, respectively. These latter values describe the particulate entering the ejector.

Test Results

Ejector Air Moving Efficiency

Gas moving efficiency of ejectors can be calculated from the ratio of the pressure and flow produced in the aspirated fluid to the energy available in the motive fluid. A series of static pressure-volume tests were made with
FIG. 2 - SCHEMATIC - TEST APPARATUS, SCHUTTE + KOERTING EJECTOR
<table>
<thead>
<tr>
<th>Test Aerosol or Gas</th>
<th>Inlet Concentration</th>
<th>Methods of Generation</th>
<th>Method of Sampling</th>
<th>Method of Analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fly Ash</td>
<td>0.1 - 0.5 Grain/ft³</td>
<td>NBS (11)</td>
<td>Up: Whatman Thimble (12)</td>
<td>Weight (12)</td>
</tr>
<tr>
<td></td>
<td>0.48 µ</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>5.5 µ</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Iron Oxide Fume</td>
<td>0.03 - 0.1 Grain/ft³</td>
<td>Combustion of Fe(CO)₅ (11)</td>
<td>1106-B paper circle (12)</td>
<td>Colorimetric (16)</td>
</tr>
<tr>
<td></td>
<td>0.027 µ</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.15 µ</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nitric Acid Vapor</td>
<td>10-1850 ppm</td>
<td>Heat 70% Acid</td>
<td>Sintered Glass Absorber with water (14)</td>
<td>Conductivity (15)</td>
</tr>
<tr>
<td>Iodine Vapor</td>
<td>8-90 mg/m³</td>
<td>Sublimation</td>
<td>Sintered Glass Absorber with 5% KI solution (8)</td>
<td>Colorimetric (8)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Crystal of ACS grade (8)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nitrogen Dioxide Gas</td>
<td>3-10 ppm</td>
<td>Commercial Cylinder gas</td>
<td>Sintered Glass Absorber with modified Griess-Ilosvay reaction (10)</td>
<td>Colorimetric (10)</td>
</tr>
<tr>
<td>Sulfur Dioxide Gas</td>
<td>5000 ppm</td>
<td>Commercial Cylinder gas</td>
<td>100 ml syringe grab sample</td>
<td>Beckman GC-2 Gas Chromatograph Benzy Ether Column</td>
</tr>
</tbody>
</table>
the 3 inch fume scrubber to evaluate its air-flow pressure characteristics, Figure 3. Manufacturer's reported performance for a 4 inch unit are also indicated. Our test data, Figure 3, were obtained with a 9 inch long (3 inch diameter) extension pipe attached to the exit of the Venturi section. The extension pipe, illustrated in dotted outline in Figure 2, was found to increase ejector efficiency. Ejector efficiency as a function of air of liquid volume flow rate, \( \frac{Q_A}{Q_W} \) (designated by \( R_f \) in this report) is indicated in Figure 4. Maximum efficiency occurs at about the same air-water rate ratio (\( R_f = 55 \)) for jet pressures investigated over the range 30 to 80 psig.

Generally, efficiencies of the unit were low, 9 per cent at best. Tests with a 30 inch extension (shown in solid outline in Figure 2) indicated a maximum efficiency of 13 per cent at 100 psig, and an \( R_f \) value of 75. Equivalent efficiency at 100 psig with no extension was 8.5 per cent. In general, efficiencies ranged from 5 to 13 per cent for \( R_f \) values of 50 to 75, and water pressures of 30 to 100 psig. By throttling the inlet gas stream to maintain a constant 1 inch water resistance, air flows ranged from to 10 to 60 cubic feet per minute for jet pressures of 30 to 80 psig, respectively.

**Fly Ash**

Weight collection efficiencies for a fly ash aerosol (mean size by count 0.48 \( \mu \), mean size by weight 5.5 \( \mu \) immediately upstream of the inlet to the jet scrubber) are shown in Table 2 for variations in air and water flow rates of 22 to 55 cubic feet per minute and 2.23 to 3.8 gallons per minute, respectively. Based upon previous studies (4) the range of fly ash loadings used, 0.1 to 0.5 grains per cubic foot, was too small to show any concentration-efficiency effects.

The range of efficiencies for fly ash, 90 to 99 per cent, was about the same as that reported in past tests by this laboratory for other coarse spray (500 micron droplet range) wet scrubbing devices (4) (5). The water rates required to attain these efficiencies (70 to 150 gallons per 1,000 cubic feet of gas) were considerably higher, however, than those customarily used, 5 to 15 gal./1000 cu. ft. per min.

Analysis of test results in terms of expected performance showed qualitative agreement with theory. The fractional efficiency of a wet scrubbing unit for a specified particle size may be expressed by the general equation

\[
E = 1 - e^{-\frac{3Q_w \bar{U} \gamma t}{2Q_a DT}}
\]

where \( Q_w \) and \( Q_a \) = volume flow rates for liquid and gas, respectively, \( \bar{U} \) = relative velocity between dust particles and water droplets, \( DT \) = liquid
FIG. 3-AIR FLOW-STATIC PRESSURE CHARACTERISTICS FOR 3" AND 4" SK JET SCRUBBER (4010)
FIG. 4-AIR PUMPING EFFICIENCY OF SCRUBBER WITH 3" DIAMETER, 9.5" LONG EXTENSION

- $P_n = 31.0$ PSIG - 2.65 GPM
- $P_n = 41.1$ PSIG - 3.03 GPM
- $P_n = 51.1$ PSIG - 3.28 GPM
- $P_n = 71.1$ PSIG - 3.78 GPM
- $P_n = 83.1$ PSIG - 4.10 GPM

Graph showing the relationship between volume of air and percent efficiency.
<table>
<thead>
<tr>
<th>Test Aerosol of Gas</th>
<th>Liquid Flow Rate Gal/min</th>
<th>Air Flow CFM</th>
<th>Inlet Conc Grains/cu ft</th>
<th>Collection Efficiency Weight Per Cent</th>
<th>Remarks</th>
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<tr>
<td>Fly Ash</td>
<td>3.2</td>
<td>22</td>
<td>0.14</td>
<td>99.1</td>
<td>5.5 u</td>
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<tr>
<td></td>
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<td>98.4</td>
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<td>81.7</td>
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<td>28</td>
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<td>0.03</td>
<td>19</td>
<td>Size By</td>
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<td>88</td>
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<td>31</td>
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<td>Iodine Vapor</td>
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<td>40</td>
<td>mg/cu. m</td>
<td>99.9 + 8%</td>
<td>NaOH Scrubbing</td>
</tr>
<tr>
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<td>3.2</td>
<td>20</td>
<td>26</td>
<td>99.9 + 8%</td>
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<tr>
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<td>99.9 + 8%</td>
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<td>6</td>
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<td>6</td>
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<td>Sulfur Dioxide</td>
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<td>5000</td>
<td>70</td>
<td>Original Ejector</td>
</tr>
<tr>
<td></td>
<td>3.5</td>
<td>20</td>
<td>5000</td>
<td>70</td>
<td>Water</td>
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<td>61</td>
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<td>Blank</td>
<td>40</td>
<td>5000</td>
<td>52</td>
<td></td>
</tr>
</tbody>
</table>

a Grains per cubic foot for dust and fume, parts per million by volume for nitric acid vapor and gases, mg/m³ for iodine vapor

