EXPERIENCE IN TRAPPING IODINE - 131 AND OTHER FISSION PRODUCTS RELEASED FROM IRRADIATED AGR - TYPE FUEL ELEMENTS

by

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SUMMARY

Iodine-131 and other fission products have been released from irradiated stainless steel-clad uranium dioxide under conditions relevant to potential accidents in the Advanced Gas-Cooled Reactor.

Measurements have been made of the penetration of Iodine-131, Tellurium-132, Caesium-137 and Strontium-90, released in the above manner, through a trapping system consisting of an absolute filter, followed by a 4" deep bed of charcoal and a final absolute filter, in series. No significant penetrations have been observed except in the case of iodine-131 in the form of alkyl iodides. Some small fraction of the iodine released from the irradiated fuel is in this form and investigations have shown some conditions in which this form can be produced from iodine initially present in other forms.

Experiments with macroscopic amounts of methyl iodide, representing the above form of iodine, have shown that it is only delayed on activated charcoal at temperatures up to 100°C, but part of it can be permanently retained at 200°C. Oxidising catalysts show some promise of effectively retaining methyl iodide, but the presence of carbon monnoxide interferes.

INTRODUCTION

An efficient fission product trapping, or clean-up plant is an essential part of the containment system of the AGR and of various other types of reactor. If we can establish confidence in the ability of containment systems to prevent the spread of fission products in the remote event of a reactor accident, relaxation in reactor siting requirements could follow. A programme of experimental work to prove the ability of the trapping plant to remove dangerous fission products in the case of the AGR is now in hand at the UKAEA Reactor Group Laboratories at Windscale.

The fission product which causes greatest concern is iodine-131 so that it is chiefly the behaviour of iodine which determines the design of trapping plants. Other fission products that have to be considered include Tellurium-132, Caesium-137 and Strontium-90. The rare gas isotopes would be extensively released in a reactor accident, but, as they can be fairly rapidly dispersed from a high stack, they do not present a major hazard with current sizes of reactor, and current designs of trapping plant for gas-cooled reactors are not intended to retain them.

It was known from earlier work, notably at AERE(1) and at ORNL(2), that the trapping plant should include an absolute filter and a bed of activated charcoal. In addition, a heat sink would be necessary to ensure that the operating temperature limits of the charcoal were not exceeded. Other traps, for example, aqueous alkaline scrubbers or copper mesh, are effective against I_2 vapour, but workers at AERE have shown that when elemental iodine vapour is released into the atmosphere at low concentrations, part adsorbs on solid aerosols and part forms chemical compounds with trace impurities. Against iodine emissions of this nature, the combination of absolute filter and charcoal bed provides the best available barrier. More recently, it has been shown at AERE(3) that two types of iodine compounds are formed in such releases: inorganic compounds which are reactive and easily removed by caustic soda and organic compounds which include methyl, ethyl and higher alkyl iodides and which are not easily removed from the gas.

The chemical and physical forms in which fission product iodine would be released from heated irradiated uranium dioxide in carbon dioxide atmospheres were not known at the outset of this work and were expected to depend upon the burn-up of the fuel and the conditions in which it was heated.

Two representative types of reactor accidents are taken as the basis of the experimental study:-

(a) pressure circuit rupture, leading to escape of fission product activity into the containment volume and a need for clean-up by recirculation through the trapping plant.

(b) channel blockage, which leads to overheated fuel and release of fission products into an intact pressure circuit which may be cleaned up by direct blowdown through the trapping plant.

In both these cases, a trapping plant which restricts the penetration of iodine to less than 1 part in 10^4 of that presented to it is considered to provide satisfactory protection of the environment.

EXPERIMENTAL

The experimental approach is to release fission products from irradiated fuel elements, to measure the amounts and forms in which they are released and, in the same experiment, to measure their penetration through scaled-down trapping systems. This work is augmented by studies of techniques by which the forms of iodine released from irradiated fuel may be simulated with separated iodine isotopes. These techniques are then used to study iodine behaviour and to test trapping systems on a larger scale.

The experiments fall into three main categories:

(a) Laboratory-scale experiments in which activity is released into gas at atmospheric pressure in glass apparatus and presented, with little time delay, to a gas treatment system. Irradiated miniature fuel elements can be handled up to a total activity of about 50 curies in a lead-shielded cell (Fig. 1). Similar experiments are carried out in unshielded apparatus in which iodine is released from trace-irradiated miniature fuel elements or from identical unirradiated fuel elements to which separated iodine-131 has been artificially added. This work has been in progress for about 18 months.

(b) Larger scale experiments in which iodine can be allowed to age, under conditions appropriate to its release in a reactor accident, and is then presented to commercial-sized filters and charcoal beds. This work includes experiments in a large steel filter test rig (Fig. 2), which has only recently been brought into operation, and allows reactor accidents to be simulated with separated iodine-131 or 132, or with iodine released from trace irradiated fuel.

(c) A single highly active experiment in a small steel rig (Fig. 3) in which fission products are released from irradiated AGR fuel elements at ~2000 MWD/te burn-up, on the scale of about 4000 curies total activity. After the release of fission products into gas at high pressure, the performance of a correctly-scaled trapping system is measured under conditions simulating (i) direct blowdown of the pressure circuit through the trapping system (ii) rupture of the pressure circuit and release of its contents into the containment volume, with subsequent recirculation of the containment volume contents through the treatment system. This experiment has been carried out but final evaluation of its results is not yet complete.

A fourth aspect of the programme is the testing of the installed trapping plant of the Windscale AGR with artificially generated releases of iodine-132. Some tests were carried out earlier this year, but further work is planned.

Trapping System Components

Unless otherwise noted, the trapping system used in the experiments consisted of a coarse pre-filter (Vokes glass fibre material), an absolute filter (Vokes VAF 76 glass fibre paper), a 4" deep x 1" dia. bed of activated charcoal maintained at 100°C (SCII coconut charcoal - 8 + 12 B.S.S. mesh size), and final coarse and absolute filters.

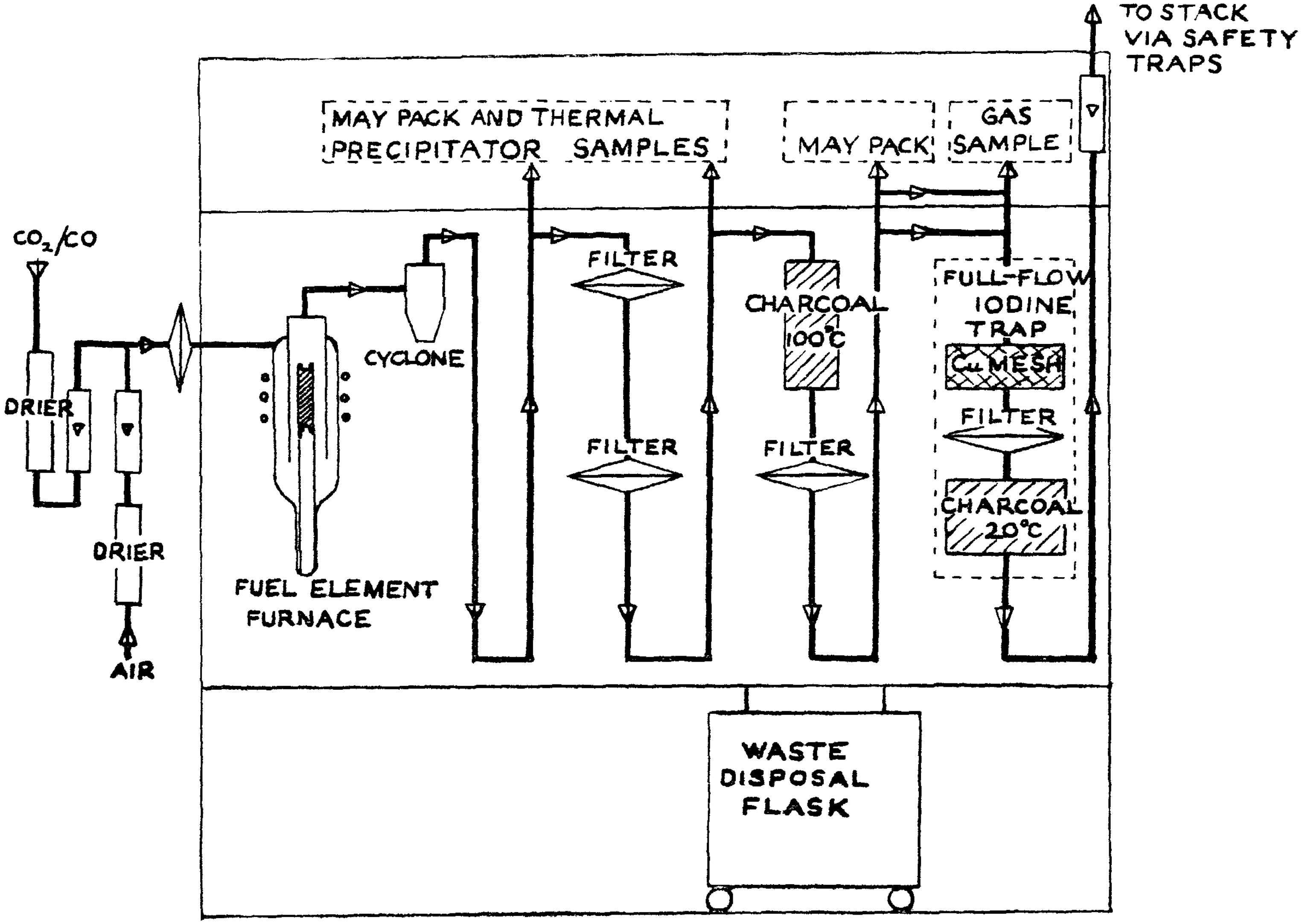
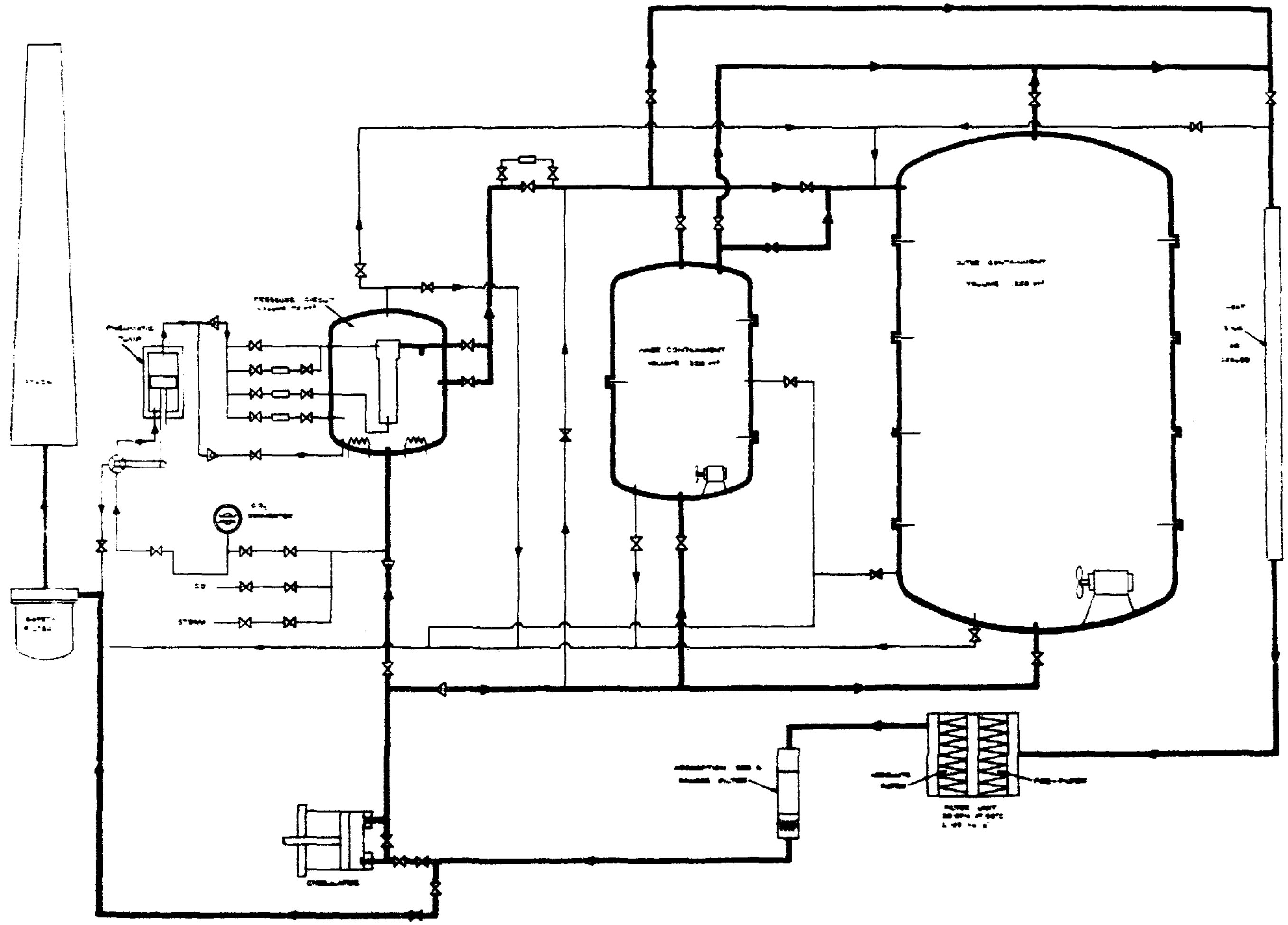

TO STACK VIA SAFETY TRAPS
MAY PACK AND THERMAL PRECIPITATOR SAMPLES
MAY PACK
GAS SAMPLE
CO$_2$/CO
DRIER
DRIER
AIR
FUEL ELEMENT FURNACE
CYCLONE
FILTER
FILTER
CHARCOAL 100°C
FILTER
FULL-FLOW IODINE TRAP
Cu MESH
FILTER
CHARCOAL 20°C
WASTE DISPOSAL FLASK

Figure 1



AG.E FILTER TEST RIG

Figure 2

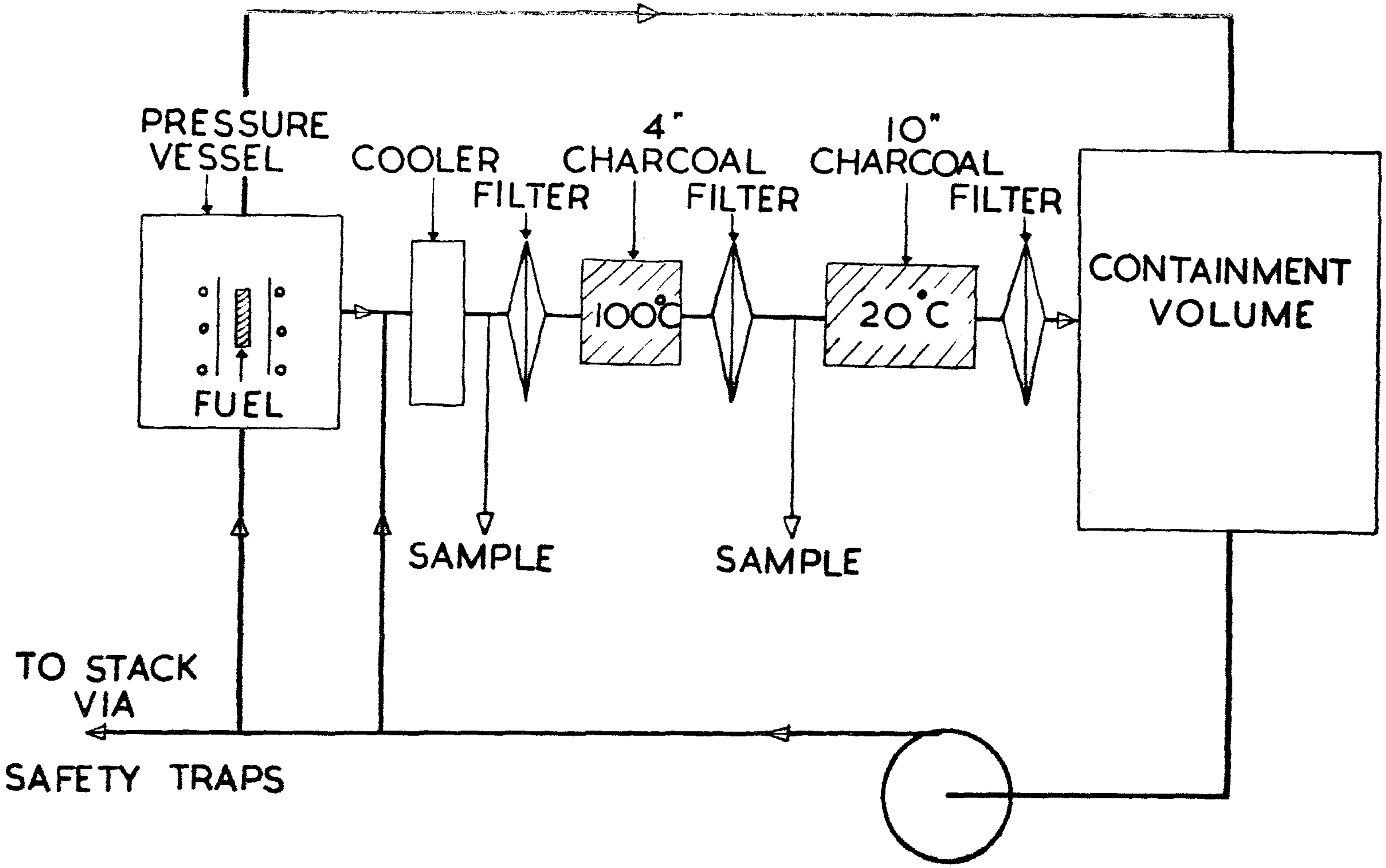


Figure 3

HIGHLY ACTIVE TEST

Iodine Samplers

The fission product activity in the gas is measured at points before and after components of the trapping system by sampling packs designed principally to distinguish between different forms of iodine. These are called "May Packs" after F. G. May of A.E.R.E. who originated them.

The version we use (Fig. 4) consists of the following components which the gas passes in succession as it traverses the pack.

(a) Four layers of fine copper gauze, especially cleaned before use and effective against elemental iodine vapour.

(b) Type PH millipore filter, which is highly effective against all sizes of particles and consequently gives a good measurement of particulate iodine.

(c) Two carbon-impregnated glass fibre papers (Whatman Type ACG/B). Activated carbon is the best known adsorber for ill-defined compounds of iodine that can penetrate elemental iodine adsorbers (such as copper) and particulate filters. By using these papers, the equivalent of only a very shallow bed is presented so that only the more reactive compounds are retained.

(d) Two successive 2" deep beds of SCII charcoal (-18 + 52 B.S.S. mesh size). These beds are effective against the less reactive iodine compounds.

The components of the pack are separated by PVC gaskets and the whole assembly is welded inside a PVC envelope to prevent leakage. Holes to permit gas passage are cut in the envelope just before use. The pack is designed to take a gas flow of 600 cc/min. The separate components are analysed by gamma spectrometry after use in the experiment.

Particle size analysis

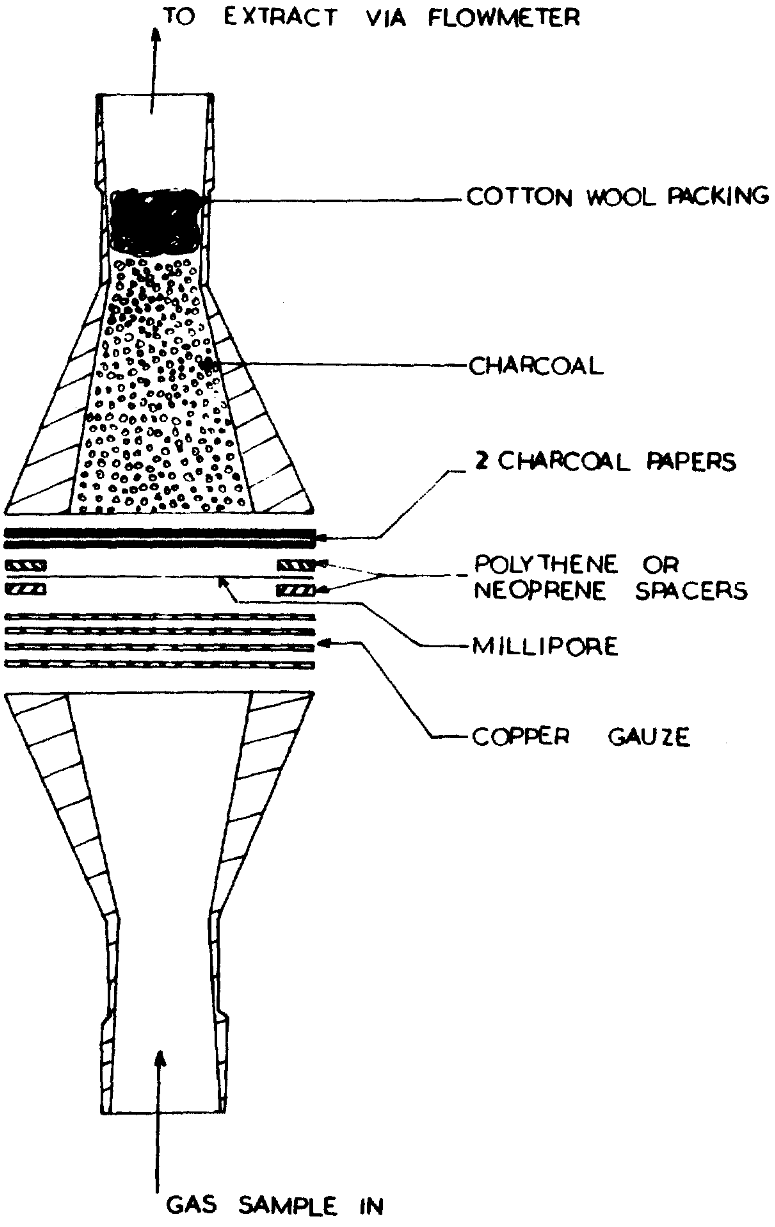
The particle emission from the heated fuel element is sampled by thermal precipitators before and after the coarse filter and first absolute filter. The particles are deposited on a silicon monoxide film which gives a very clean background when examined in the electron microscope (only 10-100 particles per hundred fields of view at x 50,000). Samples taken before the filter on type PH millipore filters and on thermal precipitators give particle size distributions that agree very well with one another. However, millipore filters cannot be used after the absolute filter because their background contamination is too high for sensitive detection of particles which are present at very low concentrations in the gas.

Fuel Elements

The AGR fuel elements consist of uranium dioxide pellets canned in stainless steel and miniature elements are made up from three full size UO_2 pellets sealed inside a short length cut from an actual AGR stainless steel can. Fission products are released by induction heating the elements to above the melting point of the can, either in carbon dioxide containing some carbon monoxide, which is the normal coolant in the reactor, or in mixtures of these gases with air which, in some fault conditions, may be present in the gas surrounding the fuel elements.

MAY PACK

Figure 4



RESULTS AND DISCUSSION

General Summary

Growing knowledge of the complex behaviour of iodine when released into the atmosphere as elemental iodine vapour, has led one to expect at least equally complex behaviour when the release is from irradiated fuel elements into a reactor circuit and containment. It is therefore encouraging to find in many experiments that the combination of absolute filter and charcoal bed provides adequate performance.

Only one class of iodine compounds has been found with which the above trapping system may not be able to deal effectively. These are the alkyl iodides, of which methyl iodide is the predominant and most penetrating member.

The alkyl iodides appear to form only a very small proportion of the primary products in the emission of iodine from irradiated uranium dioxide fuel at about 1500°C to 1700°C in CO_2, but it is possible that the amounts produced may be sufficient to penetrate activated charcoal when the total amount of iodine released is such that the loading on the charcoal exceeds a few µg iodine per gram of charcoal.

In experiments with small quantities of irradiated fuel, iodine released at 1500°C to 1700°C and carried immediately to a trapping system, penetrates the system to an extent not exceeding a few parts in 10^4 at low levels of iodine release (about 10^{-2} $\mu g/m^3$ in the gas stream) and dropping to about one part in 10^5 at concentration levels of relevance to serious reactor accidents (about 500 $\mu g/m^3$). The penetration of iodine through the system is not significantly increased by continued passage of gas for at least 24 hours after the iodine has been presented to the system and is independent of variations in gas composition ranging from pure CO_2 to CO_2 containing 50% CO and CO_2 containing 50% air. These experiments cover a range of iodine loadings on the charcoal from 5×10^{-4} $\mu g/g$ to 2 $\mu g/g$.

In a larger scale experiment the total iodine loading on the charcoal was about 40 µg/g and the penetration amounted to a few percent of the iodine on the charcoal; that coming off being identified as a mixture of alkyl iodides. This behaviour may have resulted from the larger quantity of alkyl iodides presented to the charcoal simply because of the larger amount of irradiated fuel used in the experiment. On the other hand, the conditions of the experiment were such that a large proportion of the iodine released from the fuel deposited on vessel walls and other surfaces at temperatures up to about 400°C and it has been shown in other experiments that iodine desorbed from surfaces in these conditions contains an enhanced proportion of alkyl iodides.

The extent to which alkyl iodides can be formed by this latter route will be related to the ease with which the primary forms, released from the fuel, deposit on the surfaces (especially steel surfaces) where the secondary production of alkyl iodide appears to occur. Iodine released from irradiated fuel elements into a fairly rapid flow of carbon dioxide (about 600 cm/sec. past the specimen), does not deposit readily on pipework and other surfaces and hence is unlikely to result in the secondary production of significant quantities of alkyl iodides. On the other hand, slow gas flow past the heated fuel, which may encourage the iodine-containing particle emission from the fuel element to grow to sizes at which it deposits out readily, or the presence of air, which encourages the release of readily-deposited elemental iodine vapour, may favour the secondary production of alkyl iodides after the primary forms have deposited on surfaces.

The need for a specific trapping process for methyl iodide and other, similar, organic iodides will depend upon the limit that can be placed on the production of alkyl iodides in specific reactor accidents. The development of such a process is currently receiving attention but no really satisfactory solution can be specified at present.

Of the other fission products, none, except the rare gases, has been found to penetrate the trapping system. Caesium-137 and Tellurium-132 are released from irradiated uranium dioxide fuel elements virtually entirely as solid aerosols, of which less than one part in 10^4 penetrates the absolute filter. Strontium-90 is also released in a particulate form but so little is released from the fuel that it is below the limit of detection after the absolute filter.

The above general statement of the present position derives from experimental results that are considered in more detail below.

Fission product release from overheated fuel elements

The forms and amounts in which the dangerous fission products are released from the fuel have been studied under controlled conditions in the laboratory. For practical reasons, the miniature fuel element (containing 25 g. UO_2) is heated at atmospheric pressure in a gas flow equivalent to about half the normal mass flow per unit weight of fuel in an AGR channel. (i.e. in a flow of 17.5 l/minute which passes over the specimen surface at 600 cm/sec.). The gas composition, fuel element temperature and irradiation dose have been varied.

If the initial fission product content of the fuel is calculated from the neutron dose, the percentage release of the dangerous fission products in 30 minutes at a temperature of about 1500 to 1700°C is as given in Table I.

The release factors do not appear to vary systematically with fuel burn-up in the range $\lessdot$1MWD/te to ~2000 MWD/te, although there may be an effect of cooling period, since, of two specimens irradiated to about 2000 MWD/te, one, cooled 5 days, released about 70% of its iodine while the other, cooled 30 days, released only about 10%.

At lower temperatures the release of iodine is much slower. Work with trace-irradiated miniature fuel elements shows that release begins to be measurable at between 1200 and 1400°C and continues at the rate of a few percent per hour up to about 1550 to 1600°C where there is a sharp increase in rate and substantial release occurs in a few minutes. This behaviour seems to be independent of whether air is present in the CO_2 or not.

TABLE I

FISSION PRODUCT RELEASE FROM IRRADIATED UO_2 HEATED AT ABOUT 1500°C TO 1700°C FOR 30 MINUTES AT ATMOSPHERIC PRESSURE

Atmosphere	Iodine 131	Tellurium 132	Caesium 137	Strontium 90
95% CO_2/5% CO	~30	~30	~50	~0.01%
50% air/50% CO_2	~ 30	~ 30	~10*	not available

*only 10 mins. above 1500°C.

The forms in which iodine is carried in the gas stream leaving the furnace have been classified by the May Pack. In the absence of air, the majority of the released iodine is attached to solid aerosols and the remainder is present as undefined vapour compounds of iodine. The presence of air causes a larger fraction of the iodine to appear as elemental vapour and, although the proportion attached to solid aerosols is much reduced, a significant proportion is still as vapour compounds of iodine (Table II). When the iodine is released from the fuel in CO_2/CO mixtures about 90% of it is retained by the first absolute filter and the remainder is over 99% in the form of vapour compounds of iodine. Methyl iodide has recently been identified in the gas at this point, to the extent of a few percent of the total iodine present (a few tenths of a percent of the iodine released from the fuel). The identity of the remaining compounds is not yet known: they are not retained in a sodium carbonate scrubber but they are apparently strongly held by charcoal. In air/CO_2, 80% of the iodine which penetrates the absolute filter is elemental iodine vapour and the remaining 20% has not yet been examined although, again, it is apparently strongly held on charcoal.

The particulate fraction is associated with the solid particles emitted from the heated fuel element components and graphite. These particles consist of agglomerates of primary particles which lie in the size range about 0.005 to about 0.1 micron; the agglomerates themselves range in size from 0.01 to over 1 micron diameter. The mean frequency size of the primary particles tends to decrease with increasing irradiation dose to the fuel elements and, to a lesser extent, with increasing temperature. Experiments with unirradiated fuel element components have shown that the mean frequency size of the primary particles increases almost linearly with pressure in the range 0-500 p.s.i.g.

In the oxidising atmospheres which were used for these experiments and which are generally relevant to potential accidents in an AGR, the released Tellurium, Caesium and Strontium were associated virtually entirely with the particles and did not penetrate the absolute filters in significantly measurable amounts.

At temperatures below the melting point of the stainless steel can, particulate emission from the fuel element is very much reduced and any iodine which is released tends to be in the form of elemental iodine vapour and vapour iodine compounds. As the temperature rises, the particulate fraction increases at the expense of the other two forms, although in the presence of air, even at a concentration of only 1% in CO_2, elemental iodine predominates to higher temperatures (Table III).

Trapping plant performance

The effectiveness of the trapping plant against iodine released from irradiated miniature fuel elements as described in the above paragraphs and carried immediately in the gas stream to the plant, is shown in Table IV.

TABLE II

FORMS OF IODINE RELEASED AT 1500°C TO 1700°C FROM IRRADIATED STAINLESS-STEEL-CLAD UO_2 IN CO_2/CO AND AIR/CO_2

(2" x 0.4" dia. fuel element, containing 25g UC_2, heated in a gas flow of 17.5 l/min. passing over the specimen surface at ~ 600 cm/sec.)

Gas	Gas-borne iodine concentration $\mu g/m^3$		Distribution of iodine on May Pack components			
			Copper	Millipore	Charcoal paper	Charcoal granules
air/CO_2	before filter	~0.001	83.5	8	8.5	-
	after "		not measured			
CO_2/CO	before "	~ 0.05	7	66	13	14
	after "	~ 0.01	0.5	0.5	50	49
CO_2/CO	before "	~ 0.5	8	65	7	20
	after "	~ 0.05	< 0.1	< 0.1	50	50
air/CO_2	before "	~0.5	60	30	9.5	0.5
	after "	~0.2	80	2	16.5	1.5
CO_2/CO	before "	2-4	12	66	17	5
	after "	~0.5	0.25	0.75	67	32
CO_2/CO	before "	7-9	11	75	12	2
	after "	~1	0.5	0.5	42	57
CO_2/CO	before "	196	9.5	76.5	13.2	0.8
	after "	~ 20	1.0	0.5	71	27.5
CO_2/CO	before "	980	12	81.8	6	0.2
	after "	~100	~0.5	~ 0.5	50	49

Table III

SUMMARY OF RESULTS SHOWING EFFECT OF TEMPERATURE ON IODINE FORMS RELEASED FROM TRACE-IRRADIATED UO_2-FUEL IN DEFECTED STAINLESS STEEL CANS

Temp. °C	Distribution of iodine forms on May Pack components								
	in CO_2			in 1% air/CO_2			in 50% air/CO_2		
	Copper	Millipore	Charcoal	Copper	Millipore	Charcoal	Copper	Millipore	Charcoal
1000-1100	67	6	27	-	-	-	92	3.5	4.5
1200-1300	46	36	18	65	15	20	95	2	3
1400-1500	13	69	18	60	33	7	87	7	6
1600-1800	13	80	7	33	62	5	71	23	6

The overall penetration of iodine through the system is measured by three methods, from which the highest result obtained is given in Table IV. The May Pack analysis on an aliquot of the gas stream is backed up by analysis of a deep charcoal bed which takes the full flow of gas through the system and an independent check is provided by direct gamma-spectrometry of a sample of the residual gas obtained after chemically absorbing carbon monoxide and carbon dioxide. The last method allows a limit to be placed on the amount of iodine which could be present in a form not held on charcoal and hence not detected by the other two methods. In almost every case the May Pack analysis gave the highest results and duplicate May Packs agreed to well within a factor of 2.

The measured penetrations decrease with increasing amounts of iodine released from the fuel; from a few parts in 10^4 at about 10^{-3} µg to less than one part in 10^5 at about 1 mg. This is also illustrated in Fig. 5 in which the linearity of the slope suggests that, under the conditions of these experiments, a more or less constant, small amount of iodine is being produced in a form which will penetrate the trapping system. If the recent measurement of the proportion of methyl iodide in the release is held to b representative of the whole series of experiments, the amount of methyl iodide presented to the charcoal increases in these experiments from about 10^{-6} µg to about 2.5 µg. Penetration of 10 to 20% of the assumed methyl iodide component could account for the observed penetrations in the sub-microgram emmisions, but it is then difficult to explain why much larger amounts of methyl iodide are more efficiently retained in the later experiments. However, it may be that the amount of iodine present in other forms can influence the behaviour of the methyl iodide.

The fraction penetrating the system at the lower concentrations was reduced by a factor of between 5 and 10 by increasing the depth of charcoal from 4" to 12".

The highest additional penetration observed in this series of experiments when various clean gas mixtures of CO_2, CO and air were passed through the charcoal bed (at 100°C) for up to 30 hours after the presentation of iodine, was less than 1 part in 10^4 of that presented to the system.

At very low iodine concentrations in the gas (less than 10^{-3} µg/m^3) inconsistent behaviour occurs and some very high penetrations (up to 10%) have been observed. These are not reproducible, however, and are believed to result from reaction of iodine with trace impurities over which the experimenter has no direct control. It is possible that a high proportion of the total iodine is converted to methyl iodide on the occasions when very high penetrations occur.

None of the other hazardous fission products could be detected in significant amounts downstream of the first absolute filter, in these experiments. The measured penetrations of Tellurium-132 do not exceed a few parts in 10^5 of that presented to the system and the vast majority of the released Tellurium is trapped on the absolute filter.

Few measurements have so far been made of Caesium-137 and Strontium-90 penetrations. These showed a Caesium-137 penetration of only a few parts in 10^6 with virtually complete retention on the first absolute filter, whether in CO_2/CO or in air/CO_2. The Strontium-90 release from fuel was so small ($\sim$0.01%) that measurements of its concentration in the gas stream do not have a very high level of reliability. After the absolute filter the concentrations were below the limit of detection, which sets an upper limit to the penetration of about 1% of that presented to the system (1 part in 10^4 of that in the fuel initially).

PRACTICAL DATA ON TRAPPING OF IODINE RELEASED FROM IRRADIATED AGR FUEL

TRAPPING SYSTEM : ABSOLUTE FILTER ⟶ 4" CHARCOAL BED ⟶ ABSOLUTE FILTER

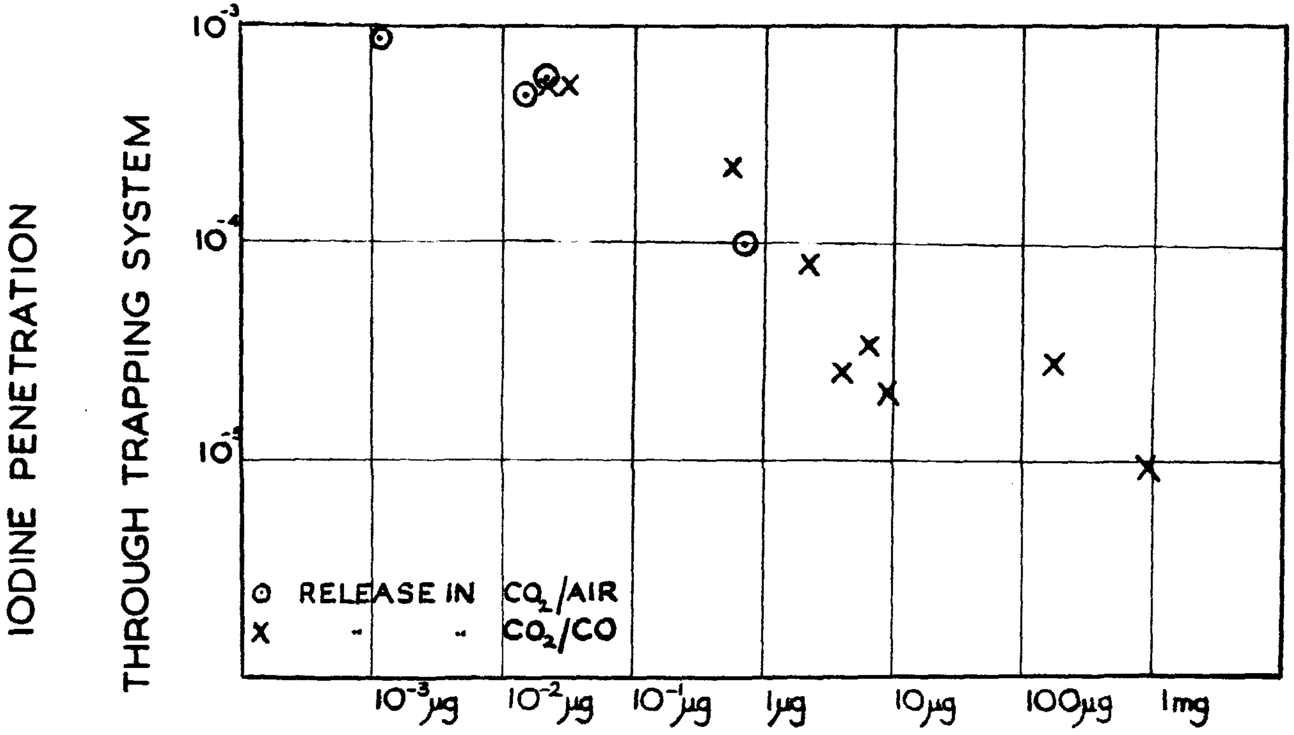


Figure 5

TABLE IV

PENETRATIONS OF IODINE-131, RELEASED FROM IRRADIATED, STAINLESS STEEL CLAD UO_2 AT 1500 TO 1700°C, THROUGH ABSOLUTE FILTER + 4 in. CHARCOAL BED at 100°C + ABSOLUTE FILTER

Average total iodine concentration in gas presented to trapping system $\mu g/m^3$	0.001	0.015	0.02	0.02	0.03	0.05	0.05	0.5	0.6	2.0	4.5	7.1	8.9	196	980
Iodine 131 penetration through trapping system parts in 10^4	9	4	4.5	4.3	4.0	0.7*	0.5*	2	1	0.8	0.2	0.25	0.16	0.42	0.085
Gas composition 50% air/50% CO_2 <u>or</u> 95% CO_2/5% CO	CO_2	air/ CO_2	air/ CO_2	CO_2/ CO	CO_2/ CO	CO_2/ CO	CO_2/ CO	CO_2/ CO	air/ CO_2	CO_2/ CO	CO_2/ CO	CO_2/ CO	CO_2/ CO	CO_2/ CO	CO_2/ CO

*increased depth of charcoal

In the larger-scale highly active test, a much greater quantity of iodine was presented to a trapping system of the same size as that used in the laboratory-scale experiments, giving an iodine loading of about 40 µg/g charcoal.

The iodine was released from irradiated fuel elements by heating them to about 2000°C inside a graphite crucible, supported within a steel pressure vessel which contained a CO_2/CO mixture at 400 p.s.i.g. The walls of the pressure vessel and the bulk of the gas inside it were maintained at about 400°C. Fission products released from the fuel elements were passed to the trapping system either a) directly, to simulate pressure circuit blow-down, or b) via. a containment volume, holding some air, to simulate pressure circuit rupture followed by recirculation. The results showed first, that an unexpectedly high proportion (over 90%) of the released iodine was retained within the pressure vessel or on connecting pipework and secondly, that, of the amount which reached the charcoal bed, a significant fraction penetrated it within a few hours of recirculation.

The penetration in the brief blow-down phase amounted to about 1 part in 10^5 of the iodine initially present in the fuel and about 50 times this amount penetrated the system in the first hour of recirculation during which gas continued to pass over the heated fuel. Fuel heating was then stopped and no more gas was passed through the pressure vessel but gas continued to be recirculated between the containment volume and the trapping system for another 20 hours. During this time iodine continued to arrive at the charcoal bed, presumably by desorption from the walls of the circuit, until it finally reached 5% of the amount originally present in the fuel. In the first 14 hours of this recirculation the penetration amounted to 0.35% of the iodine originally present in the fuel and the last six hours contributed a further 0.07%. A sample of the iodine leaving the bed was identified by gas chromatography as a mixture of organic iodides, containing a predominance of methyl iodide together with ethyl iodide and higher alkyl iodides.

It is believed that conditions within the pressure vessel encouraged the growth of considerably larger particles than are formed in the laboratory experiments and that these then settled out on surfaces in the pressure vessel carrying much of the iodine with them, which would account for the retention of much of the iodine in the pressure vessel. However, two explanations are possible for the high penetration of the trapping system by the iodine that was not retained in the pressure vessel. The first is that, if the release is assumed to contain the same proportion of methyl iodide as that observed in a recent laboratory experiment, the amount of methyl iodide presented to the charcoal bed would have been about 50 µg, and this may have been sufficient to cause breakthrough. The second possibility is that reactions taking place between the iodine deposited on surfaces and organic impurities present in the gas or on the surfaces, may have resulted in the conversion of a much larger fraction of the iodine to organic iodides, part of which, subsequently penetrated the charcoal bed.

Both explanations suggest that in certain conditions of reactor accidents, sufficient methyl iodide or similar organic iodides may be formed to result in penetration of the installed charcoal beds. It may therefore prove necessary to develop alternative trapping plant, capable of retaining alkyl iodides.

Deposition behaviour of iodine released from irradiated fuel elements

After the release of iodine from the fuel elements its retention by deposition within the reactor core and containment could provide a mechanism by which the amount available to leak to atmosphere is reduced.

TABLE V

DISTRIBUTION OF IODINE FORMS RELEASED ARTIFICIALLY FROM UNIRRADIATED FUEL ELEMENTS AT 1500-1700°C, AND THE PENETRATION OF THE RELEASED IODINE THROUGH THE STANDARD TRAPPING SYSTEM

Gas Composition		CO_2					50% air/CO_2	
Iodine concentration in gas $\mu g/m^3$		0.03	0.05	0.1	15	2.6×10^4	0.05	0.1
Distribution of iodine on May Pack components	Copper	12	9	7	12	69	90	91
	Millipore	75	83	75	70	27	5	4
	Charcoal paper	6.5	4	6	16.5	3.9	4	4.5
	Charcoal granules	6.5	4	12	2.5	0.1	1	0.5
Penetration of iodine through trapping system parts in 10^4		1	0.3	3	0.9	0.5	0.5	0.1

On the other hand, as we have seen in the highly active test , its deposition on surfaces could result in the production of a high proportion of the penetrating alkyl iodides.

Measurements have been made of the rate of deposition of iodine released from irradiated miniature fuel elements heated at 1500 to 1700°C in a fairly rapid flow of 95% CO_2/5% CO, both in a long cold mild steel pipe with many bends, and on to cold flat specimens of mild steel, stainless steel, graphite and concrete. The measured velocity of deposition on to the cold flat surfaces at a gas velocity of $\sim$100 cm/min. was in the order of 10^{-3} cm/sec., while on graphite maintained at 400°C (a tubular specimen with the gas passing through it) the deposition velocity was a factor of 10 higher. Only about 3% of the total gas-borne iodine deposited on 33 feet of cold mild steel pipe at a gas velocity in the pipe of $\sim$70 cm/sec., which again corresponds to an overall deposition velocity in the order of 10^{-3} cm/sec.

These measurements are consistent with the absence from the release of elemental iodine vapour, which would have been expected to have a deposition velocity in these conditions at least two orders of magnitude higher, and with the predominance of iodine in the forms of (a) particle agglomerates with a mean frequency size of about 0.05 microns and (b) relatively unreactive vapour compounds. They are not, however, consistent with the high retention of iodine in the pressure vessel in the highly active test. It is considered that the higher pressure in this experiment, the higher concentration of vapourized fuel element material and the virtually static gas surrounding the heated fuel elements must inevitably have resulted in the production of much larger particles than have been observed in the laboratory-scale experiments. Such particles are likely to have settled out in the pressure vessel, carrying much of the released iodine with them to the walls.