NOTE: System Tygon coated for all gas and vapor tests.
droplet diameter, \( t \) = retention time of gas in spray zone and \( \eta \) = target efficiency of spray droplet for a specified particle size.

For dust particles less than 10\( \mu \) in diameter, particle velocity, \( U_p \) is essentially the same as that of the gas stream, \( U_a \). The target efficiency, \( \eta \), may be defined by the relation (6)

\[
\eta = f \frac{P_p D_p^2 \bar{U}}{18 \mu D_T}
\]

where \( P_p \) = particle density, \( D_p \) = particle diameter, \( \mu \) = gas viscosity, and \( \bar{U} \) and \( D_T \) are the same as in Equation 1.

An attempt was made to apply Equation 1 to the jet scrubber system by making the simplifying assumptions that (a) all particle removal took place within the Venturi section and (b) that the Venturi expander was cylindrical rather than tapered.

Agreement between observed and predicted efficiencies was poor apparently due to the problems of defining (a) the true droplet size and the quantity of water lost as "effective" spray by impingement on the Venturi walls.

However, comparison of experimental data at constant air flow rates, (20, 40, and 60 cubic feet per minute) showed a consistent increase in efficiency when water rates were increased.

Corresponding collection efficiencies for the range of 2.23 to 3.8 gallons per minute liquid rate were 93 to \( \geq \) 99 per cent, 91.7 to 98.7 per cent, and 91 to 98.3 per cent.

A similar comparison at constant water rates showed consistent decrease in efficiency with increasing air flow rates of 20 to 60 cubic feet per minute, i.e. 93 to 91 per cent at 2.23 gallons per minute, 99 to 97.5 per cent at 3.25 gallons per minute and 99 to 98.3 per cent at 3.8 gallons per minute.

It is pointed out that target efficiencies, \( \eta \), calculated by Equation 2 showed that significant contacting between dust particles (1 to 5 micron diameter) and spray droplets were attainable, i.e. \( \eta \) values of 0.2 to 0.9. These data were obtained by assuming (a) that the relative velocity, \( \bar{U} \), between spray droplets and dust particles was defined by droplet terminal velocity, (b) fly ash density was 2.0 g per cc, and the average droplet diameter was 500 microns.

Substitution of the above \( \eta \) values in Equation 1 indicate theoretical collection efficiencies in the order of 50-90 per cent rather than the observed levels of 90 to 99 per cent, depending upon the assumptions made with regard
to droplet size and the effective retention time, \( t \).

**Iron Oxide Fume**

Iron oxide fume collection ranged from 15 to 30 per cent for inlet concentrations of 0.03 to 0.1 grain per cubic foot, at water pressures of 20 to 80 psig (2-4 gpm) and air flow rates of 20 to 55 cfm. No distinct correlations between water and air flow rates were indicated in contrast to results of fly ash tests. On the basis of the size characteristics of the iron oxide fume (mean size by count 0.03 micron, mean size by weight 0.15 micron) very little collection would be predicted by inertial or impaction mechanisms. According to Equation 2 target efficiencies would appear to be in the order of \( 10^{-3} \) to \( 10^{-4} \). Estimation of target efficiency by diffusion and interception \( \eta_d \) by Chen (7),

\[
\eta_d = f \left( \frac{D_{BM}}{\tilde{U} D_T} \right)
\]

where \( D_{BM} \) = particle diffusivity and \( \tilde{U} \) and \( D_T \) are as defined previously, again showed low \( \eta_d \) values, \(<10^{-2}\). It was concluded therefore that major iron oxide removal probably occurred within the 55 gallon drum where fine spray droplets, 10 to 50 microns diameter, entrained in the low velocity gas stream would favor diffusional capture.

Previous tests (5) also showed similar iron oxide efficiencies which were higher than predicted by theory. The complexity of the aerodynamic system does not permit further analysis of the present data.

**Nitric Acid Vapor**

Nitric acid vapor collection by the SK jet scrubber showed a marked dependence on the inlet gas concentration over the concentration range of 6 to 2000 parts per million by volume (Figure 5 and Table 2). Results of preliminary tests with the interior steel surfaces uncoated, Figure 5 - Curve 1, indicated that collection efficiency decreased from 95 to 75 per cent over the concentration range 7 to 200 parts per million. Results were essentially the same for both water and sodium hydroxide sprays. Dry runs with the above system (no liquid sprays and with the same liquid level in the 55 gallon drum) showed considerable HNO₃ removal by chemical reaction with the iron surfaces, i.e. up to 66 per cent at low inlet concentration, 10 parts per million HNO₃. Consequently, all interior surfaces were coated with Tygon paint which reduced dry or "blank" efficiency to less than 8 per cent. Tests with the coated system, Figure 5 - Curve 2, followed the same pattern shown in Curve 1, i.e., a decrease in efficiency from 93 to 67 per cent over the inlet concentration range, 10 to 300 parts per million. However, when HNO₃ concentration was increased from 300 to 2000 parts per million
FIG. 5—NITRIC ACID COLLECTION BY SK JET SCRUBBER
LIQUID TEMPERATURE = 21°C
the efficiency rose steadily to an observed value of about 97 per cent at 2000 parts per million. It should be noted that Curves 1 and 2 can be compared only with respect to trends. Interior surfaces differed and the air flow rate was higher for data reported in Curve 2 (40 vs. 20 cubic feet per minute).

The concentration-efficiency relation was explained by the fact that HNO₃ remains in the vapor state in low concentration so that its collection is controlled by diffusion through the gas film surrounding the spray droplets. Lowered liquid temperature which retards release of water vapor to the liquid film favors increased diffusional capture. According to our tests with the NaOH scrubbing system, collection efficiency for HNO₃ in the concentration range, 60 to 150 parts per minute was 19, 43, and 87 per cent when liquid temperatures were 38, 21, and 11°C.