The deposition behaviour to be expected in a reactor accident cannot yet be predicted. In the case of a blocked channel accident it seems likely that behaviour similar to that observed in the highly active test would occur, with much of the iodine being retained within the channel; but of that which escaped, an appreciable proportion might be in the form of penetrating alkyl iodides. In the case of a pressure circuit rupture, much would depend on the gas flow that could be maintained through the core and the temperature reached by the fuel elements.

Experience with iodine generated artificially

Work with irradiated fuel elements is slow and difficult and can only be done on a large scale at great expense. It is therefore very desirable to be able to simulate the behaviour of iodine released from fuel by artificial methods of generation. A technique used at Windscale is to deposit carrier-free iodine-131 (or with a known amount of stable iodine) inside a short length of stainless steel fuel can, to add unirradiated UO_2 pellets and then to reseal the can. The can is then heated in the same way as an irradiated specimen and iodine is released when the can melts.

When the specimen is heated in a rapid flow of CO_2 in glass apparatus, the distribution of iodine forms and the penetration of iodine through the trapping system are both closely similar to those obtained from irradiated fuel in similar conditions, although there is not a pronounced difference in penetration over a wide range of concentrations. In the presence of air the distribution is qualitatively similar to that obtained from irradiated fuel, but the proportion of elemental iodine vapour is rather higher and that of particulate iodine rather lower (Table V).

The identity of the iodine compounds which penetrate to the charcoal components of the May Pack is not known at present, but they have recently been shown in one experiment to contain some alkyl iodides.

In view of the association of a relatively high penetration of the trapping system in the highly active test with the presence of a relatively high concentration of alkyl iodides, it is of interest to discover under what conditions the production of alkyl iodides from other forms of iodine can be enhanced.

Eggleton and Atkins at AERE (3) were the first to identify alkyl iodides in various iodine emissions used for testing trapping systems and showed that it is formed in small amounts when milligram quantities of iodine crystals are dispersed in very large volumes of air; when carrier-free NaI^{131} is oxidised in air; and when fission product iodine-131 is released from irradiated uranium wire at about 800°C in air.

We have found a variety of conditions in which up to 10^{-3} μg to 10^{-2} μg iodine can be produced in the form of alkyl iodides. These include

a) iodine deposited from CS_2 solution on ultra-clean glass-ware and heated in argon or helium at temperatures in the range 100°C to 400°C.

b) iodine deposited from aqueous sodium iodide solution on ultra-clean glassware and heated in argon or helium at temperatures from 200°C to 400°C.

c) iodine deposited from CS_2 solution or from aqueous sodium iodide solution on stainless steel surfaces and heated in inert gas, air or carbon dioxide/carbon monoxide atmospheres at temperatures from 400°C to ~1300°C.

d) iodine attached to particles released from unirradiated fuel element components at about 1800°C, captured on a stainless steel sinter and subsequently desorbed from this surface in air or argon at about 500°C.

In each of these cases, when the amount of iodine added was in the order of 10^{-3} to 10^{-2} μg, or less, 50% or more of the iodine was found in the form of alkyl iodides (chiefly methyl iodide) but when amounts of stable iodine equal to 10 μg or more were added the proportion present as alkyl iodides was so small that it could no longer be detected.

The readiness with which methyl iodide is formed at very low iodine concentrations makes it tempting to ascribe the unreproducible high penetrations, which have been observed at these levels, to its presence. However, as pointed out earlier in this paper, it is difficult to reconcile this idea with the detection of much greater amounts of methyl iodide in the gas presented to the charcoal bed in a recent experiment in which no measurable penetration was detected, unless the retention of small amounts of methyl iodide on charcoal is dependent upon the presence of a sufficient quantity of other, more easily retained, forms of iodine. So far no method has been found of producing percentage quantities of alkyl iodides artificially in an emission anywhere near as concentrated as that which emerged from the pressure vessel in the highly active test (several hundred $\mu g/m^3$ in the gas).

Whatever the source, however, it seems likely that sufficient alkyl iodides were produced in the highly active test to cause penetration of the 4" deep charcoal bed. Since their production to a similar degree in reactor accidents cannot be excluded, it is prudent to develop an improved method of trapping them.

Methyl iodide trapping

Methyl iodide is produced in much larger quantities than any other alkyl iodide and is likely to be the most intractable. Experiments are therefore being carried out with iodine-131-labelled methyl iodide to find a suitable trapping method for reactor application.

It has been illustrated that, under the same gas flow conditions as used in the fission product trapping experiments, milligram quantities of methyl iodide penetrate a 4 " deep by 1" diameter bed of SCII charcoal in a matter of minutes when the bed is at 100°C and in about one and a half hours when the bed is at 20°C. Further experiments are in hand to discover whether retention occurs at some lower level of methyl iodide loading.

The search for alternative methods of methyl iodide trapping is at present based on a) methods involving oxidation b) methods involving hydrolysis c) methods involving specific reactions in organic media. Only methods in category a) have so far received significant attention.

It has been found that the commercial preparation HOPCALITE (basically MnO_2+CuO) at 100°C appears to retain about 90% of the methyl iodide presented to it in 100% CO_2. Most of the iodine which penetrates the HOPCALITE is in the form of molecular iodine and is retained on SCII charcoal at room temperature. About 99.9% of the methyl iodide was retained by the two beds in series.

In the presence of carbon monoxide, however, the HOPCALITE'S efficiency is reduced and only about 50% of the methyl iodide is retained during a few hour's passage of 5% CO/CO_2. The methyl iodide which is not adsorbed on HOPCALITE is unchanged chemically and is only retained for an hour or so on the charcoal bed. The presence of carbon monoxide is almost inevitable in any accident to an AGR so that these observations are not encouraging.

The use of a silver-coated copper knitmesh bed at 300°C in CO_2 had no effect on the behaviour of methyl iodide.

50% retention of about 100 mg. methyl iodide has been found on passage through SCII charcoal held at about 200°C. This suggests that oxidation can occur at some sites on charcoal and lends hope that a charcoal of higher ignition temperature may be even more effective above 200°C. It also raises the possibility that rather higher penetrations of small amounts of methyl iodide might be observed through a charcoal bed maintained at room temperature than through one allowed to operate at 100°C, as in the experiments described in this paper.

CONCLUSIONS

The present form of fission product trapping plant, consisting of a combination of absolute filters and a bed of activated charcoal, provides adequate protection against the escape of dangerous fission products from the Advanced Gas-Cooled Reactor, with the possible exception of iodine-131 in the form of alkyl iodides.

Pending further elucidation of the mechanism and extent of formation of alkyl iodides and of conditions which may enhance their production, it is prudent to develop a specific method of trapping them.

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3. A. E. J. EGGLETON, and ATKINS, D. H. (A.E.R.E.) Identification of radio-iodine compounds formed on releasing carrier-free I-131 in air. Paper given at 1963 Annual Meeting of American Nuclear Society at Salt Lake City . (also AERE-M1211).

DISCUSSION AND COMMENT

We have thought about decomposing the methyl iodide with high temperature and one of the first experiments tried was to pass the methyl iodide through a heated bed of alumina, about 400° C. to give an extended surface for the reaction to occur. Although some decomposition occurred, it only gave a factor of about five. We are looking at oxidative catalysts which are showing much more promise, and one of these, if I can just enlarge on this point now, is hopcalite, with which you are probably familiar and it does seem to be particularly effective in decomposing methyl iodide. From our point of view one snag is that the presence of carbon monoxide, which is almost inevitable in any graphite moderated reactor, seems to interfere. We have some evidence this can be overcome provided the carbon monoxide is first reduced to a very low percentage, and then enough air is added to the gas to the equivalent of the carbon monoxide, so that the hopcalite is not used up, and is therefore free to continue to decompose methyl iodide. But these results are still preliminary, and we do not regard this as the most attractive system.

At the Salt Lake City American Nuclear Society Conference earlier this year, a Harwell Colleague, Dr. Eggleton, described the manner in which he discovered that the iodine was in the form of methyl iodide. We extended the use of the method to analyze the gas obtained from the release of fission products from irradiated uranium dioxide, and we found the same peaks. In fact, we sent a sample of our gas to him and he confirmed our result by his method; we were happy that these cross-checks confirmed that the material is in fact methyl iodide. But, it is only a small constituent of the total iodine release. There are more major constituents of the release which are not elemental and which are not particulate, and which we have not yet identified. It is very important that we should have information on these, particularly if we wish to design on a rational basis trapping systems which do not depend upon activated charcoal. The only reason I have not enlarged upon this form of iodine is that activated charcoal does seem to be very effective in stopping it. But activated charcoal beds may not be the most appropriate form of reactor trap in every reactor system, and we would like to know all the iodine compounds with which we have to deal.

Session Chairman: Thank you, Mr. Collins.

Our next paper was prepared by Robert B. Boise, of the National Lead Company of Ohio, and is titled, "Exhausting Corrosive Fumes with Quick Draft Eductors."

EXHAUSTING CORROSIVE FUMES WITH QUICKDRAFT EDUCTORS

Robert B. Boies

National Lead Company of Ohio
Cincinnati, Ohio

1. ABSTRACT

The uses of Quickdraft eductors in handling and diffusing various types of corrosive fumes are described. A number of specific applications are discussed and illustrated.

2. INTRODUCTION

In the processing of uranium compounds to metal and the fabrication of uranium metal to fuel cores, many noxious and corrosive fumes are evolved. The Laboratory, Pilot Plant, and production areas, all generate these fumes, which include nitric, perchloric, hydrochloric, and hydrofluoric acid vapors; nitrous and nitric oxide; sulfur dioxide; and ammonium chloride fumes. The fumes are passed through electrostatic precipitators, dust collectors, scrubbers, and other devices to render them harmless before release to the atmosphere.

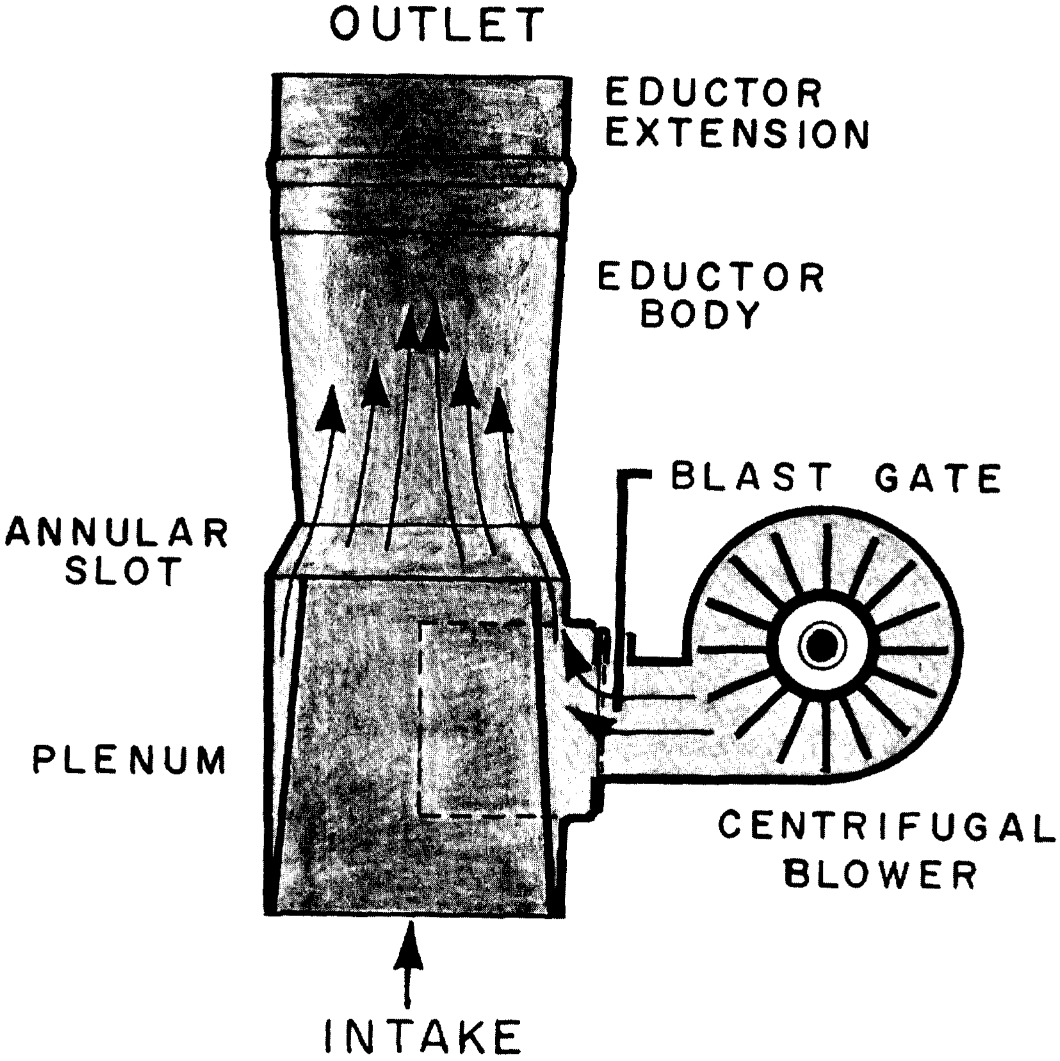
Air eductors have many advantages over conventional exhaust fans in handling such corrosive fumes. These advantages will be described.

3. DESCRIPTION OF EQUIPMENT

The eductors used at the National Lead Company of Ohio for corrosive service are manufactured by the Quickdraft Corporation of Canton, Ohio. This eductor is a unique, patented type, operating on a modification of the venturi principle. Air is introduced by a blower into the eductor through a slotted annulus. This can be seen in the cutaway drawing, Figure 1. This slotted annulus is the principle characteristic by which this unit differs from a regular venturi using a jet or nozzle for its motive force. The principal parts of the eductor are:

1. The eductor body, which is designed for the air flow needed.
2. The eductor extension, which increases the efficiency of the eductor.
3. The plenum, which acts as a distributor for the blower air to the annular slot.
4. The annular slot, through which air is discharged around the periphery of the eductor.
5. The blast gate, which controls the air flow from the blower.

Quickdraft eductors are available in a variety of sizes, with volumes ranging from 100 cubic feet per minute to



QUICKDRAFT EDUCTOR

CUTAWAY VIEW

Fig. 1

90,000 cubic feet per minute and static pressures from 0.02 inches of water to 140 inches of water. Figure 2 is a graph showing the performance of a standard 6-inch-diameter Quick-draft eductor at various blower horsepower. As can be seen, a wide selection of volumes and static pressures are available. By installing a blast gate at the blower outlet any desired combination of pressure and volume on a given horsepower curve may be attained. Models are available in black iron, vitreous enameled steel, stainless steel, hastelloy, fiberglass- or dynel-reinforced polyester or epoxy resins, and unplasticized polyvinyl chloride, or other plastic coatings designed for use in hot and corrosive atmospheres.

4. PRINCIPLE OF OPERATION

1. The annular method of discharging the blower air into the eductor causes a velocity pattern different from that in a normal venturi (see Figure 3). In the Quick-draft eductor, air velocity is greater at the outer periphery than at the center of the unit, just the opposite of the velocity pattern in a regular venturi. Two significant gains are realized from this phenomenon: (1) corrosive fumes are kept essentially in the center of the exhaust system, away from possible condensation and corrosion on the walls; and (2) in material handling operations, the amount of abrasion is greatly reduced. This action can be demonstrated simply by holding a string at the intake of one of these units and watching it progress from intake to outlet. This is shown in Figure 4, a clear plastic model of an eductor unit.

Performance Curves for Std. 6" Dia.

QUICK DRAFT EDUCTOR

at Various Blower H.P.

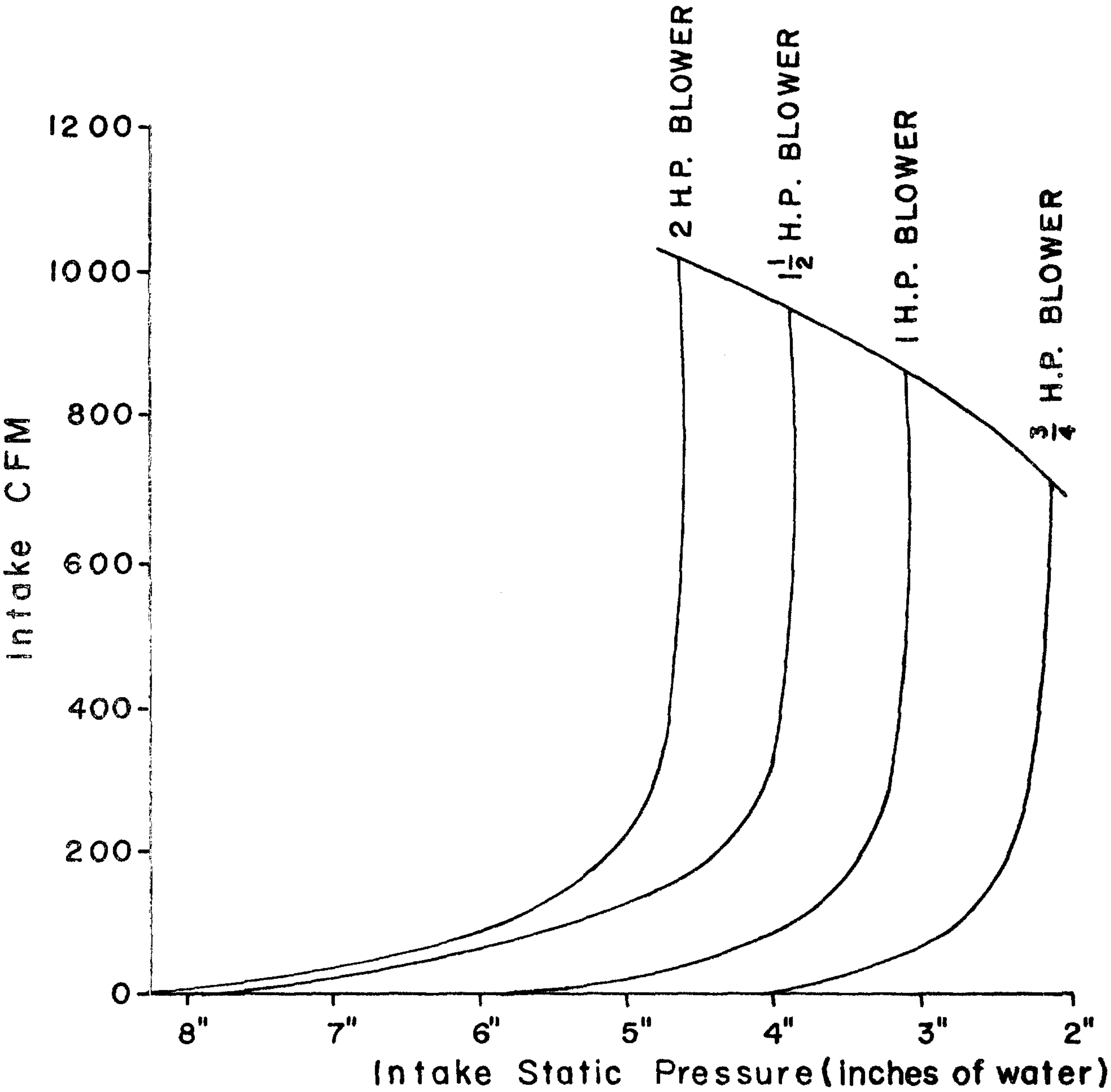
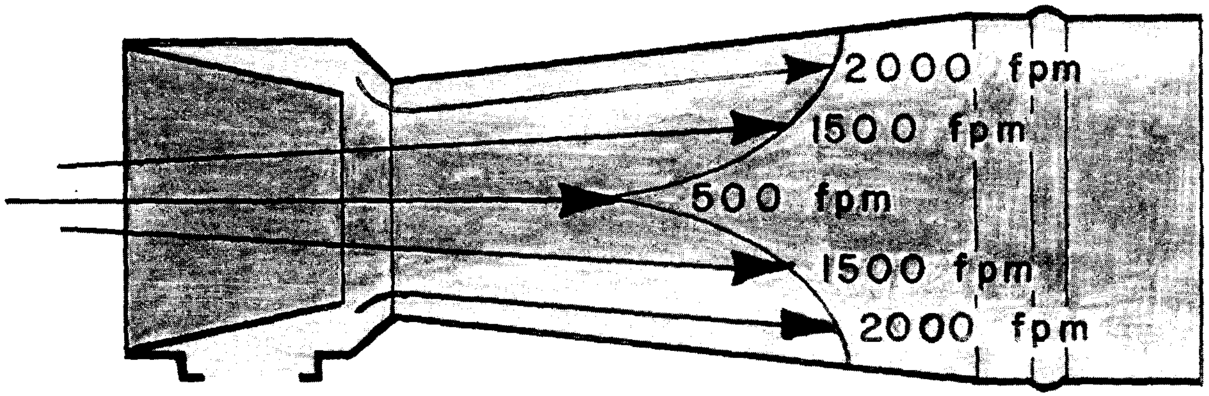


Fig.(2)



VELOCITY PATTERN INSIDE QUICKDRAFT UNIT

Fig. 3

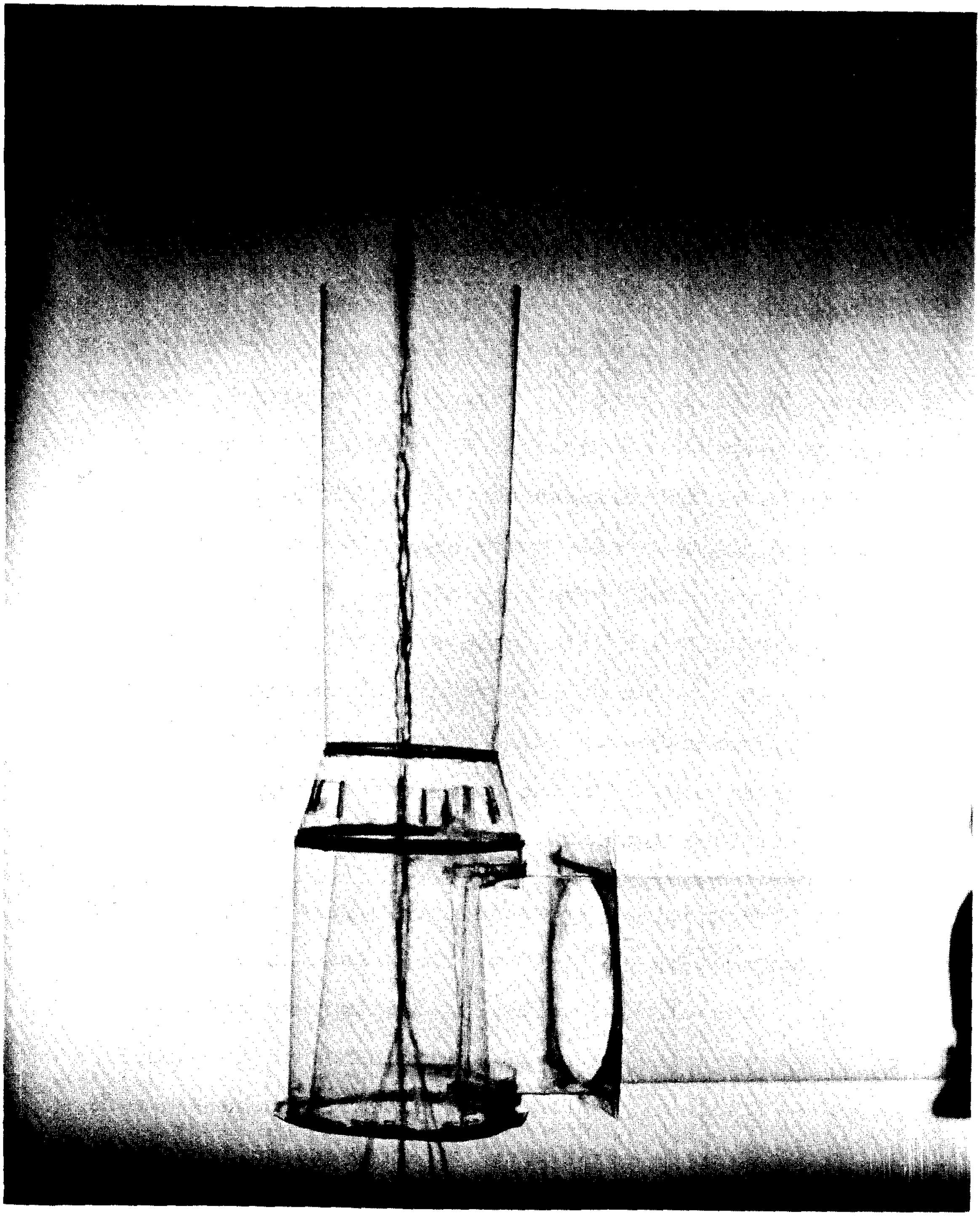


FIGURE 4 Plastic Operating Model of Quickdraft Unit; Yarn Shows Path of Intake Air

We have not yet had an opportunity to use the Quickdraft eductors in material handling applications at this site. However, I have seen these units in use at several paper mills, an aluminum sheet manufacturer, and a mylar thread manufacturer. In all of these installations, the principle of the material being transported in the center of the air stream is employed to great advantage.

5. GENERAL USAGE

One of the greatest problems in a chemical plant is disposal of fumes. Dilution into the atmosphere is usually the most economical method. The Quickdraft unit is particularly adapted to such fume dispersion. Heavy white ammonium chloride and nitrous oxide fumes emanating from a chemical operation in our plant have been almost completely dispersed by use of a Quickdraft unit as an exhauster. Thus plume effect from an exhaust duct is either eliminated or greatly reduced. The unit has an added advantage in that it atomizes large liquid particles in the gas stream, thereby speeding their vaporization into the atmosphere.

The ratio of air intake at the eductor to air intake at the blower is quite high at lower static pressures. At eductor intake pressures of 0.02 inches to 2.5 inches of water, the ratio is approximately 4 to 1; at 10 inches to 15 inches of water, the efficiency of the unit drops to around a 1 to 3 ratio. This drop in efficiency, which means an increase in the relative amount of blower air in the unit's exhaust, is put to good advantage in the dilution and dispersion of exhaust fumes, especially ammonium chloride and hydrogen. Cooling of hot exhaust gases is yet another advantage.

6. APPLICATIONS AT THE NATIONAL LEAD COMPANY OF OHIO

Described and illustrated on the following pages are a few of the installations of these eductors at our plant. In one interesting application a Quickdraft unit is used to exhaust a hydrogen-rich atmosphere. In this particular installation, an emergency source of power for the eductor is required for safety. This emergency supply is provided by 150 psi steam connected into the eductor plenum (see Figures 5 and 6).

These eductors are also used at this site to exhaust laboratory fumes. Perchloric acid fumes, which are particularly difficult to handle, are exhausted by Quickdraft eductors (see Figure 7).

In Figure 8, is shown an application of a Quickdraft unit in a forced-air furnace exhaust through a scrubber. A problem was encountered in this application, in that ammonium chloride was being carried over from the furnace through the caustic scrubber. This material flash-dried in the eductor until the throat was completely plugged. By introducing a warm water spray into the driven air stream (see Figure 9) this buildup was eliminated.

The solution of the above problem also provided us with a possible means of mixing and atomizing a liquid or solid into a gas stream for production blending purposes.

In Figures 10 and 11 are shown Quickdraft units which are dissipating the hot exhausts from three corrosive scrubbers, two of which exhaust streams include HF fumes.