Elevated liquid temperature and decreased air temperature lead to nitric acid fog formation which is further enhanced by increased HNO₃ concentration. It was expected that the growth of the fog droplets from molecular size to approximately 0.1 to 0.2 micron diameter would produce an aerosol progressively more difficult to capture by diffusion and too small to be collected by impaction. It is postulated that the fog droplet size at the minimum point on Curve 2 was in the order of 0.1 to 0.2 micron. By increasing the vapor concentration, however, significant growth occurs so that droplets are capturable by inertial mechanisms.

Ordinarily, a caustic solution would be used for acid vapor scrubbing since the neutralization process would reduce gas back-pressure at the interface. However, in the concentration ranges employed during our studies, test duration was not sufficiently long to saturate the water phase (where reversibility would be expected at high concentrations). Similar performance data for both water and caustic scrubbing solutions appeared to confirm this fact.

Iodine Vapor

Collection of iodine vapor, sublimed from C. P. crystalline iodine in air concentrations ranging from 8 to 90 mg. per m³, is shown in Table 2. Results of tests with an 8 per cent NaOH spray (3.25 gallons per minute) showed greater than 99 per cent efficiency for all air flow rates up to 60 cubic feet per minute. Reduction of spray rate to 2.23 gallons per minute indicated a gradual decrease in iodine removal as the air flow rate increased, i.e., 93, 86, and 80 per cent at 20, 40, and 60 cubic feet per minute. Substitution of water as the spray liquid at a constant flow rate of 3.25 gallons per minute showed the same efficiencies as reported for caustic at 2.23 gallons per minute.

Blank runs without liquid spray indicated approximately 30 per cent iodine removal by the dry system alone. Previous evaluation of iodine collectors by this laboratory (8) has shown that the molecular or vapor form prevails at
concentration levels >1 mg/M^3. Therefore, the observed reduction in iodine collection efficiency with reduced caustic spray rate, 3.25 to 2.23 gallons per minute was attributed to lack of spray contacting surface and not to adsorption of iodine on sub-micron dust particles.

In the case of caustic absorption, chemical fixing of iodine reduces the partial pressure of iodine vapor at the droplet surface to negligible levels. Limited solubility of iodine in water, however, leads to a significant back pressure which reduces the over-all transfer of iodine molecules through the gas film.

It is emphasized that the high iodine efficiencies reported here apply only to the indicated concentration levels. Past experience has shown that large percentages of iodine may be bound through chemical reaction or physical adsorption to sub-micron particles when concentrations are in the 10^-6 to 10^-2 mg. per m^3 range (8). In the latter cases, effective iodine removal depends upon capture of the iodine-contaminated particulate material.

**Nitrogen Dioxide**

Nitrogen dioxide introduced in concentrations ranging from 1 to 10 ppm was collected at very low efficiencies, 1 to 3 per cent, for water rates of 2.23 to 4.0 gal. per min. Parallel tests with NaOH scrubbing solutions were discontinued due to sampling problems. The above data confirmed Peters experiments (9) with NO_2 - water scrubbing systems which showed that NO_2 removal was controlled mainly by the reaction of NO_2 and water. Since this is a slow reaction at low concentrations, poor results would be expected unless extended gas-droplet contact time were possible.

**Sulfur Dioxide**

Sulfur dioxide absorption is now under investigation using gas chromatography for gas analyses. Preliminary tests, Table 2, indicated a trend toward decreasing collection efficiency when water pressure (and water rate) was increased while air flow remained constant. SO_2 absorption efficiency, for inlet concentrations of 5000 ppm ranged from 70 to 52 per cent for water pressures of 40 to 80 psig respectively. Ordinarily, the reverse would be expected since higher water rates imples greater droplet contact surface.

**CALCINER TESTS - EFFLUENT EVALUATION**

Tests described in the following section were conducted to determine basic operating parameters for the combined calcining-scrubber system shown schematically in Figure 1. The evaporation and calcining vessel was not designed to optimize the liquid waste treatment process with regard to heat transfer or quality of the final product. Its sole function was to produce an effluent which, in composition and concentration, would simulate that
evolving from a pot calcination process.

In contrast to open system tests reported previously, which demonstrated the SK jet scrubber performance with minimal gas treatment times, the equipment arrangements considered for calciner effluent treatment afforded lengthy contact time between the calciner effluent and the spray liquid. Two methods of gas treatment were investigated, a recycle system and a closed system.

Recycle or Loop System

The recycle system apparatus was essentially the same as that shown in Figure 1 except that the scrubber effluent was recycled to the calciner vessel. Consequently, gas flow rates were similar to those reported previously for open circuit tests. Although low efficiencies were excepted for any one transit of the gas through the scrubber, multiple exposures through recycling were expected to produce high level decontamination.

At the completion of calcination, all volatile decomposition products would have been transferred to the scrubber liquid, while residual non-condensable gases above the calcined product would consist of saturated air and trace rare gas isotopes.

Our preliminary tests indicated that water carry over, due to droplet entrainment and condensation led to sampling and reheat problems. Although in practice, gas temperature in the effluent return line to the scrubber could be readily kept above the dew point, several modifications would have been required for accurate effluent sampling. Therefore, the recycle phase of this study was temporarily discontinued and the apparatus reconstructed for closed systems tests.

Closed System

Operating Procedure

The closed system described in Figure 1, involves only a transfer of water and gaseous, liquid, and solid decomposition products from the calciner vessel to the scrubbing tank. Expansion of air or other non-condensable gas within the system is compensated for by the gas pressure balancing tank which, by proper counter weighing, maintains a slight negative pressure on the discharge side of the ejector. During the evaporation phase of the process, the gas entering the scrubber inlet consists of an air-steam mixture with progressive steam enrichment occurring as the water evaporation phase nears completion. Gas flow at approximately 100-125°C averages about 5 to 10 cu. ft. per min. during this period. Depending upon the composition of the waste as to volatile acid content, the effluent may contain several soluble acid gases prior to any decomposition of metal salts from fuel
element claddings. Following water evaporation, calciner temperature then rises to pre-controlled levels during which period nitrate salts, i.e., \( \text{Al(NO}_3\text{)}_3 \) are decomposed to form the oxide, i.e., \( \text{Al}_2\text{O}_3 \) and nitric acid gases. Calcining operations are discontinued when analyses of the scrubber gas phase show no oxides of nitrogen and total nitrogen content of scrub liquor shows no further increase.

During Tests 1 - 6 described in Tables 3 and 4, the simulated waste consisted of an 11.4 per cent by weight water solution of \( \text{Al(NO}_3\text{)}_3 \) (1.0 kg. \( \text{Al(NO}_3\text{)}_3 \cdot 9\text{H}_2\text{O} \) added to 4.0 liters of water). In Test 7, 110 grams of sodium hydroxide were added to the above solution. The scrubbing solution in all runs was a 0.5 per cent NaOH solution.

Scrub liquid rates were maintained constant throughout all tests at 3.2 gal. per min. Spray temperature was also maintained constant at approximately 27°C by varying the water rate in the cooling coil located in the 55 gal. drum.

The major variable during Tests 1 - 6 was the calcination temperature, 230-570°C, determined by placing a thermocouple 0.5 inch from the bottom of the calcining vessel. The couple was at all times immersed either in the solution or in the dried cake. A second variable was the variation in calcining time, 60-125 min.

**Sampling and Analysis**

Sodium hydroxide, 0.5N, was used in the fritted bubblers for gas sampling. Analyses for nitric acid and nitrogen dioxide in the effluent samples were performed (a) by titrating one portion of the bubbler sample with nitric acid to determine the nitrate present and (b) by treating the second portion with a modified Griess-Ilosvay reagent (10) to determine the nitrite. The combined procedures indicated total nitrogen, but did not distinguish between the nitric oxide, \( \text{NO} \), and the nitrogen dioxide, \( \text{NO}_2 \). Scrubbing liquid analyses were performed by the same methods with one modification, i.e., the nitrite results were checked by titrating a separate portion of the scrubbing liquid with potassium permanganate.

**Effect of Temperature**

The water evaporation phase of all tests required approximately 40 min. during which time calciner temperatures rose from 100-130°C. At the end of this period calciner temperature rose rapidly to equilibrium levels and was maintained as constant as possible until completion of the test. Calcining periods shown in Table 3 were varied from 60 to 120 minutes. The effectiveness of the calcining process was evaluated in terms of (a) the percent conversion of the waste to aluminum oxide and (b) quantity of nitrate retained in the scrubbing liquid.
### TABLE 3

GAS ABSORPTION FOR CLOSED CALCINER SCRUBBING SYSTEM

<table>
<thead>
<tr>
<th>Test No.</th>
<th>Evaporation Time</th>
<th>Calcination Time</th>
<th>Temp</th>
<th>Distribution of Evolved Gases</th>
<th>55 gal drum NO$_3^-$ NO$_2^-$ Total</th>
<th>1st Bubbler NO$_3^-$ NO$_2^-$ Total</th>
<th>2nd Bubbler NO$_3^-$ NO$_2^-$ Total</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Min</td>
<td>Min</td>
<td>°C</td>
<td>Mols</td>
<td>Mols x 10$^2$</td>
<td>Mols x 10$^3$</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>20</td>
<td>90</td>
<td>230</td>
<td>3.32</td>
<td>2.74</td>
<td>6.06</td>
<td>0.0254</td>
</tr>
<tr>
<td>2</td>
<td>23</td>
<td>60</td>
<td>415</td>
<td>4.24</td>
<td>3.47</td>
<td>7.71</td>
<td>5.64</td>
</tr>
<tr>
<td>3</td>
<td>67</td>
<td>60</td>
<td>525</td>
<td>4.40</td>
<td>3.04</td>
<td>7.44</td>
<td>2.87</td>
</tr>
<tr>
<td>4</td>
<td>30</td>
<td>125</td>
<td>400</td>
<td>4.54</td>
<td>2.89</td>
<td>7.43</td>
<td>2.4</td>
</tr>
<tr>
<td>5</td>
<td>40</td>
<td>100</td>
<td>500</td>
<td>5.18</td>
<td>2.67</td>
<td>7.85</td>
<td>2.28</td>
</tr>
<tr>
<td>6</td>
<td>35</td>
<td>120</td>
<td>570</td>
<td>4.95</td>
<td>3.23</td>
<td>8.18</td>
<td>2.60</td>
</tr>
<tr>
<td>7*</td>
<td>40</td>
<td>110</td>
<td>550</td>
<td>4.17</td>
<td>2.25</td>
<td>6.42</td>
<td>1.77</td>
</tr>
</tbody>
</table>

* 110 grams sodium hydroxide were added to Test 7.

NOTE: Simulated waste - 1.0 Kg Al(NO$_3^-$)$_3$ · 9H$_2$O in 4 l. water - nitrate molarity = 8.00
TABLE 4
COMPOSITION OF CALCINED ALUMINUM NITRATE WASTE

Weight Per Cent

<table>
<thead>
<tr>
<th>Test No.</th>
<th>Calcination Temp °C</th>
<th>Al_2O_3</th>
<th>Al(OH)_3</th>
<th>H_2O</th>
<th>Al(NO_3)_3</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>230</td>
<td>49.2</td>
<td>9.2</td>
<td>13.4</td>
<td>28.2</td>
</tr>
<tr>
<td>2</td>
<td>415</td>
<td>84.4</td>
<td>7.0</td>
<td>7.6</td>
<td>1.0</td>
</tr>
<tr>
<td>3</td>
<td>525</td>
<td>87.0</td>
<td>6.0</td>
<td>6.0</td>
<td>1.0</td>
</tr>
<tr>
<td>4</td>
<td>400</td>
<td>91.1</td>
<td>3.7</td>
<td>5.0</td>
<td>0.2</td>
</tr>
<tr>
<td>5</td>
<td>500</td>
<td>90.0</td>
<td>5.1</td>
<td>4.4</td>
<td>0.5</td>
</tr>
<tr>
<td>6</td>
<td>570</td>
<td>91.5</td>
<td>4.6</td>
<td>3.5</td>
<td>0.4</td>
</tr>
<tr>
<td>7*</td>
<td>550</td>
<td>27.2</td>
<td>10.1</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* NaAlO_2 - 14.4%, NaNO_3 - 47.6%, NaNO_2 - 0.74%

TABLE 5
DENSITY OF CALCINED ALUMINUM NITRATE

<table>
<thead>
<tr>
<th>No.</th>
<th>Bulk Density g/cc</th>
<th>Powder Density g/cc</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.17</td>
<td>0.84</td>
</tr>
<tr>
<td>2</td>
<td>0.17</td>
<td>0.86</td>
</tr>
<tr>
<td>3</td>
<td>0.17</td>
<td>0.77</td>
</tr>
<tr>
<td>4</td>
<td>0.16</td>
<td>0.74</td>
</tr>
<tr>
<td>5</td>
<td>0.16</td>
<td>0.79</td>
</tr>
<tr>
<td>6</td>
<td>0.16</td>
<td>0.75</td>
</tr>
<tr>
<td>7</td>
<td>0.23</td>
<td>1.08</td>
</tr>
</tbody>
</table>
Test 1 indicated that 28.2 per cent of the Al(NO₃)₃ remained unconverted after 90 minutes calcination at 230°C. By increasing the temperature to 415-525°C, (Tests 2-3) Al(NO₃)₃ residual was reduced to 1 per cent even though the actual calcination time was less (1 hour). A significant decrease in Al(NO₃)₃ decomposition was observed, Tests 4-6, when calcining times were extended to the 100-125 minute range. In terms of the desired final product, Al₂O₃, approximately 90-91 per cent conversion was attained in Tests 4-6, whereas 84-87 per cent was obtained in Tests 2-3.