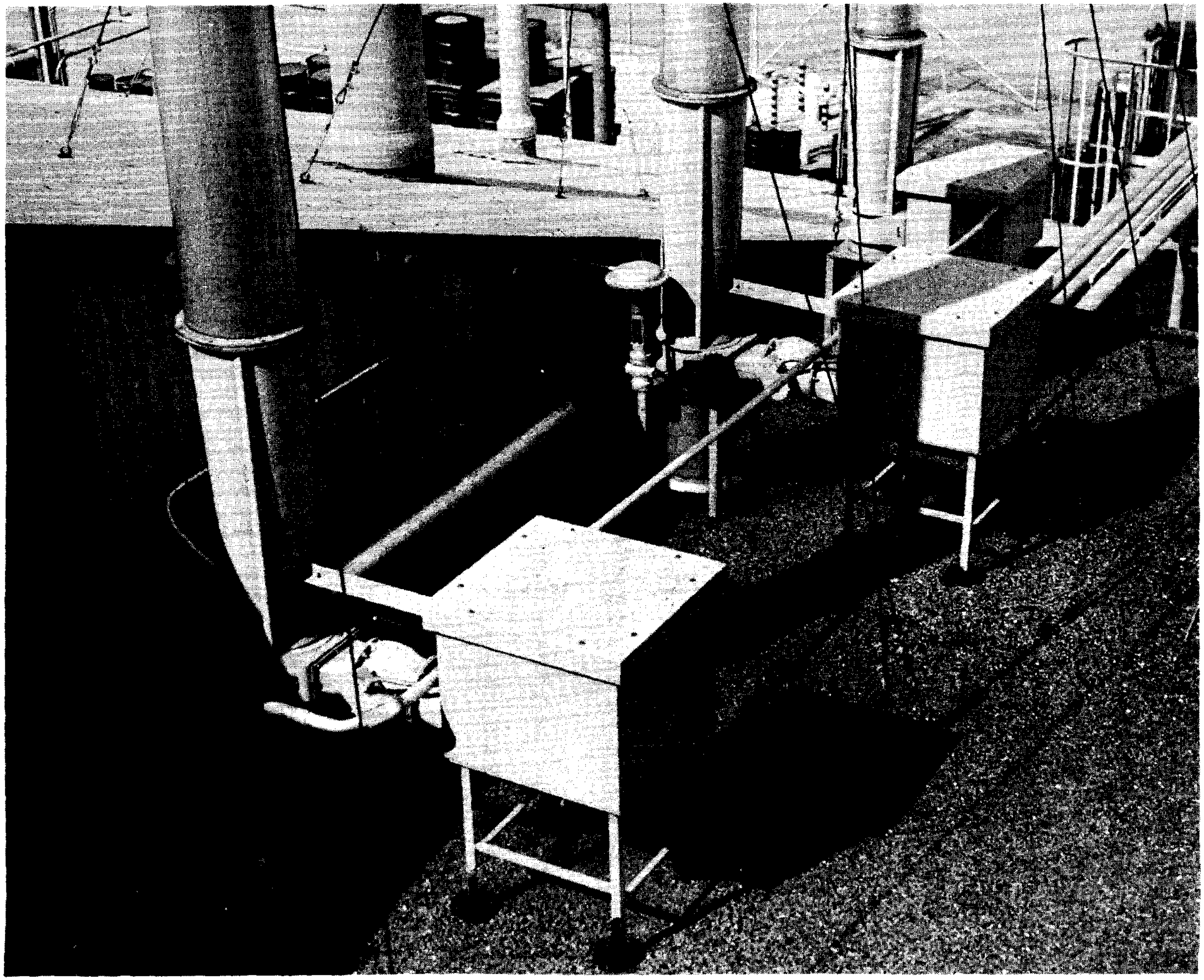


FIGURE 5 Roof-Mounted Eductors Exhausting Hydrogen-Rich Fumes

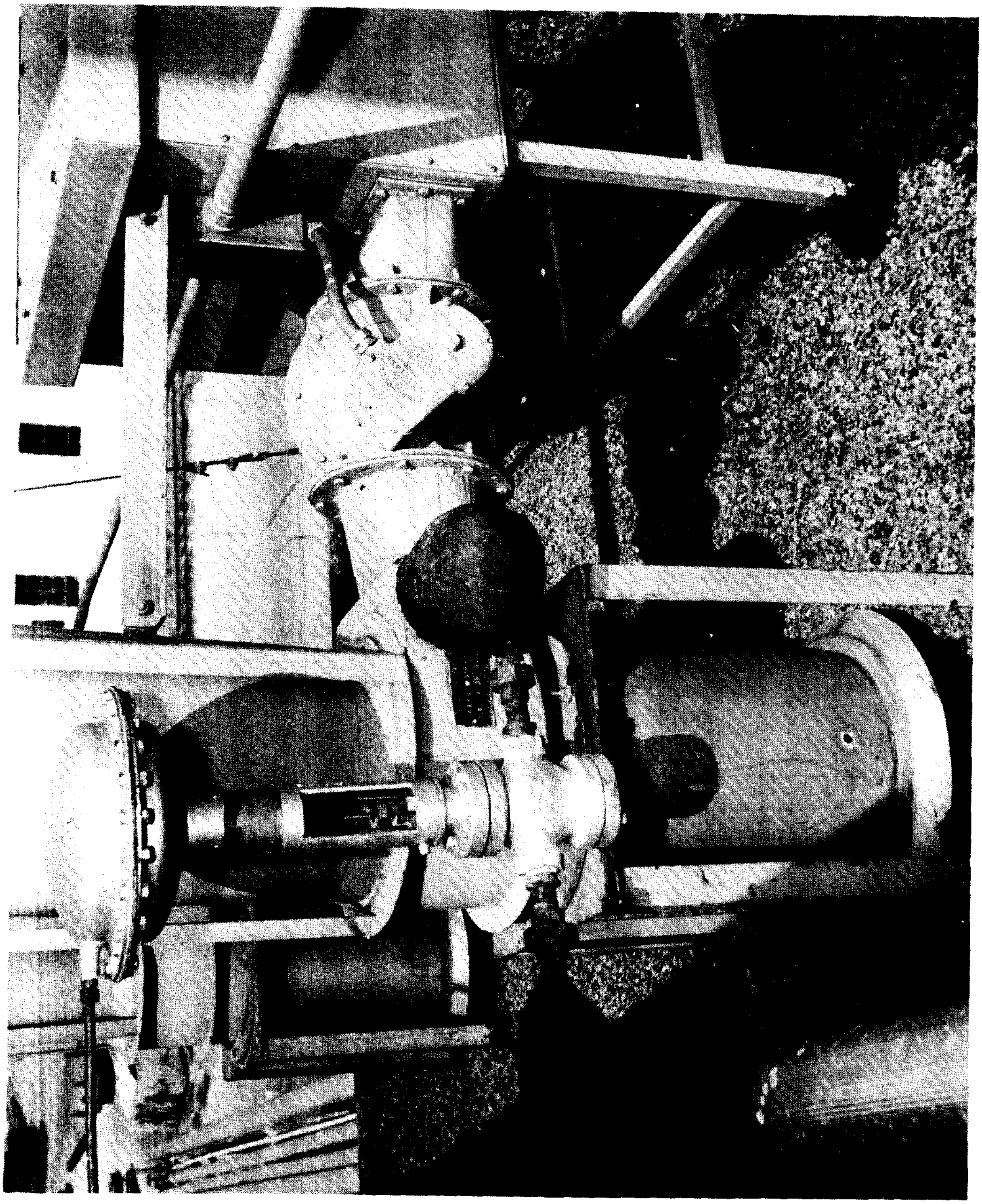


FIGURE 6 Close Up of Emergency Steam Supply for Hydrogen Exhaust Eductor

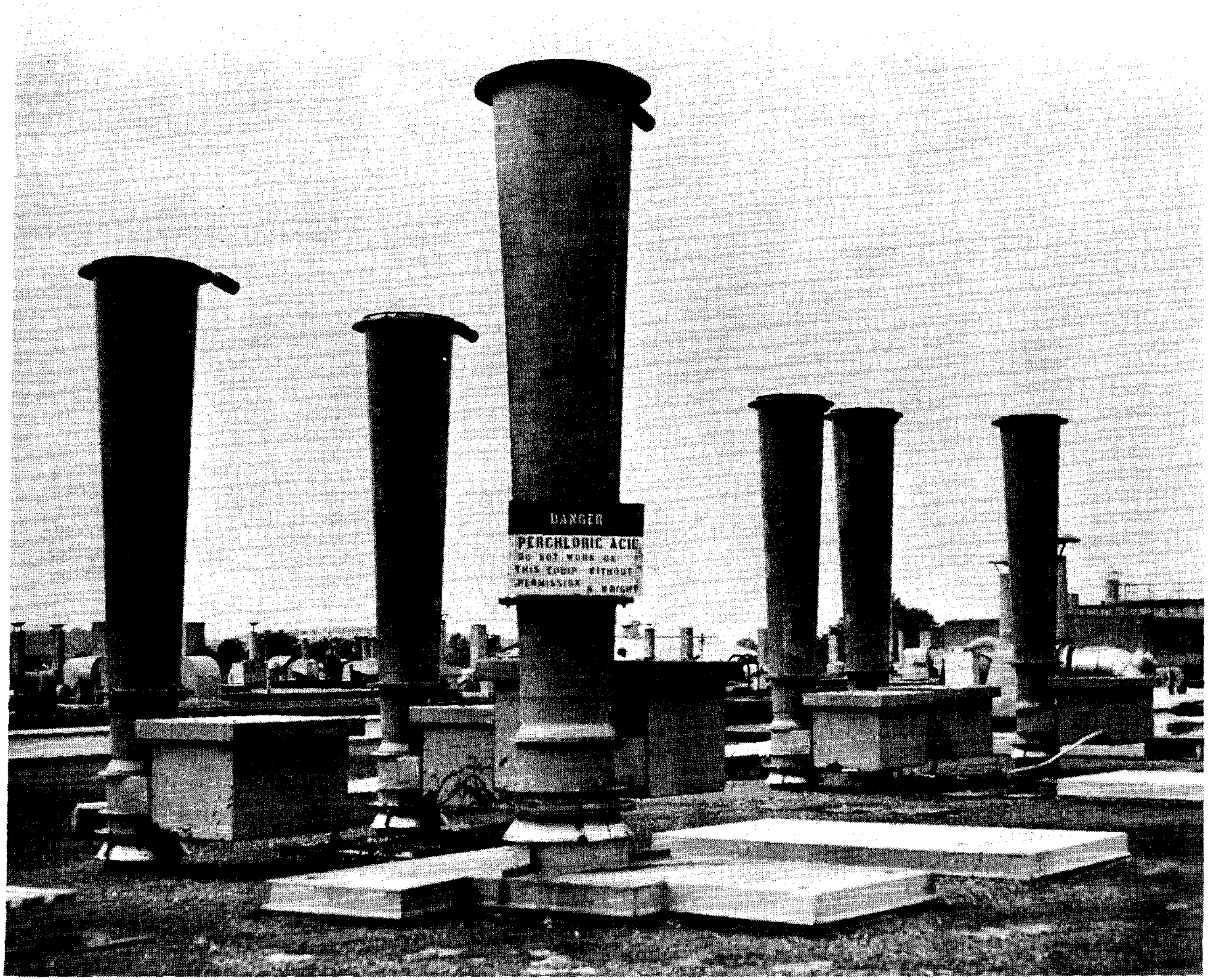


FIGURE 7 Quickdraft Eductors on Roof of Laboratory

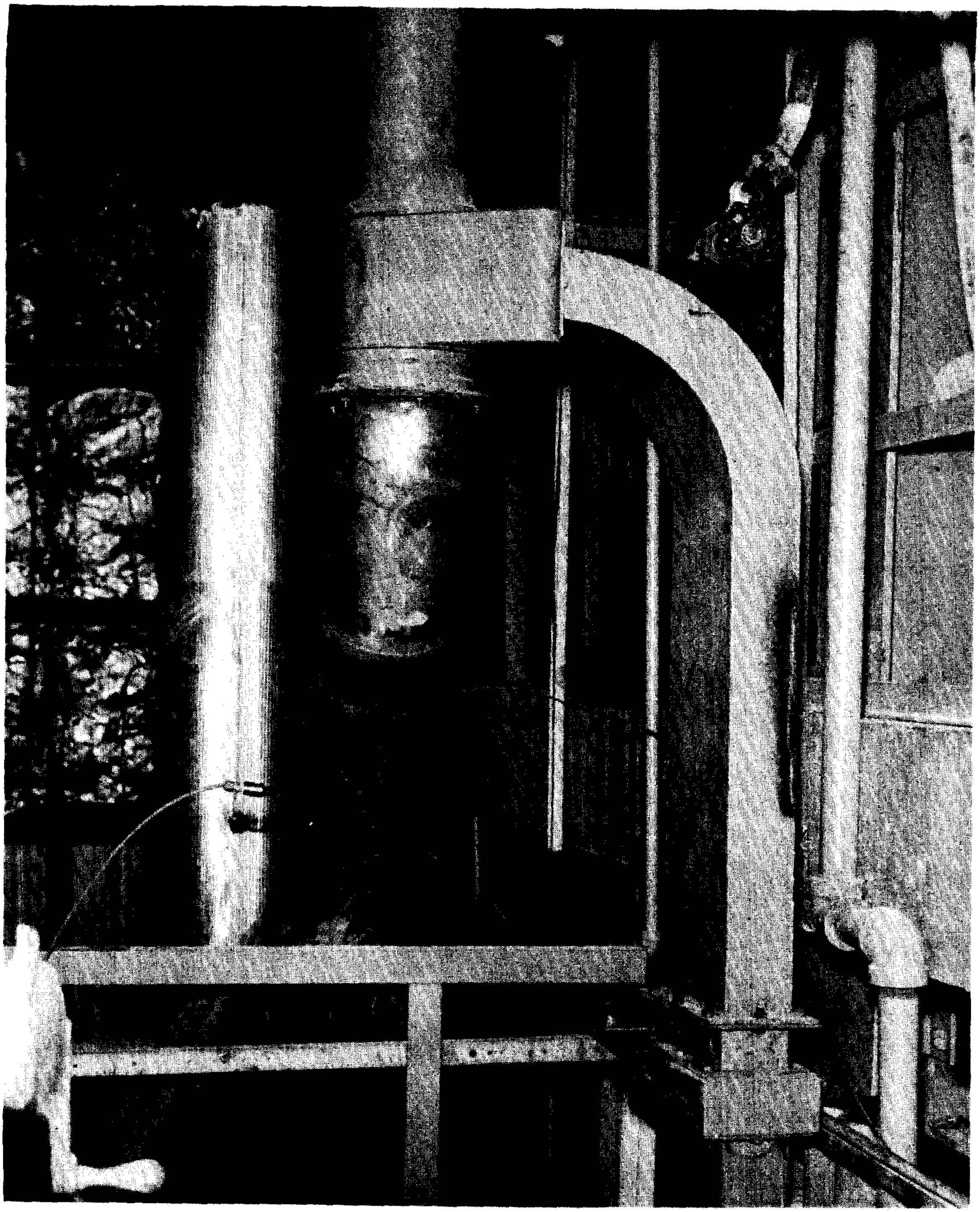


FIGURE 8 Furnace Scrubber Exhaust Eductor Showing Remote Blower Mounting

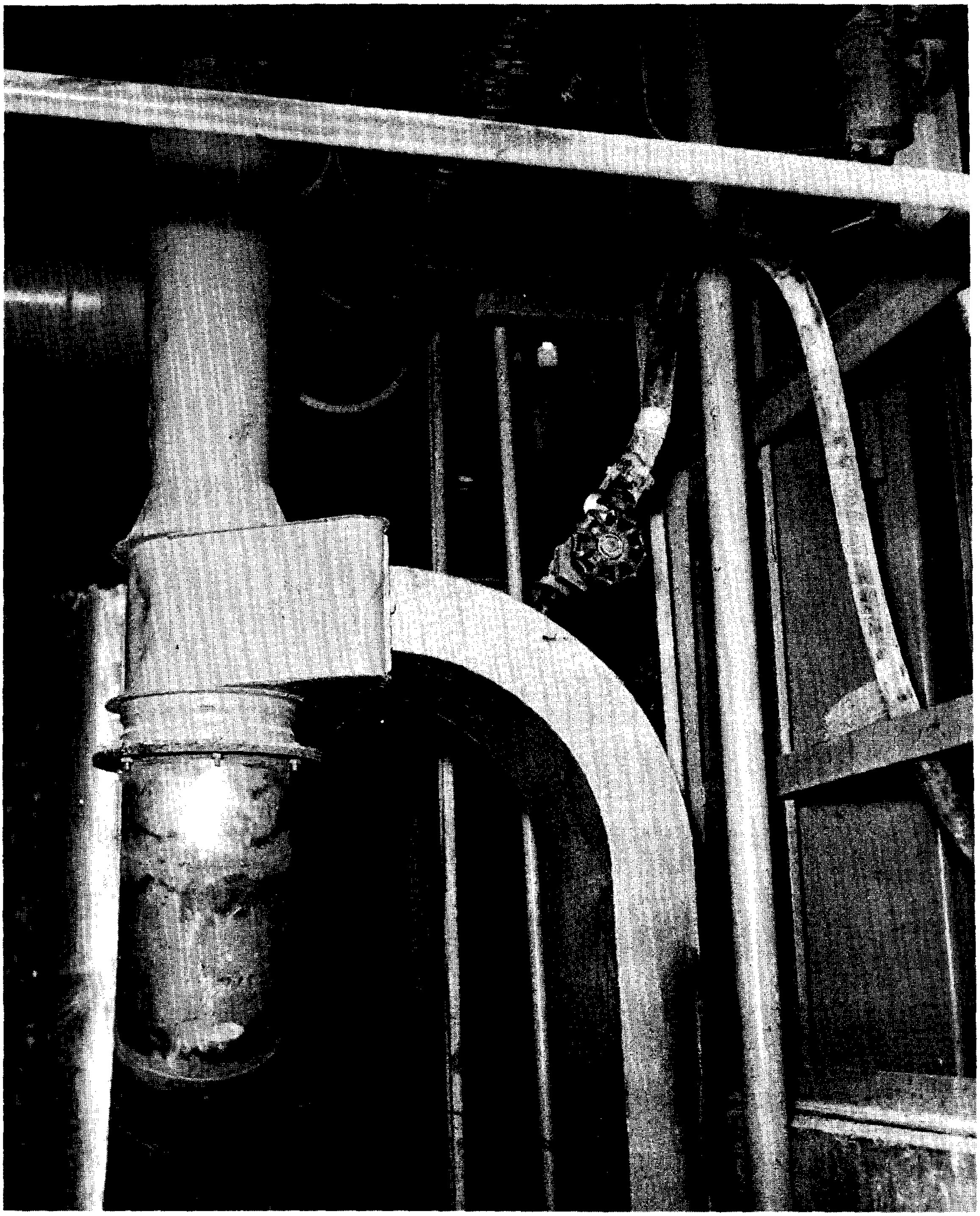


FIGURE 9 Close Up of Eductor Showing Warm Water Valve at Blower Discharge

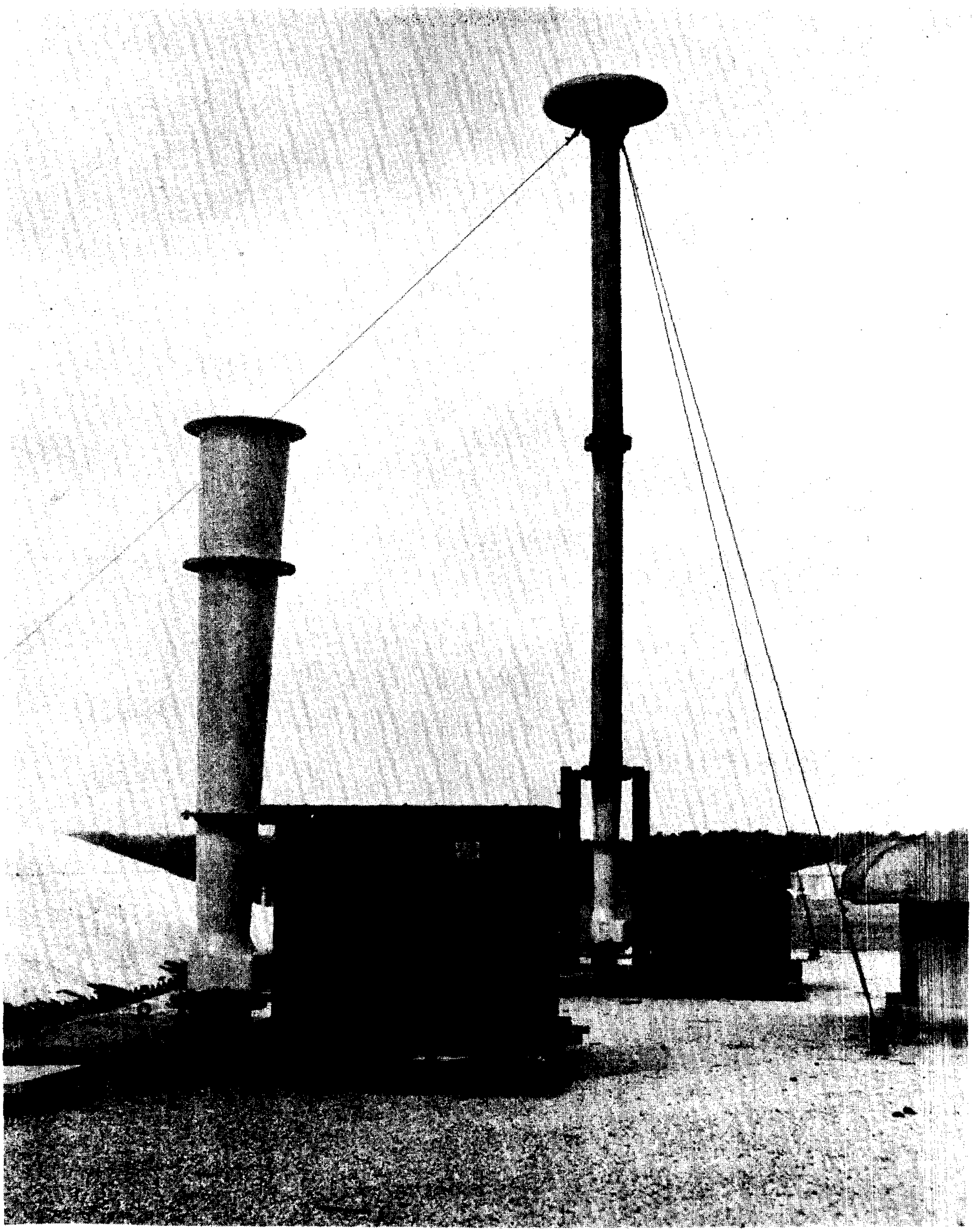


FIGURE 10 Corrosive Scrubber Exhausts Using Dynel-Reinforced Polyester Resin Quickdraft Eductors and Ductwork

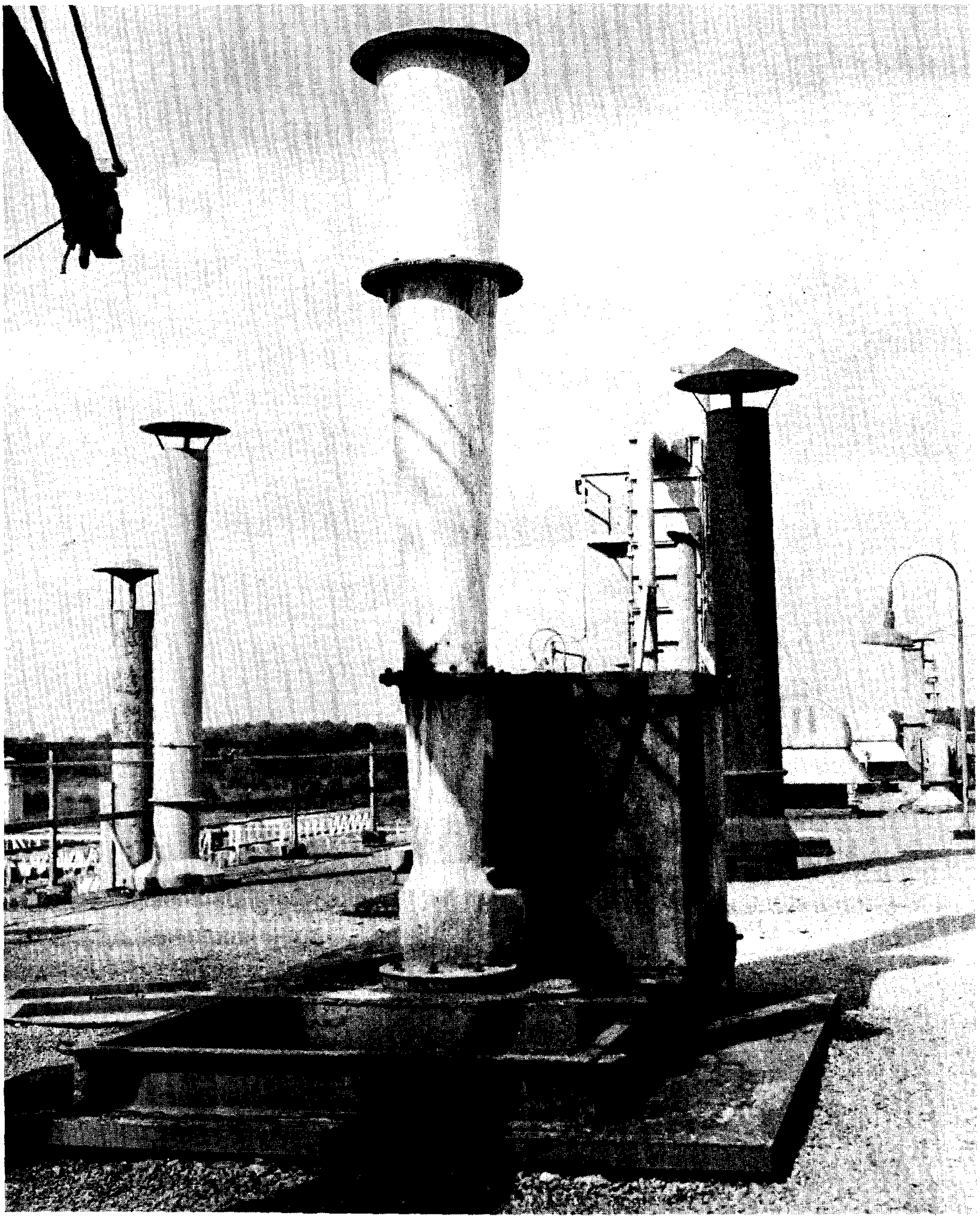


FIGURE 11 Corrosive Scrubber Exhaust Using Fiberglass-Reinforced Polyester Resin Quickdraft Eductor and Ductwork

Prior to the installation of the Quickdraft units, these scrubbers were exhausted by centrifugal blowers with plastic wheels. These wheels required replacement approximately every three weeks. The Quickdraft units have eliminated the cost of replacing these units as well as expensive downtime. To resist the very corrosive HF, the units are constructed of Duverre 22 polyester resin reinforced with dynel cloth.

Figure 12 shows a small Quickdraft eductor venting a zirconium decladding tank. This unit has outlasted three blowers in this installation.

The final application of these units to be discussed is shown in Figure 13. In this application the eductor is being used to exhaust corrosive fumes from Eimco filters. The units are fabricated fiberglass-reinforced polyester resin. The sharp elbow in the duct immediately below the Quickdraft unit is not recommended practice, but it does permit the use of the eductor in close quarters.

7. CONCLUSIONS

I have attempted to point out in this paper the numerous advantages of this unit as a ventilation source with special emphasis on its usage in handling corrosive and noxious fumes where all moving parts of the air mover must be kept out of the corrosive stream. The Quickdraft eductor is not a replacement for exhaust fans and blowers but, rather, is a valuable new tool to be used by air pollution and environmental control

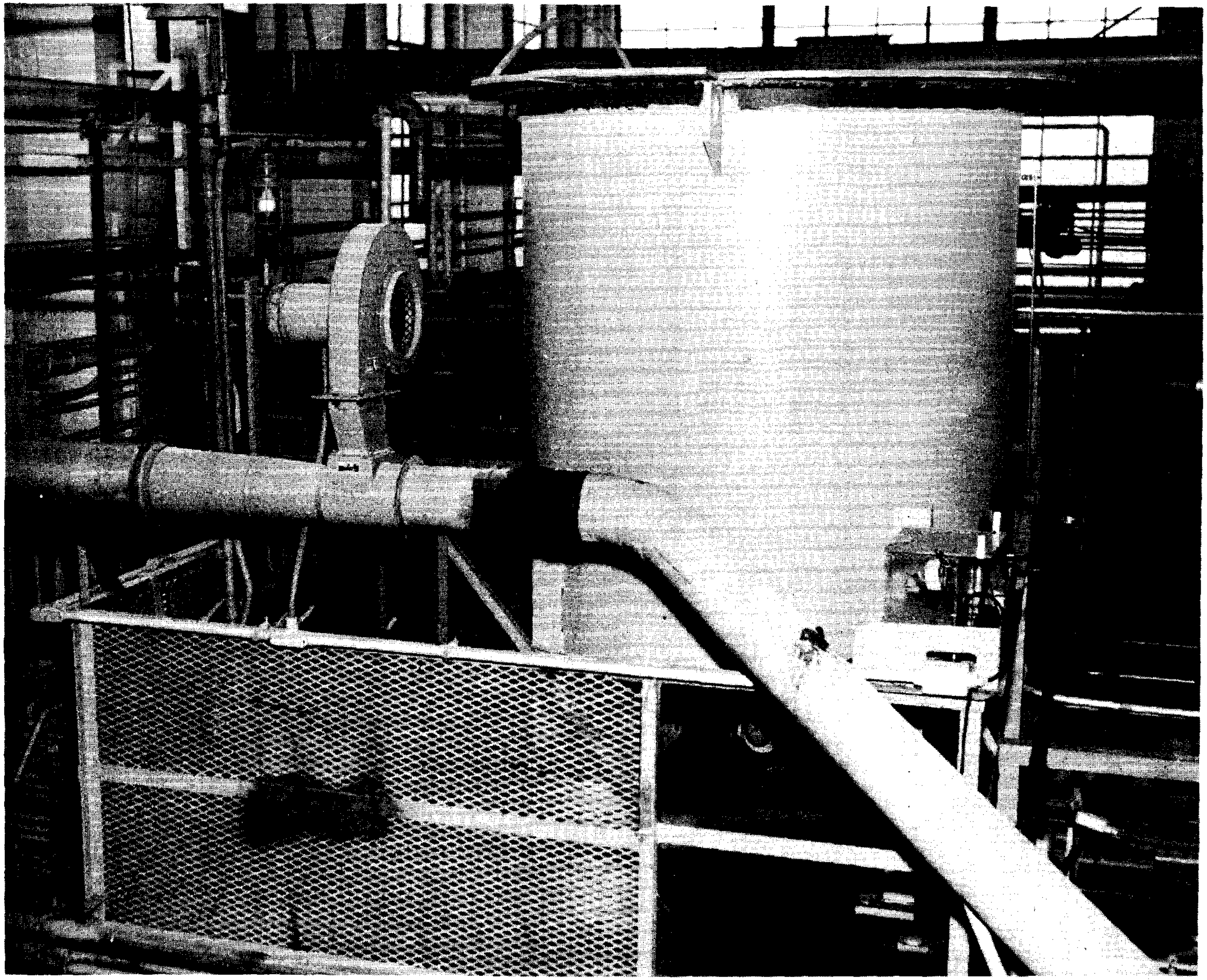


FIGURE 12 Zirconium Decladding Tank Exhaust; Horizontally Mounted Quickdraft Unit

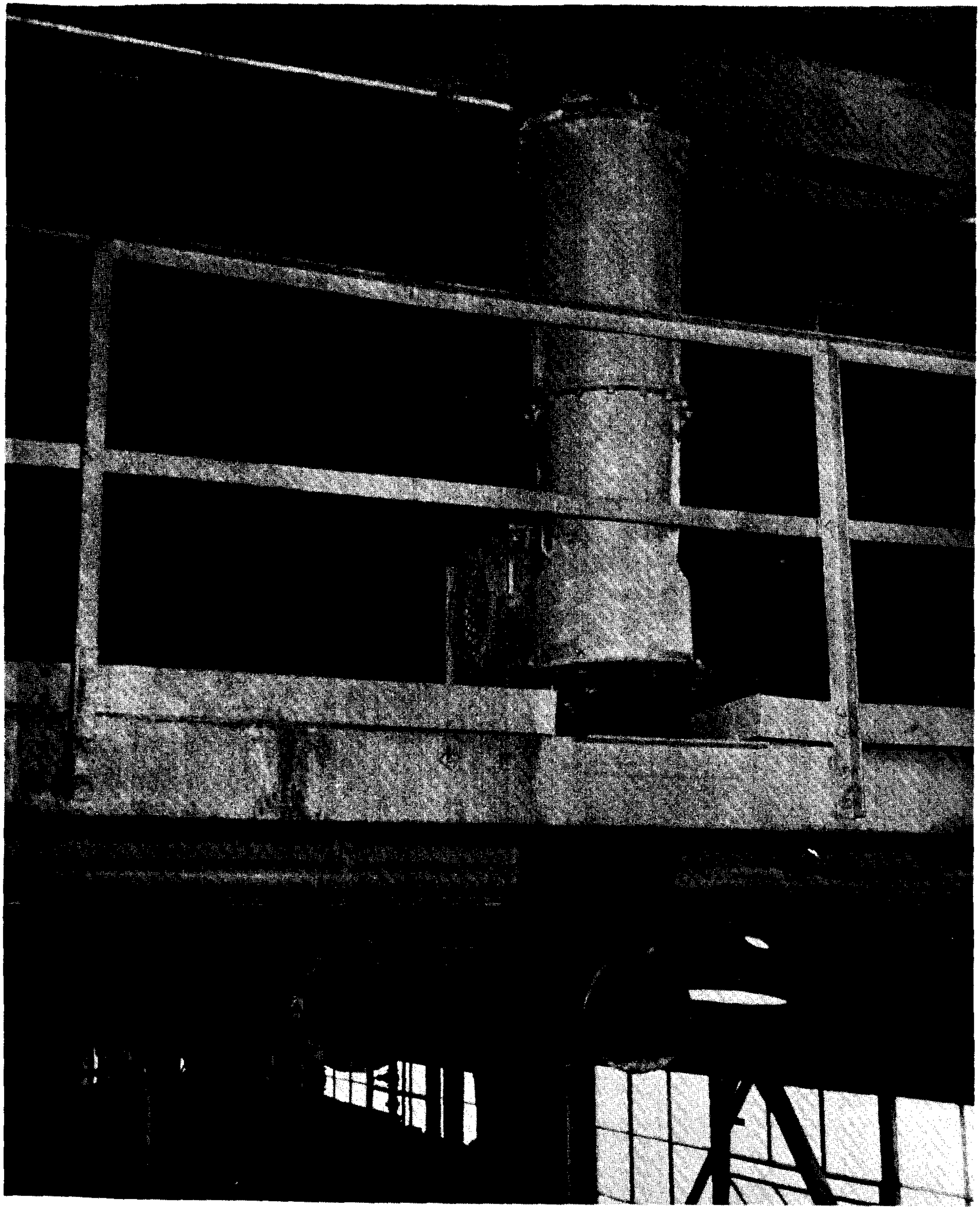


FIGURE 13 Eimco Filter Exhaust Fiberglass-Reinforced Polyester Resin Quickdraft Eductor and Duct

engineers in certain problems which were heretofore difficult to solve.

DISCUSSION AND COMMENT

The units run somewhat higher than 60% efficiency due to the manner in which the air is inducted. However, the efficiency drops off terrifically as higher statics are approached.

The proper way to evaluate may be to determine the mechanical efficiency of the power put into the system in the primary blower, and the power out in terms of the amount of air flow and the static. No figures were given on true mechanical efficiency but it was revealed that this would vary accordingly to the particular unit installed. It was estimated that the efficiency would be somewhere about or slightly below a regular blower.

The life of #304 stainless steel with perchloric acid was extended about three times. Nitric acid showed practically no attack. PVC, unplasticized, will be used from now on.

The units at lower static pressures using a quarter to one-half inch static are being purchased economically at approximately or slightly less than the price of a regular blower. The favorable purchase price was explained by the employment of a direct drive, high rpm unit.

Session Chairman: The next paper is "Final Results of Economic Survey of Air Cleaning Equipment Used at AEC Contractor Sites." Dr. M. W. First, Associate Professor of Industrial Hygiene at the Harvard Air Cleaning Laboratory, will present the paper.

FINAL REPORT OF ECONOMIC SURVEY OR AIR AND GAS CLEANING OPERATIONS WITHIN THE AEC

M. W. First

and

L. Silverman

Harvard University School of Public Health

INTRODUCTION

The Harvard Air Cleaning Laboratory has been conducting an economic survey of AEC air cleaning costs over a period of several years. Reports on the progress of this survey have been presented at past AEC Air Cleaning Conferences and they are available in the volumes of published proceedings (1,2,3). The economic survey has been completed and all of the data and results will become available soon as a numbered Harvard Air Cleaning Laboratory AEC Report is prepared.

The cost of owning and operating air cleaning equipment is a matter of interest and concern to all users and especially to organizations such as the AEC that handle exceedingly toxic materials that must be carefully and closely controlled regardless of cost because of the serious threat to life that would otherwise exist. Even in situations where health considerations take precedence over costs, it makes good sense to provide the needed protection at the least possible expense. It has been suggested by experienced engineers (4) that the development of power reactors in the U. S. is being seriously retarded by the cost of excessive safety provisions and that certain minimal risks should be accepted in the interest of low cost nuclear power production. Air cleaning is only one factor in the overall safety program, and perhaps a minor one with many types of reactors, but this type of thinking points up the necessity for a rigorous examination of costs as they relate to air cleaning effectiveness, regardless of how one responds to the idea of relaxing existing safety standards.

The exact purchase price of specific air cleaning devices is readily determined by consulting the manufacturer but the costs of erection plus the purchase of associated equipment such as ducts, stack, blower, water pumps, etc. often represent a far greater cost than the purchase price of the air cleaner. A rule of thumb that has been suggested is that the installation cost will not exceed two and one half times the purchase price of the air cleaner; but of course this is quite variable and often it is desirable to analyze the individual cost components.

Even less information is available on operational costs than on purchase and installation costs. One reason for this may be the great variations in operating conditions which exist from industry to industry and from plant to plant within the same industry. Undoubtedly, nonuniformity in accounting practices is responsible for some apparent differences in what would seem to be closely similar operations.

In spite of efforts to collect accurate and up-to-date cost information, it must be emphasized that the data represent broad averages and that significant deviations may be anticipated when out-of-the-ordinary requirements must be satisfied or when installation and servicing present special difficulties by reason of difficult terrain or unusual toxicity. When conditions do not deviate markedly from the norm, we believe the figures given here will provide a basis for performing planning functions and preparing budgetary estimates for a wide range of air and gas cleaning applications.

At the last conference (3) survey results on the cost of installing and operating AEC absolute filters and dry fibrous prefilters were reviewed. At this time, some of the data pertaining to (a) cleanable industrial fabric filters, (b) electrostatic precipitators, (c) dry mechanical dust collectors, and (d) scrubbers will be summarized. In the interests of brevity, the general methods of analysis that were discussed in the previous published progress reports to which I have already referred, will not be repeated.

CLEANABLE INDUSTRIAL FABRIC DUST FILTERS

Industrial fabric filters differ principally in the cleaning methods used to remove the accumulated dust cake and restore the permeability of the fabric. In the older of the two basic methods, the dust-laden filter is isolated from the air stream and cleaned by shaking, vibrating and/or reversing air flow through the fabric. In the other method, small fractions of the dirty fabric are cleaned continuously (while filtration proceeds) with the aid of a traveling jet of high velocity reverse flow air. Details of these cleaning methods have been published elsewhere(5).