Qualitatively, the greater the conversion of nitrate to oxide, the higher the total recovery of nitric acid gases in the scrubbing liquid. Table 3 shows the actual distribution of nitrate and nitrite between the spray liquid and the bubbler samplers. With the exception of Test 1, > 99 per cent of the nitric gases appeared in the scrub liquid. This indicates that the bubblers, which were operated continuously, were not responsible for significant system decontamination. Incremental analyses of bubbler contents throughout the test period showed no apparent increase in total nitrogen content at the end of the calcination period. A material balance between total feed and total recovery in the scrubbing liquid and bubblers showed about 95 per cent accountability. It was presumed that the remaining material was distributed throughout the system.

The calcined product was in all cases a highly porous material with an average bulk of density of 0.18 grams per cc. When powdered in a mortar and pestle it was possible to compact it to a bulk density of about 0.8 grams per cc (See Table 5). It is emphasized again that these experiments were not designed to produce an optimum density cake but rather to generate a realistic aerosol. Since most tests resulted in nearly complete evolution and collection of gaseous decomposition products, the simulation was considered satisfactory.

CONCLUSIONS

Open Cycle Tests

The following conclusions are based upon partial evaluation of the SK jet scrubber system on open cycle tests.

1. Coarse particulates, > 5 micron, were collected at efficiencies ranging from 90 to 99 per cent depending upon gas and spray liquid flow rates, 60 to 20 cubic feet per minute and 2.23 to 3.8 gallons per minute, respectively.

2. Iron oxide fume, mean size by weight 0.15 microns, was collected at low efficiencies, 15 to 30 per cent.

3. The collection of condensable gases, i.e., HNO₃ vapor, was directly related to gas stream temperature and inversely related to scrubbing liquid
temperature as well as to the solubility of the gas in the scrubbing liquid.

4. A minimum HNO₃ efficiency was attained for low inlet concentrations (300 parts per million) when the mist formed was in the size range of 0.1 to 0.2 microns.

5. At higher HNO₃ concentrations (2000 parts per million) mist formation caused droplet growth to sizes collectable by impaction mechanisms. Resulting HNO₃ efficiency exceeded 97 per cent.

6. Collection of iodine vapor, non-condensable in low concentrations, was dependent upon the solubility of the gas in the scrub liquid. Greater than 99 per cent efficiency was attained with NaOH and as low as 80 per cent with a water rate of 2.23 gallons per minute and an air flow rate of 60 cubic feet per minute.

7. Nitrogen dioxide gas was collected at very low efficiencies, 1 to 3 per cent, due to its low solubility in water sprays. Alkali results were inconclusive.

8. The over-all performance of the SK jet scrubber in terms of collection efficiency was comparable to that of several commercial wet washers using low pressure spray systems.

9. Liquid to gas ratios were considerably higher for the SK jet scrubber in comparison to many wet washers, i.e., 70-150 vs. 5-15 gal. per min. per 1000 cu. ft. per min. of gas handling capacity.

10. The gas moving capacity of the SK jet scrubber showed low effective "fan" efficiency, less than 13 per cent.

11. Sulfur dioxide absorption tests now in progress indicate that spray patterns and degree of spray washout within the SK ejector have a strong influence on performance. Further tests are proposed to define these relationships.

Calcination Tests - Effluent Evaluation

The following conclusions are based on preliminary closed system calciner effluent evaluations.

1. A 95 per cent conversion of a simulated Al(NO₃)₃ liquid waste, 11.4 per cent by weight, was attained in the calcining vessel when temperatures exceeded 500°C and calcining times were at least one hour.

2. Gaseous decomposition products, HNO₃, NO₂, NO, and other oxides of nitrogen were considered representative of typical effluents from a waste
corresponding to our simulant.

3. Collection of nitric acid gases approached 100 per cent when a 0.5N NaOH spray was used since the extended gas retention time in the closed system was sufficient to compensate for the problems of HNO₃ mist collection and the slow absorption rate of NO₂.

4. No data are yet available on particulate or liquid droplet carry over from the calcining vessel. Further tests will include measurement of carry over of typical trace contaminants under various operating conditions.

REFERENCES


2. Ibid., Vol. 2.


AIR MONITORING FOLLOWING THE SL-1 ACCIDENT

George Wehmann
U. S. Atomic Energy Commission
Health and Safety Division
Idaho Falls, Idaho

ABSTRACT

This paper describes the air monitoring program following the SL-1 accident. The types of sampling equipment and media utilized are discussed. Air concentration data and resultant doses due to iodine-131 are presented.

As a routine part of the environmental monitoring program at the National Reactor Testing Station (NRTS), air samples are taken continuously at nine locations on-site and thirteen locations off-site. Additional sampling equipment is utilized in the event of pre-planned or accidental releases. This paper describes the air monitoring program for the SL-1 accident and the results that were obtained.

Planning meetings for emergencies, as well as past experiences at the NRTS, have indicated that the capability for sampling for gaseous radiiodines must be incorporated in the monitoring equipment. This has been achieved at the NRTS and it should be pointed out that the two samplers described in this paper were not developed as the result of SL-1 accident, but have been in use at the site for several years.

Before describing the sampling equipment, the station locations will be discussed. Figure 1 shows the location of the thirteen off-site continuous air sampling station utilizing low volumetric equipment. In addition, nine of the stations were equipped with high volumetric samplers which were activated by a radio signal. Siting of these stations was dependent on three factors: (1) the prevailing wind direction, (2) population distribution surrounding the site, and (3) operations on the NRTS. Inasmuch as the NRTS lies in the Snake River Valley, the local wind conditions are influenced by the topographic features. Climatological records indicate that a southwesterly wind blows approximately 45 per cent of the time and 35 per cent of the time the wind blows from the northeast. The southwest winds are generally associated with lapse temperature conditions and as a rule occur during the day time. Inversion conditions occur at night and are associated with northeast winds. However, variations in these generalities do occur, and, in fact, did occur for the first five days following the accident. During these five days a general anti-cyclonic weather pattern predominated over the area. This resulted in a light
variable wind from the north-to-northeast during daylight hours with a
capping inversion above 1000 feet. The remainder of the time a strong,
deep inversion extending from the surface to 3000 feet existed along
with a 5-15 mph wind from the northeast.

The lack of sampling stations adjacent to the southwest boundary
of the site is due to the sparsity of residents in the area and also
to the inaccessibility to the area, which is predominately covered with
lava.

Following the accident the on-site low volume sampling stations
were supplemented by eleven special stations utilizing high volumetric
sampling equipment. One station, the nearest off-site town, Atomic
City, was started the night of January 3, five were started on
January 14, four more on January 21, and one on January 28. The locations
of these stations are shown in Figure 2. A line of six of these
special stations was placed along U. S. Highway #20 which passes about
3/4 of a mile to the south of SL-1.

The continuously operated stations consisted of a one cfm pump
mounted in an aluminum box for protection from the weather (Figure 3).
The filtering device is a small carbon cartridge that was developed
by the Analytical Branch, Health and Safety Division, Idaho Operations
Office (2). The cylinder consists of a flexible acetate plastic, 5/8
inch in diameter by 2 inches in length and open at both ends. The
cylinder is filled with 12-30 mesh activated carbon with a 5/8 inch
circle of 60 mesh brass screen to hold the carbon in place and a piece
of 5/8 inch tygon tubing about 3/16 inch long as a retaining ring to
hold the screens in place. A 5/8 inch disk of all purpose dust filter
paper is inserted at the one end between the screen and retaining
ring to serve as a particulate filter. These filters are routinely
changed weekly and the pre-filter removed and beta counted. The carbon
cartridge is gross gamma counted. The data is fed into an electronic
computer and air concentration values and resultant thyroid doses
obtained.

As mentioned previously, special sampling stations were established
around the SL-1 after the accident. If conventional power was not
available, gasoline operated electrical generators mounted in two-wheel
trailers provided the necessary power. The sampler, shown in Figure 4,
has a flow rate of 20-25 cfm. Without the adaptor head the flowrate
would be 15-20 cfm, which is not sufficient to prevent the motor from
over-heating. In addition, the screening in the adaptor provides very
good support for the filter during periods of rain or snow. Essentially,
the sampling media is the same as is used in the low volume samplers.
That is, it consists of a carbon cartridge, commercially available,
with an all purpose dust filter to remove the particulate. Approximately
.003 inch of the sampler head must be milled out for the carbon cartridge
to fit but this is not exceedingly difficult to have done. The "O"
ring provides a seal between the carbon cartridge and the sampler. These
samplers were changed daily and analyzed in the same manner as previously
described. The special stations were discontinued on March 6, 1961,
when air concentrations approached the lower limit of detection.

Of prime interest are the results of the sampling program. From
this point on, the discussion will be concerning iodine-131 since
gamma spectral analysis indicated this to be the predominant isotope
in both the initial cloud and subsequent release. Both air and vege-
tation sampling indicated that less than 10 curies of iodine-131 were
released during the first 16 hours and 70 curies of iodine-131 over the following 30 day period. This can be compared with a 49.5 curies release of iodine-131 in less than 30 minutes during a RAIA run (dissolution of short-cooled MTR fuel elements) at the Idaho Chemical Processing Plant in 1959. However, this RAIA release occurred during very favorable weather conditions.

The average air concentration for the period January 4-11, 1961, is shown in Figure 5. Ordinarily the limited number of air sampling stations would not permit drawing this map. However, a good correlation between air sampling data and vegetation sampling data permitted the use of results later to fill in the areas where air samples were not available.

A unique program was established using the data from the special sampling stations. Arrangements were made to have the samples analyzed as soon as they were brought in from the field each day. The air concentrations were immediately computed and then maps were drawn of the air concentrations surrounding SI-1.

These maps, which in a manner of speaking were similar to weather maps, provided a simple but very efficient method of evaluating the situation from day to day. In addition to showing the effect of wind direction on the distribution of radioactivity, comparison of different maps provided a quick and easy method of determining the trend in release rates. Maps were drawn daily from January 16 until February 13. A typical map is shown in Figure 5. For this particular day the predominant wind direction was from the northeast with a few hours of southwest winds during the afternoon of the 17th.

The air concentrations of iodine-131 at the nearest off-site population, Atomic City, are shown in Figure 7. The peak shown for the week of December 6-12, 1961, is due to a RAIA run made during this period. It is important to note that the average air concentration was a factor of six lower than the radioactivity concentrations guide (RCG) for off-site populace for continuous exposure to iodine-131. From these concentration values the calculation of the dose to the thyroid due to inhalation of iodine-131 indicates that the cumulative dose for the period January 3 to February 20 was approximately 35 millirads.

In conclusion, it can be stated that the pre-planning philosophy regarding the type of sampling equipment paid off. That is, provisions for monitoring of radio-iodines were necessary to adequately determine the release from SI-1. As a direct result of this capability, it can be conclusively stated that no significant dose resulted from the approximately 80 curies of iodine-131 that was released.

REFERENCES


Figure 1  Off-site air sampling station locations

Figure 2  On-site air sampling station locations
Following the SL-1 Accident
Figure 3  Low volume air sampling equipment

Figure 4  High volume air sampler
Figure 5  Iodine-131 Air Concentration - January 4–11, 1961

Figure 6  Iodine-131 Air Concentrations near SL-1

Figure 7  Iodine-131 Air Concentrations At Atomic City
DENNIS: I am certain that Mr. Hays will, on an individual basis, tell you who manufactured filter "A" and filter "B". I would also like to point out that it is not the function of this agency to act as a sales promotion group for any one manufacturer; we attempt to be as unbiased as possible. Since, there was some labeling on a slide indicating that the Cambridge Corporation is one manufacturer producing high efficiency filters, we should point out that in the United States there are three major fabricators, all supplying filters to the AEC, including the Cambridge Filter Corporation, the Mine Safety Appliance Company and Flanders Filters.