In Table I are shown acquisition and operating costs for 90 cleanable fabric filters in service at AEC installations; 82 reverse jet cleaned filters and 8 shaken-bag units. The filter units are also grouped with respect to the size of the installation, i.e., cubic feet per minute of air flow capacity. From the Table, it may be seen that on the average the cloth-to-air ratio (cfm per sq. ft. of fabric) increases with increasing size of installation for both types of collectors. The reasons why this should be so are not known with certainty. The fact that acquisition and operating costs tend to decrease rather sharply with increasing size of installation suggests that small units are not only more expensive per unit of capacity (this was found to be the case for

Table I

Costs for Industrial Fabric Filters

Capacity	Number Install'ns	Total Ann. Cost per 1,000 cfm		Cloth-Air Ratio-Av.	Cost-Analysis - % Total Cost		
cfm		Aver.	Min.-Max.	cfm/sq.ft.	Purchase & Install'n	Power	Maint. & Repairs
A. Reverse-jet Cleaned Filters							
<1,000	5	1,947	333-3,131	5.8	31.7	7.9	60.4
1,000 - 5,000	34	1,168	222-4,481	8.2	18.8	10.6	70.6
5,000 -10,000	22	989	267-3,456	11.6	9.5	21.0	69.5
10,000 -20,000	17	1,213	192-5,221	14.9	7.2	21.9	70.9
>20,000	4	196	142- 259	21.9	29.6	29.6	40.8
B. Shaken Bag Filters							
1,000 - 5,000	6	520	302- 748	3.5	29.0	18.9	52.1
5,000 -10,000	2	1,723	224-3,221	4.2	7.8	5.3	86.9

AEC filters and prefilters, also) but in addition, that small installations tend to be associated with out-of-the-ordinary and difficult applications (such as vacuum cleaning, incinerator effluent cleaning, etc.) that produce abnormally high dust loadings or dust deposits with high air flow resistance. From this it may be concluded that small size, high dust loadings, difficult operating conditions, low air-to-cloth ratios, and high costs tend to occur together. Most of the fabric filter installations that have been reported are located in plants processing uranium ores. For many units, filter bags are removed each 3-6 months and incinerated to recover the metal accumulated in the fabric. This practice has a profound influence on total operating costs and largely accounts for the abnormally high costs of the reverse jet collectors in the 10,000 - 20,000 cfm size class. The most costly of the 90 cleanable fabric filters recorded in Table I occur in this group.

The number of shaken bag units reported (eight) is far fewer than for the reverse jet type and averages are not as meaningful. For the 1,000 - 5,000 cfm size group, the total costs of shaken bag units are about half those of reverse-jet filters whereas for the 5,000 - 10,000 cfm size group the cost relationship is reversed. It seems probable that these differences reflect special use conditions rather than an inherent cost differential between the types. A recent study (6) indicates that annual unit costs are closely similar when all types of fabric filters are employed on more usual industrial applications. It is evident from Table I that reverse jet cleaned fabric filters have average air-to-cloth ratios that are 2½ times those of shaken bag units.

For both types of fabric filters, maintenance and repairs account for over 50% of the annual unit costs, except for units in excess of 20,000 cfm capacity. Again, this reflects special AEC applications of a difficult nature.

LOW-VOLTAGE TWO-STAGE ELECTROSTATIC PRECIPITATORS

Two-stage electrostatic precipitators are used both for supply and exhaust air cleaning applications at AEC sites. Supply air cleaning is, by far, the more common use for this type of unit. In Table II, costs are analyzed by the nature of the application and it is evident that unit costs are much higher for treating exhaust air. Whereas the two-stage precipitator usually performs well with low loadings of fine dust customarily encountered in outside and recirculated indoor air, it is not constructed to handle high dust loadings, moist or corrosive atmospheres, abnormal temperatures, or any of the other unfavorable conditions commonly encountered with effluent process air and gas streams. That a two-stage precipitator is a misapplication for an exhaust air stream is revealed by high total costs as well as by the high percentage of these costs (> 90%) that goes into maintenance and repairs.

The supply air precipitators show a normal decrease in unit operating costs with increasing size and maintenance; and repairs account for 25% or less of the total cost.

Table II

Costs for Two-Stage Electrostatic Precipitators

	Supply Air Units								Exhaust Air Units							
Capacity	Number Install'ns	Total Ann.Cost $/1000cfm/yr			Cost Analysis- % Total Cost			Number Install'ns	Total Ann.Cost			Cost Analysis % Total Cost		
cfm		Ave.	Min.	Max.	Purchase & Install'n	Power	Main. & Repair		Ave.	Min.	Max.	Purchase & Install'n	Power	Main. & Repair
1,000-5,000	1	130			61.6	13.8	24.6	1	164			71.3	10.5	18.2
5,000-10,000	5	76	32	144	53.3	20.0	26.7	0	-	-	-	-	-	-
10,000-20,000	1	24	--	--	45.8	33.3	20.9	3	680	201	920	6.8	1.8	91.4
>20,000	1	27	--	--	55.5	33.3	11.2	2	7,094	45	14141	0.6	0.2	99.2

DRY MECHANICAL DUST COLLECTORS

Costs were reported for ten dry mechanical dust collectors ranging in type from a simple gravity settling chamber to a small diameter multi-cyclone installation. Unit size ranged from 800-8,000 cfm. Cost data for each installation are shown in Table III as there are not enough representatives of each type and size to make average cost figures meaningful.

In general, overall annual unit cost is high when "maintenance and repairs" represents a large percentage of total cost. When total cost is low, power costs represent a sizeable fraction of the total annual cost. With the exception of the gravity settling chamber, this class of dust collectors has an air flow resistance in the range of 3-6 in. w.g. and when there are no unusual maintenance costs (because of erosion, corrosion, or special difficulties associated with the disposal of the collected dust), power tends to be a major fraction of the total cost.

It is, perhaps, somewhat surprising to learn that operating costs for simple mechanical dry dust collectors may, under unusually severe operating conditions, exceed $500.00 per 1,000 cfm of capacity per year. For normal operating conditions, annual costs of under $200.00 per 1,000 cfm of capacity per year are customary in conventional industrial plants and some of the AEC installations shown in Table III are within this cost figure.

WET DUST COLLECTORS

Cost data for four type "N" Rotoclones are shown in Table IV. Three have closely similar total annual unit costs; for the fourth, costs are approximately 300% greater. "Maintenance and repairs" represents 82% of the total for this latter unit and it may be assumed that unusually severe operating conditions make it necessary to give this collector constant servicings. "Maintenance and repairs" for the two Rotoclones handling exhaust air from a machine shop and foundry (a usual industrial application for these units) is only 5% of the total whereas power is 2/3 of the total cost, indicating a more normal "maintenance and repairs" situation for these dust collectors.

Total annual unit costs for type "N" Rotoclones are only slightly greater than those for the dry mechanical dust collectors, in spite of the fact that the collection efficiency of the type "N" for small particles is far better.

Table V shows annual purchase and installation costs for 32 Schutte-Koerting Scrubbers installed in a uranium refinery. Operating costs, other than for water requirements, are not available.

These eductor units are commonly used as a combination pump and scrubber when the corrosive properties of the gases and vapors make the use of mechanical blowers and exhausters impractical. As the mechanical efficiency of the unit when used as a blower is 10% or less, the quantities of water that must be pumped are large.

Table III

Costs for Dry Mechanical Dust Collectors

Type Unit	Capacity cfm	Total Ann. Cost $/1000cfm/yr	Cost Analysis- % of total cost			Service Conditions
			Purchase & Install'n	Power	Maint. & Repairs	
Settling chamber	4,000	103	3.7	17.5	78.8	Exhaust from graphite machining
Aerodyne	5,900	92	41.4	25.4	33.2	Incinerator flue gas.
Rotoclone "D"	4,000	593	12.2	28.3	59.5	Exhaust from graphite machining
Cyclone	3,570	203	16.3	34.6	49.1	Exhaust from carpenter shop
(2) Cyclone(Ducon)	2,200	409	33.1	22.9	44.0	Incinerator flue gas
(2) Cyclone (A,B,C)	2,200	170	22.9	55.3	21.8	Exhaust from U-machining
Cyclone (K & B)	8,090	135	17.8	69.6	12.6	Exhaust from U-machining
Multicyclone (Dustex)	800	511	46.5	18.3	35.2	Exhaust from sintering furnace

Table IV

Cost of Wet Dust Collectors Installed at AEC Sites

Type Unit	Capacity	Total Ann. Cost	Cost Analysis- % of Total Cost			Service Conditions
	cfm	$/1000cfm/yr.	Purchase & Install.	Power & Water	Maint. & Repairs	
Rotoclone "N"	3,390	505	32.3	62.5	5.2	Exhaust from machine shop and foundry
"	5,500	422	20.5	74.8	4.7	" " "
"	5,900	1,399	2.1	16.2	81.7	Exhaust from U-refinery (U_3O_8)
"	13,000	547	4.9	41.5	53.6	" " "

Table V

Purchase and Installation Costs - Schutte-Koerting Scrubbers (Uranium Refining)

Capacity cfm	Liquid rate gal/1000 cf air	Initial equipment cost $/1000 cfm/yr.*	Initial installation cost $/1000 cfm/yr.*	Total equipment annual cost $/1000 cfm/yr.*
128	70	483	388	871
300	60	167	134	301
400	60	130	103	233
750	60	66	53	119
1700	100	45	36	81
2100	150	39	67	106

* 30-year write off period.

The cost of pumping water in dollars per year per 100 gpm, is 16.1 x H, where H is the pumping head in feet. (Power is assumed to cost $.015/KWH and annual use to be 8000 hours). For a 30 foot pumping head (a commonly-used figure for low pressure applications), the annual cost per 1,000 cfm of air capacity will be $290.00 to $750.00 depending on the water requirements of each unit, as shown in Table V. This sum, plus the cost of maintenance and repairs, must be added to the annual equipment cost figures shown in the Table.

SUMMARY

Cost analyses of the many different types of dust collectors used at AEC sites show that installations that are critical from a health and safety standpoint as well as those involving the handling of heavy dust loadings and highly corrosive gases and vapors, result in unusually high annual unit costs. These conditions are not greatly different from those encountered in more conventional industrial experience except that unfavorable operating conditions and critical health problems are much more frequent within the AEC.

All types of units show lower unit costs as the size of the installation increases, up to about 10,000 - 20,000 cfm. For larger units, costs tend to be the same. For units of the same air flow capacity, treatment of exhaust air streams is usually 2-3 times as costly as treatment of ventilation air streams. For supply air units of 10,000 cfm capacity, dry fiber throw-away prefilters cost under $50/1000 cfm/yr.; two-stage electrostatic precipitators, $76/1000 cfm/yr.; and AEC absolute filters, $175/1000 cfm/yr.

For exhaust air cleaners of the same capacity (10,000 cfm), most dry and wet medium efficiency mechanical dust collectors will cost in the vicinity of $500/1000 cfm/yr., and cleanable fabric dust collectors about double this figure.

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CHARACTERIZATION OF GAS-BORNE FISSION PRODUCTS

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R. D. Ackley
M. D. Silverman

ABSTRACT

Two methods for determining the form of radioactivity in gases have been investigated. In the first method, diffusion coefficients for fine particles and for radioactive vapors such as iodine and its compounds are determined by measuring the distribution of radioactivity on the walls of a channel previously exposed to gas carrying radioactive materials and flowing under laminar conditions. This method has been utilized in investigating the nature and behavior of volatile iodine compounds. These have been observed to occur, in certain situations, in conjunction with the elemental form. Information on such compounds may be needed to design properly removal devices for radioiodine and to interpret deposition behavior. Composite diffusion tubes having, in the direction of flow, inner surfaces of silver, rubber, and, activated carbon have shown the presence of two non-elemental species of iodine in addition to the elemental. The compound or compounds which are retained by a rubber surface have an indicated diffusion coefficient in air of around 0.05 cm^2/sec, and that retained by activated carbon, about 0.10 cm^2/sec. These results together with data obtained by other means have provided clues as to the identity of the iodine compounds and further work is planned in order to identify them more explicitly.

The second method for measurement of the characteristics of radioactive aerosols makes use of fibrous filters. This method measures aerosols in terms of their response to the processes of inertial impaction, interception, and diffusion by determining their distribution versus depth in fibrous filters under carefully controlled conditions. The velocity dependence of the slope of distribution curves on a semilog plot identifies the mechanism of filtration, and the magnitude of the slopes indicates the intensity of the filtering action and thus the character of the particles. Experiments where diffusion is the controlling mechanism have been quantitatively treated and particle sizes in the 50 to 500 A^o range determined by this technique compare favorably with those obtained from concentrations and flow data and electron photomicrographs.

INTRODUCTION

The behavior of gas-borne materials depends markedly on their form; consequently, the prediction of their dispersal or transport or the selection and design of systems for their removal from gases requires information as to the distribution of radioactivity between the different forms which occur. Two methods for determining the form of radioactivity in gases have been investigated.

DIFFUSIONAL DEPOSITION OF IODINE COMPOUNDS

The first method for characterizing aerosols has been described previously (1). In this method, diffusion coefficients for fine particles and for vapors, such as iodine, are determined by measuring the distribution of radioactivity on the walls of a channel previously exposed to gas carrying radioactive materials and flowing under laminar conditions. As the diffusion coefficients are related to molecular properties or particle sizes, pertinent information as to the form of the fission products may thereby be obtained.

More recently, the diffusional deposition technique has been utilized in an effort to determine the identity of various iodine compounds which have been observed to occur in the diffusion tube experiments along with the elemental form. British workers in this field have previously reported that their data indicated some formation of iodine compounds when elemental iodine is released into atmospheric air (2, 3). Their results, however, correspond to considerably lower concentrations of iodine in air and so are probably not directly comparable. Some of the iodine compounds variously encountered may have very different deposition behavior in a gas coolant system and also are very likely removed with much lower efficiency than elemental iodine in adsorbers and scrubbers, and, consequently, knowledge of their behavior is needed to design properly radioiodine removal devices.

The probable presence of iodine compounds became apparent in this work when distributions in diffusion tubes did not conform to either those previously obtained or to those anticipated. In the earlier experiments corresponding to these observations, particles had been generated and labeled with I-131, but later on, in order to concentrate attention on the compounds, deliberate particle generation was omitted and the source air, which was the carrier gas in these studies, was rather elaborately filtered. Detailed examination of the diffusion tubes, which were silver-plated copper tubes, and of associated rubber tubing and activated carbon traps, then indicated the presence of two distinct species of iodine other than elemental. One of the species appeared to be quantitatively retained by the inner surface of rubber tubing and the other by activated carbon, but neither by silver metal. Both the origin and the importance of these two species are speculative but considering the important role of iodine in nuclear safety, further elucidation was deemed highly desirable. An experiment was then performed which included rubber tubing diffusion tubes; from the I-131 distribution in these a diffusion coefficient for one of the species in air was calculated to be about 0.05 cm^2/sec indicating a molecular weight (or weights) of about 300 amu. Of the total I-131 activity involved in the experiment, around 6% was estimated to be in the form of compounds. Also, the source iodine, which had I-127 as carrier, remaining after this experiment was analyzed by mass spectrometry and was observed to contain two compounds having masses well in excess of that of elemental iodine, the masses being 284 and 338. Two compounds having these weights are SiH_2I_2 and $C_6H_{12}I_2$, respectively, although this is not to suggest the identities but only to provide examples of the possibilities.

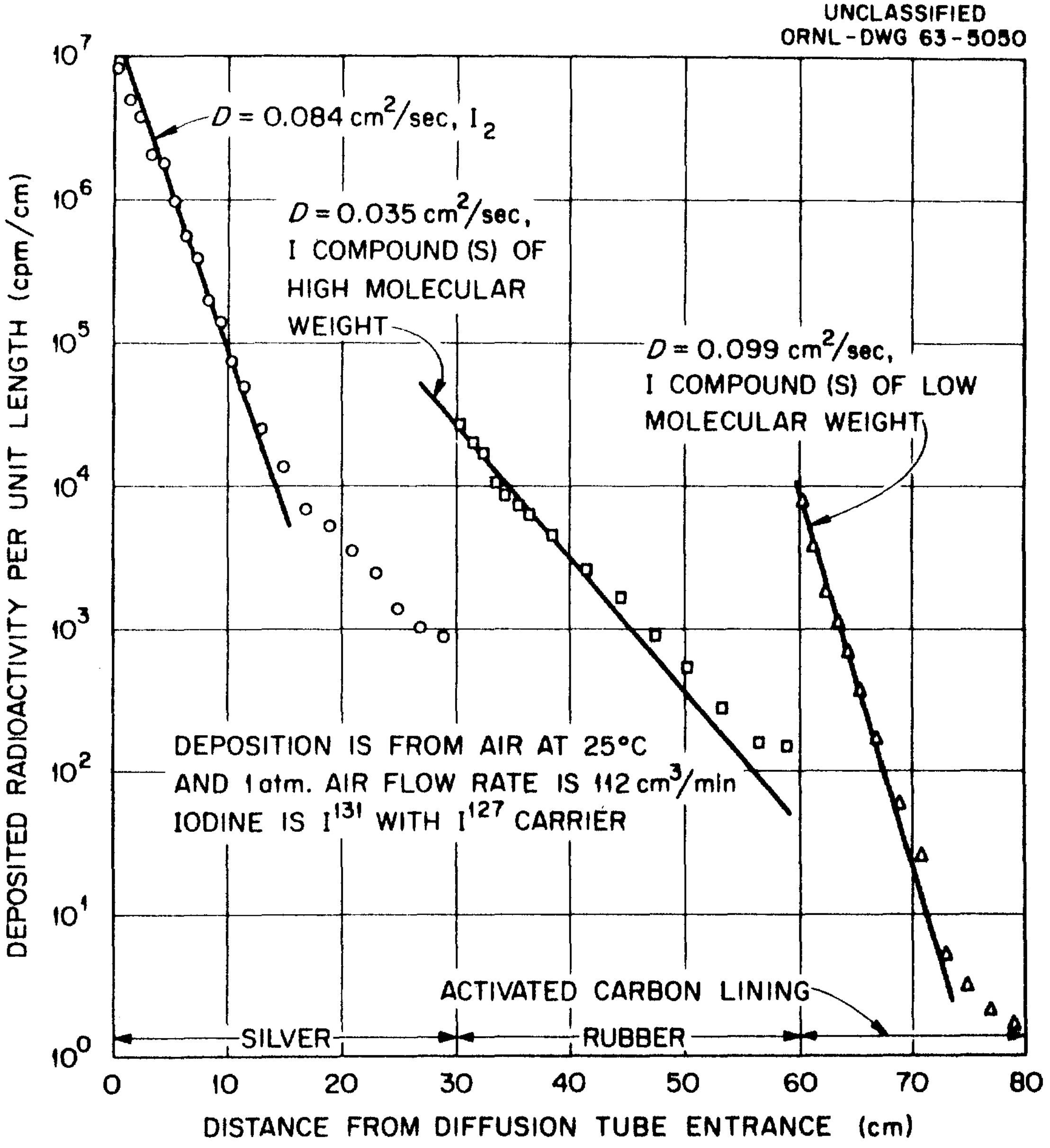
In another approach to this problem, the initial objective was to be able to prepare pure radioiodine and subsequently, to allow such material to contact other gases and various surfaces, and then to determine if volatile radioactive compounds were produced. The method of iodine preparation investigated was similar to that described by British workers (2, 3) and involved the evolution of iodine from melted potassium dichromate containing NaI-131 and KI-127. The purity was determined by means of the behavior of the iodine in diffusion tubes. With helium as a carrier gas, a purity of 99.6% was achieved; but with air, the purity was only 98.8% and the principal impurity had a diffusion coefficient in air of 0.05 cm^2/sec, as had been obtained earlier. Since these observations indicated the air source was at least partially responsible for the appearance of compounds, attention was directed toward it. The air used in the test yielding 98.8% was plant air passed through an activated carbon trap. Since the activated carbon itself might have desorbed gases which subsequently reacted with iodine, as only traces would be required, it was replaced in the next experiment with a trap filled with Linde Molecular Sieves Type 5A. Also, the diffusion tube train had, in addition to silver-plated and rubber tubing, a following section of tube lined with activated carbon dust. Virtually all of the I-131 radioactivity which evolved from the dichromate melt and which entered the diffusion tube train was retained in the train and, of this iodine, 99.3% was indicated to be elemental. Most of the remaining 0.7% deposited on the rubber tubing and exhibited a diffusion coefficient in air of about 0.035 cm^2/sec, not greatly different from the previous results for rubber tubing. About 0.1% penetrated the silver-plated and the rubber tubing and deposited in the tube lined with activated carbon to yield a diffusion coefficient of 0.10 cm^2/sec which corresponds to HI or a slightly larger molecule than HI. These deposition results are shown in Fig. 1. A small portion of the evolved radioactivity did not reach the diffusion tube but deposited, probably due to a gas-phase reaction, as a yellow solid at a point where the air stream carrying the radioactivity was diluted by the main air stream. Preliminary indications are that the deposit was an iodine-oxygen compound.

As may be seen from the preceding, a variety of information has been obtained pertaining to the compounds of iodine which may occur in conjunction with the elemental form, and further studies toward this objective are planned.

CHARACTERIZATION OF AEROSOLS BY FIBROUS FILTERS

The second method for measurement of the characteristics of radioactive aerosols makes use of fibrous filters. The transport of an aerosol through an array of fibers brings into play the processes of inertial impaction, interception, and diffusion. Since all these processes have important effects on the behavior of radioactive materials in gases, it would be useful to be able to characterize radioactive aerosols by measuring their behavior in fibrous beds. This method measures aerosols in terms of their response to these processes by determining their distribution versus depth under carefully controlled conditions. A filter was developed having uniform fiber diameter to permit theoretical analysis and having a layered structure to facilitate separation of the fiber bed into discrete layers for radioassay after exposure to the aerosol. A radioactive aerosol of 0.004-0.03μ particles, containing Zn-65 was developed for initial tests of this method of characterizing aerosols.

Experiments have been conducted over a wide range of linear flow rates, from 0.2 to 44 cm/sec. The data are analyzed by means of graphs (Fig. 2) in which the log of the activity collected per layer is plotted against depth in the filter expressed as number of layers of material. Each layer contains approximately 10^4 cm of fiber per cm^2 of filter area, evaluated from disk weights and the density of the fiber. In the low flow region where Brownian diffusion is the dominant process for particle transport to the fiber, the efficiency of filtration decreases with increasing velocity. In the high flow region, where inertial impaction is the principal process for particle transport, the efficiency increases with increasing velocity. In the intermediate flow region, the interception range where geometrical considerations involving particle size and fiber size are very important, the filtration efficiency is largely independent of velocity. Thus, the velocity dependence of the slope of these distribution curves identifies the mechanism of filtration, and the magnitude of the slopes indicates the intensity of the filtering action.



Diffusional Deposition of Elemental Iodine and Iodine Compounds.