We have several interesting questions that can be asked. I am going to set the bell for ten minutes and during this interval I hope that the pertinent questions relating to the papers presented by Mr. Anton, Mr. Hays and Mr. Riley can be covered.

BILLINGS: This service that Mr. Hays talks about, why aren't they using a stripping system to get this moisture out? This looks like a misapplication to me.

HAYS: We have had this in mind and tried a few of these. We took the attitude, "Let's see what they will do."

FIRST: This is not a question but a comment on your results: There are two types of water repellancy to be considered. If, for example, you treat a paper with a silicone oil, or something similar, it will shed liquid water. However, if this same paper is subjected to high humidity, you may find that in a few hours the paper becomes soaking wet and useless from adsorbed water vapor against which the water repellant treatment gives no protection. Therefore, moisture repellancy, as well as water repellancy, is a desirable property in a filter. A problem arises in trying to get both properties in a single filter paper. It may be that the user will have to insist on the characteristic which he needs most.

WEITZEL: What can you say about the position of the filter and the direction of flow with relation to its ability to shed water; i.e., updraft or downdraft?

HAYS: These tests were run on the filter in the vertical position.
SHAVER: I wonder about manufacturer "C". We had "A" and "B". With the current status of filter papers, there is a situation where we are getting new products -- do you plan to test further materials, and so forth?

HAYS: Yes. The point you made is very good. They seem to be changing almost day by day and manufacturer "C" was omitted because during the early stages of our testing we showed that the water-repellency used by manufacturer "C" at that time was very poor.

I didn't mean, or don't intend to make a big difference between the two manufacturers. Both of them are poor on this test. Both of them are down to about 0.1 of your flow at four inches pressure drop. We have not shown anything to indicate that "A" or "B" have the filter we want. I would like to clear that up, if there is some idea of saying that filter "B" is, by far, the best filter shown. They are both below what we need.

SILVERMAN: I would like to ask John Riley what he does in case of fire in the charcoal bed, or what kind of information they have about this secondary hazard.

RILEY: First of all, we try to prevent it by putting in the heat sink where we are expecting high temperature gases.

SILVERMAN: Any one of them are subject to fire, depending on failure of your sink or heat exchanger.

RILEY: The beds would be totally enclosed in suitable vessels that could be completely isolated in the event of a fire.

DENNIS: Can you tell us what limiting temperature can be handled by the heat sink and the heat exchanger in the case of a catastrophe condition?

RILEY: The medium design inlet temperature of the bed depends on the ignition temperature of the particular type of charcoal used. In the present case a maximum temperature of 100°C has been assumed.

DENNIS: 300°C entering the heat exchanger and 100°C entering the fine filter circuit?

RILEY: Yes.

SCHNELLER: I presume that your heat sink system is on the line at all times?

RILEY: Yes.
SCHNELLER: Then in utilizing the recirculating system how is the heat sink line closed off and the recirculating system put on the line? I'm interested in both the instrumentation and mechanical means of effecting the change-over.

RILEY: The heat sink line is only brought into use when the valve leaks. The extra line will be brought in by some sort of instrumentation based on pressure surges or on activity releases. It will be mainly brought into action by excess pressure and will shut off the ventilation plants at the top. In the event of a serious emergency, you would want this one shutting off to prevent the direct discharge to the atmosphere.

KEIGHER: I would like to remind all attendees that four years ago, particularly at the Boston meeting, our concern was strong in the area of fire resistance. We were having many fires in filter installations all over the Commission. We seem to have swung now to the problem of waterproofing and water resistance. I would like to continue the real concern that there are very few situations where the fire problem has been fully eliminated or is negligible, so please keep this in balance when trying to compromise these requirements, whether it is moisture resistance you want or fire resistance. The fire problem is still with us and will be in all filter installations.

DENNIS: I will have to curtail further questions. There are several persons here who will be glad to discuss unresolved points on an informal basis.

Originally we had scheduled one hour's time for a summary of the conference and future business. At this time, with a little pressure on Dr. Silverman, I am going to request that he treat these subjects with the emphasis on future business.

SILVERMAN: It is pretty hard to summarize this conference, except to point out that we have seen filters exposed to everything from snow to steam. We expect that the exposures will keep increasing in number and variety. We have also seen that there are many new techniques that are useful for assessing performance of filters. There are many and varied problems in gas contamination as well as on the particulates in the air.

I would like to simply digress for a moment to say to Mr. Chester that we are not as ignorant as we may seem about sophistication in our analyses. Actually, in the carbon bed problem this has been subject to derivation of isotherms and attempts to correlate these with performance. If you can see the differences that Dr. Adams pointed out between carbon; and the difference in carbon preparations as to pore size etc., you can see that you have problems of multiplication and variation. Add to that the problem of multiple gases and aerosols, combined, where there are varied characteristics of the gas and the aerosol, increases the complexity. We can
derive fairly simple cases and I think Mr. Chester had a good example of one where we are dealing with fairly simple gases, carbon monoxide and hydrogen, in a fairly simple system. Still he had to take the coward's way out and put his results into a digital computer. We can and have done the same thing in a few cases and we certainly appreciate his comment.

In regard to this conference, many agree with me that these are very useful. I hope you all have arrived at the same opinion, but not everybody is agreed as to what should be covered. It seems to be axiomatic that the larger the conference gets, the more formal it becomes. I noticed that some of our more vocal attendees, people who had been more vocal in the past, in smaller groups were restrained somewhat, Mr. Thaxter in particular.

The more papers put on a program, the more pressed you are for time. An inverse relationship between formality and time also seems to show up. I hope that at the next conference, and there have been some favorable expressions in this regard, we will reserve perhaps half a day for questions and discussion.

As to future meeting suggestions, the first one would be frequency of meeting. We all feel that the intervals we have had for these in the past have not been too bad. Others think the meetings ought to be held yearly. We are prepared to take an informal vote on this matter. I would say that we have also had requests as to the content of the conference, whether we should have more panel discussions and, also, whether we should repeat the training discussions. The latter was one frequent question: "When are you going to repeat the training type discussion you had at Harvard in 1957 and 1951?" This will depend on the general interest and requests from people.

There have been some expressions that we ought to point out that the basic filter and aerosol mechanisms are not well understood. I think this is a remark that goes to almost every conference, but maybe it was more obvious here, when we deal with some of these complex heterogeneous mixtures of gases and aerosols. Certainly we ought to consider more theoretical discussions, but many people come to these conferences with problems and they would like answers at the earliest opportunity.

I think Mr. Hays is the most recent example of that situation.