Figure 1

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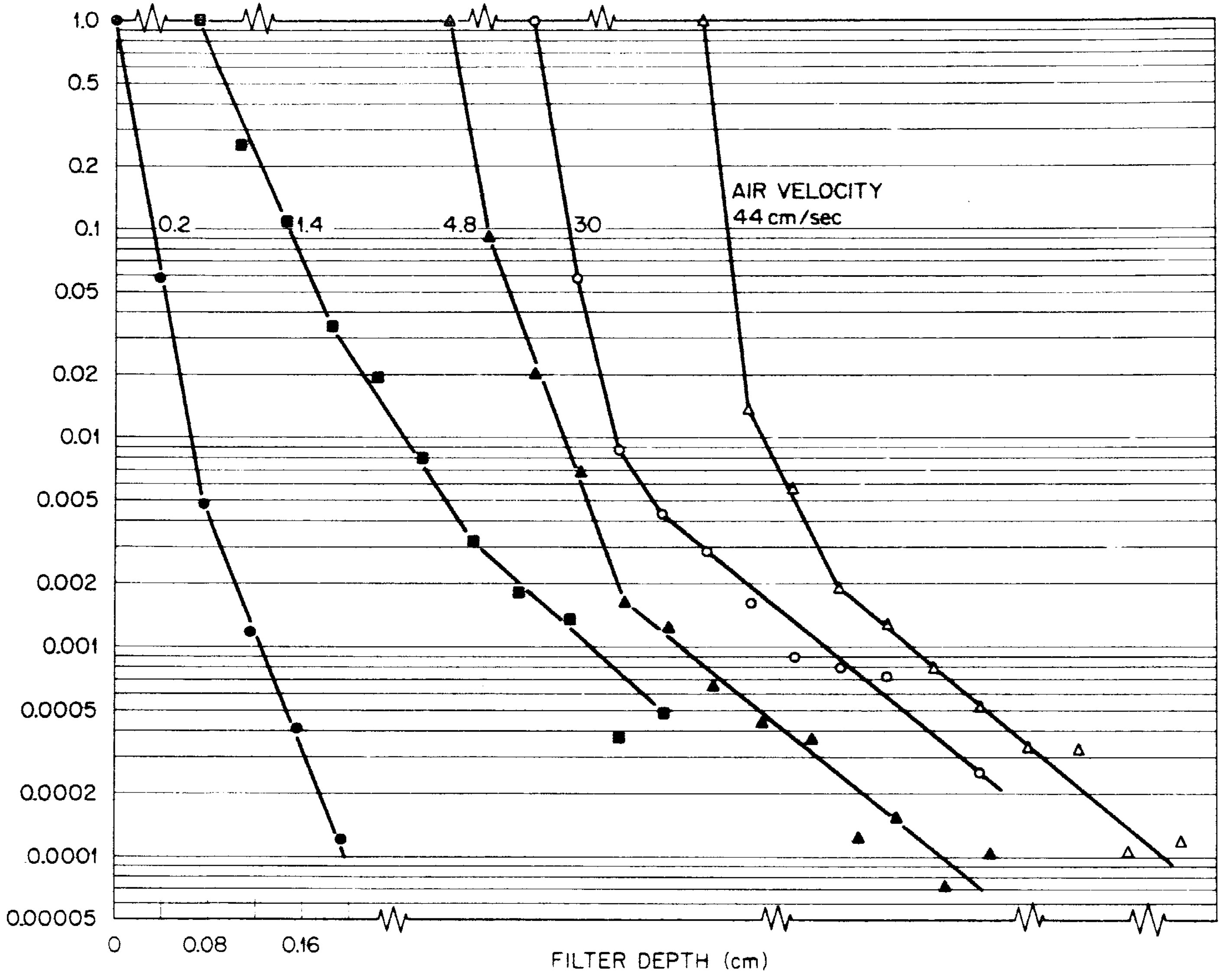

Distribution of Zn Activity vs Depth in Filter.

Figure 2

The filtering action of a fiber mat is the cumulative effect of particle removal by individual fibers. The collection efficiency of an individual fiber, η_d, is defined as the ratio of the cross-sectional area of the aerosol stream from which particles are removed to the projected area of the fiber in the direction of flow and may be obtained from the slope of the distribution curve on the semi-log plot. This slope may be substituted directly for $(\ln N/N_o)/L$ in the expression derived by Langmuir (4) and confirmed by Davies (5) and by Chen (6) for the single fiber efficiency:

$$\eta_d = - \ln \frac{N}{N_o} \frac{\pi(1 - \alpha)d_f}{4\alpha L} , \qquad (1)$$

where N_o and N are the upstream and downstream particle concentrations, respectively, α is the volume fraction occupied by the fibers, L is the thickness of mat, and d_f is the fiber diameter, all in consistent units. Fiber efficiencies, evaluated for those experiments conducted at linear velocities of 0.2 to 2.5 cm/sec showed that η_d is proportional to $1/\text{velocity}^x$. This decrease of efficiency with increasing velocity indicates that, in this flow region, diffusion is the primary mechanism of filtration.

Stairmand (7), using the same approach as Langmuir, based on diffusion of particles to the fiber surface in a fluid within a time equal to $\pi d_f/2v$, derived the following expression for the diffusion collection efficiency:

$$\eta_d = \left(\frac{8D}{vd_f}\right)^{1/2} , \qquad (2)$$

where v is the fluid velocity and D is the diffusion coefficient calculated from the Einstein expression (8):

$$D = \frac{CkT}{3\pi\mu d_p} . \qquad (3)$$

Here d_p is the diameter of the particle, C is the Cunningham coefficient, μ is the viscosity of the fluid, and the other symbols have their conventional meanings. The quantitative agreement between Eq. (2) and the experimental velocity dependence is confirmation that diffusion is the primary filtration mechanism under the specific conditions treated.

Particle diameters calculated from the experimental data using Eq. (2) are shown in Table I for experiments at several different velocities. The double sets of figures represent the two groups of particle sizes demonstrated by the separate portions of the curves in the semi-log radioactivity distribution plots discussed above. The use of Eq. (2) emphasizes the smaller size end of the particle spectrum and the results agree well with the lower size end of a particle distribution plot obtained from electron photomicroscopy. In the third column of the table under the heading "radiochemical data" another set of diameters, weighted toward the larger particle sizes, was calculated as follows: Radioassay results from the filter mats, along with specific activity data from the irradiated zinc foil electrodes, were combined to give the total weight of zinc removed by the filters. The number of particles produced was calculated from the volume of air transported and a particle concentration for the aerosol (read from a condensation nuclei counter, Gardner Associates). The concentrations observed are consistent with those which would be expected from the geometry of the experiment, and coagulation theory expounded by Whytlaw-Gray (9). From the weight of aerosol filtered (assuming a density of 1) and the number of particles per experiment, an average diameter was calculated for the particles in each experiment. It should be noted that at 2.5 cm/sec, the particle diameters

Table I. Comparison of Particle Sizes Determined by Different Methods

Gas Velocity (cm/sec)	Calculated Particle Diameter (A^o) From Diffusion Equation (2)	From Radiochemical Data
0.2	104	180
	168	
0.35	100	160
	180	
0.5	52	135
	98	
0.85	36	105
	70	
1.4	35	80
	105	
2.5*	13	65
	28	

*An aerosol prepared at 2.5 cm/sec, examined by electron microscopy, had a median diameter of 120 A^o and a geometric standard deviation of 2.0.

calculated theoretically are low compared with the radiochemical value, because the interceptional process is beginning to contribute to the filtration. The agreement between the three different methods for determining particle size is good.

Similar analyses of the distribution curves are being made for the inertial and interceptional regimes.

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Session Chairman: The last paper to be presented during Session III is by L. A. Haack of ORNL and is entitled "Analysis of a Possible Influence of Particle Size on Iodine Removal Efficiency of a Charcoal Filter."

ANALYSIS OF A POSSIBLE INFLUENCE OF PARTICLE SIZE ON THE IODINE REMOVAL EFFICIENCY OF A CHARCOAL FILTER

L. A. Haack

INTRODUCTION

The purpose of this paper is to show that to describe properly the efficiency of a filter in removing contaminants, especially iodine-131, from an air stream not only must the amount of impurities be specified (such as the number of curies or grams of iodine-131 per unit volume or parts per million) but also that the size distribution of particles in the air stream must be specified. Only in this way can the efficiency of a single charcoal filter, or of a system composed of a series of filters of various types, be properly described. The size distribution of particles is important since these particles might have iodine-131 associated with them.(a)

It is recognized that statements of this general type have been made previously by others. For example, Linderoth, Hilliard, and Healy stated:

> "...an investigation of particle emission should be made. This would include the determination of the types of fission products associated with the particles, the activity exhibited, and the size distribution of the aerosol."(1)

Dennis, et al (2) and Browning and Ackley(3) have made similar statements.

The importance of these statements is emphasized by the main feature of this paper; namely, a calculation in which it is shown that the efficiency of an assumed model of the filtering process compared quite closely with the efficiency which had been measured for an actual filter system. Based on the success of this agreement, the efficiencies of filters of two other situations were calculated:

1. Assuming that the air contained 10% of the total amount of iodine-131 associated with particles less than 0.3-micron diameter. (Absolute filters do not remove particles less than 0.3 micron with high efficiency.)

2. Injecting elemental iodine into the air upstream from a filter.

It is further suggested that it is quite easy for an exhaust stream to contain a significant percentage of particles smaller than 0.3 micron; namely, that such particles are produced by man and normally exist in the atmosphere. This fact,

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(a)By "associated" it is meant that the iodine-131 is adsorbed or absorbed by a particle, or that the iodine-131 has formed a compound with one or more other elements, or a combination of the preceding. For the purpose of this paper it is not important to distinguish which is the case; rather, it is only important that a certain percentage of the total iodine-131 activity behaves in the filtering process as though it were particulate in the size range from about 0.001 to 0.3 microns.

as pointed out by Dennis, et al,(4) could greatly complicate and confuse the laboratory work of very accurately measuring the efficiency of filters since the amount of such particles varies widely from locality to locality and even varies with time at a given locality.

This paper is largely heuristic in nature, and therefore certain assumptions are made without attempting to justify them.

CALCULATIONS AND COMPARISONS

One of the main concerns in the operation of a nuclear reactor is that in case of an accident the materials which are released by the accident not cause a hazard to personnel or great damage to property. Specifically, the main concern in many reactors is the biological effect of the radioiodines, especially iodine-131.

A common method of limiting the potential biological hazard is to exhaust the air and its contaminants from the reactor building, pass it through a set of filters, and discharge it to the atmosphere from a high stack.

The key link in such a scheme is the filter system through which the air passes and which, ideally, removes the hazardous materials. This process must be very efficient for most reactors since as little as about 0.01% of the iodine-131 inventory contained within the fuel elements of some reactors, if released directly to the atmosphere, would exceed the present maximum permissible concentration.

Since the air exhausted is likely to contain particles having a wide range of sizes, the filter system usually consists of different types of filters--normally roughing filters, absolute filters, and charcoal filters.

Roughing filters usually consist of a fibrous material and can usually remove all particles larger than ~100-microns diameter. Absolute filters usually consist of asbestos-impregnated paper and can remove 99% of the particles of greater than 0.3-micron diameter. Charcoal filters usually consist of 6-14 mesh charcoal particles and can usually remove more than 99.99% of the elemental iodine. Figure 1 summarizes these commonly accepted filter efficiencies.

Apparently there is a particle size range for which no type of filter is completely effective. The range is that below the absolute filter (0.3 microns) and above the charcoal filter (elemental iodine). At first glance, this gap might appear to be relatively unimportant since this range is such a very small percentage of the total. As previously stated, though, it is the main point of this paper to show that this range may be a very critical one that influences considerably the over-all efficiency of a reactor air-handling filter system. As can be seen from Figure 2,(5) several processes produce particles in this size range; for example, smoke from various sources and chemical mists.

Model Used in Calculations and Comparison with Actual Filter Efficiency Measurements

The model used in the calculations is quite simple. Essentially it assumes the following:

1. The air stream passing through the filter is composed of two components:
    - a. Iodine in elemental form, and
    - b. Iodine associated with particles less than 0.3 microns in diameter.

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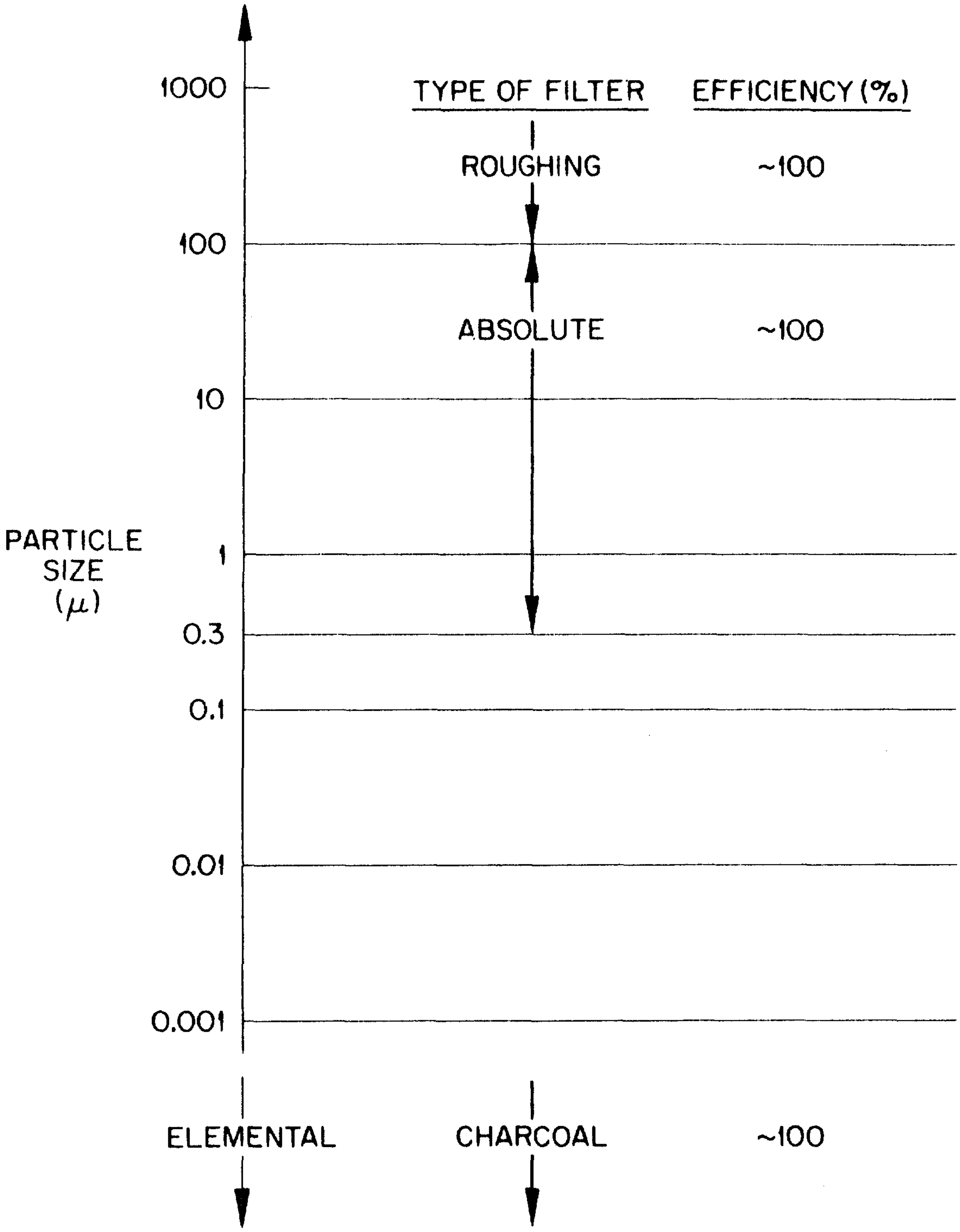

Fig. 1. Efficiency of Various Types of Filters for Range of Particle Sizes.

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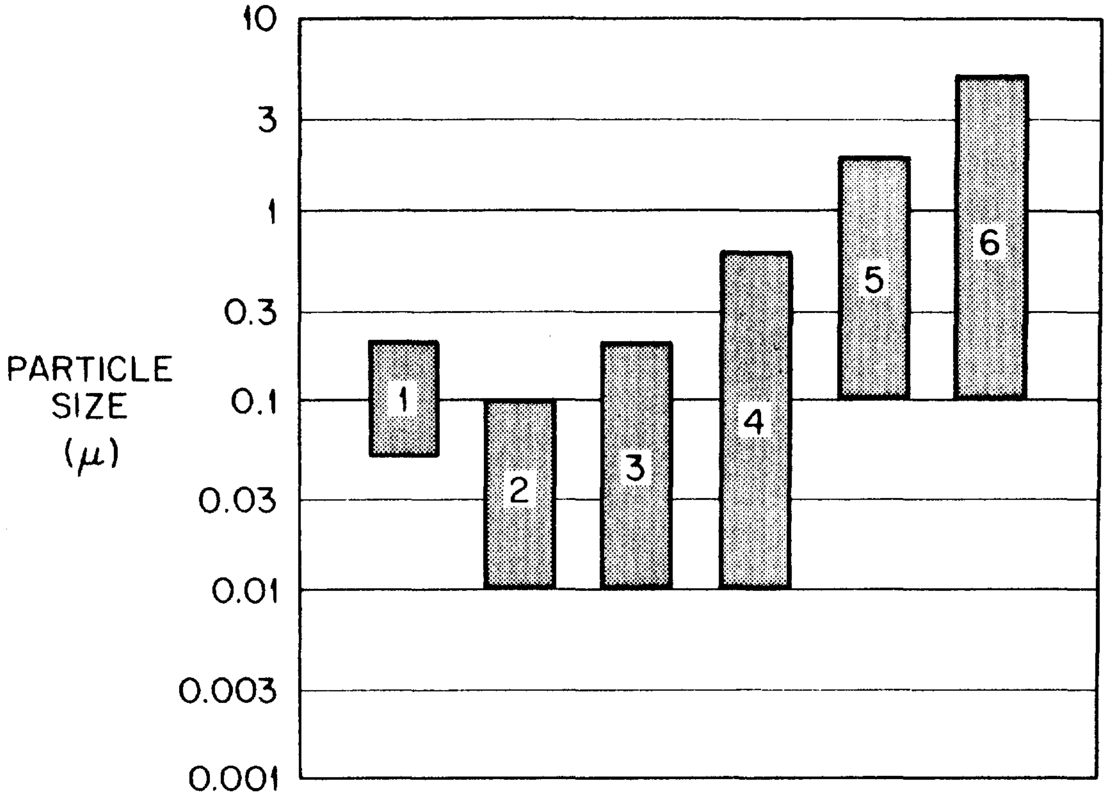

1. EXHAUST FROM AUTOMOBILE ENGINE (REF 11)
2. EXHAUST FROM BESSEMER CONVERTER (REF 10)
3. TOBACCO SMOKE (REF 5)
4. MAGNESIUM OXIDE SMOKE (REF 5)
5. AMMONIUM CHLORIDE FUMES (REF 5)
6. ALKALI FUMES (REF 5)

Fig. 2. Sizes of Selected Types of Particles.

2. Each of these two components is removed with a distinctive efficiency (it might be called a "partial efficiency" or "component efficiency"). In the case of elemental iodine the efficiency is 99.9%, for iodine associated with particles something much less.(b)

3. The distinctive efficiency of each component remains unchanged for subsequent filters of the same type in series.

The air stream could be separated into more than two components, each of which could be filtered with a particular efficiency. However, it is believed that this is a refinement that is not justified on the basis of present experimental data.

As was pointed out by Browning and Ackley,(6) the ability of a charcoal filter to remove particles smaller than 100 Å is considerably less than its ability to remove elemental radioiodine. For example, they reported efficiencies for a 3/4-in.-thick charcoal bed of 75% for 60 Å particles.

Based on these reported values it is interesting to calculate the combined and individual efficiencies of three charcoal filters in series for an air stream that contains, for example, radioiodine, 99.95% of which is elemental radioiodine and ~0.05% of which is associated with particles smaller than 0.3 micron. To arbitrarily choose workable numbers for the calculation, suppose that the exhaust stream has 10,000,000 arbitrarily-sized activity units of elemental iodine and 5,000 units associated with particles smaller than 0.3 micron. Further, suppose that the charcoal filter removes elemental iodine with an efficiency of 99.9% and the particles with an efficiency of 10%.

By using these efficiencies, we find that 10,000 units of elemental activity and 4,500 units of particulate activity penetrate the first charcoal filter (Figure 3a). The efficiency of this filter in removing radioiodine from the stream, regardless of its form, is

$$\text{Efficiency} = 1 - \frac{10,000 + 4,500}{10,000,000 + 5,000} = 99.9\%.$$

If these same efficiencies are assumed for the two iodine-131 components for the second filter, 10 activity units of elemental iodine and 4,050 activity units of particulate will penetrate (Figure 3b). The efficiency of this second filter is then calculated to be

$$1 - \frac{10 + 4,050}{10,000 + 4,500} = 72.0\%.$$

If this stream now passes through a third charcoal filter with the same efficiencies, 0.01 activity units of elemental iodine and 3,645 activity units of particulate will penetrate (Figure 3c). The efficiency of this third filter is

$$1 - \frac{0.01 + 3,645}{10 + 4,050} = 10.1\%.$$

These calculated efficiencies are compared in Table I with those reported by Browning and Adams.(7)

(b) The thickness of the charcoal and nominal velocity of the air stream passing through it need not be rigorously specified for the calculations, since the general method will be applicable to a wide range of thicknesses and velocities. A 3/4-in. thickness of charcoal and velocity of 25 fpm are reasonably typical to use for the efficiencies in the calculations.