Questions have been raised about very specific items: Dust check systems, prompt release detection at stacks, and continuous monitoring for stacks.

For the future meeting, we have had people say we should emphasize basic phenomena, particle and vapor deposition, more work on low iodine concentrations, more work on instrumentation, more emphasis on panel conferences, which I have mentioned, and somebody said we should have a bibliography on air cleaning publications.
To the last one, I can answer, "Thank heavens, the Library of Congress has this responsibility, under money provided under Public Law 159 of the Public Health Service." I don't believe that many things are not already covered in their bibliographies which have been made available in recent years. The continuing updating of these would call for a particular effort by the AEC. However, we will entertain the subject, again in the hope that if a volume that brings everything up to date is what the individual has in mind.

Just to throw out three items for vote, perhaps, we might take as the first one: Frequency of meeting? It looks like some place between one and two years is the consensus of the group.

The next question is as to location, and I would perhaps put this out on a priority basis.

Taking the Chair's prerogative, I would put down Oak Ridge as the first possibility and find out all those in favor of going to Oak Ridge. I will recognize no vote from an Oak Ridger in this regard. All those in favor of going to Hanford? It looks like Oak Ridge won; UCRL is second; and, Hanford is third.

This brings me to the third point, and that is whether or not the next conference should incorporate a training session. Speaking for myself and my colleagues, the frequency of these, at four to five-year intervals, has been desirable, because we take a lot of time and trouble to prepare for these training sessions. We try to bring things up to date, so we would not particularly care to do this at Oak Ridge, away from home. My own demonstration yesterday is a good point. At any rate, the Chair will entertain any sentiment in this regard.

BILLINGS: The sessions we went through this summer, are we going to do it again two years from now?

SILVERMAN: I think Mr. Billings is referring to something which was perhaps stimulated by these AEC conferences. It was asked for by the American Industrial Hygiene Association. We held an Air Cleaning Institute at Harvard, which lasted a week, in which we extended the training you might have been exposed to at the AEC air cleaning conferences to five days. One of the days included contributions from the industrial people who were present.

This particular type of sessions we might be asked to do again in the interim and we would certainly be happy to entertain applications from other people. There is, however, a difference, namely, that there is a charge for this service, whereas, under our AEC work, we consider it an obligation of our contract. We will inform you if we plan to repeat this Institute soon. Having just finished one, I must say that I am not happy to repeat it again on the same basis, without an equivalent regeneration period. Let me just sound out the audience. How many in the audience would like to have us hold a two-day training session at the time or just prior to the next conference,
even though it couldn't be at Oak Ridge?

It looks like about eight. You would probably be interested in sending some other people from the same facility, so that we might consider one.

Can we get a show of hands from the people who just put up their hands? Two from Oak Ridge, Hanford — Oak Ridge and Hanford seem to want it, and Los Alamos. I think we have enough of an opinion here to give this some very serious consideration.

Well, that is about the extent of what I wanted to say. I am sorry that we don't have time to take up all possible answers to many questions that have been raised. We are going to have a lot of interest in air cleaning. There is much interest in monitors and in rare gas removal. These items will be on the agenda again, without fail, next time. This might stimulate us to having the meeting a little sooner, because of the greater interest in these items.

CULKOWSKI: I have a suggestion for the Oak Ridge meeting. Due to a tremendous number of meetings that are held there, and the added factor of foreign visitors, notice of future meetings should be as far in advance as possible.

SILVERMAN: Six months is what we would like to think about. You can see here, and it is certainly no fault of the parties concerned, but even three months, which is about the lead time that we had for this meeting, is not quite enough. Of course, you do get data right off the griddle. It may be burned or fried, but still it is fresh! Nevertheless, it is important to a lot of people to get clearance and to get authorization for travel and matters of this kind. I can say amen to this business of lead time. We ought to have six months.

COSTELLO: You might make reference to the vacation time. It was a bad time to get information out while people were on vacation.

CULKOWSKI: Down there we feel that the Fall would be an ideal time from the esthetic standpoint and the ease of obtaining reservations. With vacations over, papers should be easier to prepare.

SILVERMAN: Columbus Day seems to be a good time. We observe the holiday, which makes it nicer, because we are not missing anything in Boston, except a day off. From the time of the year standpoint, it makes it bad for academicians who get involved with classes and things of that nature. We are in the minority however and we won't squeal too loudly.

WESTERN: I wonder if you could notify us individually six months in advance, as well as through the department heads and letting it filter down?
SILVERMAN: This is a pretty absolute filter when it hits the department head.

COSTELLO: We are supposed to go through channels. However, there is no reason why we couldn't send you a carbon copy. We finally did notify everyone by air mail special delivery and to expedite this I put about 200 envelopes in the Washington mail box myself.

SILVERMAN: In closing, and it looks like we are at that point, we want to again thank our host, the Brookhaven National Laboratory. Some of you will still be dependent upon them for the next twenty-four hours. We want to express our appreciation to Mr. Costello and Mr. Belter, who got things under way and kept them rolling, and to all the speakers who answered our requests for recent information and came, sometimes on short notice, to replace others, and all the rest of you for being such a patient audience and contributing to the discussions from the floor.

I would like to have Walter Belter close the meeting with the official AEC blessing.

BELTER: Unfortunately, we ran out of time again today, just as we do in all of our meetings. We had hoped to have an hour to an hour and a half this morning in which we could have had some active, lively discussion, similar to the night before last, between five and six o'clock. We did not think the agenda for this morning would be crowded, but it turned out that this three hours slipped away more quickly than anticipated. I think we shall make a note of Dr. Silverman's suggestion that at our next meeting we plan to have about one-half day with no formal papers or anything scheduled at all and leave the time open for discussion and active participation from everyone in the audience. It seems even though we set up these agendas to have something like a half hour discussion at the end of papers, we are never able to adhere to this type of schedule. Fortunately, you didn't mind the last two evenings breaking up at approximately six o'clock, and that did give us some extra time for discussion.

In conclusion, on behalf of the AEC in Washington, I certainly want to thank Dr. Silverman and his staff at Harvard for their tremendous contribution to making these meetings a success. It is difficult to envision an air cleaning conference without their leadership and active participation. We certainly are appreciative of their efforts.

For all the people who presented papers -- we want to thank each of you for taking the time to prepare this material and to come to BNL.

I think the applause that was given to Lee Gemmell a short time ago was certainly due him and his staff for making all these many arrangements that are necessary in conducting a meeting of this type. You have our sincere thanks and may we hope that we will be able to come back to Brookhaven again very soon. With that final note, we will now adjourn.