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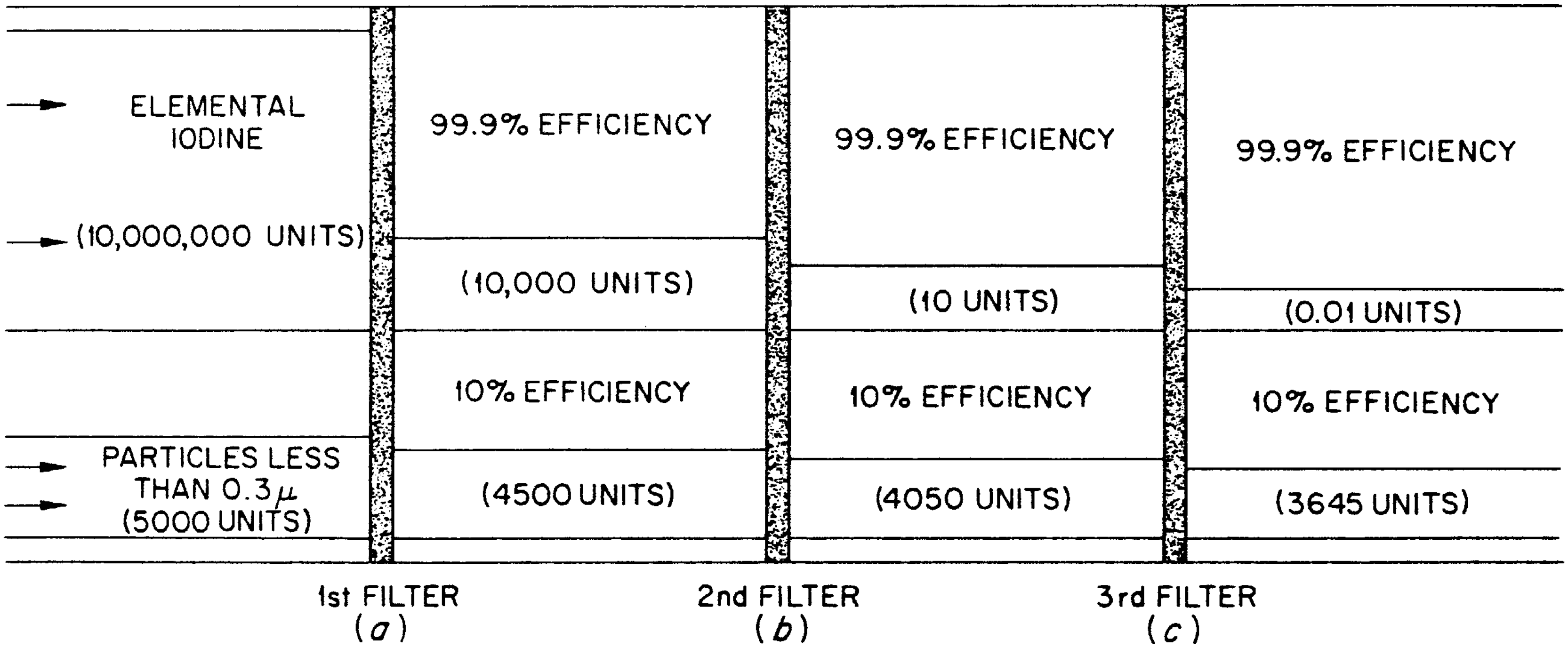

Fig. 3. Calculated Efficiencies of Three Charcoal Filters in Series for Approximately 0.05 Percent of the Total Amount of Iodine Associated with Particles Less than 0.3 Micron Dia.

Table I. Comparison of Calculated Filter Efficiencies with Measured Efficiencies

	Browning-Adams	This Paper
1st filter	99.99%	99.9%
2nd filter	74.6%	72.0%
3rd filter	14.5%	10.1%

The comparison between the two is striking!

The calculated combined efficiency of the three filters then is

$$1 - \frac{0.01 + 3{,}645}{10{,}000{,}000 + 5{,}000} = 1 - 3.64 \times 10^{-4} = 99.96\%.$$

Moreover, if the relative amount of iodine associated with particles is greatly changed, the efficiency of each individual filter and of the filter combination is changed greatly, too. This is shown in the next section.

Admittedly, the model used in the preceding calculation is quite crude, and the quantities of activity and efficiencies which were assumed are quite arbitrary and fortuitous. Nevertheless, the general idea is demonstrated adequately.

Calculated Filter Efficiency for Air Stream Containing Approximately 10 Percent of Total Iodine-131 Activity Associated with Particles Less Than 0.3-Micron Diameter

Since the efficiencies calculated by the proposed model compared favorably with experimental data, it would seem of interest to use the model for other situations. One interesting situation is that of an air stream which has a fairly high percentage of activity associated with particulate in the range 0.001 to 0.3 microns.

For example, suppose that the exhaust stream has 10,000,000 arbitrarily-sized activity units of elemental iodine and 1,000,000 units associated with particles smaller than 0.3 micron. (The actual percentage of activity associated with the particles is 9.1% of the total activity. The 10,000,000 and 1,000,000 were chosen for convenience.)

For the first charcoal filter, 10,000 activity units of elemental iodine and 900,000 units of particulate iodine penetrate (Figure 4a). The efficiency is

$$1 - \frac{10{,}000 + 900{,}000}{10{,}000{,}000 + 1{,}000{,}000} = 91.7\%.$$

For the second filter the efficiency is (Figure 4b)

$$1 - \frac{10 + 810{,}000}{10{,}000 + 900{,}000} = 11.0\%.$$

For the third filter the efficiency is (Figure 4c)

$$1 - \frac{0.01 + 729{,}000}{10 + 810{,}000} = 10.0\%.$$

The over-all efficiency for the three filters in series is

$$1 - \frac{0.01 + 729{,}000}{10{,}000{,}000 + 1{,}000{,}000} = 93.4\%.$$

This over-all efficiency would probably not be acceptable for many installations.

Based on the preceding calculations an important conclusion can be drawn; namely, if a fairly large percentage of iodine activity is associated with particles, or behaves as particles, in the range 0.001 to 0.3 microns, the efficiency of a charcoal filter may not be acceptable.

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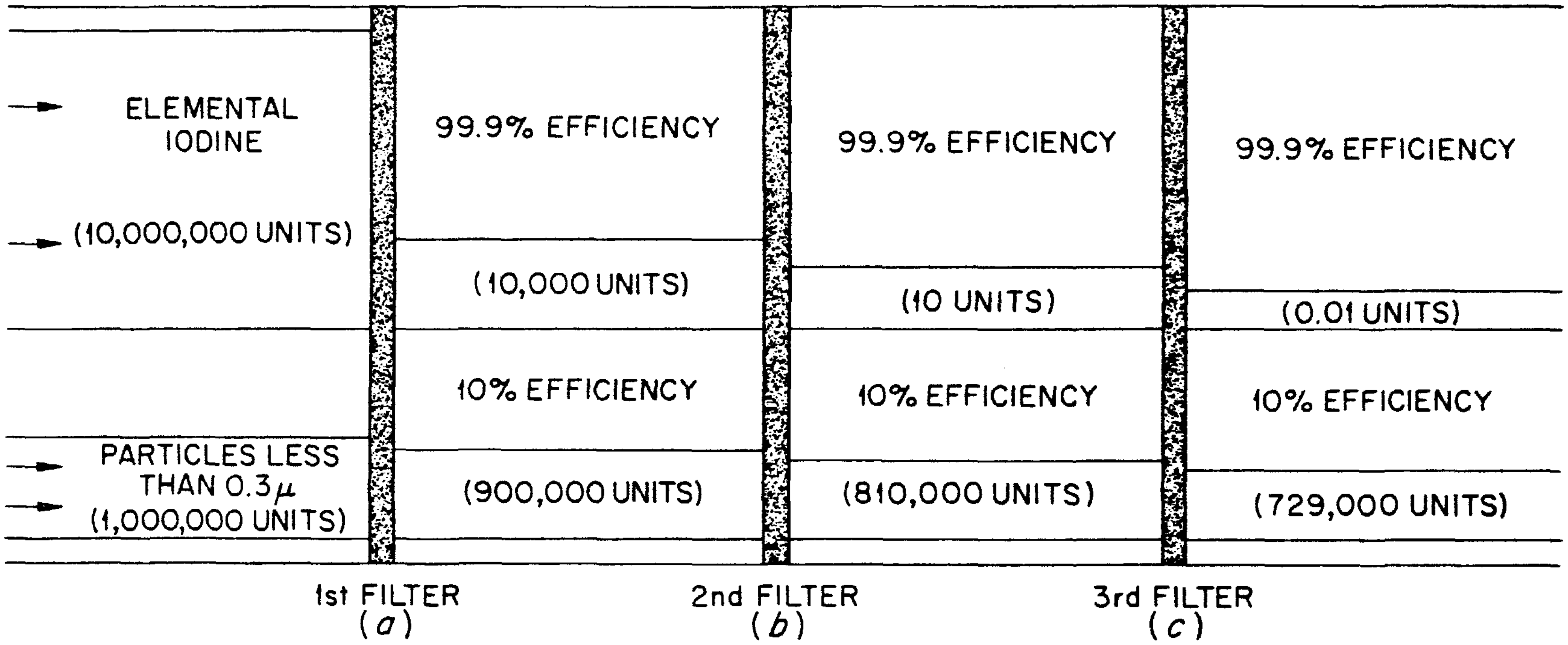

Fig. 4. Calculated Efficiencies of Three Charcoal Filters in Series for Approximately 10 Percent of the Total Amount of Iodine Associated with Particles Less than 0.3 Micron Dia.

It is natural to speculate on conditions when an air stream, especially in a full-sized plant, contains iodine-131 activity a large fraction of which is associated with particles in the size range 0.001 to 0.3 microns. Several conditions which could lead to this situation are:

1. Certain types of fuel elements when melted under certain conditions produce particles in this size range.

2. The atmosphere contains, at all times, particles in this size range.

3. Fuel-reprocessing plants and isotope-recovery plants in their normal operation produce chemical mists in this size range.

Regarding the first condition, particles in this size range have been observed to be produced by the melting of some types of fuel elements. For example, Parker(8) measured the size distribution of particles produced when UO_2 was melted in several different atmospheres. He found that in air 3% of the particles produced were less than 0.0250 microns, in helium 15% were less than 0.007 microns, and in carbon dioxide 30% were less than 0.040 microns. (This is not to imply, however, that all types of fuel elements or conditions causing melting are likely to produce particles of these sizes.)

It can be postulated that a certain fraction of iodine-131 released in elemental form somehow becomes associated with these particles. (We leave unsettled the question of what percentage of the original amount of iodine-131 in the fuel element was released upon melting.) These particles will not be removed by any of the three types of filters with a high efficiency.

If the total amount of iodine-131 activity associated with these particles is quite small, say 0.05%, the situation may not be serious. On the other hand, if the amount associated with the particles is higher, say 10%, the situation could indeed become serious.

Regarding the role of naturally occurring particles in the atmosphere, it has been found that the atmosphere contains at all times particles in a wide range of size and of chemical form. As shown in Table II,(9) the amounts and types of particles depend very much upon locality.

Table II. Occurrence of Particles in the Atmosphere for Various Localities During 1957 to 1958

Locality	Maximum Quantity of Suspended Particulate Observed ($\mu g/m^3$)	Maximum Quantity of Benzene Soluble Organic Matter Observed ($\mu g/m^3$)
Akron, Ohio	431	40.0
Gary, Indiana	945	46.8
Knoxville, Tennessee	245	33.7
Minneapolis, Minnesota	722	18.1
Los Angeles, California	361	56.1
Baldwin County, Alabama	49	7.6
Butte County, Idaho	56	3.8

Unfortunately, the sizes of the particles were not listed; however, we can gain knowledge on the probable sizes of many of these from other sources. For example, the size of many particles emitted from a Bessemer converter has been found to be in the range from 0.01 to 0.1 micron.(10) Also, particles exhausted from automobile engines have been found to be in the range from 0.05 to 0.1 micron.(11)

These numbers are not meant to imply that the quantities quoted in Table II consist entirely of particles from steel mills or automobile engines; they are meant to imply, however, that some of the particulate matter most likely consists of particles in this size range.

The air which is drawn through the reactor containment system will contain these particles even if the inlet air to the reactor building is filtered with roughing and absolute filters which, for practical reasons, is usually not the case anyway.

Thus any elemental iodine released to the containment system may readily become associated with some of these particles. Therefore, even though a particular type of fuel element, upon melting, may not produce small particles there is still the distinct possibility that iodine-131 will become associated with difficult-to-remove particles and thus be transported through the filter system to the atmosphere. The seriousness of such a situation depends, in part, upon the quantity and type of contaminants existing in the atmosphere.

Contaminants in the atmosphere have been suggested(12) as contributing to the difference in efficiencies measured by various workers for the same type of filter under apparently the same test conditions.

Furthermore, even if an extremely pure atmosphere is used in laboratory tests the fact remains that the ultimate users of filter systems (namely, reactors, isotope-recovery plants, and fuel-reprocessing plants), for practical reasons, may be unable to use anything except the unfiltered, or only slightly filtered, natural atmosphere. For example, the ventilation system at the Oak Ridge Research Reactor is designed to exhaust about 6,000 cfm of air during an emergency. To achieve this flowrate an equivalent quantity of air must also enter the building. The major portion of the air enters through two intake ducts which contain roughing filters; a smaller portion enters through various leaks, such as around the doors, and is not filtered.

It is also suggested that there are factors that can cause concentrations higher than the maximums quoted in Table II. The exhaust from a specific source of the particles, a diesel-generator unit, for example, causes higher concentrations in its immediate area. If such a unit were used as the emergency power source for a reactor and if the exhaust of the unit were discharged near the intake of a reactor containment system, then even more particles than normal would be available. It is likely that some of the iodine-131 would become associated with these particles.

The third situation, where more than the normal amount of particles less than 0.3-micron diameter are likely, is in a fuel-reprocessing plant or in an isotope-recovery plant (in the latter case, especially, recovery of iodine-131). Of course, if the fuel at a fuel-reprocessing plant had decayed for a sufficiently long time, the iodine-131 presents no problem. However, in the case of the isotope-recovery plant, there is a problem since iodine-131 is often an important product. During portions of the recovery process at the ORNL Iodine-131 Processing Facility, certain chemicals are used. Among these are NaOH, HNO_3, and H_2SO_4.(13) It is likely that in this process chemical mists containing particles of less than 0.3-micron diameter are produced, as can be seen by referring to Figure 2.

Such particle production could be a major contributor to the lower-than-expected efficiencies of charcoal filters in the iodine-131 isotope-recovery plants.(13) Specifically, it is likely that more than 0.05% of the total iodine-131 activity is somehow associated with particles of less than 0.3-micron diameter.

Effect of Adding Elemental Iodine to an Air Stream In an Effort to Increase Filter Efficiency

The efficiency of a charcoal filter is often low when the iodine concentration in the air stream is low. Since the low efficiency is suggested to be caused somehow by the low iodine concentration, it seems reasonable to attempt to increase the efficiency of a filter by adding more iodine to the air stream.

However, it is believed by the author that iodine concentration per se cannot completely account for the low efficiency. The proposed model of the filter can be used to show that injecting iodine in elemental form upstream from a filter does not improve the efficiency of the filter. Figure 5, an extension of Figure 3c, shows this situation.

Assume that the iodine is injected between the third and fourth filters as elemental iodine only and that there is no increase in the amount of activity associated with the particles. The efficiency of the fourth filter is then (Figure 5)

$$1 - \frac{1{,}000 + 3{,}280}{1{,}000{,}000 + 3{,}645} = 99.6\%.$$

This is a tremendous increase over the 10.1% efficiency of the third filter. However, we have merely succeeded in releasing more iodine from the system, as tabulated below:

Leaving third filter; 0.01 + 3,645 = 3,645 units.

Leaving fourth filter; 1,000 + 3,280 = 4,280 units.

Granted, we need not have injected as much as 1,000,000 elemental units nor need it have been radioiodine. However, the calculation does show that simply injecting elemental iodine to increase the concentration of iodine-131 in the air stream probably produces no increase in the efficiency of the filter. This calculation further illustrates the main purpose of this paper; namely, that the form of the iodine must also be specified.

Of course, if the injected elemental iodine did somehow cause the iodine associated with the particulate to escape and become elemental iodine or did cause the particulate to agglomerate to particles larger than 0.3 micron, the efficiency of the fourth filter would be improved.

CONCLUDING COMMENTS

This paper is not, and was not intended to be, a complete and extensive review of the problems in achieving high efficiencies with filters in removing contaminants, especially iodine-131, from an air stream. Its only purpose is to focus attention on the role that particles in the size range from about 0.001 to 0.3 microns apparently play in the efficiency of filter systems, particularly in full-size operating plants.

It naturally follows, then, that the usual method of expressing the concentration of iodine-131 in an air stream (namely, mg/ft^3) is not sufficient if one is to take these smaller particles into account properly. Rather, as has been suggested by others, it is necessary that information on the particle-size distribution and the amount of iodine-131 activity apparently associated with them should also be given when describing the efficiency of filters.

Further, this paper neglects the effect of deposition of iodine-131 on the walls of a duct. The main reason for doing so is that the percentage of iodine which deposits on the walls of the duct is probably not significant in comparison to

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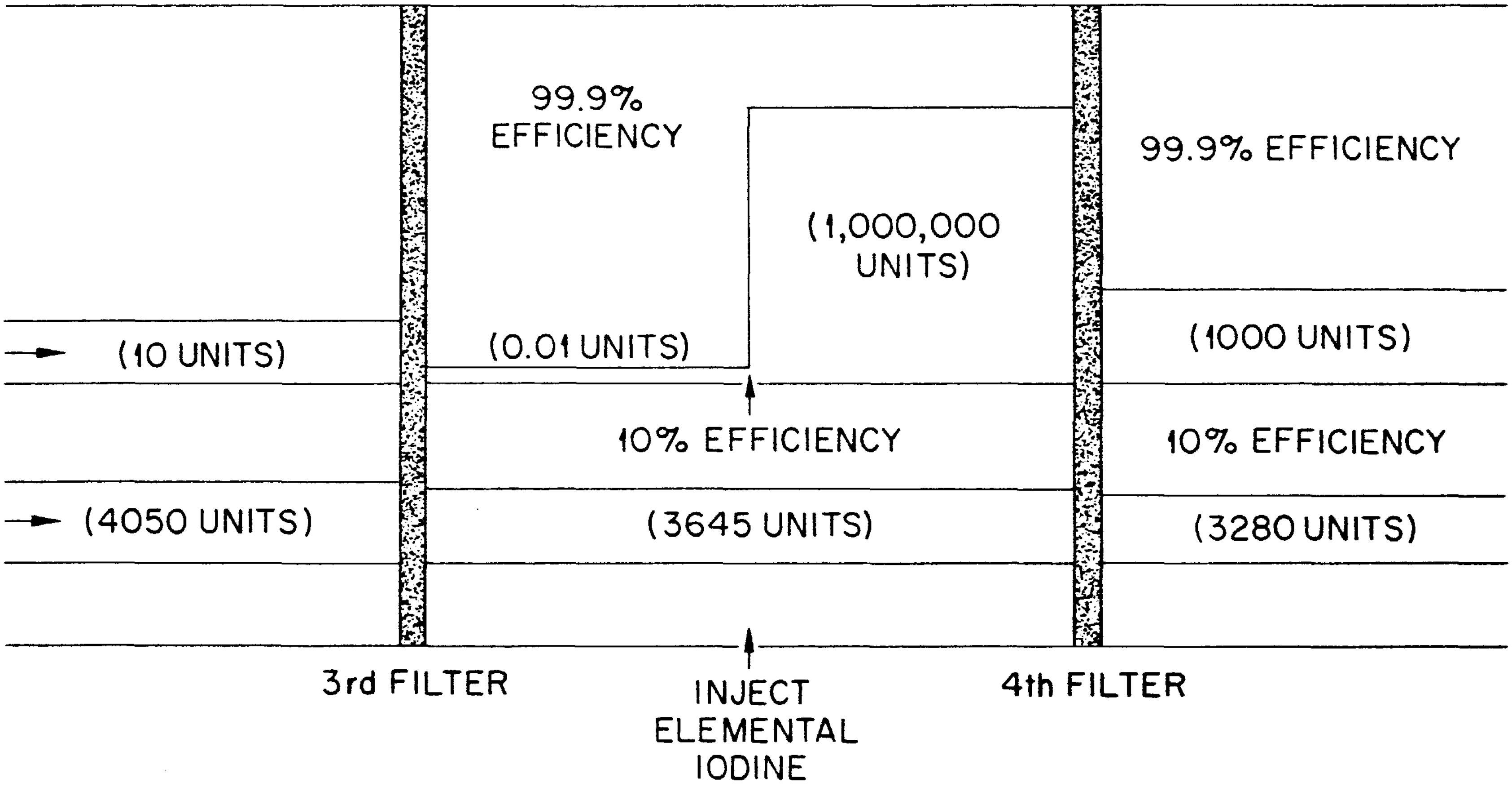

Fig. 5. Calculated Efficiency of a Charcoal Filter when Elemental Iodine is Injected into the Air Stream Ahead of the Filter (Extension of Fig. 3).

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the required removal efficiency of the filter. Furthermore, it is not yet clear whether the iodine so deposited is in elemental form, which would be removed by a charcoal filter anyway, or is associated with the difficult-to-filter particles. It seems more prudent to assume the former until there is proof to the contrary.

It is emphasized that the specific mechanism by which the iodine is associated with the particles is of no great interest for the purpose of this paper. That is to say, it makes no difference whether the iodine is in compound form or attached as elemental iodine to some particle. It is only necessary for the purpose of this paper that the iodine somehow becomes attached to particles of this size or that the particles behave as though they were in this size range. Of course, in actual practice the form of the iodine would most likely be important if attempts were made to somehow increase its particle size or to convert it to elemental iodine. Also, certain forms may attach to particles more readily than others.

Last, it must be remembered that the major purpose of the vast amount of work currently in progress regarding air cleaning is for a specific end result; namely, use in plant-size facilities where extremely large amounts of biologically harmful materials exist. Even in case of a major accident, the design must be such that not more than a very small percentage of these materials can be released to the atmosphere. This means that information which can be used in the design of a practical, over-all filter complex must be available to the designers of these systems. Such information would include the distinctive efficiency of the various components of the air stream for the different types of filters and the amount and types of particles which could be expected from various types of reactor incidents or meltdowns, isotope-recovery plants, and fuel-reprocessing plants. From this information, a "tailor-made" filter system could be designed. This could be a formidable task for some reactors, such as research reactors, which usually have more than one type of fuel either in various test loops or as the reactor fuel. What may be practical in small-scale laboratory work may not be practical in a large plant.

DISCUSSION AND COMMENT

The complete penetration of the 0.3 micron particles, and below, and efficiencies were assumed in this paper.

Session Chairman: If there are no further questions, I will turn the podium over to Dr. First who will be the Chairman of Session IV.

SESSION IV - AIR AND GAS CLEANING METHODS

Afternoon - 22 October 1963

M. W. First, HACL, Chairman

Session Chairman: Since we are running considerably behind time, as I am sure you are aware, instead of taking a stretch as we might like, I am going to continue right on.

Session IV concerns Air and Gas Cleaning Methods. I am sure I do not have to introduce the first speaker, who will discuss "Performance of Diffusion Board for Radioactive Gases and Particulates." If I were to say the speaker is Mr. Air Cleaning I am sure you would understand I am talking about Les Silverman. He is Professor of Environmental Health, Engineering and Radiation Hygiene at Harvard University, and is the Head of the Department of Industrial Hygiene.